International Atomic Energy Agency

INDC(NDS)-428
Distr. G+L

INDC INTERNATIONAL NUCLEAR DATA COMMITTEE

LONG TERM NEEDS FOR NUCLEAR DATA DEVELOPMENT

Texts of papers presented at the Advisory Group Meeting

IAEA Headquarters
Vienna, Austria
28 November – 1 December 2000

Compiled by
M. Herman
IAEA Nuclear Data Section
Vienna, Austria

August 2001

IAEA NUCLEAR DATA SECTION, WAGRAMERSTRASSE 5, A-1400 VIENNA

Reproduced by the IAEA in Austria
August 2001

INDC(NDS)-428
Distr. G+L

LONG TERM NEEDS FOR NUCLEAR DATA DEVELOPMENT

Texts of papers presented at the Advisory Group Meeting

IAEA Headquarters

Vienna, Austria

28 November – 1 December 2000

Compiled by

M. Herman

IAEA Nuclear Data Section
Vienna, Austria

August 2001

FOREWORD

This report contains the texts of the invited presentations delivered at the Advisory Group Meeting on Long Term Needs for Nuclear Data Development. The meeting was organized by the International Atomic Energy Agency (IAEA) and held at IAEA Headquarters, Vienna, Austria, 28 November – 1 December 2000. The texts are reproduced here, directly from the Author's manuscripts with little or no editing, in the order in which the presentations were made at the meeting. For the main conclusions refer to the Summary Report, published as INDC(NDS)-423.

TABLE OF CONTENTS

CROSS SECTION DATA FOR PRODUCTION OF DIAGNOSTIC AND THERAPEUTIC RADIONUCLIDES

S.M.QAIM

Institut für Nuklearchemie, Forschungszentrum Jülich GmbH
D-52425 Jülich, Germany

ABSTRACT

The status of nuclear reaction cross section data for production of medically interesting radionuclides is discussed. The needs for short- and long-term investigations are considered. Whereas short-term investigations refer to improvements in the utilization of known radionuclides, long-term studies pertain to the development of new radionuclides. The major interest is directed to longer-lived β^+ emitters and low-range radiation emitting therapeutic radionuclides. Some challenges involved in the relevant nuclear data work are outlined.

1 Introduction and Present Status

The importance of nuclear reaction cross section data in medical radionuclide production is well known [1,2]. In particular, data are needed to optimise a production process, i.e. to maximise the yield of the desired product and to minimise the level of radionuclidic impurities. In recent years, considerable amount of standardisation work has been done under a Co-ordinated Research Programme (CRP) of the IAEA, and evaluated excitation functions of about 50 nuclear reactions have been reported in a Technical Document [3]. Those reactions cover the major nuclear processes used for monitoring charged particle beams (p, d, ^{3}He and ^{4}He) as well as for production of commonly used diagnostic radionuclides ^{67}Ga, ^{111}In, ^{123}I, ^{201}Tl, etc. in *Single Photon Emission Computed Tomography* (SPECT) and ^{11}C, ^{13}N, ^{15}O, ^{18}F etc. in *Positron Emission Tomography* (PET). In general, a fairly reliable data base has been established. With the exception of a few discrepancies, e.g. data for the ^{124}Xe(p,x)^{123}I process, or a few cases where only single measurements exist, for the present the data needs for routine production of commonly used medical radionuclides at cyclotrons appear to be satisfied. In the case of reactor produced diagnostic radionuclides (^{99m}Tc, ^{125}I, etc.) the data base is anyway quite strong.

Regarding the therapy radionuclides, the situation is somewhat different. These radioisotopes emit corpuscular radiation (α, β^- or Auger electrons) and are commonly produced in a nuclear reactor. The available production data are generally good, especially for the β^- emitting radionuclides like ^{31}P, ^{89}Sr, ^{90}Y, ^{111}Ag, ^{131}I, ^{153}Sm, ^{166}Ho, ^{177}Lu, etc. and the Auger electron emitter ^{125}I. However, to date no effort has been devoted to the evaluation and standardisation of those data.

2 Future Trends

Despite the fact that radionuclide production technology is well established, constant research work is going on either to improve the utilization of the known radionuclides or to develop new radionuclides for novel medical applications. In either case there is considerable need of nuclear data research. In this regard, several considerations have been recently elaborated [4]. Here some salient features are emphasized.

2.1 Short-term investigations

These refer to improvements in the utilization of known radionuclides. They may involve the optimisation of a useful production method or the development of an alternative production route. The formation of ^{124}I, a relatively new β^+ emitting radionuclide, furnishes a typical example. The radionuclide was generally produced via the ^{124}Te(d,2n)^{124}I process. An alternative route involving the ^{124}Te(p,n)^{124}I reaction had been neglected on the basis of some older data [5]. It was considered to be a very low-yield process. Some recent accurate measurements, however, revealed [6] that the threshold of the reaction is shifted to lower energies by about 2.5 MeV. The two sets of data are shown in Fig.1. The new results established the fact that ^{124}I could be produced advantageously via the ^{124}Te(p,n)-reaction. The product is of high radionuclidic purity and a small-sized cyclotron is adequate for production purposes.

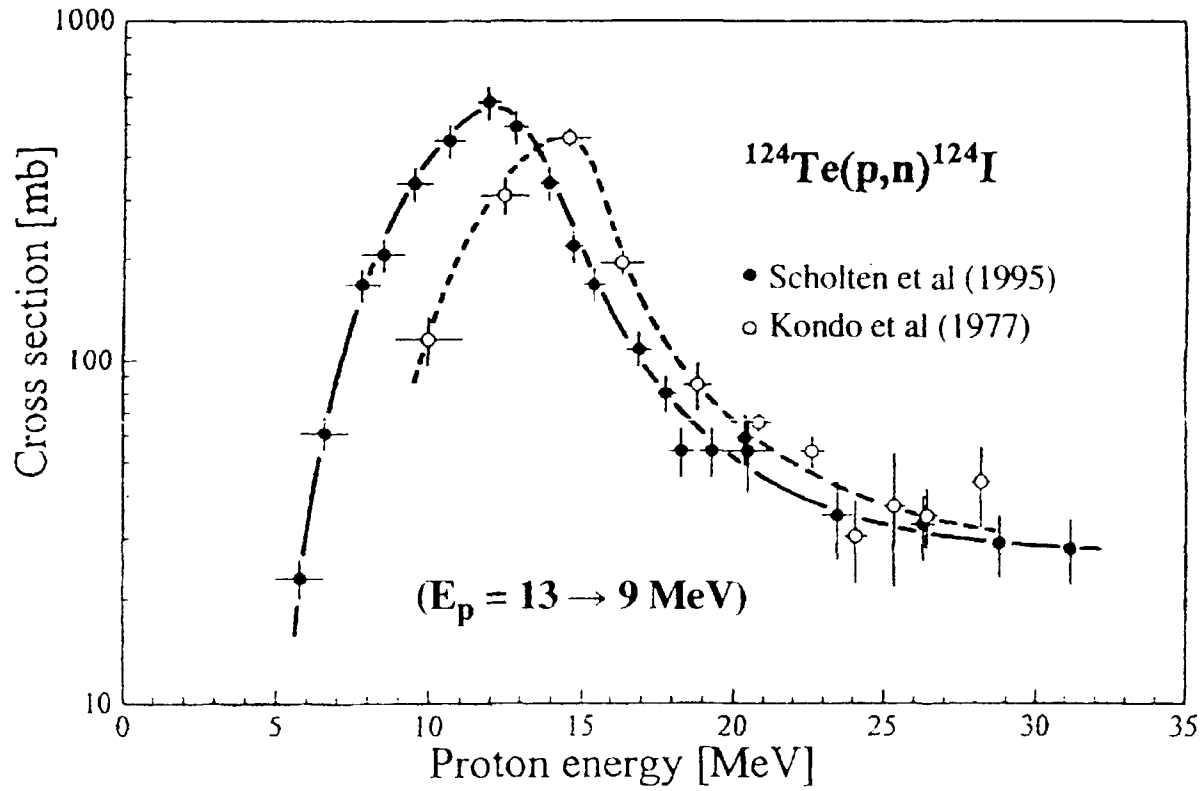


Fig.1 Excitation function of the ^{124}Te(p,n)^{124}I reaction. The older measurements (Kondo et al, 1977, ref. [5]) covered only the energy region above 10 MeV. The new measurement (Scholten et al, 1995, ref. [6]) showed a shift to lower energies. The optimum energy range for production of ^{124}I is E_p = 13 → 9 MeV. With the increasing demand of ^{124}I it is essential to increase its productivity.

Since the $^{124}Te(p,n)^{124}I$ reaction is a relatively low-yield process, the development of yet another route, resulting in higher yield, would be very fruitful. In this regard, recently the $^{125}Te(p,2n)^{124}I$ reaction was investigated [7] and the results are shown in Fig.2. The suitable energy range is E_p = 21 → 15 MeV. The theoretically expected yield of ^{124}I amounts to 81 MBq/μA·h as compared to 16 MBq/μA·h in the $^{124}Te(p,n)^{124}I$ reaction; the level of ^{125}I, however, is higher and careful checks are necessary to clarify whether this level of ^{125}I-impurity is acceptable.

The case of ^{124}I production illustrates typically the data needs regarding diagnostic radionuclides which have to be met on a short-term basis. Other cases would include removal of discrepancies wherever they exist and provision of more extended and quality-assured data base for the production of rather well-established radionuclides.

The short-term nuclear data work regarding the therapeutic radionuclides would consist of a thorough evaluation of the production routes of the commonly used β^- emitters. In most of the cases the (n,γ) reaction is utilized. However, occasionally double neutron capture, (n, charged particle) and (n,f) processes are employed. A few therapy related radionuclides are also produced at a cyclotron.

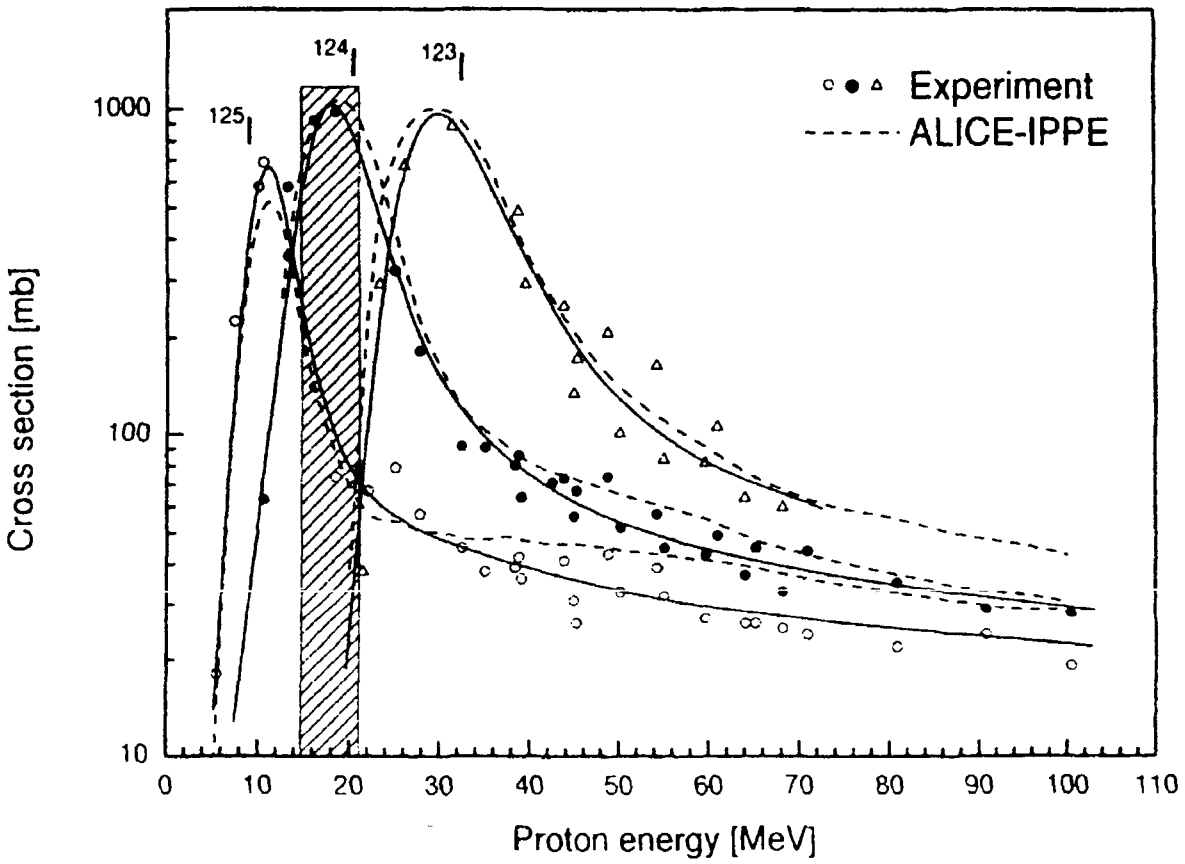


Fig.2 Excitation funtions of $^{125}Te(p,xn)^{123,124,125}I$ reactions. The solid lines are eye-guides. The broken lines show the results of nuclear model calculations using the Code ALICE-IPPE. The shaded area gives the suitable energy range for the production of ^{124}I (diagram taken from Hohn et al, 2001, ref.[7]).

2.2 *Long-term studies*

These pertain to the development of new radionuclides for medical applications. To date the emphasis has been on organic elements. Recent radiopharmaceutical researches have, however, shown that even some metal-complexes could be of considerable interest, both for diagnosis and therapy. Efforts regarding the development of some potentially interesting radionuclides have therefore been intensified in recent years and it is expected that the data needs will enhance as new applications emerge.

The development of new radionuclides is progressing in two directions :

- Longer-lived β^+ emitters for diagnostic purposes
- Therapy related radionuclides

A discussion of the nuclear data work in the two directions is given below.

• *Longer-lived β^+ emitters*

With the increasing significance of PET in diagnostic nuclear medicine, the need for longer-lived β^+ emitters is enhancing. In contrast to the commonly used short-lived β^+ emitters, the longer-lived β^+ emitting nuclides could be employed to study slow metabolic processes like cell proliferation, amino acid metabolism, etc. Furthermore, the corresponding radiopharmaceuticals could be transported to relatively long distances under the "satellite" concept. A few examples of radionuclides under this category are ^{64}Cu ($T_{½}$ = 12.4 h), ^{73}Se ($T_{½}$ = 7.1 h) and ^{76}Br ($T_{½}$ = 16.0 h).

Another application of longer-lived β^+ emitters is as analogue tracers. In this connection, a β^+ emitter could be applied for quantification of SPECT-radiopharmaceuticals. Useful examples are furnished by the systems : ^{94m}Tc for ^{99m}Tc and ^{120g}I for ^{123}I. Similarly, in endotherapy with β^- emitting particles, therapy planning and dosimetry could be done advantageously using a β^+ emitting analogue. Typical examples are ^{86}Y for ^{90}Y and ^{83}Sr for ^{89}Sr.

A summary of the useful or potentially useful β^+ emitting radionuclides is given in Table 1. It is not an exhaustive compilation but a list of some typical radionuclides which have been either recently developed for medical applications or which are in development. Besides decay data (taken from the Table of Isotopes), some potentially useful production routes are given. In some cases good measurements have already been reported. For many others, however, considerably more experimental data will be needed than presently available. Also worth pointing out is that reactions induced by all the four light charged particles (p, d, ^{3}He, α) could be utilized and the energy range would extend up to about 100 MeV. In recent years even the use of heavy ions has been propagated [cf.8]. The yields are, however, low.

The cross section data for the formation of ^{94m}Tc are shown in Fig.3 [9]. It constitutes a typical case regarding the new β^+ emitters. Worth mentioning here is the formation of the isomeric state. The suitable energy range for production is E_p = 13 → 7 MeV. The yield of ^{94m}Tc is high and the level of the ^{94g}Tc impurity amounts to 7 %. In many other nuclides as well, isomeric states may occur. Measurements on the formation of those states appear to be mandatory to keep the level of impurities as low as possible.

Table 1. Nuclear data of some useful or potentially useful longer-lived β^+ emitters

Nuclide	Decay data			Useful production routes	
	$T_{½}$	E_{β^+} [MeV]	I_{β^+} [%]	Nuclear reaction	Energy range of interest [MeV]
^{45}Ti	3.1 h	1.04	86.0	^{45}Sc(p,n)	5 - 20
				^{45}Sc(d,2n)	5 - 20
^{48}V	16.0 d	0.69	49.6	^{48}Ti(p,n)	5 - 20
				^{48}Ti(d,2n)	5 - 20
				^{50}Cr(d,α)	5 - 20
				^{48}Ti(^{3}He,t)	10 - 40
				^{45}Sc(α,n)	10 - 25
^{49}Cr	42.0 min	1.54	~100	^{51}V(p,3n)	20 - 50
				^{48}Ti(^{3}He,2n)	15 - 30
				^{46}Ti(α,n)	10 - 25
^{51}Mn	46.2 min	2.21	~100	^{50}Cr(d,n)	5 - 15
				^{52}Cr(p,2n)	20 - 40
^{52}Mn	5.6 d	0.57	28.0	^{52}Cr(p,n)	5 - 20
				^{52}Cr(d,2n)	5 - 20
				^{54}Fe(d,α)	5 - 20
				^{52}Cr(^{3}He,t)	10 - 40
				^{51}V(^{3}He,2n)	15 - 30
^{52}Fe	8.3 h	0.80	56.5	^{55}Mn(p,4n)	40 - 70
				^{52}Cr(^{3}He,3n)	20 - 40
				^{50}Cr(α,2n)	15 - 35
^{55}Co	17.6 h	1.51	77.0	^{56}Fe(p,2n)	10 - 30
				^{58}Ni(p,α)	5 - 20
				^{54}Fe(d,n)	3 - 15
^{57}Ni	36.0 h	0.84	40.0	^{59}Co(p,3n)	20 - 50
				^{56}Fe(^{3}He,2n)	15 - 30
				^{54}Fe(α,n)	10 - 25
^{61}Cu	3.4 h	1.20	62.0	^{61}Ni(p,n)	5 - 20
				^{61}Ni(d,2n)	5 - 20
				^{59}Co(α,2n)	15 - 35
^{64}Cu	12.7 h	0.57	19.3	^{64}Ni(p,n)	5 - 20
				^{64}Ni(d,2n)	5 - 20
^{66}Ga	9.4 h	4.15	56.5	^{66}Zn(p,n)	5 - 20
				^{66}Zn(d,2n)	5 - 20
				^{65}Cu(^{3}He,2n)	15 - 30
				^{63}Cu(α,n)	10 - 25
^{72}As	26.0 h	3.33	77.0	^{72}Ge(p,n)	5 - 20
				^{72}Ge(d,2n)	5 - 20
				^{71}Ga(^{3}He,2n)	15 - 30

Table 1. continued

^{73}Se	7.1 h	1.68	65.0	^{75}As(p,3n)	20 - 50
				^{72}Ge(^{3}He,2n)	15 - 30
				^{70}Ge(α,n)	10 - 25
^{75}Br	1.6 h	1.74	75.5	^{74}Se(d,n)	3 - 15
				^{76}Se(p,2n)	10 - 30
				^{78}Kr(p,α)	5 - 20
				^{75}As(^{3}He,3n)	20 - 40
^{76}Br	16.0 h	3.98	57.0	^{76}Se(p,n)	5 - 20
				^{76}Se(d,2n)	5 - 20
				^{77}Se(p,2n)	10 - 30
				^{75}As(^{3}He,2n)	10 - 30
^{77}Kr	1.2 h	1.87	~80	^{79}Br(p,3n)	20 - 50
				^{76}Se(^{3}He,2n)	10 - 30
^{82m}Rb	6.2 h	0.80	25.6	^{82}Kr(p,n)	5 - 20
				^{82}Kr(d,2n)	5 - 20
				^{81}Br(^{3}He,2n)	15 - 30
				^{79}Br(α,n)	10 - 25
^{83}Sr	32.4 h	1.23	24.0	^{85}Rb(p,3n)	20 - 50
				^{82}Kr(^{3}He,2n)	10 - 30
^{86}Y	14.7 h	3.15	34.0	^{86}Sr(p,n)	5 - 20
				^{88}Sr(p,3n)	20 - 50
				^{85}Rb(^{3}He,2n)	10 - 30
^{90}Nb	14.6 h	1.50	53.0	^{90}Zr(p,n)	5 - 20
				^{90}Zr(d,2n)	5 - 20
				^{89}Y(^{3}He,2n)	10 - 30
^{94m}Tc	52.5 min	2.47	72.0	^{94}Mo(p,n)	5 - 20
				^{93}Nb(^{3}He,2n)	10 - 30
				^{92}Mo(α,pn)	15 - 40
^{110m}In	1.1 h	2.25	62.0	^{110}Cd(p,n)	5 - 20
				^{110}Cd(d,2n)	5 - 20
				^{109}Ag(^{3}He,2n)	10 - 30
				^{107}Ag(α,n)	10 - 25
^{120g}I	1.3 h	4.0	56.0	^{120}Te(p,n)	5 - 20
				^{120}Te(d,2n)	5 - 20
				^{122}Te(p,3n)	20 - 50
^{124}I	4.2 d	2.14	23.0	^{124}Te(p,n)	5 - 20
				^{124}Te(d,2n)	5 - 20
				^{125}Te(p,2n)	10 - 30
^{152}Tb	17.5 h	2.80	13.0	^{181}Ta(p,spall)	800
				^{nat}Nd(^{12}C,xn)	50 - 120

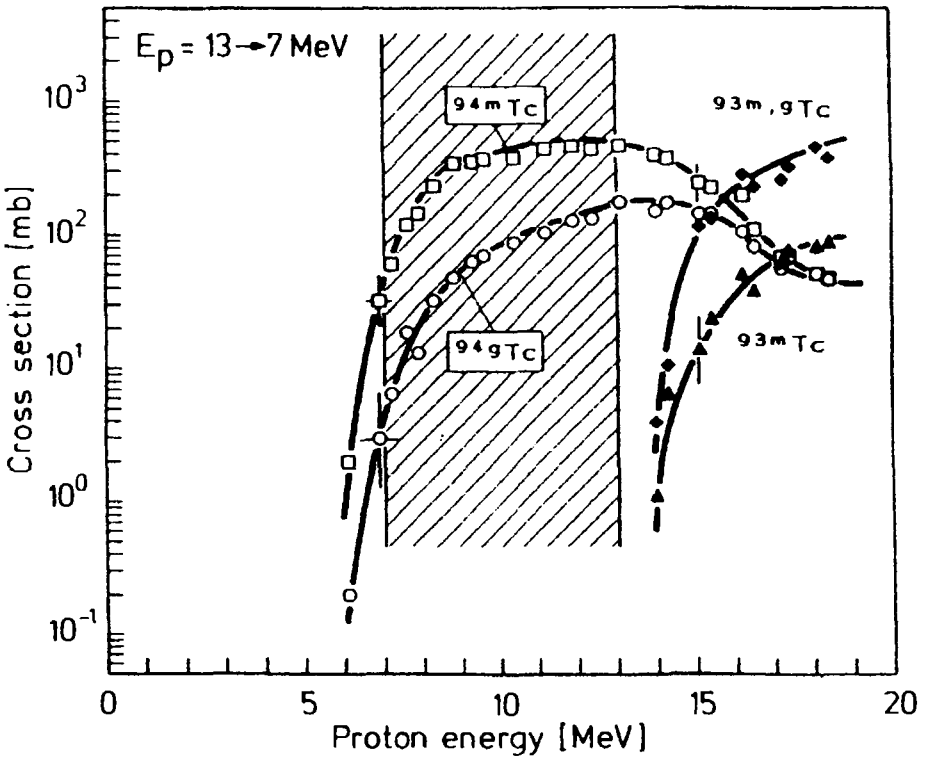


Fig.3 Excitation functions of proton induced nuclear reactions on highly enriched ^{94}Mo leading to the formation of ^{94m}Tc, ^{94g}Tc, ^{93m}Tc and ^{93m,g}Tc. The shaded area gives the suitable energy range for the production of ^{94m}Tc. (diagram taken from Rösch and Qaim, 1993 [ref. 9]).

• ***Therapy related radionuclides***

Many of the potentially useful therapy radionuclides are β^- emitters and their production routes will mainly consist of (n,γ) and double neutron capture processes, the latter in high flux nuclear reactors. Two important examples of the double neutron capture processes are :

$$^{164}\text{Dy}(n,\gamma)^{165}\text{Dy}(n,\gamma)^{166}\text{Dy} \xrightarrow{\text{generator}} {}^{166}\text{Ho}$$
$$(3.4\ \text{d}) \qquad (2.4\ \text{h})$$

$$^{186}\text{W}(n,\gamma)^{187}\text{W}(n,\gamma)^{188}\text{W} \xrightarrow{\text{generator}} {}^{188}\text{Re}$$
$$(69.0\ \text{d}) \qquad (17.0\ \text{h})$$

For these two cases the data are known but, in general, for developing other systems, new measurements would be necessary.

The (n,γ) reaction leads to a product of low specific activity. Efforts are therefore constantly under way to increase the specific activity via the formation of a precursor or generator system. Another method involves the use of a charged particle induced reaction. The recent development works related to the production of ^{64}Cu and ^{186}Re provide good examples. Instead of the (n,γ) process, ^{64}Ni(p,n)^{64}Cu [10] and ^{186}W(p,n)^{186}Re or ^{186}W(d,2n)^{186}Re [11] reactions are successfully applied. All such development studies demand extensive nuclear data measurements.

The need of high specific activity of therapeutic radionuclides, may be partly met via the use of the (n,p) process. This reaction has generally a high threshold and hence the fission type neutron spectrum is not very effective. If a high-intensity d/Be

Table 2. Possibility of production of some therapeutic radionuclides in no-carrier-added form using high-intensity breakup or spallation neutrons

Therapeutic radionuclide	$T_{½}$	Presently used reaction in a nuclear reactor	Alternative reaction using high energy neutrons
^{64}Cu [a]	12.7 h	$^{63}Cu(n,\gamma)$	$^{64}Zn(n,p)$
^{67}Cu [a]	61.9 h	$^{67}Zn(n,p)$ [b]	$^{67}Zn(n,p)$
^{89}Sr	50.5 d	$^{88}Sr(n,\gamma)$	$^{89}Y(n,p)$
^{153}Sm	46.3 h	$^{152}Sm(n,\gamma)$	$^{153}Eu(n,p)$
^{159}Gd	18.5 h	$^{158}Gd(n,\gamma)$	$^{159}Tb(n,p)$
^{166}Ho	26.6 h	$^{165}Ho(n,\gamma)$	$^{166}Er(n,p)$
^{177}Lu	6.7 d	$^{176}Lu(n,\gamma)$ [c]	$^{177}Hf(n,p)$

[a] This radionuclide is better produced at a cyclotron using a charged particle beam.
[b] This process gives a no-carrier-added product but the yield is low.
[c] An alternative route is $^{176}Yb(n,\gamma)^{177}Yb \rightarrow ^{177}Lu$ which gives no-carrier-added product. (1.9 h)

neutron spectral source (such as the one used in therapy) or if a spallation neutron source is available, the use of the (n,p) reaction may be of some advantage. Table 2 gives a list of some radionuclides which could be produced via the (n,p) process. Evidently, a full knowledge of the excitation function, or at least the spectrum averaged cross section, will be needed. The cross section is expected to be low and hence the yield will also be low. The method could therefore not compete with the (n,γ) process as far as the yield is concerned. However, in special cases where very high specific activity is desired, and no other method is available, the suggested process may be worth attempting. It is anticipated that this suggestion will gain more attention when intense fast neutron sources are more readily available.

The radiation dosimetry in an endoradiotherapeutic application is rather empirical if the biodistribution data are not accurately known. For the required tracer studies, therefore, new radionuclides serving as analogues of the therapy radionuclides need to be developed. The analogue radionuclides could be β^+ emitters (see above) or photon emitters which could be measured via SPECT. In the case of the therapy nuclide ^{159}Gd ($T_{½}$ = 18.5 h), for example, ^{147}Gd ($T_{½}$ = 38.1 h) was developed as an analogue SPECT-radionuclide [12]. Evidently, further development of such tracers would require considerable amount of reaction cross section work.

In recent years considerable interest has been aroused in radionuclides emitting low-range but high-intensity and highly-ionising radiation (high LET values) for internal therapy. Thus the trend is shifting from high-energy β^- emitters to low-energy β^--particle, α-particle, X-ray and Auger electron emitters. The related nuclear data problems have been recently discussed in detail [13].

The production data of the commonly used α-particle emitter ^{211}At as well as of the widely used Auger electron emitter ^{125}I are well known. Some other Auger electron emitters like ^{67}Ga and ^{111}In are also well studied since they are commonly used in low

Table 3. Possible production routes of some useful or potentially useful low-range radiation emitting radionuclides

Nuclide	$T_{½}$	E_{max} of emitted particle [MeV]	Useful production routes	
			Nuclear reaction	Energy range of interest [MeV]
^{149}Tb	4.1 h	3.97 (α)	^{181}Ta(p,spall)	800
			^{nat}Nd(^{12}C,x)	50 - 120
^{225}Ac	10.0 d	5.8 (α)	^{226}Ra(p,2n)	10 - 30
^{103}Pd	17.0 d	Auger electrons	^{103}Rh(p,n)	5 - 20
			^{103}Rh(d,2n)	5 - 20
			^{nat}Ag(p,x)	20 - 80
^{140}Nd	3.4 d	Auger electrons	^{140}Ce(^{3}He,3n)	20 - 40
			^{141}Pr(p,2n)	10 - 30
^{195m}Pt	4.0 d	Auger and conversion electrons	^{192}Os(α,n)	10 - 25
^{193m}Pt	4.3 d	Auger and conversion electrons	^{193}Ir(p,n)	5 - 20
			^{192}Os(^{3}He,2n)	10 - 30
			^{190}Os(α,n)	10 - 25

doses as diagnostic radionuclides. Their potential in therapy is being increasingly realized. A few other useful or potentially useful radionuclides are listed in Table 3. Their possible production routes are given. The list will certainly increase in the next few years.

It should be emphasized that cross section measurements on the formation of low-range particle emitters are often very challenging since very clean radiochemical separations and thin source preparation are required. In the spallation process, e.g. in the case of ^{149}Tb, on-line mass separation is mandatory [cf.14]. For producing ^{225}Ac, the radioactive target ^{226}Ra needs very careful handling [cf.15]. The counting methods may involve low-level α-spectrometry, β counting, X-ray spectrometry or liquid scintillation counting. The data for the formation of ^{103}Pd, for example, have been recently measured via X-ray spectrometry [16, 17]. Thus interdisciplinary techniques are of great significance in these studies.

3. Conclusions

As discussed above, the data base for most of the radionuclides commonly used in diagnostic studies is good. Regarding the therapeutic radionuclides, however, no evaluated data file has been developed. The short-term nuclear data investigations refer to improvements in the utilization of known radionuclides. In the case of diagnostic radionuclides this would involve the removal of discrepancies in the existing data, some integral tests of the recently evaluated data, and some new measurements in search of alternative routes of production. In the case of therapeutic radioisotopes, a critical look at the available data and evaluations similar to those for the diagnostic

radioisotopes are urgently needed. The long-term data studies would be related to the development of longer-lived β^+ emitters for diagnostic studies, and low-energy β^--particle, α-particle, Auger electron and X-ray emitters for internal therapy. Extensive interdisciplinary studies would be mandatory to establish the required data bases.

References

[1] S.M. Qaim, Radiochimica Acta **30** (1982) 147.

[2] S.M. Qaim, Radiochimica Acta **89** (2001) 189.

[3] K. Gul, A. Hermanne, M.G. Mustafa, F.M. Nortier, P. Oblozinsky, S.M. Qaim, B. Scholten, Y. Shubin, S. Takács, F. Tárkányi, Y. Zhuang, *"Charged particle cross section data base for medical radioisotope production: diagnostic radioisotopes and monitor reactions"*, IAEA-TECDOC-1211 (2001) pp. 1-285.

[4] S.M. Qaim, Radiochimica Acta **89** (2001) 223.

[5] K. Kondo, R.M. Lambrecht, A.P. Wolf, Int.J.Appl.Radiat.Isot. **28** (1977) 395.

[6] B. Scholten, Z. Kovács, F. Tárkányi, S.M. Qaim, Appl. Radiat.Isot. **46** (1995) 255.

[7] A. Hohn, F.M. Nortier, B. Scholten, T.N. van der Walt, H.H. Coenen, S.M. Qaim, Appl.Radiat.Isot. **55** (2001) 149.

[8] B.J. Allen, G. Goozee, S. Sarkar, G. Beyer, C. Morel, A.P. Byrne, Appl.Radiat.Isot. **54** (2001) 53.

[9] F. Rösch, S.M. Qaim, Radiochimica Acta **62** (1993) 115; Erratum **75** (1996) 227.

[10] F. Szelecsényi, G. Blessing, S.M. Qaim, Appl.Radiat.Isot. **44** (1993) 575.

[11] F. Szelecsényi, S. Takács, F. Tárkányi, M. Sonck, A. Hermanne, J.Labell.Compd. Radiopharm. **42** (1999) 912.

[12] F.-O. Denzler, F. Rösch, S.M. Qaim, Radiochimica Acta *69* (1995) 209.

[13] S.M. Qaim, Radiochimica Acta **89** (2001) 297.

[14] G.J. Beyer, University Hospital, Geneva, Switzerland, private communication (2001).

[15] W. Janssens, Institute for Transuranium Elements, Karlsruhe, Germany, private communication (2001).

[16] M. Faßbender, F.M. Nortier, I.W. Schroeder, N. van der Walt, Radiochimica Acta **87** (1999) 87.

[17] A. Hermanne, M. Sonck, A. Fenyvesi, L. Daraban, Nucl.Instr.Methods B **170** (2000) 281.

LONG TERM NUCLEAR DATA NEEDS FOR INTERNAL RADIATION DOSIMETRY

T.W. BURROWS[1]

[1]Brookhaven National Laboratory, USA

Abstract

The Evaluated Nuclear Structure Data File (ENSDF) is the principle source of nuclear data for internal radiation dosimetry and is, therefore, described briefly. Nuclear data needs and accuracy requirements for internal radiation dosimetry are summarized. Currently available sources of internal radiation dosimetry data are outlined and the need for traceability and documentation of these data is discussed.

1 Evaluated Nuclear Structure Data File

The National Nuclear Data Center (NNDC), Brookhaven National Laboratory, USA on behalf of the IAEA-sponsored International Nuclear Structure and Decay Data Network maintains the Evaluated Nuclear Structure Data File (ENSDF) [1]. There are ~3200 datasets of possible interest to internal radiation dosimetry contained in this database. The ENSDF has been the starting point for most publications and databases relevant to internal radiation dosimetry since about 1978.

Some limitations in this file related to the evaluation philosophy and current formats do require additional work to satisfy some of the dosimetry needs. In terms of philosophy, the decay datasets in the ENSDF usually reflect the "best" information for that decay mode; therefore, the energy of the same γ-ray observed in different decays may differ. At present the decay datasets contain only the total internal conversion coefficients for each atomic shell; this may result in insufficient detail for some radiations. In many cases, for the metastable states populated in the decay the datasets do not contain sufficient information for direct use in dosimetry applications.

2 Nuclear Data Needs

The nuclear and related data required for internal radiation dosimetry are the following:

- Branching ratios (*e.g.*, $\%\alpha$ or $\%\beta^-$), half-lives ($T_{½}$), independent or cumulative spontaneous fission yields
- Radiation information:
  - Electromagnetic radiations (γ-rays):
    - Photon energies and intensities
    - Conversion coefficients — These are required to calculate the Auger- and conversion-electron and x-ray energies and intensities
    - Conversion-electron energies and intensities
    - Electron/positron pair formation coefficients
  - α-ray energies and intensities and the recoil energies of the daughter nuclide
  - β^-, β^+, and electron capture:
    - $\beta^\pm$ average and end-point energies and intensities — for many problems, involving doses over short distances, one needs the actual spectrum to get a true picture of the dose distributions.
    - Electron capture fractions — these are required to calculate the Auger-electron and x-ray energies and intensities.
  - Atomic radiations associated with conversion electrons and electron capture
    - Auger-electron energies and intensities — Auger electrons, along with α's, are very useful in nuclear medicine since they exhibit linear energy transfer. Therefore, in some cases, a very detailed description of the Auger electron spectrum is required.
    - X-ray energies and intensities
  - Internal bremsstrahlung from $\beta^\pm$ and electron capture
  - Spontaneous fission prompt neutron and γ-ray average energies and intensities
  - External bremsstrahlung associated with $\beta^\pm$ and Auger and conversion electrons — the spectrum is dependent on the media surrounding the radioactive source.

Estimates of the number of radionuclides for which data are required range from about 250 to 1800. The lower figure of 250 represents those nuclides presently being used or studied in nuclear medicine while the higher figure of 1800 probably represents the needs of radiation protection. Including possible contaminants and daughter nuclei, the total number of radionuclides for nuclear medicine range from about 400 to 800. New production methods (possibly including Radioactive Ion Beam facilities or Accelerator Driven Systems (ADS)) may increase the number of radionuclides useful to nuclear medicine. Also, ADS and advanced accelerator designs will probably increase the number of radionuclides whose data will be required for radiation protection.

The nuclear data used in internal radiation dosimetry should at least be consistent with the current experimental and theoretical data (*e.g.*, In 1978 $T_{1/2}(^{79}Se)$ was 65,000 y; the current value is 1.1×10^6 y.). Where accuracy may be important, the nuclear data should be current.

3 Nuclear Data Accuracy Requirements

Generally, the accuracy requirements of the nuclear data are not very stringent since two other factors dominate in the dose equations. These factors are the biokinetic data and the absorbed fractions (energy emitted in a source that is absorbed in a target). These values can have quite large uncertainties, which sometimes dwarf, by orders of magnitude, the uncertainties in the nuclear data. This may change as the knowledge of these processes improves.

For the ~250 nuclides used or being studied for diagnostic purposes, the accuracy of the nuclear data should be as good as possible. This will allow a more precise calculation of the dose, improving the "quality of life" for the patient.

4 Accessibility of Internal Radiation Dosimetry Data

Internal radiation dosimetry data are available in printed and electronic form. There are also several computer programs that are useful. Following are summaries of some of these sources.

Publications		
Publication	**ENSDF[a]**	**Comments**
DOE/TIC-11026 [2]	1978	Nuclear data are over twenty years old but data still extensively cited.
ICRP-38 [3]	1978	1. Nuclear data are over twenty years old but data still extensively cited. 2. Fairly detailed Auger electron data. 3. Estimates of the prompt neutron and γ-ray average energies from spontaneous fission.
MIRD: Radionuclide Data and Decay Schemes [4]	1987?	Fairly detailed Auger electron data.
Table of Radioactive Isotopes [5]	1984?	1. Fairly detailed Auger electron data. 2. Continua $\beta^{\pm}$ and internal bremsstrahlung data tables but probably insufficient detail.

[a] Approximate year of the ENSDF data used in the publication.

Electronic Access		
Database	**Centers or Organizations[b]**	**Comments**
MIRD	NNDC, IAEA-NDS, CJD	1. Generated "on the fly" from ENSDF. 2. Similar format to ICRP-38 or MIRD.
MIRD	SNM	Site established in June 1999 but no data available as of January 7, 2001.
NuDat	NNDC, IAEA-NDS, CJD	1. Updated from ENSDF every six months. 2. Similar format to DOE/TIC-11026. 3. All radiations with an intensity greater than 10^{-12}% listed. 4. PC version also available.
WWW Table of Radioactive Isotopes	Lund, LBNL-IP	Last updated February 1999.
Evaluated Nuclear Data File (ENDF)	NNDC, IAEA-NDS, CJD, NEADB	1. ENDF/B-VI, JEF-2, JENDL-3, *etc.* 2. Extensive set of decay data for fission-product, actinide, and other radionuclides. 3. Spontaneous fission yields and prompt neutron and γ-ray intensities and average energies. 4. Well-documented and computer-readable format. 5. Format allows storage of continua spectra.

[b] The database is available by the World Wide Web or TELNET at the centers indicated.

CJD — Russian Nuclear Data Center (Center Jadernykh Dannykh), A.I. Leipunski Institute of Physics and Power Engineering (rndc.ippe.obninsk.ru).

IAEA-NDS — IAEA Nuclear Data Section, Vienna, Austria (www-nds.iaea.or.at).

LBNL-IP — Isotope Project, Lawrence Berkeley National Laboratory, Berkeley, USA (ie.lbl.gov).

Lund — Lund University, Lund, Sweden (nucleardata.nuclear.lu.se/nucleardata).

NEADB — OECD Nuclear Energy Agency Data Bank, Paris, France (www.nea.fr/html/databank).

NNDC — National Nuclear Data Center, Brookhaven National Laboratory, Brookhaven, USA (www.nndc.bnl.gov).

SNM — Society of Nuclear Medicine, USA (www.snm.org).

Computer Programs		
The following programs all use ENSDF-formatted [1] files as input.		
Code	**Laboratory or Center**	**Comments**
EDISTR	Oak Ridge National Laboratory, USA	1. Detailed calculation of conversion electrons, Auger electrons, and x-rays. 2. Continua $\beta^{\pm}$ and internal bremsstrahlung spectra. 3. External bremsstrahlung in different absorbing media of interest.
RadList	NNDC, Brookhaven National Laboratory, USA	1. Descendant of MEDLST (ORNL). 2. Less detailed calculation of conversion electrons, Auger electrons, and x-rays than EDISTR. 3. Continua $\beta^{\pm}$ and internal bremsstrahlung spectra. 4. Multiple output formats (ENDF, MEDLST, MIRD, and NuDat).
SDF2NDF	CEA Bruyeres-le-Chatel, Service de Physique Nucleaire, France	1. Descendent of RadList. 2. Primary emphasis on ENDF.

5 Documentation and Traceability

The original source of the nuclear data (usually ENSDF of a certain date), auxiliary data used (*e.g.*, Auger-electron and x-ray yields), and calculational methods employed need to be well documented. This is generally the case in the sources outlined in 4. Limitations in the nuclear data or their vintage may require revisions to the original data. These revisions need to be documented and archived in a manner, which will make this information readily accessible.

6 Summary

Although it has a few limitations, the Evaluated Nuclear Structure Data File (ENSDF) [1] has been used as a starting point for publications and databases relevant to internal radiation dosimetry for over twenty years. The nuclear data required for dosimetry applications include half-lives, branching ratios, and the energies and intensities of the emitted radiations. For selected nuclides, a more detailed representation of the Auger-electron spectra and continuum $\beta^{\pm}$ than in the past is required. The nuclear data used should at least be consistent with current knowledge and, in specific instances, should be

current. As new methods of radioisotope production are developed and new designs of critical reactors and accelerator driven systems mature, the number of radionuclides for which data are needed will probably increase both for nuclear medicine and radiation protection applications.

7 Acknowledgements

I thank Dave Kocher (Oak Ridge National Laboratory), Mike Stabin (Vanderbilt University), and my colleagues at the National Nuclear Data Center for their input and the many users of the NNDC systems for their useful feedback.

This work has been funded by the U.S. Department of Energy under contract number DE-AC02-98CH10886 with Brookhaven Science Associates.

References

[1] J.K.Tuli, report BNL-NCS-51655-Rev.87, Brookhaven National Laboratory (1987).

[2] D.C.Kocher, report DOE/TIC-11026, U.S. Department of Energy, Oak Ridge (1981).

[3] ICRP (International Commission on Radiological Protection), ICRP Publication 38, Ann. ICRP 11-13 (1983).

[4] D.A.Weber, K.F.Eckerman, L.T.Dillman, and J.C.Ryman, MIRD: ***Radionuclide Data and Decay Schemes*** (Society of Nuclear Medicine, New York, 1989).

[5] E.Browne, R.B.Firestone, and V.S.Shirley, ***Table of Radioactive Isotopes*** (John Wiley & Sons, New York, 1986).

Nuclear Data For Fast Neutron and Proton Therapy

M. B. Chadwick[1], D. T. L. Jones[2], H. H. Barschall[3+], R. S. Caswell[4], P. M. DeLuca Jr.[3], J-P. Meulders[5], A. Wambersie[5], H. Schuhmacher[6], P. G. Young[1], L. J. Cox[7], G. M. Hale[1], U. J. Schrewe[6], J. V. Siebers[8*]

[1]Los Alamos National Laboratory, Los Alamos NM, USA
[2]National Accelerator Centre, Faure, SOUTH AFRICA
[3]University of Wisconsin, Madison WI, USA
[4]National Institute of Standards and Technology, Gaithersburg MD, USA
[5]Université Catholique de Louvain, Louvain-la-Neuve, BELGIUM
[6]Physikalisch-Technische Bundesanstalt, Braunschweig, GERMANY
[7]Lawrence Livermore National Laboratory, Livermore CA, USA
[8]Loma Linda University Medical Center, Loma Linda CA, USA

+Deceased
*Current address: Medical College of Virginia, Richmond, VA, USA

ICRU Report 63 entitled "Nuclear Data for Neutron and Proton Radiotherapy and for Radiation Protection" has recently been published. The present paper presents an overview of this report, along with examples of some of the results obtained for evaluated nuclear cross sections and kerma coefficients. These cross sections are evaluated using a combination of measured data and the GNASH nuclear model code for elements of importance for biological, dosimetric, beam modification and shielding purposes. In the case of hydrogen both R-matrix and phase-shift scattering theories are used. Neutron cross sections and kerma coefficients were evaluated up to 100 MeV and proton cross sections up to 250 MeV.

Introduction

An International Commission on Radiation Units and Measurements (ICRU) report which describes nuclear data that are needed in fast neutron and proton radiotherapy studies and for radiation protection has recently been published (ICRU Report 63 [19]). Neutron cross sections and kerma coefficients are presented up to 100 MeV and proton cross sections up to 250 MeV. Potential uses of these data include their implementation in radiation transport and treatment planning computer codes to optimize dose delivery to the treatment volume; studies of the impact of nuclear reactions on the relative biological effectiveness of neutron and proton therapy beams; determination of radiation shielding requirements; and use of kerma coefficients to determine absorbed dose for a given neutron energy distribution, and to convert the absorbed dose, measured with a dosimeter of a given material composition, to absorbed dose in tissue.

The nuclear cross sections are evaluated using a combination of measured data and GNASH [36] nuclear model calculations. Measurements that have determined cross sections and kerma coefficients are reviewed, but since there are only a limited number of experimental data sets for biologically-important target elements, theoretical predictions are needed to supplement these data. Numerous benchmark comparisons are presented that compare the model predictions with measured data to validate the calculations of energy- and angle-dependent emission spectra, as well as total, non-elastic, and elastic scattering cross sections. For hydrogen, an evaluation is described that uses both R-matrix and phase-shift scattering theories to represent neutron-proton reaction data. Kerma coefficients are derived from the evaluated neutron cross sections and presented for individual elements as well as for ICRU muscle, A-150 tissue-equivalent plastic and other substances.

The evaluated cross sections and kerma coefficients are tabulated in the report for neutron- and proton-induced reactions on H, C, N, O, Al, Si, P, Ca, Fe, Cu, W, and Pb. Most detailed information is provided for the most important elements, with less information for the others. However, complete tabulations on a fine incident-energy grid are provided for all target elements as electronic files on an accompanying compact disc (CD).The CD also contains the same cross section evaluations in the Evaluated Nuclear Data File (ENDF) format which are useful for implementation in radiation transport calculations.

Evaluation Methods

The evaluated nuclear cross sections and kerma coefficients which are presented in the report were determined by nuclear model calculations using the GNASH code [36] and

were benchmarked to available measurements [8, 9, 10]. The GNASH nuclear model code applies theories for compound nucleus, pre-equilibrium and direct reaction mechanisms. Optical model calculations serve to determine total, non elastic and elastic scattering cross sections. Making use of experimental data is particularly important for the analysis of the interaction of neutrons and protons with biologically-important elements, since an accurate theoretical description is generally difficult because of the non-statistical properties of light nuclei (caused by their widely-spaced nuclear levels). However, while there have been some useful recent measurements which have expanded the experimental database, the experimental information above 15 MeV is still relatively sparse, and for this reason nuclear theories and models provide a valuable tool for supplementing the measured data. Additionally, a nuclear-model computer code can generate the large amount of information specifying a reaction (cross sections for the secondary reaction products at all out going energies and angles) in a manner which conserves energy, angular momentum, parity, and unitarity (flux conservation).

There are a number of previous studies which aimed at determining neutron cross sections above 20 MeV for biologically-important elements, using nuclear model calculations. The most important of these are the calculations of Brenner and Prael [3], and Dimbylow [12], both of which show extensive comparisons of their results with experimental data. The present work represents an advance over these earlier approaches principally in two ways: (1) it makes use of recent improvements in nuclear model calculations and optimizes them for describing nuclear reactions in the 0-250 MeV energy region; and (2) new experimental information, which has become available since the earlier studies, is used extensively to guide the calculations. Perhaps the most important measurements of non elastic processes are the neutron cross section measurements by Benck et al. [2], Haight et al. [14], Nauchi et al. [25], Slypen et al. [31, 32] and Subramanian et al., [33, 34] and the proton cross section measurements by Bertrand and Peelle [2], Cowley [11], Förtsch et al. [13] and Meier et al. [21, 22].

Results and Comparisons with Measurements

ICRU Report 63 [19] provides extensive comparisons between the evaluated nuclear data and measured cross sections. In this paper we provide two illustrative examples taken from these comparisons: one particularly relevant to neutron therapy studies (neutron-induced emission spectra for oxygen); and one particularly relevant to proton therapy studies (proton-induced total nonelastic cross sections up to 250 MeV). In neutron therapy, calculations of energy deposition, and secondary neutron, photon, and charged particle production, depend critically on the accuracy of the nuclear reaction cross sections. Figure

1 shows the calculated proton, deuteron, and alpha-particle angle-integrated emission spectra for 27, 40 and 60 MeV neutrons incident on oxygen. Experimental data are shown from Benck et al. [2] and Subramanian et al. [33, 34], and the agreement is seen to be good. Calculations by Brenner and Prael [5], shown as a dashed line, are also shown for comparison. An accurate description of such microscopic reaction cross sections is important in radiation transport simulations of radiation therapy since oxygen is the most abundant tissue element (by mass), and a significant fraction of the energy deposited by neutrons in tissue is due to light secondary charged particles produced in neutron nonelastic interactions.

In proton therapy, protons lose energy mainly through electromagnetic interactions with atomic electrons, and because of the relatively small mass of the electron the protons lose only a small fraction of their energy and are only deflected by small angles in each interaction. The probability of nuclear reactions occuring increases with proton energy, but is still relatively small in the therapeutic energy range. However, these reactions in general remove primary protons from the beam and result in the production of secondary particles such as neutrons, gammas, protons, light ions and heavier recoil nuclei which can be emitted at large angles to the incident proton beam thus reducing the energy deposition along the beam path. The relative energy depostion of the neutron-induced heavier secondary charged particles is highest at, or just beyond, the maximum range of the primary proton beam [18]. Their contribution to the integral absorbed dose is small, but their enhanced biological effect can complicate dosimetric, biological and clinical phenomena. Particles emitted out of the beam can also result in small, but undesirable absorbed doses to a patient outside the target. Figure 2 shows the evaluated total nonelastic cross sections for protons on C, O, Fe and Pb up to 300 MeV compared with experimental data [6]. The evaluated results are based on optical model calculations, but small modifications to the calculated results were made to improve agreement with measurements. Carbon and oxygen are two of the most important tissue elements; iron and lead are present in accelerator collimators, shielding, and beam modifiers.

Kerma coefficients are derived from the evaluated microscopic neutron cross sections. ICRU Report 63 [19] presents comparisons between these kerma coefficients and experimental kerma coefficient data, obtained from both integral measurements of the ionization due to the secondary charged particles and from cross section measurements. Results are shown for both individual elements, and for mixtures and compounds, and the agreements are found to be good in most cases. An example is shown in Figure 3 for A-150 tissue-equivalent plastic (to make A150 electrically conductive carbon essentially replaces the tissue oxygen). Results from a previous compilation [7] are also shown for comparison. A particularly important quantity in neutron therapy is the kerma ratio for ICRU-muscle to

A150 plastic, since this can be used to relate A150 plastic ionization chamber dose measurements to dose in muscle. By convoluting this quantity with the neutron therapy source fluence spectra, and averaging the results for the National Accelerator Centre (South Africa), Fermi National Accelerator Laboratory (USA), and a p(66)/Be neutron fluence spectrum assumed by Awschalom et al. [1] a value of 0.94 is obtained. This is in good agreement with the recommendation of 0.95 in both the international neutron dosimetry protocol [24] and in ICRU Report 45 [17]. A direct measurement of this ratio (using microdosimetric techniques) in the National Accelerator Centre's p(66)/Be beams gave 0.92 ± 0.02 [4]. Although the above ratios are consistent within the uncertainties there is a need for more accurate data for clinical dosimetry purposes. There are significant discrepancies based on microscopic data evaluations for C and particularly for O.

Two selected examples of comparisons of measured data with different Monte Carlo code calculations are shown in Figures 4 and 5. In both cases the agreement is excellent.

Conclusion

The current state of knowledge of the most important nuclear data required for calculations pertaining to fast neutron and proton therapy appears to be adequate. Predictions of nuclear models fit the microscopic experimental data reasonably well. Integral neutron experimental measurements are consistent with calculations using evaluations of microscopic data. Monte Carlo calculations using currently available nuclear data evaluations also agree well with various measurements made in therapy beams.

Neutron interactions on C and O are the only reactions for which better data may be required to provide more accurate values of the A150/muscle kerma ratio which is an important parameter in neutron dosimetry.

References

1. Awschalom M, Rosenberg I, Mravca A. Kerma for various substances averaged over the spectra of fast neutron therapy beams: a study in uncertainties. Med. Phys 1983; 10: 395-409.

2. Benck S, Slypen I, Meulders J-P, Corcalcuic V. Light charged particle production induced by fast neutrons on carbon, oxygen and aluminium. In: Reffo G, Ed. Proc of the International Conference on Nuclear Data for Science and Technology, Trieste, Italy, 1997. Bologna, Italy: ENEA .

3. Bertrand FE, Peelle RW. Complete hydrogen and helium particle spectra from 30 to 60 MeV proton bombardment on nuclei with A=12 to 209 and comparison with the intranuclear cascade model. Phys Rev 1973; 8: 1045 -1064.

4. Binns PJ. Personal Communication, 1998.

5. Brenner DJ, Prael RE. Calculated differential secondary-particle production cross sections after nonelastic neutron interactions with carbon and oxygen between 15 and 60 MeV. Atomic Data and Nuclear Data Tables 1989; 41: 71-130.

6. Carlson RF. Proton-nucleus total reaction cross sections and total cross sections up to 1 GeV. Atomic and Nuclear Data Tables 1996; 63: 93-116.

7. Caswell RS, Coyne JJ, Randolph ML. Kerma factors for neutron energies below 30 MeV. Rad Res 1980; 83: 217-254.

8. Chadwick MB, Blann M, Cox LJ, Young PG, Meigooni AS. Calculation and evaluation of cross sections and kerma factors for neutrons up to 100 MeV on carbon. Nucl Sci Eng 1996; 123: 17-37.

9. Chadwick MB, Young PG. Calculation and evaluation of cross sections and kerma factors for neutrons up to 100 MeV on $^{16}0$ and ^{14}N. Nucl Sci Eng 1996; 123: 1-16.

10. Chadwick MB, Young PG, MacFarlane RE et al. Cross section evaluations to 150 MeV for accelerator-driven systems and implementation in MCNPX. Nucl Sci Eng 1999; 131: 293-328.

11. Cowley AA. Personal Communication,1997.

12. Dimbylow PJ. Neutron cross-section and kerma value calculations for C, N, O, Mg, Al, P, S, Ar and Ca from 20 to 50 MeV. Phys Med Biol 1982; 27: 989-1001.

13. Förtsch SV, Cowley AA, Pilcher JV et al. Continuum yields from $^{12}C(p-p)$ at incident proton energies of 90 and 200 MeV. Nucl Phys 1988; A485: 258-270.

14. Haight RC, Lee TM, Sterbenz SM et al. Alpha-particle emission from carbon bombarded with neutrons below 30 MeV. In: Dickens JK, Ed. Proc of the International Conference on Nuclear Data for Science and Technology, Gatlinburg, TN. La Grange Park IL: American Nuclear Society, 1994: 311-313.

15. Hartman Siantar C, Chandler WP, Rathkopf JA et al. Peregrine: An all-particle Monte Carlo code for radiation therapy. In: Proc. of the International Conference on Mathematics and Computations, Reactor Physics and Environmental Analyses, Portland, Oregon, La Grange Park IL: American Nuclear Society, 1995: 857-865.

16. Hughes HG, Adams KJ, Chadwick MB, et al. Status of the MCNP/LCS merger project. In: Proc. of the 1998 Radiation Protection and Shielding Division Topical Conference, Los Alamos National Laboratory, La Grange Park, IL: American Nuclear Society, 1998: 188-195.

17. ICRU Report 45. Clinical neutron dosimetry. Part 1: Determination of absorbed dose in a patient

treated by external beams of fast neutrons. Bethesda, MD: International Commission on Radiation Units and Measurements, 1989.

18. ICRU Report 59. Clinical proton dosimetry. Part 1: Beam production, beam delivery and measurement of absorbed dose. Bethesda MD: International Commission on Radiation Units and Measurements, 1997.

19. ICRU Report 63. Nuclear data for neutron and proton radiotherapy and for radiation protection. . Bethesda, MD: International Commission on Radiation Units and Measurements, 2000.

20. McDonald JC, Cummings FM. Calorimetric measurements of carbon and A-150 plastic kerma factors for 14.6 MeV neutrons. Rad Prot Dosim 1988; 23: 31-33.

21. Meier MM, Clark DA, Goulding CA et al. Differential neutron production cross sections and neutron yields from stopping-length targets for 113-MeV protons. Nucl Sci Eng 1989; 102: 310-321.

22. Meier M M, Amian WB, Goulding CA, et al. Differential neutron-production cross-sections for 256 MeV protons. Nucl Sci Eng 1992; 110: 289-298.

23. Menzel H, Bühler G, Schuhmacher H, et al. Ionization distributions and A-150 plastic kerma for neutrons between 13.9 and 19.0 MeV measured with a low pressure proportional counter. Phys Med Biol 1984; 29: 1537-1554.

24. Mijnheer BJ, Wootton P, Williams JR, et al. Uniformity in dosimetry protocols for therapeutic applications of fast neutron beams. Med Phys 1987; 14: 1020-1026.

25. Nauchi Y, Baba M, Sanami T et al. Measurement of (n,xp) and (n,xd) double differential cross sections of Al and C for ten's MeV neutrons. In: Reffo G, Ed. Proc of the International Conference on Nuclear Data for Science and Technology, Trieste, Italy, 1997. Bologna, Italy: ENEA

26. Pihet P, Guldbakke S, Menzel HG, Schuhmacher H. Measurement of kerma factors for carbon and A-150 plastic: neutron energies from 13.9 to 20.0 MeV. Phys Med Biol 1992; 37: 1957-1976.

27. Schrewe UJ, Brede HJ, Gerdung S et al. Determination of kerma factors of A-150 plastic and carbon at neutron energies between 45 and 66 MeV. Rad Prot Dosim 1992; 44: 21-24.

28. Schrewe UJ, Newhauser WD, Brede HJ, DeLuca PM Jr. Neutron kerma factor measurements in the energy range between 5 MeV and 66 MeV. In: Reffo G, Ed. Proc of the International Conference on Nuclear Data for Science and Technology, Trieste, Italy, 1997. Bologna, Italy: ENEA .

29. Schuhmacher H, Brede HJ, Henneck R et al. Measurement of neutron kerma factors for carbon and A-150 plastic at neutron energies of 26.3 MeV and 37.8 MeV. Phys Med Biol 1992; 37: 1265-1281.

30. Shin K, Yoshiaki I, Miyahara K, et al. Transmission of intermediate-energy neutrons and associated gamma rays through iron, lead, graphitee and concrete shields. Nucl Sci Eng 109; 1991: 380-390.

31. Slypen I, Corcalciuc V, Meulders J-P. Proton and deuteron production in neutron-induced reactions on carbon at E_n=42.5, 62.7 and 72.8 MeV. Phys Rev 1995; C51: 1303-1311.

32. Slypen I, Corcalciuc V, Meulders J-P, Chadwick MB. Triton and alpha-particle production in neutron-induced reactions on carbon at E_n=42.5 MeV, 62.7 MeV and 72.8 MeV. Phys Rev 1996; C53: 1309-1318.

33. Subramanian JL, Romero JL, Brady FP et al. Double differential inclusive hydrogen and helium spectra from neutron-induced reactions on carbon at 27.4, 39.7 and 60.7 MeV. Phys Rev 1983; C28: 521-528.

34. Subramanian TS, Romero JL, Brady FP et al. Double differential inclusive hydrogen and helium

spectra from neutron-induced reactions at 27.4, 39.7 and 60.7 MeV: oxygen and nitrogen. Phys Rev 1986; C34: 1580-1587.

35. Wuu C, Milavickas L. Determination of the kerma factors in tissue-equivalent plastic, C, Mg and Fe for 14.7 MeV neutrons. Med Phys 1987; 14: 1007-1014.

36. Young PG, Arthur ED, Chadwick MB. Comprehensive nuclear model calculations: introduction to the theory and use of the GNASH code. Technical Report LA-12343-MS. Los Alamos NM: Los Alamos National Laboratory, 1992.

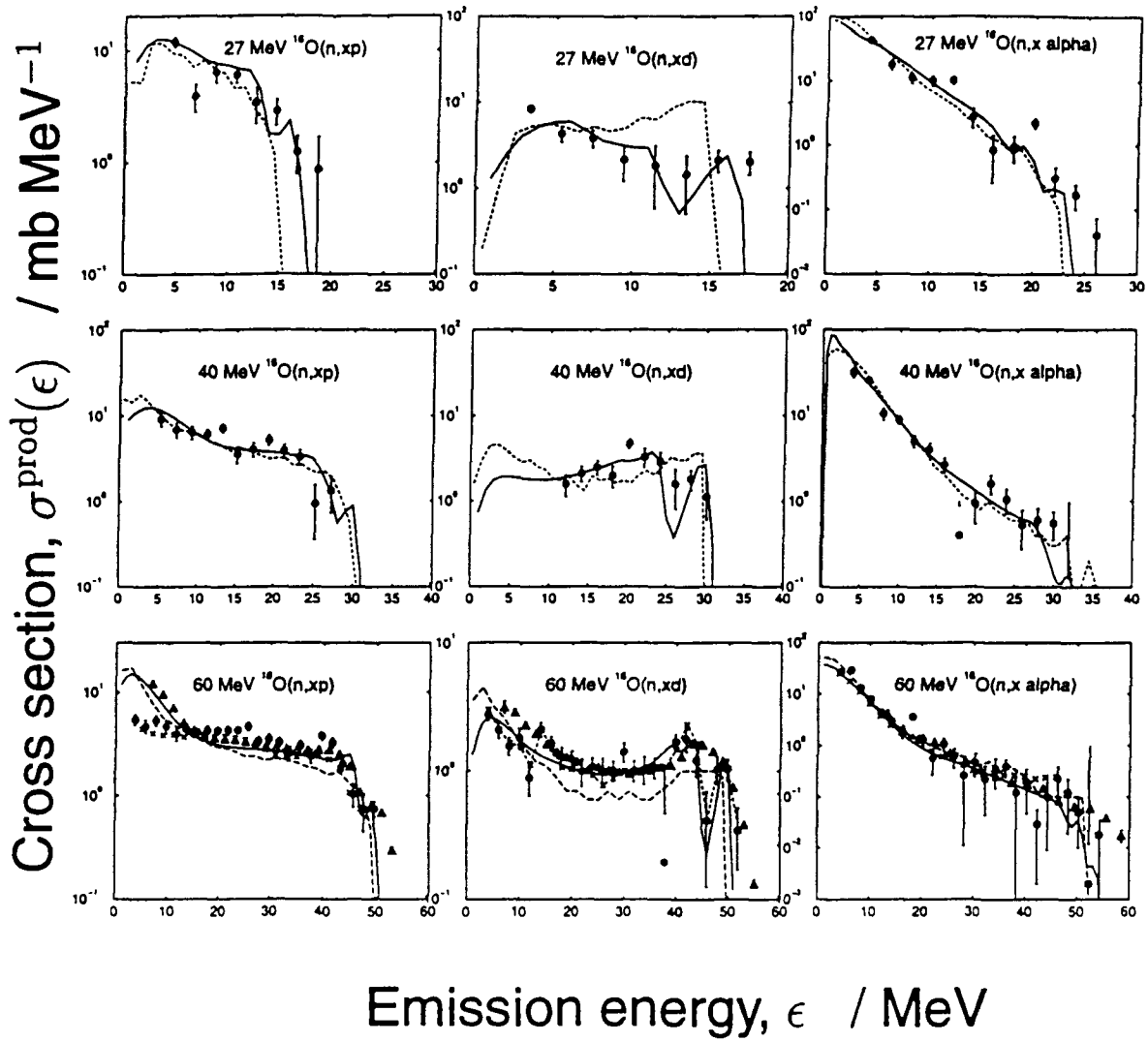


Figure 1. Evaluated angle-integrated emission spectra for n+^{16}O from GNASH [36] model calculations (full line), compared with experimental data and Brenner and Prael's calculations [5] (dashed lines) for 25, 40 and 60 MeV. Triangles represent the data of Benck et al. [2], circles the data of Subramanian et al. [34] and crosses the re-determination of Subramanian's data at 60 MeV by Chadwick et al. [9].

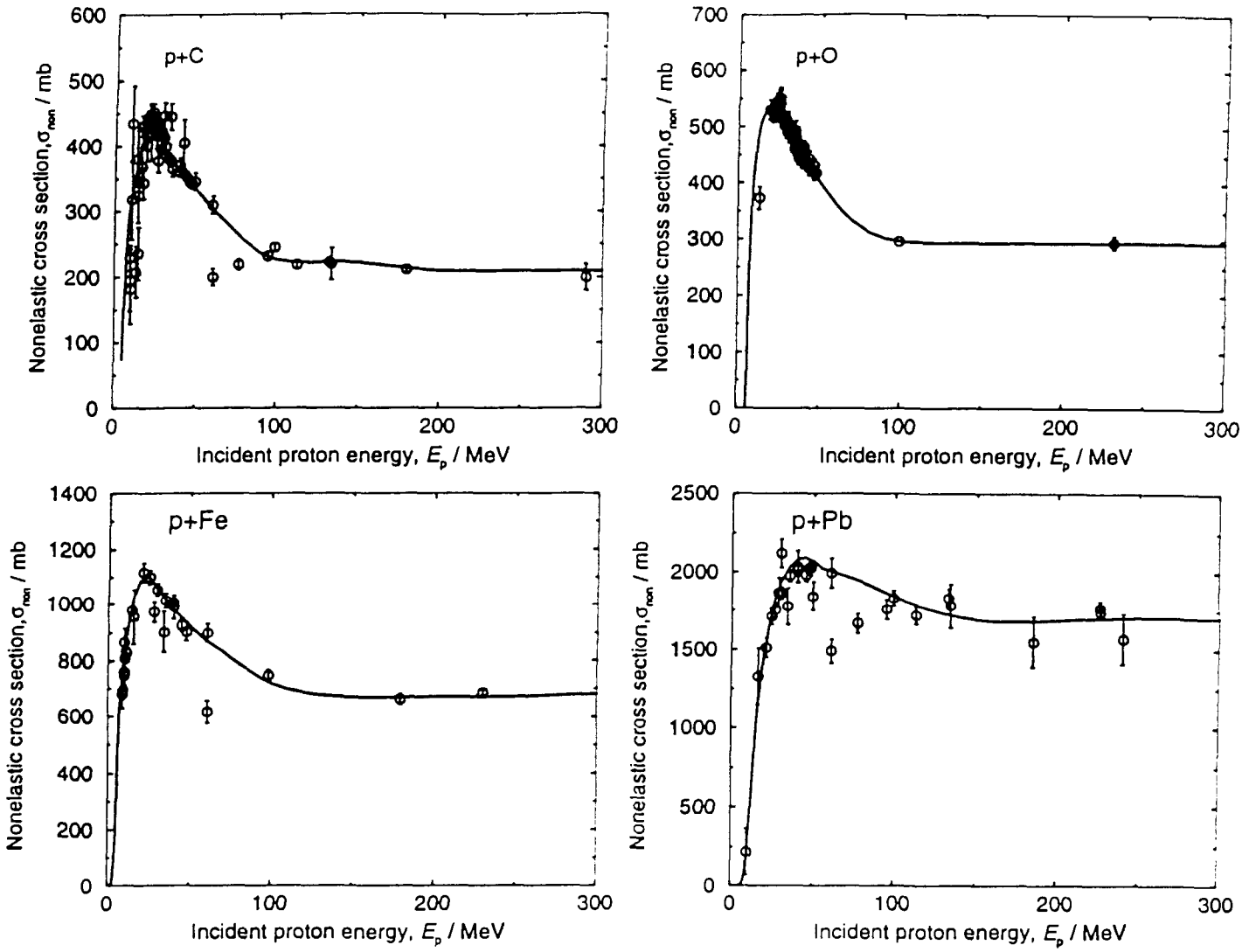


Figure 2. Evaluated total nonelastic cross sections for protons incident on C, O, Fe and Pb compared with measurements [6].

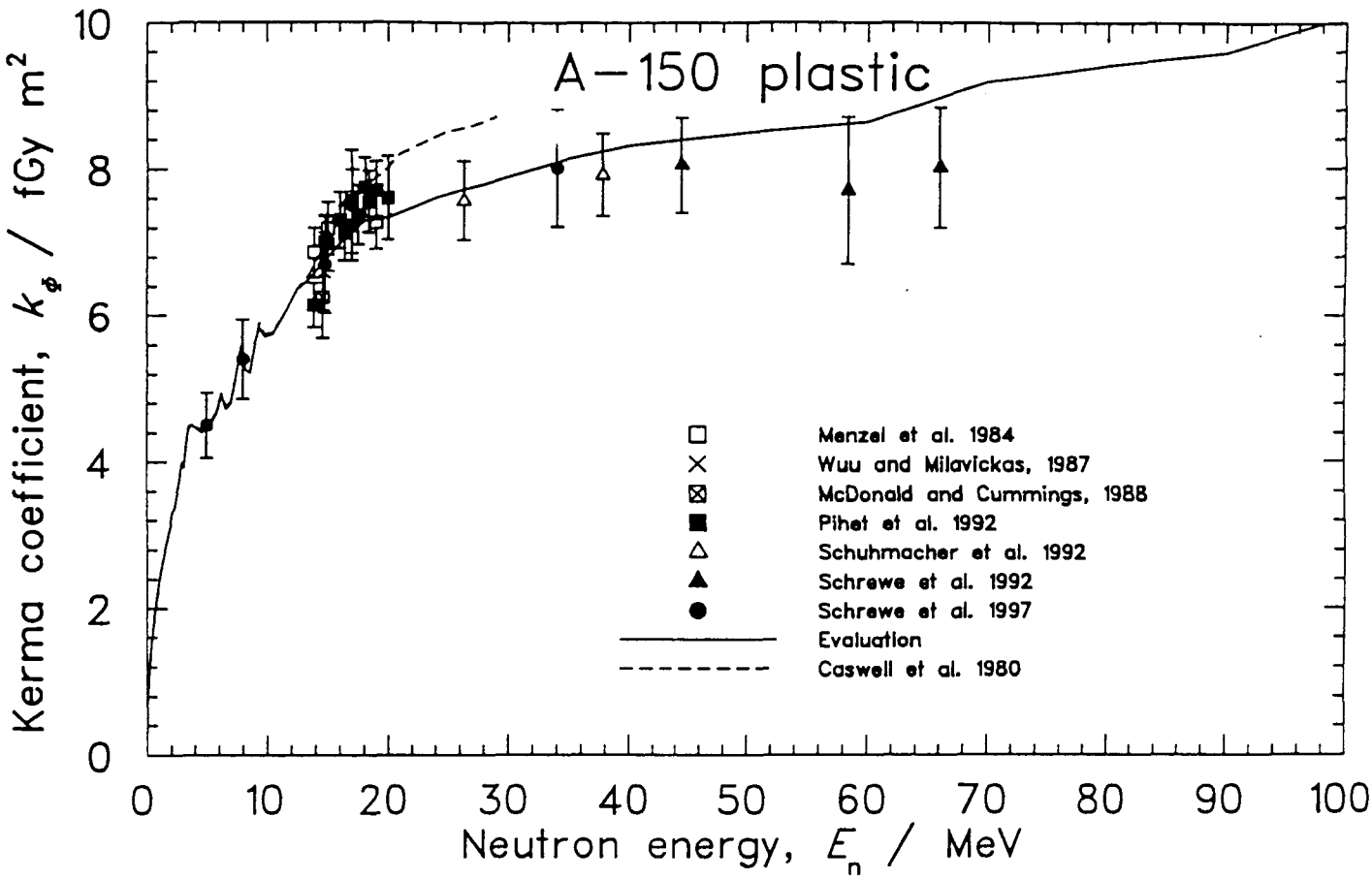


Figure 3. Total kerma coefficient for A-150 tissue-equivalent plastic, k_Φ, as a function of the energy of incident neutrons, E_n. The evaluated data (full line) are compared with an earlier evaluation (dashed line) by Caswell et al. [7] and with data (symbols) measured by Menzel et al. [23], McDonald and Cummings [20], Pihet et al. [26], Schrewe et al. [27, 28], Schuhmacher et al. [29] and Wuu and Milavickas [35].

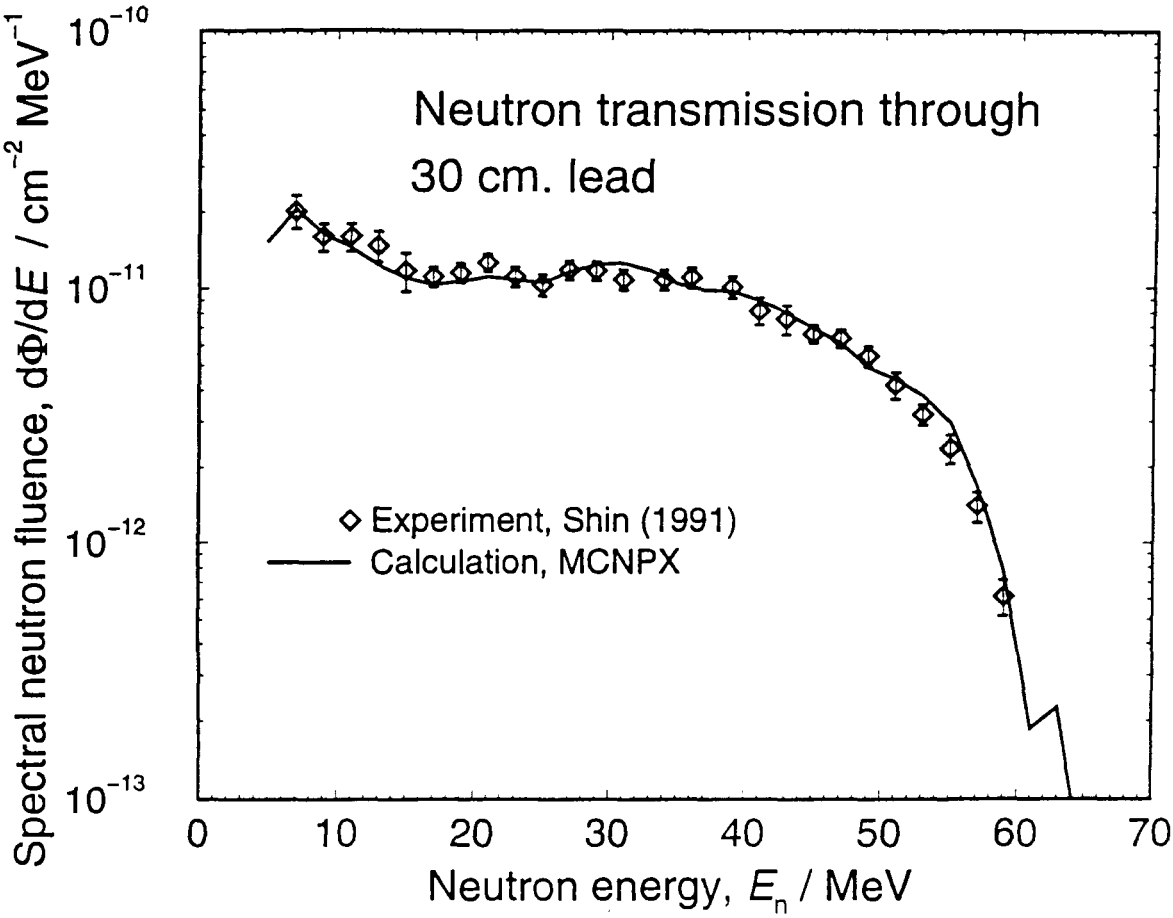


Figure 4. Comparison of measured neutron fluence through 30 cm of lead, with the results of a MCNPX [16] transport calculation.

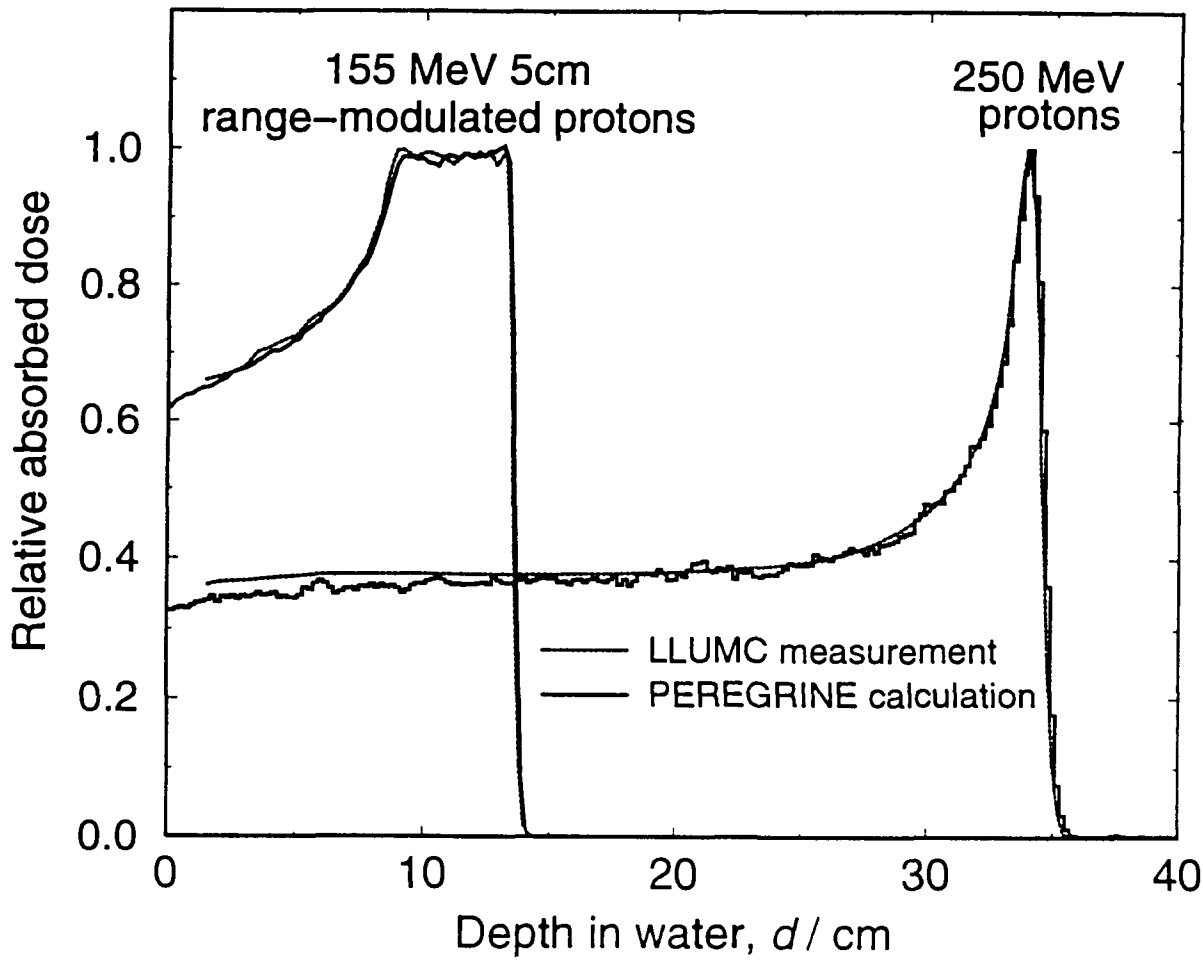


Figure 5. Comparison between measured and calculated [15] proton central axis depth dose distributions for 250 MeV protons in water, and for 5 cm range-modulated 155 MeV protons in water.

NUCLEAR DATA NEEDS IN ION BEAM ANALYSIS

I. C. VICKRIDGE

Groupe de Physique des Solides, UMR 7588 du CNRS, France

Abstract

After a presentation of the basic characteristics of Ion Beam Analytical methods, illustrated by selected examples, the strategic needs for evaluated nuclear reaction cross section data are evaluated in view of the possibilities and appropriateness of activities coordinated by the IAEA. It is suggested that a priority be placed on i) development and application of theoretical models allowing reliable interpolation amongst well-chosen existing experimental data, and ii) robust archivage and dissemination of nuclear reaction cross sections to allow access by IBA practitioners to appropriate cross sections in a format suitable for use in existing IBA spectrum simulation programs.

1 Introduction.

Ion Beam Analysis (IBA) refers to a suite of analytical techniques in which various prompt interactions between a beam of charged particles and a target are exploited in order to infer compositional and structural properties of materials to be analysed. Most commonly, beams of light particles (isotopes of hydrogen and helium) of energy ranging from about 100 keV to a few MeV are used. Less commonly, beams of other ions and energies are applied – for example in heavy ion elastic recoil analysis in which heavier incident ions (e.g. ^{28}Si or ^{128}I) are used at energies of 1-3 MeV/amu. Common to all of the techniques is the need to know the shapes and magnitudes as a function of incident energy and detection angle of the interaction cross sections that are exploited. In this document I explore the present and future needs for nuclear data for the various techniques that make up IBA.

2 IBA Techniques and definition of scope

The names of the various techniques mostly reflect the nature of the interaction that is directly exploited for analytical purposes. Table 1 lists some common acronyms (following the recommendations of [1]) and the associated interactions.

Acronym		Interaction
PIXE	Particle-Induced X-ray Emission	Characteristic X-ray emission following inner_shell ionization by the primary beam
PIGE	Particle-Induced Gamma Emission	Prompt gamma emission during ion beam irradiation
RBS	Rutherford Backscattering Spectrometry	Elastic scattering at backward angles, in which the cross section is Rutherford
NRA	Nuclear Reaction Analysis	Nuclear reaction between incident beam and nuclei in the target, producing a light charged particle
NRP or r-NRA	Nuclear Resonance Profiling or Resonant Nuclear Reaction Analysis	Exploitation of narrow nuclear resonances via scanning of the incident beam energy
ERDA or FRS	Elastic Recoil Detection Analysis or Forward Recoil Spectroscopy	Elastic recoil at forward angles, not necessarily Rutherford

In addition to these well-established techniques, there are variations that have not yet established widely accepted acronyms. The two most important are non-Rutherford elastic scattering with light particle beams – principally protons and alpha particles, and heavy-ion high-energy elastic recoil. These higher energy variants are clearly included under the 'IBA' umbrella, but it is unclear whether the lower energy methods - Low Energy Ion Scattering (LEIS) and Medium Energy Ion Scattering (MEIS) – should be considered 'IBA' methods, since they generally use beams produced by implantation-type machines rather than the MeV accelerators that are a traditional element of IBA. Furthermore, and perhaps more importantly in the present context, in these techniques, the interactions between the incident beams and the target are dominated by inelastic energy-loss processes (i.e. energy loss to target electrons) and the (screened) Coulomb interaction between the incident beam and the

target nuclei. The data needs here are therefore not 'nuclear' and so LEIS and MEIS are not considered for the purposes of the present exercise.

The PIXE technique is a mature sub-field of IBA, with its own conferences, established simulation codes, and data bases. Furthermore, since the primary interaction is between an incident particle and the core electron shells, PIXE data does not really fall within the scope of 'Nuclear Data Needs'. Therefore, the data needs for PIXE need not be considered within the framework of the present Advisory Group Meeting.

The RBS technique, in which incident alpha particles of up to a few MeV are elastically back-scattered from target nuclei, is governed by the analytical Rutherford cross-section with well-established screening corrections [2] and so may be considered as requiring no further work. This is also the case for the heavy-ion variant, in which the major requirements are for improved energy straggling and multiple scattering data.

Another technique which is on the margins of conventional IBA is Charged Particle Activation Analysis (CPAA), the charged particle analogue of Neutron Activation Analysis, in which a target is analysed via the energies and half-lives of radioactive nuclei produced by irradiation with a charged particle beam. Here, we adopt the convention that IBA is concerned with prompt radiations and so exclude CPAA.

Finally, it should be noted that common to all of the techniques cited above is the need for accurate stopping power data. This is a complex field, but in all of the IBA techniques, the stopping power is determined almost exclusively by energy loss of the incident ion to electrons in the target. The comprehensive pioneering work of Ziegler and coworkers [3] remains a central pillar of the field, used almost universally in IBA and ion implantation. For specific cases there are recent new proposed stopping power semi-empirical fits, for example for RBS [4]. Independently of whether or not one considers stopping power data to be 'nuclear data' it is my view that for IBA, collation and evaluation of stopping power values by the IAEA is a substantial task that will add little value to the existing work, and so evaluation of stopping power data fails the cost/benefit test.

We are concerned, then, with differential cross sections for NRA, non-

Rutherford elastic scattering and PIGE.

3 Evaluation of needs

3.1 NRA

In order to illustrate the cross section data needs for NRA, I consider two limiting representative case studies. In the first the analysis concerns films which are sufficiently thin that the shapes of spectral features in the NRA spectrum are determined by the analysis system. In the second, the shapes of the spectral features of the NRA spectrum also include information about the shape of the nuclear reaction cross section and the concentration depth distribution in the sample being analysed.

3.1.1 Thin film NRA analysis

One of the first and still perhaps one of the most widely used NRA methods is the determination of ^{16}O by the $^{16}O(d,p_1)^{17}O$ reaction [5]. Figure 1 shows the measured cross section of this reaction for a 150° detection angle. For analysis of films in which

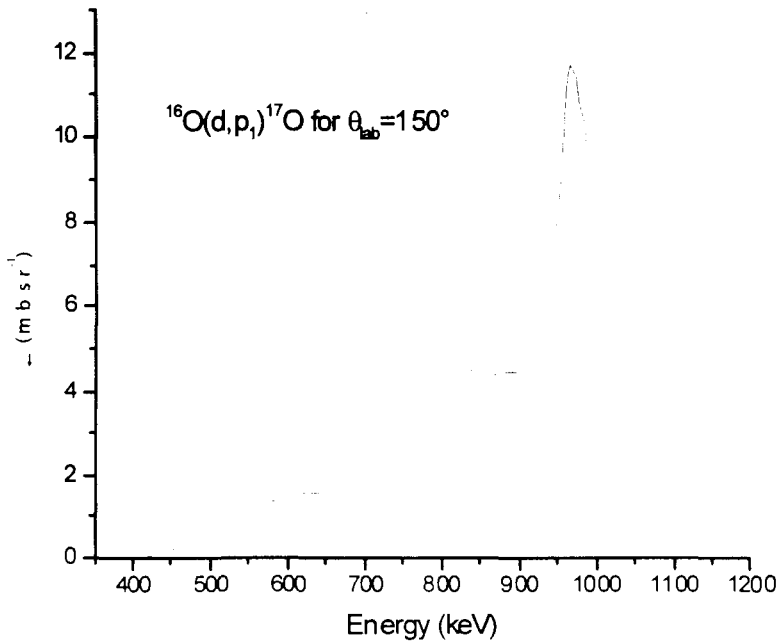


Figure 1. Differential cross section of the $^{16}O(d,p_1)^{17}O$ nuclear reaction measured at 150° in the laboratory frame.

the incident 900 keV deuteron beam loses less than about 70 keV, the plateau between about 850 keV and 920 keV ensures that the number of detected protons is strictly

proportional to the number of ^{16}O atoms in the film. Figure 2 shows the typical charged particle spectrum observed with a silicon semiconductor detector at 150° when a thin anodic oxide of tantalum is irradiated with 850 keV deuterons. A thin mylar foil is placed in front of the detector to exclude the high flux of elastically scattered deuterons, whilst letting the more energetic protons through. The number of detected protons N may be expressed by :

$$N = n\Omega\varepsilon \int_{E_{inc}}^{E_{final}} C(x)\sigma(E)dE \qquad (1)$$

where n is the number of incident deuterons, Ω the detector solid angle, ε the detector efficiency, C(x) the concentration of ^{16}O atoms in the thin film, and $\sigma(E)$ the differential cross section of $^{16}O(d,p_1)^{17}O$. Note that dx and dE are related by the

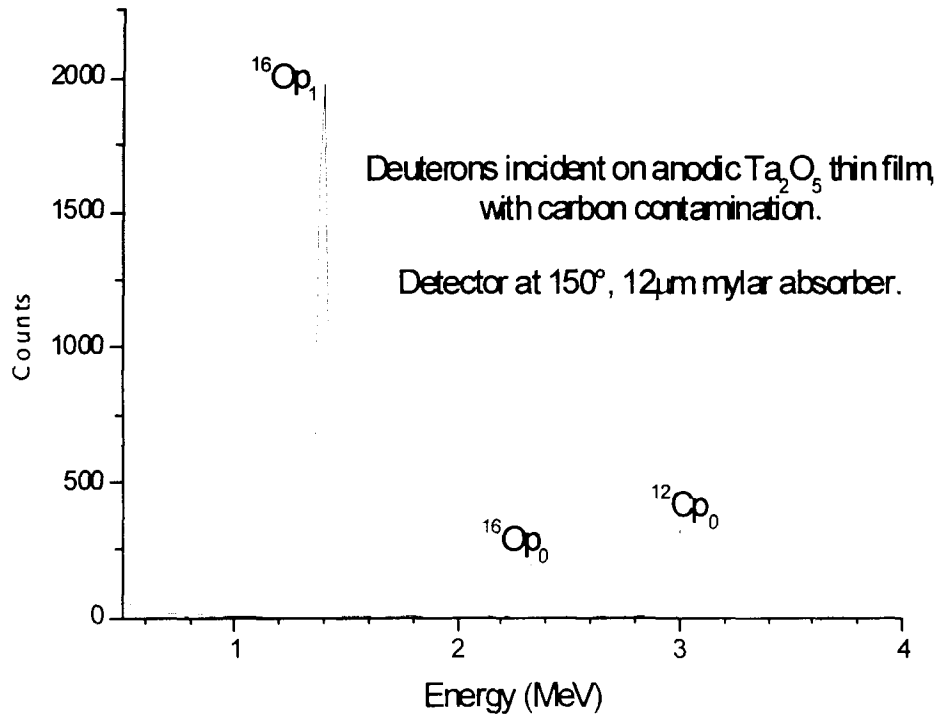


Figure 2. The detected prompt charged particle spectrum obtained from a thin Ta2O5 layer bombarded by 850 keV deuterons.

stopping power S(e)=dE/dx dE/dx. It may be shown by appropriate substitution of variables that when $\sigma(E)$ does not vary over the thickness of the film being analysed

the integral term in equation 1 is strictly independent of the stopping power and proportional to the projected number of atoms of ^{16}O per cm^2. We refer to this as the areal density of ^{16}O, which we represent by $\{^{16}O\}$. Furthermore, if the shape of $\sigma(E)$ is known then corrections can be applied even if $\sigma(E)$ is not constant. In principal all of the terms in equation 1 can be determined experimentally: the efficiency of solid state charged particle detectors is 1; the solid angle can be defined by suitable collimators (to avoid edge effects in the detector) and accurately measured; the incident beam charge can be accurately measured to determine *n*, and in principle $\sigma(E)$ can be accurately determined. Thus, in principle, NRA should be a standardless absolute method for $\{^{16}O\}$. However accurate *absolute* beam current integration is rather difficult and requiring precise knowledge of Ω for all experimental arrangements is a serious constraint. In practice it is far more convenient and probably more accurate to rely on a reference thin film containing a well-known areal density of ^{16}O. Thin anodic oxides of tantalum may be prepared in which $\{^{16}O\}$ is known to about 2% absolute accuracy [6]. In this case, after measurements of N_U and N_R with incident beam doses of n_U and n_R respectively for the unknown and reference films, $\{^{16}O\}_U$ in the film of unknown thickness is deduced from $\{^{16}O\}_R$ in the reference film by :

$$\{^{6}O\}_U = \frac{N_U n_R}{N_R n_U} \{^{6}O\}_R \qquad (2)$$

Here, the measurement is independent of the absolute value of $\sigma(E)$ – in fact the only thing we need to be sure of about $\sigma(E)$ is that it does not vary appreciably over the energy range corresponding to that lost by the incident particle in the thin film being analysed. An approximate knowledge of the absolute value is useful to guide the choice of experimental parameters to achieve reasonable measurement times, count rates and incident beam doses for the films to be analysed. It is worthwhile noting here that obtaining suitable reference targets can be problematic. The approach of Davies and coworkers [7-9] has been to determine accurate ratios of cross sections of $^{12}C(d,p)^{13}C$, $D(^3He,p)^4He$, $^{14}N(d,\alpha)^{12}C$, $^{14}N(d,p)^{15}N$, $^{15}N(d,\alpha_0)^{13}C$ and $^{15}N(p,\alpha_0)^{12}C$ to that of the $^{16}O(d,p_1)^{17}O$ reactions by using stoichiometric frozen gas targets of CO_2, NO and D_2O. This enables the reliable and robust Ta_2O_5 reference targets to be used as

a reference for NRA determinations of {D}, {^{12}C}, {^{14}N} and {^{15}N}.

The above discussion implicitly assumes that the detector solid angles employed are sufficiently small that $\sigma(E)$ may be considered constant over the angular span of the detector. This may not be the case when very large solid angles are used – for example with annular detectors in which solid angles up to near 2π can be achieved. In this case detailed knowledge of $\sigma(E,\theta)$ is required if the integral of equation 1, which would now become a double integral over both energy and angle, is to be evaluated. Reliable experimental cross section data over both the required energy and angular range is seldom available, and the number of IBA practitioners using large solid angles is very small. In my view, the huge experimental effort required to accurately fill all the gaps is probably not justified by the needs, however a more modest theoretical effort enabling reliable interpolation amongst critically evaluated measured data would certainly be very worthwhile.

In the above case study a very simple system was deliberately chosen in order to illustrate the fundamental nuclear data requirements for thin film analysis by NRA. Similar considerations apply for analysis of other isotopes in thin films – a useful reference for nuclear microanalysis of light elements may be found in [8]. In many cases, however, the thin film to be analysed may be of more complicated composition, and the substrate may also give rise to nuclear reaction products : when designing and interpreting an NRA measurement the possibility of interfering nuclear reactions needs to be considered. For example, when determining {^{16}O} in the presence of ^{14}N, the numerous proton groups from $^{14}N(d,p_{0\text{-}7})^{15}N$ reactions interfere with the $^{16}O(d,p_1)^{17}O$ protons. In this particular case analysis of ^{16}O in the presence of ^{14}N may proceed by using the $^{16}O(d,p_0)^{17}O$ group which is free of interference, but has a lower cross section. The maximum energy of a potential interfering charged particle group in the NRA spectrum may easily be calculated from tabulated reaction Q-values, however to assess the relative amplitude of the interfering groups the relevant *cross section ratios* are required. Furthermore, if the interfering group comes from a thick component of the system (most commonly the substrate) then the relevant $\sigma(E)$ is required. For most of the cases of practical importance data of sufficient accuracy for guiding experimental design is available in the literature, however such data needs to be accessible in a form easily useable by the IBA analyst.

3.1.2 Intermediate and thick film NRA analysis.

Samples of sufficient thickness that the energy loss of the incident beam, and of the outgoing nuclear reaction products from the region being investigated is larger than the energy resolution of the charged particle detection system may no longer be considered thin. In this case information about the concentration profile C(x) is also

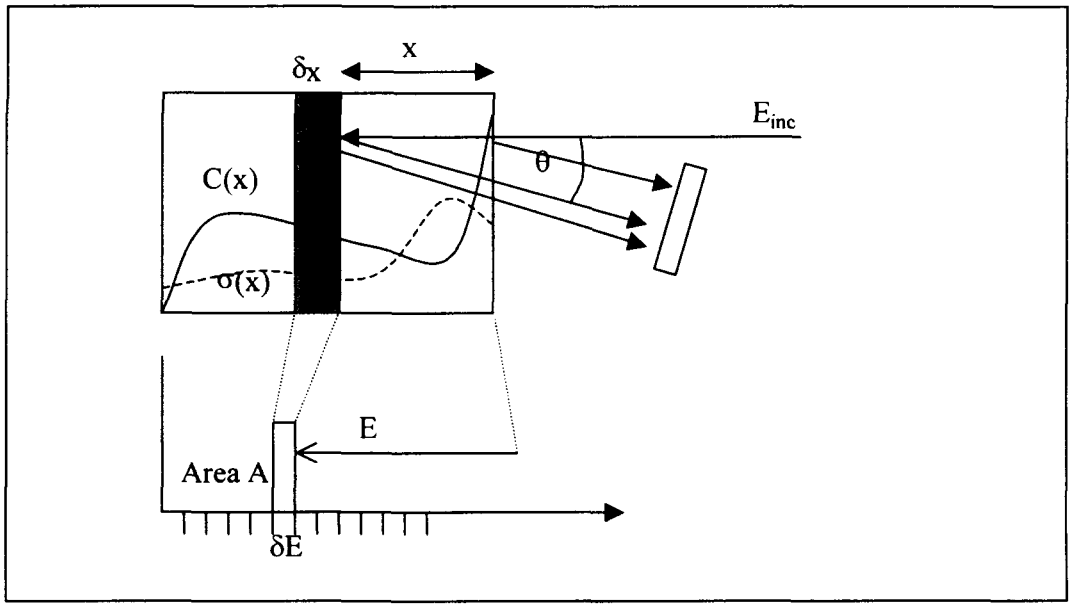


Figure 3. Definition of some variables related to NRA of thick targets.

contained in the *shape* of the charged particle spectra. A detailed mathematical description of the relationship between the shape of the spectrum and C(x), σ(E) and S(E) is beyond the scope of this report, however considering Figure 3, and assuming for simplicity that the stopping powers of the incident and outgoing particles do not vary with energy, and ignoring energy straggling and detector resolution, the most important points for our purposes are :

i) a channel of width δE_c at energy E_c in the spectrum corresponds to a slice of width δx at depth x in the sample, with Ec and δEc being inversely related to x and δx through a linear combination of the stopping powers for the incident and outgoing particles,

ii) the number of particles accumulated into that histogram bin is proportional to C(x), δx, and $\sigma(E_x)$, where E_x is the energy of the incident beam when it gets to depth x.

Since the height of the spectrum, and the conversion from the energy scale to a depth scale depend directly on the stopping powers, accurate stopping powers are obviously of primary importance for the accurate determination of concentration depth profiles by NRA. It is also clear that the more accurately the cross section is

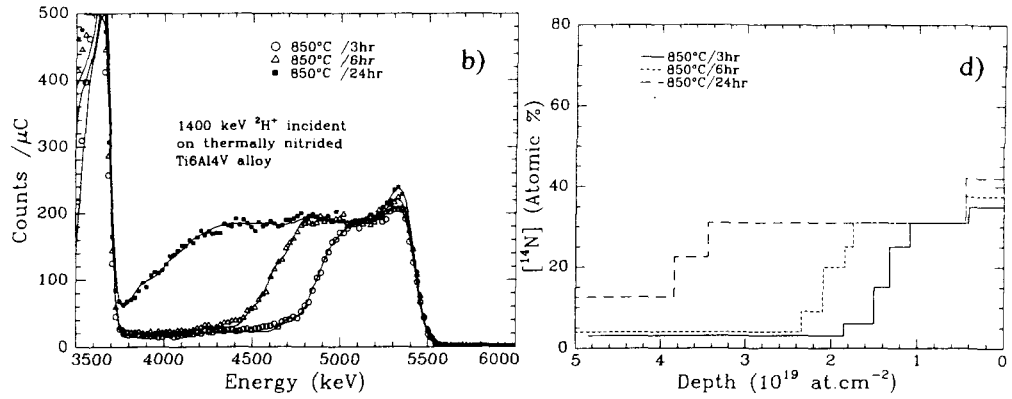


Figure 4. $^{14}N(\alpha,p)$ spectra and deduced depth profiles from thermally nitrided Ti6Al4V alloy. Taken from Reference

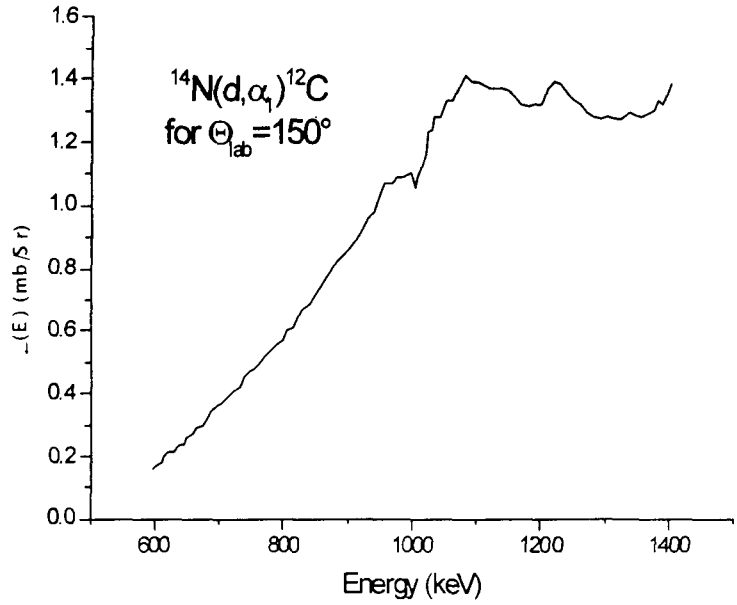


Figure 5. Differential cross section of the $^{14}N(d,\alpha_1)^{12}C$ nuclear reaction, showing the broad plateau exploited in the measurements of Figure 4.

known, the more accurately the concentration may be deduced. However, as in the case of the analysis of thin layers, accurate knowledge of the shape of the cross-section is more important than accurate absolute values, since in most cases of practical interest, the spectra are measured to within an arbitrary multiplicative factor,

and this factor is then determined by the use of a reference sample.

A typical example is given in Figure 4, which shows a portion of the deuteron-induced prompt particle spectra obtained from titanium thermally nitrided for various times in dry N_2 [11]. Figure 5 shows the cross section of the $^{14}N(d,\alpha_1)^{12}C$ reaction [12]. In this case the broad plateau in $\sigma(E)$ was deliberately chosen since in this case the $^{14}N\alpha_1$ spectral shape may be simply interpreted to first order by visual inspection. More detailed analysis requires computer synthesis of theoretical spectra corresponding to assumed forms of C(x), and iterative adjustment of C(x) until the simulated spectra satisfactorily match the measured spectra.

3.1.3 Non-Rutherford elastic scattering cross sections.

The considerations given above apply equally well to the non-Rutherford (α,α) and (p,p) cross sections. This topic is dealt with more fully, and recent theoretical and evaluation work described, in a report by A. F. Gurbich [13].

3.2 PIGE

PIGE differs from NRA and elastic scattering methods in that the outgoing particles do not suffer energy loss as they traverse the target, and their energy is almost always independent of that of the incident particle. Also, in contrast to the PIXE method in which outgoing photons of 1keV to 30keV are detected, PIGE typically exploits photons of 100keV up to 10 MeV, and so the linear attenuation coefficients are sufficiently small that absorption of the gammas between the point of production in the target and the detector can be completely ignored. Thus, all depth information is lost, and the gamma yield Y from a thick target may then be written :

$$Y = n\Omega\varepsilon \int_{x=0}^{x=R_p} C(x)\sigma(E)dx \qquad (3)$$

where R_p is the projectile range in the target and the other variables are the same as for equation (1). Assuming that C(x) is constant and equal to C and changing the integration variable to E, we may rewrite equation 3 :

$$Y = n\Omega\varepsilon C \int_{E=E_{inc}}^{E=0} \frac{\sigma(E)}{S(E)} dE \quad . \qquad (3a)$$

It has been shown that the integral in equation 3a may be approximated by

$$\int_{E=E_{inc}}^{E=0} \frac{\sigma(E)}{S(E)} dE \cong \frac{1}{S} \int_{E=E_{inc}}^{E=0} \sigma(E) dE \qquad (4)$$

where S is the stopping power at the incident ion energy E_{inc} [14], or at $E_{1/2}$, which is that incident ion energy at which the thick target gamma-yield is ½ of that for ions incident with energy E_{inc} [15]. It is now possible to write down an equation analogous to equation (2) above, in which bulk concentration of an element A, [A], assumed uniform in the region probed by the beam, is deduced from comparison of the gamma yields from a standard and an unknown sample :

$$[A]_U = [A]_S \frac{Y_U n_R}{Y_S n_U} \frac{S_R}{S_u} \qquad (5)$$

Examination of equations 3-5 allows us to make some observations. Firstly, in contrast to the case of charged particle detection, $\varepsilon\Omega$ is rather difficult to measure accurately for the case of gamma detection. In particular, the efficiency of scintillator and semiconductor detectors varies with gamma energy, and may vary with time with 'dead zones' in the detector resulting in incomplete charge detection in semiconductor detectors. Thus the use of reference samples is virtually universal in PIGE. Secondly, knowledge of the stopping powers of both the standard and the unknown samples is essential - much more important than knowing even the shapes of the cross sections. However, knowledge of the cross section shapes is useful when assessing the characteristics of the analytical method - for example determining the 'analysed depth' corresponding to the PIGE analysis. Knowledge of the thick target yield as a function of energy is useful when designing a measurement experiment, in order to calculate expected count rates. This information has been measured and published for the two most important cases of proton [16, 17] and deuteron [18] bombardment. The uncertainties the detector efficiencies and in the stopping powers for thick target PIGE mean that there is little point for IBA in obtaining highly accurate cross sections for PIGE, and as far as nuclear data needs are concerned there is little work that now needs to be done.

3.3 Narrow Nuclear Resonances.

Narrow resonances in proton-induced reactions may be used to determine

concentration depth profiles [19, 20]. The reaction product yield of a nuclear reaction is monitored as a narrow isolated resonance in $\sigma(E)$ is scanned inside the target by scanning the incident beam energy. The excitation curve Y(E) obtained is then a deformed image of C(x). The community regularly applying this rather specialized IBA technique is limited to just a few laboratories, and the pertinent characteristics (widths, energies and strengths) of the most widely used resonances are sufficiently well known for analytical application. As with PIGE and NRA, in virtually all cases reference targets are used in order to fix the vertical scale and the energy calibration of the accelerator, which is crucial for these measurements. The physics that relates C(x) toY(E) is well established and available in a PC program [21], however reliably extracting the best estimate of C(x) from Y(E) requires accurate knowledge of stopping powers, and also accurate knowledge of the energy straggling of the incident beam. It has been found that many of the resonance widths measured in the context of nuclear physics studies or for studies of stellar combustion rates in astrophysics have been overestimated, mainly because either beam energy resolution was too poor, or targets were too thick. An example is the $^{18}O(p,\alpha)^{15}N$ narrow resonance near 151 keV, which was initially reported to be 0.5 keV wide [22]. A more recent measurement showed it in fact to be less than 0.1 keV - and consequently very useful for high resolution depth profiling [23]. There is, therefore, a clear need for repeating further resonance width measurements, however given the specialized nature of the measurements and the relatively small 'client base' I suggest that this activity be left to the groups concerned rather than being undertaken with IAEA supervision.

4 Discussion

It is clear that there are two main data needs for IBA : accurate stopping powers for elemental and compound solids, and a comprehensive and accessible collection of reliable nuclear reaction cross sections for NRA. Of these, the second falls within the scope of the guidelines of this AGM, which is to identify areas in emerging applications where data improvement activities could be coordinated by the IAEA.

The IBA community itself has already undertaken some work in this area. As a workshop held as part of the Third International Conference on Chemical Analysis in Namur, Belgium in 1991 [24] a common data format for storage and dissemination of

nuclear reaction cross sections for IBA was proposed [25]. Shortly afterwards a number of cross-sections were scanned and digitized by G. Vizkelethy and made available on the Sigmabase FTP site, which later developed into the Sigmabase web site [26], which is now mirrored in Hungary [27]. More recently, a collection of the main NRA nuclear reaction cross sections has been prepared together with bibliographic details and some evaluation in the form of a PC computer program, the NRABASE [28]. The most recent IBA Handbook [29] contains printed graphs of numerous non-Rutherford cross sections and nuclear reaction cross sections. Mostly, these efforts have been those of a small number of individuals, and mostly with only passive support from their institutions.

5 Conclusions

There is a clear role for the IAEA in coordinating and supporting the evaluation of experimental nuclear reaction and non-Rutherford elastic scattering cross-sections, and development of theoretical tools that will allow confident interpolation between and even extrapolation beyond the experimental dataset. Such a programme would also be of great help to identify the most critical needs for re-measurement of cross sections. There is also a need for the nuclear reaction and elastic scattering cross sections of importance to IBA to be archived in an internationally stable database from which it should be easily available in suitable form to IBA practitioners. Substantial progress towards meeting these two most important strategic needs of the IBA community could be made with even a modest investment of resources on the part of the IAEA : it is my view that the cost-benefit analysis is favorable.

6 Acknowledgements

It is with pleasure that I thank G. Amsel (GPS, Paris, France), D. Cohen (ANSTO, Sydney, Australia) and A. F. Gurbich (IPPE, Obninsk, Russia) for most helpful discussions. I also thank here my many colleagues in the IBA and other communities, with whom I have had the privilege of learning, developing and applying IBA techniques.

7 References

[1] G. Amsel, Nucl. Instr. And Meth. B118 (1996) 52.

[2] J. L'Ecuyer, J. A. Davies and N. Matsunami, Nucl. Instr. And Meth. 160 (1979) 337.

[3] J. F. Ziegler, J. P. Biersack and U. Littmarck. The Stopping and Range of Ions in Solids. Pergamon (1985).

[4] G. Konac, S. Kalbitzer, C. Klatt, D. Niemann and R. Stoll, Nucl. Instr. And Meth. B 136-138 (1998) 159.

[5] G. Amsel and D. David. Analytical Chemistry 39(14) (1967) 1689.

[6] G. Amsel and J. A. Davies, Nucl. Instr. And Meth. 218 (1983) 177-182.

[7] J.A. Davies and P.R. Norton, Nucl. Instr. And Meth. 168 (1980) 611-615.

[8] J. A. Davies, T. E. Jackman, H. Plattner and I. Bubb, Nucl. Instr. And Meth. 218 (1983) 141-146.

[9] J. A. Sawicki, J. A. Davies and T. E. Jackman. Nucl. Instr. And Meth. B15 (1986) 530-534.

[10] G. Demortier (Ed). Microanalysis of light elements (Hydrogen to Neon) using charged particle accelerators. Proceedings of the Third International Conference on Chemical Analysis, Namur, Belgium, 8-12 July 1991, Nucl. And Instr. Meth B 66 (1992).

[11] I. C. Vickridge, W. J. Trompetter, I. W. M. Brown and J. E. Patterson. Nucl. Instr. And Meth. B99 (1995) 454.

[12] G.Amsel and D.David, Revue de Physique Appliquee, 4(1969), 383.

[13] A. F. Gurbich, this meeting.

[14] M. J. Kenny, J. R. Bird and E. Clayton. Nucl. Instr. And Meth. 168 (1980) 115.

[15] G. Deconninck and G. Demortier, J. Radioanal. Chem. 12 (1872) 727.

[16] A. Antila, R. Hanninen and J. Räisänen, J. Radioanal. Chem. 62(&-2) (1981) 293-306.

[17] Á.Z. Kiss, E. Koltay, B. Nyakó, E. Somorjai, A. Antila and J. Räisänen, J. Radioanal. Nucl. Chem. Articles 89 (1985), 123.

[18] Á.Z. Kiss, I. Biron, T. Calligaro and J. Salomon. Nucl. Instr. And Meth B. 85 (1994) 118-122.

[19] G. Amsel and D. Samuel, J. Phys. Chem. Solids 23 (1962) 1707.

[20] B. Maurel, G. Amsel and J. P. Nadai, Nucl. Instr. And Meth. 197 (1982) 1.

[21] I. C. Vickridge and G. Amsel. Nucl. Instr. And Meth. B45 (1990) 6.

[22] H. Lorenz-Wirba, P. Schmalbrock, H. P. Trautwetter, M. Weischer, C. Rolfs and W. S. Rodney, Nucl. Phys. A313 (1979) 356.

[23] G. Battistig, G. Amsel, E. d'Artemare and I. Vickridge, Nucl. Instr. And Meth. B61 (1991) 369-376.

[24] I. C. Vickridge, Nucl. Instr. And Meth. B66 (1992) 303.

[25] I. C. Vickridge, *Proposed ASCII Format for Communication of Reaction Cross Sections in the IBA Community*, (1991), DSIR Physical Sciences Report 33, Geological and Nuclear Sciences Ltd, Lower Hutt, New Zealand.

[26] http://ibaserver.physics.isu.edu/sigmabase/

[27] http://www.mfa.kfki.hu/sigmabase/

[28] A. F. Gurbich, A. V. Ignatyuk, *in* G. Reffo, A. Ventura and C. Grandy (Eds.), Nuclear Data for Science and Technology, Conf. Proc. Vol. 59, SIF, Bologna, 1997, p1740.

EVALUATION AND CALCULATION OF CHARGED PARTICLE NUCLEAR DATA FOR ION BEAM MATERIALS ANALYSIS

A.F. GURBICH

Institute of Physics and Power Engineering, Obninsk, Russia

Abstract

The needs of the IBA community in charged particle nuclear data are briefly reviewed. The recent results obtained in the evaluation of the cross sections for IBA are presented and the problems which should be resolved in order to establish a reliable basis for the IBA analytical work are discussed. It is shown that evaluating cross sections by combining a large number of different data sets in the framework of the theoretical model enables excitation functions for analytical purposes to be calculated for any scattering angle, with reliability exceeding that of any individual measurement. The ways to provide the IBA community with a reliable source of the nuclear data are outlined.

1 Introduction

The Ion Beam Analysis (IBA) techniques [1] have become more and more widespread. At present hundreds of laboratories both in industrial and developing countries employ these techniques for materials characterization. Distinct of activation analysis based on the induced radioactivity IBA uses the prompt radiation accompanying a nuclear interaction for determining elemental concentrations. The information about the composition and the structure of the sample is deduced from the spectrum of the accelerated ions undergoing an interaction within the sample. The linear dependence of the registered signal on the atomic concentration and on the cross section results in obvious constraints on the required accuracy of the employed data. It is evident that the concentration cannot be determined with the accuracy exceeded that of the cross section. There are a number of different IBA methods based on the registration of elastically scattered particles or the products of nuclear reactions and a reliable source of cross section data is needed for all of them except for Rutherford backscattering for which the cross section can be calculated according to the known formula.

Most of the IBA work to date has been in the detection of light elements for which charged particle induced reactions are particularly suitable. Although the officially accepted list of required nuclear data for IBA does not exists it is a safe assumption that such a list should comprise first of all (though not only) the differential cross sections for proton and ^{4}He non-Rutherford elastic scattering and nuclear reactions for p, d, and ^{3}He with energy $E < 5.0$ MeV interacting with $A \leq 40$ nuclei. In view of the number of possible exit channels it appears that the number of the required data is tremendous. An attempt to prepare a detailed inventory of all reactions of interest or potential interest to IBA has been reported in Ref.[2]. Although the number of the required data is great some cross sections of a primary importance for IBA can

be indicated. From the author's point of view they are non-Rutherford proton and ^{4}He elastic scattering cross sections for A<40 nuclei and deuteron induced reactions data for carbon, nitrogen, and oxygen. To provide the charged particles cross sections for IBA is the task which resembles the problem of nuclear data for other applications in all respects save one. Differential cross sections rather than total ones are needed for IBA.

Whatever actual needs the requirements of analytical work favour the use of those only reactions for which adequate information already exists. Many differential nuclear reaction cross sections were measured in the fifties and sixties. Most of those data are available from the literature but mainly as graphs. Besides, the energy interval and angles at which measurements were performed are often out of range normally used in IBA. Therefore, although a large amount of cross section data seems to be available, most of it is unsuitable for IBA. Because of lack of required data many research groups doing IBA analytical work started to measure cross sections for their own use every time when an appropriate cross section was not found. The Internet site SigmaBase was developed for the exchange of measured data. However, all these data should be evaluated prior to their widespread use. The reasons are as follows. The analysis of the information compiled in NRABASE data base [3] revealed numerous discrepancies in the reported cross sections values which are far beyond quoted experimental errors. In addition, because of cross sections dependence on a scattering angle the available data are valid only in the case of a scattering geometry very close to the geometry used in the cross sections measurements. Due to historical reasons charged particles detectors are fixed in different laboratories at different angles in the interval approximately from 130° to 180°. Meanwhile, the cross section may strong depend on a scattering angle. Though in some cases measured data were parametrized using empirical expressions it is essential that the parametrization should represent cross sections not only at measured energies and angles but also provide a reliable extrapolation over all the range of interest. So a theoretical evaluation of the cross sections grounded on appropriate physics seems to be the only way to resolve the problem of nuclear data for IBA. In order to meet the needs of the IBA community the evaluation of some most urgent data was recently made. The aim of the present report is to present the obtained results and to discuss the problems which should be resolved in order to establish a reliable basis for the IBA analytical work.

2 Evaluation

The evaluation procedure consisted of the following generally established steps. Firstly, a search of the literature and of nuclear data bases was made to compile relevant experimental data. Data published only as graphs were digitized. Then, data from different sources were compared and the reported experimental conditions and errors assigned to the data were examined. Based on this, the apparently reliable experimental points were critically selected.

Free parameters of the theoretical model, which involve appropriate physics for the given scattering process, were then fitted in the limits of reasonable physical constraints. The model calculations were finally used to produce the optimal theoretical differential cross section, in a statistical sense. Thus, the data measured under different experimental conditions at different scattering angles became incorporated into the framework of the unified theoretical approach. The final stage was to compare the calculated curves to the experimental points used for the model and to analyze the revealed discrepancies.

2.1 Proton elastic scattering

It has been generally established that proton elastic backscattering spectrometry is a valuable analytical tool. Difficulties arise however in use of this technique since the cross sections for proton scattering are as a rule non-Rutherford and they cannot be calculated from a simple analytical formula. There is every indication to believe that direct potential scattering and resonant scattering are the mechanisms which should completely account for the (p,p) elastic cross section at low proton energies. The interaction between the proton and a nucleus can be represented, in the center of mass of the equivalent single particle problem, by an optical model potential. The optical model potential of the standard form was used except for the real central part which was modified by adding a surface term to the Saxon-Woods potential to take account of channel coupling [4]. A real central part of the modified potential is given in the usual notation by

$$U_R(r) = -V_R f(r, r_R, a_R) + 4a_S V_S \frac{d}{dr} f(r, r_S, a_S),$$

where the subscript S denotes parameters of the surface term, $f(r,r_x,a_x)$ is a Saxon-Woods form factor $f(r, r_x, a_x) = [1 + \exp(r - R_x)/a_x]^{-1}$ with a half value radius $R_x = r_x A^{1/3}$ and a diffuseness parameter a_x, A is a target mass number, V_R and V_S are the depths of the real central volume and surface potentials respectively.

Resonance scattering was taken into account by addition of Breit-Wigner resonanses to the optical amplitude. The differential cross section can be written in terms of amplitudes $f_l^{\pm}$ which are related to the S-matrix elements by

$$f_l^{\pm} = (S_l^{\pm} - 1)/2i$$

For spin zero nuclei the diagonal elements of the scattering matrix were assumed to be of the form

$$S_l^{\pm} = \exp(2i\lambda_l^{\pm}) \left[\exp(-2\mu_l^{\pm}) + \exp(2i\phi_p) \frac{i\Gamma_p}{E_0 - E - \frac{1}{2}i\Gamma} \right],$$

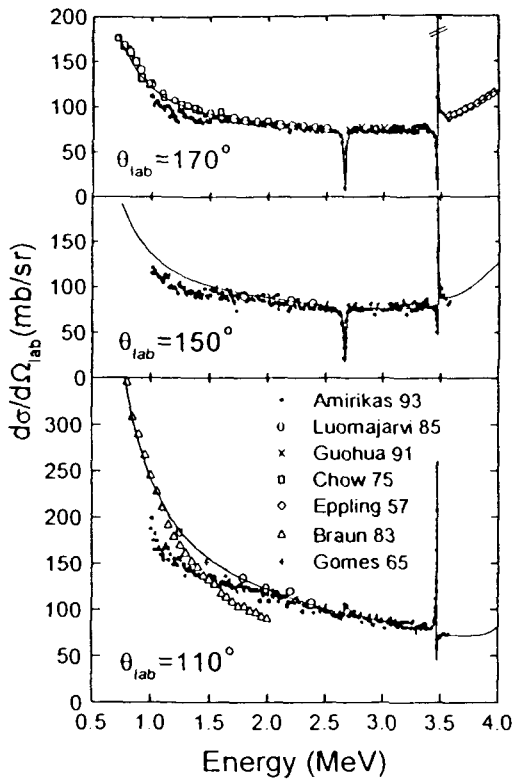


Fig. 1. The evaluated differential cross sections and the available experimental data at 110°, 150°, 170° for proton elastic scattering from oxygen.

where $\lambda_l^{\pm} + i\mu_l^{\pm}$ is the off-resonance nuclear phase shift describing the elastic scattering of protons of energy E. The quantities E_0, Γ and Γ_p are the energy, total width and partial elastic width, respectively. The subscript l is the relative angular momentum of the proton and the target in units of $\hbar$. The plus sign refers to the case when $J=l+1/2$ and the minus sign to the case when $J=l-1/2$. The quantity ϕ_p is a resonance phase shift. The off-resonance scattering matrix elements are those determined in the framework of the optical model. The optical model computer program SCAT2 [5] has been modified according to the above equations to take into account resonance scattering and the surface term in the real potential. An optimization procedure which used the modified code as subroutine has been developed for the model parameters search. Alternatively the R-matrix theory can be applied to reproduce the cross sections in the field of the IBA interests [6].

The results obtained for proton elastic scattering from oxygen [7] are presented in Fig. 1. The evaluation was also made for carbon [8] and silicon [9]. In the case of carbon the discrepancy between the evaluated cross sections and the results of the new measurement [10] was analyzed [11] and the experimental data were then corrected [12]. For silicon new measurement was undertaken in order to investigate a discrepancy between theoretical and experimental data. The obtained experimental results appeared to be in agreement with the evaluated cross sections [13]. In principle there is no problem to extend the work made so far on the whole range of the nuclei interested for IBA.

2.2 Elastic scattering of ^{4}He ions

The differential cross sections for elastic backscattering of ^{4}He ions from light nuclei are among the most important data for IBA. The utilization of ^{4}He beams with energies at which the elastic scattering cross section for light elements is conditioned by nuclear rather than electrostatic interaction has become very common over the past years. A dramatic enhancement of the cross section which becomes comparable with the Rutherford one for the heaviest nuclei is observed for low Z elements at energies ≥2 MeV. At these energies the excitation functions for elastic scattering of ^{4}He from light nuclei have as a rule both relatively smooth intervals

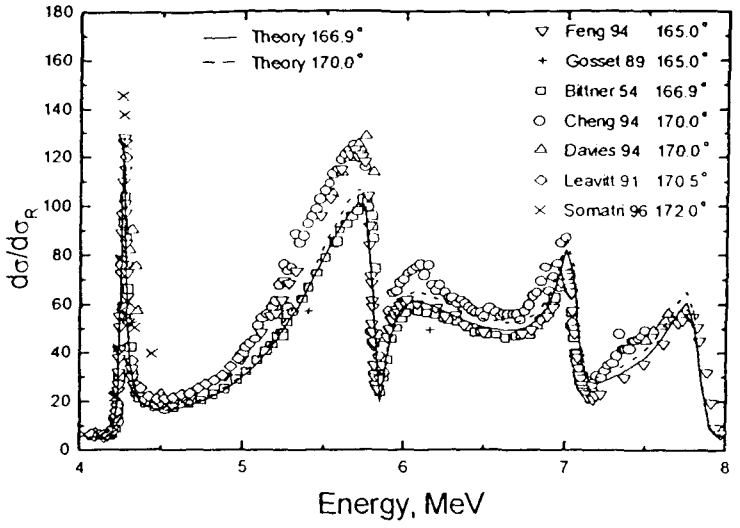


Fig. 2. The available experimental data and the evaluated excitation function for ^{4}He elastic scattering from carbon in the energy range from 4.0 to 8.0 MeV.

convenient for elastic backscattering analysis and strong resonances suitable for resonance profiling. In order to take advantage of these remarkable features the precise knowledge of the non-Rutherford cross sections over a large energy region is required.

In the course of the model adjustment it was found that a different depth V of the nucleus potential well is required in order to fit single particle resonances of different angular momentum l. To reproduce simultaneously all the data the depth V was split in respect of partial waves. The physical reasons for the use of the l-split potential in the case of scattering of complex particles can be found elsewhere [14]. The results of the evaluation of the differential cross sections for ^{12}C(^{4}He,^{4}He)^{12}C scattering [15] are shown in Fig. 2. The work to evaluate cross sections for ^{4}He elastic scattering from oxygen and nitrogen is now under way.

2.3 Deuteron induced reactions

For the reactions induced by low energy deuterons at light nuclei it was assumed that the main contribution to the cross section of the process is given by the following three mechanisms: direct stripping, resonant mechanism and in some cases a compound nucleus mechanism. It was accepted, that the complete amplitude T of process is $T=D+R$, where D is the amplitude of the direct process of stripping, which was calculated within the framework of a method of deformed waves without the account of effects of a recoil nucleus, and R-is the amplitude of resonant process, calculated in frameworks of a single level approximation. The compound mechanism contribution if any is incoherent and it may be simply added. Complete and partial width of formation

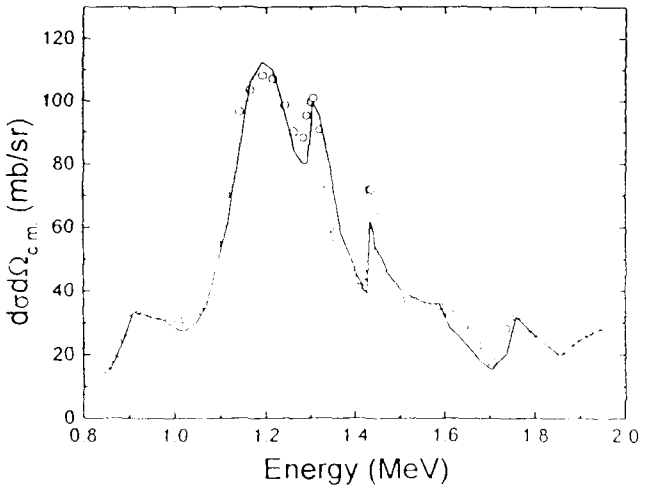


Fig. 3. The results of ^{12}C(d,p$_0$)^{13}C differential cross section calculations for 165°. Points – experimental data from Ref. [16].

and disintegration of resonances in the system, which are necessary in order to calculate the amplitude of R, were defined by fitting the model predictions to the available experimental cross sections of elastic deuteron scattering and (d,p)-reaction. The satisfactory description of the experimental data for $^{12}C(d,p_0)^{13}C$ reaction was obtained (see Fig.3). However, for a reliable description of a whole set of (d,p)-reaction data a development of the model in several directions is required.

First, it is necessary more strictly to determine a role of a compound nucleus mechanism in (d,p)-reaction in a wide kinematics range. In calculations performed within the framework of Hauser-Feshbach-Moldauer model the contribution of the compound mechanism at backward scattering angles is about of 50 % of total cross section. However, as appear, the contribution of this mechanism is essentially overestimated for the several reasons: (i) owing to low binding energy of a deuteron and its electric charge distribution asymmetry; (ii) due to the fact that since light nuclei have low number of particles and the quantum mechanics forbidden rules are strong for these nuclei they have low internal degrees of freedom and consequently the equilibrium processes in the particle system of this type are essentially suppressed.

Secondly, there are problems in the description of a direct component of an interaction. For the DWBA calculations of reactions with light fragments it is necessary to take into account a final radius of interaction and the effects of a multiple projectile-target exchange of nucleons.

Thirdly, at a collision of deuterons with nuclei in addition to the compound nucleus mechanism and the direct (d,p) stripping reactions also direct exchange processes of knock-out and heavy stripping are possible.

In - fourth, it is necessary correctly to evaluate a role of a resonant mechanism in case of strongly overlapped resonances. The problem of taking into account the close lying resonances interference is far beyond the limits of the simplified approach of single level approximation employed in the performed calculations. Actually the interaction of disintegrating states should be taken into account. To choose that or other physical concept for the analysis of the area of a spectrum, where the levels are strongly overlapped, it is necessary to resolve a number of important problems, requiring further research.

3 Calculations

Once the differential cross section for a given nucleus is evaluated the required excitation function for analytical purposes may be calculated for any scattering angle. It is worth noting that these data are needed not only for the use in spectra processing programs but also in order to choose the optimal experimental conditions. For instance, it is seen from Fig.4 that only in the vicinity of 100° is the excitation function for $^{28}Si(p,p_0)^{28}Si$ devoid of strong resonance features, thus providing a favorable condition for routine backscattering analysis. The evaluated excitation functions for ^4He elastic scattering from carbon calculated in the virtually important

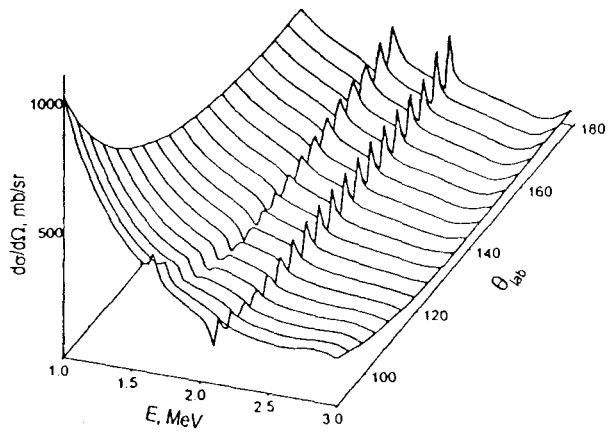


Fig. 4. The calculated differential cross sections for proton elastic scattering from silicon.

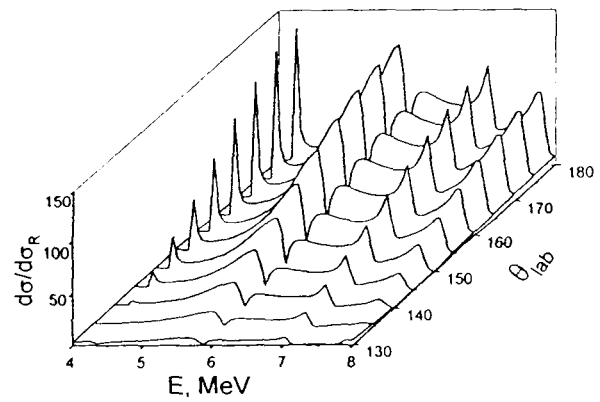


Fig. 5. The calculated differential cross sections for ^{4}He elastic scattering from carbon.

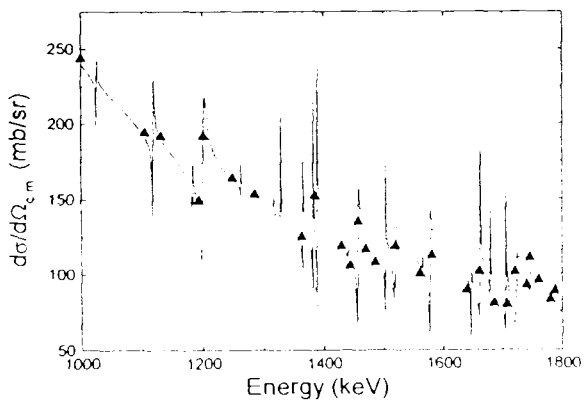


Fig. 6. The ^{27}Al(p,p$_0$) differential cross sections at 170°. The solid line is the *R*-matrix calculations with the parameters taken from Ref.[17]. Experimental points are from Ref.[18].

angular interval from 130° to 180° with step of 5° in the energy range of 4.0 – 8.0 MeV are presented in Fig. 5. As is seen from Fig. 5 the significant enhancement of the cross section is observed exclusively at the very back scattering angles. The cross section at these angles has a strong angular dependence that should be taken into account while designing an experiment.

In some papers measured data were closely reproduced by the theoretical fit. If there is enough information in the paper as for details of calculations and the parameters used then it is possible to obtain the excitation function suitable for IBA solely by calculations. An example is shown in Fig. 6. High resolution proton resonance measurement for ^{27}Al(p,p$_0$) scattering followed by *R*-matrix theory fit was reported in Ref. [17]. The excitation function presented in Fig. 6 by solid line was retrieved by means of the similar calculations. The experimental data for ^{27}Al(p,p$_0$) were obtained later in Ref. [18] devoted to the cross section measurements especially for the use in IBA. These data are also shown in Fig. 6. It is seen that in the sparse points measurements [18] the excitation function is sporadically influenced by resonances whereas the fine structure is completely missed. It was proved by calculations that this fine structure is unfortunately important in order to adequately simulate the spectrum obtained in the IBA experiment. Thus the calculated excitation function is superior in this case.

In order to provide the IBA scientist with a tool for computing the required excitation

functions a software ("SigmaCalc") is under development. The SigmaCalc is based on the already published and some new results of the data evaluation. A user friendly environment enables the IBA scientist having no expertise in nuclear physics to perform the calculations of the required smooth curves $d\sigma(E)/d\Omega$ presented in optional units. The excitation function for non-Rutherford proton scattering for carbon, nitrogen, oxygen, aluminum, silicon, and sulphur and for ^4He scattering from carbon and oxygen can be at the moment calculated by the SigmaCalc for any scattering angle in the energy range suitable for IBA.

4 Conclusion

It has been clearly shown that evaluating cross sections by combining a large number of different data sets in the framework of the theoretical model enables excitation functions for analytical purposes to be calculated for any scattering angle, with reliability exceeding that of any individual measurement. Low energy nuclear physics is regarded nowadays as a sufficiently studied field. Reaction mechanisms are known and appropriate models have been developed. However, further development of the models is still needed. Though theory is unable to provide the cross section *a priory* prediction, a particular cross section can be as a rule reliably represented by adjusting model parameters. In some important for IBA cases the reaction mechanisms are in general known but no code which provide necessary calculations is available. So some work is still needed in this field.

With SigmaCalc software being released the problem of non-Rutherford proton elastic scattering cross sections for IBA would be resolved. However this work is made without any support and so may take a lot of time. Some kind of cooperation in this project could help to finish it in the nearest future. The problem of deuteron induced reactions seems to be another challenging subject for coordinated efforts both in experimental and theoretical fields. The needs of the IBA community in these data are without doubt, the ways how to resolve the problem are clear, the time needed for this is reasonable. The benefits will be brought by this work both to the IBA community because of obtaining a firm basis for analytical work and to nuclear physicists due to the opportunity to implement new investigation on the still interesting subject.

Acknowledgments

The author would like to thank Dr. N.N.Titarenko for his contribution to the section 2.3 and for useful discussions.

References

[1] J.R. Tesmer, M.Nastasi (Eds.), Handbook of Modern Ion Beam Materials Analysis, MRS,

Pittsburg, PA, 1995.

[2] J.P.F. Selschop, S.H. Connel, Nucl. Instr. and Meth., B 44 (1994) 1.

[3] A.F. Gurbich, A.V. Ignatyuk, in: G. Reffo, A. Ventura, C. Grandy (Eds.), Nuclear Data for Science and Technology, Conf. Proc., vol. 59, SIF, Bologna, 1997, p. 1740.

[4] B. Gyarmati, R.G Lovas, T. Vertse, P.E. Hodgson, J. Phys. G 7 (1981) L209.

[5] O. Bersillon, Centre d'Etudes de Bruyeres-le-Chatel Note CEA-N-2227 (1981).

[6] E. Berthoumieux, B. Berthier, C. Moreau, J.P. Gallien, A.C. Raoux, Nucl. Instr. and Meth. B 136-138 (1998) 55.

[7] A.F. Gurbich, Nucl. Instr. and Meth. B 129 (1997) 311.

[8] A.F. Gurbich, Nucl. Instr. and Meth. B 136-138 (1998) 60.

[9] A.F. Gurbich, Nucl. Instr. and Meth. B 145 (1998) 578.

[10] S. Mazzoni, M. Chiari, L. Giuntini, P.A. Mando, N. Taccetti, Nucl. Instr. and Meth. B 136-138 (1998) 86.

[11] A.F. Gurbich, Nucl. Instr. and Meth. B 152 (1999) 403.

[12] S. Mazzoni, M. Chiari, L. Giuntini, P.A. Mando, N. Taccetti, Nucl. Instr. and Meth. B 159 (1999) 191.

[13] M.J.F. Healy, A.F. Gurbich, Nucl. Instr. and Meth. B 161-163 (2000) 136.

[14] N.S. Zelenskaya, I.B. Teplov, Exchange Processes in Nuclear Reactions, MSU, 1985 (in Russian).

[15] A.F. Gurbich, Nucl. Instr. and Meth. B 161-163 (2000) 125.

[16] E. Kashy, R.R. Perry, J.R. Risser, Phys. Rev. 117 (1960) 1289.

[17] R.O. Nelson, E.G. Bilpuch, C.R. Westerfeldt, G.E. Mitchell, Phys. Rev. C 29 (1984) 1656.

[18] E. Rauhala, Nucl. Instr. and Meth. B 40/41 (1989) 780.

NEUTRON DATA NEEDS IN ASTROPHYSICS

F. KÄPPELER

Forschungszentrum Karlsruhe, Institut für Kernphysik
Postfach 3640, D-76021 Karlsruhe, Germany

Abstract

Neutron cross sections are of relevance for a variety of nucleosynthesis scenarios, from the Big Bang, during the He burning phase of stellar evolution up to supernova explosions. Following a brief discussion of the existing data base this contribution is focussed on the remaining requests for a comprehensive description of neutron capture nucleosynthesis.

1 Introduction

Practically all elements heavier than the Fe/Co/Ni group have been produced in neutron reactions, either in the slow neutron capture process (s process) that occurs during stellar He burning or in the rapid (r) process which is associated (presumably) with supernova explosions. Both processes are distinguished by their characteristic time scales compared to typical β-decay half-lives. An additional small abundance contribution comes from the p process in supernovae, which accounts for the rare isotopes on the proton rich side of the stability valley as a result of explosive Ne/O burning.

The s process is characterized by temperatures of several hundred million degrees corresponding to the conditions of the related He burning scenarios. The relatively low neutron densities of $\approx 10^8$ cm^{-3} imply neutron capture times between a few months and a few years, slow compared to the average β-decays. Accordingly, the reaction path follows the stability valley and involves mostly the stable isotopes in the mass region between H and Bi. Accordingly, a comprehensive data base has been established by now, based on a large number of experimental results. These data allow quantitative studies of the observed s-process abundances as well as sensitive tests of currently proposed stellar models. The remaining gaps and deficiencies of this data base are discussed in Sec. 3.

In the Big Bang, neutron reactions occur under a similar temperature regime though at much higher neutron densities. Therefore, the cross sections for describing the origin of the lightest isotopes up to ^{7}Li are required in the same energy window as for the s process. (For cross section requests see contribution by M.S. Smith).

In contrast to the s process, explosive nucleosynthesis implies an extended reaction network off the stability valley, comprising thousands of reactions between unstable nuclei. Neutron reactions are mostly important during the rapid decline of temperature and density at the end of the explosive episodes, i.e., during freeze-out of the respective abundance distributions. Since the freeze-out occurs before the β-decay chains have reached the stable nuclei, such calculations have to rely on theoretical reaction rates obtained by statistical model approaches. These models, however, need to be tested and improved by comparison with experimental data.

2 Stellar Cross Sections

In astrophysics, experimental cross section data have to be converted into proper averages over the thermal velocity distribution in the stellar plasma. The stellar rate, $\lambda_{j,k}$, for reactions between two particles j, k (e.g. proton, neutron, or alpha capture processes on heavier mass particles) can be derived by the convolution between the energy dependent reaction cross section $\sigma(E)$ and the Maxwell-Boltzmann distribution of the interacting particles,

$$\lambda_{j,k} = \left[\frac{8}{\mu\pi}\right]^{1/2} (kT)^{-3/2} \int_0^\infty E\sigma_{j,k}(E) \cdot exp(-E/kT)dE, \tag{1}$$

which depends on particle energy E, stellar temperature T, and the reduced mass of the target projectile system μ.

For neutron induced processes Maxwellian-averaged cross sections $\langle\sigma\rangle$ are commonly given instead of reaction rates. These are defined as

$$\langle\sigma\rangle_{kT} = \frac{\langle\sigma v\rangle}{v_T} = \frac{2}{\sqrt{\pi}} \frac{\int_0^\infty \sigma(E_n)\, E_n \exp(-E_n/kT)\, dE_n}{\int_0^\infty E_n \exp(-E_n/kT)\, dE_n}, \tag{2}$$

where $E_n = E_{n,lab}(A/(A+1))$ is the total kinetic energy in the center-of-mass system, $E_{n,lab}$ is the laboratory neutron energy, and $v_T = \sqrt{2kT/\mu}$ is the mean thermal velocity. Typical thermal energies for neutron capture nucleosynthesis range from kT = 8 keV during the s process to about 100 keV for the freeze-out phase of the explosive r- and p-process scenarios [1]. Accordingly, neutron cross sections are required over an energy interval from 0.1 to 500 keV.

Under stellar conditions, the Maxwellian-averaged cross sections of a number of isotopes have to be corrected by a temperature-dependent stellar enhancement factor

$$SEF(T) = \frac{\langle\sigma\rangle^{star}}{\langle\sigma\rangle_{kT}}. \tag{3}$$

which accounts for neutron capture on excited states in thermally equilibrated nuclei.

3 Neutron Cross Section Data for the s-Process

Since the duration of the s-process episodes are sufficiently long, reaction flow equilibrium is established over almost the entire mass range from Fe to Bi. This has the consequence that the resulting s abundances are inversely proportional to the

respective stellar (n,γ) cross sections. The only exceptions from this behavior are the neutron magic nuclei, since their small capture cross sections act as bottle necks for the reaction flow. Accordingly, (n,γ) cross sections are essential for quantitative *s*-process studies, for deriving the abundance pattern as well as for characterizing the reaction flow.

Because the *s*-process path follows the stability valley, the involved isotopes are easily accessible to laboratory experiments and have been determined with increasing accuracy. This reliable data base opens the possibility for detailed investigations not only of the gross properties of the *s* process (abundances, overall neutron exposure, seed nuclei) but even of the physical conditions at the stellar site (analyses of branchings in the reaction path). Hence, the *s* process provides a direct link between experimentally measured nuclear physics data and quantities related to the stellar environment: an ideal tool for testing the yet uncertain stellar models of the Red Giant stage.

Depending on the particular problem, cross section uncertainties of less than 2% are required for meaningful analyses. This is particularly stringent for cases where accurate abundance information is available, i.e. for isotopic patterns of elements with pure *s* nuclei, the well defined elemental abundances of the rare earth elements, and the wide new field of pure *s* material identified in stellar dust grains which survived in meteorites [2].

The current status of these important data [3] is illustrated in Fig. 1 showing the stellar (n,γ) rates between Fe and Pb for a characteristic thermal energy of kT = 30 keV. Significant progress in experimental techniques in the last decade has led to increasingly accurate data. This is evident from the fact that many of the data points exhibit uncertainties smaller than the $\pm$4%, indicated by the size of the symbols in Fig. 1, even in the deep minima at magic neutron numbers. Many of the more recently reported measurements helped to resolve discrepancies among previous results. Nevertheless, additional and more precisely measured cross sections are still needed, i.e. in the mass region from Mo to Pd where large uncertainties persist. In some cases, experimental data are yet missing, namely for isotopes of Ge and Se. Until recently, this was also true for the important *s*-only nuclei ^{128}Xe, ^{130}Xe, and ^{192}Pt. Meanwhile, first measurements on th latter isotopes have been completed and are under analysis.

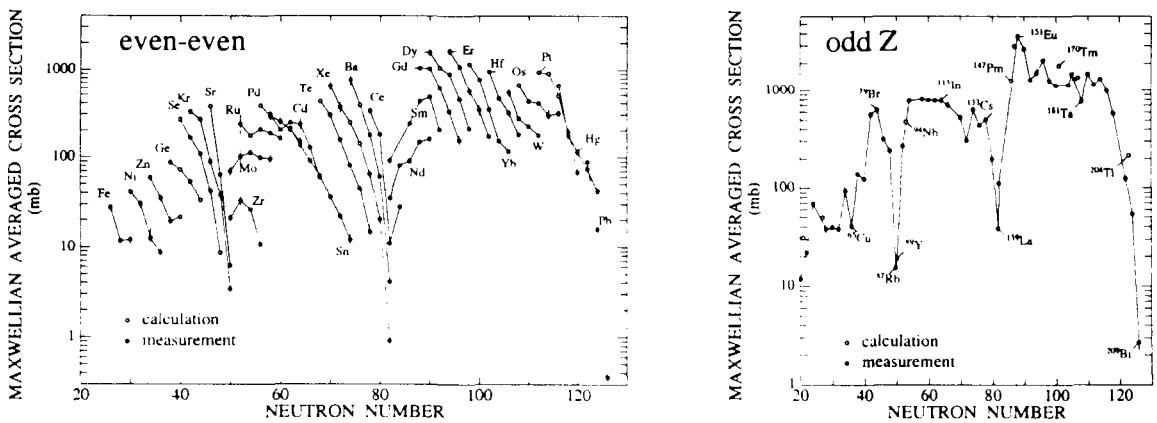


Figure 1: The 30 keV Maxwellian-averaged (n,γ) cross sections along the *s*-process path. Experimental and theoretical data are indicated by black and open symbols, respectively. In the left panel, the solid lines connect isotopes of the same element.

Experimental techniques have been improved with respect to accuracy, sensitivity, and spectroscopic quality. An impressive example for the accuracy that can be presently achieved are the difficult measurements of the small stellar (n,γ) cross sections of the barium isotopes, where a consistent data set could be established [4] from three independent experiments with final uncertainties of about 3%. This case illustrates that different techniques [5, 6, 7] can agree within the quoted uncertainties, and that the overall quality of the data can be improved by combining complementary methods. Even smaller uncertainties of typically 1% were achieved in experiments with the Karlsruhe 4π BaF_2 detector (see e.g. [8, 9]).

Experiments of high sensitivity permit the study of small cross sections and of cases where only small samples could be used. This includes time-of-flight (TOF) measurements on light isotopes, e.g. the very small (n,γ) cross sections of ^{12}C [10] and ^{16}O [11], which show the importance of the direct capture (DC) mechanism in this mass region. Also, determination of the small cross sections of neutron magic nuclei could be improved by high-resolution TOF measurements, e.g. on ^{138}Ba and ^{208}Pb [12] as well as by series of activation measurements [13, 14].

The standard activation method has been extended by the fast cycling technique [15]. With this new method, which can be used to investigate short-lived residual activities with half-lives down to 1 s, the small cross sections of light neutron-rich isotopes from ^{14}C to ^{50}Ti were successfully determined [16]. The excellent sensitivity of the activation technique also allowed a first measurement of the (n,γ) cross section of ^{155}Eu, a radioactive branch point nucleus with a half-life of less than 5 yr [17]. The main difficulties of such measurements, namely the high γ-background due to the sample activity, the radiation hazards related to sample handling, and the fact that suitable samples are not easily available, can be considerably reduced in activation measurements where samples of about 100 ng or some 10^{14} atoms are sufficient. Therefore, such studies could benefit from future radioactive ion beam facilities where intensities of 10^9 s^{-1} allow the production of appropriate samples within a few hours, presumably with considerably better purity than can be achieved by radiochemical methods.

For the first time neutron capture events leading to isomeric states could be identified in a TOF experiment covering the keV region. Due to the good energy resolution of the Karlsruhe 4π BaF_2 detector, capture γ-ray cascades to the ground and isomeric states could be distinguished in several Yb isotopes by their different sum energies [8], a novel feature that was not available to previous techniques using Moxon-Rae detectors or the pulse height weighting method. The spectroscopic quality of these new experiments will allow the study of important effects of long-lived isomers on some s-process abundance patterns, since these are determined by the respective partial cross sections. Moreover, these data are also useful as tests of the γ-decay spectra predicted by statistical model calculations.

The present status of (n,γ) data for the s process is summarized in Fig. 2 showing the respective uncertainties over the mass range from H to Bi. From this figure two main conclusions can be drawn:

(i) Experimental techniques have reached a stage where the 1% accuracy level required for meaningful analyses of particular abundance patterns has been met.

(ii) This quality has been achieved so far only for a minority of the relevant isotopes.

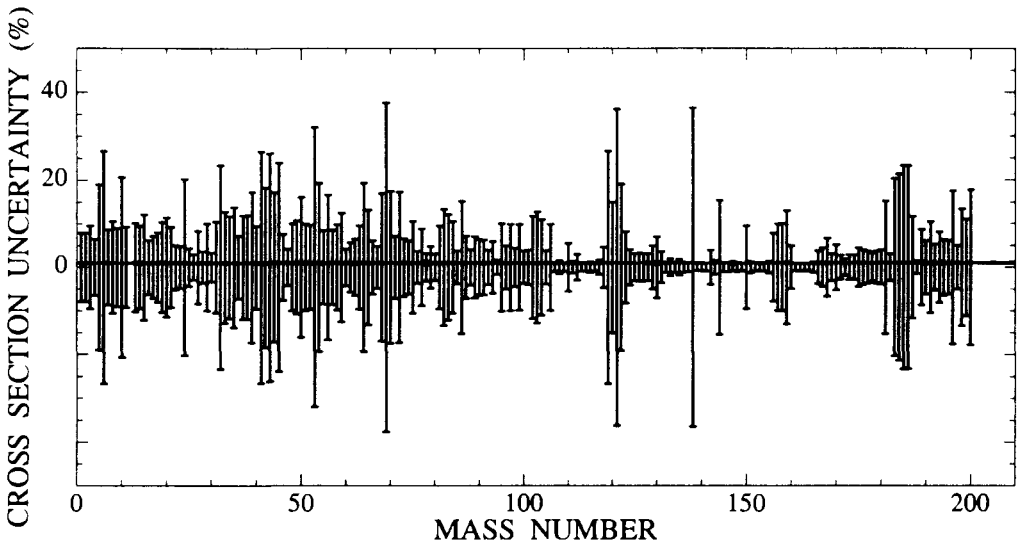


Figure 2: Experimental uncertainties of the stellar (n,γ) cross sections at s-process temperatures (kT=30 keV). Note that the mass region below A=120 requires substantial improvement.

4 Neutron Data for Explosive Scenarios

Nucleosynthesis in explosive scenarios occurs off the stability valley. The rapid neutron capture process (r process) presumbly related to supernova explosions or neutron star mergers (see contribution by S. Goriely) is characterized by neutron densities in excess of 10^{22} cm^{-3} and temperatures of about 1 to 2 billion degrees. Accordingly, neutron captures are much faster than β-decays, resulting in a reaction path close to the neutron drip line. The contributions from the r process equal approximately those from the s process and constitute about 50% of the isotopic abundances between Fe and Bi as well as all of the actinides.

The remaining rare isotopes on the proton rich side of the stability valley are assigned to the p process, which is assumed to take place in type II supernovae. The shock wave of the explosion heats the outer zones and ignites explosive burning of the Ne/O layer leading to temperatures of up to 3 billion degrees. At this point, the thermal photon bath becomes hot enough to photodesintegrate the preexisting s isotopes in this region, and hence to produce the proton-rich nuclei.

In contrast to the comparably stable situation of the s process, the complex explosive nucleosynthesis scenarios imply huge reaction networks including several thousand reactions. So far, the respective reaction rates have been exclusively obtained by statistical model calculations, which could only be tested by experimental data for stable nuclei. In order to verify the necessary extrapolation to the region of unstable nuclei, experimental data for as many unstable isotopes as possible would, therefore, be highly desirable.

5 Recommendations

5.1 Data for the *s* Process

The persisting data needs for quantitative *s*-process analyses concentrate on requests for (n,γ) measurements in the following areas:

- All *s*-only nuclei, since these are the key isotopes for any *s*-process investigation including the analysis of the *s*-process branchings. The *s*-process branchings are important because they provide direct clues with respect to stellar neutron density, temperature, and pressure and allow to characterize the He-burning zones, where the *s* process actually takes place. So far, the necessary accuracy of 1% has been reached only for half of the 33 *s*-only nuclei between ^{70}Ge and ^{204}Pb.

- Elements for which isotopic anomalies have been found in meteoritic inclusions. These signatures are characteristic of specific nucleosynthetic processes and should be investigated with particular emphasis. For this purpose, the cross sections have to be determined with uncertainties of 1% for decoding the full information contained in the respective abundance patterns. However, the present status is far from being adequate, particularly for the lighter elements oxygen, neon, magnesium, silicon, calcium, titanium, and zirconium. In this group, data for about 70 isotopes have to be determined.

- Nuclei at or near magic neutron numbers N=50, 82, and 126, which act as bottlenecks for the reaction flow in the main *s*-process region between Fe and Bi. These data should be known with uncertainties of better than 3%.

- Abundant light isotopes below Fe, which may constitute crucial neutron poisons for the *s*-process. Of particular importance are ^{16}O, ^{18}O, and ^{22}Ne.

- Nuclides for which the DC process contributes a significant fraction of the astrophysical reaction rate. For example, two thirds of the ^{208}Pb *s*-process (n,γ) rate is due to direct capture which is very difficult to detect in TOF measurements. Similar cases with significant DC contributions are ^{14}C, ^{16}O, ^{88}Sr, and ^{138}Ba.

- Nuclei with as yet unmeasured cross sections. These gaps in the experimental data, as well as the uncertain cross sections of numerous nuclei, especially in the mass region below Fe, around A=100, and near the end of the *s*-process region from Pt to Bi should be determined to the 5% level.

- Last, but not least, enhanced efforts should be directed to measurements of unstable nuclei of relevance for the reliable interpretation of the branchings in the *s*-process reaction path. In addition to the activation technique, the very high neutron fluxes available at spallation neutron sources appear to be promising options for such studies [18, 19]. From a list of possible measurements, priority should be given to the important branch points ^{79}Se, ^{147}Pm, ^{151}Sm, ^{163}Ho, ^{170}Tm, ^{171}Tm, ^{179}Ta, ^{204}Tl, and ^{205}Pb. These cases are of immediate relevance to *s*-process analyses and should not present unexpected experimental problems.

Besides the total (n,γ) cross sections listed above, partial cross sections leading to long-lived isomers are important for several branchings. A well-known example is the population of the 10.8 yr isomer in ^{85}Kr, which determines the probability for neutron captures to the neutron magic isotope ^{86}Kr. So far, this partial cross section has been determined by activation for a particular thermal energy only. Therefore, complementary TOF measurements with a 4π γ-detector such as the Karlsruhe BaF_2 array are important for determining the energy dependence of partial cross sections. This information is crucial for describing the branching pattern in the complex He burning scenarios suggested by stellar models.

Elastic and inelastic scattering data for a variety of isotopes are definitely needed for establishing a quantitative set of stellar enhancement factors, in analogy to the treatment of the Os isotopes [20, 21].

Finally, the neutron producing reactions in the *s* process, i.e. the (α,n) reactions on ^{13}C and ^{22}Ne, have to be improved considerably. Both cross sections exhibit large uncertainties and are not yet directly measured in the relevant stellar energy range. The best way for extrapolating the existing data to stellar energies is to combine as many reaction channels as possible in an R-matrix analysis. Accordingly, measurements of the (n,α)-cross sections of ^{16}O and ^{25}Mg would provide a significant contribution to this problem.

Parallel to the efforts for completing the experimental data base, necessary improvements on the theoretical side would include the following:

- *Local systematics* of nuclear properties relevant to the calculation of astrophysical reaction rates close to the stability valley should be extended. This includes level density systematics (and the energy dependence of parity distributions), parametrization of giant dipole resonance (GDR) widths and energies, and neutron potentials. Experimental data should be obtained and then described in simple, phenomenological models, taking into account a possible extrapolation towards very neutron-rich nuclei.

- The consistent treatment of *SEFs and superelastic scattering* should be emphasized by using elastic and inelastic scattering data.

- Efforts towards *improved microscopic calculations* of relevant properties, such as masses, shell and microscopic corrections for level density calculations, level schemes, and optical potentials should be intensified.

- *Comprehensive DC cross sections* should be calculated based on systematics of neutron optical potentials for DC and of scattering phase shifts. Also spectroscopic factors and information on nuclear levels, either from microscopic models or from experiment are important in this context.

5.2 Neutron Data for Explosive Nucleosynthesis

In *r*-process calculations, neutron cross section data have a direct impact for scenarios with comparably low neutron densities as well as during freeze-out, where they contribute to smoothen the pronounced odd-even effects predicted for the primary yields. In principle, several unstable nuclei on the neutron-rich side of the stability

valley could be studied experimentally, e.g. ^{90}Sr, ^{123,126}Sn, ^{182}Hf, ^{226}Ra, and a number of higher actinides.

Similarly, such data would clearly improve the description of freeze-out effects in the *p* process, where neutrons are liberated by (γ,n) reactions during the explosive burning of the Ne/O shell. The strong impact of these reactions on the final abundance distribution has already been demonstrated [22]. Furthermore, (n,γ) cross sections of proton-rich nuclei would be most useful in determining the inverse rates by detailed balance. Experimentally feasible cases include about 25 unstable isotopes between ^{53}Mn and ^{202}Pb.

Apart from measurements on unstable nuclei, even data for stable isotopes are urgently required for improving the reaction rates used in explosive nucleosynthesis. In particular, complete data sets for long isotope chains are important for testing the results obtained by statistical model calculations and to verify the necessary further extrapolation to the region of unstable nuclei. This means that stellar (n,γ) cross sections should be determined also for all *r*- and *p*-only nuclei.

At this point it should be mentioned that there are only very few experimental (p,γ) and (α,γ) cross sections at astrophysically relevant energies in the mass region of the *p* process. This holds for stable isotopes, not to speak of data on unstable nuclei.

Especially, the (α,γ) and (α,p) rates represent significant nuclear physics uncertainties in understanding the *p*-process abundances. Since direct (α,γ) and (α,p) measurements are difficult and time-consuming, the calculated rates are poorly constrained by experimental data. In particular, the α-nucleus potential used in statistical model calculations seems to be rather uncertain. A series of (n,α) measurements at astrophysically meaningful energies could help to solve this persisting problem.

References

[1] F. Käppeler, F.-K. Thielemann, and M. Wiescher, Ann. Rev. Nucl. Part. Sci. **48**, 175 (1998).

[2] F. Käppeler, Prog. Nucl. Part. Phys. **43**, 419 (1999).

[3] Z. Bao *et al.*, Atomic Data Nucl. Data Tables **76**, 70 (2000).

[4] F. Käppeler, in *Nuclei in the Cosmos V*, edited by N. Prantzos and S. Harissopulos (Editions Frontières, Paris, 1998), pp. 174 - 180.

[5] F. Voss, K. Wisshak, and F. Käppeler, Phys. Rev. C **52**, 1102 (1995).

[6] P. Koehler *et al.*, Phys. Rev. C **54**, 1463 (1996).

[7] P. Mutti, F. Corvi, K. Athanassopoulos, and H. Beer, Nucl. Phys. **A621**, 262c (1997).

[8] K. Wisshak *et al.*, Phys. Rev. C **61**, 065801 (2000).

[9] F. Voss *et al.*, Phys. Rev. C **59**, 1154 (1999).

[10] T. Ohsaki *et al.*, Ap. J. **422**, 912 (1994).

[11] M. Igashira *et al.*, Ap. J. **441**, L89 (1995).

[12] H. Beer, F. Corvi, and P. Mutti, Ap. J. **474**, 843 (1997).

[13] F. Käppeler, K. Toukan, M. Schumann, and A. Mengoni, Phys. Rev. C **53**, 1397 (1996).

[14] F. Käppeler, W. Zhao, H. Beer, and U. Ratzel, Ap. J. **355**, 348 (1990).

[15] H. Beer *et al.*, Nucl. Instr. Meth. A **337**, 492 (1994).

[16] H. Beer *et al.*, in *Capture Gamma-Ray Spectroscopy and Related Topics*, edited by G. Moln'ar, T. Belgya, and Z. R'evay (Springer, Berlin, 1997), p. 489.

[17] S. Jaag and F. Käppeler, Phys. Rev. C **51**, 3465 (1995).

[18] A. Michaudon and S. Wender, Technical report, Los Alamos National Laboratory, New Mexico, USA, report LA-UR-90-4355.

[19] S. Abramovich *et al.*, Technical report, CERN, Geneva, Switzerland, report CERN/SPSC 99-8; SPSC/P 310.

[20] R. Winters, R. Carlton, J. Harvey, and N. Hill, Phys. Rev. C **34**, 840 (1986).

[21] R. Winters, R. Macklin, and R. Hershberger, Astron. Astrophys. **171**, 9 (1987).

[22] M. Rayet, N. Prantzos, and M. Arnould, Astron. Astrophys. **227**, 271 (1990).

International Atomic Energy Agency Nuclear Data Section
Advisory Group Meeting on Long Term Needs for Nuclear Data
Vienna, Austria, Nov. 28 – Dec. 3, 2000

NUCLEAR DATA NEEDS IN NUCLEAR ASTROPHYSICS: CHARGED-PARTICLE REACTIONS

MICHAEL S. SMITH

Physics Division, Oak Ridge National Laboratory, Oak Ridge, TN, USA

Abstract

Progress in understanding a diverse range of astrophysical phenomena - such as the Big Bang, the Sun, the evolution of stars, and stellar explosions - can be significantly aided by improved compilation, evaluation, and dissemination of charged-particle nuclear reaction data. A summary of the charged-particle reaction data needs in these and other astrophysical scenarios is presented, along with recommended future nuclear data projects.

1 Importance of Nuclear Data in Astrophysics Studies

Nuclear astrophysics involves the study of the origin of the elements and the evolution of the astrophysical sites where this synthesis occurs. Systems as diverse as the early Universe, the interstellar medium, red giant stars, novae, and supernova explosions are the focus of many vigorous observational, theoretical, and laboratory research programs worldwide. These programs address some of the most interesting questions in nature: What are the origins of the elements that make up our bodies and our world, that make life on earth possible? How did the solar system, the sun, the stars, and the galaxy form, and how do they evolve? What is the total density of matter in the universe, and will the universe eventually collapse or expand forever? Nuclear data is the physical basis for models used to address these questions. It is also needed to interpret a wealth of new observations by ground-based telescopes such as the Keck Telescopes, by space-borne observatories such as the Hubble Space Telescope and the Chandra X-Ray Observatory, and by large subterranean detectors such as the Sudbury Neutrino Observatory and SuperKamiokande.

A diverse set of nuclear data is required to model the composition changes and energy release in a wide range of astrophysical environments. For example, rates of fusion reactions, transfer reactions, and nuclear decays are all needed, as are nuclear masses. Given the predominance of H and He in the Universe, many of the nuclear reactions occurring in astrophysical environments are induced by charged particles. Where available, the relevant nuclear reaction rate information is derived from laboratory measurements. For thousands of reactions, however, there is little experimental data. Some of these rates are calculated using theoretical analyses of indirect measurements and nuclear structure information such as excitation energies, spins and parities, resonance decay modes, and ground state half-lives. Other rates are calculated using statistical reaction models such as Hauser-Feshbach.

There have been a number of exciting advances in laboratory nuclear physics, including the availability of beams of radioactive nuclei, the ability to measure solar fusion reactions at their appropriate stellar energy in underground low-background laboratories, and sophisticated arrays of high-resolution gamma-ray and charged-particle detectors. Such advances, coupled with the evolution of fast, inexpensive computers, has enabled

extremely complex astrophysical calculations of increasing realism to be carried out. For example, calculations coupling the time-dependent synthesis of hundreds of isotopes via thousands of linking reactions with multi-dimensional simulations of stellar and explosive environments are now beginning. These new codes require significantly more, and more accurate, nuclear data than ever before to keep pace with the stunning new capabilities of the Hubble, Chandra, Keck, and other observatories. Progress in many fundamental problems in astrophysics requires the best predictive power of astrophysical models.

In many instances, this predictive power has a strong dependence on the input nuclear data. There are numerous examples of the significant impact that new, more precise assessments of nuclear data can have on astrophysical studies. One example is the rate of the $^{3}H(\alpha,\gamma)^{7}Li$ reaction producing ^{7}Li during the Big Bang. The standard Big Bang Nucleosynthesis (BBN) model predictions for ^{7}Li production are extremely sensitive to the model's one "free" parameter - the amount of "normal" matter in the universe (the baryon density) - as well as to the input nuclear physics. Consequently, comparisons of the primordial ^{7}Li abundance inferred from observations to BBN model predictions allows a constraint to be put on the amount of normal matter in the universe. An evaluation [1] of the rate of the $^{3}H(\alpha,\gamma)^{7}Li$ reaction was a factor of two lower at temperatures corresponding to the Big Bang than previous work. When put into a BBN model, this resulted in a production of ^{7}Li which was 20 % lower than previous model predictions. When compared to observations, this resulted in a limit on the baryon density of the universe which was 50 % higher than the previous limits. Therefore, an improvement in one nuclear reaction rate out of the many used in Big Bang models resulted in increasing the estimate of the amount of material in the Universe by 50 %.

Numerous other cases of the critical role of nuclear data in astrophysical models can be cited - many of them involving charged-particle reactions. For example, there is a very strong dependence of the abundances produced in supernova explosions on the value of the $^{12}C(\alpha,\gamma)^{16}O$ reaction rate [2]. This rate, one of the most important in all of nuclear astrophysics, determines the ratio of C to O after core He burning, and determines the subsequent evolution of a massive star into a neutron star or black hole. For this reason, it is the subject of much experimental and theoretical attention. The rates of reactions that produce and destroy ^{26}Al have a significant impact [3] on the interpretation of observations of ^{26}Al in the interstellar medium [4] and in meteorites [5]. The uncertainty in the rate of the $^{7}Be(p,\gamma)^{8}B$ reaction determines the current uncertainty in neutrino oscillation "solutions" to the Solar Neutrino Problem [6]. Uncertainty in the rates of proton-capture reactions on radioactive isotopes contribute to a factor of ~300 uncertainty in the production of the long-lived radioisotope ^{18}F, which can act as an observational tracer of nova explosions [7]. This uncertainty makes it difficult to determine the sensitivity of multi-million dollar observatories for measuring gamma rays from these explosions. These are only a few examples of the significant impact that nuclear data can have on nuclear astrophysics studies.

2 Motivation for Expanded Nuclear Data Activities in Astrophysics

The user community for nuclear astrophysics data consists of scientists using a combination of sophisticated astrophysics modeling codes to decipher the most detailed, precise, and costly astrophysical observations ever made. Measurements in the nuclear laboratory form the empirical foundation for the current models of element synthesis. Astrophysics models therefore have a crucial dependence on the completeness, precision, and timeliness of substantial quantities of input nuclear data. Evaluations of nuclear reaction data for astrophysics was initiated and nurtured by Willy Fowler and his collaborators (see, e.g. [8,9]), but unfortunately he and his program are no longer with us. There is currently little manpower devoted to continuing his effort. Indeed, there is a growing recognition that the data needs of the research community cannot be provided by the voluntary efforts of any single research group alone. The little voluntary work that is being done is fragmentary, poorly coordinated, and most importantly, not nearly enough. The result is a developing break down in communication

between scientists in the nuclear laboratory and those using the data in their codes. This results in an extremely poor utilization of important, difficult, and costly nuclear physics experiments by the astrophysicists for whom they were measured.

There is also a growing recognition that the data needs in nuclear astrophysics have a strong overlap with nuclear data needs in other basic and applied nuclear physics fields. Furthermore, the nuclear data community has developed considerable expertise and technical resources [10] that could be used to address some of these astrophysics data needs. In fact, some resources in the U.S. nuclear data community have been shifted towards work in this frontier field. However, there is currently no systematic, coordinated, long-term solution in the U.S. or internationally to the problem of providing the best nuclear data in forms needed for astrophysical models.

Indeed, the current situation in nuclear astrophysics parallels the situation in reactor physics and nuclear weapons design in the U.S. 25 years ago. Astrophysicists often have their own sets of nuclear data produced by the individual scientist from scattered data collections of their choosing. This makes it difficult to compare results, and also makes the uniform updating of codes with new evaluations difficult. Significant progress was realized in these other fields when standardized sets of data were widely available to researchers, allowing the model codes to be decoupled from the input data. While some astrophysicists will always prefer to use proprietary data sets, a set of freely available, regularly updated, communal data sets would lead to a standardization that would help advance the field and would encourage the development and improvement of the input data sets themselves in a synergistic way.

There are, however, some difficulties in producing these data sets. For example, laboratory measurements often probe the astrophysical reactions of interest only indirectly. Relevant information (e.g., nuclear resonance widths and energies) must be extracted and used to calculate useful quantities such as reaction rates. This requires extra work before the data can be used in astrophysical models. Even when this work is done and published, it still must be evaluated, compared with other measurements, and disseminated to the community. For this and many other reasons, many of the existing sets of nuclear data for astrophysics studies are incomplete, both in their scope (e.g., not enough nuclides) and their depth (e.g., missing some crucial information). For example, some of the most sophisticated new astrophysical observations cannot be used to distinguish between competing astrophysical models without significantly improving calculations of uncertainties in model predictions. Uncertainties in the input nuclear physics, which are often not provided in available evaluated nuclear data sets, are required in order to calculate the uncertainties in model predictions of astronomical observables.

Dedicated effort is required to address all of these problems and to provide usable, accurate, and significant amounts of nuclear data in a timely fashion. A new initiative is required, and the benefit will be progress in many fundamental problems in nuclear astrophysics. The data stewardship activities which are needed include: making high-quality data evaluations, complete compilations, and timely and useful disseminations; and using nuclear reaction and structure models to extend existing measurements to unmeasured reactions, energy ranges, and isotopes.

3 Nuclear Data Used in Nuclear Astrophysics

In general, astrophysical models require the rates of and energy released in nuclear reactions occurring in astrophysical sites [11]. Many of the relevant nuclear reactions are induced by charged particles because of the overwhelming presence of H and He in the Universe. The rates are derived from laboratory measurements of cross sections convoluted with the thermal (Maxwell-Boltzmann) relative velocity distribution of the interacting

particles. The released energies of the relevant reactions (Q-values) and nuclear masses are derived from measurements and calculations. The types of charged-particle reactions occurring in astrophysical scenarios include particle capture (fusion), particle exchange, particle transfer, spallation, and (to a lesser extent) three body reactions. Additionally, weak interactions (electron and positron decays and captures), photodisintegrations, and beta-delayed particle emissions occur. For example, heavy isotopes are synthesized from the fusion of lighter isotopes with the "fuel" (e.g., protons, alphas) present in stellar environments. The relevant energies of these reactions depend on the astrophysical site, but range from approximately 0.01 - 2 MeV/u for reactions with charged particles. The nuclides relevant for each of these reaction types varies greatly with the phenomenon studied. For example, charged-particle reactions on stable isotopes for masses up to the Fe group and beyond are important for massive star evolution, whereas reactions on radioactive isotopes near the proton drip line are important for explosive nucleosynthesis in novae, supernovae, and X-ray bursts. Details of important reactions in different scenarios are given in Section 4. There are a number of existing data sets relevant for nuclear astrophysics studies [10], some of which need significant expansion, updating, or other improvements. These data sets and projects for their improvement are discussed in Section 5.

Since the thousands of nuclear reactions occurring in some astrophysical environments cannot all be measured, nuclear models play a central role in providing the information needed for astrophysics models. Nuclear models, used to calculate the cross section and the energy release (Q-value) of unmeasured reactions, require substantial nuclear structure information as input, such as masses, the parameters of resonances near particle capture thresholds of the relevant isotopes, optical model parameters, one- and two-particle separation energies, and single-particle energy levels. Other quantities such as nuclear wave functions, level densities and partition functions are also required as input. The nuclear modeling needs for nuclear astrophysics will be discussed in a separate article in these proceedings.

4 Nuclear Data Needs for Specific Astrophysics Scenarios

A very diverse set of information on nuclear reactions and nuclear properties is required for nucleosynthesis models, and the particular information varies significantly for different astrophysical phenomena. Examples of the information needed for studies of the Big Bang, stellar evolution, stellar explosions, and other scenarios are given below.

4.1 Big Bang Nucleosynthesis

Approximately 3 minutes after the beginning of the Universe in the Big Bang, the temperature had cooled to roughly 10^9 K, enabling protons and deuterons to fuse to form deuterium and initiating the synthesis of nuclei up to ^{7}Li. The twelve most important reactions in the synthesis of nuclei in homogeneous models of the early Universe are ^{1}H(n,γ)d, ^{2}H(p,γ)^{3}He, ^{2}H(d,n)^{3}He, ^{2}H(d,p)t, ^{3}He(n,p)t, ^{3}H(d,n)^{4}He, ^{3}He(d,p)^{4}He, ^{3}He(α,γ)^{7}Be, ^{3}H(α,γ)^{7}Li, ^{7}Be(n,p)^{7}Li, ^{7}Li(p,α)^{4}He, and the decay of the neutron [1]. These reactions have been recently evaluated at temperatures appropriate for the Big Bang (laboratory energies up to ~1 MeV) [1,12]. Currently, the ^{1}H(n,γ)d, ^{2}H(p,γ)^{3}He, ^{3}He(α,γ)^{7}Be, and ^{7}Li(p,α)^{4}He need additional work, along with a critical, statistically robust assessment of the uncertainties of all the important rates considering all previous measurements. These uncertainties are important because they are input into models employing a Monte Carlo technique to determine uncertainties in the synthesized abundances.

There are, however, models of the early universe where the phase transition from quarks and gluons to hadrons could possibly cause proton-rich and neutron-rich regions to form [13]. These Inhomogeneous Big Bang Nucleosynthesis (IBBN) models involve nuclear reactions on some light unstable isotopes which produce different abundances than in standard Big Bang models. Examples of important reactions are ^{8}Li(α,n)^{11}B and

$^{8}Li(d,n)^{9}Li$ [14], as well as neutron captures on neutron-rich isotopes of carbon. Recent observations of the spectrum of the cosmic microwave background radiation power spectrum [15] suggest a universal mass density that is consistent with some IBBN models but outside the range normally quoted for standard Big Bang models [16]. This suggests the importance of evaluating the nuclear reactions in IBBN models.

4.2 Evolution of Low Mass Stars

Stars are born when clouds of gas and dust in the interstellar medium gravitationally collapse and ignite nuclear reactions at their core. These reactions turn H into He and generate energy, preventing further gravitational collapse and causing the stars to shine [11]. Low-mass stars like our Sun burn hydrogen through sequences of reactions called the pp-chain, while those a few times the mass of the Sun burn hydrogen through the CNO cycles. These stars eventually exhaust their hydrogen fuel at the core - leaving behind a He core surrounded by a thin shell where H is burned and finally an outer convective layer. A rich nucleosynthesis occurs when these stars go through a thermally-pulsing asymptotic giant branch (TP-AGB) phase [18]. Some of the nuclides created are dredged up to the surface by convection and ejected into space via strong winds. There are significant uncertainties in the rates of a number of nuclear reactions such as $^{14}N(p,\gamma)^{15}O$, $^{15}N(p,\gamma)^{16}O$, $^{17}O(p,\gamma)^{18}F$, and $^{17}O(p,\alpha)^{14}N$ occurring during this evolution. These uncertainties make it difficult to understand the time for core H depletion, the abundances of the C, N, and O isotopes, and other evolutionary aspects [17,18]. Reactions in the NeNa Cycle also occur in the hydrogen-burning shell, such as $^{22}Ne(p,\gamma)^{23}Na$ and $^{23}Na(p,\gamma)^{24}Mg$, which need to be better understood to describe the observed abundances of Na isotopes on stellar surfaces. Similarly, in the MgAl Cycle, the $^{25}Al(p,\gamma)^{26}Si$ and $^{25}Mg(p,\gamma)^{26}Al$ reactions influence the abundance of Al isotopes observed on stellar surfaces and the amount of the long-lived radioactive isotope ^{26}Al in the interstellar medium (observed by the Compton Gamma Ray Observatory) [4]. The $^{12}C(\alpha,\gamma)^{16}O$ reaction [19] determines C / O ratio after core He burning and thereby strongly influences the subsequent evolution of these stars. Improved measurements, theoretical calculations, and new evaluations are needed for this reaction. The $^{13}C(\alpha,n)^{16}O$ and $^{22}Ne(\alpha,n)^{25}Mg$ reactions are the source of neutrons for the s-process occurring in these stars, and the current large uncertainties in the $^{22}Ne(\alpha,n)^{25}Mg$ rate influences the production of elements heavier than Fe. The $^{22}Ne(\alpha,n)^{25}Mg$ reaction also influences the depletion of Ne - and therefore plays a role in the overproduction of Na in current stellar models.

4.3 Evolution and Explosions of Massive Stars

Stars with masses more than ~ 8 times that of the Sun go through a number of burning phases (H, He, C, O, Ne, Si) at their core resulting in a core of Fe. This core then quickly undergoes gravitational collapse, changing the material to primarily neutrons and compressing the material to super-nuclear densities. The core rebounds, sending out a shock wave responsible for a violent explosion generating ~ 10^{51} ergs of energy in less than a second [20]. These explosions are most likely responsible for the synthesis of approximately half of the isotopes heavier than iron via the rapid neutron capture process (r-process). The current scenario has a series of alpha-induced reactions assembling nuclides up to mass 80 (the alpha-process), followed by fast (n,γ) reactions on neutron-rich unstable isotopes out to the dripline, all occurring in the high-entropy wind off the surface of the newly-formed protoneutron star created in the explosion [21]. Supernovae are also thought to be the site of the p-process, which creates heavy proton-rich stable nuclides most likely by photodissociation of heavier nuclides.

Understanding this model of a core collapse supernova and all the nucleosynthesis requires significantly improved nuclear physics, including many charged-particle reactions [22]. The nucleosynthesis is more sensitive to the pre-explosion star than to the details of the explosion mechanism. The $^{12}C(\alpha,\gamma)^{16}O$ reaction is the most crucial reaction for these studies [2], but there are many others. For example, screening effects at low energies need to be understood for the $^{12}C + ^{12}C$ reaction in pre-collapse core carbon burning. Also, p-induced & α-induced reactions on C – Si nuclei influence the energy generated in CNO burning, the abundances of

stable nuclides such as ^{17}O, ^{18}O, and ^{22}Ne, and the abundances of long-lived radioisotopes such as ^{26}Al and ^{22}Na. The propagation of a supernova shock wave into the outer layers of the star may heat and compress material sufficiently to ignite brief occurrences of explosive burning [23]. The nuclear reactions involve captures of protons and alphas on proton-rich radioactive isotopes similar to those occurring in very hot novae or X-ray bursts (see Section 4.1). In the C-Si mass range, the reactions are not amenable to statistical model calculations because of the low level densities of the compound nuclei. Therefore, improved experiments and evaluations of individual reactions are needed.

For nuclides for $14 < Z < 50$, p-induced & α-induced reactions on unstable & stable nuclides are needed for supernova models, and these can be treated with statistical model calculations since the level densities are often sufficiently high [22]. These reactions influence the explosive burning that may occur in outer envelopes of supernovae, p-process nucleosynthesis, (the beginning of) r-process nucleosynthesis, and the abundances of radioisotopes ^{44}Ti and ^{60}Fe. Improved knowledge of Gamow-Teller resonances in (p,n) reactions in Si - Ni mass range is needed to determine e^- capture rates, as well as better rates for (α,γ) reactions on α-nuclei from ^{28}Si - ^{44}Ti. Furthermore, there is almost no experimental data on (p,γ) or (α,γ) reactions in the Gamow window of the p-process, for unstable or stable isotopes. Some experimental information, coupled to the best statistical model reaction calculations, are needed for work in this area.

4.4 Explosions in Accreting Binary Systems

4.4.1 Novae

Nova explosions are accretion-driven explosions caused by the transfer of mass from one star to a white dwarf companion star [24]. The mass transfer and subsequent rise in temperature and pressure can initiate a violent runaway thermonuclear explosion (~ 10^{38}- 10^{45}ergs released), resulting in the synthesis of elements up to mass ~ 40 and their subsequent ejection into space. Novae are thought to be sources of nuclides such as ^{13}C, ^{15}N, and ^{17}O which are difficult to produce in other astrophysical environments. The explosion also influences the subsequent evolution of the binary star system. These catastrophic stellar events are characterized by extremely high temperatures ($> 10^8$ K) and densities ($> 10^3$ g/cm^3). Under such conditions, (p,γ) and (α,p) reactions can rapidly (on timescales of nanoseconds to minutes) produce unstable nuclei on the proton-rich side of the valley of stability. Any such nuclei (decaying via e^+-emission) produced with half-lives longer than, or comparable to, the mean time between nuclear reactions can potentially undergo subsequent nuclear processing.

Reactions on proton-rich radioactive nuclei are crucial in these explosions [25,23], producing abundances which are very different than those from the hydrogen burning occurring in non-explosive environments [26,27] and generating energy up to 100 times faster than in quiescent stars. Some radioactive nuclei (those with lifetimes greater than 100 s) synthesized in explosions may be carried by convection to the top of the envelope before they decay. Observations of the γ - ray lines (especially the 511- keV emission of ^{18}F) resulting from such radioactive decays in the envelope may provide stringent tests of nova models [28,29]. The γ-ray emissions depend sensitively on the amount of radionuclides synthesized by nuclear reactions in the explosion, which in turn depends on the rates of nuclear reactions on radioactive isotopes [30,31]. Examples of important reactions include $^{17}F(p,\gamma)^{18}Ne$, $^{18}F(p,\alpha)^{15}O$, $^{19}Ne(p,\gamma)^{20}Na$, $^{20}Na(p,\gamma)^{21}Mg$, $^{21}Na(p,\gamma)^{22}Mg$, $^{22}Na(p,\gamma)^{23}Mg$, and $^{25}Al(p,\gamma)^{26}Si$ [25,32]. These can lead to hydrogen burning through the rapid proton capture process (rp-process), involving (p,γ) reactions near the proton dripline competing with e^+-decay and reaction cycles (e.g.,

the Ne-Na and Mg-Al cycles). The rates of such reactions on unstable isotopes are needed to understand the nova phenomenon. Even though evaluations of these reactions was listed as one of the top priorities by a steering committee on nuclear astrophysics data in 1996 [33], only a few reactions have been examined since that time. These evaluations can have significant impact on the interpretation of observations made by multi-million dollar astrophysical devices, especially those of the INTEGRAL satellite to be launched in 2002. The uncertainties of these rates are also needed because of the ability of Monte Carlo techniques to estimate uncertainties in the synthesized abundances from the input nuclear physics uncertainties [34].

4.4.2 X-ray Bursts and X-ray Pulsars

Other accretion-driven phenomena important in astrophysics include X-ray bursts and X-ray pulsars. These can occur when material is accreted onto the surface of a neutron star. The temperatures and densities can reach over 10^9 K and 10^6 g/cm^3, respectively [35,36], and the ensuing explosive hydrogen burning can synthesize isotopes with masses up to 80 - 100 or beyond [36,37,38] via reactions in the αp- and rp-processes. Recent studies of nucleosynthesis in these violent explosions suggest that their X-ray luminosity and neutron star crust composition are influenced by the nuclear reactions (most involving proton-rich radioactive isotopes) used in the model [39]. For example, the rates of reactions in the sequence $^{12}C(p,\gamma)^{13}N(p,\gamma)^{14}O(\alpha,p)^{17}F(p,\gamma)^{18}Ne(\alpha,p)^{21}Na(p,\gamma)^{22}Mg$ are crucial because they give the maximum flux of X-rays and serve as the gateway to the synthesis of heavier nuclei [36,40]. Properties of nuclei along the proton dripline are important, along with proton capture reaction rates, to understand these violent explosions.

4.5 Other Astrophysical Environments

There are other astrophysical sites [37] - e.g., the accretion disk around black holes [40] - where temperatures and densities may be sufficient for explosive hydrogen burning to occur. In these environments, reactions on proton-rich radioactive isotopes may play an important role, as they do in novae and X-ray bursts. Supermassive stars [41,42] are another possible site of explosive H and He burning via reaction sequences such as $p(p,e^+\nu)d(p,\gamma)^3He(\alpha,\gamma)^7Be(p,\gamma)^8B(\alpha,p)^{11}C(p,\gamma)^{12}N(e^+\nu)^{12}C$. This sequence produces ^{12}C via an alternate pathway to the slow triple-alpha process. This additional ^{12}C can, via hydrogen burning through the Hot CNO cycles, generate sufficient energy to possibly alter the evolution of these exotic stars. The reactions involved include those in quiescent stellar hydrogen burning (but at higher temperatures) as well as those in explosive stellar hydrogen burning. There is a renewed interest in these stars because of the recent discovery of many supermassive black holes at the centers of galaxies.

5 Nuclear Data Sets for Nuclear Astrophysics

One example of charged-particle nuclear data sets which are extremely valuable for studies in nuclear astrophysics are collections of reaction rates derived from measured or calculated cross sections. These collections typically include rates as analytic functions of temperature and be parameterized in formats easily input into astrophysical models. A list of data sets available (up to 1995) can be found in ref. [10]. Examples include: the 1988 collection of Caughlan and Fowler [9] with 160 reactions; an update of 86 of these reactions by the NACRE collaboration [43] in 1999; a new collection of 56 charged-particle reaction rates for stable and proton-rich unstable isotopes in the mass 20 - 40 region by Iliadis et al. [44]; collections of rates derived from statistical model cross section calculations by Rauscher and Thielemann [45], Woosley and Hoffman [46], and Goriely [47]; and the REACLIB rate library [48] which includes approximately ~ 8000 reactions, most from Hauser-Feshbach statistical model calculations. Some of these collections [9,43,44,48] include rates based on evaluated cross sections. However, these rates were based on evaluations of the total cross section only, over a

limited energy range - sufficient to determine a reaction rate over temperatures appropriate for a wide range of astrophysical environments. Complete ENDF-style cross section evaluations are not necessary for this type of work. The NACRE work includes bibliographic information and cross section data from all measurements of the reactions, something missing in the Caughlan and Fowler work, and rate uncertainties (although not quoted as 2σ limits) as well as calculated astrophysical S-factor. The Iliadis et al. work has similar features with much attention given to the rate uncertainties. In these two collections and that of Caughlan and Fowler, each rate has a different analytical form – a disadvantage when hundreds of these reactions are needed in astrophysical models. REACLIB, however, uses the same analytical form for all reactions, and therefore astrophysical models using it gain significantly in speed over the other using rate collections. One rate collection exclusively uses tabular rate values [47], while others [9,43] include both formulae and tabular values.

Other useful data sets include experimental cross sections, nuclear model calculations, evaluated cross sections, and bibliographic information. A list of data sets available (up to 1995) can be found in ref. [10]. A reference list of resources was also compiled by a steering committee [33] and posted online [49]. Compilations of unevaluated experimental cross sections of astrophysically relevant reactions are valuable, for example, because such compilations are the first step in obtaining evaluated cross sections. These compilations are also valuable for detailed studies of a particular reaction - for example, where different fits to the data are explored, where the data is fit over a different energy range, or where the data are rescaled to account for systematic uncertainties. Cross section uncertainties are needed in these compilations, since they are necessary to generate reaction rate uncertainties. The Cross Section Information Storage and Retrieval System (CSISRS) [50] at the U.S. National Nuclear Data Center (NNDC) is an online compilation of cross sections in the internationally accepted EXFOR format. This database has relatively good coverage of neutron-induced reactions, but sparse coverage for charged-particle reactions.

Furthermore, there is a strong need for evaluated cross sections for capture, transfer, exchange, and some other types of reactions occurring in astrophysical environments, since they are used to calculate reaction rates for direct input into astrophysics codes. The evaluated data sets are produced by combining a set of measurements, augmented by model results, into a cross section as a function of energy which is formatted in a standardized, well-documented manner. Access to the cross sections (as opposed to the rates) are useful for a number of reasons: for calculating reaction rates over a non-standard temperature range; for generating astrophysical S-factors; for examining the extrapolation of S-factors to unmeasured energy ranges; and for examining the quality of particular evaluations. Cross section uncertainties are also necessary because of their usefulness in determining reaction rate uncertainties. As mentioned above, ENDF [51] and similar general purpose evaluated cross section libraries [10] contain, in general, more information than is needed for the determination of thermonuclear reactions in astrophysics.

6 Nuclear Astrophysics Data Projects

A number of projects to address current deficiencies in existing charged-particle nuclear data sets for nuclear astrophysics are discussed below.

6.1 Explosive Hydrogen Burning Reaction Evaluations

It is very important to evaluate the rates along the proton dripline for the Hot CNO cycle and the beginning of the rp-process ($A < 20$), combining indirect and direct measurement information. These reactions are important for diagnosing observations of nova explosions and X-ray bursts, and are not contained in the Caughlan and

Fowler, NACRE, or Iliadis et al. rate collections. The REACLIB rates for these reactions are, in many cases, quite old and desperately need updating. This project was given a high priority by a steering committee [33].

6.2 Expanding the Caughlan and Fowler Rate Collection

While 86 of the 160 rates in the Caughlan and Fowler collection [9] were updated by the NACRE collaboration [43], there is a strong need to update the other approximately 80 rates which are now 12 years or more out of date. Since these rates are used in studies of a wide range of astrophysical phenomena, this project would have a wide impact on the field.

6.3. Selectively Modifying Rates in the NACRE Collection

Some rates in the Caughlan and Fowler collection [9] updated by the NACRE collaboration [43] are still preferred by some because they were based on both direct and indirect information. Other rates in the NACRE collection use polynomial fits to S-factors rather than R-matrix fits, which may be more appropriate in some cases. Finally, the NACRE rates use parameterization different from that in the REACLIB or Caughlan and Fowler collections. The parameterizations of the NACRE rates using the REACLIB format would be very useful, as would a very selective updating of some NACRE rates to include R-matrix fits of experimental data and indirect reaction information (e.g., transfer reaction measurements) when necessary.

6.4 Heavy Radioisotope Production Reaction Evaluations

The Iliadis et al. rate collection [44] almost fully addresses one of the high priority items cited by a steering committee [33]. However, it may be very useful to selectively extend this type of work to mass 40 - 60, to cover reactions important in the synthesis of long-lived radioisotopes ^{44}Ti and ^{60}Fe that play a role in diagnosing supernova explosions. While some reactions in this mass range are amenable to a statistical model treatment, others are dominated by individual resonances and need to be treated on an individual basis. Also, a determination of the uncertainties of some of these rates, in the manner done by Iliadis et al., would be very valuable.

6.5 Inhomogeneous Big Bang Nucleosynthesis Reaction Evaluations

As discussed in Section 4.1, there is renewed interest in IBBN models because of the latest observations in cosmology. Data from the MAXIMA and BOOMERANG devices is still being analyzed, and the MAP platform will be launched soon – all of which promises more information on the early Universe and may give even more motivation to study IBBN models. However, the rates for IBBN reactions have never been thoroughly evaluated, and this should be done to enable the best prediction of element synthesis is the early Universe when the assumption of a homogeneous composition is dropped.

6.6 Supermassive Star Reaction Evaluations

As discussed in Section 4.5., there is renewed interest in the evolution of supermassive stars, because of recent observations and because some of these reactions are now addressable with beams of radioactive isotopes. The last evaluation of supermassive star reactions was in 1989 [42], and a new evaluation of some of these reactions (e.g., those involving ^{11}C) may be very useful.

6.7 Modifying the REACLIB Collection

REACLIB includes approximately ~ 8000 reactions, most from Hauser-Feshbach statistical model calculations. Because REACLIB uses the same analytical form for all reactions, astrophysical models using it gain significantly in speed over those using rate collections with different parameterizations for each reaction (e.g., NACRE, Caughlan and Fowler). REACLIB is a very valuable and extensive collection of rates for astrophysics studies [48]. There are, however, some features that could be improved. For example, there is little information about the source of the rates, there is no uncertainty information on the rates, and no cross section or S-factor information is compiled. Additionally, the latest Hauser-Feshbach statistical model calculations [45] need to be incorporated into REACLIB, as do the NACRE rates and those from the new Iliadis et al. collection. An important long-term project is therefore to address these shortcomings of the REACLIB collection.

6.8 Expanding the CSISRS Cross Section Compilation

The CSISRS database [50] has relatively good coverage of neutron-induced reactions, but sparse coverage for charged-particle reactions. It has, however, been significantly updated in recent years with additional charged-particle reactions, most notably those in the NACRE collection, by an effort at the NNDC. Continued updating of the CSISRS database with charged-particle reactions is very important for the field.

6.9 The ENDF Evaluated Cross Section Database

As discussed above, full ENDF-style evaluations are generally not needed for astrophysics studies, however; only evaluations sufficient to determine thermonuclear reaction rates are necessary. Therefore, updating ENDF and similar evaluated cross section databases is not a high priority for nuclear astrophysics. However, whenever cross sections are evaluated (in a less extensive manner), they should be put in an ENDF-compatible format so that full evaluations can be pursued at a later date if warranted.

6.10 Dissemination Projects

There are a number of dissemination projects that would be very beneficial for nuclear astrophysics; many of these are detailed in Appendix 1 of ref. [10]. One of the most important is the establishment of a central archive for specialized sets of nuclear data for astrophysics models that would be accessible via the World Wide Web.

7 Summary

Progress in understanding a diverse range of astrophysical phenomena - such as the Big Bang, the Sun, red giant stars, massive star evolution, and stellar explosions - can be significantly aided by improved compilation, evaluation, and dissemination of charged-particle reactions. The charged-particle reaction data needs in these and other astrophysical scenarios is summarized, and some of the existing nuclear data sets for nuclear astrophysics are described. A number of projects to address current deficiencies in these data sets are described, including: evaluating reactions for explosive hydrogen burning, supermassive star evolution, inhomogeneous big bang nucleosynthesis, and the production of heavy radioisotopes in stellar explosions; expanding and / or modifying the CSISRS cross section compilation and NACRE and REACLIB reaction rate collections, and dissemination projects such as establishing a central archive for specialized sets of nuclear data for astrophysics models.

Acknowledgements

ORNL is managed by UT-Battelle LLC for the U.S. Department of Energy under contract DE-AC05-00OR22725. Gerry Hale, Victoria McLane, Eric Norman, Donald Smith, Roger White, and Stan Woosley contributed to Sections 1 & 2. Thanks to Rob Hoffman, Stephane Goriely, Franz Kaeppeler, Rob Cavallo, and Jeff Blackmon for useful discussions.

References

[1] M.S. Smith, L.H. Kawano, R.A. Malaney, Ap. J. Suppl. **85** (1993) 219.
[2] T.A. Weaver, S.E. Woosley, Physics Reports **227** (1993) 65.
[3] A. Coc et al., Astron. Astrophys. **299** (1995) 479.
[4] R. Diehl et al., Astron. Astrophys. **298** (1995) 445.
[5] S.S. Russell et al., Science **273** (1996) 757.
[6] E.G. Adelberger et al., Rev. Modern Phys. **70** (1998) 1.
[7] A. Coc, M. Hernanz, J. Jose, and J.-P. Thibaud, Astron. Astrophys. **357** (2000) 561.
[8] W.A. Fowler, G.R. Caughlan, B.A. Zimmerman, Ann. Rev. Astron. Astrophys. **5** (1967) 525.
[9] G.R. Caughlan, W.A. Fowler, At. Data Nucl. Data Tables **40** (1988) 283;
http://www.phy.ornl.gov/astrophysics/data/cf88/index.html.
[10] M.S. Smith, F.E. Cecil, R.B. Firestone, G.M. Hale, D.C. Larson, D.A. Resler, "U.S. Nuclear Data Resources for a Coordinated U.S. Effort in Nuclear Data for Nuclear Astrophysics", http://www.phy.ornl.gov/astrophysics/data/task/taskforce_report.html (1995).
[11] C.E. Rolfs, W.S. Rodney, *Cauldrons in the Cosmos: Nuclear Astrophysics* (University of Chicago Press, Chicago, IL) 1988.
[12] S. Burles, K.M. Nollett, J.W. Truran, M.S. Turner, Phys. Rev. Lett. **82** (1999) 4176.
[13] R.A. Malaney, G.J. Mathews, Phys. Rept. **229** (1993)145.
[14] R.A. Malaney, W.A. Fowler, Ap. J. **333** (1988) 14; **345** (1989) L5.
[15] A.H. Jaffe et al., http://xxx.lanl.gov/abs/astro-ph/0007333 (2000).
[16] K. Kainulainen, H. Kurki-Suonio, E. Sihvola, Phys. Rev. **D59** (1999) 083505.
[17] R.M. Cavallo, A.V. Sweigart, R.A. Bell, Ap. J **492** (1998) 575.
[18] M. Forestini, C. Charbonnel, Astron. Astrophys. Suppl. Ser. **123** (1997) 241.
[19] L. Buchmann, Ap. J. Lett. **468** (1996) L12712.
[20] D. Arnett, *Supernovae and Nucleosynthesis*, (Princeton University, Princeton, NJ) 1996.
[21] B.S. Meyer, Ann. Rev. Astron. Astrophys. **32** (1994) 153.
[22] R.D. Hoffman, S.E. Woosley, T.A. Weaver, T. Rauscher, F.-K. Thielemann, Ap. J. **521** (1999) 735.
[23] A.E. Champagne, M. Wiescher, Ann. Rev. Nucl. Part. Sci. **42** (1992) 39.
[24] S. Starrfield, Phys. Rept. **311** (1999) 371.
[25] R.K. Wallace, S.E. Woosley, Ap. J. Suppl. **45** (1981) 389.
[26] K.M. Vanlandingham et al., Mon. Not. R. Aston. Soc. **282** (1996) 563.
[27] Y. Chin, C. Henkel, N. Langer, P. Mauersberger, Ap. J. Lett. **512** (1999) L143.
[28] M.J. Harris et al., Ap. J. **522** (1999) 424.
[29] M. Hernanz et al., Ap. J. **526** (1999) L97.
[30] M. Wiescher et al., Astron. Astrophys. **160** (1986) 563.
[31] J. Jose, A. Coc, M. Hernanz, Ap. J. **520** (1999) 347.
[32] M. Wiescher, J. Goerres, H. Schatz, J. Phys. **G25** (1999) R133.
[33] P.D. Parker, C. Barnes, L. Buchmann, G. Hale, F. Kappeler, S. Kubono, C.E. Rolfs, M.S. Smith, R. Stokstad, F.-K. Thielemann, M. Wiescher, S.E. Woosley, *Nuclear Astrophysics Data Steering Committee Meeting Minutes*, Feb. 1996 (unpublished).
[34] W.R. Hix, M.S. Smith, A. Mezzacappa, S. Starffield, D.L. Smith, *Proc. 10th Annual Astrophysics*

Conference in Maryland - Cosmic Explosions, 11-13 October 1999, eds. S.S. Holt, W.W. Zhang AIP Press (1999).
[35] R.E. Taam, S.E. Woosley, T.A. Weaver, D.Q. Lamb, Ap. J. **413** (1993) 324.
[36] H. Schatz, L. Bildsten, A. Cumming, M. Wiescher, Ap. J. **524** (1999) 1014.
[37] H. Schatz et al., Phys. Rept. **294** (1998) 167.
[38] M. Wiescher, H. Schatz, A.E. Champagne, Philos. Trans. R. Soc. London **A356** (1998) 2105.
[39] O. Koike, M. Hashimoto, K. Arai, S. Wanajo, Astron. Astrophys. **342** (1999) 464.
[40] K. Arai, M. Hashimoto, Astron. Astrophys. **254** (1992) 191.
[41] G. Fuller et al., Ap. J. **307** (1986) 675.
[42] M. Wiescher et al., Ap. J. **343** (1989) 352.
[43] C. Angulo et al., Nucl. Phys. **A656** (1999) 3; http://pntpm.ulb.ac.be/nacre.htm.
[44] C. Iliadis et al., Ap. J. Suppl. (2000) in press.
[45] T. Rauscher, F.-K. Thielemann, Atomic Data Nuclear Data Tables **75** (2000) 1; http://quasar.physik.unibas.ch/~tommy/reaclib.html#access
[46] R.D. Hoffman, S.E. Woosley, *Stellar Nucleosynthesis Data from the Tables of Reaction Rates for Nucleosynthsis Charged Particle, Weak, and Neutrino Interactions, Version 92.1* (1992) unpublished; http://ie.lbl.gov/astro/hw92_1.html
[47] S. Goriely, in *Proc. Capture Gamma-Ray Spectroscopy and Related Topics*, 30 August – 3 Sept. 1999, ed. S. Wender (American Inst. Physics Conf. Proc. 529, New York) 1999, p. 287; http://www-astro.ulb.ac.be/Html/hfr.html
[48] F.-K. Thielemann et al., Adv. Nucl. Astro. **525** (1987) 1; http://ie.lbl.gov/astro/friedel.html
[49] P.D. Parker, C. Barnes, L. Buchmann, G. Hale, F. Kappeler, S. Kubono, C.E. Rolfs, M.S. Smith, R. Stokstad, F.-K. Thielemann, M. Wiescher, S.E. Woosley, *Reference List for Nuclear Astrophysics Data* (1996), http://ie.lbl.gov/astro/astroref.html (unpublished).
[50] CSISRS: http://www.nndc.bnl.gov/nndc/exfor/
[51]ENDF: http://www.nndc.bnl.gov/nndc/endf/

THE ROLE OF NUCLEAR MODELS IN PROVIDING NUCLEAR DATA FOR ASTROPHYSICS

S. GORIELY

IAA – Université Libre de Bruxelles, Brussels, Belgium

Abstract

Although important effort has been devoted in the last decades to measure decay half-lives and reaction cross sections, major difficulties related to the specific conditions of the astrophysical plasma remain (capture of charged particles at low energies, large number of nuclei and properties to consider, exotic species, high-temperature and/or high-density environments, ...). In many astrophysical scenarios, only theoretical predictions can fill the gaps. The nuclear ingredients to the reaction or weak interaction models should preferentially be estimated from microscopic global predictions based on sound and reliable nuclear models which, in turn, can compete with more phenomenological highly-parametrized models in the reproduction of experimental data. The latest developments made in deriving the nuclear inputs of relevance in astrophysics applications are reviewed. It mainly concerns nuclear structure properties (atomic masses, deformations, radii, etc...), nuclear level densities, nucleon- and α-nucleus optical potentials, γ-ray and Gamow-Teller strength functions. Emphasis is made on the possibility to make use of reliable microscopic models for practical applications.

1 Introduction

Nuclear astrophysics is a vastly interdisciplinary field. There is a large number of different problems invoked calling for a variety of different and complementary research fields (for a review, see [1, 2]). Impressive progress has been made for the last decades in the various fields related to nuclear astrophysics. Factors contributing to these rapid developments include progress in experimental and theoretical nuclear physics, as well as in ground-based or space astronomical observations and astrophysical modellings. In spite of that success, major problems and puzzles remain, which challenges continuously the nuclear astrophysics concepts and findings. To put them on a safer footing requires in particular a deeper and more precise understanding of the many nuclear physics processes operating in the astrophysical environment. Among the numerous objectives in nuclear astrophysics, let us mention some major issues:

- the nature, structure, composition and evolution of objects such as low- and intermediate-mass stars, massive stars, supernovae of type I or II, white dwarfs, neutron stars, novae, X-ray bursts, γ-ray bursts, ...

- the nucleosynthesis of elements lighter than iron, and in particular species like ^{3}He, ^{7}Li, ^{15}N, ^{19}F, ^{23}Na, ^{26}Al, in low- and intermediate-mass stars, massive stars, novae, supernovae of type I or II, ...
- the nucleosynthesis of elements heavier than iron by
  - the slow neutron-capture process (or s-process) in low- and intermediate-mass AGB stars and massive stars,
  - the rapid neutron-capture process (or r-process) possibly, in exploding massive stars or neutron star mergers, and
  - the p-process in the oxygen/neon layers at the presupernova stage or during the stellar explosion of massive stars.

All these scientific questions raise major nuclear challenges that up to now have been addressed in a more or less satisfactory way. Astrophysics needs in nuclear data are defined by the astrophysics modelling and by the sensitivity of the astrophysics predictions to the nuclear input. Depending on the level of reliability of the astrophysics model, a relevant answer about the impact of nuclear uncertainties on the astrophysics observables can be given. Our ability to model astrophysics sites varies with the complexity to describe numerically the corresponding physical mechanisms. Schematically, four classes of models can be defined according to their level of reliability: *(i)* state-of-the-art 3D self-consistent parameters-free models, *(ii)* realistic self-consistent 1D models, *(iii)* parametrized (semi-realistic) 1D models and *(iv)* phenomenological fully-parametric site-independent models. Nowadays, almost no astrophysics simulation is performed on the basis of 3D self-consistent models, and as regards nuclear uncertainties, not a single study has ever been made in such refined calculations. Major modelling difficulties still need to be solved, in particular in the description of convection, radiation and neutrino transport, plasma equation of state, mass losses by stars, rotation and magnetic effects, ... For this reason, in all the still-open nuclear astrophysics questions, nuclear physics must be regarded as a necessary, but not a sufficient condition to solve the problems. This fundamental issue should be kept in mind when estimating the importance of nuclear physics for astrophysics applications. Extensive studies about the impact of nuclear physics, for example, on non-explosive H- and He-burning phases can be found in [3], explosive nucleosynthesis in type II supernovae in [4], s-process nucleosynthesis in [5, 6, 7], r-process nucleosynthesis in [8, 9, 10], nucleo-cosmochronometry in [11, 12, 13], p-process nucleosynthesis in [10, 14, 15].

Strong, weak and electromagnetic interaction processes play an essential role in nuclear astrophysics (for a review, see [2, 16]). Nuclear reactions concern thermonuclear as well as spallation reactions. The thermonuclear reactions taking place in stars include the capture of nucleons or α-particles at relatively low energies (far below 1 MeV for neutrons and the Coulomb barrier for charged particles). They are of pivotal importance for the stellar energy balance, as well as for the bulk composition of the galaxies and for the peculiar abundances observed at the surface of stars of certain classes. The spallation reactions act in low temperature and density media through the interaction of particles (non-thermally) accelerated up to energies in excess of some tens of MeV per nucleon with the interstellar medium, or with the material (gas or grains) at stellar surfaces or in circumstellar shells. β-decay rates, as well as electron

or positron captures are also crucial for our understanding of specific scenarios in stellar evolution (e.g presupernova and supernova models) and nucleosynthesis (e.g the r-process). Although important effort has been devoted in the last decades to measure decay half-lives and reaction cross sections, major difficulties related to the specific conditions of the astrophysical plasma remain, and only theoretical predictions can fill the gaps. Charged-particle induced reactions at stellar energies (far below the Coulomb barrier) have cross-sections that are not too low to be measured at the present time. Stellar reactions often concern unstable or even exotic (neutron-rich, neutron-deficient, superheavy) species for which no experimental data exist. Given astrophysical applications (e.g the r- or p-processes of nucleosynthesis) involve a large number (thousands) of unstable nuclei for which many different properties have to be determined (including ground and excited state properties, strong, weak and electromagnetic interaction properties). In high-temperature environments, thermalization effects of excited states by electron or photon interactions, as well as ionization effects significantly modifies the nuclear properties in a way that it remains difficult or almost impossible to simulate in the laboratory. For example, electron screening in the laboratory conditions complicates the experimental determination of the bare cross-section characterizing the ionised stellar environments. The contribution of thermally populated excited states, as well as atomic effects in the strongly ionised stellar plasma can modify by orders of magnitude the laboratory β-decay and electron capture half-lives. Finally, in high-density environments, the basic definition of the nucleus is lost and nuclear binding must be understood in terms of a nuclear equation of state. For all these specificities found in astrophysical plasmas, theoreticians are requested to supply reliable determination of all relevant quantities to the experimentally unreachable energy and mass regions. For some specific applications (e.g the r-process), no experimental data are available at all.

To fulfill these specific requirements, when estimating the different nuclear inputs for astrophysics applications, two major features of the nuclear theory must be contemplated, namely its *microscopic* and *universal* aspect. A microscopic description by a physically sound model based on first principles ensures a reliable extrapolation away from experimentally known region. On the other hand, a universal description of all nuclear properties within one unique framework for all nuclei involved ensures a coherent prediction of all unknown data. For these reasons, when the nuclear ingredients to the reaction (e.g Hauser-Feshbach) or weak interaction (e.g the Quasi-Particle Random Phase Approximation or QRPA) models cannot be determined from experimental data, use is made preferentially of microscopic or semi-microscopic global predictions based on sound and reliable nuclear models which, in turn, can compete with more phenomenological highly-parametrized models in the reproduction of experimental data. The selection criterion of the adopted model is fundamental, since most of the nuclear ingredients in rate calculations need to be extrapolated in an energy and mass domain out of reach of laboratory measurements, where parametrized systematics based on experimental data can fail drastically. Global microscopic approaches have been developed for the last decades and are now more or less well understood. However, they are almost never used for pratical applications, because of their lack of accuracy in reproducing experimental data, especially when considered globally on a large data set. As we defined different classes of astrophysics model according to their reliability, different classes of nuclear models can be contemplated, starting from local macroscopic approaches up to global microscopic approaches. We find in between these

two extremes, approaches like the classical (e.g liquid drop, droplet), semi-classical (e.g Thomas-Fermi), macroscopic-microscopic (e.g classical with microscopic corrections), semi-microscopic (e.g microscopic with phenomenological corrections) and fully microscopic (e.g mean field, shell model, QRPA) approaches. In a very schematic way, the higher the degree of reliability, the less accurate the model reproduces the bulk set of experimental data. The classical or phenomenological approaches are highly parametrized and often successfull in reproducing experimental data, or at least much more accurate than microscopic calculations. The low accuracy obtained with microscopic models mainly originates from computational complications making the determination of free parameters by fits to experminental data time-consuming. This reliability vs accuracy character of nuclear theories are detailed below for most of the relevant quantities needed to estimate reaction or β-decay rates, namely nuclear masses, nuclear level densities, optical potentials, γ-ray and Gamow-Teller strength functions. As schematized in Fig. 1, phenomenological, as well as microscopic models are available for each of these ingredients. Nowadays, microscopic models can be tuned at the same level of accuracy as the phenomenological models, renormalized on experimental data if needed, and therefore could replace the phenomenogical inputs little by little in practical applications. The needs for further theoretical investigations in each of these fields are also stressed in the following sections.

2 Towards global microscopic predictions

2.1 Prediction of ground state properties

Among the ground state properties, the atomic mass $M(Z, A)$ is obviously the most fundamental quantity and enter all chapters of nuclear astrophysics. Their knowledge is indispensable to estimate the rate and energetics of any nuclear transformation. Although masses for more than about 2000 nuclei are known experimentally [17], important nuclear astrophysics applications, like the r- or p-processes, involve exotic neutron-rich and neutron-deficient nuclei for which no experimental data exist. The calculation of the reaction and decay rates also requires the knowledge of other ground state properties, such as the deformation, density distribution, single-particle level scheme, pairing force, shell correction energies, ... for which nuclear structure theory must provide predictions. The impact of the different mass models on the r- and p-processes predictions are discussed, for example, in [10].

Attempts to develop formulas estimating the nuclear masses of nuclei go back to the 1935 "semi-empirical mass formula" of von Weizsäcker [18]. Being inspired by the liquid-drop model (LDM) of the nucleus, this is the macroscopic mass formula *par excellence*. Improvements have been brought little by little to the original mass formula, leading to the development of macroscopic-microscopic mass formulas [19], where microscopic corrections to the liquid drop part are introduced in a phenomenological way. In this framework, the macroscopic and microscopic features are treated independently, both part being connected exclusively by a parameter fit to experimental masses. Later developments included in the macroscopic part properties of infinite and semi-infinite nuclear matter and the finite range character of nuclear forces. Until recently the atomic masses were calculated on the basis of one form or another of the liquid-drop model, the

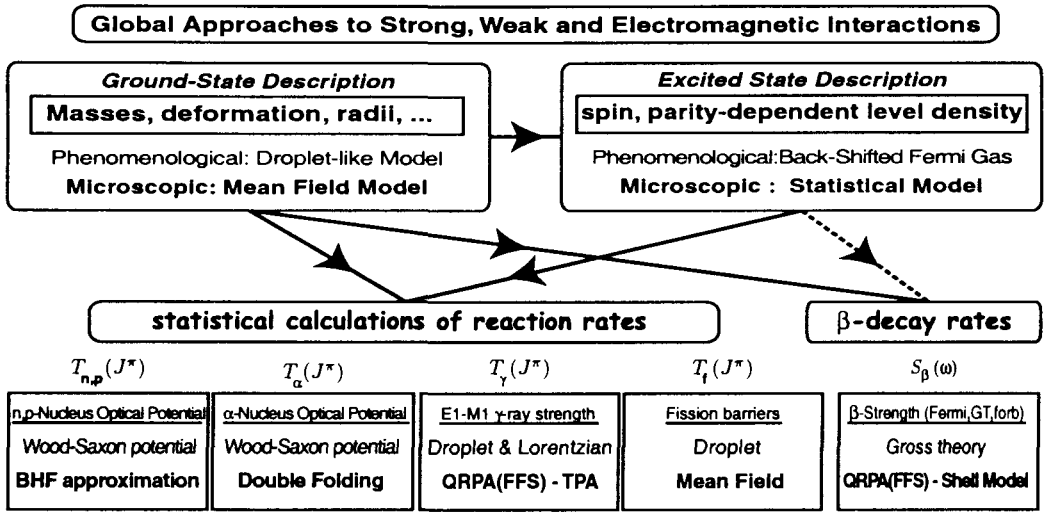


Figure 1: Global phenomenological and microscopic theories used to estimate the ground- and excited state properties, as well as transmission coefficients T and β-strength function of relevance in the Hauser-Feshbach and β-decay rates calculations. Details are given in the text.

most sophisticated version of which is the "finite-range droplet model" (FRDM) [20]. Despite the great empirical success of this formula (it fits the 1888 $Z \geq 8$ masses with an rms error of 0.689 MeV), it suffers from major shortcomings, such as the incoherent link between the macroscopic part and the microscopic correction, the instability of the mass prediction to different parameter sets, or the instability of the shell correction. There is an obvious need to develop, for astrophysics applications, a mass formula that is more closely connected to the basic nuclear interactions [10]. Two such approaches can reasonably be contemplated at the present time, one being the non-relativistic Hartree-Fock (HF) method, and the other the relativistic Hartree method, also known as the relativistic mean-field (RMF) method. Progress in the HF and RMF mass models has been slow, presumably because of the computer-time limitations that arose in the past with deformed nuclei. Nuclear forces are traditionally determined by fitting to the masses (and some other properties) of less than ten or so nuclei. The resulting forces give rise to rms deviations from the 1888 experimental masses well in excess of 2 MeV. This is far from reaching the level of precision found by droplet-like models (around 0.7 MeV).

The result is that the most microscopically founded mass formulas of practical use were till recently those based on the so-called ETFSI (extended Thomas-Fermi plus Strutinsky integral) method [21]. The ETFSI method is a high-speed approximation to the HF method based on Skyrme forces, with pairing correlations generated by a δ-function force that is treated in the usual BCS approach. In the latest version of the ETFSI mass model (ETFSI2), eleven parameters are found to reproduce the 1719 experimental masses of the $A \geq 36$ nuclei with an rms deviation of 0.709 MeV [16] comparable with the one obtained with the droplet-like formula. The ETFSI model remains an approach of the macroscopic-microscopic type, although it provides a high

degree of coherence between the macroscopic and microscopic terms through the unifying Skyrme force underlying both parts. A logical step towards improvements obviously consists in considering now the HF method as such. It was demonstrated very recently [22, 23] that HF calculations in which a Skyrme force is fitted to essentially all the mass data are not only feasible, but can also compete with the most accurate droplet-like formulas available nowadays. The force used in the latest HFBCS mass calculation of [23] is a conventional 10-parameter Skyrme force, along with a 4-parameter δ-function pairing force. The Skyrme and pairing parameters are determined by fitting to the full data set of 1719 $A \geq 36$ masses, leading to an rms error of 0.702 MeV.

The quality of the mass models available is traditionally estimated by the rms error obtained in the fit to experimental data and the number of free parameters. However, this overall accuracy does not imply a reliable extrapolation far away from the experimentally known region in view of the possible shortcomings linked to the physics theory underlying the model. Major progress has been performed recently by replacing the macroscopic-microscopic models by more fundamental approaches, such as HF. The accuracy achieved by the recent HFBCS mass table makes it competitive for pratical applications. However, this development must be regarded as a first step towards more microscopic global approaches. In particular, pairing correlations treated in the BCS approach neglects the fact that the scattering of nucleon pairs between different single-particle states under the influence of the pairing interaction will actually modify the states. This problem is avoided in the HF-Bogolyubov (HFB) method which puts the pairing correlations into the variational function, so that the single-particle and pairing aspects are treated simultaneously and on the same footing. Wigner corrections for nuclei with $Z \simeq N$, as well as rotational corrections for deformed nuclei are so far treated phenomenologically and remained to be described in a more microscopic way. Finally, more fundamentally, mean field models need to be improved, so that all possible observables can be estimated coherently on the basis of one unique effective force. For example, empirical values of the nucleonic effective mass or the Landau-Migdal parameters can be in contradiction with the values deduced from the existing forces. These universality aspects are extremely complicate to reconcile coherently and will most probably be the focus of fundamental nuclear physics research for the coming decades.

2.2 Nuclear level densities

As for the determination of the nuclear ground state properties, until recently, only classical approaches were used to estimate nuclear level densities (NLD) for practical applications. Although reliable microscopic models (in the statistical and combinatorial approaches) have been developed for the last four decades, the back-shifted Fermi gas model (BSFG) approximation–or some variant of it– remains the most popular approach to estimate the spin-dependent NLD, particularly in view of its ability to provide a simple analytical formula. However, it is often forgotten that the BSFG model essentially introduces phenomenological improvements to the original analytical formulation of Bethe, and consequently none of the important shell, pairing and deformation effects are properly accounted for in such a description. Drastic approximations are usually made in deriving analytical formulae and often their shortcomings in matching experimental data are overcome by empirical parameter adjustments. It is well accepted that

the shell correction to the NLD cannot be introduced by neither an energy shift, nor a simple energy-dependent level density parameter, and that the complex BCS pairing effect cannot be reduced to an odd-even energy back-shift (e.g [24]). A more sophisticated formulation of NLD than the one used in the BSFG approach is required if one pretends to describe the excitation spectrum of a nucleus analytically, especially because of the very high sensitivity of NLD to the different empirical parameters. For these reasons, large uncertainties are expected in the BSFG prediction of NLD, especially when extrapolating to very low (a few MeV) or high excitation energies ($U \gtrsim 15$MeV) and/or to nuclei far from the valley of β-stability.

Several approximations used to obtain the NLD expressions in an analytical form can be avoided by quantitatively taking into account the discrete structure of the single-particle spectra associated with realistic average potentials [25]. This approach has the advantage of treating in a natural way shell, pairing and deformation effects on all the thermodynamic quantities. The computation of the NLD by this technique corresponds to the exact result that the analytical approximation tries to reproduce, and remains by far the most reliable method for estimating NLD (despite some inherent problems related to the choice of the single-particle configuration and pairing strength). A NLD formula based on the ETFSI ground state properties (single-particle level scheme and pairing strength) has already been proposed [24]. Though it represents the first global microscopic formula which could decently reproduce the experimental neutron resonance spacings, some large deviations, for example in the Sn region, are found. These deficiencies are cured in the new HFBCS-based model [26] which predicts all the experimental s-wave resonance spacings with an accuracy comparable to the one obtained by the phenomenological BSFG formula. The microscopic NLD formula also gives reliable extrapolation at low energies where experimental data on the cumulative number of levels is available. Furthemore, the microscopic model is renormalized on experimental (neutron resonance spacings and low-lying levels) data to account for the available experimental information. The HF-BCS-based model can now be used in practical applications with a high degree of reliability. NLD's are provided in a tabular form in order to avoid the loss of precision with analytical fits. The complete set of HFBCS-based NLD tables on a large energy and spin grid is available at *http://www-astro.ulb.ac.be.* Important effort still has to be made to improve the microscopic description of collective (rotational and vibrational) effects, and the disappearance of these effects at increasing energies. Coherence in the pairing treatment of the ground- and excited-state properties also needs to be worked out more deeply.

2.3 Optical potential

Due to the specific requirements in astrophysics, the phenomenological potential of Woods-Saxon type have been replaced by the nucleon-nucleus optical potential [27, 28] derived from a Reid's hard core nucleon–nucleon interaction by applying the Brückner–Hartree–Fock approximation. This semi-microscopic potential gives satisfactory results, though some improvements might be required in the low-energy description of the potential and the treatment of deformed nuclei.

Regarding the α-nucleus optical potential, the situation is less optimistic. The very-low energies of relevance in astrophysical environments (far below the Coulomb barrier) make the extrapolation of global potentials quite hazardous as shown by the results in

the ^{144}Sm$(\alpha,\gamma)^{148}$Gd experiment [29]. For these reasons, new global parametrizations of Woods-Saxon [30] or double folding type [31] were proposed in order to take into account the strong energy dependence and nuclear structure effects affecting the imaginary part of the potential at low energies ($E \lesssim 20$MeV). However, experimental data at low energies [scattering data, α-capture or (n,α) cross sections] are scarce making the predictive power of the new parametrizations still uncertain. Cross section predicted with different potentials can differ by one order of magnitude. Much theoretical and experimental work remains to be done in this area.

2.4 γ-ray strength function

The total photon transmission coefficient from a compound nucleus excited state is one of the key ingredients for statistical cross section evaluation. It strongly depends on the low-energy tail of the giant dipole resonance (GDR). In addition to the generalized Lorentzian model [32, 33], improved description of the $E1$-strength function can be derived from the thermodynamic pole approach (TPA) [34], RPA calculations or the theory of finite Fermi systems (FFS) [35]. Dipole transitions to bound states investigated by means of the nuclear resonance fluorescence confirmed the systematic existence of a so-called pygmy $E1$-resonance at energies below the neutron separation energy [36]. Pygmy resonances have been observed in fp-shell nuclei as well as in heavy spherical nuclei near closed shells (Zr, Mo, Ba, Ce, Sn and Pb). The pygmy resonance is associated with the out-of-phase motion of surface neutrons with respect to the neutron-proton core. The total $E1$ strength in the pygmy resonance is small (around a few percent of the total GDR strength), but, if located well below the neutron separation energy, can significantly increase the radiative neutron capture cross-section [33]. The quasi-particle phonon model has been successful in explaining the fine structure and fragmentation of the $E1$ strength and the presence of the pygmy resonance [36]. Nevertheless, much work remains to be done to estimate reliably its systematic impact on the neutron capture rates by exotic neutron-rich nuclei.

2.5 β-decay rates

The knowledge of weak interaction processes is crucial for understanding the early stage of the core collapse of massive stars, and also the nucleosynthesis of the heavy elements by the r-process. Under stellar conditions, the weak interaction rates are dominated by Gamow-Teller (GT) transitions. The presupernova electron capture and β-decay rates for nuclei in the mass range $A = 45 - 60$ were till recently estimated by a GT phenomenological parametrization based on the independent particle model [37]. Modern shell models have been used for a reliable estimate of weak rates. However, the determination of the GT distribution in nuclei requires large shell-model diagonalizations which account for all correlations among the valence nucleons in a major oscillatory shell. For this reason, shell-model calculations are restricted to light or intermediate-mass nuclei (in particular, sd- and fp-shell nuclei) [38].

Regarding the β^--decay of relevance in the r-process nucleosynthesis, different nuclear models have been proposed for practical applications. In the gross theory [39], statistical arguments are used to estimate the smooth energy dependence of the GT and first forbidden strength function. Recent improvements in the so-called semi-gross

theory [40] accounts for the inclusion of shell effects in the parent nucleus and the dependence of the one-particle strength function with the spin and parity of the decaying nucleon. The limitations of the method are essentially related to the neglect of coherent effects due to the effective NN-interaction.

A second simple, though microscopic, approach to nuclear β-decay properties is based on the proton-neutron QPRA. A complete model hamiltonian includes single-particle and pairing components, as well as a schematic separable NN-interaction in the particle-hole (ph) and particle-particle (pp) channels. The most popular version of this model is the FRDM model with a folded Yukawa single-particle potential, a constant BCS pairing and a separable GT interaction in the ph channel [41]. The main advantage of the pnQRPA is a physically sound description of the low-lying structures of the β-strength function. The limitations of the method are mainly due to the use of the first order QRPA, a phenomenological one-body single-particle potential and a simple separable spin-isospin effective NN-interaction. In particular, the latter does not provide a universal treatment of the spin-isospin excitations of different multipolarities.

A fully self-consistent Hartree-Fock-Bogolyubov plus QRPA approach applicable to large-scale calculations of the ground state and β-decay properties has not been achieved yet (mainly for computational limitations). A practical step in this direction is found in the approximate microscopic approach ETFSI plus continuum QRPA [42]. Based on the ETFSI ground state description, the strength function of the charge-exchange excitations and the resulting β–decay rate is calculated within the spherical continuum QRPA with the exact account for the single-particle continuum in the ph channel. In this approach, the mass-independent finite-range effective NN-interaction ensures a universal description of the spin-isospin excitations of arbitrary multipolarity. Fully self-consistent models for spherical and deformed nuclei need to be developed. The impossibility to describe at the moment the ground-state and the spin-isospin excitation with the same value of the Landau-Migdal constant (as extracted from experimental data) has to be investigated in deeper details. In addition, the influence of forbidden transitions and high-order QRPA effects on the β–decay rates still remain to be studied systematically.

3 Conclusions

Although important effort has been devoted in the last decades to measure decay half-lives and reaction cross sections, major difficulties related to the specific conditions of the astrophysical plasma remain (charged-particle capture at low energies, large number of nuclei and properties to consider, exotic species, high-temperature and/or high-density environments, ...). In many astrophysical scenarios, only theoretical predictions can fill the gaps. The extrapolation to exotic nuclei or energy ranges far away from experimentally known regions constrains the use of nuclear models to the most reliable ones, even if phenomenological approaches sometime present a better ability to reproduce experimental data. A subtle compromise between the reliability, accuracy and applicability of the different theories available has to be found according to the specific application considered. Microscopic models can now be adjusted to reach a level of accuracy similar to (or better than) the phenomenological models. Renormalized on experimental data if needed, these microscopic models can replace the phenomenogical

approaches little by little in practical applications. In addition, further investigations should aim at describing reliably and accurately all nuclear properties within one unique framework. This universality aspect of the microscopic predictions corresponds to one of the major challenges of fundamental nuclear physics research for the coming decades.

A continued effort to improve our predictions of the reaction and β-decay rates is obviously required. Priority should be given to a better description of the ground-state properties, nuclear level density and the α-nucleus optical potential, as well as a better understanding of given nuclear effects affecting exotic neutron-rich nuclei, such as the soft dipole modes. Fully self-consistent models for the description of β-decay properties of spherical and deformed nuclei still need to be developed. This continued effort to improve the microscopic nuclear predictions is concomitant with new measurements of masses and β-decay half-lives far away from stability, but also reaction cross sections on stable targets.

Acknowledgments: S.G. is FNRS senior research assistant

References

[1] M.Arnould et al., In: Impact and Applications of Nuclear Science in Europe (NuPECC report) ed. A. van der Woude (1994) p.157.

[2] M.Arnould and K.Takahashi, Rep. Prog. Phys. 99 (1998) 1.

[3] M.Arnould, S.Goriely and A.Jorissen, Astron. Astrophys. 347 (1999) 572.

[4] R.D.Hoffman et al., Astrophys. J. 521 (1999) 735.

[5] F.Käppeler, H.Beer and K.Wisshak, Rep. Prog. Phys. 52 (1989) 945.

[6] S.Goriely, Astron. Astrophys. 342 (1999) 881.

[7] S.Goriely and N.Mowlavi, Astron. Astrophys. 362 (2000) 599.

[8] S.Goriely and M.Arnould, Astron. Astrophys. 312 (1996) 327.

[9] S.Goriely, Astron. Astrophys. 325 (1997) 414.

[10] S.Goriely, Hyp. Int. (2000) in press.

[11] M.Arnould and K.Takahashi, In: Astrophysical Ages and Dating Methods (Editions Frontières) eds. E.Vangioni-Flam et al. (1990) p.325.

[12] S.Goriely and B.Clerbaux, Astron. Astrophys. 346 (1999) 798.

[13] F.Bosch et al., Phys. Rev. Lett. 77 (1996) 5190.

[14] M.Rayet et al., Astron. Astrophys. 298 (1995) 517.

[15] V.Costa et al. Astron. Astrophys. 358 (2000) L67.

[16] S.Goriely, In: Capture Gamma-Ray Spectroscopy and Related Topics (AIP) ed. S. Wender (2000) p.287.

[17] G.Audi and A.H.Wapstra, Nucl. Phys. A595 (1995) 409.

[18] C.F.von Weizsäcker, Zeit. Phys. 99 (1935) 431.

[19] W.D.Myers and W.J.Swiatecki, Nucl. Phys. 81 (1966) 1.

[20] P.Möller et al., At. Data Nucl. Data Tables 59 (1995) 185.

[21] Y.Aboussir et al., At. Data Nucl. Data Tables 61 (1995) 127.

[22] F.Tondeur et al., Phys. Rev. C62 (2000) 024308.

[23] S.Goriely et al., At. Data Nucl. Data Tables **77 (2001), 311.**

[24] S.Goriely, Nuc. Phys. A605 (1996) 28.

[25] P.Decowski et al., Nuc. Phys. A110 (1968) 129.

[26] P.Demetriou and S.Goriely, Nucl. Phys. A **688 (2001) 584; Nucl. Phys. A (2001) in press.**

[27] J.P.Jeukenne et al., Phys. Rev. C16 (1977) 80.

[28] E.Bauge et al., Phys. Rev. C58 (1998) 1118.

[29] E.Somorjai et al., Astron. Astrophys. 333 (1998) 1112.

[30] C.Grama and S.Goriely, In: Nuclei in the Cosmos (Editions Frontières) eds. N.Prantzos et al. (1998) p.463.

[31] P.Mohr, Phys. Rev. C61 (2000) 045802.

[32] J.Kopecky and R.E.Chrien, Nucl. Phys. A468 (1987) 285.

[33] S.Goriely, Phys. Lett. B436 (1998) 10.

[34] Reference Input Parameter Library, IAEA-Tecdoc-1034 (1998) (also available at *http://iaeand.iaea.or.at/ripl*).

[35] S.G.Kadmenskii et al., Sov. J. Nucl. Phys. 37 (1983) 165.

[36] K.Govaert et al., Phys. Rev. C57 (1998) 2229.

[37] G.M.Fuller et al., Astrophys. J. 293 (1985) 1.

[38] K.Langanke and G.Martinez-Pinedo, Nucl. Phys. A673 (2000) 481.

[39] T.Tachibana et al., Prog. Theor. Phys. 84 (1990) 641.

[40] H.Nakata et al., Nucl. Phys. A625 (1997) 521.

[41] P.Möller et al., At. Data Nucl. Data Tables 66 (1997) 131.

[42] I.Borzov and S.Goriely, Phys. Rev. C62 (2000) 035501.

LONG TERM NUCLEAR DATA NEEDS FOR SAFEGUARDS

John W Boldeman, Australian Nuclear Science & Technology Organisation,
Private Mailbag 1, Menai, NSW, 2234, Australia

ABSTRACT

Possible nuclear data needs for the effective application of international safeguards in the next 20 - 25 years are discussed.

1. INTRODUCTION

The Treaty for the Non-Proliferation of Nuclear Weapons (NPT), concluded in 1968, places a number of obligations on signatory states, including: Non-Nuclear-Weapon States (NNWS) agree to forego the development of nuclear weapons or other nuclear explosive devices, and to accept IAEA safeguards on all their nuclear activities to verify that they are used for exclusively peaceful purposes; and the Nuclear-Weapon States (NWS) agree not to assist the NNWS to develop such weapons or devices. The NPT was extended indefinitely by consensus in 1995, with several important principles endorsed by the NPT Review and Extension Conference. One of these was the adoption of the principle that universal adherence to the Treaty is an urgent priority, and all non-parties, particularly States that operate unsafeguarded nuclear facilities, were called on to accede to the Treaty at the earliest date. Another, more relevant to this paper, was the endorsement of the proposals from the IAEA's "Programme 93 + 2" which aimed at substantially strengthening safeguards following political events in the Persian Gulf and elsewhere.

The IAEA's Programme 93 + 2 was a broad based effort that touched on most aspects of IAEA safeguards, with the emphasis on the development of strengthened measures to provide increased assurance regarding the absence of undeclared activities. As before, material accountancy remained the cornerstone of IAEA safeguards but it now has a much wider inspection regime. The strengthened measures include:

- Broad access to information involving expanded declarations from States that include a complete description of their nuclear programme in addition to nuclear material holdings, information on the import-export of certain equipment and material and information from national technical means,
- Broad physical access both to declared locations and to a range of other locations,
- The conduct of unannounced inspections where the State is not given advance notification regarding timing, location and activities associated with an inspection,
- The application of environmental monitoring for the detection of undeclared activities.

As indicated previously, the cornerstone for safeguards has been the concept of materials accounting whereby all materials entering or leaving a specific location or crossing a "materials balance area" boundary are recorded in a materials ledger with the extensions outlined above. In support of materials accounting records, there is a need for materials verification measurements whose accuracy, frequency and timeliness are determined

according to safeguards criteria. These measurements require special instruments which clearly involve the application of nuclear data. To these traditional activities, the process of environmental monitoring and the relevant measurement techniques must now be included.

2. SAFEGUARDS MEASUREMENT TECHNIQUES

Safeguards measurements can be conveniently divided into two classes of techniques - Destructive Assay Techniques (DAT) and Non Destructive Techniques (NDAT). The former, generally applied at major analytical laboratories, e.g. Siebersdorf Analytical Laboratory to verify field measurements, are normally the most accurate but lack timeliness. Typical techniques include isotope dilution mass spectrometry, X-ray fluorescence, alpha counting and gamma ray, neutron resonance absorption and accelerator massspectrometry. Non destructive assay techniques are generally performed in the field by IAEA inspectors to verify data supplied by the operators. The appropriate instrumentation is generally small and portable but often must be fairly sophisticated particularly as the inspectors may be obliged to apply measurement techniques to almost an infinite variety of samples. The figure from Dragnev[1], Figure 1, illustrates the range of NDAT. Table 1 from the review paper by Lammer[2] provides a summary of the traditional safeguards techniques that have made use of nuclear data.

In addressing the question of the needs for nuclear data for effective international safeguards, it is convenient to divide the topic into sub groups

- Nuclear data requirements for the current regime of safeguards activities
- Data requirements for possible changes in the areas in which international safeguards will have to be applied.

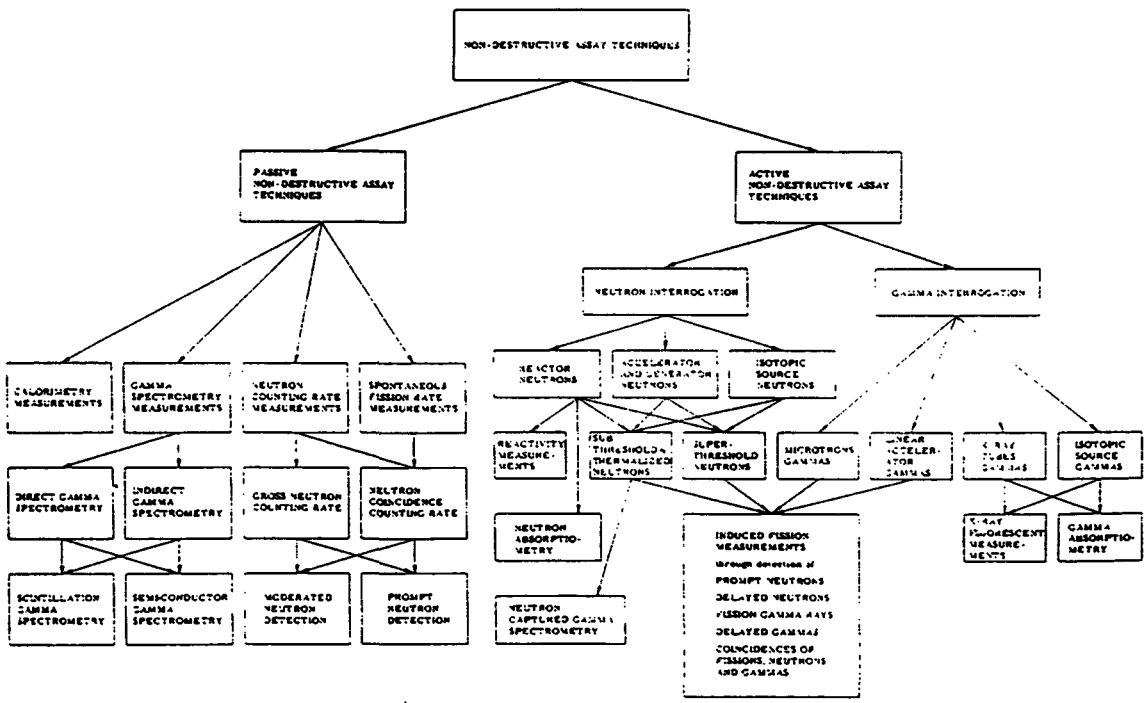


Figure 1 - The structure of non destructive assay measurements (from Ref.1).

TABLE 1

CURRENT SAFEGUARDS METHODS THAT USE NUCLEAR DATA (from Ref.2)

Method	Nuclear Data	Purpose
(1) Fresh Fuel Assay		
Gamma spectrometry of recycled U fuel	Half-life of U-232, γ-ray energies and intensities of its daughter products	Correction for interference of U-232 daughters with U-235 γ-rays
Gamma spectrometry of Pu containing fuel	Half-lives of Pu isotopes and Am-241, γ-ray energies and intensities of their α-decay daughters	Quantitative analysis of the γ-ray spectrum of Pu containing fuel
Active neutron interrogation (standards)	Library of yields, half-lives, γ-ray energies and intensities of FP's	Investigation of activation, build-up in calibration standards
Coincident counting techniques	Prompt γ and prompt neutron multiplicity distributions from fission of U-235, Pu-239, (Pu-141) and spontaneous fission of Pu-238, 240, 242. Possibly delayed neutron yields as a function of time (induced fission)	Optimisation of coincident counting instrumentation layout
X-ray fluorescence	X-ray energies and intensities of Th, U, Pu	Spectrum analysis
(2) Spent Fuel Assay		
FP γ-ray spectroscopy	Thermal fission yields of Zr-95, Ru-106, Cs-133, Cs-137, Ba-140, Ce-144, Eu-153 from U-233, U-235, Pu-239 (Pu-241). Half-lives and γ-ray intensities of Zr-Nb-95, Ru-Rh-106, Cs-134, Ca-137, Ba-La-140, Ce-Pr-144, Eu-154; capture cross-sections of Ca-133, Eu-153	γ-spectrum analysis, interpretation of measured activities and their ratios
Passive neutron assay	Pu-238, 239, 240, 242, Am-241, Cm-242, 244: α-decay and spontaneous fission half-lives, $\bar{\nu}$ for spontaneous fission; fission and capture cross-sections also for U-238, Pu-241, Am-242, Cm-243 and half-lives for the last 3 nuclides; O^{18} (α,n) cross section	Calculation of neutron emission from irradiated fuel for a better understanding of the method and interpretation of the results
(3) Dissolved Fuel (reprocessing plant)		
Isotope correlations	Fissions and capture cross-sections of U-234, 235, 238, Pu-238 to 242; cumulative fissions yields from U-235, Pu-239, Pu-241; capture cross sections for: Kr-82 to 84, 86, Xe-131 to 136, Nd-143 to 146; half lives of Xe-133, 135	Help to resolve discrepancies between measured and calculated correlations

3. NUCLEAR DATA REQUIREMENTS FOR CURRENT SAFEGUARDS APPLICATIONS

It can be seen from Figure 1 and Table 1 that one of the principal nuclear data requirements for safeguards applications is nuclear decay data. Nuclear decay data are generally more than adequate for current safeguards applications and the use of calibrated reference standards reduces even further any need for an improvement in the accuracy. The priority here within the safeguards research community is focussed on the development of more sophisticated detector systems and recording apparatus. The second major area where nuclear data are needed is in neutron measurements. As before the current nuclear data are generally more than adequate, however, in a small, yet important number of applications, generally where fairly complex measurements are required, improved nuclear data can contribute to the effectiveness of safeguards.

During the last decade and a half, there was some evidence to suggest that the efficiency and accuracy of safeguards instrumentation would benefit from an improvement in the quality and the application of selected nuclear data. Accordingly, steps were taken, particularly within the NDS, to determine which nuclear data were most important in safeguards applications and subsequently, to produce a comprehensive handbook[3,4,5] which listed the best evaluated data for those parameters which were in common use in safeguards applications or could be considered useful in inspection activities. Other studies[6-8], including evaluations, suggested that better data could be of value in the critical area of neutron coincidence counting.

3.1 Neutron Coincidence Counting

Neutron coincidence counting is the technique that is generally employed in the non-destructive assay of bulk quantities of nuclear material and in particular, quantities of plutonium. Figure 2, taken from the review article by Menlove[9], illustrates the range of application of the technique. These have been extended in recent times following disarmament agreements and the transfer of some strategic nuclear weapon material into the safeguards regime.

The technique makes use of the multiplicity of neutrons emitted in the fission process whether from spontaneous fission in passive systems or from neutron induced fission in active systems. In the application of neutron coincidence counting, the sample is surrounded by a moderating medium incorporating neutron sensitive detectors. The neutrons are emitted in the fission process within 10^{-16}s of fission, however, the detection process generally requires moderation of the neutrons and their detection in time following fission is determined principally by the lifetime of the neutron in the system comprising the detector and sample. Most neutron coincidence counting systems employ a number of ^{3}He detectors embedded in, typically, polyethylene moderators. Recently, systems involving proton recoil and scintillator detectors have been studied. The principal problems restricting the accuracy of neutron coincidence counting systems include matrix effects, (α,n) backgrounds and multiplication caused by both fission neutrons from the primary event and by the (α,n) background. Since the first application of NCC, there have been many efforts internationally to improve the performance of neutron coincidence counters and to extend its applicability[10-14]. Some of these have concentrated on increasing the efficiency of the detectors so that more of the neutrons are detected per fission event leading to higher multiplicities, effectively more data, and, as a consequence, the opportunity to correct for some of the multiplication effects. To assist this improvement in the application of NCC and its family of

techniques, the nuclear data community committed some resources to the improvement of the relevant nuclear data. An important contribution was a series of independent evaluations of the neutron emission probabilities, P_ν for spontaneous and thermal neutron fission, some new measurements and the identification of areas where the nuclear data could certainly be improved.

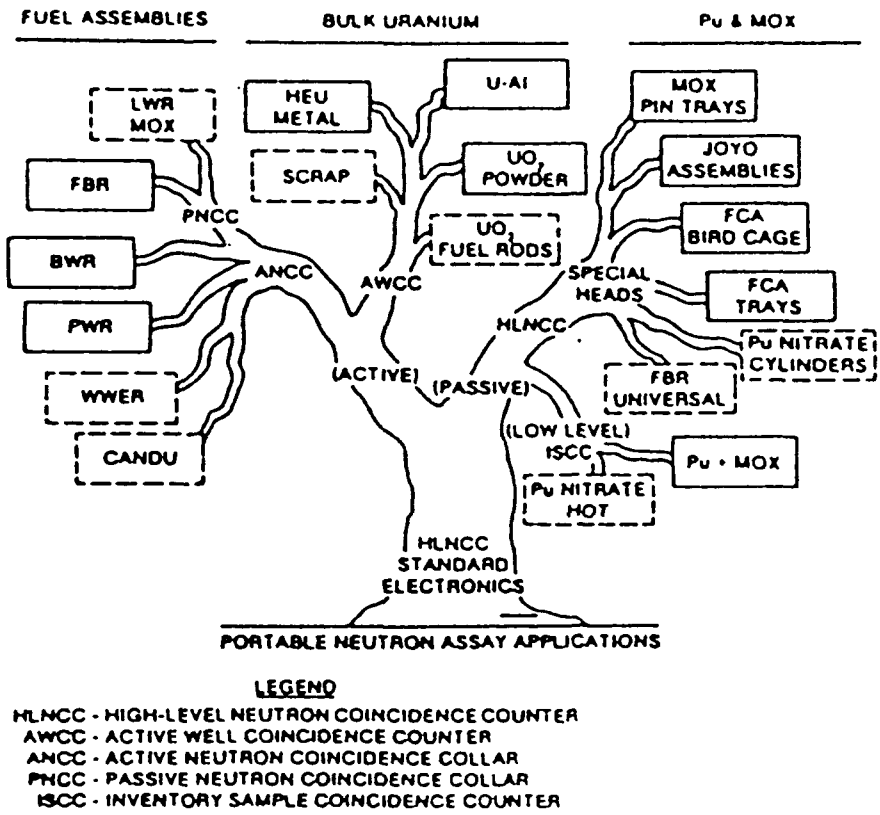


Figure 2 - "Family Tree" diagram of active and passive neutron coincidence systems and applications based on the standard shift-register electronics package developed for the HLNCC (from Ref.9).

In principle, the assay of plutonium in neutron coincidence counting proceeds as follows. The isotopic composition of the sample is determined by gamma ray spectroscopy and the amount of ^{240}Pu is determined by neutron coincidence counting. Although in practise the amount of plutonium is not determined absolutely from a combination of the experimentally determined spontaneous fission half life and the neutron emission probabilities, it was considered that as instruments became more sophisticated rate accurate data for these parameters would lead to intrinsically better detector systems. Therefore, a very accurate measurement was made of the spontaneous fission half-life of ^{240}Pu, and, as indicated previously, comprehensive evaluations were made of the neutron emission probabilities. Many samples also contain ^{242}Pu and less significant amounts of

other spontaneously fissioning isotopes. The concept of effective equivalent amounts of ^{240}Pu for these isotopes are needed. A reasonably accurate way to determine these parameters would be through a combination of the spontaneous fission half-lives of the isotope relative to ^{240}Pu and a similar comparison of the P_ν distributions for these isotopes. As it happens, the one precision measurement[15] of the P_ν distributions for ^{240}Pu also had equivalent precision for those parameters for the spontaneous fission of ^{242}Pu.

The techniques above are called passive methods. For the measurement of isotopes where there is not an inclusion of an isotope with a significant spontaneous fission yield of time-correlated neutrons, active methods involving the use of externally generated neutrons such as ^{252}Cf sources, Pu(Li or Be) sources or neutron generators are used to induce fission in the sample under measurement. The detection can involve gamma ray counting as well as neutron counting and delayed coincidence techniques can be useful with for example the ^{252}Cf sources. The application of these techniques is most appropriate for the measurement of quantities of highly enriched uranium. A variety of names have been applied to such devices and include active well coincidence counters, Cf shufflers (working on delayed fission neutrons), Fast Active Neutron Coincidence Counters[16-18].

Not all of the improvements in nuclear data that were considered relevant to NCC and similar techniques have been addressed and there are several areas where it is still considered that improvement in the data would be valuable. A great deal of current development in this area of measurement involves the application of the techniques to so-called difficult to measure materials[16]. Some of the relevant data that could be improved include those listed below.

(a) Despite the efforts of evaluators and reviews of previous measurements, better information for the prompt fission neutron multiplicity distributions, P(ν), for neutron induced fission of ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{235}U and ^{238}U as a function of the energy of the fission inducing neutron are required. Because the moderating systems are limited in size, the fast fission component of the multiplication process tends to be significant and therefore improved data here would improve the Monte Carlo calculations of detector response. Zucker and Holden have pointed out that there is only one available data set for $n+^{235}U$, $n+^{238}I$ and $n+^{239}Pu$ between 1.36 and 25 MeV and this set was unpublished, possibly because of poor statistics. The data for thermal neutron fission are in excellent shape following extensive evaluations.

(b) The P(ν) data for ^{238}Pu and ^{238}U effectively do not exist and corrections for these elements are required in modern instruments.

(c) As the applicability of neutron coincidence counting increases in its scope, it is now feasible to consider the separation of Pu and Cm isotopes. To do this, some data for P(ν) distributions for the spontaneous fission of ^{242}Cm and ^{244}Cm would be needed.

(d) Improved six group decay constants and delayed neutron yields for ^{234}U and ^{236}U and Np isotopes are required for background corrections.

(e) As indicated previously, a major source of background in neutron coincidence counting derives from (α,n) reactions of the αs from the Pu isotopes with light contaminant isotopes such as F, Na, Mg and Cl. Improved data on the energy dependence of the (α,n) reaction rate for such isotopes is required.

In general, the NCC instrument makers and the users of these instruments do not see any problems with the data as reference material standards are extensively used in the industry.

3.2 Environmental Monitoring

The concept of environmental monitoring relies upon the assumption that undeclared nuclear activities will result in the routine or accidental release of radionuclides and other material into the environment. The subsequent identification of these signatures could then be used as an indication of such an undeclared facility. The measurement of these signatures requires very high sensitivity trace element detection methods such as accelerator mass spectrometry[20-23]. At present, it is not possible to visualise any need for more accurate nuclear data in this area since the measurements aim to provide essentially a signature of undeclared activities. However detection methods rely on the appropriate choice of charge states in the ion sources of the accelerator systems. For standard activities, the use of reference standards and long experience with the techniques means that issues of charge state are not important. However, for non standard activities, a better understanding of charge state distributions could be useful.

4. POTENTIAL LONG TERM NEEDS FOR NUCLEAR DATA

The long term needs for nuclear data for safeguards applications depend entirely on how nuclear and for that matter fusion power develop in the future. Certainly diminishing world oil supplies, their increased cost, the aftermath of the Kyoto Protocol and potential carbon credits etc combined with increasing efforts to make nuclear power more socially acceptable, all suggest that there may be a new dawn for nuclear power generation. Furthermore, if note is taken of limited low cost world uranium resources there should also be increasing efforts to utilise as efficiently as possible uranium resources. These questions of the role of emerging nuclear power options and the nuclear data requirements for them are addressed in detail in other parts of this meeting. These of course are relevant from the safeguards perspective however the principal interest should be on how this effects safeguard strategies and the instruments that are used to verify materials etc.

As a guide to possible new nuclear power activities, reference has been made to many papers at the recent Tenth International Conference on Emerging Nuclear Energy Systems[24]. Some of the prospective power sources discussed there include

- Accelerator driven nuclear power systems
- Accelerator driven transmutation
- New concepts in fast reactors e.g BREST reactor, Super-PRISM Reactor
- U^{233} - Th thermal breeder reactors.

In addition, there are possible variants of existing nuclear fuel arrangements such as

- Proliferation -resistant Fuels
- Radkowsky Thorium Fuel Concept

Finally, there new approaches to reprocessing methods which include

- Co-processing of FBR material

- DUPIC process
- Pyro-electro-chemical reprocessing.

A review of these options indicates that there would be substantial changes in the fuel and moderating systems of the next generation of nuclear based power sources. In the fuel stocks etc of new generation nuclear power sources the range of isotopes extends from thorium isotopes to perhaps beyond the curium isotopes. Therefore appropriate accurate nuclear data must be in place to ensure that, for the safeguards regime, a reasonable picture can be held of the range of isotopes that might be in samples that require measurement to verify adherence to international safeguards.

As before, the nuclear decay data for all of the isotopes referred to above are in excellent shape and are adequate for any perceived safeguard application. For verification of strategic material in bulk or enclosed samples etc., it is not likely that there will be any substitute for neutron coincidence counters (either active or passive) in the future. The possible data improvements referred to previously for neutron coincidence counting are likely to be more pressing in this prospective environment and reinforce the recommendations listed previously. Furthermore, the α backgrounds will also certainly be higher and for these detectors to achieve a satisfactory accuracy appropriate corrections must be made for the induced background. Thus some improvement in the data for (α, n) reactions with light element components of the sample could be required.

5. SUMMARY AND RECOMMENDATIONS

In general, the current state of nuclear data is adequate for the present regime of safeguards activities with only a modest number of areas where improved data might ultimately prove useful. Therefore there is no need to convene any activity specifically directed at the generation or evaluation of nuclear data for current safeguards applications. The series of handbooks on relevant nuclear data for safeguards are an excellent resource and should be updated in the future at appropriate intervals as new information becomes available.

For the long term future, it is clear that there are many new emerging nuclear power systems that could be quite different from those currently in operation. The safeguards requirements for these facilities will need to be addressed by the safeguards community. The nuclear data community can contribute to this process by being vigilant in following the development of these new technologies and their implication for international safeguards so that appropriate evaluations and possibly new measurements can be recommended to satisfy any deficiency.

ACKNOWLEDGEMENTS

The author is indebted to Dr V. Bragin, Mr J. Carlson, Dr N. Ensslin and Dr M. Lammer, for extremely valuable advice for the preparation of this document.

REFERENCES

1. T. Dragnev, "Non Destructive Assay Techniques for Nuclear Safeguards Measurements". At.*En. Review,* **11** 341 (1973).

2. M. Lammer. "Nuclear Data for Safeguards", INDC/P(81)-24 (1981).
3. M. Lammer and O. Schwerer, *Handbook of Nuclear Data for Safeguards,* INDC(NDS)-**248** (1991).
4. N. Kocherov, M. Lammer and O. Schwerer, INDC(NDS) –**376** (1997).
5. V. Goulo, private communication.
6. N. Holden and M. Zucker, "Prompt Neutron Emission Multiplicity Distribution and Average Values (Nubar) at 2200 ms^{-1} for the Fissile Nuclei", *Nucl. Sci. and Eng.,* **98**, 174 (1988).
7. M. S. Zucker and N.E. Holden, BNL-48109 (1993).
8. J. Boldeman, *"Nuclear Data for safeguards and a Possible Comprehensive Test Ban Treaty"*, Proc. Int. Conf. On Nuclear Data for Science and Technology, Gatlinburg, USA, 2, 1108 (1994).
9. H.O. Menlove, "Standardization of Portable Assay Instrumentation - the Neutron Coincidence Tree", *Proc. Fifth Annual Symposium on Safeguards and Nuclear Material Management,* ESARDA, Versailles, France, April (1983).
10. D.G. Langner, N. Dytlewski and M.S. Krick, "Pyrochemical Multiplicity Counter Development", (1992).
11. N. Dytlewski, M.S. Krick and N. Ensslin, "Measurement Variances in Thermal Neutron Coincidence Counting", *Nucl. Instr. and Methods,* **A327**, 469 (1993).
12. L. Bondar, "Passive Neutron Assay by the Euratom Time Correlation Analyser", *International Symposium on International Safeguards,* Vienna, 14-18 March (1994).
13. J. E. Stewart, H. O. Menlove, D. R. Mayo, K. E. Kroncke, F. A. Duran, B. S. Cordova, D. G. Langner and C. D. Rael, "The Epithermal Neutron Multiplicity Counter (ENMC); faster Plutonium Assays by a Factors of 5 - 20", *Proc. of the Inst of Nuclear Materials Management - 40th Annual Meeting,* Phoenix, Arizona, to be published, (1999).
14. D. R. Mayo, N. Ensslin, W. H. Geist, R. F. Grazioso, A. S. Heger, H. Y. Huang, M. C. Miller, A. P. Romano and P. A. Russo, "Design of a Fiber/Scintillator Neutron-Capture Counter to Assay Impure Plutonium", *Proc. of the Inst of Nuclear Materials Management - 40th Annual Meeting,* , Phoenix, Arizona, to be published, (1999).
15. J. W. Boldeman and R. L. Walsh, "Prompt Nubar Measurements for the Spontaneous Fission of ^{240}Pu and ^{242}Pu", *J. of Nuclear Energy, 22,* 63.
16. S. Croft, P. M. J. Chard, I. G. Hutchinson and D. J. Lloyd, "Design and Performance of the Harwell N33 Active Well Coincidence Counter", *Proc. of the Inst of Nuclear Materials Management - 36th Annual Meeting,* Palm Desert, California, p 1135 (1995).
17.
18. W. H. Geist, N. Ensslin, L. A. Carillo, M. S. Krick, D. R. Mayo and P. A. Ruso, "Initial Results of Fast Active Neutron Coincidence Measurements", *Proc. of the Inst of Nuclear Materials Management - 40th Annual Meeting,* , Phoenix, Arizona, to be published, (1999).
19. N. Ensslin, Private communication.
20. G. Andrew, "Prospects for Environmental Monitoring in International Safeguards", *Proc. International Symposium on International Safeguards,* Vienna, 14-18 March (1994).
21. K.W. Nicholson, C.L. Rose, J.A. Garland, W.A. McKay and LR. Pomeroy, "Environmental Sampling for the Detection of Undeclared Nuclear Activities", *AEA* FS 0241(H) (1994).

22. N.A. Wogman, R.W. Perkins and G.R. Holdren, "Environmental Sampling and Analysis as a Safeguards Tool", *Proc. International Symposium on International Safeguards,* Vienna, 14-18 March (199.1).

23 M. A. C. Hotchkiss, D. Child, D. Fink, G. E. Jacobsen, P. L. Lee, N. Mino, A. M. Smith and C. Tuniz, 'Measurement of ^{236}U in Environmental Media", *Nuclear Instruments and Methods in Physics Research,* **B 172,** 659 (2000).

24. The Tenth International Conference on Emerging Nuclear Energy Systems, Petten, The Netherlands, September, 2000.

AGM Nuclear Data Needs, IAEA Vienna, 28 Nov-1 Dec 2000
Session "Nuclear Safeguards and Related Applications"

NUCLEAR DATA NEEDS FOR MATERIAL ANALYSIS

Gábor L. Molnár
Institute of Isotope and Surface Chemistry, Chemical Research Centre, H-1525 Budapest, P. O. Box 77, Hungary

Abstract

Nuclear data for material analysis using neutron-based methods are examined. Besides a critical review of the available data, emphasis is given to emerging application areas and new experimental techniques. Neutron scattering and reaction data, as well as decay data for delayed and prompt gamma activation analysis are all discussed in detail. Conclusions are formed concerning the need of new measurement, calculation, evaluation and dissemination activities.

New trends in material analysis

Neutron-based methods are very powerful tools for characterising the physical and chemical properties of materials. The methods of neutron scattering, diffraction and reflectometry reveal various physical properties. On the other hand, methods based on neutron absorption-induced radiations elucidate the chemical composition. In the latter category belong delayed-gamma neutron activation analysis (NAA), prompt-gamma activation analysis (PGAA), as well as neutron depth profiling (NDP).

There is a continuous need for improvement of the available nuclear data, thanks to emerging application areas and new experimental techniques. Such new frontiers of material analysis are the areas of environment and biology. To the most important new experimental techniques belong the methods utilising intense beams of cold neutrons from nuclear reactors, e.g., cold neutron-induced PGAA, NDP, and neutron reflectometry. Resonance capture of neutrons in the 10-100 eV energy range is also gaining importance due to the extremely high selectivity of this process, and its direct relevance to the transmutation of nuclear waste. Therefore, in the followings special attention will be paid to the data needs of those emerging applications.

Available data

In this chapter, the basic nuclear data available for neutron-based techniques of material analysis are reviewed. For any kind of activation analysis, the basic quantities are the cross-sections characteristic of the nuclear interaction producing the activity, and the gamma-ray energies and emission probabilities of the activation products. The energies are used to qualify, while the product of cross-section and emission probability to quantify the element

(or more precisely, its isotope) in a sample. There are other important parameters, such as decay half-life, which refine the picture. For neutron scattering, the information is carried by the coherent (and incoherent) neutron scattering lengths, rather than the cross-sections, as their sign plays a role in the interference of neutron waves. Besides, magnetic moments of nuclei are important for the study of magnetic structures.

Cross-sections

For neutron activation analysis, the relevant quantities are the thermal neutron-capture cross section, σ_0, and the resonance integral, RI, entering the ratio, $Q_0=RI/\sigma_0$, frequently used for correction of epithermal activation. The evaluation by Mughabghab et al. [1,2] is still the recommended source of information, although it is quite old and contains many data of low accuracy. In half of the cases the uncertainties on σ_0 exceed 10%, and there are only a few examples when the accuracy is better than 0.5%. Although several revisions [3] have been made, the original publication is still the only one giving detailed enough information on the kind of absorption reaction - (n,γ), (n,p), (n,α), (n,f) -, and the type of neutron spectrum. It also provides cross-section values for the activation of individual isomeric states. The most up-to-date and easily available source of the Mughabghab cross sections and resonance integrals is the NuDat database [4], available on the Internet. For certain nuclides, the thermal cross-section deviates from the *1/v* law due to the existence of a low-energy resonance. The correction is given by the Westcott *g*-factor. Even if their data are partly out of date, the compilation by Gryntakis *et al.* in ref [5] is still a useful reference concerning thermal activation cross sections, g-factors and resonance integrals, as well as effective resonance energies calculated by F. De Corte for 128 target isotopes. A more detailed discussion can be found in a recent review article [6], covering data for activation analysis.

Fast neutrons also induce inelastic scattering and other reactions. The cross-sections for these have characteristic energy dependence and a well-defined threshold. The situation with directly measured data is best for 14 MeV neutrons [5,7]. As to other neutron reactions, still the most comprehensive source is the old IAEA handbook on nuclear activation data [5], followed by a more specialised handbook on data for borehole logging [8]. More recent calculated values for thermal, Maxwellian average, fission-spectrum average and 14-MeV cross sections, as well as resonance integrals are available in tabular form, but without information on the uncertainties [9]. The same is true for a recent source of calculated energy-differential capture cross sections [10]. The old "Barn Book" with plotted experimental energy differential cross-section data [11] is still in use. More recent data can be viewed together with various evaluations (ENDF-6, JENDL, BROND, etc.) so far only electronically [12]. Another resource of electronic tables and plots is the KAERI Table of the Nuclides [13], which is restricted to evaluated data. A most recent handbook [14] presents experimental data and evaluated curves together for photonuclear reactions on 164 isotopes.

The evaluation for bound coherent neutron scattering lengths has recently been updated [15]. It should be used instead of the older reference by Sears [16], although the latter resource is complete with incoherent scattering lengths as well.

Decay data

The most comprehensive source of decay data is the ENSDF file [17]. It contains the adopted decay schemes and half-lives, the energies, relative and absolute intensities of the

gamma radiations, as well as the total intensities including electron conversion (when applicable) for the various decay branches of radionuclides.

For the radionuclides of interest in NAA, most of the decay half-lives are known with sufficient precision. The ENSDF values are included in the periodically updated Nuclear Wallet Cards [18] and in NuDat [4]. For about two thirds of the 66 elements involved in NAA there is at least one radionuclide with a half-life accurate to at least 0.1%, see ref. [6] for details. On the other hand, fewer than one tenth of the 130 radionuclides (including isomers) considered there have a half-life less accurate than 1%.

Gamma-ray energies are used for element (nuclide) identification, or qualitative analysis, and the relative intensities of the transitions are helpful when correcting for spectral interference. Accurate absolute intensities or, in other words, photon emission probabilities enable quantitative analysis by the absolute method, provided cross sections and other parameters are also known with a sufficient accuracy. While the energy values from ENSDF [17] are usually very accurate, this is not always so for the absolute gamma-ray intensities. The relative uncertainty of normalisation is very often several percent, and sometimes even larger than ten percent. This uncertainty is added, of course, to the cross-section uncertainty quadratically when calculating the activity.

In NAA, the product of cross-section and emission probability is the important quantity, hence attempts have been made to measure a nuclear constant – called k_0-factor – proportional to this product directly with an accuracy of a few percent. A comparison between measured [19,20,21] and calculated k_0-factors has shown that there may be problems even with some of the directly measured k_0-factors, see ^{24}Na and other examples in ref. [6].

For PGAA, one has to look into to the ENSDF file [17] for thermal neutron capture gamma-ray data. Until recently, such data were available only for nuclides with mass number greater than 44. While the energies are mostly accurate, the intensity data are in most cases unsuitable for analytical work. The normalisation of absolute intensity is rarely more precise than 10% relative standard deviation, and sometimes it is even lacking. Probably for this reason, in a compilation of ENSDF capture data by Tuli [22] only relative intensities have been included uniformly, and all uncertainties have been omitted. Besides, the Tuli table is nuclide oriented and the contributions of the individual isotopes of a given element have to be combined by involving elemental abundances and cross sections. The only prompt gamma-ray library for elements is still the old compilation by Lone et al. [23]. However, it does not meet present-day standards, in that the data are old and of poor quality, with contaminant peaks (e.g., Cd) admixed to many elemental sets. Moreover, important low-energy gamma rays are missing. Only the ranges of uncertainty is indicated for the gamma-ray energies, and the values are rather large, sometimes above 3 keV, for high-energy gamma rays. Moreover, the uncertainties for absolute emission probabilities have not been given explicitly, they have been estimated to be below 20%. Very recently, that high-quality gamma ray data have appeared [24] and the prompt k_0 method has also been implemented for PGAA [25,26].

Gamma-ray spectrum libraries for various detectors, especially semiconductor detectors, are much needed for designing and conducting an experiment in activation analysis. Radioactive decay spectra for NAA [27,28] and (n,n'γ) spectra for reactor fast neutrons [29] are the only available resources.

Data needs

There is a definite need for more data or just better data in the areas discussed above. Data are almost entirely lacking for cold neutron capture gaining importance rapidly in PGAA, and for resonance capture in the 20-120 eV energy range. More data are needed for fast neutron reactions as well, for borehole logging and illicit trafficking applications.

One needs specifically

- catalogues of prompt γ-ray spectra for cold, thermal, resonance (10-100 eV) and fast neutrons
- low energy cross-sections for non-1/v nuclides (only a few are measured)
- standards to characterise cold neutron beams

In many cases, improvement of the existing data is necessary by doing new evaluations and measurements, depending on the case. Here we try to summarise the most important problems communicated to us.

Cross-sections

- remeasure some thermal activation and absorption cross-sections, e.g., $^{93}Nb(n,\gamma)^{94m}Nb$, $^{23}Na(n,\gamma)^{24m}Na$
- better I_0, Q_0 values, especially for $^{114}Cd(n,\gamma)^{115}Cd$
- improve thermal, epithermal spectrum characterisation (Westcott convention)
- re-evaluate neutron resonances
- re-evaluate some neutron scattering lengths (incoherent)
- re-evaluate data for interfering fast neutron reactions
- remeasure fast neutron cross-sections for $^{16}O(n,n')$, $^{12}C(n,n')$, etc.

Decay data

- remeasure , re-evaluate half-lives (short-lived: ^{24}Na, ^{76}As; long-lived: ^{14}C)
- determine missing absolute normalisations and their uncertainties for decay γ rays (^{238}U chain: ^{234m}Pa, ^{214}Pb, ^{214}Bi)
- remeasure k_0-factors for NAA (90% of data from 2 laboratories)
- remeasure k_0-factors for PGAA (most from 1 laboratory only)
- use k_0-factors for consistency check of activation cross-sections

Fission data

- improve accuracy of individual fission yields for short-lived products (^{134}Te, ^{132}I, etc.)
- improve data on the yield and spectral composition of delayed neutrons (for U, etc. determination)

Data dissemination needs

Specialised databases or specifically designed interfaces for retrieval are needed for material analysis. Those databases should contain

- all relevant data in one place
- all uncertainties

Whichever solution is realised, traceability of the data source should be maintained. As to the dissemination format, both hardcopy and electronic form remain necessary. The continuation of IAEA handbooks is of vital importance. For instance, a new reference library for neutron activation has long been expected [30]. As to electronic formats, Web and CD-ROM have their own merits, hence both remain needed. On the other hand. it would be desirable to abandon printer-oriented formats in favour of computer readable formats, such as spreadsheet or popular computer code inputs (MCNP, etc.).

Summary and conclusions

The status nuclear data for material analysis with neutron-based methods has been reviewed with respect to current research trends and specific needs expected in the future. It is evident, that environment and biology are becoming increasingly important application areas. As to emerging experimental techniques, cold neutron methods, such as PGAA and NDP, are gaining importance rapidly. Improvements on nuclear data have to be made urgently to meet these challenges.

There is a need for new measurements of prompt γ-ray spectra, anomalous cold neutron cross-sections, some fast neutron cross-sections and cold as well as thermal neutron k_0-factors. The same applies to decay half-lives of some extremely short- and long-lived nuclides, respectively. Individual fission yields for short-lived products, and characteristics of delayed neutron emitters should be remeasured with higher precision. New evaluations for thermal neutron cross-sections, neutron resonances and incoherent scattering lengths are called for. New methods imply new data needs. For cold neutron PGAA, reference standards are needed to characterise the spectrum of the cold neutron beam.

New concepts have to be developed in data dissemination. Easy availability and traceability of data, and handy ways of retrieval are the most important criteria. There are some good examples already, see refs. [12,31,32].

Finally, the author would like to thank the numerous comments and suggestions from a number of colleagues. Special thanks are due to A. V. Andreev, V. R. Burmistrov, G. Gorini, W. D. James, A. Jehouani, G. Kennedy, G. Krexner, R. M. Lindstrom, S. Mukhammedov, T. M. Nakanishi, A. Simonits, B. Smodis, J. Uyttenhove, T. Weizhi, G. Wermann, R. D. Wilson, and R. Zeisler.

References

1. S.F Mughabghab, M. Divadeenam and N.E. Holden, Neutron Cross Sections from Neutron Resonance Parameters and Thermal Cross Sections, Vol 1, Part A, Z=1-60; Academic Press, New York, 1981; http://isotopes.lbl.gov/isotopes/ngdata/sig.htm
2. S.F. Mughabghab, Neutron Cross Sections, Vol. 1, Neutron Resonance Parameters and Thermal Cross Sections, Part B, Z=61-100, Academic Press, New York, 1984; http://isotopes.lbl.gov/isotopes/ngdata/sig.htm
3. N. E. Holden, Neutron Scattering and Absorption Properties (Revised 1996), in CRC Handbook of Chemistry and Physics, 78th edition, CRC Press, Boca Raton, 1997-1998, p. 11-147.

4. R. R. Kinsey, The NuDat Program for Nuclear Data on the Web, National Nuclear Data Center, Brookhaven National Laboratory; http://www.nndc.bnl.gov/nndc/nudat/
5. Handbook on Nuclear Activation Data, Technical Report Series No. 273, International Atomic Energy Agency, Vienna, 1987.
6. G.L. Molnár, J. Radioanal. Nucl. Chem. 244 (2000) 27.
7. G. Elayi, in: Activation Analysis, Vol. II, Z. B. Alfassi, ed. (CRC Press, Boca Raton, 1990) pp. 73-141.
8. Handbook on Nuclear Data for Borehole Logging and Mineral Analysis, Technical Report Series No. 357, International Atomic Energy Agency, Vienna, 1993.
9. Table of simple integral neutron cross section data from JEF-2.2, ENDF/B-VI, JENDL-3.2, BROND-2 and CENDL-2, JEF Report 14, OECD Nuclear energy Agency, Paris, 1994; http://www-nds.iaea.or.at/indg_eval_therm.html
10. J. Kopecky, NGAtlas – Atlas of Neutron Capture Cross Sections, Report INDC(NDS)-362, International Atomic Energy Agency, Vienna 1997; http://iaeand.iaea.or.at/ngatlas/main.htm
11. V. McLane, Ch. L. Dunford, Ph. F. Rose, Neutron Cross Sections, Vol. 2, Neutron Cross Section Curves, Academic Press, New York, 1988.
12. Nuclear Data Viewer, Los Alamos National Laboratory, T-2 Nuclear Information Service; http://t2.lanl.gov/data/data.html
13. KAERI Table of the Nuclides; http://www.dne.bnl.gov/CoN/index.html, also mirrored at http://sutekh.nd.rl.ac.uk/CoN/, http://www.nes.ruhr-uni-bochum.de/CoN/index.html
14. Handbook on photonuclear data for applications. Cross-sections and spectra, IAEA-TECDOC-1178, International Atomic Energy Agency, Vienna, 2000.
15. H. Rauch and W. Waschkowski, in: LANDOLT-BÖRNSTEIN, New Series I/16A, Ed. H. Schopper (Springer, Berlin, 2000), Chap.6; http://www.ati.ac.at/~neutropt/scattering/table.html
16. V.F. Sears, Neutron News 3 (1992) 29; http://rrdjazz.nist.gov/index.html?page=/main.html
17. Evaluated Nuclear Structure Data File (ENSDF) - a computer file of evaluated experimental nuclear structure data maintained by the National Nuclear Data Center, Brookhaven National Laboratory; http://www.nndc.bnl.gov/~burrows/ensdf/
18. J. K. Tuli, Nuclear Wallet Cards, 5th ed., National Nuclear Data Center, Brookhaven National Laboratory, July 1995; subsequent updates by J. K. Tuli from the Evaluated Nuclear Structure Data File (ENSDF), http://www.nndc.bnl.gov/wallet/
19. F. De Corte, A. Simonits, J. Radioanal. Nucl. Chem. 133 (1989) 43.
20. F. De Corte, A. Simonits, F. Bellemans, M. C. Freitas, S. Jovanovic, B. Smodis, G. Erdtmann, H. Petri, A. De Wispelaere, J. Radioanal. Nucl. Chem. 169 (1993) 125.
21. S. Roth, F. Grass, F. De Corte, L. Moens, K. Buchtela, J. Radioanal. Nucl. Chem. 169 (1993) 159.
22. J. K. Tuli, Thermal Neutron Capture Gamma Rays, in: Prompt Gamma Neutron Activation Analysis, eds. Zeev B. Alfassi, Chien Chung, CRC Press, Boca Raton, 1995, p. 177; http://www.nndc.bnl.gov/wallet/tnc/capgam.shtml
23. M. A. Lone, R. A. Leavitt, D. A. Harrison, Atomic Data Nucl. Data Tables 26 (1981) 511.
24. G.L. Molnár, Zs. Révay, T. Belgya, R.B. Firestone, Appl. Radiat. Isot. 53 (200) 527.
25. G. L. Molnár, Zs. Révay, R. Paul, R. M. Lindstrom, J. Radioanal. Nucl. Chem. 234 (1998) 21.
26. Zs. Révay, G. L. Molnár, T. Belgya, Zs. Kasztovszky, and R. B.Firestone, J. Radioanal. Nucl. Chem. 244 (2000) 383.

27. R. L. Heath, Gamma-Ray Spectrum Catalog - Ge(Li) and Si(Li) Spectrometry, Report ANCR-1000-2, Aerojet Nuclear Company, 1974.
28. R. G. Helmer, R. J. Gehrke, J. R. Davidson, INEL CD-ROM version of the Ge(Li)-Si(Li) Gamma-Ray Spectrum Catalog, INEEL, 1999; http://id.inel.gov/gamma/samples.html
29. M.R. Ahmed, S. Al-Najjar, M.A. Al-Amili, N. Al-Assafi, N. Rammo, A.M. Demidov, L.I. Govor, and Yu.K. Cherepantsev, Atlas of gamma-ray spectra from the inelastic scattering of reactor fast neutrons (Atomizdat, Moscow, 1978)
30. Reference Neutron Activation Library, IAEA TECDOC, in preparation.
31. S.Y.F. Chu, L.P. Ekström and R.B. Firestone, The Lund/LBNL Nuclear Data Search, Version 2.0, February 1999; http://nucleardata.nuclear.lu.se/nucleardata/toi/
32. JANIS (JAva Nuclear data Information System), OECD Nuclear Energy Agency; http://www.nea.fr/html/dbdata/janis/

NUCLEAR DATA NEEDS FOR NON-INTRUSIVE INSPECTION

D.L. SMITH AND B.J. MICKLICH

Technology Development Division, Argonne National Laboratory,
Argonne, Illinois, U.S.A.

Abstract

Various nuclear-based techniques are being explored for use in non-intrusive inspection. Their development is motivated by the need to prevent the proliferation of nuclear weapons, to thwart trafficking in illicit narcotics, to stop the transport of explosives by terrorist organizations, to characterize nuclear waste, and to deal with various other societal concerns. Non-intrusive methods are sought in order to optimize inspection speed, to minimize damage to packages and containers, to satisfy environmental, health and safety requirements, to adhere to legal requirements, and to avoid inconveniencing the innocent. These inspection techniques can be grouped into two major categories: active and passive. They almost always require the use of highly penetrating radiation and therefore are generally limited to neutrons and gamma rays. Although x-rays are widely employed for these purposes, their use does not constitute "nuclear technology" and therefore is not discussed here. This paper examines briefly the basic concepts associated with nuclear inspection and investigates the related nuclear data needs. These needs are illustrated by considering four of the methods currently being developed and tested.

1 Introduction

There is no doubt that detailed and accurate knowledge of nuclear data is important for successful application of nuclear techniques to non-intrusive inspection. That having been said, in our experience we have observed the emergence of two fundamental truths: First, it is often very difficult to specify exactly what data are needed, and to what level of accuracy, until the development of a particular technique is well in progress. Second, it is amazing how many gaps still exist in what should be very basic - and often not so difficult in principle to determine - knowledge about the interactions of neutron and gamma radiation with matter. Another point worth mentioning is that often there exist administrative barriers to establishing and communicating specific nuclear data needs that are imposed by security concerns. Nuclear-based

inspection is a field that straddles those research domains that are designated as classified for security purposes and those that are open to broad inspection.

The first point raised above should not surprise the reader. Rarely does one know exactly what problems will emerge in any practical situation until one embarks on investigating the details. In the application of scientific knowledge to technology, there are often large disparities between what is "scientifically feasible" and what can be exploited practically in the field given limitations associated with cost, complexity, size and weight, and overall robustness of a particular approach. Little can be said here about those obstructions to the communication between users and producers of nuclear data imposed by security issues other than to emphasize that it is a reality that cannot be overlooked or avoided.

Continuing the discussion, one should not be puzzled by the gap between what information has been provided from scientific investigations and what is needed for practical applications. The motivations that drive the efforts of basic and applied scientists are quite different. An example: Many basic science studies of (CP,n) neutron-source reactions leading to the formation of product nuclei in discrete excited states have been performed using thin targets. The motivation was, quite reasonably from the perspective of the basic scientists, to learn about the properties of these excited nuclear states. However, in applied science the need is generally for neutron emission information corresponding to thick, stopping-target CP (charged-particle) reactions. Since less can be learned about the physics of these processes from thick target experiments it is not surprising that such information is frequently lacking in the scientific literature. Basic research scientists have often inadvertently discarded "raw" information derived from their experiments even though one day these results might prove valuable for applications. Generally, the intent of their research is to answer a specific scientific question and then to publish these academic results. An example: Establish the spin, parity, and isotopic spin of a particular excited nuclear state. Influenced by pressure from peers and rules fixed by scientific journals, attempts are seldom made to provide what pure-science communities tend to view as pedestrian, non-interpreted data or systematic data of broad scope.

In some cases, there may exist nuclear data that could satisfy the requirements for applications but that have not been adequately evaluated or, if evaluated, not processed into suitable form for use in computer codes such as those that perform Monte Carlo simulations of specific interrogation processes. Clear distinctions need to be made on this issue to avoid launching expensive nuclear data development programs that end up proving unnecessary.

Finally, there exists the well-known problem that there is frequently limited funding available for conceiving and developing new ideas pertinent to nuclear-based inspection. Potential users generally want to be able to acquire and field a device that has already been proven in the laboratory rather than to allocate scarce funding (as well as valuable time) to exploring the potential of a particular concept and eliminating technical "glitches". In short, resources for R&D tend to be limited in the coffers of user organizations. Because of the issues mentioned above, the present authors realized early in their quest to define nuclear data requirements for non-intrusive inspection that it would be difficult to provide definitive lists of universally acknowledged "nuclear data needs".

2 Basic Concepts of Inspection

Non-intrusive inspection can introduce a degree of technical difficulty that may not be encountered in non-destructive (*e.g.*, the luggage can be opened) or destructive techniques. All inspection concepts benefit from (or are limited by) the physics of neutron and/or gamma ray production by (or origination from) certain materials, and by their subsequent interactions with other materials as well as their detection. The major physics issues are as follows: radiation types, radiation sources, radiation intensities, radiation coincidences, radiation energy spectra, detector efficiencies, and background sources. The practical issues associated with applications are as follows: cost, complexity, safety, environmental impact, and - yes - political and legal questions. This paper will focus on the physics issues although it is difficult to de-couple the physical from the practical matters. Central to all inspection concepts is the idea of a "signal" or "signature". One defines what attribute of the interrogated object is sought (*e.g.*, type and quantity of contraband material and its location inside the sealed container) and attempts to identify what nuclear process could yield a signal or signature that is clearly discernable above background. Sometimes background issues are relatively minor concerns, but in most cases they are significant and often overwhelming. The reliability of a particular approach generally hinges on the ability to distinguish the signal clearly from the background. The lower the background the greater the sensitivity and specificity of a particular approach.

Passive techniques rely entirely on observing radiation emanating from materials residing in a container and shielded from direct visual observation. This approach is limited to detecting radioactive materials. Usually the signature involves the measurement of characteristic gamma radiation, but in some cases the observation of delayed neutron emission or spontaneous fission neutrons and γ-rays provides a signature. There is obviously a need to detect the signature radiation and to analyze absorption and scattering of this radiation.

Active techniques require that the interrogating radiation be produced externally and that the interrogated object, when exposed to this radiation, will generate a signal in the form of emitted, characteristic secondary radiation that then has to be detected. This approach offers many more conceptual options for nuclear inspection, but systems based on active techniques tend to be more expensive and complex to operate and, consequently, somewhat less reliable. In this paper we will consider data needs for these two approaches separately, but sometimes the boundaries are blurred so that similar nuclear data requirements emerge from both passive and active techniques.

3 Data Needed for Passive Inspection Techniques

Passive inspection techniques are employed in many applications: routine monitoring of radiation levels at nuclear facilities, assay of new and spent nuclear fuel, characterization of radioactive waste, personnel radiation dosimetry, control of nuclear materials transport, detection of clandestine nuclear materials (*e.g.*, nuclear warheads), monitoring of planned or inadvertent release of nuclear materials from the confines of nuclear facilities, identification of nuclear explosions and determination of the nature of the implicated devices, and general-purpose environmental monitoring of natural or man-made radioactivity to insure the safety of populations. The required apparatus is often relatively simple (*e.g.*, hand-held monitors) but it can be quite sophisticated (*e.g.*, for measurements that involve distinguishing very low-level

signals from background). All of these methods are based on detecting one or more of the following types of radiation emitted directly from suspect materials: α, β, γ, n (delayed), and n (SF). Consequently, the requisite nuclear data are associated with establishing signatures for specific nuclear materials, *i.e.*, defining the energies and the relative and absolute intensities of specific types of radiation, as well as determining the operating characteristics of radiation detectors. Most of these data are related to nuclear structure issues and can be found in the Chart of the Nuclides [1], Evaluated Nuclear Structure Data File (ENSDF) [2], or handbooks based on these sources. It is important that analyses be performed using the most recent evaluated results, wherever possible. The compilation and evaluation of nuclear structure data is an ongoing international activity.

Because of the wide range of applications and associated data requirements, it is difficult to define specific needs in the space allotted to this paper. One aspect of this subject, data needs for safeguards, has already been reviewed quite extensively by Boldeman [3]. Tables 1 and 2 based on his work provide examples of data needs in this area. It is seen that the needs are not very specific but they generally involve fissionable actinides, fission products, and tritium that is inevitably produced when neutrons are present from fission reactors or nuclear explosions.

4 Data Needed for Active Inspection Techniques

As mentioned in Section 2, nuclear technologies proposed for active non-intrusive inspection applications tend to be rather complex. The materials most often sought by these techniques are fissionable isotopes, illicit drugs, and explosives. Neutrons or energetic gamma rays are produced externally and in practice impinge on the object to be interrogated in order to stimulate the emission of a characteristic radiation that is detected to provide a "signal" or "signature" of the presence of a contraband material. In some cases, an indication of the quantity and location of this material is also provided. It is immediately obvious that the metrology system utilized must distinguish between the primary and secondary radiation. The former is always overwhelming in intensity relative to the latter. The process of distinction is especially difficult if the primary and secondary radiation is the same, *e.g.*, neutrons. The nuclear data needed for successful application of these technologies fall into three broad categories: production of primary interrogating radiation, scattering and absorption of primary and secondary radiation, and radiation-induced nuclear reactions. These processes must be understood not only for contraband materials but also for the benign materials that accompany them in realistic situations. As is the case for passive techniques, knowledge of nuclear structure and decay processes is required for several of these technologies.

Tables 3 and 4 provides some expressed needs for neutron interrogation and gamma interrogation applications, respectively, as gleaned from the authors' own experiences, private communications, and literature sources. In most cases they are listed here because they have not been satisfied. For neutron absorption, scattering, and induced reactions, one can refer to evaluated nuclear data files such as ENDF [4], JEF [5], JENDL [6], CENDL [7], and BROND [8] to obtain the latest available evaluations. Although these evaluated data libraries are far from complete, their content does offer the possibility of satisfying or nearly satisfying a rather large number of data needs once they are suitably processed for use in analysis codes.

5 A Look at Some Active Inspection Techniques

Many different active inspection techniques have been suggested. It would be impossible to review them all in the space and time available. Therefore, four distinct methods have been selected to demonstrate the interplay between concept and nuclear data in practical applications. Three of these methods involve the use of neutrons as an interrogating radiation while the third utilizes energetic gamma rays.

5.1 Fast Neutron Transmission Spectroscopy (FNTS)

Argonne National Laboratory [9] and several other laboratories have investigated the use of fast-neutron transmission spectroscopy to locate and reliably identify both explosive materials and illegal drugs in containers by determination of elemental content (see Figure 1). The measurements involve an accelerator, pulsed-beam time-of-flight techniques, and fast-neutron detectors. This technique is heavily dependent on computer simulation and the application of tomographic methods that require two types of nuclear data as input. First, one needs to use energy-dependent spectra and angular distributions of thick-target, light-element, white-spectrum neutron source reactions such as Be(d,n) and (p,n) and ^{7}Li(d,n) and (p,n) in order to estimate the efficiency of the method. Analysis of the technique at ANL was undertaken using the Be(d,n) source, and at the outset of this project it was found that the neutron-production data for this reaction was not adequately known for deuterons from 0.5 – 10 MeV. Some measurements have been performed that allowed estimation of the effectiveness of the interrogation procedure [10] (see Figure 2). In some applications the Be(p,n) reaction might be a better choice, but knowledge of this reaction remains inadequate pending publication of recent results from Ohio University.

Other required data for the application are neutron total cross sections from 0.5 – 10 MeV for H, C, O, N, Fe, Ni, Cu, Cl, S, and other elemental materials found in drugs, explosives, and benign materials included in luggage and containers. As can be seen from Figure 1, the interrogating neutron beams are never perfectly collimated so the neutron transmission through an interrogated object can be affected by small-angle scattering. Thus, effective transmission spectra cannot be adequately reproduced by simply energy-averaging high-resolution neutron total cross section results found in data libraries such as ENDF [4] even though these evaluated results are generally quite well known. Libraries of effective neutron-transmission cross sections for realistic geometries have to be developed, either by direct measurements or sophisticated calculations that take into account small-angle scattering, in order to adequately "unfold" elemental-abundance values for the inspected objects.

5.2 Pulsed Fast-Neutron Analysis (PFNA)

In this neutron inspection technique, approximately mono-energetic beams of fast neutrons with energies of $\approx$ 8.5 MeV are produced using the ^{2}H(d,n)^{3}He source reaction [11,12]. There are also variants of the method that have been suggested that employ 14-MeV neutrons from D-T neutron generators. Neutrons impinge on the object to be interrogated and they produce gamma rays by (n,n'-γ) reactions (see Figure 3). Individual isotopes (and thus elements) are identified and quantitatively measured by observing the characteristic gamma rays that follow prompt de-excitation of specific levels in C, O, N, Cl, and several other elements. The gamma-ray measurements are usually made using NaI scintillation detectors that provide both pulse-height and timing information with good efficiency. In PFNA, contraband objects are

identified by observing signature ratios for gamma ray yields from key elements. The method requires use of an accelerator and pulsed-beam time-of-flight techniques. It is particularly vulnerable to the effects of small angle neutron and gamma-ray scattering as well as absorption.

Properties of the neutron source reaction appear to be adequately known. The inelastic γ-ray yield data available data for C are relatively consistent and may also be adequately known (see Figure 4). However, the angular distribution information available for C in evaluated libraries appears to be inadequately represented. Furthermore, what is clearly inadequately known are the excitation functions for gamma-ray production from O, N, and Cl (*e.g.*, see Figure 5). Finally, angular distributions of the gamma rays are generally not well enough represented for most elements based on the evaluated libraries. As mentioned above, in the case of angular distributions part of the problem may be inadequate formats for representation of these evaluated data, *i.e.*, shortcomings in the Legendre-polynomial representation scheme used in ENDF [4]. The neutron elastic scattering results are probably adequate for the important materials. However, inelastic scattering, which also distorts the primary neutron spectrum and influences background gamma-ray spectra from benign materials, is generally not sufficiently well defined.

5.3 Fissile Interrogation using Gamma Rays from Oxygen (FIGARO)

A non-intrusive inspection technology based on use of 6 – 7 MeV gamma rays from the $^{19}F(p,\alpha\text{-}\gamma)^{16}O$ reaction is under development at Argonne National Laboratory [13] and it has been found to be effective in locating fissile material and other nuclear materials such as ^{6}Li, Be, and deuterium hidden in containers (see Figure 6). Success of this method relies on the fact that photons in the 6 – 7 MeV range can induce photo-nuclear and/or photo-fission neutron production on the above-mentioned nuclear materials whereas neutron emission from most benign materials is not energetically allowed. It is possible to detect the secondary neutrons in the presence of intense gamma-ray fields by using a gamma-insensitive detector system with good efficiency, such as arrays of BF_3 counters in a polyethelene moderator.

When this project began it was found that the available thick-target photon yield data from the $^{19}F(p,\alpha\text{-}\gamma)^{16}O$ reaction were completely inadequate to predict feasibility. Therefore they were measured to the required accuracy from 1.5 – 4 MeV [14] (see Figure 7). Full exploitation of this technique is still limited by inadequacy of thick-target (p,n) neutron data needed to assess background neutron-source problems. Furthermore, knowledge of photo-fission and photo-neutron cross sections for actinides is poor in the region below 10 MeV preventing, *e.g.*, a clear distinction between ^{235}U (HEU) and ^{238}U (DU).

5.4 Multi-Detector Analysis System (MDAS)

In this concept, waste containers to be inspected are irradiated with intense low-energy neutron fields (generally, ≤ 0.5 MeV) [15]. The neutrons induce fission in actinide materials and the prompt gamma rays emitted from de-exciting fission fragments are observed with an array of HPGe detectors. Signatures of fission in specific actinide isotopes are provided by observing selected gamma rays emitted from the fragments in pair-wise coincidence. If it is known that a particular gamma-ray line corresponds to prompt de-excitation of a particular FP nucleus, then many gamma rays observed in coincidence should correspond to emission from the other fragment. Relying on conservation of total charge number, one can narrow the range of possibilities to isotopes of a particular element. In principle, by also performing coincident neutron multiplicity measurements, and relying on conservation of total neutron number, one can

identify the specific sister FP nucleus and thereby establish which actinide isotope actually underwent fission. This method relies on use of radioactive (*e.g.*, ^{252}Cf SF), a reactor, or accelerator neutron source, extensive computer analysis of coincident gamma-ray spectra, and computer simulation to deal with geometry problems, neutron and photon absorption, *etc.*

Obviously, the method requires detailed knowledge of the energies and relative intensities of gamma rays associated with the decay of excited states of FP nuclei. These data may not be well known for many of the FP isotopes far from the line of stability. Knowledge of neutron fission cross sections and prompt neutron (nu-bar) and γ-ray multiplicities as a function of neutron energy is necessary for all actinide materials likely to be found in interrogated waste containers.

6 Prognosis for Satisfying the Nuclear Data Needs

Most of the nuclear data needs outlined in very general terms in this paper could in principle be satisfied in a straightforward manner using existing facilities, techniques, and theoretical and experimental expertise available around the world. In some cases, the data needs stated here may have already been met. An example: New evaluations for Bismuth (already released in ENDF) and for Oxygen (soon to be released in ENDF) should lead to improvements in the situation for these elements. However, the overall prognosis for significant progress to made in the foreseeable future is not encouraging. There are attitude barriers to overcome, particularly in the basic research community. Facilities where applied work is usually done are generally under-staffed and under-funded, or their resources are devoted to other tasks. Funding is a key issue. Agencies charged with carrying out non-intrusive interrogation for various purposes generally do not have research budgets that are adequate for this task and they are often inadequately informed concerning nuclear data issues.

Should a turn of events lead to a higher priority being assigned to obtaining nuclear data for nuclear inspection purposes, the first order of business would be to examine very carefully the adequacy of the existing databases in the context of intended applications. This is not a trivial job. To do it right would involve several man-years of skilled specialist effort (not necessarily restricted to a single individual). Due to security issues and the highly diverse nature of the methods and their practitioners in this field, it is not clear whether this review will take place in the foreseeable future. Another problem is that many of the specialists who are skilled in performing the needed measurements, theoretical calculations, and evaluations are retiring and these skills are not being replaced. The longer-term consequences of this evolving situation for the future of all nuclear technologies are potentially quite negative.

7 Summary

It is seen that there are extensive nuclear data needs for both passive and active non-intrusive inspection applications. Generally, the requirements are not very carefully defined or documented suggesting that there is a need for detailed sensitivity studies to be performed and reported. However, due to security concerns and funding limitations this important work is not likely to be completed in a comprehensive fashion in the foreseeable future. For passive applications, the needs generally relate to nuclear structure properties and to the decay of

radioactive species. For active techniques, such data are also needed along with information on production, scattering, absorption, and nuclear transmutation information for the neutrons and gamma rays that are both employed as the interrogating radiation and are detected as emitted secondary radiation. In many cases, information that satisfies these needs adequately can be found in libraries of evaluated structure and cross section data but may not be suitably processed.

Decay data are generally reasonably well known for the major isotopes of structural materials such as Al, V, Fe, Cu, Ni, *etc.*, and for the major fissile materials and fission products, as long as the half lives are neither too short nor too long. For less common materials, for cases where the half lives are very short or very long, or for those where the decay schemes do not favor convenient measurement, the information is less reliable. Neutron scattering data for such common elements as C, Al, Fe, Ni, *etc.*, are reasonably well known. It is surprising that similar data for O are not as well known and that the most recent ENDF evaluation for Cl (a major constituent of illicit drugs) dates to 1974 and is both uncertain and incomplete. Gamma-ray production data are generally less well known than neutron scattering data. Neutron and γ-ray source data, particularly for thick targets are available only in a very limited number of situations. There would be a clear benefit obtained from comprehensive measurements made of thick-target neutron yields, angular distributions and spectra from (p,n) and (d,n) reactions on all stable materials up to Fe for incident particle energies from a few hundred keV up to perhaps 10 MeV. Finally, both photo-fission and photo-neutron (γ,n) measurements ought to be made on a variety of benign as well as fissionable and other nuclear materials from threshold to about 10 MeV. In particular, the ability to distinguish between ^{235}U (in HEU) and ^{238}U (in DU) based on photon-induced neutron production would be extremely valuable.

Acknowledgments

The authors are indebted to the following individuals who through private communications offered material for use in preparing this paper: Z. Bell (Lockheed-Martin Energy Systems), J. Bendahan (Ancore), Y. Dardenne (LLNL), C. Fink (ANL), J. Hall (LLNL), S.D. Harvey (PNL), J. Mihalzco (ORNL), D. Spears (DOE), and T. Valentine (ORNL). This work was supported by the U.S. Department of Energy, Science Programs, under Contract W-31-109-ENG-38.

References

[1] J.R. Parrington *et al.*, *Nuclides and Isotopes: Chart of the Nuclides 15th Edition*, G.E. Nuclear Energy, 175 Curtner Avenue, San Jose, CA 95125 (1996).

[2] ENSDF, "Evaluated Nuclear Structure Data File", National Nuclear Data Center, Brookhaven National Laboratory, U.S.A. Internet: http://www.nndc.bnl.gov (2000).

[3] J.W. Boldeman, "Nuclear Data for Safeguards and a Possible Comprehensive Test Ban Treaty", *Proceedings of the International Conference on Nuclear Data for Science and Technology*, ed. J.K. Dickens, American Nuclear Society, p. 1108 (1994).

[4] ENDF/B-6, "Evaluated Nuclear Data File", National Nuclear Data Center, Brookhaven National Laboratory, U.S.A. Internet: http://www.nndc.bnl.gov (2000).

[5] JEF-2, "Joint Evaluated File", NEA Nuclear Data Bank, Nuclear Energy Agency, Paris, France (1998).

[6] JENDL-3, "Japanese Evaluated Nuclear Data Library", Nuclear Data Center, Japan Atomic Energy Research Institute, Tokai-mura, Japan (1994).

[7] CENDL-2, "Chinese Evaluated Nuclear Data Library", Nuclear Data Center, Institute of Atomic Energy, Beijing, China (1992).

[8] BROND-2, "Russian Evaluated Nuclear Data Library", Nuclear Data Center, Obninsk, Russia (1997).

[9] B.J. Micklich, M.K. Harper, L. Sagalovsky, and D.L. Smith, "Nuclear Data Needs and Sensitivities for Illicit Substance Detection Using Fast-Neutron Transmission Spectroscopy", *Proceedings of the International Conference on Nuclear Data for Science and Technology*, ed. J.K. Dickens, American Nuclear Society, p. 1102 (1994).

[10] J.W. Meadows, "The Thick-Target ^{9}Be(d,n) Neutron Spectra for Deuteron Energies Between 2.6 and 7.0 MeV", ANL/NDM-124, Argonne National Laboratory (1991).

[11] J. Bendahan, R. Lovemen, and T. Gozani, "Nuclear Data for Non-Intrusive Inspection Systems", *Proceedings of the International Conference on Nuclear Data for Science and Technology*, ed. J.K. Dickens, American Nuclear Society, p. 1093 (1994).

[12] J. Bendahan, R. Loveman, and T. Gozani, *Nuclear Instruments and Methods In Physics Research* **A353**, 205 (1994).

[13] B.J. Micklich, D.L. Smith, T.N. Massey, D. Ingram, and A. Fessler, *Proceedings of the 16th Conference on Applications of Accelerators in Research and Industry*, Denton, Texas (November 2000).

[14] A. Fessler, T.N. Massey, B.J. Micklich, and D.L. Smith, *Nuclear Instruments and Methods in Physics Research* **A450**, 353 (2000).

[15] G.M. Ter-Akopian *et al.*, *Physical Review* **C55**, 1146 (1997). Also, J.D. Cole, INEEL, Idaho, U.S.A. (private communication).

[16] G. Andrew, "Prospects for Environmental Monitoring in International Safeguards", *Proceedings of an International Symposium on International Safeguards*, Vienna, 14-18 March (1994).

[17] M. Lammer, "Nuclear Data for Safeguards", INDC/P(81)-24 (1981).

Table 1. Some nuclear data needs for passive monitoring of small releases to the environment. [a]

Materials	Nuclear Data Required	Application
^{3}H, ^{85}Kr, Pu (α), ^{241}Am, U, ^{95}Zr	Radioactive decay data. [b]	Air
^{129}I, Pu (α), U	Radioactive decay data. [b]	Deposition
^{90}Sr, ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{129}I, Pu (α), ^{241}Am, U	Radioactive decay data. [b]	Soil/Dust
^{3}H, ^{129}I, ^{95}Zr, ^{95}Nb, U	Radioactive decay data. [b]	Vegetation (Terrestrial)
^{3}H, ^{125}Sb, ^{129}I, ^{137}Cs, ^{237}Np, Pu (α), ^{241}Pu, ^{241}Am	Radioactive decay data. [b]	River Water
^{95}Nb, ^{110m}Ag, Pu (α), ^{241}Pu, ^{241}Am, Cm	Radioactive decay data. [b]	Sediment (Freshwater)
^{95}Nb, ^{110m}Ag, Pu (α), ^{241}Pu, ^{241}Am	Radioactive decay data. [b]	Vegetation (Freshwater)
Pu (α), ^{241}Am, ^{237}Np	Radioactive decay data. [b]	Sea Water
^{95}Zr, ^{95}Nb, ^{144}Ce, ^{237}Np, Pu (α), Cm	Radioactive decay data. [b]	Sediment (Marine)
Pu (α), ^{241}Pu, ^{241}Am	Radioactive decay data. [b]	Marine Algae

[a] Information taken from J. Boldeman [3] and attributed to G. Andrew [16].
[b] Half lives, energies, and intensities of emitted radiation: α, β, γ, n (delayed), and n (SF) as applicable.

Table 2. Some nuclear data needs for passive assay of fresh and spent nuclear fuel. [a]

Materials	Nuclear Data Required	Application
^{232}U and its daughter products	^{232}U half life. γ-ray energies and intensities fordaughter products.	Assay recycle U fuel. Correction for ^{232}U daughter interference with ^{235}U γ-rays.
Pu isotopes, ^{241}Am, and their daughter products	Pu isotope and ^{241}Am half lives. Energies and intensities of γ-rays from α-decay daughters.	Quantitative analysis of γ-ray spectrum of Pu containing fuel.
$^{238,239,240,242}Pu$, ^{241}Am, $^{242,244}Cm$	α-decay and SF half lives. Nu-bar for SF.	Passive analysis of neutron emission from irradiated fuel.
^{238}U, ^{241}Pu, ^{242}Am, ^{243}Cm	Neutron fission and capture cross sections.	Passive analysis of neutron emission from irradiated fuel.
^{241}Pu, ^{242}Am, ^{243}Cm	Decay half lives.	Passive analysis of neutron emission from irradiated fuel.
^{18}O	(α,n) cross section.	Interference effects in passive analysis of neutron emission from irradiated fuel.

[a] Information taken from J. Boldeman [3] and attributed to M. Lammer [17].

Table 3. Some nuclear data needs for active neutron non-intrusive inspection applications. [a]

Materials	Nuclear Data Required	Application
Bismuth, Cesium, Chlorine, Germanium, Iodine	Neutron activation data from 0 – 16 MeV.	Neutron dosimetry. Neutron activation. For use in analysis of HPGe, NaI, CsI, and BGO detector response and background. PFNA, FNA, *etc.*
Bismuth, Cesium, Chlorine, Germanium, Iodine	Prompt neutron-induced γ-ray production data.	Neutron response. For use in analysis of HPGe, NaI, CsI, and BGO detector data. PFNA, FNA, *etc.*
Bismuth, Carbon, Chlorine, Germanium, Iodine, Nitrogen, Oxygen	Neutron differential elastic and inelastic scattering data from 0 – 16 MeV. Evaluations for Chlorine and Germanium are incomplete and very old.	Neutron transport calculations. TNA, PFNA, FNA, NES, *etc.*

Nitrogen, Oxygen, Chlorine	Inelastic γ-ray production yields and angular distributions.	Analysis of signature γ-ray production. TNA, PFNA, FNA, *etc.*
Major actinide elements	Prompt fission neutron emission data for multiplicity and correlation analysis. Nu-bar and neutron energy spectra. Data are especially poor for ^{237}Np and ^{232}Th.	Actinide identification measurements using neutron multiplicity detectors.
Major actinide elements	Prompt fission γ-ray emission data for multiplicity and correlation analysis. Gamma-ray energies, intensities and angular distributions. Results for ^{235,238}U and ^{239}Pu are lacking and are particularly important.	Actinide identification measurements using γ-ray multiplicity detectors. This may prove to be a more sensitive approach than neutron detection.
Germanium	Neutron inelastic and capture γ-ray production cross sections. Activation reaction data. Evaluated data for Germanium are incomplete and very old.	Identification of background interference in monitoring γ-rays with HPGe detectors in the presence of neutrons from nuclear weapons or reactors.
C, O, Al, Cl, Fe, Ni, and Hg	Neutron differential elastic and inelastic scattering data.	Characterization of nuclear waste by PFNA, NES and related techniques.
C, N, O, Al, Fe, Cu, Cl, and others	Neutron differential elastic and inelastic scattering data.	Monte Carlo simulation in PFNA for the detection of explosives and narcotics.
Carbon	Angular distribution of 4.44-MeV γ-ray produced by neutron inelastic scattering. Representation in evaluated files is inadequate.	Signature measurements of the presence of carbon by PFNA.
Fission products, light elements and structural materials	Neutron yield from (α,n) reactions.	Data needed to estimate neutron production from α-particle induced reactions on various materials in the presence of α-active actinides. Resolution of background problems.
Actinides	Yields of FP nuclei as a function of neutron energy. Of particular interest are thermal neutron fission yields of ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{106}Rh, ^{133}Cs, ^{137}Cs, ^{140}Ba, ^{140}La, ^{144}Ce, ^{153}Eu, ^{134}Cs, ^{140}La, ^{144}Ce, ^{144}Pr, ^{154}Eu.	Data needed for use in active assay of spent fuel.
^{133}Cs, ^{153}Eu	Neutron-capture cross sections.	Data needed for the active assay of spent nuclear fuel.
H, C, N, O, F, Na, Al, Si, Cl, K, Fe, and Cu	Transmission-derived neutron total cross sections in realistic geometries for 0.5 – 10 MeV.	Unfolding of elemental abundances by FNTS in geometries where small-angle scattering effects are influential.

[a] Information derived from literatures sources [3,9,11,12,15] and private communications.

Table 4. Some nuclear data needs for active gamma-ray non-intrusive inspection applications.

Materials	Nuclear Data Required	Application
Common benign materials, especially those with low photo-nuclear reaction thresholds	(γ,n) cross sections from threshold to about 15 MeV. Knowledge of emitted neutron spectra would also be useful.	Photon interrogation concepts such as FIGARO [13] and those which employ white-spectrum gamma sources.
Major actinides, especially ^{235}U, ^{233}U, ^{238}U, and Pu isotopes	(γ,f) and (γ,n) cross sections from threshold to about 15 MeV measured with sufficient accuracy to distinguish different actinide species.	Photon interrogation concepts such as FIGARO [13] and those which employ white-spectrum gamma sources.

F, Al, Au, Fe, Ni, S, Ca, Mg and other elements found in target assemblies and target compounds.	Thick-target (p,γ) yields, angular distributions, and spectra. Production of primary interrogation gamma rays as well as background gamma rays	Photon interrogation concepts such as FIGARO [13] and those which employ white-spectrum gamma sources.
Target structure, target compound, and accelerator beam line materials such as Fe, C, S, etc., with low (p,n) thresholds.	Thick-target (p,n) neutron yields, angular distributions, and spectra. Data needed to identify sources of neutron background.	Photon interrogation concepts such as FIGARO [13] and those which employ white-spectrum gamma sources.

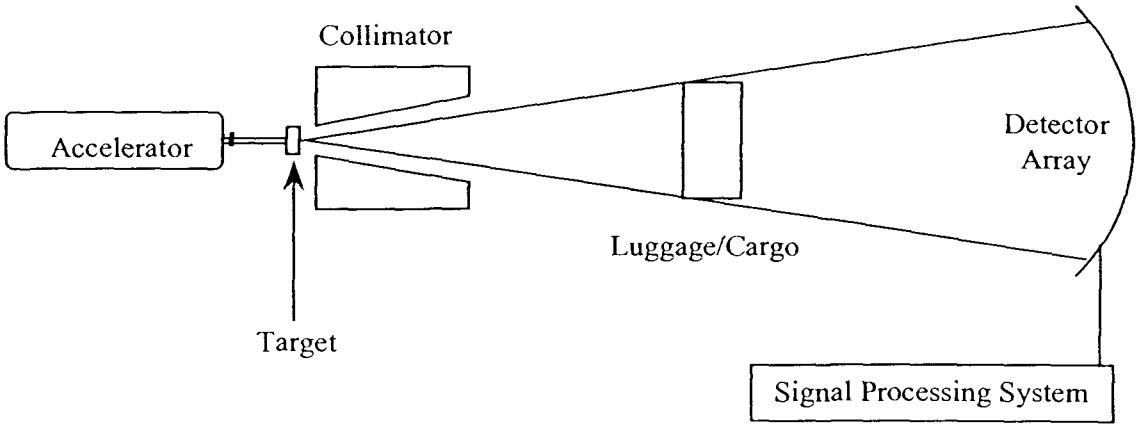


Figure 1. Schematic diagram of Fast Neutron Transmission Spectroscopy (FNTS).

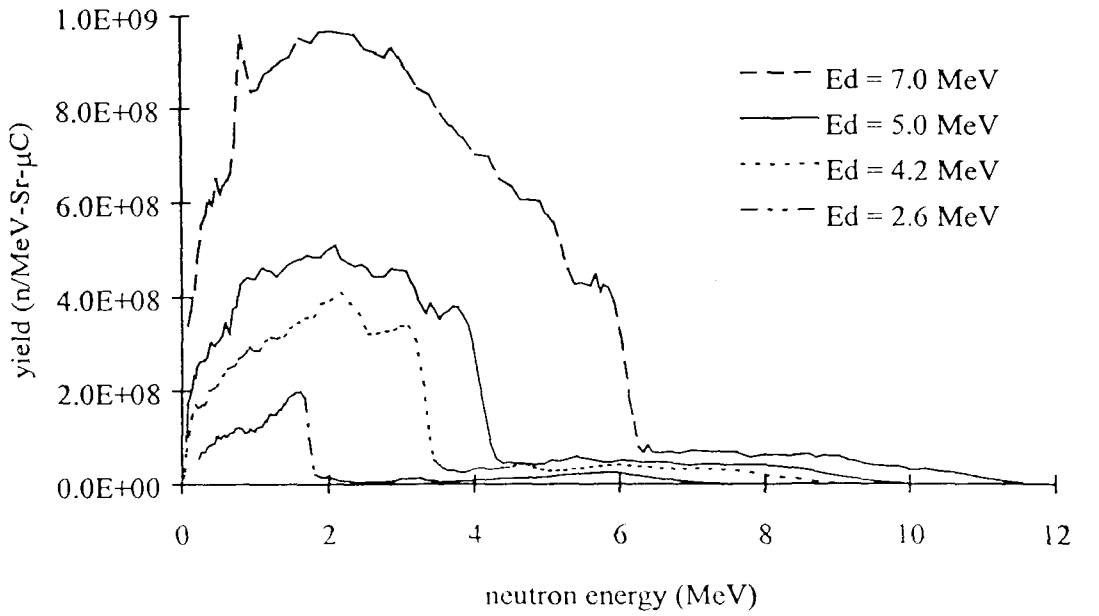


Figure 2. Neutron source spectrum for the ^{9}Be(d,n) reaction for selected incident deuteron energies.

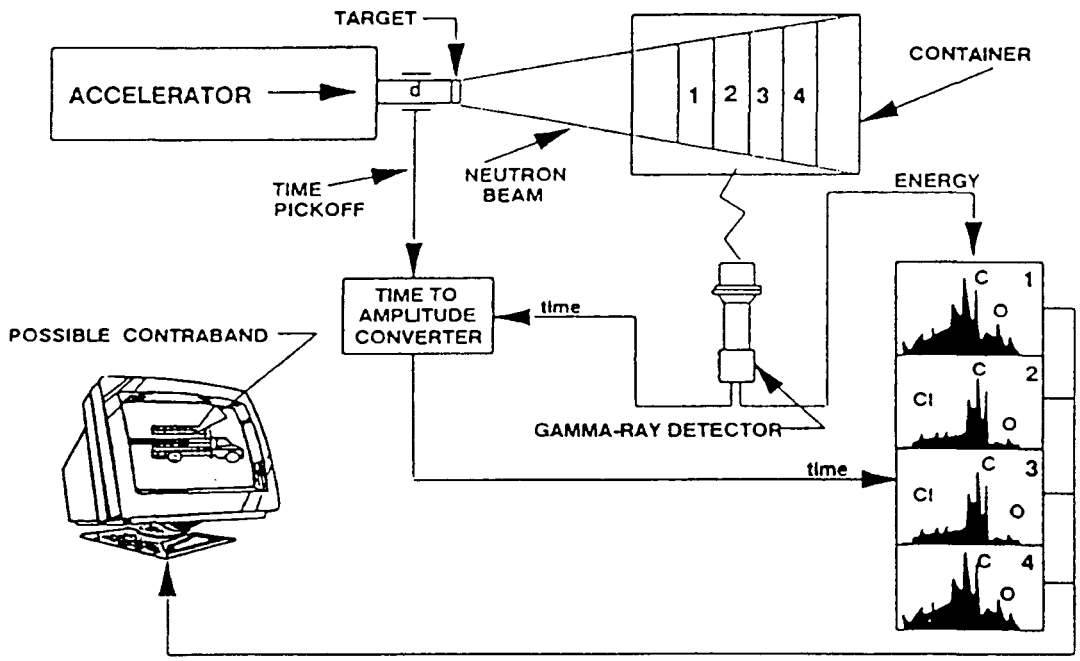


Figure 3. Schematic diagram of PFNA (Pulsed Fast Neutron Analysis).
(Figure published by J. Bendehan *et al.* [11])

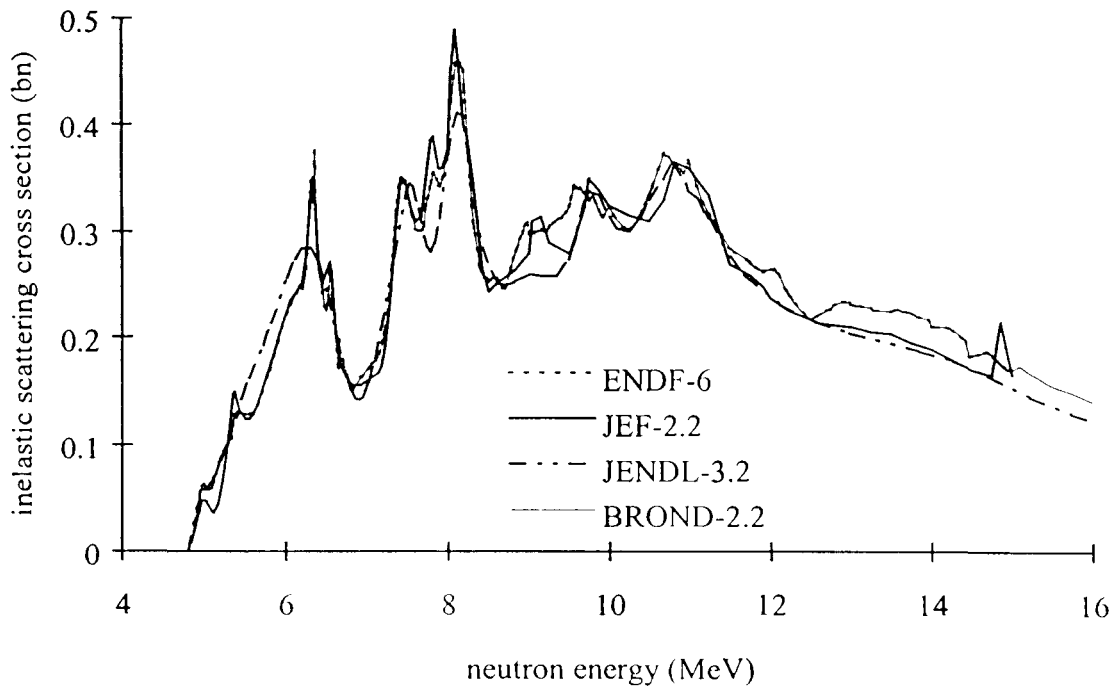


Figure 4. Inelastic scattering cross-section to the first excited state of ^{12}C.

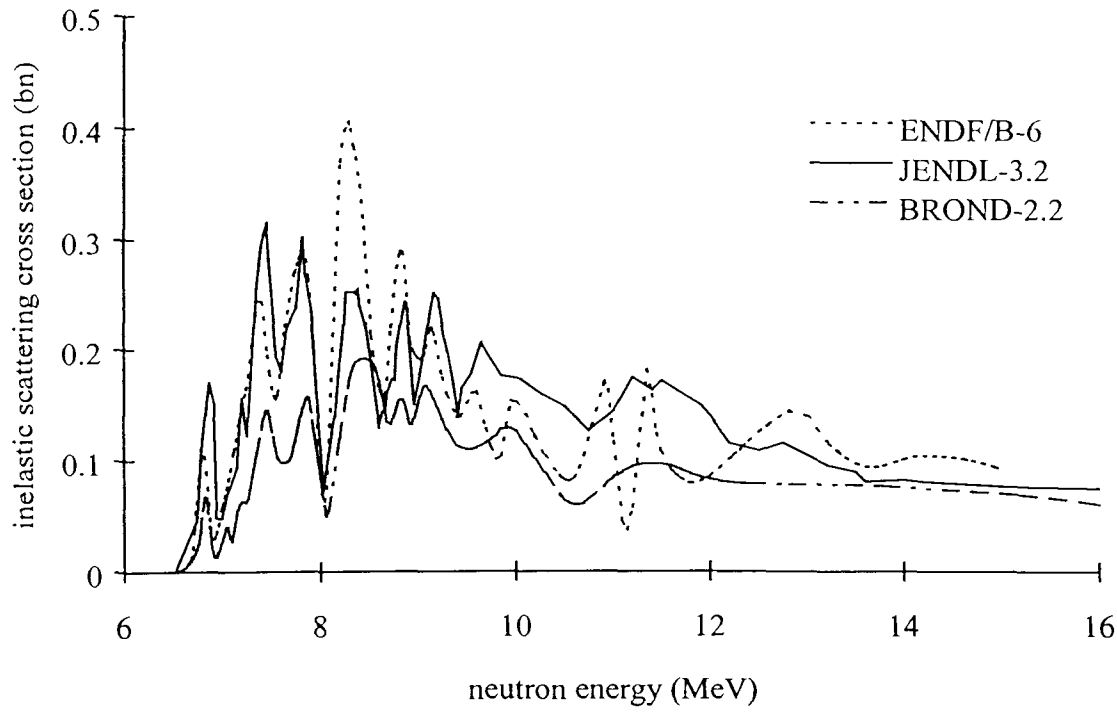


Figure 5. Inelastic scattering cross-section to 2nd excited state of ^{16}O (JEF-2.2 is identical to ENDF/B-6).

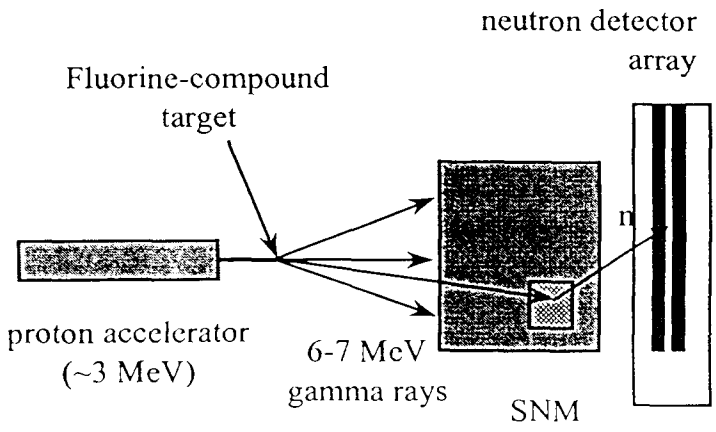


Figure 6. Schematic diagram of FIGARO (**F**issile **I**nterrogation using **G**amma **R**ays from **O**xygen).

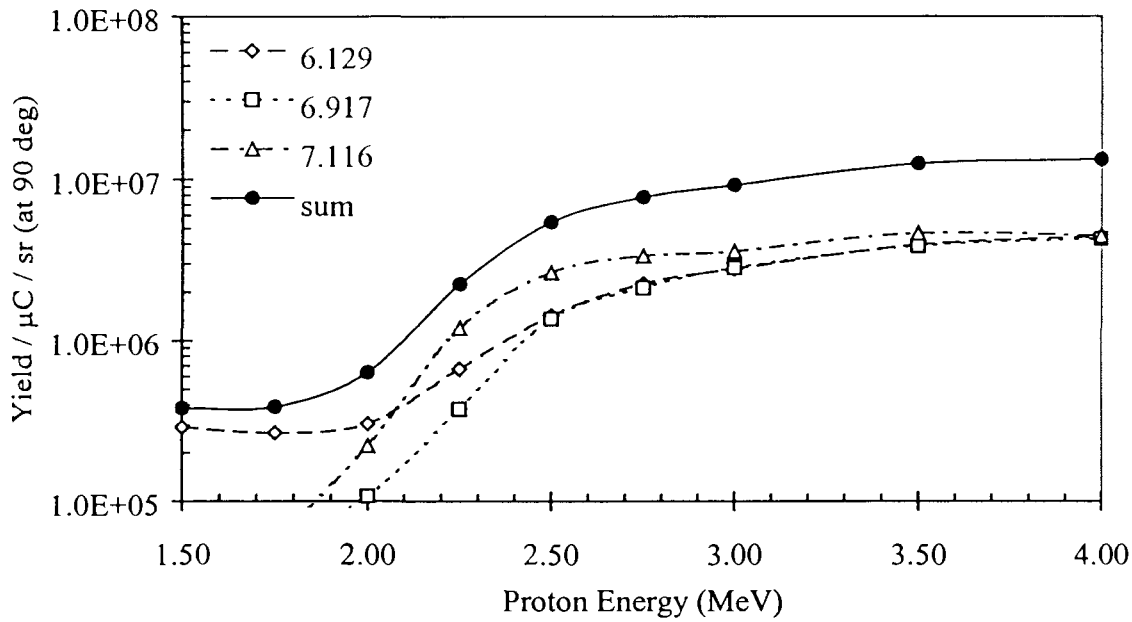


Figure 7. Gamma yield curve for the $^{19}F(p,\alpha\text{-}\gamma)^{16}O$ reaction as a function of proton energy

NUCLEAR DATA FOR ADVANCED FAST REACTORS

N.S.Rabotnov

Ministry of the Russian Federation for Atomic Energy, Moscow, Russia

ABSTRACT

Interest revives to fast reactors as the only proven technology obviously able of satisfying human energy needs for next millenium by using full energy content of both natural uranium resources and of vast stocks of depleted uranium. This interest stimulates revision and improvement of fast reactor ND. Progress in reactor calculations accuracy due to better codes and much faster computers also increases relative importance of the input data uncertainties, especially in case of small reactivity margin and fuels of equilibrium compositions. The main objects of corresponding R&D efforts should be minor actinides and heavy liquid metal coolant. Data error bands and covariance information also gains importance as necessary component of neutron physics calculations.

1. INTRODUCTION

After prolonged stagnation in nuclear power industry development which was triggered by TMI and Chernobyl accidents and resulted in postponement of the plans to start full scale introduction of fast reactors there are signs now of revived interest in FR technology. The interest is stimulated by gradual realization of following basic facts:

1. The mankind is consuming organic fuels million times faster than Nature was creating them. Peak production level of the most important one – conventional oil – may be reached in a few years and followed by decline so steadily growing demand could be met only by non-conventional oil at much higher prices. Recent signals from the oil market should not be misinterpreted.
2. The resources of natural uranium to be enriched and used in PWR type thermal reactors only could support nuclear power for less than a century even if spent nuclear fuel is recycled with full use of regenerated U and Pu.
3. Fast reactors are the only proven technology obviously able of satisfying human energy needs for next millenium by using full energy content of both natural uranium resources and of vast stocks of depleted uranium.
4. Fast reactors can burn minor actinides destroying the most hazardous component of radwaste and provide excess neutrons for transmutation of long-lived fission products so, in principle, radiation balance may be reached between excavated and buried radioactivity in long term closed nuclear fuel cycle.

Operating fast reactors were designed a few decades ago so nuclear data libraries should be revisited and critically analyzed to make fast neutron physics calculations of future advanced units more reliable. Some most prominent problems are discussed below.

2. BASIC PRINCIPLES AND CHALLENGES OF ADVANCED FAST REACTORS DESIGN

Following requirements to nuclear technologies of future large scale nuclear power industry may be formulated:

- higher safety level based primarily on natural laws, not on engineering barriers (natural safety);
- guaranteed fuel supply for thousands of years;
- proliferation resistance;
- competitiveness with organic fuels;
- solution of the problem of long-lived radwaste.

Physical and chemical principles of natural safety allowing to meet the above requirements:

- neutron rich cores with hard fast spectrum;
- reactivity effects of appropriate signs and values;
- breeding ratio slightly above unity without uranium blankets, producers of high grade Pu;
- inert, high-boiling, low activating coolant;
- closed NFC without Pu separation and with transmutation of long-lived radionuclides.

A set of engineering solutions for implementation of natural safety principles was proposed recently in Russia [1-2] and chosen by Minatom as focus of development efforts:

- dense mononitride fuel with equilibrium actinide composition;
- heavy liquid metal coolant (Pb);
- double circuit cooling;
- BR≈1.05 without uranium blankets;
- total reactivity margin within $\approx \beta_{eff}$ for the whole fuel life-time;
- fuel regeneration without U and Pu separation.

Possibility to apply the same principles (except the coolant choice, of course) for modification of Na-cooled BN-800 fast reactor design is now under intense investigation [3] and first results indicate that parameters close to those expected with Pb coolant may be achieved and the first BN-800 unit may incorporate the new features.

3. STATUS AND NEEDS IN THE SECTOR OF NUCLEAR DATA FOR FAST REACTORS

Recent progress in the accuracy of fast reactor neutron physics calculations is estimated in Table 1. Up to now final uncertainties were determined mainly by software algorithmic quality and by computers' speed but with their substantial progress the uncertainties of input nuclear data begin to show.

Table 1. Target and achieved accuracy of fast reactors neutronic calculations [4].

	Target Accuracy	Achieved Accuracy	
		1990	2000
K-eff	**0.5 %**	2.5 %	1.0 %
Void Reactivity Effect	**0.2 %Δk/k**	1.1 %Δk/k	0.4 %Δk/k
Doppler Effect	**10 %**	20 %	12 %
Power Release	**2 %**	5 %	3 %
Control Rods Efficiency	**5 %**	20 %	15 %
Breeding Gain	**0.02**	0.06	0.04

Following main directions of FR nuclear data improvements may be pointed out:

3.1 Minor actinides. FR equilibrium fuel itself contains MA with concentrations much higher than SNF of PWRs. If heterogeneous transmutation mode is applied MA concentrations in the target sub-assemblies will be still higher so needed data accuracy for MA should be close to the accuracy of traditional fuel nuclides. A list of achieved and target accuracies of minor actinides data aimed at transmutation needs and published recently [5] is given in Table 2.

Table 2. Actinides fast neutrons cross sections accuracy, achieved and needed, per cent [5].

	Capture		**Fission**		**Inelastic**	
	Achieved	**Target**	**Achieved**	**Target**	**Achieved**	**Target**
Np-237	15	5	7	5	30	10
U-238	5	3	3	3	10	10
Pu-238	25	10	10	5	40	
Pu-239	6	4	3	3	20	15
Pu-240	10	5	5		20	15
Pu-241	15	5	5	3	20	
Am-241	10	5	10	5	30	10
Am-242m	30	10	15	5	40	
Am-243	30	10	10	5	30	
Cm-242	50	10	15	5	30	
Cm-243	50	10	15	5	30	
Cm-244	50		10	5	30	

MA nuclear data are important not only for reactivity calculations but probably even more so for closed nuclear fuel cycle calculations because MA concentrations in the fuel to be reprocessed determine technology, safety and economy of the process as well as the level of irretrievable losses and radiotoxicity of final radwaste.

Dozens of specimens of actinide isotopes and isotopic mixtures were irradiated in BN-350 fast reactor [6]. Full time of irradiation was 781-797 days, fluence varied from 1.5 to $2.0 \times 10^{23} cm^{-2}$, full burn up from 9.3 to 23 per cent, principal isotope burn up from 11 to 35 per cent. Some results compared with calculated values are presented in Table 3. C/E ratio is

considerably less than unity in most cases, the lowest value is below 50 per cent. Preliminary recommendations resulting from this analysis are: uncertainties pile up in long chains which probably indicates on correlation of the data used in calculations; fast neutron capture cross sections for some actinides are underestimated and should be increased (^{240}Pu by 8%, ^{241}Am by 10%, ^{242}Cm by 20%); covariation matrices of the uncertainties should be estimated.

Table 3. Accumulation of Am and Cm in actinide specimens irradiated in BN-350.

Nuclide	Cell 89	Cell 243
	C/E	C/E
^{242m}Am	1.11±0.02	-
^{243}Am	0.78±0.09	-
^{242}Cm	0.89±0.10	0.83±0.09
^{243}Cm	0.67±0.03	0.58±0.01
^{244}Cm	0.47±0.023	0.75±0.70

3.2. Burn-up credit. One of the well known problems in the reactor fuel cycle calculations is burn-up credit. It's important due to influencing the efficiency of SNF storage and transportation. PWR case is, in a sense, simpler: spent fuel unloaded from reactor is placed in the same environment – water - in "wet" storage. With fast reactors situation is different, and the difference is especially important for the fuels of equilibrium compositions. First, water moderating and reflecting properties are drastically different from those of liquid metal coolants. Second, small reactivity margin during the whole fuel life time in the core means that SNF practically maintains its criticality properties so, due to changing environment, burn up credit may turn into "burn up debit" which should be calculated carefully.

Extensive OECD/NEA benchmark activity on burn up credit in the nineties was summarized in a series of publications (see, for example, [7-9]). The status of some important data used in the calculations is illustrated by the tables 4-9. There is four-fold difference in Np-237 thermal cross section values used by different participants (see Table 4) which is obviously unacceptable. The discrepancies in the properties of important fission product absorbers (cumulative yields and capture cross sections, Tables 6-9) are also prominent.

The data presented in Tables 4-5 indicate that in fact uncertainties estimated in Table 2 as "achieved" in some cases still are to be achieved. One more example of this kind is presented in Fig.1. Controversial data on the fission cross section of Cm-243 hardly support 15% accuracy labeled as "achieved" in Table 2.

Table 4. Comparison of ^{243}Am thermal capture cross sections used in NEA benchmark.

Participant	Thermal cross section, b	Participant	Thermal cross section, b
AEA	68	BNFL	40
NUPEC/INS	51	ORNL-27g	52
ORNL-44g	40	PSI	74

Table 5. Comparison of ^{237}Np thermal capture cross sections.

Participant	Thermal cross section, b	Participant	Thermal cross section, b
AEA	159	BNFL	132
NUPEC/INS	38	ORNL-27g	77
ORNL-44g	89	PSI	89

Table 6. Comparison of ^{149}Sm thermal capture cross sections.

Participant	Thermal cross section, b	Participant	Thermal cross section, b
AEA	6.15E+04	BNFL	4.50E+04
NUPEC/INS	-	ORNL-27g	4.35E+04
ORNL-44g	4.70E+04	PSI	4.16E+04

Table 7. Comparison of ^{151}Sm thermal capture cross sections.

Participant	Thermal cross section, b	Participant	Thermal cross section, b
AEA	1.25E+04	BNFL	4.38E+03
NUPEC/INS	-	ORNL-27g	4.03E+03
ORNL-44g	3.93E+03	PSI	4.16E+04

Table 8. Cumulative fission yield data for ^{151}Sm.

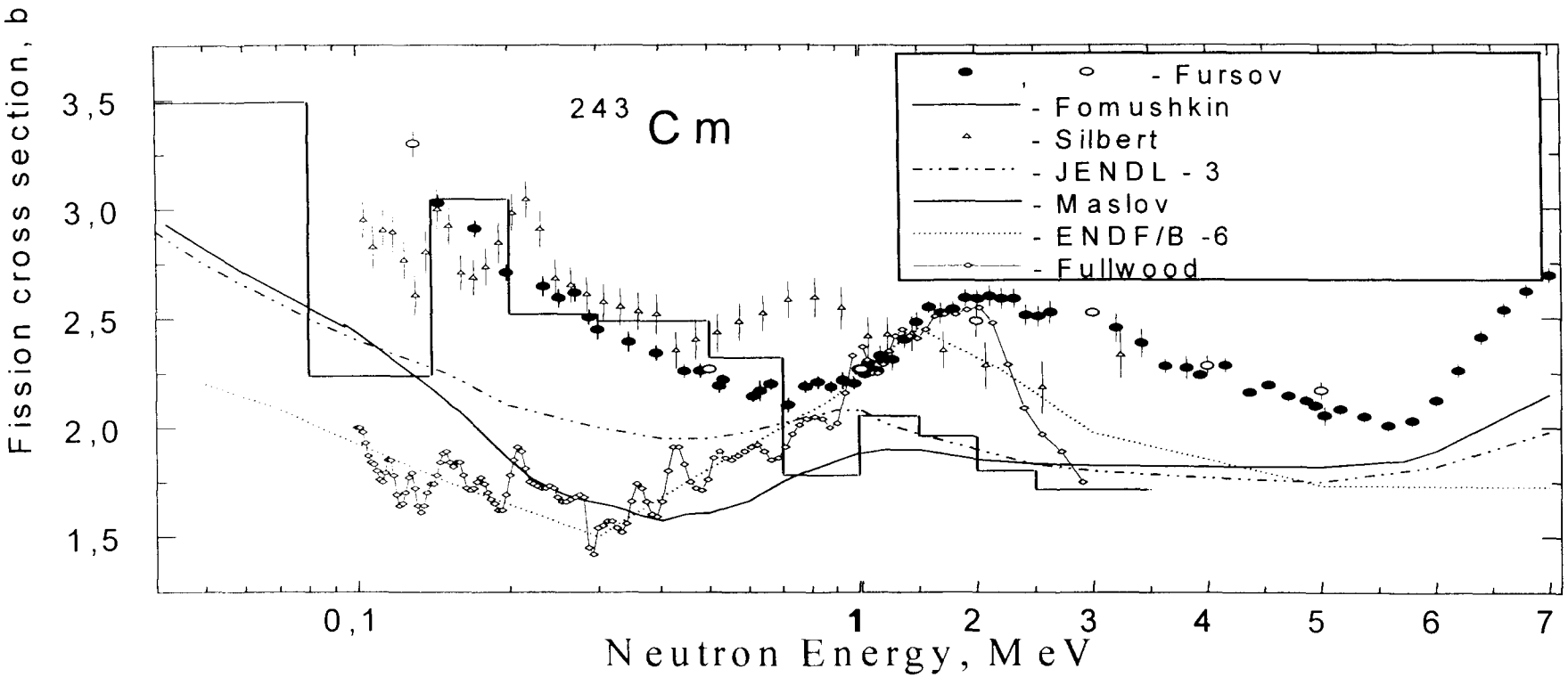
	AEA	CEA	BNFL
U-235	4.16E-03	4.16E-03	3.12E-03
U-238	8.09E-03	8.05E-03	3.31E-03
Pu-239	7.62E-03	7.55E-03	6.37E-03
Pu-241	8.55E-03	9.11E-03	7.33E-03

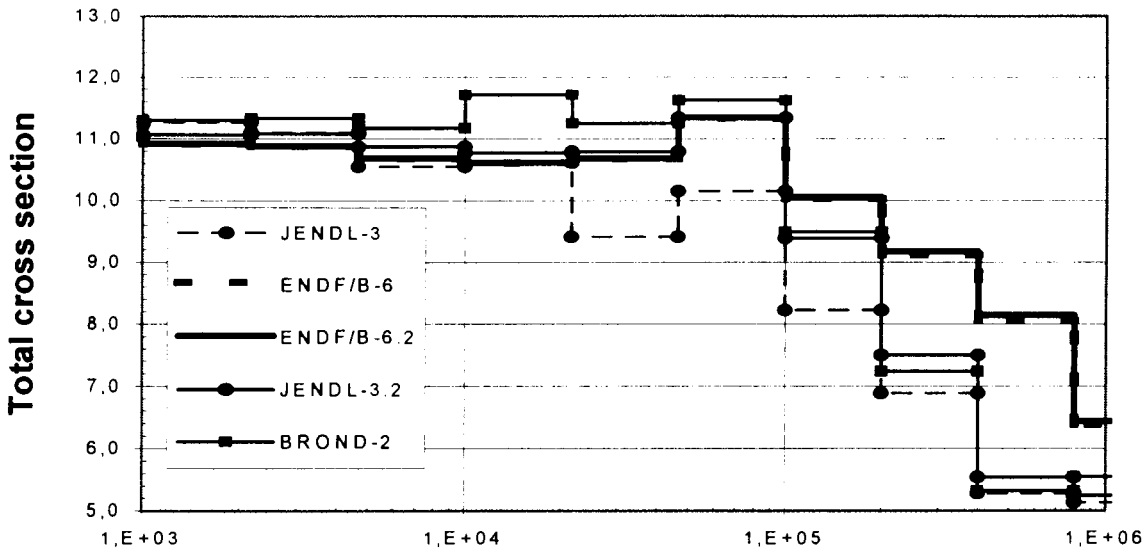
Table 9. Cumulative fission yield data for ^{149}Sm.

	AEA	CEA	BNFL
U-235	1.05E-02	1.07E-02	2.42E-03
U-238	1.66E-02	1.62E-02	1.04E-03
Pu-239	1.25E-02	1.23E-02	3.81E-03
Pu-241	1.46E-02	1.47E-02	5.63E-03

3.3. Heavy liquid metal coolant. Pure lead is rather neutron transparent substance. Its relative nuclear neutrality was probably the reason why Pb did not attract special attention of ND community until recently. The results of experiments and analysis presented in [4] indicate that there are considerable discrepancies between various nuclear data libraries on basic cross sections of lead. This situation is illustrated by Figs.2-5 and Table 10 below.

Fig.1. Experimental data on Cm-243 fission cross section.





ig. 2. Data on ^{nat}Pb total cross section taken from different ND libraries.

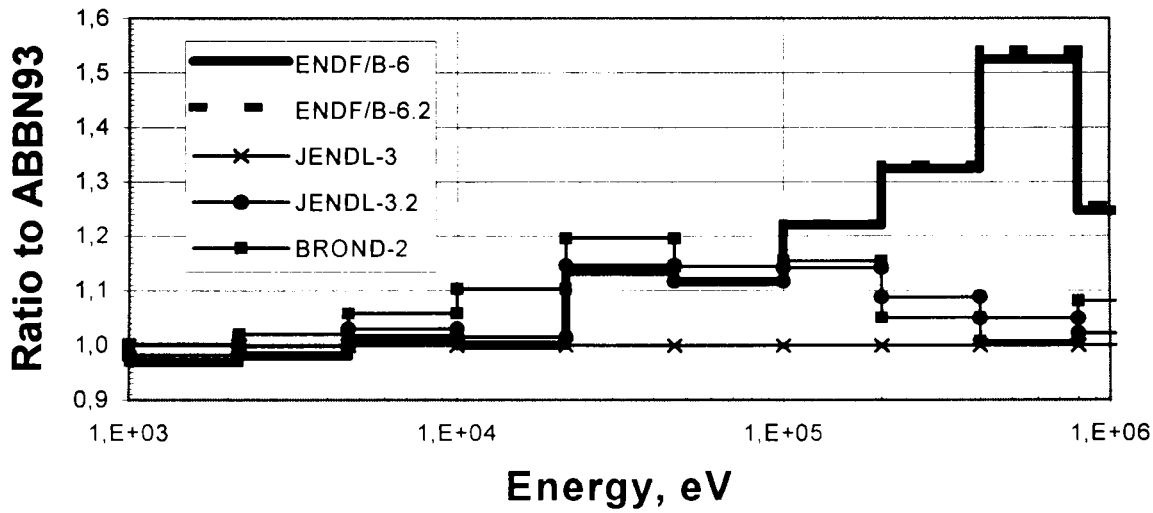


Fig. 3. The same as in previous Figure relative to ABBN93.

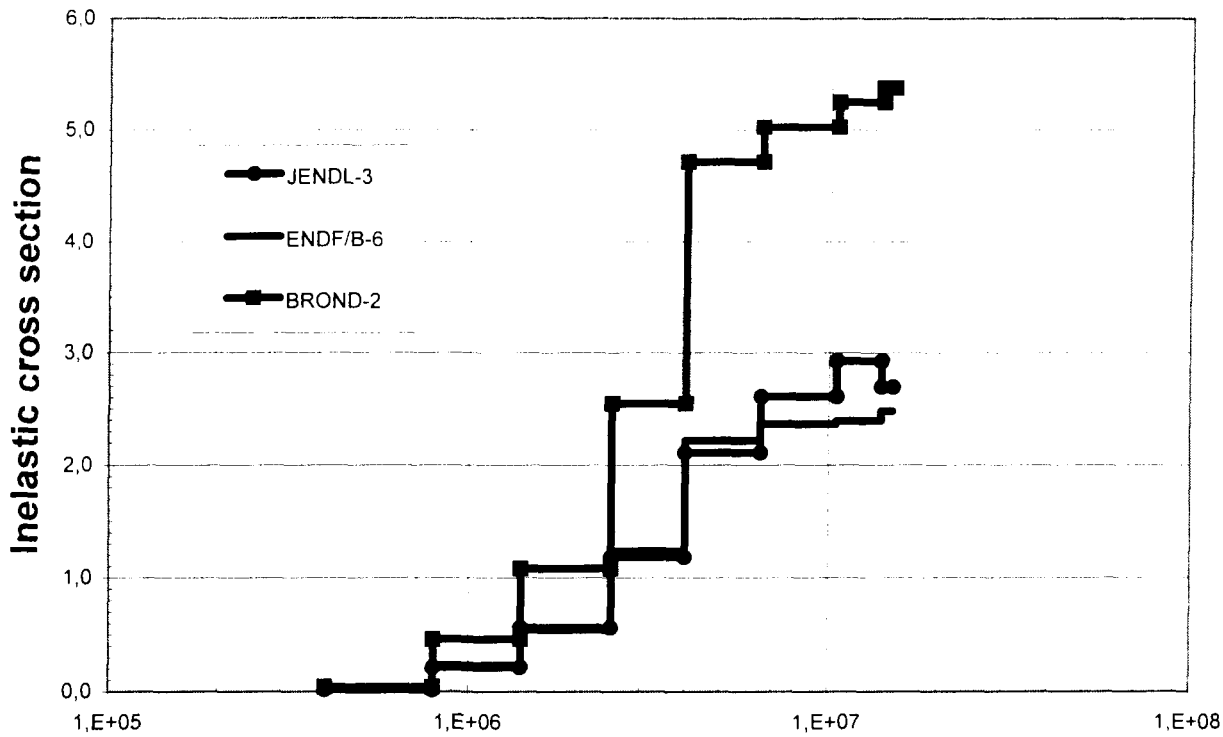


Fig. 4. Data on ^{nat}Pb inelastic cross section taken from different ND libraries.

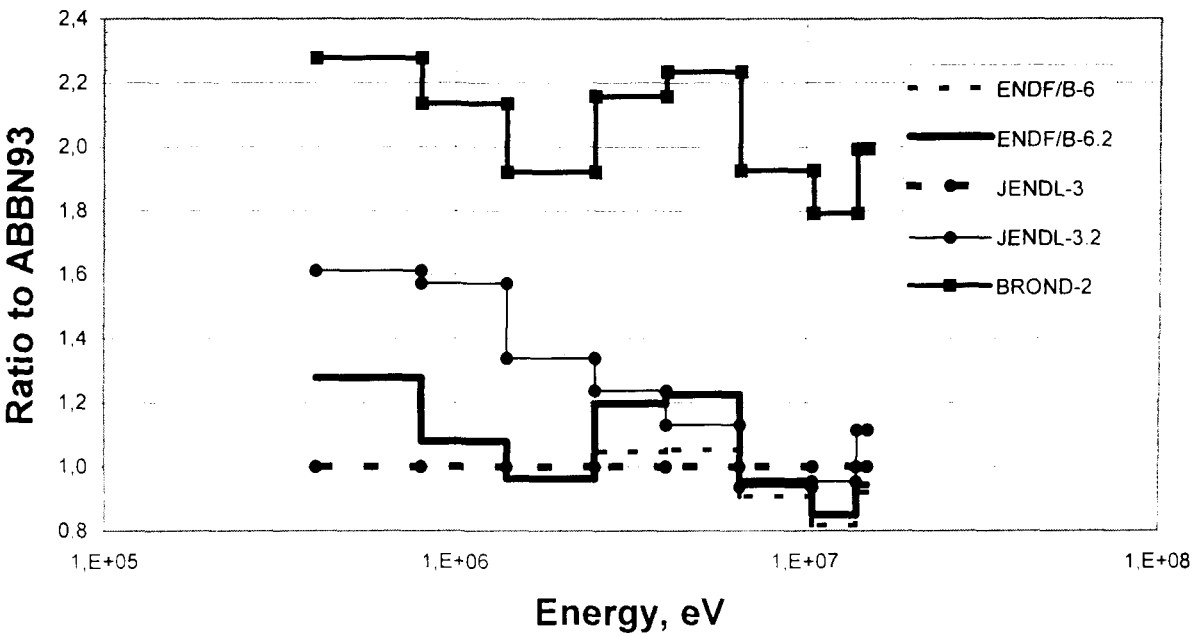


Fig. 5. The same as in previous Figure relative to ABBN93.

Table 10. C/E ratio of sigma remove for Pb^{nat} [4].

Data Library	C / E		
	$U^{238}(n,f)$	$Np^{237}(n,f)$	$Al^{27}(n,p)$
BROND-2	1.70	1.90	1.09
ENDF/B-6	1.07	1.19	0.99
ENDF/B-6.2	1.15	1.30	1.13
JENDL-3	***0.97***	***1.00***	***0.99***
JENDL-3.2	1.51	2.00	1.08

3.2. Error bands and covariances. The accuracy of calculated reactor parameters is of vital importance for safe and reliable operation of the units. The uncertainties of nuclear data used in those calculations are starting ground for any evaluation of calculations accuracy. At the same time it's widely known that standard statistical methods applied to generate error bands of evaluated ND from primary experimental information produce grossly underestimated values. This situation is probably best illustrated by comparison of purely statistical and expert evaluations of nuclear standard data [10] given in Table 11. One of the reasons of this discrepancy are highly correlated systematic errors of experimental data. The algorithms taking them into account are now being developed and there are some hopeful results both for error bands and for covariance matrices.

Table 1. The uncertainties of recommended standard cross sections. I – the results of purely statistical analysis of American Cross Section Evaluation Working Group (CSEWG). II – final expert evaluations by the members of the same Group.

$^{10}B(n,\alpha_0)^7Li$

Energy range, KeV	I, %	II, %	II/I
5 - 30	0.38	3	7.9
30 - 90	0.38	5	13.2
90 - 150	0.86	10	11.6
150 - 200	0.86	12	14.0
200 - 250	0.79	15	19.0

$^6Li(n,t)^4He$

Energy range, KeV	I, %	II, %	II/I
1 - 10	0.14	0.7	5.0
10 - 50	0.14	0.9	6.4
50 - 90	0.25	1.1	4.4
90 – 150	0.25	1.5	6.0
150 – 450	0.29	2.0	6.9

$^{197}Au(n,\gamma)^{198}Au$

Energy range, KeV	I, %	II, %	II/I
200 - 500	1.31	3.0	2.3
500 - 1000	2.10	3.5	1.7
1000 - 2500	2.00	4.5	2.3

$^{235}U(n,f)$

Energy range, KeV	I, %	II, %	II/I
150 - 600	0.19	1.5	7.9
600 - 1000	0.60	1.6	2.7
1000 - 3000	0.60	1.8	3.0
3000 - 6000	0.69	2.3	3.3
6000 – 10000	0.69	2.2	3.2
10000 - 12000	1.14	1.8	1.6
12000 - 14000	1.14	1.2	1.1
14000 - 14500	0.55	0.8	1.5
14500 - 15000	0.55	1.5	2.7
15000 - 16000	0.97	2.0	2.1

CONCLUSIONS

1. Revision and improvement of nuclear data for fast reactors are needed, progress in reactor calculations accuracy due to better codes and much faster computers increases relative importance of the input data uncertainties.
2. Better accuracy is especially important in the case of small reactivity margin and fuels of equilibrium compositions characteristic for the cores of enhanced safety and proliferation resistance.
3. The main objects of R&D efforts should be minor actinides and heavy liquid metal coolant where data discrepancies still are large.
4. Data error bands and covariance information also gains importance as necessary component of neutron physics calculations so adequate methods of their generation should be developed.

REFERENCES

1. White Book of Nuclear Power. General Editor Prof. E.O.Adamov. First Edition. Edited by RDIPE, Moscow, 1998.
2. A.V.Lopatkin, V.V.Orlov. Fuel cycle of a New Generation of Fast Reactors Based on the Principles of Non-Proliferation and Radiation Equivalent Disposition of Radwaste. Paper presented to International Seminar "Cost Competitive, Proliferation Resistant, Inherently and Ecologically Safe Fast Reactor and Fuel Cycle for Large Scale Power" Ministry of Russian Federation for Atomic Energy, Moscow, 2000. 29 May-1 June 2000.
3. V.M.Poplavsky et al. On Real and Imagine Dangers of Fast Reactor Sodium Coolant. Paper presented to 10-th Annual Conference of the Russian Nuclear Society, Obninsk, 28 June - 3 July, 1999.
4. I.P.Matveenko, A.M.Tsyboulya, G.N.Manturov, M.Yu. Semenov, V.N.Koscheev (IPPE), V.S.Smirnov, A.V. Lopatkin, V.G.Muratov (NIKIET), P.N.Alexeyev (RRC Kurchatov Institute). Experimental Studies of BREST-OD-300 Reactor Characteristics on BFS Fasilities. Paper presented to International Seminar "Cost Competative, Proliferation

Resistant, Inherently and Ecologically Safe Fast Reactor and Fuel Cycle for Large Scale Power". Ministry of Russian Federation for Atomic Energy, Moscow, 2000.

5. V.N.Koscheev, G.N.Manturov, M.N.Nikolaev et al. Nuclear Data for Plutonium and Minor Actinides. - In Proc. of the 3rd Intern. Conf. on Accelerator Driven Transmutation Technologies and Applications, Praha 1999, Czech Republic, p. 1194.
6. V.I.Golubev, V.V.Dolgov, V.A.Dulin et al. Investigations of Actinides Transmutation in Fast Reactors. Atomnaya Energiya, 1993, v.74, N1.
7. M.C. Brady, M. Takano, M.D. DeHart, H. Okuno, A. Nouri, E. Sartori . Findings of the OECD/NEA Study on Burn-up Credit. Paper presented at Physor'96, Mito, Japan.
8. M.C. Brady, H. Okuno, M.D. DeHart, A. Nouri, E. Sartori. International Studies on Burn-up Credit Criticality Safety by an OECD/NEA Working Group. Paper presented at the Intl. Conference on the Physics of Nuclear Science & Technology, Long Island, NY, 5-8 Oct. 1998.
9. M.D. DeHart (ORNL), M.C. Brady (SNL), C.V. Parks (ORNL) OECD/NEA Burn-up Credit Calculational Criticality Benchmark Phase-IB: Isotopic Prediction: Final Report. June 1996, ORNL-6901, NEA/NSC/DOC(1996).
10. "Nuclear Data Standards for Nuclear Measurements", NEA/OECD, 1992.

Data Needs for Modeling of Fuel Cycle Concepts for Fast Lead Cooled Reactors

N.P. Kocherov, A.A. Rimski-Korsakov
V.G. Khlopin Radium Institute
St. Petersburg, Russia

Abstract

Calculations of different conceptual schemes of external fuel cycle for future development of atomic power industry within the framework of Minatom strategy plans for the beginning of the XXI century are described. Requirements for accuracy of nuclear data needed for such calculations are described.

In May 2000 the Government of Russia has endorsed the document "Strategy of Nuclear Power Industry Development in the First Half of the XXI Century" /1/. According to this document the main activities in the period till 2010 will be concentrated in the following areas:

- Extension of service life of nuclear power reactors;
- Completion of construction of reactors which are now under construction with a high or medium degree of readiness;
- Construction of third generation reference power units (VVER-A-1000 and BN-800) at existing sites;
- Reconstruction of the nuclear spent fuel recycling plants, enlargement of fuel storage facilities both for liquid and dry storage of spent fuel;
- Construction of liquid sodium BN-800 power reactor with nitride fuel;
- R&D work on closed fuel cycle technologies;
- Development and construction of a demonstration reactor unit with intrinsic safety (fast reactor with liquid lead coolant BREST-A-300 and experimental fuel cycle facilities for it).

A large volume of calculations will be needed especially for the last two items of this program. It is due to the fact that the BREST-A-300 reactor differs in many ways from presently existing fast reactors. This makes it necessary to reconsider a number of aspects, including technology of spent fuel recycling. The fuel of a BREST reactor during the initial charging consists of depleted uranium nitride (85%) and plutonium nitride recycled from nuclear power reactor fuel (15%). After the end of reactor campaign its fuel is recycled, only fission products are separated and the rest is used as a fuel for the next cycle. Up to 20 cycles are considered. During recycling natural or depleted uranium nitride is added as a feeding material for electricity production. This type of recycling leads to build up of minor actinide quantities till they reach equilibrium values. In this case the equilibrium concentration of minor actinides (MA) in the fuel is about 0.7% of heavy atoms, their contribution to the total number of fission events in the reactor is about 1%. It is possible to add some quantities of MA from thermal reactors during recycling. In this case BREST reactor will operate as a transmuter of MA. MA content in the fuel will increase. It was estimated that the MA content could be increased up to 5% of heavy atoms without deterioration of the safety parameters of the reactor. The contribution of MA fission events will increase in this case up to 9% /2/.It should be noted that significant amounts of MA will stay permanently in the reactor core (stored), and after the end of the service life they should be transferred to another installation for transmutation.

In order to find optimal schemes of organizing the fuel cycle processes for nuclear energy systems, consisting of thermal and fast reactors, it is necessary to make calculations also for other possible types of installations for transmutation of MA. There were many proposals to use thermal and fast reactors, special molten salt reactors and accelerator driven systems for this purpose.

Calculations of accumulation and burnout of radioactive nuclides are usually performed with the help of computer codes such as Origen2 /3/ or Corout /4/ or other. Cross-section libraries for these codes are specially prepared in the form of one or two group spectrum averages. Neutron spectra are calculated by Monte Carlo computer programs for needed locations in reactor installations. To calculate the dynamics of accumulation and burnout of MA spectrum averages for (n,γ), (n,f), (n,2n), (n,3n), (n,γ)Ex, and (n,2n)Ex (Ex – isomer production) reaction cross-sections for for 40 nuclides from thorium to californium are needed. For these 150 cross-sections experimental data exist only for 70 – 80. In other cases one has to use the results of theoretical model calculations, the accuracy of the latter is not sufficiently high. The accuracy of calculated averages depends both on the quality of initial cross-section data as well as on the accuracy of the neutron spectra. This dependence is clearly seen in Table 1, where spectrum averaged cross-sections calculated with one set of initial cross-section data (ENDFB) for reactor installations with different neutron spectra are given.

Table 1. Fission and capture cross-sections for some nuclides averaged over different reactor neutron spectra.

Nuclide/ Reactor	Np-237		Am-241		Am-242m		Am-243	
	σ_{cap}	σ_f	σ_{cap}	σ_f	σ_{cap}	σ_f	σ_{cap}	σ_f
Thermal*	181	0.018	621	3,15	1349	6652	75,34	0,074
Candu**	58,6	0,29	314,7	2,19	849	4166	43,8	0,065
Pwru**	32,1	0,52	105,8	1,12	98	466	6,49	0,396
Lmfbr**	1,43	0,39	1,32	0,345	0,364	3,9	0,55	0,472
Brest***	0,39	0,92	0,405	0,79	0,04	2,39	0,373	0,58

Nuclide/ Reactor	Pu-238		Pu-239		Pu-240		Pu-241		Pu-242	
	σ_{cap}	σ_f	σ_{cap}	σ_f	σ_{cap}	σ_f	σ_{cap}	σ_f	σ_{cap}	σ_f
Thermal*	563	17,1	271,4	748	287,6	0,064	361,3	1012	19,3	0,001
Candu**	142,	5,09	123	267	144	0,33	116	339	11,8	0,025
Pwru**	34,7	2,46	58,6	106,2	104	0,58	38,6	181,1	8,7	0,14
Lmfbr**	0,69	1,19	0,47	1,8	0,48	0,42	0,44	2,44	0,4	0,31
Brest***	0,24	1,65	0,087	1,68	0,13	0,95	0,14	1,65	0,12	0,76

Nuclide/ Reactor	Cm-242		Cm-243		Cm-244		Cm-245		Cm-246	
	σ_{cap}	σ_f	σ_{cap}	σ_f	σ_{cap}	σ_f	σ_{cap}	σ_f	σ_{cap}	σ_f
Thermal*	16,9	3.0	58,2	693	10,4	0,6	343	2228	1,3	0,06
Candu**	12,1	1,68	36,4	416	23,2	0,7	201,5	1143	2,56	0,185
Pwru**	5,8	0,56	8,39	71,7	13,8	0,87	29,1	171	2,9	0,58
Lmfbr**	0,31	0,2	0,23	2,6	0,79	0,49	0,3	2,6	0,22	0,32
Brest***	0,05	0,46	0,04	2,0	0,24	1,16	0,08	1,6	0,07	0,83

The cross-sections were taken from ENDFB-6 (*), Origen2 (**) libraries and calculated by us for Corout code (***) using BREST neutron spectrum /5/. As can be seen from the Table the effective cross-sections change by hundred times. Therefore the accuracy of neutron spectra can significantly change the results of spectrum average calculations. Data on neutron spectra which were obtained during design stages or in special experiments often are not published in journals and are difficult to obtain in digital form. It would be very useful to compile information on neutron spectra in different reactor installations in the form of an atlas with graphs and numerical data. Such data would be very helpful for specialists performing conceptual studies of technological schemes of nuclear fuel cycle.

A similar approach could also be used for calculations of dynamics of nuclide accumulation and burnout for homogeneous transmutation of MA, if their quantities in the fuel are not large, and their contribution to the total number of fission events per unit volume does not exceed 10 – 15%. Homogeneous transmutation means that MA are evenly distributed in the reactor fuel. Such an approach usually does not cause much trouble from the point of view of reactor safety, but becomes inconvenient during the radiochemical treatment of spent fuel and refabrication. In this case the radioactivity of fuel increases considerably due to higher concentrations of MA and accumulation of Pu-238 in the process of irradiation in reactors, and all technological processes during chemical treatment and refabrication should be remotely controlled.

Alternative, so called heterogeneous, approach means that the transmuted material is placed in separate pellets or capsules which are irradiated in blankets or as separate fuel elements in the core. The chemical reprocessing of these materials after irradiation is performed in separate cells, thus eliminating the need of multiple recycling of large volumes of fuel with increased concentration of MA.

According to our calculations the specific heat, generated in these elements nearly equals that in the standard fuel elements both in thermal and fast neutron spectra, being somewhat higher in the latter. This means that for safety calculations of the behavior of such elements in the case of heterogeneous transmutation it is necessary to have initial nuclear data for Np-237, Am-241, 242m, 243 with the same accuracy as for the fuel constituents. In the existing technologies of spent fuel treatment (Purex process and its modifications) neptunium is separated as in one fraction and americium and curium stay together in another.

Therefore in fast reactors cross-sections for Np-237 should be known with the following accuracy: $\sigma(n,f)$ – 1%, $\sigma(n,\gamma)$ – 3%, $\sigma(n,n')$ – 5-10%, $\sigma(n,2n)$ – 10%, $\sigma(^1H,^3H,^4He$ production) – 15%, and ν - 0.5%. These requirements for the accuracy of data for the main fuel nuclides were formulated in /6-9/.

The americium – curium fraction consists approximately of 62% Am-241, 29% Am-243 and of 9% curium, its composition depends on the details of reactor campaign and duration of storage. Therefore the accuracy requirements could be formulated as follows.

For Am-241: $\sigma(n,f)$ – 2-3%, $\sigma(n,\gamma)$ – 6-8%, $\sigma(n,n')$ – 10-20%, $\sigma(n,2n)$ – 20%, $\sigma(^1H,^3H,^4He$ production) – 30%, and ν - 1%.

For Am-243: $\sigma(n,f)$ – 3%, $\sigma(n,\gamma)$ – 10%, $\sigma(n,n')$ – 20-30%, $\sigma(n,2n)$ – 30%, $\sigma(^1H,^3H,^4He$ production) – 45%, ν - 1.5%.

No definite decision on the fate of curium was made. It can be separated from americium and stored for a long period, or be transmuted together with americium. In

the latter case accuracy requirements for curium will be less stringent than for americium.

The accuracy of cross-section data in evaluated libraries are lower than what is required. Especially unsatisfactory is the evaluations of ν for these nuclides which is usually based on Madland-Nix model calculations and errors are not defined. It is important to use experimental database more extensively and determine the precision. New measurements are still needed.

References.

1. Strategy of Development of Atomic Power in Russia in the First Half of XXI century, MINATOM, Moscow, 2000.
2. E.O. Adamov, I.H. Ganev, A.V. Lopatkin et al., Transmutational Fuel Cycle for Large Scale Nuclear Power in Russia, Report NIKIET, Moscow, 1999.
3. A.G. Croff, A User's Manual for the Origen2 Computer Code, Report ORNL-TM-7175, (1980).
4. A.A. Rimski-Korsakov, A.S. Roschin, S.G. Yavshits, Computer Code for Radioactive Inventory Calculations, Report INDC(NDS)-355/R, p. 89, IAEA, Vienna, 1996.
5. A.V. Lopatkin, Private Communication, 1998.
6. S.M. Zaritskij, M.F. Troyanov, Proc. Conf. On the Physics of Fast Reactors, p. 261, BNES, London, 1969.
7. S.M. Zaritskij et al., Proc. Conf on Neutron Physics, v. 1, p. 53, 1973, Kiev.
8. Yu.G. Bobkov, L.N. Usachev, Nuclear Constants, v. 16, p. 5,1975.
9. L.N. Usachev et al., Proc. Conf. On Neutron Physics, v. 1, p. 64, 1976, Kiev.

Paper presented in IAEA AGM on Long Term Needs for Nuclear Data Development,
28 November to 1 December, 2000
At IAEA, Vienna

Indian Advanced Heavy Water Reactor for Thorium Utilisation and Nuclear Data Requirements and Status

R. Srivenkatesan, Umasankari Kannan, Arvind Kumar,
S. Ganesan and S.B. Degwekar
Bhabha Atomic Research Centre
Mumbai 400 085
INDIA

Abstract

BARC is embarking on thorium utilisation program in a concerted and consistent manner to achieve all round capabilities in the entire Thorium cycle under the Advanced Heavy Water Reactor (AHWR) development program. Upgrading our nuclear data capability for thorium cycle is one of the main tasks of this program.

This paper gives a brief overview of the physics design features of the AHWR.

The basic starting point of the analysis has been the lattice simulation of the fuel cluster employing the WIMS-D4 code package with 1986 version of 69 group library. For the analysis of thorium cycle, the present multi group version contains the three major isotopes viz., ^{232}Th, ^{233}U and ^{233}Pa. To correctly evaluate the fuel cycle we require many more isotopes of the Th burnup chain. With the help of NDS, IAEA many other isotopes of interest in AHWR, actinides in the thorium burnup chain, burnable absorbers, etc., were generated. Some of them were added to the WIMS-D4 library and the results are discussed.

The WIMSD-4 library is also being updated as part of the IAEA coordinated research project on Final Stage of WLUP with international cooperation. India is also taking part in CRP. The evaluation of AHWR lattice with this new library is presented.

Some comments regarding the fission product data being used in WIMS libraries are given, which are tuned to U-Pu cycles. The measurements for ^{233}U are rather old. Measurements in high energies are also very sparse. More attention by nuclear data community is required in this regard as well.

India has also begun a modest program to assess the ADS concepts, with the aim of employing thermal reactor systems, such as AHWR. A one way coupled booster reactor concept is being analysed with available code systems and nuclear data. A brief summary of this concept is also being discussed in this paper.

A general survey on the quality of the evaluated nuclear data of the major and minor isotopes of thorium cycle is also given. A major international effort is necessary to bring the nuclear database for the uranium-thorium cycle up to the reliability level of that for the uranium-plutonium fuel cycle.

1. Introduction

The long term objective of thorium utilisation on a large scale is one of the most distinguishing features of the Indian Nuclear Power Program. The inherent nuclear charcteristics of ^{233}U, particularly the eta value in thermal energies make heavy water reactors as good option for thorium utilisation. Due to our extensive experience in PHWRs we would like to keep the technological advantages of reactor design and operation. At the same time one should build-in new safety features to be consistent with the modern perceptions of advanced reactors. Hence the AHWR is being developed in India for utilising thorium for power generation with passive safety features. [1,2]

AHWR is a vertical pressure tube type reactor cooled by boiling light water and moderated by heavy water. The reactor has been designed to produce 750 MW(th) at a discharge burnup of about 25,000 MWd/te. Negative void coefficient has to be achieved, in spite of using boiling light water coolant. Another design objective is to be self-sustaining in ^{233}U with most of the power from the conversion of thorium fuel while using plutonium as the external fissile feed. Also one has to minimise the external Pu feed in the equilibrium fuel cycle, such that major fraction of power is produced by the in situ conversion of Th and burning of ^{233}U.

AHWR incorporates several advanced passive safety features, e.g., heat removal through natural circulation, ECCS injection directly into the fuel cluster through a dedicated water channel inside the cluster, passive containment isolation etc. [2]

In this paper after briefly summarising the physics design features of AHWR, the current methodologies used in the lattice physics simulations and nuclear data status are discussed. Then we comment on the nuclear data requirements for the thorium cycle in general.

2. Physics Design Features of AHWR

The fuel design of AHWR has followed an evolutionary path ranging from a seed and blanket concept to a simplified composite cluster to achieve a good thermal hydraulic coupling. The composite cluster design also has changed from a square cluster to a circular cluster inside a circular pressure tube. [3]

The aim was to design a single cluster, which is capable of achieving a negative void coefficient as well as satisfy the thermal hydraulic requirements. If all the fuel assemblies are similar and the core has a flatter power distribution, it would be possible to achieve optimised coolant conditions from the point of view of heat removal by natural circulation. The power density should also be low to get optimum heat flux ratios with the boiling coolant driven by natural circulation.

With this in view, a composite cluster has been designed using two kinds of fuel namely, (Th-^{233}U) MOX and (Th-Pu) MOX [3,4]. Plutonium has to be kept in the outer pins of a fuel cluster facing the moderator from reactivity and void coefficient considerations, while ^{233}U can be kept inside in a relatively harder spectrum without losing reactivity.

The ^{233}U content in the composite cluster has to be higher than the self-sustaining limit of about 1.5 % from reactivity as well as local peaking factor considerations. The overall self sustaining feature of ^{233}U is achieved by its production in the outer (Th-Pu) MOX pins.

The void reactivity is made negative in spite of using light water coolant by incorporating a large central (coolant displacer) rod with a slow burning absorber such as dysprosium – about 3 wt% dysprosium oxide in zirconium oxide. The cluster design is given in Fig. 1. The displacer rod contains a central hole through which the ECCS water is injected from the top and the water jets emerge all along the cluster. The use of such an absorber rod compromises the excellent neutron economy of a HWR, but offers inherent safety, which is the overriding principle.

The core consists of 500 lattice locations, of which 452 will contain fuel, while the rest of the channels are used for Shut Down System –1 and control (adjuster) rods. The core has a height of about 5 meters, of which the fuel length is 3.5 m. The core diameter is about 8 m. The core layout is given in Fig. 2.

The void reactivity of the core is nearly zero under nominal operating conditions, being within –3 mk for coolant density variation from inlet value of about 0.75 to 0.05 gm/cc. The coolant temperature coefficient variation of about +3 mk from cold to hot-stand-by condition, and a negative power coefficient of about 6 mk from hot-stand-by to hot operating condition, constitute the parameters that will dictate stability of the reactor.

An important aspect of AHWR is also its Pu burning capability. The following table shows the initial and discharge vectors of Pu isotopes. The initial charge is from the reprocessed Pu from Indian PHWR employing natural uranium fuel (average exit burnup of about 6500 MWD/t).

	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}
Initial	68.8 %	24.6 %	5.3 %	1.3 %
Discharge	10.3 %	51.9 %	19.9 %	17.9 %

With each recycle of ^{233}U, the amount of ^{232}U will increase leading to inconvenient handling problems. Suitable development strategies are being evolved to cope with these problems. AHWR also offers an option to burn some minor actinides such as ^{231}Pa, ^{237}Np etc. Some results of our analysis are given in § 3.2.

We are also planning a low power critical facility, which will validate our simulation strategies for the novel thorium fuel including nuclear data employed.

3. Calculational methodology and strategies for AHWR

The WIMS D4 code and library has been used for lattice simulation of AHWR. This 69 group library has been extensively validated for the plutonium-uranium cycles. The thorium burnup chain is shown in Fig. 3. The 1986 version WIMS library we have used

has data tables only for three isotopes of the thorium cycle namely, ^{232}Th, ^{233}Pa and ^{233}U. The WIMS library released this year as part of the IAEA-CRP also contains only these isotopes [5]. This is not sufficient to fully analyse the fuel cycle issues of thorium cycle or AHWR. Since AHWR will operate in a closed fuel cycle, we have to recycle the uranium isotopes generated in AHWR. This demands the assessment of shielding for the remote handling facilities for reprocessing of the discharged fuel and fabricating new clusters for future fuel loading.

Although from the spent fuel point of view the generation of higher actinides is less than that of the uranium cycle, the isotopes having shorter half lives and significant absorption cross section cannot be ignored. For example, ^{232}U is important from the point of view of spent fuel handling as the daughter products of ^{232}U are highly radioactive. The concentrations of ^{232}U has to be determined with at least 10 % accuracy [6]. ^{232}Pa is another isotope which has got significant fission cross section for thermal neutrons ~700 barns. (There are some difference of opinions about the significance of this isotope.) Other isotopes of radioactivity significance in these processes are ^{228}Th, ^{229}Th, ^{230}Th, ^{231}Pa, ^{234}U etc.

One normally employs a code like ORIGEN for end-of-cycle mass and activity estimations, for fission products and actinides, as a function of burnup and storage time. Such simulations are also used for toxicity evaluations. But there are always large differences in the masses during burning in a code like ORIGEN which uses one group cross-sections with homogeneous fuel distribution and a multi group lattice code like WIMS which simulates the cluster explicitly. However the list of isotopes (FP and actinides) in the lattice code library is tailored to only those which have reactivity implications. To meet the current demands of integrated design, the front as well as back end of the fuel cycle requirements have to be explicitly addressed in nuclear data also. Such a trend is observed in the ADSS studies. Often these new studies use direct Monte Carlo core simulations with burnup with about a thousand nuclei and point data directly from evaluated libraries. [7] They often combined the Monte Carlo codes such as MCNP with versions of ORIGEN itself to simulate burnup [8].

Our strategy is to include the above isotopes in the WIMS library, - the present as well as the WLUP versions – generate the masses of actinides at discharge and analyse the long term decay characteristics with ORIGEN. The WLUP library contains more isotopes (see § 3.2). If we include the other isotopes we have generated to WLUP library, our current requirements would be satisfied to a large extent.

3.1 Lattice evaluations

We discuss here some of our work in dealing with the nuclear data for AHWR

- The burnup chain of Thorium fuel cycle has to be expanded taking into account the other significant isotopes such as ^{232}U, ^{231}Pa ^{232}Pa etc. This is recognized as an essential task under the Task force on Nuclear Data for AHWR in BARC. For our immediate requirement of AHWR analyses, some of these isotopes have been processed at the IAEA and is to be incorporated into the present WIMS library. The

isotopes of the thorium cycle namely, ^{232}Th, ^{230}Th, ^{231}Pa, ^{232}Pa, ^{233}Pa, ^{232}U, ^{233}U and ^{234}U were processed using the NJOY code system at the Nuclear Data Section of the IAEA and the multigroup cross section set in the WIMS library format have been generated [9].

- Currently the fission spectrum is the same for all isotopes, which is a restriction from the conventions in WIMSD-4 code and library. Basic libraries do contain them.
- As mentioned above dysprosium is used in the AHWR cluster designs. The WIMS library contains only ^{164}Dy, which has an abundance of 28.2% only. The other isotopes ^{160}Dy (2.34%), ^{161}Dy (18.9%), ^{162}Dy (25.5%) and ^{163}Dy (24.9%) have significant resonances and affect the design parameters. These were also generated in IAEA during the above exercise. In a typical cluster design study we found that the amount of Dy required reduces from 3.5wt% to 3 wt% if one uses all the isotopes [10].
- Also there is a proposal to use isotopically tailored/denatured Zirconium by removal of ^{91}Zr in order to reduce the reactivity loads in PHWRs. ^{91}Zr alone is present in the WIMS library. In order to estimate the burnup penalty, the nuclear data of individual isotopes of Zircaloy have to be treated explicitly. This is applicable to U-Pu cycles as well.

3.2 Simulations with WLUP Library for AHWR lattice

As part of the on-going CRP of IAEA for Final Stage of WIMS Library Update Project, two WIMS libraries based on ENDF-B/6 and JEF 2.2 have been distributed. These libraries have over 140 elements comprising of 21 actinides with Americium and Curium isotopes built in the burnup chains of plutonium. Also, there are about 40 individual fission products. Isotopes like, Niobium, Magnesium have also been included as a result of requests from participating countries. A thorough review of the input options of the data processing code NJOY has been done under the WLUP. The actinide profile of U-Pu cycle can thus be estimated more accurately than the earlier libraries. As mentioned above our task is to add more isotopes for thorium cycle.

In BARC, Pa^{231} and U^{232} have been introduced into the WIMS-1986 library. This required a special procedure as these isotopes are basically formed by (n,2n) reactions and (n,2n) reactions are not present explicitly in the WIMS library structure. The production of these isotopes has been accounted for by introducing spectrum dependent reaction rate ratios for their buildup as a pseudo fission product. Similar exercise has to done with WLUP library as well.

3.2.1 Simulations with AHWR D-3 cluster

The WLUP libraries were benchmarked with the AHWR D-3 cluster *(this cluster design has since been given up due to its high positive reactivity at higher burnups; See Ref. 4 for details)*. The D-3 cluster earlier designed for the AHWR consists of 52 pins placed in a square array. The inner 32 pins are composed of (Th-^{233}U) MOX pins with a ^{233}U enrichment of 2.94 % and the outer 20 consist of (Th-Pu) MOX pins with a plutonium enrichment of 2.7 %. A burnup analysis was done to study the reactivity, void reactivity

and the isotopic compositions for this cluster using the WIMSD-4 code system with the different WLUP libraries obtained. The results are tabulated in Table 1.

Table 1. Analysis of AHWR lattice with different datasets

Parameter	ENDF-B/6 (WLUP-2000)	JEF 2.2 (WLUP-2000)
K-infinifty 0.0 MWd/Te	1.23227	1.24212
8000.0 MWd/Te	1.09029	1.09833
20000.0 MWd/Te	0.97252	0.97765
K-effective 0.0 MWd/Te	1.19652	1.20589
8000.0 MWd/Te	1.05785	1.06539
20000.0 MWd/Te	0.94259	0.94740
Void Reactivity (ΔK_{eff}) mk (100 %change)		
0.0 MWd/Te	-1.8	+0.75
8000.0 MWd/Te	+6.1	+8.99
20000.0 MWd/Te	+18.4	+22.04
Isotopic compositions wt% U^{233} in (Th,U^{233})MOX pin		
0.0 MWd/Te	2.94	2.94
8000.0 MWd/Te	2.61	2.61
20000.0 MWd/Te	2.18	2.16
Pu^{239} 0.0 MWd/Te	1.857	1.857
8000.0 MWd/Te	0.707	0.709
20000.0 MWd/Te	0.087	0.087
Pu^{240} 0.0 MWd/Te	0.664	0.664
8000.0 MWd/Te	0.746	0.748
20000.0 MWd/Te	0.483	0.486
U^{233} in (Th,Pu)MOX pin		
0.0 MWd/Te	0.0	0.0
8000.0 MWd/Te	0.526	0.505
20000.0 MWd/Te	1.058	1.043

The JEF 2.2 data set seems to be consistently giving a higher reactivity and the difference is maintained through the burnup regime. The isotopic vector does not show much

deviation between the two libraries. Similar results have been reported earlier by Jagannathan et al for another reactor type based on thorium. [5,11]

3.2.2 Actinide profile of PHWR cluster

Using the WLUP library, the actinide profile of 19-rod PHWR cluster at discharge was calculated in order to decide on the plutonium feed enrichment required for the AHWR cluster. The average discharge burnup for PHWR cluster was taken to be 7000 MWd/Te and the actinide concentrations were generated. The plutonium isotopes contribute about 97% of the actinides. The rest are mainly composed of the Neptunium isotopes, the long lived isotope, ^{237}Np contributes 0.8%, and ^{239}Np which has a very short half-life contributes about 2.1%. The ^{238}Pu constitutes 0.13% of the total plutonium. We are also studying repcyclin issues whereby such isotopes can be added along with Pu into AHWR. The reactivity penalty due to other actinides of PHWR along with plutonium used in AHWR lattice is about 1.5 to 2.0 mk.

Table 2. Actinide profile (wt %) of PHWR fuel at discharge

Burnup	U^{234}	U^{235}	U^{236}	U^{237}	U^{238}	Np^{237}
0.0		0.71			99.29	
4000.0	1.07E-7	0.3818	4.98E-2	1.27E-4	98.897	1.32E-3
7000.0	9.68E-7	0.206	7.55E-2	1.69E-4	98.494	3.20E-3

Burnup	Pu^{238}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}	Am^{241}
0.0						
4000.0	1.1E-4	0.2007	0.0426	8.04E-3	9.61E-4	4.38E-5
7000.0	4.96E-4	0.26002	0.10515	0.025	7.04E-3	2.68E-4

Burnup	Am^{243}	Cm^{242}	Cm^{244}
0.0			
4000.0	1.99E-5	3.63E-6	7.19E-7
7000.0	3.32E-4	4.70E-5	2.68E-5

In Table 3 we give the results for the AHWR D-3 cluster using the above actinide profile of PHWR. The Pu isotopic composition has to be adjusted to get the same initial reactivity without other actinides.

We notice in Table 3 that ^{237}Np reduces by a factor of two in the discharged fuel. However Am and Cm isotopes increase. These isotopes will have to be again recycled in AHWR. However there will be reactivity penalty especially due to ^{241}Am. These studies have to be done in more detail.

Table 3 Actinide in AHWR D-3 cluster

Actinide	0.0 MWd/te wt %	25,000 MWd/Te wt %
^{237}Np	0.021	0.010
^{238}Pu	0.003	0.017
^{239}Pu	1.767	0.028
^{240}Pu	0.714	0.348
^{241}Pu	0.170	0.153
^{242}Pu	0.048	0.222
^{241}Am	0.002	0.013
^{243}Am	0.002	0.051
^{242}Cm	3.20E-4	4.04E-3
^{244}Cm	1.82E-4	0.018

We have also added magnesium to the WIMS-1986 library. Natural Mg is used as dopant in Thorium fuel for ease in dissolution during reprocessing. Estimated of the penalty is about 2 mk.

The pressure tube material in PHWRs and AHWR is an alloy of Zirconium and 2.5% Niobium. The Nb cross sections are not present in the WIMS-1986 library we are using. However it is available in the WLUP version. Inclusion of Nb data gives a reactivity penalty of about 4 mk, compared to the current practice of increasing the zirconium density by about 5% based on thermal cross sections.

3.3 Fission Product Data

The fission product yield data in the WIMS libraries are given independently for each fissile nuclide. There are about 40 individual fission products and a lumped fission product. The fission product selection is based on the absorption and the decay characteristics. Certain criteria have been taken to define the explicit fission products and grouped as highly absorbing nuclides ($\gamma_p\sigma_p$ > const./φ), short lived nuclides($\sigma_p\lambda$ < const./φ), nearly stable nuclides ($\sigma_p\lambda$ > const. /φ) and highly absorbing capture products. The yields are taken from basic data files and averaged with a weighting spectrum. The basic data is tabulated at 3 major energies, namely 14 MeV,0.5 MeV and 0.025 eV). The WIMS-D/4 code calculates one group reaction rates and evaluates the burnup dependent parameters. The earlier 1986 WIMS library had only 35 individual fission products. In the WLUP library, the I^{135} isotope has been separated from the cumulative Xe yield [12]

- The separation of individual and pseudo-fission products is based on uranium and ^{239}U isotopes' yield data.
- Applicability to ^{233}U and thorium systems has to be validated.

- The experimental measurements of FP yield data for ^{233}U in thermal fission has been measured long back. However measurements in high energy regions are sparse. BARC has recently measured FP yield data for ^{233}U at 0.5 MeV for 27 FP isotopes. More such measurements are required and have to be properly evaluated [13].

4. Accelerator Driven Sub-critical Systems (ADSS):

In recent years several countries have embarked upon Accelerator Driven Subcritical Systems (ADSS) projects aimed towards waste transmutation, weapon Pu burning and energy production. Many of these projects propose to use Thorium as it is more abundant and uniformly distributed in the world than Uranium, produces very little Pu and other minor actinides and is not considered relatively a proliferation risk. Thorium utilisation in ADSS represents the main reason for Indian interest in ADSS. Particularly we are interested in employing the thermal reactor concept based on AHWR for possible application. However it is well known that thermal systems require a lower k_{eff} than fast systems, which affects the gain. Hence we have done investigations on a coupled core concept, which employs a thermal reactor zone, coupled in one direction to a fast source zone. A brief summary is given here.

4.1 BARC : One Way Coupled Booster Reactor Concept

In the conventional design of ADSS the required constant current of 10 to 20 mA for a 1GeV proton beam is one of the most challenging tasks. For this reason, there have been suggestions for reducing the current to levels that are presently achievable [14]. Besides lower accelerator performance requirements, lower current schemes would place lesser demands on window and target performance.

Degweker et al [15,16], have proposed a one way coupled booster-reactor system which could be operated at currents as low as 1-2 mA. The basic idea is to enhance the importance of the spallation neutrons by placing a booster region around the source located at the centre with the main reactor region surrounding the booster. The booster contains a fuel having a relatively higher enrichment than the main reactor and is intended to enhance the spallation source neutrons. To obtain a large gain in the booster, it is essential to have its K_{eff} as close to unity as possible. To ensure that the overall K_{eff} of the booster reactor combination does not exceed unity, the arrangement has to be designed so as to have one way coupling between the two parts. Two practical ways of getting such a coupling were studied. The first of these envisaged a fast booster and a fast (main) reactor separated by a large gap to get effective decoupling between the two. In the second concept, there was a fast booster and a thermal (main) reactor region separated by a large gap, and a thermal lining around the booster. In both these arrangements, neutrons from the booster enter the main reactor but very few neutrons from the latter return to the booster, ensuring a one way coupling. Very similar ideas have been independently proposed by some Russian groups [17,18].

Our studies on these systems indicate that it may be possible to have a fairly large power of about 750 MWth with an accelerator current in the range 1-2 mA, while maintaining

adequate sub criticality margin, which ensures that the system does not become critical under various conditions. One difficulty with the fast scheme is that the power is very sharply peaked in the booster region while the outer main region has a very low power density, making the scheme unviable. The fast-thermal combination also has a power density sharply peaked at the centre of the booster. However the specific power of the outer main reactor region is comparable to typical thermal reactors while in the inner region it does not exceed that of fast reactors. Our earliest study was based on a simple one dimensional model using group diffusion theory with suitable boundary conditions at the material vacuum and for the absorber lining. More refined calculations based on multigroup transport theory in one dimensional spherical and two dimensional cylindrical geometries have supported the results of our earlier studies. Our calculational models and nuclear data require further improvements, which we are pursuing.

5. Status of Nuclear Data for Thorium Fuel Cycle

We will largely draw from reviews done in connection with an IAEA Consultants Meeting held in April 1999 on "Assessment of Nuclear Data Needs for Thorium and other Advanced Nuclear Cycles" [5] and a review by Ganesan [19]. We will however restrict to only thermal reactor related issues. Kuzminov and Manokhin [20] also have discussed the status of Th cycle nuclear data with more emphasis on fast reactors, which was used in the IAEA CM meeting. The general opinion is that the evaluated cross sections of the isotopes of thorium chain were based on measurements done a few decades ago, often based on a few experiments or only theory.

a) **^{232}Th :-** The capture process in thorium dictates the breeding of ^{233}U. However there are noticeable differences in the major libraries like ENDF/B-VI, JENDL3.2 and JEF2.2 in the basic evaluated point data itself. While the (n,γ) cross sections in thermal energies up to say 0.1 eV is in good agreement, large differences exist even in the resolved resonance regions and more beyond. Choosing different libraries only for ^{232}Th in lattice calculations show differences in k_∞. For a typical AHWR cluster we got the following results for k_∞: WIMS-1986 - 1.256; substituting with ^{232}Th (in WIMS format) from ENDF/B-VI - 1.253; substituting with ^{232}Th from JENDL3.2 - 1.264. This is a large difference for a single isotope. Similar studies are reported by Shiroya et al in connection with their evaluation for a critical facility. [21]

The (n, 2n) reaction cross sections which produce ^{232}U through a chain are based on a few experiments. The values given in the two libraries differ considerably near the threshold as well as beyond 15 MeV. In thermal reactors this (^{232}Th) would be a major source for ^{232}U (due to higher value of the cross section of nearly a barn and the concentration of ^{232}Th) along with those from ^{233}U (cross section of nearly 200-300 mbarn).

b) **^{233}U :-** The discrepancies in point fission cross sections between different libraries are quite large. But it may reduce while arriving at multigroup sets from the basic libraries due to "within-group" cancellation of discrepancies. While discrepancies are about –4.7% to 8.5% for 69group library when generated from

ENDF/B-VI or JENDL-3.2, they can become –63% to 97% for a 175group library. But it should be pointed out that the major region where these differences are manifest are in the 60 to 150 eV, essentially due to the extension of resolved energy region of JENDL-3.2 beyond 60 eV [19].

However Shiroya et al show that for a critical assembly that "a perturbation calculation executed by substituting ^{233}U cross sections of ENDF/B-VI for those of JENDL-3.2, the reactivity difference becomes –0.5% $\Delta k/k$ which is considered more than –1 $ in the ^{233}U fueled system". Since this magnitude of reactivity difference can be easily measured by the critical experiments, it is necessary to perform systematic critical experiments to resolve such issues [21].

The (n, 2n) reaction in ^{233}U which will directly produce ^{232}U are based on theoretical estimates. The values given in the two libraries differ considerably near the threshold as well as beyond 15 MeV.

c) **^{233}Pa :-** The capture cross sections in thermal and low energy regions agree within ± 5% between ENDF/B-VI (rel. 5) and JENDL-3.2. But the limits of resolved and unresolved resonance regions differ between both the libraries. In 100 to 700 keV energy region JENDL-3.2 cross section is higher by a factor of two. The (n,2n) cross sections also differ by a factor of 2.7.

Detailed criticality calculations have been done by Ganesan and Wienke, employing different evaluated basic data libraries show that the k_∞ value obtained by JENDL-3.2 data is 0.36853 ± 0.00063 and by ENDF/B-VI is 0.46033 ± 0.00111 [22].

Thus we notice that all the three major isotopes of the thorium cycle require more detailed experimental measurements and evaluation in the near future.

Let us now turn our attention to some of the other isotopes in the thorium burnup chain. Apart from ^{232}U, the other isotope of concern from radioactivity point of view is ^{231}Pa, which has a long half-life of $3.276X10^4$ years. In AHWR about 30 grams will be come out with the discharged fuel per year. Within a year this will be the dominant protactinium isotope if discharged as waste. It will be prudent to recycle Pa isotopes along with bred uranium isotopes in the AHWR itself, where it will get transformed to ^{232}U on neutron absorption and subsequent decay. While this will help in long term waste disposal problem, will however increase our ^{232}U burden during recycling of uranium from thorium. This aspect requires detailed study.

d) **^{231}Pa :-** The thermal capture cross sections agree well between different libraries. The libraries (ENDF/B-VI and JENDL-3.2) differ in the extent of resolved resonance regions. The (n,f) reaction has very small cross sections. Ganesan recommends the JENDL-3.2 data to the users for this isotope. Using this data k_∞ was evaluated as 0.9727 ± 0.00114, but the ENDF/B-VI evaluation gives close to 0.94. [23]

e) **^{232}Pa :-** This isotope may not have much significance in thermal reactors in spite of having large fission and capture cross sections, except for its decay with a short half life (1.31 days) to ^{232}U.

f) **^{232}U :-** It has significant fission and capture cross sections, in the thermal energy ranges, and both the above libraries agree very well in these energy ranges. But the prompt ν nu-bar differs by 22% between the libraries. Its destruction via the above processes in a thermal reactor has to be assessed properly. A recent evaluation shows that the k_∞ with ENDF/B-VI.5 is 3.07756 ± 0.00170, while with JENDL-3.2 is 2.64800 ± 0.00148. The critical masses using the two nuclear data sources differ by a factor of 3.6 [24].

5.1 Nuclear Data for ADSS Applications

The nuclear data of accelerator driven systems are much less known. The energy region of spallation neutrons covers neutron energies from 1 eV to 250 MeV, far beyond the existing thermal, fast and fusion reactor designs. Most of the absorptions in thorium occur in the resonance region, a few eV to 50 keV. Neutron emission cross sections and gas production cross sections are needed in the MeV region for radiation damage studies. With such features, the presently available ENDF/B files cannot be considered today as a totally reliable basis for designing a full scale Energy Amplifier. Nuclear data of minor actinides such a Americium, Curium and those of minor actinides of thorium fuel cycle show gigantic differences in databases. For example, there is no evaluation for ^{232}Pa in ENDF/B-VI while the evaluations in the presently available ENDF/B files, ^{238}Np, ^{242}Am, ^{242m}Am ^{243}Cm, and ^{247}Cm, ^{238}Pu, ^{244}Pu and ^{247}Cm are regarded as very weak . The data of ^{242}Pu and ^{247}Cm and ^{245}Cm are weak. The present CERN proposal to measure high-resolution cross sections in 1 eV to 250 MeV is a welcome step in the right direction [25].

As a general remark, we wish to stress that the basic experimental cross section database for isotopes of thorium fuel cycle were those measured two decades ago. These isotopes ^{230}Th, ^{232}Th, ^{231}Pa, ^{232}Pa, ^{233}U, ^{232}U etc have received several orders of magnitude less attention, efforts and funding in the past when compared to the isotopes of the U-Pu cycle. In many cases, experimental data is absent and the evaluated data files are based only on phenomenological models and systematics based on sparse experimental data.

There is a need to make a thorough assessment of the existing database for each of the isotopes of the thorium fuel cycle and encourage new evaluations using state-of-the-art evaluation tools. New measurements are required at differential level in most of the cases. As mentioned before, in the epithermal energy regions the discrepancies currently are very high.

6.0 Conclusions:

We have highlighted our work on nuclear data requirements for the analysis of AHWR. A lot more development work is required to be done to answer the issues raised by different project groups in BARC with regard to Th fuel cycle aspects. In this we will be looking forward to collaboration with IAEA under their future CRPs for improving the Th nuclear data base. As observed by experts nuclear data base of many of the isotopes of the thorium chain require a close assessment with regard to their accuracy and dependability. The required accuracies for future measurements have been brought out by the IAEA Consultants meeting in April 1999[5]. From the user (reactor physicist) point of view these measurements and accepted data sets have to be converted to multigroup libraries as undertaken by the WLUP. More critical experiments are also necessary in this regard. We are building a critical facility in which we plan to do number of such experiments. A major international effort is necessary to bring the nuclear database for the uranium-thorium cycle up to the reliability level of that for the uranium-plutonium fuel cycle.

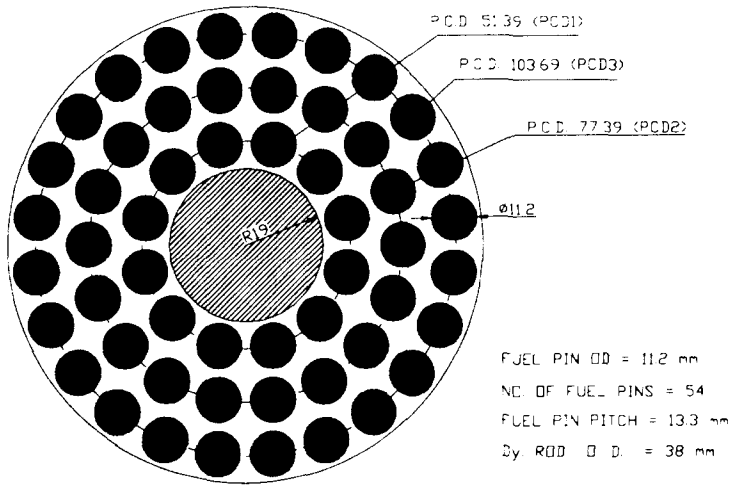
Acknowledgements

We thank Dr. A. Kakodkar, Director, BARC and Prof. S.S. Kapoor for their keen interest in this paper and their useful comments.

References:

1. R. Srivenkatesan et al, "Physics Considerations for Utilisation of Thorium in Power Reactors and Subcritical Cores", IT-2, Indian Nuclear Society Annual Conference, INSAC-2000, June 2000.
2. R.K. Sinha et al, "Design and Development of AHWR – the Indian Thorium Fuelled Innovative Nuclear Reactor", IT-4, Indian Nuclear Society Annual Conference, INSAC-2000, June 2000.
3. Arvind Kumar, " A new cluster design for the reduction of void reactivity in AHWR", poster paper presented in the Indian Nuclear Society Annual Conference, INSAC-2000, June 2000.
4. Arvind Kumar et al, "Physics Design of Advanced Heavy Water Reactor utilising Thorium", Paper presented in the Technical Committee Meeting on "Utilization of Thorium Fuel Options", IAEA, Vienna, November 1999.
5. Final Stage of WIMS-D Library Update Project, Summary Report, F1-RC-733.2, Argentina, August 2000. *Also see the web site* "http://www-rcp.ijs.si/-wlup"
6. V.G. Pronyaev, "Summary Report of the Consultants' Meeting on 'Assessment of Nuclear Data Needs for Thorium and other Advanced Nuclear Cycles', April 1999", INDC(NDS)-408, IAEA, August 99. See also R. Srivenkatesan and Umasankari Kannan, "Note on Nuclear Data requirements for Thorium Cycle, specially for ^{232}U", Paper presented to the consultants meeting.
7. Carlo Rubbia et al, "Conceptual Design of a Fast Neutron Operated High Power Energy Amplifier", CERN/AT/95-44 (ET), 29th September 1995. See also C. Rubbia, "A High Gain Energy Amplifier Operated with Fast Neutrons", AIP Conference Proceedings 346, International Conference on Accelerator-Driven Transmutation Technologies and Applications, Las Vegas, July 1994.

8. G. Fioni et al, "Experimental Studies of the Transmutation of Actinides in High Intensity Neutron Fluxes", ICENES –2000, Petten, Netherlands, September 2000
9. Umasankari Kannan, "WIMS Library update for the thorium cycle" ,RPDS/FC/18.
10. Umasankari Kannan, " Updating the WIMS library for Dysprosium isotopes and its application to AHWR design", INSAC 2000, June 2000.
11. V. Jagannathan et al, "Sensitivity Studies for a Thorium Breeder Reactor Design with Nuclear Data Libraries of WLUP", ICENES 2000, Petten, Netherlands, September, 2000.
12. A.Trkov and D.L. Aldama, "Definition of pseudo fission product for reactor calculations", paper presented as part of Final stage of WLUP.
13. A. Ramaswami et. al, " Measurement of Absolute Fission Yield in the fast neutron induced fission of ^{233}U by track etch cum gamma ray spectrometry, Paper submitted to Nuclear Chemistry and Radiochemistry sysposium (NUCAR-2001), to be held during Jan-Feb, 2001 in Pune, India.
14. Daniel H. and Petrov Yu.V., Nucl. Instr. Meth. Phys. Res. A 373, 131 (1996)
15. Degweker S.B., Lawande S.V. and Kapoor S.S., IAEA TCM on Feasibility and Motivation for Hybrid Concepts for Nuclear Energy Generation and Tansmutation, Madrid, Sept 17-19 (1997); *also see* Degweker S.B., Lawande S.V. and Kapoor S.S , Ann Nucl. Energy 26, 123 (1998)
16. Degweker S.B., Sahni D.C. and Kapoor S.S., Intl. Meeting on ADS, Moscow, Oct 11-16 (1999)
17. Kocherov B.P. and Seliverstov V.V., Int. Meeting on ADS, Moscow, Oct. 11-16 (1999)
18. Alekseev P.N. et al, IAEA TECDOC No. IAEA-6110/3 (1998).
19. S. Ganesan, "A Review of the Current Status of Nuclear Data for Major and Minor Isotopes of Thorium Fuel Cycle", Report No. BARC/2000/E/005, March 2000; Also published in PHYSOR-2000.
20. B.D. Kuzminov and V.N. Manokhin, "Status of Nuclear Data for Thorium Fuel Cycle", Report No. INDC(CCP)-416, 1998.
21. Seiji Shiroya et al, "Assessment of ^{232}Th Nuclear Data through Critical Experiments Using the Kyoto University Critical Assembly (KUCA)", Paper presented in the Technical Committee Meeting on "Utilization of Thorium Fuel Options", IAEA, Vienna, November 1999.
22. S. Ganesan and H. Wienke, "On the Criticality Property of ^{233}Pa derived from various Nuclear Data Files using the NJOY and MCNP Codes", ICENES 2000, Petten, Netherlands, September 2000.
23. S. Ganesan et al, "A Re-calculation of Criticality Property of ^{231}Pa using New Nuclear Data", Current Science, Vol 77, No. 5, pp. 667-670, Indian Academy of Sciences, Bangalore, India, September, 1999.
24. S. Ganesan and Amit Raj Sharma, "New Investigations of the Criticality Property of Pure ^{232}U", Indian Nuclear Society Annual Conference, INSAC-2000, June 2000.
25. C. Rubbia et al., "A high resolution Spallation driven facility at the CERN-PS to measure neutron cross sections in the interval from 1 eV to 250 MeV: a relative performance assessment, Report CERN/LHC/98-02 (EET)-Add. 1, Geneva, June 15, 1998.



(^{233}U+Th) MOX Pins (Pu+Th) MOX Pins

Zirconium Oxide + Dysprosium Oxide Displacer Rod

Fig. 1 The AHWR Composite Fuel Cluster

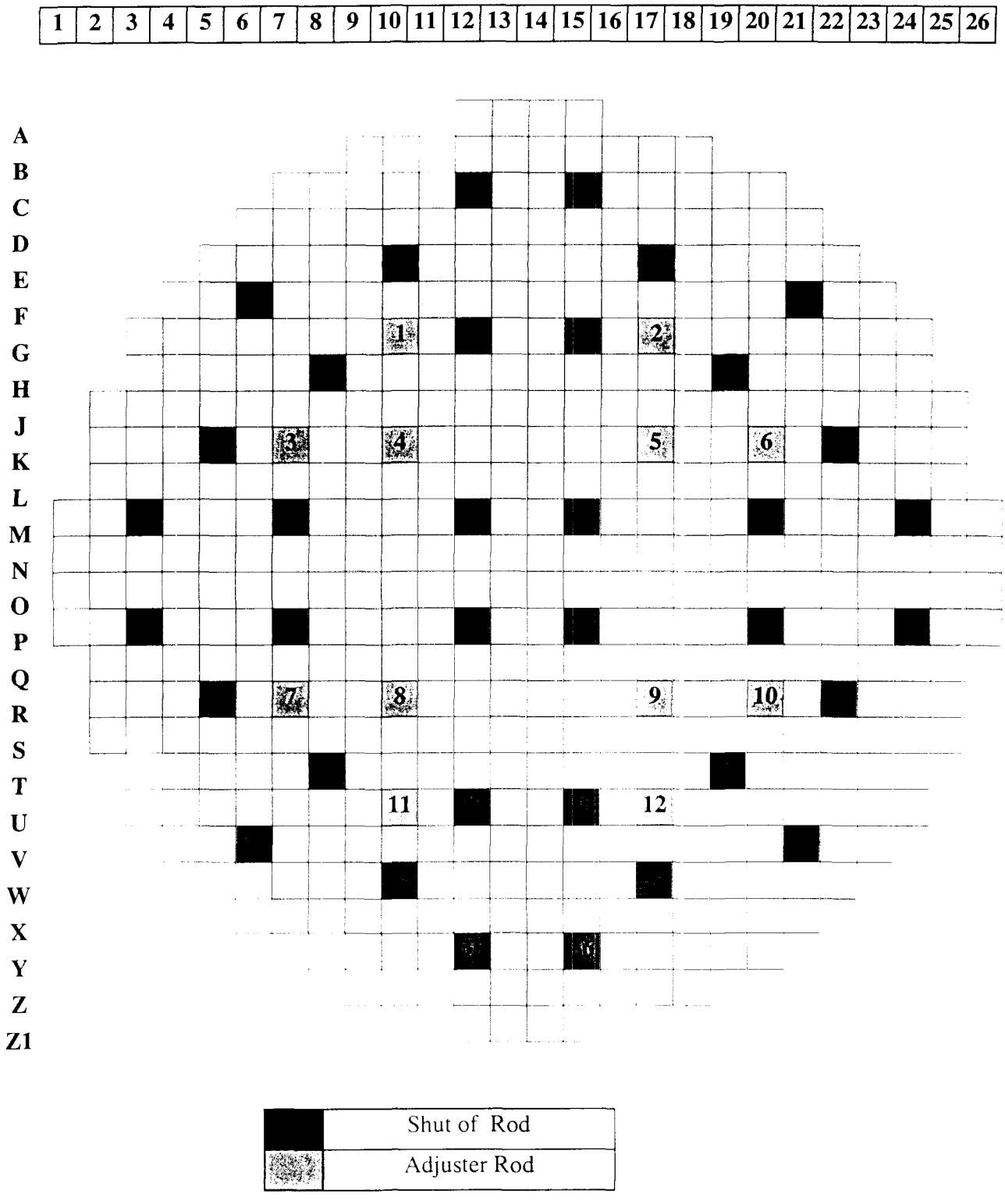


Figure.2 AHWR CORE LAYOUT

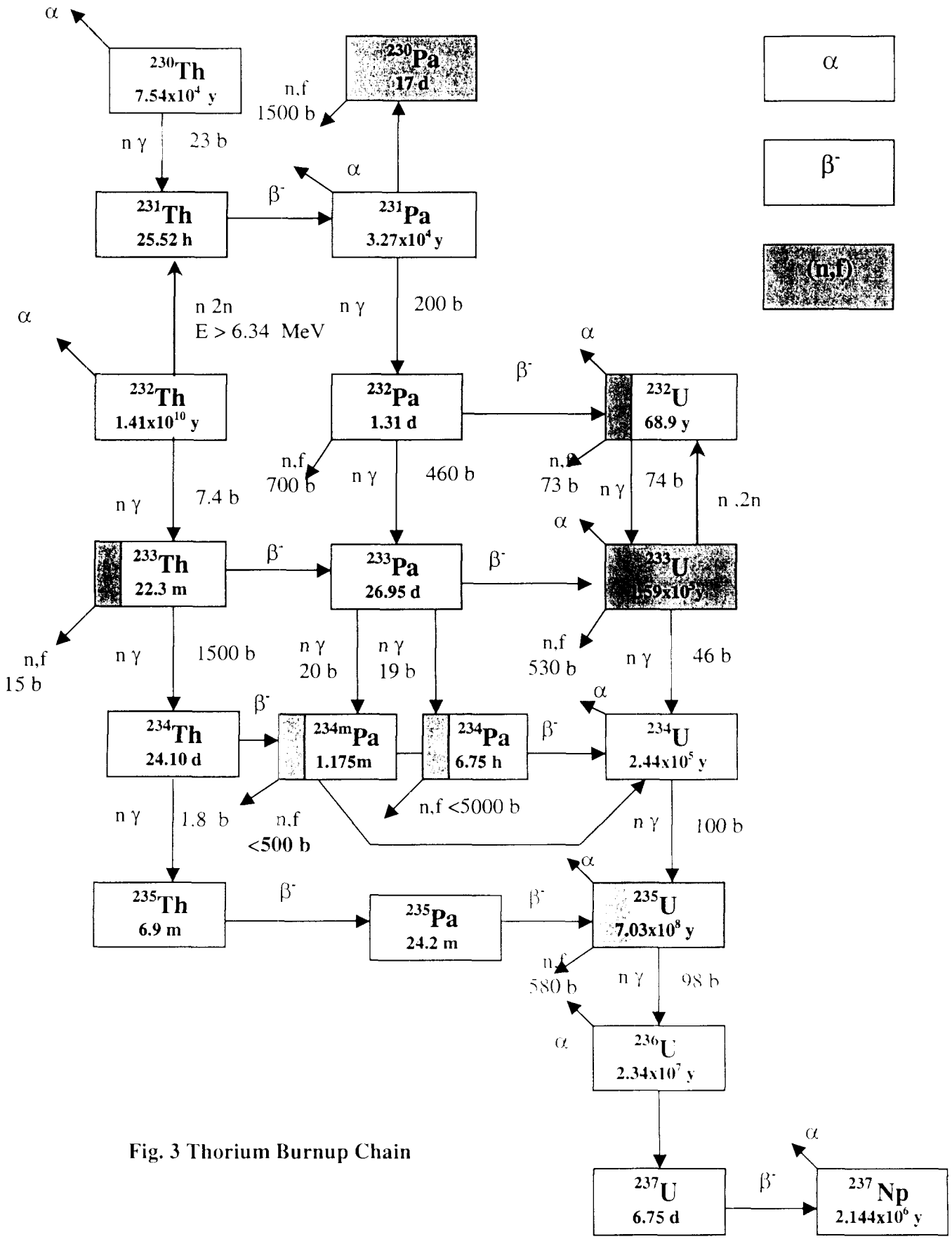


Fig. 3 Thorium Burnup Chain

Accelerator Driven Subcritical Reactors

M. Salvatores

Direction des Réacteurs Nucléaires
Bâtiment 707
Centre d'Etudes Nucléaires de Cadarache
F-13108 Saint-Paul-lez-Durance

ADS concepts have been proposed in the last decade for a variety of applications. However, there is a convergence of interest of several countries and laboratories on the application of ADS to transmutation ("burning"). This applies to plutonium, and/or minor actinides (MA) and long-lived fission products (LLFP). As far as the so-called partitioning and transmutation (P/T) strategies, it was indicated that they can be clarified according to the option taken with respect to Pu and MA, i.e., a) keep Pu and MA together, b) separate Pu from MA. Strategy a) gives rise to transmutation concepts with "homogeneous" recycling of MA in the fuel of "standard" (LWR, Fast Reactors) reactors, or to concepts devoted to Pu+MA burning (i.e., no fertile materials in the fuel), as in the ATW concepts developed in the USA. In this last case the absence of fertile nuclides (e.g., U-238) gives rise to cores with very low β_{eff} and near zero Doppler effect. These features have suggested the use of subcritical core configurations for the burning of pure Pu+MA.

Strategy b) (Pu separated from MA), gives rise to transmutation concepts like "heterogeneous" recycling (e.g. MA targets to be irradiated in "standard" critical reactors), or to the "double strata" concept (originally developed at JAERI-Japan), in which MA are handled in dedicated cores, in a separate stratum of the fuel cycle. These dedicated cores also show low β_{eff} and Doppler coefficients, and these features practically preclude the option of a critical core, and here again ADS find a relevant application.

At present several programs are going on ADS: in Japan in the frame of the Joint Project between KEK and JAERI; in the USA (the Advanced Accelerator Applications, AAA, initiative); in EUROPE, where activities in 9 countries are coordinated by a European Technical Working Group (ETWG), chaired by Carlo Rubbia, and where the European Union is sponsoring and partly funding a dozen of projects (over three years) in different areas of ADS R&D (nuclear data, neutron physics, materials, fuels and preconceptual designs). Finally, several laboratories in Russia are also active in the ADS field. As far as the implications for the definition of nuclear data needs, dedicated subcritical cores should have new type of fuels (Pu+MA in different proportions). Proposals are being worked out. For example, composite (such as ceramic-metallic or ceramic-ceramic) fuels are presently under study. The actinide oxide is dispersed in a metallic matrix (Zr, or W or Mo) or in an oxide matrix (e.g., MgO). In these cases, reliable data are required for the matrix materials. As far as coolants, Pb/Bi, Pb, and gas are considered, besides Na. Hard (or very hard) fast neutron spectrum is required.

As far as realizations, the horizon is 2010-2015 for experimental ADS (power <20 MWth) and 2020-2025 for demonstration experiments at higher power (>100 MWth).

As far as LLFP, transmutation strategies in ADS are proposed. Candidates are ^{129}I, ^{99}Tc, ^{135}Cs, but also ^{79}Se, ^{107}Pd, ^{93}Zr etc. At present, there is no clear option for their transmutation (one needs a high level of thermalized neutrons, support matrixes for target irradiation, isotopic separations, reprocessing techniques, etc.).

Finally, ADS transmutation will give rise to fuel cycles, where very active materials will be present. Cm and higher mass isotopes (up to ^{252}Cf!) will be contributors to dose and neutron source strength. This area will deserve attention in future, in order to define the relevant data needs.

Recommendations

a) Coordinated work on MA data is still a priority: ^{241}Am, ^{242m}Am, ^{243}Am and Cm isotopes, since the typical target accuracies required (i.e., $\approx$ 5% for σ_f, $\approx$ 10% for σ_c, $\approx$ 15% for σ_{in}) are not yet achieved. In this respect the following recommendations can be made:

1) Review the consistency of data and eliminate non-physical data (e.g. ^{242}Cm σ_f and ^{243}Cm σ_{in} in ENDF/B-VI).

2) Perform extensive sensitivity studies for significant parameters of the subcritical core (k_{eff}, reactivity coefficients, peak power, dpa gradients etc.) for systems with different ratios of Pu and MA in the fuel (e.g. Pu/MA=80/20 or = 40/60), for two coolants (e.g., Pb/Bi and gas) and for the initial stage and at end of cycle. This will permit agreement on target accuracies and priorities on a sound basis (to make selected experiments).

3) Review uncertainties and, more generally, the status of data for the calculation of β_{eff}, decay heat, γ-heating for the systems mentioned in 2), before suggesting new experiments.

b) 1) Review the status of data for materials which are potential new candidates as core support materials (e.g., MgO, Zr, Ti)

2) Same for Pb, Bi, N, ^{15}N (σ_t, $\sigma_{n\gamma}$, σ_{el}, σ_{inel}, σ_{np} below 20 MeV).

c) As far as major actinides in the context of transmutation:

1) Review the ^{242}Pu and ^{238}Pu data accuracy, in particular $\sigma_{n,\gamma}$.

2) Assess uncertainty on MA and Pu isotopes σ_f in the region 1-20 MeV (required accuracy: $\pm$ 5%).

Advisory Group Meeting on Long-Term Needs for Nuclear Data Development

Vienna, November 28 – December 1, 2000

NUCLEAR DATA NEEDS FOR SUBCRITICAL REACTORS WITH HEAVY-METAL COOLANT

A.V. Ignatyuk

Institute of Physics and Power Engineering, 249020 Obninsk, Russia

Abstract: Requests on improvement of evaluated data files for minor actinides (MA) are briefly reviewed. New evaluations of neutron cross sections for Np-237, Am-241 and Am-243 after the corresponding tests and verifications should satisfy the required accuracies of data for developing MA-burners. More difficult problems arise for curium isotopes, evaluated data of which are strongly divergent. International expertise of available evaluations could be very desirable. Needs in data improvements for perspective heavy-metal liquid coolants are outlined.

The nuclear data required for the analysis of subcritical reactors are generally the same as for powerful fast reactors. The main differences relate to the fuel composition and possible new types of coolants. The increase of MA component in the fuel creates more severe requests on the accuracy of MA data needed for the analysis of reactor characteristics. Effects from high-energy neutrons (E>20 MeV), coming from the spallation target, are much less important, at least for high values of the total multiplication factor of a subcritical reactor.

Current status of evaluated data files

For practical calculations of nuclear reactors the ABBN-93 group data system is widely used nowadays in Russia [1]. These group constants are based mainly on the evaluated data included into BROND-2 and FOND-2 microscopic file libraries [2].The ABBN-93 data were tested on many benchmarks and macroscopic experiment data, and it was concluded that their accuracy is good enough for the main fuel and structural materials [3].

However, the data for minor actinides remained still badly verified mainly due to the limited number of reliable macroscopic data. During the nineties, a large-scale program of neutronics experiments was realized on the first worldwide industrial fast reactor BN-350, one of the tasks of which was a verification of the MA data. The actinide samples were irradiated in different parts of the reactor core and blanket, and the measured activities were compared with the corresponding simulation calculations [4].

During the last years, some new experiments with MA were performed also on the BFS facility at the IPPE [5]. The central reactivity sample worth ratios and the central fission cross-section ratios relative to U-235 were measured for Np-237, Pu-239, Am-241. Similar experiments were performed also on the fast critical assemblies (FNS) at JAERI [6]. The full description of the FCA experiments is not available for us. Nevertheless, attempts were made to use the FCA data together with the BFS results to test the available evaluations of MA data in Russian and foreign libraries [4]. The analysis performed demonstrates a reasonable agreement between the measured and calculated values, but the macroscopic data do not permit still to make a preference between available evaluations.

Special investigations have been made to estimate the current and the required accuracy of the MA data on the basis of their effect on the neutron transport in the fast-reactor

core [4]. Calculations were performed for the BN-800 reactor, which is now under design at IPPE to be used as the MA burner. The results of the data analysis are presented in Table. 1.

Table 1. Current and required (in brackets) uncertainties of actinides cross sections

Nuclide	Capture, %	Fission, %	Inelastic, %
Np-237	15 (5)	7 (3)	30 (10)
Pu-238	25 (10)	10 (5)	40 (30)
Pu-239	6 (4)	3 (5)	20 (15)
Pu-240	10 (5)	5 (5)	20 (15)
Pu-241	15 (5)	5 (3)	20 (20)
Am-241	10 (5)	10 (5)	30 (10)
Am-242m	30 (10)	15 (5)	40 (30)
Am-243	30 (10)	10 (5)	30 (30)
Cm-242	50 (10)	15 (5)	30 (30)
Cm-243	50 (10)	15 (5)	30 (30)
Cm-244	30 (20)	10 (5)	30 (30)

Recent evaluations of neutron cross sections for minor actinides

During the last years, several groups worked actively on the improvement of MA data. New versions of data files were compiled for Np-237, Am-241,-243, Cm-242,-243,-244 in the Russian BROND-3 library [7-9]. Completely new evaluations for americium and curium isotopes were performed by the Minsk group [10, 11], and some revisions of previous evaluations were made in the ENDF/B-VI [12] and JENDL-3.2 files.

All available experimental data were analyzed under evaluations, and for Np-237 and Am-241 the new evaluations agree much better than the previous ones. However, some divergences of evaluations still remains, and to satisfy the required data accuracies (Table 1), we should understand better the main reason of the residual discrepancies.

As an example, the evaluations of neutron capture cross sections for Am-241 are shown in Fig. 1 together with the available experimental data. The careful analysis of the measurement conditions was performed for the BROND evaluation, and all experimental data were reduced to the current standard and reference reaction cross sections [13]. As a result, much better agreement between experimental data was achieved, and the uncertainties of the evaluated cross sections were estimated about 5-7% for the whole energy region shown in Fig. 1. Taking into account the small deviation between our evaluation and ENDF/B-VI [12], this estimation of uncertainties seems reasonable. We did not reach still the accuracy required for Am-241 by Table 1, but we are close enough to it.

However, returning to Fig.1, we can note an essential difference between the evaluation of the Minsk group and all others for energies below 100 keV. The reason for this relates to the theoretical model that was applied by this group without an adjustment of parameters responsible for the energy dependence of the radiative strength functions to the corresponding experimental data. Without such an adjustment, the statistical models may be used for cross-section evaluations with uncertainties of about 15 %, but for higher accuracies the adjustment of model parameters to some crucial data is always important.

The arguments given above relate also to the capture cross sections of Am-243, for which the estimated uncertainties of the BROND-3 evaluation are better than 10 % for the energy region between 1 and 400 keV [13]. The uncertainties of the new capture cross-section evaluations for Np-237 are about two times smaller, and there are no significant deviations between the evaluated cross sections for this isotope [7]. So it seems that the required accuracies for the neutron capture cross sections are almost achieved in the last evaluations of MA data.

Evaluations for the fission cross sections of Am-243 are shown together with the available experimental data in Fig. 2. A strong divergence of evaluations is connected with the use of contradictive experimental data in the previous analyses. The careful consideration of the measurement conditions and the correction of discrepant data removed the divergence of experimental data to the considerable extent [8]. As a result, the estimated uncertainties of the evaluation are reduced strongly, and we approach to the accuracy 3-5 % for the whole energy region from 100 keV up to 5 MeV. Unfortunately, the uncertainties of the evaluation increase about two times at energies below 100 keV, mainly due to essential fluctuations of the experimental data in the region of unresolved resonances.

The estimated uncertainties of the fission cross-section evaluations for Np-237 and Am-241 are rather similar to Am-243, despite the fact that much more experimental data are available for the first two isotopes. The main reason for that is obvious, the old data with relatively large errors cannot reduce significantly the resulting uncertainty of the approximating curves. For the further improvement of evaluation accuracies, the new precision measurements of fission cross sections at energies between 1 and 100 keV are very desirable.

There are much less experimental data for curium isotopes than for americium ones. Measurements of the neutron total and capture cross sections are limited by the neutron resonance region. For fast neutrons, the fission cross sections are measured only. It is reason why all evaluations of neutron cross sections are based mainly on the analysis of fission measurements, and the optical-statistical calculations are applied for other cross sections. Our evaluations of the fission, inelastic neutron scattering and (n,2n) reaction cross sections for Cm-242 are shown in Fig. 3 together with the previous ones. The discrepancies between different evaluations are considerable, and the evaluation validations are definitely required.

Several groups measured the fission cross-sections for Cu-243, but there is no agreement of their results. The data available are shown in Fig. 4 together with the main evaluations. It is necessary to note, that the precise measurements of the averaged fission cross-section were performed on the fast critical assembly with the average neutron energy about 300 keV [14]. The value of (2.651±0.090) b was obtained, which is consistent with our evaluation and differs considerably from other evaluations. Thus we consider the BROND evaluation as being consistent with the benchmark data.

The results of optical-statistical calculations of inelastic neutron scattering and the (n,2n) reaction cross sections for Cm-243, which is consistent with the evaluated fission cross section, are shown in Fig. 5. They differ considerably from the previous cross-section evaluations. The discrepancies for high-energy neutrons are not only due to the difference in the fission cross-section evaluations, but also to the discrepancies in the neutron absorption cross sections. Both for curium and americium isotopes the Minsk group used the optical potential, which results in lower values of the absorption cross section at energies above 10 MeV than the observed fission cross-sections for some curium isotopes at the energy of 14 MeV [15].

The divergence of experimental data and evaluations of the fission cross sections for Cm-244 is not so significant as for the Cm-243 [9]. However, the averaged fission cross section, calculated for the fast critical assembly with our evaluated data, is equal to 1.35 b that exceeds noticeably the measured value of (1.232±0.039) b [14]. The calculated values remain approximately the same, if the ENDF/B-VI or Minsk' evaluations are applied. At present we do not see any explanation of this contradiction between the microscopic and macroscopic data. The evaluations of fission cross sections for Cm-244 seem to require additional validations.

Taking into account the contradictions of the available experimental data on the fission cross sections and the complete deficit of measurements for other cross sections, it is difficultly to guarantee the evaluation accuracies better than the current ones in Table 1. The BROND-3 evaluations for curium isotopes seem to be the most reasonable nowadays, but their validation, nevertheless, requires additional benchmark tests.

New evaluations for lead and bismuth

The development of powerful fast reactors or an intensive spallation-neutron source with the heavy-metal liquid coolant definitely requires more accurate data than available for the neutron cross sections of lead and bismuth. Discrepancies between the evaluated data of ENDF/B-VI, JENDL-3.2 and BROND-2 for these elements amount to 40-50% in many cases, and such disagreements should be removed in data recommended for future practical applications. Evaluations carried out during the last years for BROND-3 were aimed on this problem [16].

Main attention in new evaluations was paid to the neutron inelastic scattering and (n,2n) cross sections. The analysis of all experimental data was performed, and the corrections to current standard were made for many old data. The corrected data agree much better than the original ones, and such corrections are especially important for data on discrete gamma-ray transitions between low-lying levels. The evaluated excitation functions for the first levels of lead isotopes and Bi-209 are shown in Fig. 6 together with the corresponding experimental data. The divergence of previous evaluations is so strong, that the need their improvement seems obvious.

The detailed analysis of the gamma-transitions in separate lead isotopes was done, and the resulting evaluations of gamma-ray spectra and integral production cross sections were tested additionally on the basis of more numerous data for the natural mixture of lead isotopes. The recent precision measurements of the integral gamma-ray spectra for 14.3 MeV incident neutrons [17] were considered to validate once more the evaluated gamma-production cross sections and spectra.

The renewed data files for the separated lead isotopes, the natural lead and Bi-209 were compiled and they are being tested now against the criticality benchmarks for the assemblies with the lead reflectors of different thickness [18]. The testing results will be taken into consideration in the final selection of recommended cross sections and estimation of their uncertainties.

Conclusion

The new version of BROND-3 library that is being compiled now by the Russian Nuclear Data Center should supply the developing projects of MA-burners with more accurate evaluated data than previous data systems. The main improvements of MA data were briefly

discussed in the present contribution.

For Np-237, Am-241 and Am-243 improved evaluations are being testing now intensively in experiments on critical assemblies, results of which will supply in several years both the selection of best evaluations and the validation of their uncertainties.

For curium isotopes it is difficult to hope on a crucial test of evaluations in the nearest future. Nevertheless, the careful international expertise of available evaluations could help in this case. The critical analysis of theoretical models, as well as an attraction of phenomenological systematics will be useful for the selection of the best evaluations.

Improvement of evaluated data for new types of coolants seems a simpler task. However, it should be considered and solved in accordance with accuracy requests corresponding to designing advanced reactors and powerful ADS.

References

1. G.N. Manturov, M.N. Nikolaev, A.M. Tsiboulya, BNAB-93 Group Data Library. Part I: Nuclear Data for the Calculation of Neutron and Photon Radiation Fields. Report INDC(CCP)-409/L, 1997, pp.65-110.
2. A.I. Blokhin, B.I. Fursov, A.V. Ignatyuk et al. In: Nucl. Data for Science and Technology. (Gatlinburg, 19954), Oak Ridge, 1994, v.1, p. 695.
3. G.N. Manturov, M.N. Nikolaev, A.M. Tsiboulya. Ibid., v.2, p.993.
4. V.N. Koscheev, G.N. Manturov, M.N. Nikolaev et al. In: Proc. Conf. ADTTA-99, Prague, 1999, Report We-I-15.
5. S.P.Belov, V.A. Dulin, I.A.Matveenko et al. In: Proc. Conf. on Reactor Physics (Mito, 1996), JAERI, 1996.
6. T. Mukaiyama, et. al., Actinide Integral Measurements on FCA for Evaluating and Improving Cross Section Data, 1985, Report NEACRP-A-684.
7. A.V. Ignatyuk, S.A. Badikov, A.I. Blokhin et al. Evaluationa of neutron cross sections for Np-237. VANT, Ser. Nuclear Constants, 1997, is. 3-4, pp. 59-78.
8. A.V.Ignatyuk, A.I.Blokhin, V.P.Lunev et al. New evaluations of neutron cross sections for Am-241 and Am-243. VANT, Ser. Nuclear Constants. 1998, is. 2, pp. 25-38.
9. A.I.Blokhin, A.S.Badikov, A.V.Ignatyuk et al. Evaluations of cross sections for Cm-242,-243,-244. VANT, Ser. Nuclear Constants. 1999, is. 2, pp. 53-60.
10. V.M. Maslov et al. Evaluations of neutron data for Am-241 and Am-243. Report INDC(BLR)-005 & -006, IAEA, Vienna, 1996.
11. V.M. Maslov et al. Evaluation of neutron data for Cm-243. Report INDC(BLR)-007, IAEA, Vienna, 1996.
12. P. Young, D. Madland. ENDF/B-VI, MAT 9543, 1994.
13. K.I. Zolotarev, A.V. Ignatyuk, G.Ya. Tertychny, V.A. Tolstikov. VANT, Series. Nuclear Constants, 1997, is. 4, p. 34.
14. Fomushkin et al. Nuclear Data for Science and Technology (Trieste, 1997), IPS, Bologna, 1997, p. 462.
15. E.F.Fomushkin et al. VANT, Ser. Nuclear Constants, 1992, is. 1, p. 5.
16. A.V.Ignatyuk, A.I.Blokhin, V.P.Lunev et al. In: Proc. 9th Conf. on Radiation Shielding (Tsukuba, 1999). JAERI, 2000, to be publ.
17. Nefedov Yu.Ya. et al.: ibid.
18. International Handbook of Evaluated Criticality Safety Benchmark Experiments. OECD, Paris, 2000.

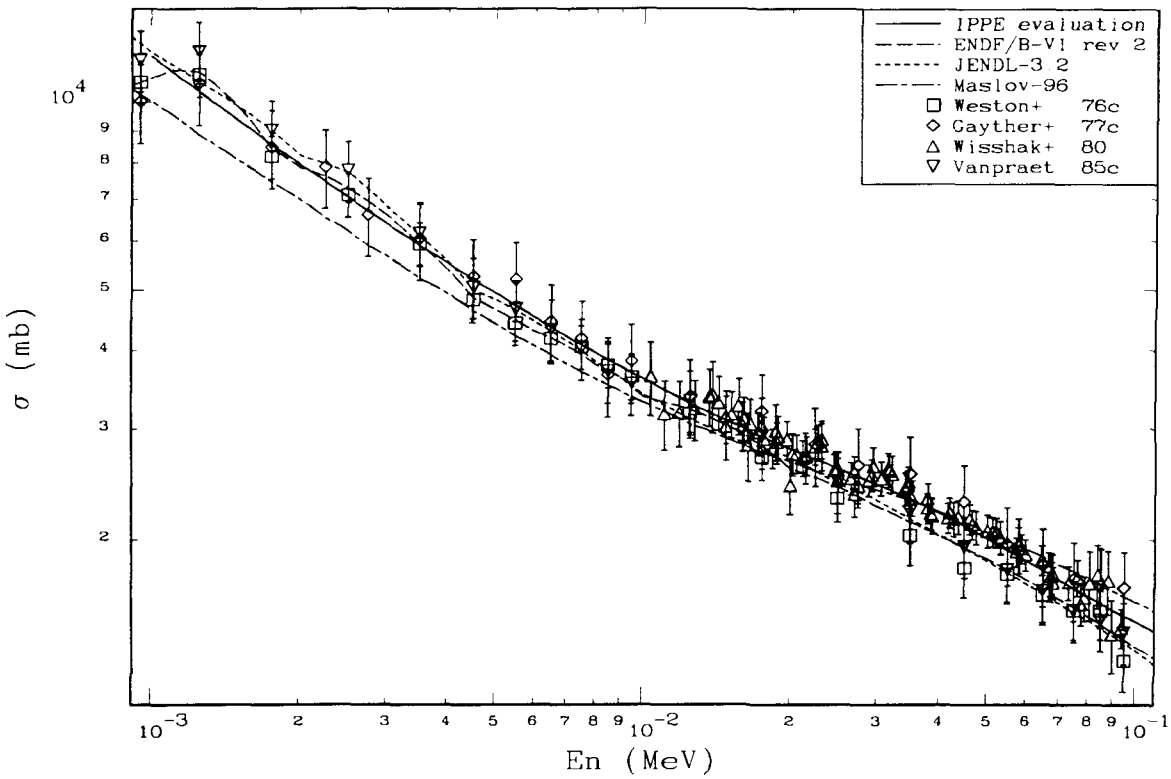


Fig. 1. Evaluated neutron capture cross sections for ^{241}Am in comparison with experimental data.

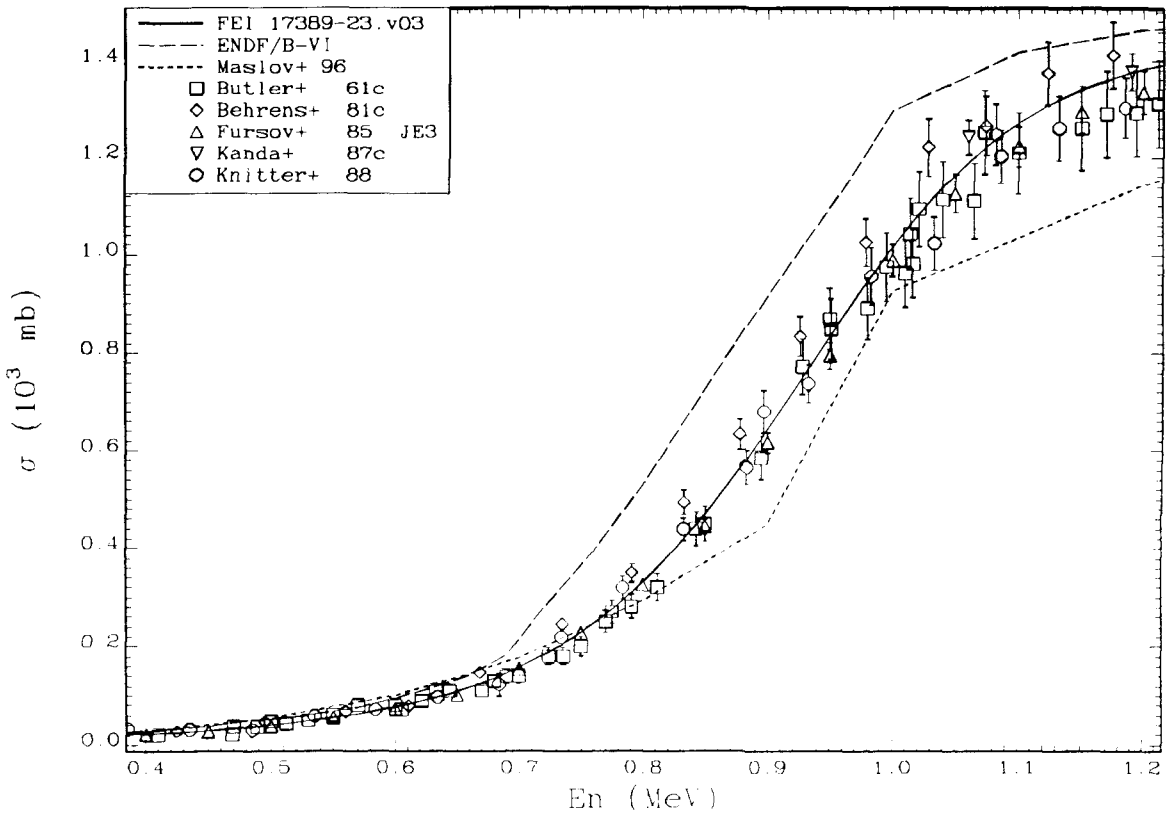


Fig. 2. Recommended fission cross sections for ^{243}Am in comparison with the corrected experimental data.

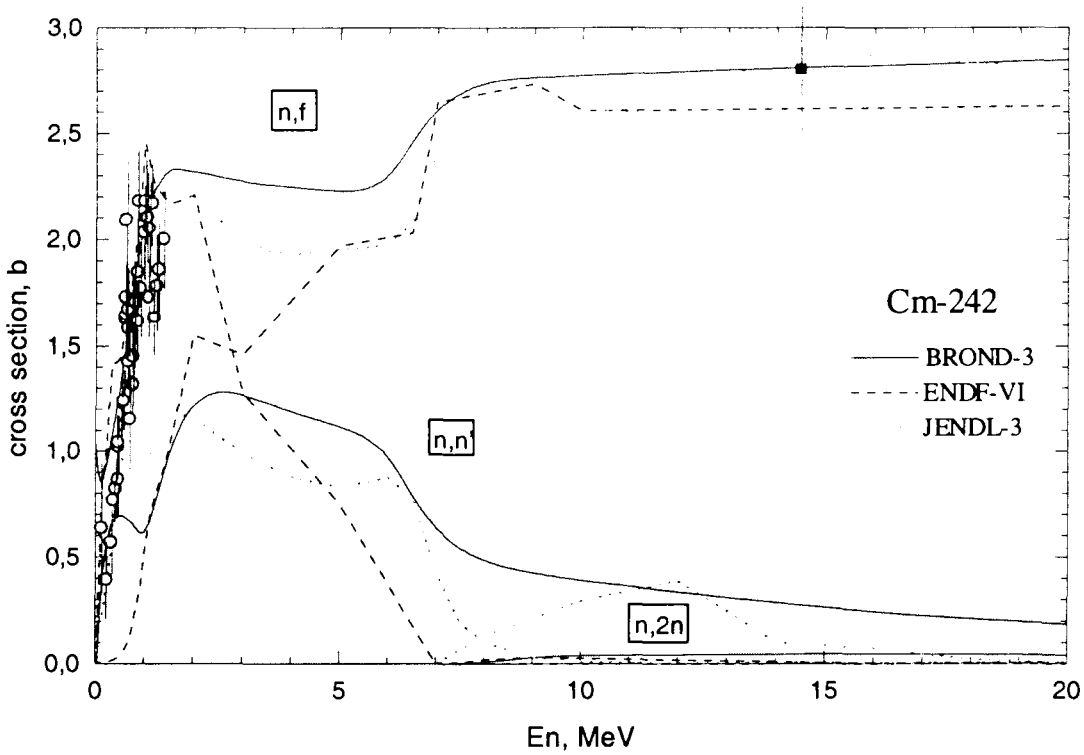


Fig. 3. Comparison of the evaluated fission, inelastic and (n,2n) reaction cross sections for ^{242}Cm.

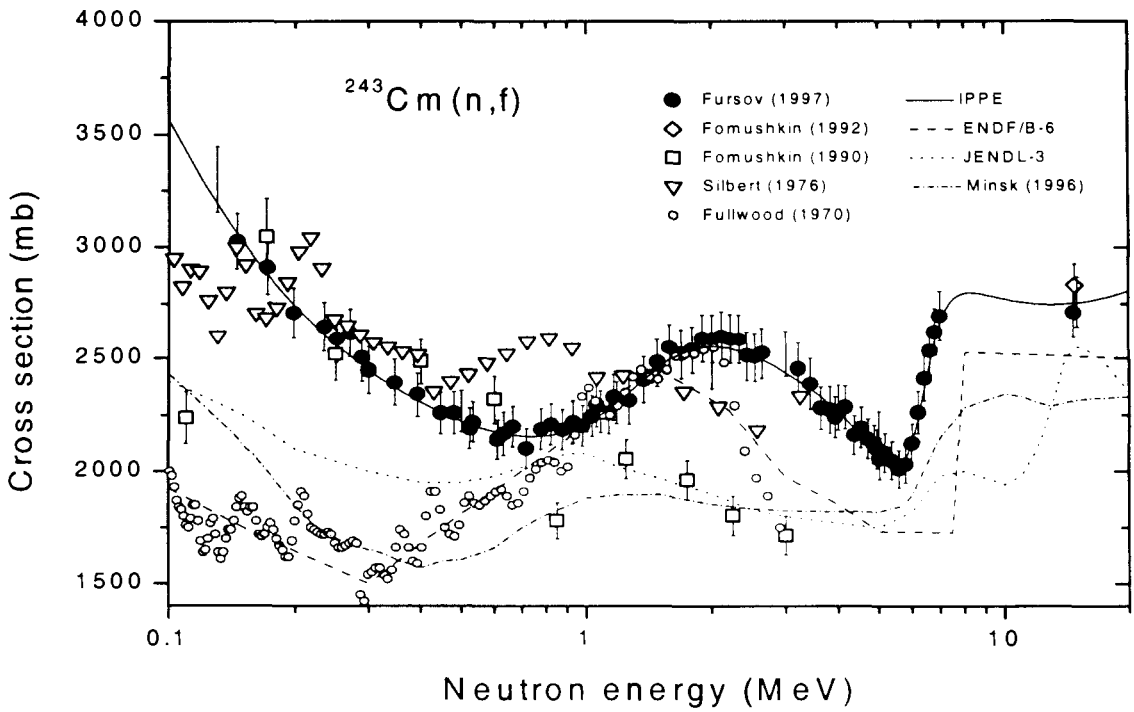


Fig. 4. Comparison of the evaluated fission cross sections for ^{243}Cm with experimental data.

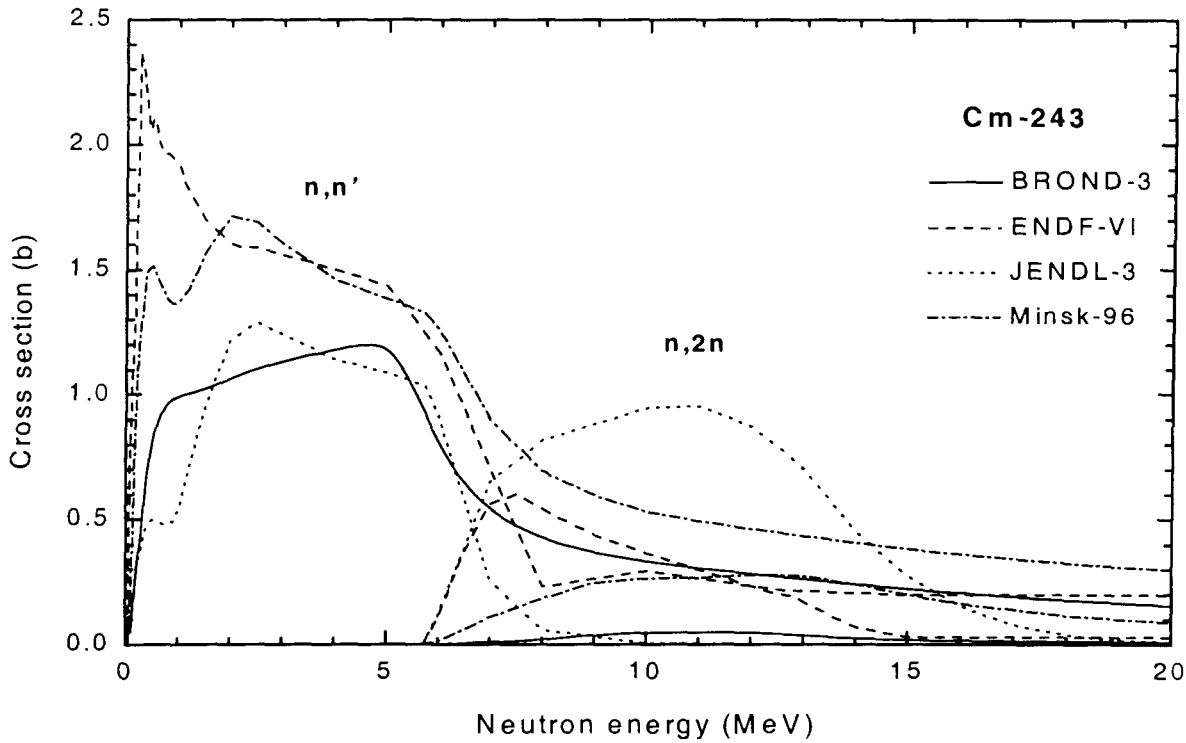


Fig. 5. Comparison of the evaluated inelastic and (n,2n) reaction cross sections for ^{243}Cm.

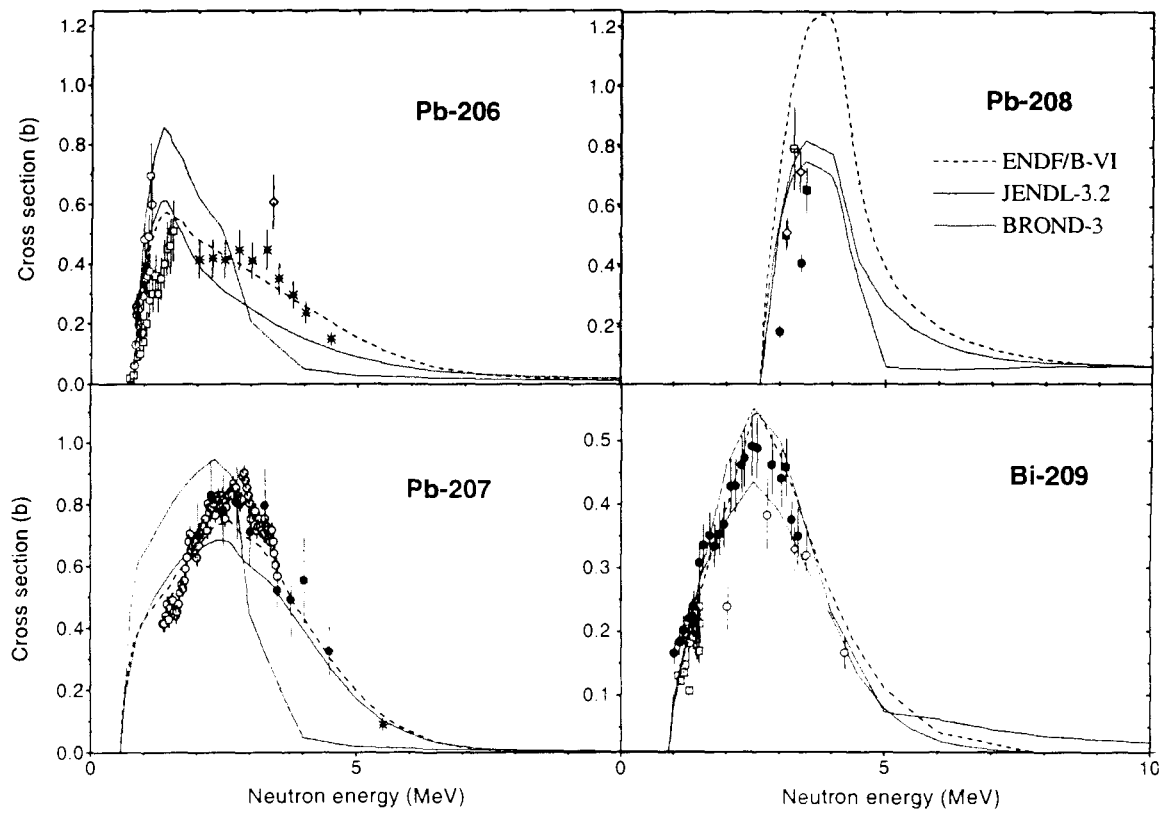


Fig. 6. Evaluated inelastic neutron scattering cross-sections for the first excited level of lead and bismuth isotopes in a comparison with experimental data.

Need for Neutron Nuclear Data for Transmutation of Nuclear Waste by Accelerator Driven Sub-critical Reactor

Zhixiang ZHAO Hongwei Yu
China Institute of Atomic Energy, P.O.Box 295(1), Beijing 102413

Abstract: Neutron nuclear data requirements for transmutation of long-lived nuclear waste produced in LWR and for production of nuclear waste in accelerator driven sub-critical reactor are described.

I, Introduction

In recent years, an innovative technological option in nuclear power exploitation, so called accelerator driven sub-critical system (ADS), is proposed by nuclear science community. Most of people believe that the ADS is a promising nuclear power system to ensure a sustainable nuclear energy supply due to it's good properties of resource friendly, environment friendly and public friendly. Several national level programs are going on in the world, such as ATW in USA[1], neutron science project in Japan[2], XADS[3] in Italy, etc..

Recently, a R&D project related ADS has been approved our government. The goal of this project during the coming five years is to perform the basic research of the physics and technology of the ADS. It includes the following contents: The optimization of the ADS, to clarify its roles, merits and perspectives in the nuclear energy sustainable development in combining with the status of the nuclear industry system in our country; To carry out the study of the physical foundation of the ADS, including the nuclear physics, the reactor physics and the physics of high intensity beam, and the suitable specific experiments for the examine of the conclusions drawn in the study; To complete the technical design of the verification facility for the comprehensive study of the ADS in the future.

In the R&D stage including conceptual research and design for ADS system, many technical options are proposed for different purpose, such as sufficient utilization of uranium resource and utilizing thorium resource, burning up military plutonium, transmute nuclear waste discharged from thermal neutron nuclear power station and producing energy. The detailed feasibility study, safety assessment and conceptual design of these strategies requires the accurate knowledge of neutron nuclear data.

Some new type coolants and moderators are suggested to be used in ADS, among them Pb/Bi eutectic seems to be a promising candidate. The uncertainty of the data for these coolants have a large impact on the performance of ADS system[4]. The structure materials of ADS have to be

different from what used in PWR or FBR to be compatible with the coolant and moderator. This demands the nuclear data for some specific nuclides.

The ADS system is driven by an external neutron source obtained through a spallation process on a heavy element target bombarded by inter-medium energy proton. The neutrons out from the target have average energy around 10 MeV and hard tail can be up to several hundreds MeV. The neutron energy spectrum in the core of the sub-critical reactor is much high than those both in thermal neutron critical reactor and in sodium cooled FBR. The existing evaluated neutron nuclear data libraries mainly service for critical reactor and fusion reactor and the neutron energy concerned in the libraries is limited below 20 MeV. So ADS research urgently demand to extend the neutron energy region of nuclear data evaluation and measurement up to several hundreds MeV.

This paper only describes the neutron nuclear data requirement for consideration of nuclear waste transmutation and production in ADS.

II, Neutron Data for Transmutation of Long-Lived Fission Products

The list of major long-lived fission products (LLFP) in spent fuel discharged from a standard light water reactor (LWR) with 1 GWe power and after 40 years operation is given in Table 1.

Table 1 LLFP Production from LWR with 1GWe after 40 Years Operation

Element	Mass, kg	$T_{1/2}$, year	Other isotopes, kg
^{79}Se	6.6	1.10×10^{6}	63
^{90}Sr	565	28.79	
^{93}Zr	810	1.53×10^{6}	4200
^{99}Tc	843	2.11×10^{5}	843
^{107}Pd	240	6.50×10^{6}	
^{147}Sm		1.06×10^{16}	
^{149}Sm		$>2\times10^{15}$	
^{151}Sm	18	90	
^{126}Sn	29.5	1.0×10^{5}	96
^{129}I	196	1.57×10^{7}	255
^{135}Cs	442	2.3×10^{6}	2500
^{137}Cs	832	30	
^{142}Ce		$>5\times10^{16}$	

LLFP can be transmuted into stable or short-lived nuclide through (n, γ) reaction. Transmutation chains for several LLFP are given in Fig.1 to Fig.5. As shown in the figures, the daughter nuclide is normally either stable or short-lived, quickly decaying into a stable species.

The (n, 2n) reaction for daughter nuclide of (n, γ) reaction is an inverse process of transmutation and also play important role in the transmutation.

Accompanying LLFP in Tab.1, the other isotopes of same element are also produced in LWR as fission product. These isotopes are not easy to be separated from LLFP to be transmuted and could be composition in transmutation element. The neutron nuclear data for these isotopes are also needed.

The transmutation rate can be expressed as

$$-dN/dt = \lambda N + \sigma \Phi \tag{1}$$

where σ is the average absorbing cross section

$$\sigma \Phi = \int \sigma(E)\Phi(E)dE \tag{2}$$

From eq.(1), we obtain

$$N = N_0 e^{-(\lambda + \sigma \Phi)t} \tag{3}$$

Defined the time when $N = N_0/2$ as effective half life $T_{1/2}(eff)$, we have

$$T_{1/2}(eff) = 0.693/[\lambda + \sigma \phi] \tag{4}$$

Very high neutron flux will need to use thermal neutron in transmutation of these LLFP because they usually have small thermal neutron capture cross section. For example, to deduce the effective half-life to three months for ^{93}Zr and ^{137}Cs the thermal neutron flux will be asked to be high up to about $10^{17}/cm^2s$, and for ^{99}Tc and ^{129}I the figure is about $5 \times 10^{15}/cm^2s$.

In the ADS system with molten heavy metal coolant such as Lead or Pb/Bi eutectic, the LLFP can be transmuted by so-called "adiabatic resonance crossing" method[5]. In this method which have been demonstrated in TARC experiment[6], the LLFP to be transmuted is diluted at low concentration in a transparent, diffusing medium of large atomic number such as Lead or Bismuth. The neutron energy is slightly reduced at each elastic scattering, thus "scanning" in very tiny energy steps through the resonance spectrum of the sample during the smooth,

Table 2 Neutron data needed in LLFP transmutation

Element	Reaction	Energy region/ data type*	Remark**
^{79}Se	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	A B
$^{77,78,80,82}Se$	(n, γ)	Below 20 MeV	B
^{90}Sr	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	B C
$^{84,86,87,88}Sr$	(n, γ)	Below 20 MeV	C
^{93}Zr	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	A B
$^{90,91,92,94,96}Zr$	(n, γ)	Below 20 MeV	C
^{99}Tc	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	A B
^{107}Pd	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	B C
$^{102,104-106,108,110}Pd$	(n, γ)	Below 20 MeV	C
^{147}Sm	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	B C
^{149}Sm	(n, γ) (n,2n), (n,xn)	Below 20 MeV /RP Below 300MeV	B C
^{151}Sm	(n, γ) (n,2n), (n,xn)	Below 20 MeV/RP Below 300MeV	B C
$^{144,148,150,152,154}Sm$	(n, γ)	Below 20 MeV	C
^{126}Sn	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	A B
$^{116,120,122,124}Sn$	(n, γ)	Below 20 MeV	C
^{129}I	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	A B
^{127}I	(n, γ)	Below 20 MeV	C
^{135}Cs	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	A B
^{137}Cs	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	B C
^{133}Cs	(n, γ)	Below 20 MeV	C
^{142}Ce	(n, γ) (n,2n), (n,xn)	Below 20 MeV/ RP Below 300MeV	B C
Ce^{140}	(n, γ)	Below 20 MeV	C

* RP: Resonance Parameter;

** "A" means data very important, "B" important, and "C" useful.

otherwise unperturbed, energy slow-down of the initially high energy (MeV) neutrons of the external source. The neutron capture efficiency can be largely enhanced through this process. Therefore the neutron cross sections in resonance region for LLFP will be very important in research on their transmutation in ADS system. To consider the temperature effect, the resonance parameter describing is necessary.

The Table 2 summary the demand for neutron nuclear data in transmutation of LLFP.

As mentioned before, the situation of the data in evaluated data files is unsatisfactory for LLFP transmutation research in ADS. There is no evaluation for some nuclides. For some important data such as Zr^{93} and Cs^{135}, a large discrepancy exists among evaluated nuclear data libraries. As example, the Table 3 shows a comparison of resonance capture integral from different evaluated data files.

Table 3 Comparison of Resonance Capture Integral in Data Files

In barn

NUCLIDE	JEF2.2	ENDF/B-VI	JENDL-3.2	BROND-2
^{79}Se			60.60	
^{90}Sr	0.4798	0.4812	0.06666	0.4901
^{93}Zr	33.01	28.04	18.12	15.36
^{99}Tc	304.2	350.4	311.1	304.2
^{107}Pd	104.8	109.8	111.3	120.7
^{126}Sn	0.1596	0.16	0.1296	
^{129}I	30.28	35.56	28.98	27.97
^{135}Cs	61.02	61.79	62.29	28.48
^{137}Cs	0.5985	0.4759	0.3339	
^{151}Sm	3465	3449	3407	3497

III, Neutron Data for Transmutation of Minor Actinides

The Table 4 shows the production yields for several minor actinides calculated based on Daya Bay nuclear power station with 4.45 enriched UO_2 fuel under 45000MWD/TU burn up.

A LWR with 1Gwe power yearly discharge spent fuel 33t including 35kg of MA (Np, Am, and Cm). After capturing a neutron, the MA becomes new MA nuclides. Therefore the best way to destroy the MA waste is through the fission process. Due to the MA nuclides usually have a large α values, the cross section ratio of capture to fission, at thermal neutron energy region, it is not a good way to incinerate the MA waste in thermal neutron reactor. In the other hand, the heavy metal cooled ADS

system have rather hard neutron spectrum in which the MA have much smaller α values, this make it become very good incinerator to destroy the MA wastes.

Table 4 Production of TRU in LWR (g/TU)

MA	Time after Shut Down					
	0	7d	0.5y	1y	3y	8y
^{234}U	204	204	205	206	209	218
^{236}U	5753	5753	5753	5753	5754	5755
^{237}Np	665	673	681	681	681	684
^{238}Pu	217	219	227	230	229	221
^{239}Pu	4947	5032	5044	5044	5044	5043
^{240}Pu	2541	2541	2541	2542	2544	2549
^{241}Pu	1387	1386	1354	1322	1201	944
^{242}Pu	574	574	574	574	574	577
^{241}Am	35	36	67	100	221	474
^{243}Am	123	123	123	123	123	123
^{244}Cm	40	40	39	38	35	29

The nuclear data uncertainty for MA can have an impact on properties of ADS to be used for transmutation of MA and therefore on proton current requirements[7]. In a fast ADS dedicated to burn MA with the fuel consisting of 37% Pu, 19% Np, 35% Am and 9% Cm, ±20% uncertainty on fission cross section of each MA isotope will lead about 1% uncertainty on K_{eff} . This can imply a 50% uncertainty on the accelerator current needed, even without having taken into account the effects due to the Pu isotope cross section uncertainties.

The MA to be transmuted in ADS will be composition of the fuel element. The temperature effect of MA cross sections will have an impact on safety properties of ADS system. Therefore the neutron resonance parameters for these nuclides are also needed.

The neutrons with high energy above 300 MeV is only a small part (less than 1%) in the spectrum of out going neutrons from the target under 1 GeV proton bombarding[8]. So the most important neutron energy region for these data is below 300 MeV.

The important minor actinide wastes to be transmuted are listed in Table 5 with nuclear data requirement. In the table 5, the data for ^{237}Np, $^{241,243}Am$ and $^{244,245}Cm$ have the first importance. The nuclear data for the nuclides in transmutation chain not appearing in the Table are also needed.

Table 6 shows the discrepancies existing among evaluated nuclear data libraries for the MA listed in Table 5. Generally speaking, the cross

sections at thermal neutron energy in different libraries are rather consistent each other, large discrepancies for (n, 2n) cross sections at fast neutron energy region and the data in resonance region exist among different evaluated nuclear data libraries.

Table 5 Neutron Data Needed in MA Transmutation

MA	$T_{1/2}$ year	Data Type	Energy Region
^{237}Np	2.14×10^{6}	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{238}Pu	87.7	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{240}Pu	6564	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{241}Pu	14.29	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{242}Pu	3.73×10^{5}	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{244}Pu	8×10^{7}	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{241}Am	432.2	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{242m}Am	141	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{243}Am	7370	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{243}Cm	29.1	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{244}Cm	18.1	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{245}Cm	8500	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{246}Cm	4760	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{247}Cm	1.56×10^{6}	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV
^{248}Cm	3.48×10^{5}	(n, γ), (n, f), (n, xn), ν, resonance parameter	Below 300MeV

Table 6 Discrepancies among Evaluated Data Libraries for MA

MA	Reaction /Energy	Data in Evaluated Neutron Data Libraries, b				
		JEF-2.2	ENDF/B6	JENDL-3.2	BROND-2	CENDL-2
^{237}Np	fiss./RI	0.2066	0.2143	0.8868		0.2088
^{238}Pu	fiss./RI	22.70	20.95	22.85	26.99	
	n2n/14MeV	0.055	0.6304	0.5147	0.1340	
^{240}Pu	fiss./RI	2.736	2.712	2.545	3.595	
	n2n/14MeV	0.4272	0.4600	0.3731	0.3670	
^{241}Pu	n γ /RI	169.2	169.0	179.2	200.6	
	n2n/14MeV	0.1781	0.1050	0.1143	0.5000	
^{242}Pu	fiss./RI	0.9418	0.2277	0.2535	16.58	
	n2n/14MeV	0.4541	0.4172	0.4333	0.2560	
^{244}Pu						
^{241}Am	fiss./RI	9.774	8.260	7.365	7.409	8.254
^{243}Am	fiss./RI	1.194	2.134	2.246	2.120	
	n2n/14MeV	0.37	0.04	0.36	0.44	
^{243}Cm	n γ /RI	283.8		198.1		
	n2n/14MeV	0.2866		0.4509		
^{244}Cm	fiss./RI	11.91		5.961	5.719	
^{245}Cm	n2n/14MeV	0.5727	0.25	0.3595		
^{246}Cm	n3n/14MeV	0.8032	0.8032	0.2451		
^{247}Cm	fiss./RI	740.3	741.0	600.3		
	n2n/14MeV	0.1	0.1	0.2413		
^{248}Cm	fiss./RI	8.731		11.28		
	n2n/14MeV	0.23		0.1479		
	n3n/14MeV	0.55		0.2356		

Note: fiss.-- fission;
n2n-- (n, 2n) reaction;
n3n-- (n, 3n) reaction;
n γ -- (n, γ) reaction;
R I -- Resonance integral.

IV, Nuclear Data for Producing Long-lived Nuclear Wastes in ADS

In a natural uranium fueled ADS system, the fissile material is ^{239}Pu, which is produced through ^{238}U+ n $\rightarrow ^{239}$U (β^-) $\rightarrow ^{239}$Np (β^-) $\rightarrow ^{239}$Pu process. In the case of ADS system fueled with natural thorium, the fissile material is ^{233}U produced through ^{232}Th + n $\rightarrow ^{233}$Th (β^-) $\rightarrow ^{233}$Pa (β^-) $\rightarrow ^{233}$U process. Due to the hard neutron spectrum in ADS, the fission from ^{238}U and ^{232}Th make great contribution to power output of ADS[9]. This situation is different from LWR where ^{235}U is the fissile material. To calculate the production of LLFP in ADS, the yields for

fission products of ^{238}U, ^{232}Th, ^{233}U and ^{239}Pu are needed.

Generally speaking, fast spectrum ADS system produces much less MA compared to LWR. This is because ^{233}U and ^{239}Pu have smaller α values, the cross section ratio of capture to fission, for fast neutron than thermal one. It means that the MA production rate in fast ADS will be much lower than those in thermal neutron reactor such as LWR at same power level. Table 7 gives the α values of ^{233}U and ^{239}Pu for several neutron spectrum. Our calculation demonstrates that the ADS working at smaller α value will have lower MA production rate[9].

Table 7 α for U^{233} and Pu^{239}

Spectrum	U^{233}			Pu^{239}		
	$\sigma_{n\gamma}$, b	σ_{nf}, b	α	$\sigma_{n\gamma}$, b	σ_{nf}, b	α
Thermal	45	531	0.085	270	747	0.362
FEA-1	0.289	2.784	0.104	0.557	1.780	0.313
FEA-2	0.280	2.815	0.100	0.537	1.861	0.288
CIAE	0.074	1.899	0.039	0.065	1.682	0.039
Fission	0.072	1.950	0.037	0.054	1.800	0.030

Note: thermal--0.0253 eV;
FEA-1---spectrum of fast energy amplifer by Rubbia[10];
FEA-2---spectrum of sodium cooled FBR with 0.455MeV average neutron energy;
CIAE---- histogram spectrum of 0.7 to 1.0 MeV;
Fission-- fission neutron spectrum.

The main long-lived minor actinides produced in ADS include ^{237}Np, $^{238,240,241,242,244}Pu$, $^{241,242,242m,243,244}Am$ and $^{242\text{-}248}Cm$. The (n, γ), (n,2n) and (n,f) cross sections for these nuclides are important. Due to fission process for these nuclides will play important role in neutron economy of the assembly, the neutron multiplicity in fission is also needed.

The calculation of MA production rate concerns the nuclide evolution in ADS. The first important nuclides are ^{232}Th and ^{233}U in Th→U cycle as well as ^{238}U and ^{239}Pu in U→Pu cycle. The (n, γ) and (n, f) cross sections for ^{232}Th, ^{238}U, ^{233}U and ^{239}Pu are basic data and have to be known very precisely in energy region up to several hundreds MeV. The neutron spectrum above 20 MeV of incident neutron energy for these four nuclides is very unclear and need to be measured immediately. In fast ADS system, (n, n') process for ^{232}Th and ^{238}U has also to be considered.

Some nuclides with short life such as ^{233}Pa and ^{239}U play important role in nuclide evolution process, their (n, γ) and (n, f) cross sections are also needed. Unfortunately, measurement for these unstable targets is

very difficult. Usually the measured data for these nuclides are unavailable and have to be calculated based on model theory. The nuclear theory code to predict cross sections for unstable nuclides should to be developed.

The Long Lived Heavy Elements (LLHE) comes from parasitic reactions which are produced on the different constituents in the fuel.

For the Th-U cycle, the important parasitic reactions to produce LLHE are[11]

$^{233}U(n,2\,n)\,^{232}U$,

$^{232}Th(n,\,2n)^{231}Th \rightarrow \beta^- + {}^{231}Pa$ and

$^{231}Pa(n,\,\gamma)\,^{232}Pa \rightarrow \beta^- + {}^{232}U$.

The ^{232}U are responsible for a large part of the short-term (few centuries) radio-toxicity and ^{231}Pa responsible for the long-term radio-toxicity.

For the U-Th cycle, the important parasitic reactions producing LLHE include

$^{238}U(n,\,2\,n)\,^{237}U \rightarrow \beta^- + {}^{237}Np$,

$^{235}U(n,\,\gamma)\,^{236}U(n,\,\gamma) \rightarrow \beta^- + {}^{237}Np$,

$^{239}Pu(n,\,\gamma)^{240}Pu(n,\,\gamma)^{241}Pu(n,\,\gamma)\,^{242}Pu(n,\,\gamma)^{243}Pu \rightarrow \beta^- + {}^{243}Am$,

$^{241}Pu \rightarrow \beta^- + {}^{241}Am(n,\,\gamma)^{242}Am$,

$^{242}Am \rightarrow \beta^- + {}^{242}Cm(n,\,\gamma)^{243}Cm(n,\,\gamma)\,^{244}Cm$,

$^{243}Am(n,\,\gamma)\,^{244}Am \rightarrow \beta^- + {}^{244}Cm$,

$^{242}Cm \rightarrow \alpha + {}^{238}Pu$ and

$^{237}Np(n,\,\gamma)\,^{238}Np \rightarrow \beta^- + {}^{238}Pu$.

It is obvious that the LLHE production is driven by (n, γ) reactions. These (n, γ) cross sections are very important but poorly known.

V, Concluding Remarks

The nuclear data need for nuclear waste transmutation have been presented. There are huge quantities of the data needed to be measured, many of them need unstable samples. To extend the neutron energy region up to several hundreds MeV, the new measurement standard should be established. The model theory, evaluation methodology and related computer code need to be developed. Any single country can not undertake this huge task. The world nuclear data community should share their research resource and make more powerful cooperation coordinated under Nuclear Data Section of IAEA.

References

1, F. Venneri, C.Bowman and S. Wender, " The Physics Design of Accelerator-Driven Transmutation System", Proc. Int. Topl. Conf. Evaluation of Fuel Cycle for Future Nuclear Systems, Versailles, France, Sep. 11-14, 1995, Vol.1, p.476

2, S.Saito, " Research and Development Program on Accelerator-Driven Transmutation At JAERI", Proc. Second Int. Conf. on ADTT, Kalmar, Sweden, June 3-7, 1996,Vol.1, p.52

3, C.Rubbia et al., "Conceptual Design of a Fast Neutron Operated High Power Energy Amplifier", CERN/AT/95-44(ET), 1995; "Summary Report on EA Demonstration Facility Reference Configuration", EA B0.00 1 200 – Rev.0, 1999

4, G.Palmioti et al., "Uncertainty Assessment for ADS", NEA Workshop on Utilisation and Reliability of High Power Accelerators, Mito, Japan, Oct. , 1998

5, C.Rubbia, "Resonance Enhanced Neutron Captures for Element Activation and Waste Transmutation", CERN/LHC/97-04(EET), 1997

6, TARC collaboration group, "Neutron Driven Nuclear Transmutation by Adiabatic Resonance Crossing", EUR 19117 EN, 1999

7, M.Salvatores, "ADS in the Frame of Waste Management Activities in France", NEA Workshop on Utilisation and Reliability of High Power Accelerators, Mito, Japan, Oct. , 1998

8, Q. Shen et al., "Calculations of Nucleon Emission and Energy Deposition of Spallation Neutron Source Induced By Intermediate Energy Protons", CNIC-01468, 2000, p.231

9, Z.Zhao et al., "Conceptual Research on Reactor Core Physics for Accelerator Driven Sub-Critical Reactor", CNIC-01468, 2000, p.17

10, C.Rubbia et al., "Fast Neutron Incineration in the Energy Amplifier as Alternative Geologic Storage: Case of Spain", CERN/LHC/97- 01(EET), 1997

11, TOF collaboration group, "Proposal for a Neutron Time of Flight Facility", CERN/SPSC 99-8, 1999

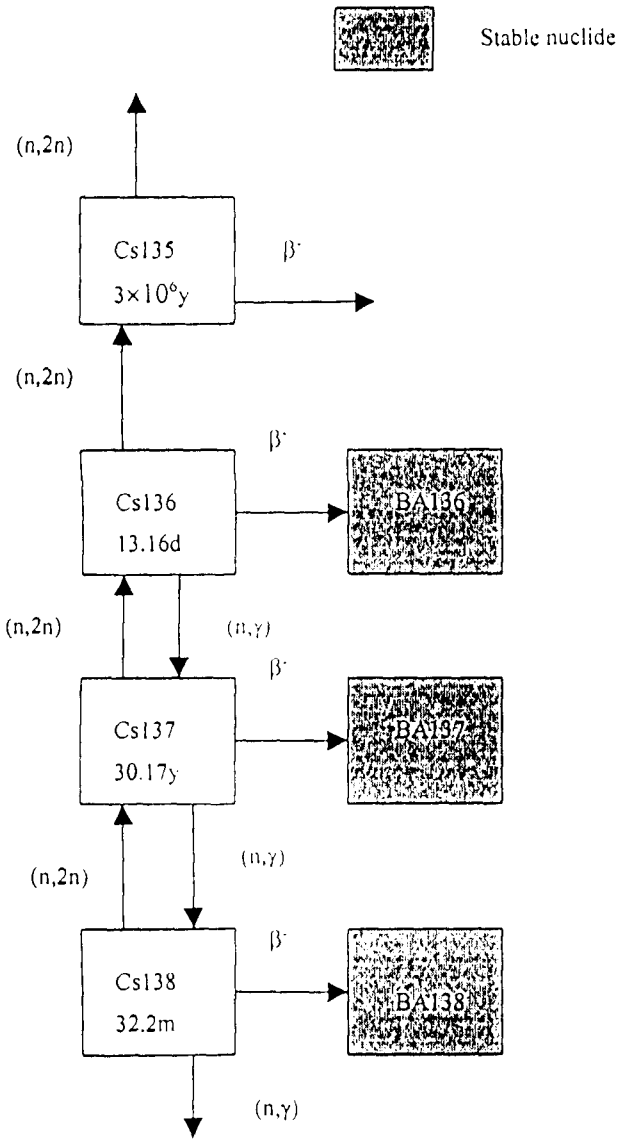


Fig.2 Transmutation chain for Cs^{137}

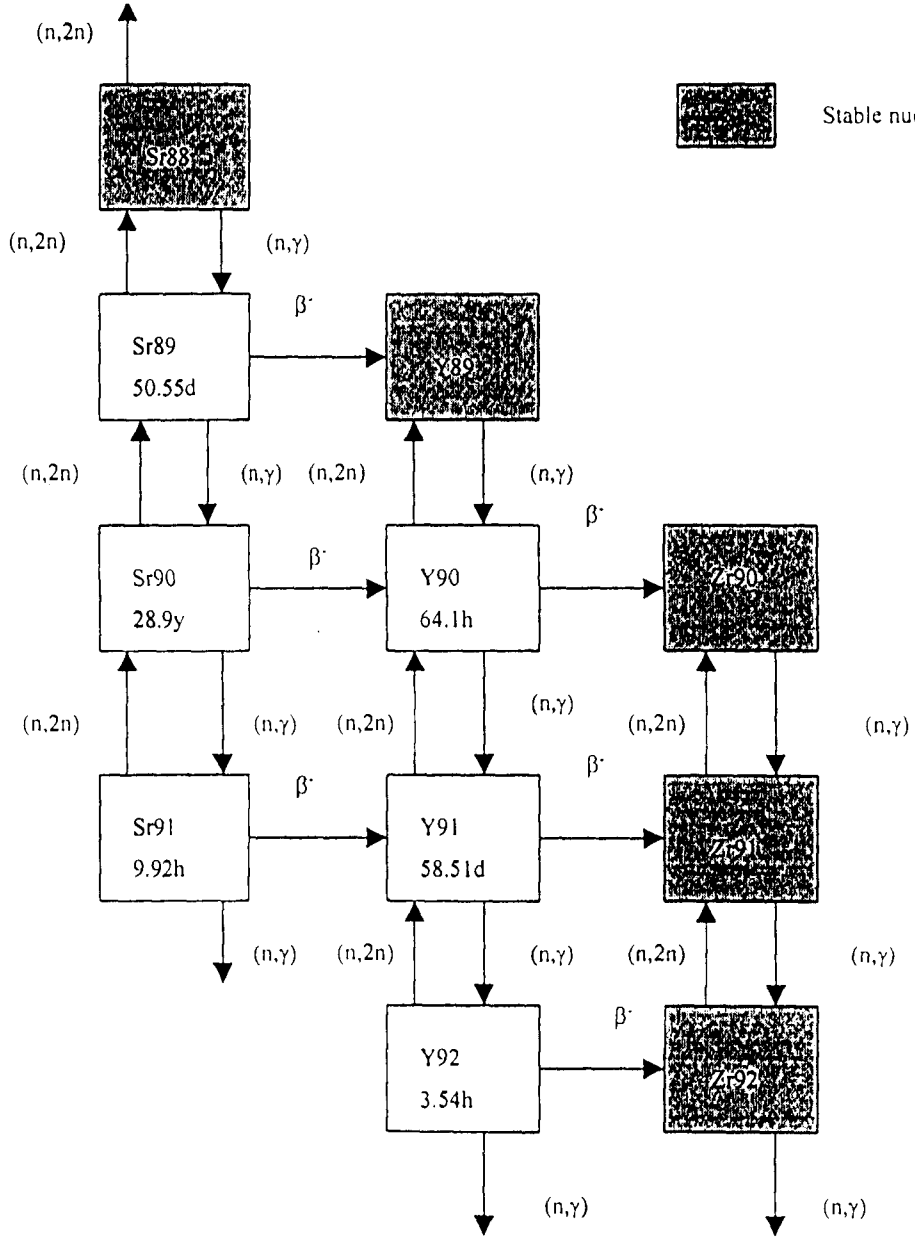


Fig.1 Transmutation chain for Sr^{90}

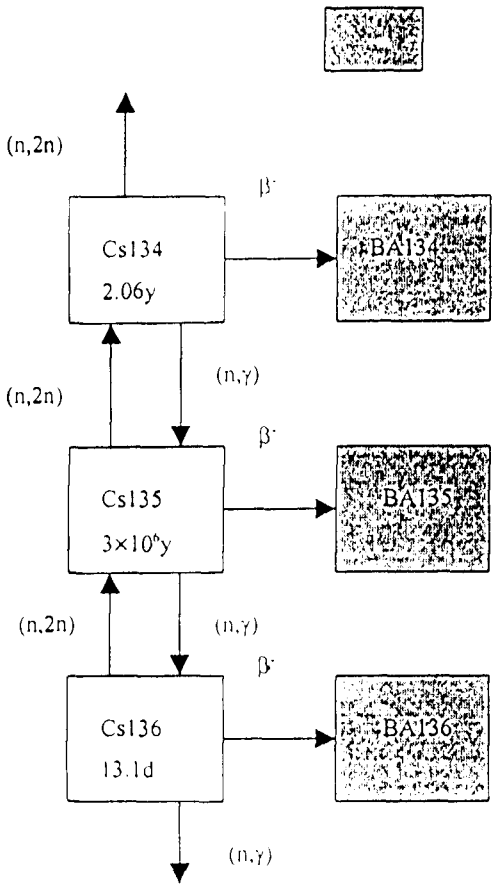


Fig.5 Transmutation chain for Cs^{135}

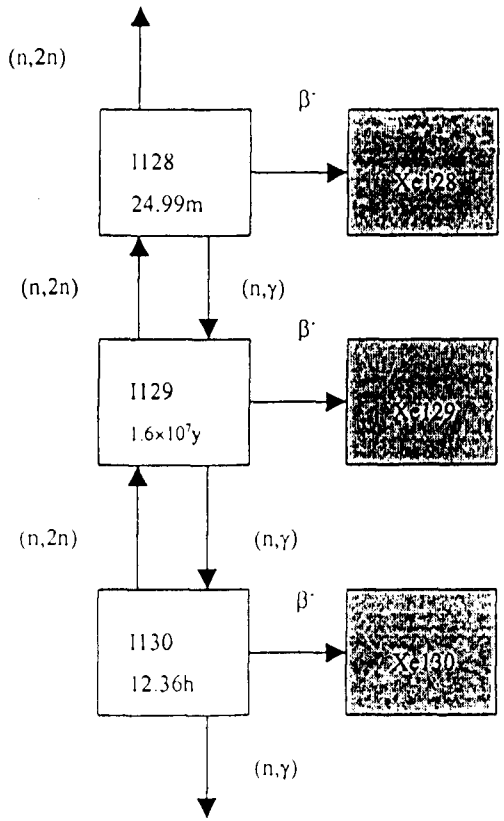


Fig.4 Transmutation chain for I^{129}

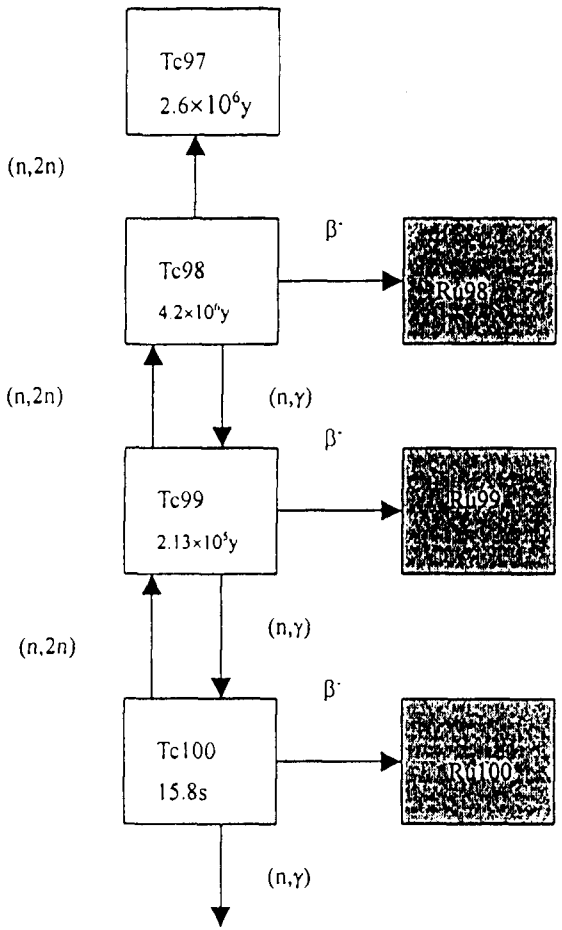


Fig.3 Transmutation chain for Tc^{99}

Nuclear Data Needs for Accelerator Driven Transmutation System

K. Tsujimoto, T. Sasa, H. Takano, T. Nakagawa

Japan Atomic Energy Research Institute, Japan

Abstract

The accelerator-driven transmutation system has been studied at the Japan Atomic Energy Research Institute. This system is a hybrid system which consists of a high intensity accelerator, a spallation target and a subcritical core region. Nuclides in high-level waste to be transmuted are minor actinides and long-lived fission products which are loaded in subcritical core and blanket in ADS, respectively. The calculations of subcriticality and burnup swing by present evaluated nuclear data were performed to investigate the effect on the performance of ADS. To compare with the integrated experimental and calculational results, analyses of actinide samples irradiated in the Dounreay Prototype Fast Reactor are presented. Recent measurements for thermal neutron capture cross section which affect transmutation efficiency are shown compared with evaluated nuclear data for some long-lives fission products.

1 Introduction

The Japanese long-term program called OMEGA has started in 1988 for research and development of new technologies for partitioning and transmutation of minor actinides and fission products. The main aims of this program are exploring the possibility to utilize high-level waste as useful resources and widening options of future waste management. Further improvements of long-term safety assurance in the waste management can be expected through establishing the partitioning and transmutation technology. Under the OMEGA Program, the Japan Atomic Energy Research Institute (JAERI) is proceeding with the research and development on proton accelerator-driven system (ADS).[1] Nuclides in high-level waste to be transmuted are minor actinides (MA) and long-lived fission products (LLFP). Transmutation of MA and LLFP was studied by using a lead-bismuth cooled ADS with 800MWth.[2] MA should be transmuted mainly through fission reactions because the transmutation of MA by neutron capture reactions has the possibility of increasing higher actinides, while the thermal capture is main transmutation reaction for LLFP. The mixture of the mono-nitride of plutonium and MA and inert matrix is used as the fuel for the subcritical fuel region surrounding the spallation target is driven by the spallation neutrons. It is possible to transmute LLFP in a thermalized region surrounding subcritical fuel region. The concepts for Pb-Bi cooled ADS are shown in Fig. 1.

The transmutation of MA and the burnup reactivity swing are especially important to estimate the performance of ADS. Nuclear data about MA is directly connected these characteristics. Fission and capture cross sections are dominant factor in transmutation and subcriticality of ADS. Neutron yield per fission and fission spectrum are also important in estimating of k-eff. The delayed neutron data which connects to operating and monitoring of subcritical level in ADS is one of important factor. For LLFP, accuracy of nuclear data affect transmutation efficiency because these nuclides are transmuted to stable nuclide by neutron absorption reaction.

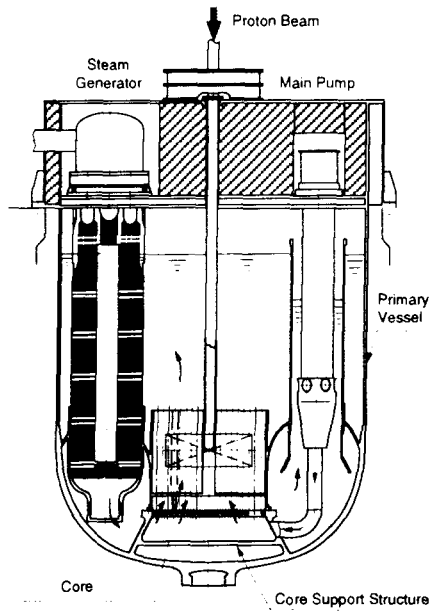


Fig. 1 Preliminary design of Pb-Bi cooled ADS plant

The effect of nuclear data for characteristics of ADS including present status of nuclear data about MA and LLFP are discussed following section.

2 Effect of Nuclear Data on Characteristics of ADS

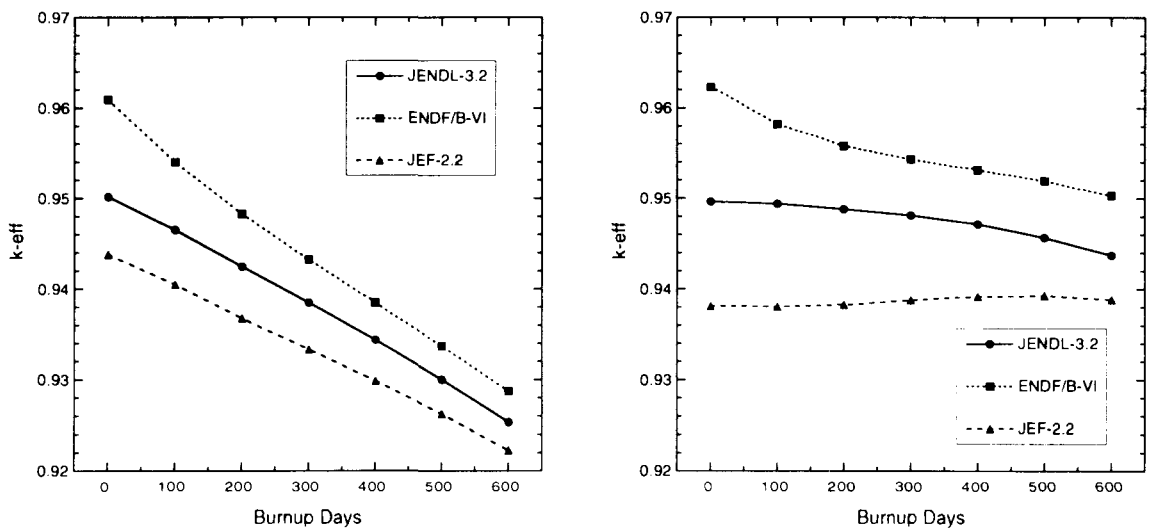
In ADS, plutonium and MA from power reactor are used as a fuel of subcritical core. Typical plutonium and MA compositions resulting from reprocessing of UO_2 and MOX fuel from PWR are showed in Table 1.[3] In all cases which are considered, seven years cooling times before reprocessing. The most different nuclide between MA from UO_2 and MOX fuel PWR is ^{237}Np. ^{237}Np is mainly produced from ^{235}U, so in MOX fuel with depleted uranium it produce less amount of ^{237}Np. These nuclides becomes initial loading fuel in ADS.

The accuracy of subcriticality and burnup swing are very important factor in ADS. The system must be subcritical in any case since ADS is driven with spallation neutron source by proton beam. The proton beam power needed to operate the ADS is connected to the multiplication factor (k-eff) of system, if the initial k-eff is 0.95, the burnup swing of 2% is corresponds to the proton beam swing of about 50%. Therefore, the minimization of the burnup swing is an important factor in operation of ADS. To investigate the effect of nuclear data on k-eff and burnup swing in ADS, burnup calculations are carried out using the ATRAS code system[4] with the JENDL-3.2, ENDF/B-VI and JEF-2.2 libraries, respectively. The burnup characteristics were investigated for ADS loading plutonium and MA from UO_2 and MOX fuel PWR with 50 GWd/t burnup. We assumed initial Pu loading as 40% for both cores because the burnup reactivity swing is minimized in the core with the initial Pu loading of 40%. The burnup calculations were done for five burnup cycles which was constructed by the burnup of two years and the cooling of three years. In the cooling period, the fission products were removed and the fresh fuel of equal mass to the fission products was added to the burnup fuel. The additional fresh fuel contained MA only, so Pu was loaded only once at the initial loading. The burnup swings in initial two years operation are showed in Fig. 2. The results show large discrepancy of about 1% for k-eff at initial core. The burnup swings, especially for Pu and MA from MOX

Table 1 Plutonium and MA compositions resulting from reprocessing of UO_2 and MOX fuel from PWR (nuclide atom %)[3]

Fuel Burnup	UO_2 33 GWd/t	UO_2 50 GWd/t	MOX 33 GWd/t	MOX 50 GWd/t
^{238}Pu	1.5	2.7	2.6	4.1
^{239}Pu	59.3	55.3	44.5	41.9
^{240}Pu	23.7	23.9	31.0	30.5
^{241}Pu	8.7	9.5	10.7	10.6
^{242}Pu	5.5	7.1	9.5	11.3
^{241}Am	1.3	1.5	1.7	1.7
Total	100	100	100	100
^{237}Np	44.6	46.4	4.5	4.4
^{241}Am	43.6	37.1	62.5	58.3
^{243}Am	9.7	12.7	24.3	26.1
^{244}Cm	2.1	3.8	8.7	11.3
Total	100	100	100	100

fuel PWR, also show different trend.



(a) Pu and MA from UO_2 fuel PWR

(b) Pu and MA from MOX fuel PWR

Fig. 2 Comparison of burnup swing during 600 days operation for ADS with fuel from UO_2 and MOX fuel PWR

To investigate the reason of discrepancy among results by different nuclear data, nuclides and reaction contributions for difference of k-eff at initial core were calculated by perturbation calculations. The results for MA from UO_2 fuel PWR at beginning of cycle (BOC) and end of cycle (EOC) are showed in Table 2 and Table 3, respectively. The values at BOC reflect the difference of nuclear data, but those at EOC include the difference of change in amount of each nuclides. The results show that contributions of capture reaction and neutron yield

are large while fission reaction seem to be smaller. The contributions of ^{237}Np and ^{241}Am between JENDL-3.2 and ENDF/B-VI, ^{241}Am between JENDL-3.2 and JEF-2.2 are dominant in the differences in capture reaction at BOC. For ^{237}Np, the differences in capture reaction in Table 2 indicates that evaluated value in JENDL-3.2 is different from other nuclear data. The capture cross section affect the burnup swing because capture reaction of ^{237}Np ,^{241}Am and ^{244}Cm produce ^{238}Pu ,^{242m}Am and ^{245}Cm which has relatively high fission cross section. ^{241}Am capture cross sections and energy break down of differences in capture reaction between nuclear data are showed in Fig. 3. It indicates that the energy region above 1 keV is dominant and contribution of capture reaction on k-eff is very sensitive to the difference of cross section. For neutron yield, the contributions of ^{243}Am and ^{241}Pu between JENDL-3.2 and ENDF/B-VI, ^{237}Np and ^{239}Pu are relatively large at BOC. The contribution of secondary produced MA, such as ^{238}Pu ,^{242m}Am and ^{245}Cm, become large with burnup. For the comparison between JENDL-3.2 and ENDF/B-VI, the difference in ^{242}Cm is large. This is because, in the evaluation of ENDF/B-VI, the fission cross section in the resonance region for ^{242}Cm is too small as shown in Fig. 4. This is one of the reason for difference of burnup swing. The branching ratio of ^{241}Am(n,γ)^{242g}Am and ^{241}Am(n,γ)^{242m}Am reactions is important. Its evaluated data, however, are only given in ENDF/B-VI, so a constant value of 0.8 for isomeric ratio of ^{241}Am(n,γ)^{242g}Am and total capture reaction was used for all calculations. A comparison with the evaluated and experimental data is showed in Fig. 5.[5,6] The data in the thermal region are satisfactory, but it is necessary to investigate in the higher energy region.

Table 2 Nuclides and reaction component for difference of initial k-eff in ADS with Pu and MA from UO_2 fuel PWR (%Δk/k)

Comparison between JENDL-3.2 and ENDF-B/VI

	Total	Σ_c	Σ_f	Σ_s	$\nu\Sigma_f$
^{237}Np	0.453	0.254	0.112	0.020	0.066
^{238}Pu	-0.041	-0.025	0.005	-0.009	-0.011
^{239}Pu	0.038	0.116	0.018	0.011	-0.107
^{240}Pu	0.287	0.135	-0.019	0.051	0.120
^{241}Pu	-0.158	0.054	0.032	-0.052	-0.190
^{242}Pu	-0.001	0.024	-0.001	-0.003	-0.021
^{241}Am	0.225	0.332	0.002	-0.077	-0.032
^{243}Am	0.347	0.187	-0.056	-0.089	0.304
^{244}Cm	0.019	-0.070	-0.005	-0.019	0.113
Total	1.170	1.008	0.087	-0.167	0.241

Comparison between JENDL-3.2 and JEF-2.2

	Total	Σ_c	Σ_f	Σ_s	$\nu\Sigma_f$
^{237}Np	0.104	0.257	0.136	0.157	-0.445
^{238}Pu	0.005	0.023	0.003	-0.009	-0.013
^{239}Pu	0.364	-0.038	-0.129	-0.007	0.537
^{240}Pu	0.109	0.037	-0.030	-0.007	0.107
^{241}Pu	-0.158	-0.122	0.006	-0.011	-0.034
^{242}Pu	-0.008	0.003	-0.004	0.015	-0.021
^{241}Am	-0.989	-1.441	0.079	0.278	0.095
^{243}Am	-0.106	-0.247	-0.036	0.144	0.033
^{244}Cm	0.060	0.060	-0.010	-0.012	0.021
Total	-0.619	-1.466	0.016	0.550	0.281

Table 3 Nuclides and reaction component for difference of k-eff at end of cycle in ADS with Pu and MA from UO_2 fuel PWR (%Δk/k)

Comparison between JENDL-3.2 and ENDF-B/VI

	Total	Σ_c	Σ_f	Σ_s	$\nu\Sigma_f$
^{237}Np	0.391	0.222	0.071	0.013	0.086
^{238}Pu	-0.333	-0.173	0.053	-0.059	-0.154
^{239}Pu	0.039	0.087	-0.007	0.008	-0.063
^{240}Pu	0.337	0.157	-0.031	0.049	0.162
^{241}Pu	-0.274	0.047	0.065	-0.037	-0.350
^{242}Pu	0.003	0.036	-0.001	-0.006	-0.046
^{241}Am	0.198	0.278	0.001	-0.057	-0.023
^{242m}Am	0.658	0.045	-0.116	0.021	0.708
^{243}Am	0.298	0.157	-0.048	-0.071	0.259
^{242}Cm	-0.880	0.052	0.252	-0.043	-1.141
^{243}Cm	-0.089	0.003	0.018	-0.002	-0.108
^{244}Cm	-0.036	-0.088	0.018	-0.023	0.057
^{245}Cm	0.134	-0.002	-0.023	-0.014	0.173
Total	0.445	0.819	0.266	-0.219	0.421

Comparison between JENDL-3.2 and JEF-2.2

	Total	Σ_c	Σ_f	Σ_s	$\nu\Sigma_f$
^{237}Np	0.106	0.152	0.057	0.106	-0.209
^{238}Pu	0.150	0.191	-0.006	-0.060	0.025
^{239}Pu	0.082	-0.010	-0.041	-0.000	0.133
^{240}Pu	0.128	0.041	-0.035	-0.010	0.132
^{241}Pu	-0.261	-0.081	0.042	-0.006	-0.216
^{242}Pu	0.008	-0.010	-0.021	0.028	0.022
^{241}Am	-0.678	-0.827	0.130	0.219	-0.200
^{242m}Am	0.279	-0.001	-0.080	-0.019	0.379
^{243}Am	-0.080	-0.162	-0.017	0.116	-0.017
^{242}Cm	-0.188	-0.009	0.026	-0.019	-0.186
^{243}Cm	0.042	0.002	-0.009	-0.000	0.050
^{244}Cm	0.161	0.086	-0.041	-0.0200	0.036
^{245}Cm	-0.068	0.006	0.029	0.004	-0.106
Total	-0.320	-0.621	-0.035	0.324	-0.057

The delayed neutron fraction is connected operating and control of ADS. Moreover, monitoring of subcritical level in ADS is important because ADS must be subcritical in any case. The subcriticality is measured by units of β_{eff} in many experimental methods. The present status of delayed neutron data for major nuclides in evaluated nuclear data file is showed in Table 4. ENDF-B/VI contains data for all nuclides except for ^{244}Cm. On the other hand, JENDL-3.2 and JEF-2.2 include only major Pu isotopes. Though JENDL-3.2 has part of delayed neutron data for all nuclides, complete set is necessary for evaluation of delayed neutron fraction. The contributions of these MA to delayed neutron fraction were investigated by calculation based on ENDF/B-VI library. As a result, it is about 30% for initial core and increase with fuel burnup. After five burnup cycles (about 10 years operation), it becomes about 60%. The complete set of these nuclides are desirable in future nuclear library from view point of MA transmutation system.

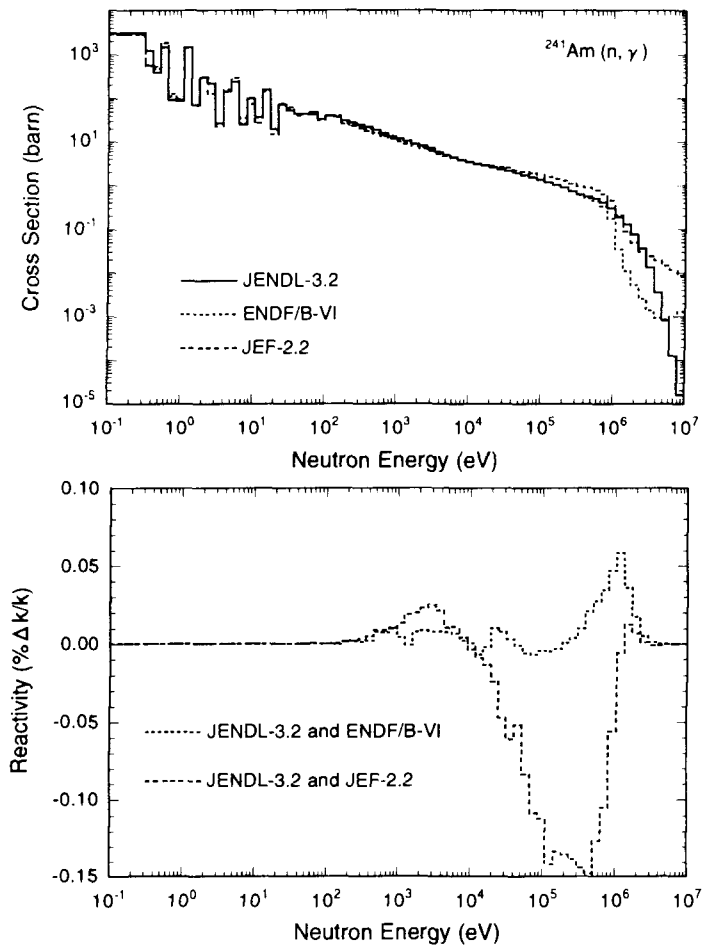


Fig. 3 ^{241}Am(n,γ) cross sections and energy break down of differences in capture reaction between nuclear data

For evaluation and improvement of nuclear data, not only differential experiments but also integrated experiments are indispensable. Actinide samples which were irradiated in the Dounreay Prototype Fast Reactor (PFR) are precious experimental data for MA.[7] This experiment was done under a joint research program between the United States and the United Kingdom, a part of solution of sample were brought to JAERI from Oak Ridge National Laboratory. The samples were milligram quantities of actinide oxides of 21 different isotopes from thorium to curium that had been encapsulated in vanadium holders and exposed for 492 effective full-power days. The results of chemical analyses and comparison with calculational results are shown in Table 5. In Table 5, difference between beginning and ending of chemical analyses for main isotopes and fission per initial metal atom (FIMA) for samples are presented. The calculations were done by ORIGEN-2 code[8] with JENDL-3.2 library. For difference during irradiation for main isotopes, the comparison with calculational results show good agreement with the experiments, except for ^{236}U, ^{238}Pu, ^{240}Pu and ^{242}Pu. For neptunium, americium and curium, there are large disagreement for FIMA while good agreement in difference for main isotopes. These results is preliminary one , so it will be needed more detailed calculation analysis to investigate the reason for these disagreement.

Some of fission products contained in residual waste from reprocessing have extremely long-term radiotoxicity. Partitioning and transmutation of the fission products are attracting considerable attention at present as an option to reduce the long-term radiological hazard of the

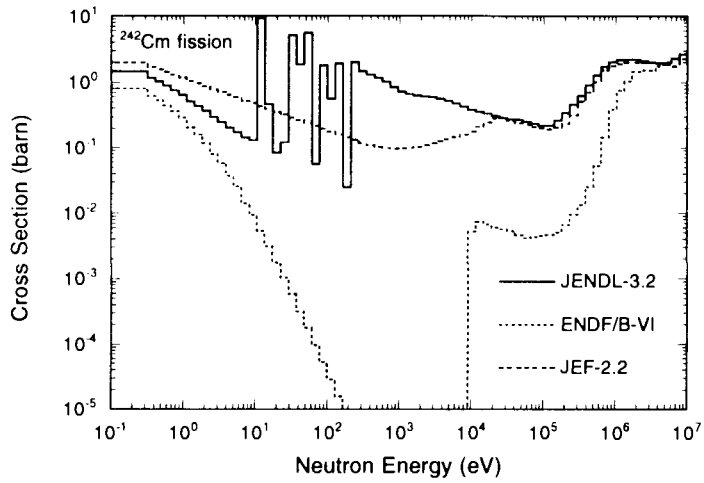


Fig. 4 ^{242}Cm fission cross section

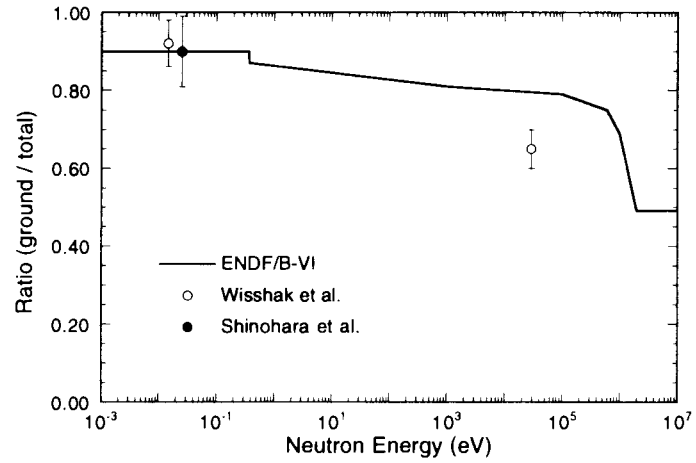


Fig. 5 Isomeric ration of ^{241}Am(n,γ)^{242g}Am and total ^{241}Am(n,γ) cross sections[5,6]

high-level nuclear waste. After ^{137}Cs and ^{90}Sr are decayed out, the still remaining toxicities are from only the 7 LLFPs (^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I and ^{135}Cs). Among this 7 LLFPs, ^{99}Tc and ^{129}I are soluble in water and the most troublesome nuclides on the geological disposal technology, though these potential hazards are smaller than those of MA. In JAERI proposed system, for example, the Iodine are loaded axially and radially with the from of NaI around the MA-fuel core in ADS. It was shown that an ADS can transmute the MA and Iodine generated from about 10 units if LWR ^{99}Tc and ^{129}I become stable nuclide ^{100}Ru and^{130}Xe according to following reactions.

$$^{99}Tc(n,\gamma)^{100}Tc \xrightarrow{16s}_{\beta} {}^{100}Ru(stable)$$

$$^{129}I(n,\gamma)^{130}I \xrightarrow[\beta]{12h} {}^{130}Xe(stable)$$

Therefore, accuracy of capture cross section for ^{99}Tc and ^{129}I directly affect transmutation efficiency. Recent measured thermal cross sections (σ_0) and resonance integrals (I_0) in Japan

Table 4 Present state of delayed neutron data for major minor actinides in evaluated nuclear library

	JENDL-3.2	ENDF-B/VI	JEF-2.2
Np-237	△	○	×
Pu-238	△	○	×
Pu-239	○	○	○
Pu-240	○	○	○
Pu-241	○	○	○
Pu-242	△	○	×
Am-241	△	○	×
Am-242m	△	○	×
Am-243	△	○	×
Cm-244	△	×	×
Cm-245	△	○	×

○: all data exit,△: only ν_{di} and λ_i,×: no data

Table 5 Results of chemical analyses on PFR-irradiated actinide sample and comparison with calculation results

Sample	atom/IMA[a]	(C/E)	FIMA[b](%)	(C/E)
^{233}U	-0.487	(0.99)	44.8	(0.99)
^{234}U	-0.185	(0.96)	9.19	(1.06)
^{235}U	-0.430	(1.01)	33.7	(1.05)
^{236}U	-0.100	(1.33)	5.93	(1.09)
^{238}U	-0.070	(0.94)	2.00	(1.01)
^{237}NP	-0.356	(1.06)	11.7	(1.45)
^{238}Pu	-0.453	(1.15)	21.9	(1.34)
^{239}Pu	-0.415	(1.03)	32.6	(1.07)
^{240}Pu	-0.206	(1.12)	10.9	(1.14)
^{241}Pu	-0.504	(1.01)	28.4	(1.13)
^{242}Pu	-0.158	(1.20)	6.25	(1.10)
^{241}Am	-0.396	(1.01)	10.1	(1.13)
^{243}Am	-0.335	(0.97)	5.66	(1.11)
^{243}Cm	-0.365	(0.97)	31.0	(1.18)
^{244}Cm	-0.519	(0.99)	11.7	(1.34)
^{246}Cm	-0.084	(0.97)	8.45	(1.24)
^{248}Cm	-0.105	(1.26)	6.95	(1.36)

[a] Difference between starting and ending atom per initial mass atom

[b] Fission per initial mass atom

Nuclear Cycle Development Institute[9,10] are shown in Table 6. Recent results of σ_0 is about 10% larger than the evaluated value. For resonance and keV region energy range, there is a few experimental data as shown in Fig. 6, so measurements with considerable precision are desired

Table 6 Thermal neutron capture cross sections (σ_0) and resonance integrals (I_0)

$^{99}Tc(n,\gamma)^{100}Tc$

		σ_0(b)	I_0
Harada *et al.*[9]	'95	22.9 ± 1.3	398 ± 38
Lucas *et al.*[11]	'77	20 ± 2	186 ± 16
JENDL-3.2		19.65	311.1
ENDF/B-VI		19.57	350.4
JEF-2.2		19.14	304.2
BROND-2		19.14	304.2

$^{129}I(n,\gamma)^{130}I$

		σ_0(b)	I_0
Nakamura *et al.*[10]	'96	30.3 ± 1.2	33.8 ± 1.4
Roy *et al.*[12]	'58	26.7 ± 2.0	36.0 ± 4.0
Block *et al.*[13]	'60	31 ± 4	
JENDL-3.2		27.01	28.98
ENDF/B-VI		27.17	35.56
JEF-2.2		33.93	30.28
BROND-2		26.93	27.97

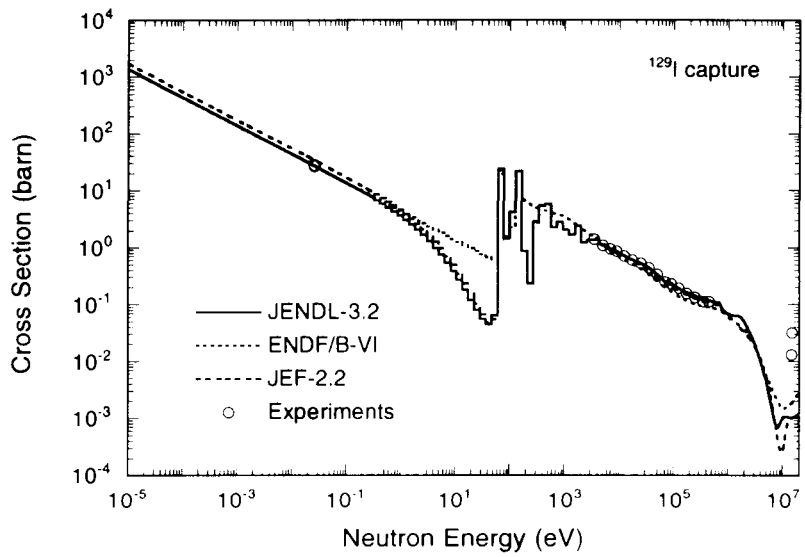


Fig. 6 ^{129}I capture cross section

3 Concluding Remarks

The conceptual design study of ADS are in progress at JAERI under the OMEGA program. Nuclides in high-level waste to be transmuted are MA and LLFP. JAERI proposed transmutation system for MA and LLFP was showed, and the effect of nuclear data for system characteristics, especially subcriticality and burnup swing, were investigated. From these results, the status of the evaluated data for the fission reaction is satisfactory for main nuclides in ADS, while large discrepancies are found in ^{242}Cm. The capture cross section are discrepant especially for ^{237}Np

and ^{241}Am. The importance of the nuclear data for secondary produces MA, ^{238}Pu, ^{242m}Am and ^{245}Cm, are also showed. About ^{242m}Am, further investigation for the ratio of ^{241}Am(n,γ)^{242g}Am and ^{241}Am(n,γ)^{242m}Am reactions in higher energy region is needed. The present status of fission neutron yield, delayed neutron data and fission neutron spectrum in present evaluated nuclear data is not sufficient. For LLFP, recent measurement results for ^{99}Tc and ^{129}I which are the troublesome nuclides on the geological disposal were presented.

References

[1] T.Takizuka, T.Sasa, K.Tsujimoto and M.Mizumoto, *Proc.* Global '97, vol.1 p.422 (1997).

[2] K. Tsujimoto, T. Sasa, K. Nishihara, T. Takizuka, H. Takano, K. Hirota, Y. Kamishima, *Proc.* ICONE-7 (1999).

[3] "Calculations of Different Transmutation Concepts", OECD/NEA Nuclear Science Committee (2000).

[4] T.Sasa, K.Tsujimoto, T.Takizuka and H.Takano, JAERI-Data/Code 99-007 (1999).

[5] K. Wisshak, J. Wickenhauser, F. Kappeler, G. Reffo, F. Fabbri, *Nucl. Sci. Eng.*, **81**, 396 (1982).

[6] N. Shinohara, Y. Hatsukawa, K. Hata, N. Kohno, *J. Nucl. Sci. Technol.*, **34**, 613 (1997).

[7] B.D.Murphy, T.D.Newton and S.Raman, ORNL-6689 (1996).

[8] A. G. Groff , ORNL-5621 (1980).

[9] H. Harada, S. Nakamura, T. Katoh and Y. Ogata, *J. Nucl. Sci. Technol.*, **32**, 395 (1995).

[10] S. Nakamura, H. Harada, T. Katoh and Y. Ogata, *J. Nucl. Sci. Technol.*, **33**, 283 (1996).

[11] M. Lucas, R. Hagemann, R. Naudet, C. Renson and C. Chevalier, IAEA-TC-119/14, 407, (1977).

[12] J. C. Roy and D. Wuschke, *Can. J. Chem.* **36**, 1424 (1958).

[13] R. C. Block, G. G. Slaughter and J. A. Harvey, *Nucl. Sci. Eng.*, **8**, 112 (1955).

Nuclear Models and their impact on ADS nuclear data *

Arjan Koning

NRG Nuclear Research and Consultancy Group
Westerduinweg 3
NL-1755 ZG Petten
Netherlands

* was presented at the meeting orally, and the main conclusions were included in the Summary Report of the meeting, published as report INDC(NDS)-423.

NECESSITY OF LONG TERM NUCLEAR DATA DEVELOPMENT FOR VARIOUS APPLICATIONS NEEDING NUCLEAR DATA

Tokio FUKAHORI

Nuclear Data Center, Japan Atomic Energy Research Institute,
Tokai-mura, Naka-gun, Ibaraki-ken, 319-1195 Japan
E-mail: fukahori@ndc.tokai.jaeri.go.jp

Abstract

Necessity of long term nuclear data development for accelerator-driven system target design, high-energy radiation shielding, medical application, space and astrophysical applications, etc. is described in this paper. For each application field needing nuclear data, considered were importance of nuclear data in determining the success or failure of the application, important gaps remaining in the nuclear data and feasibility of filling the gaps with a modest research effort. It can be concluded much more international discussions are required.

1 Accelerator-driven System Target Design and High-energy Radiation Shielding

1.1 Importance of Nuclear Data

Many kinds of accelerators are used for various applications. Working and planned accelerators in Japan are High Power Proton Accelerator for JAERI-KEK Joint Project, Heavy Ion Accelerator for Biological and Material Science (JAERI/TIARA), Heavy Ion Cancer Therapy (NIRS/HIMAC), Radioactive Beam Facility (RIKEN), Electron Accelerator for Positron Factory (JAERI/TIARA), Electron Accelerator for Photon and Photoneutron Source (JNC), Light Source (JAERI/Spring-8), Photon Factory (KEK), Research Accelerator (Tohoku University, CYRIC), Free Electron Laser (JAERI/FEL), other accelerators for radation therapy at Tsukuba University, RIKEN, RCNP, Hyogo Prefecture, etc. Though there are a lot of purposes for accelerators, items considered for a shielding design is rather similar, for example, neutron and/or radiation productions of an accelerator itself and surrounding equipments, radiation transport and activation estimation. Kinds of nuclear data related such items are activation cross sections and transport cross sections (double-differential particle production cross sections). However nuclides considered for a radiation shielding design distribute wide range: nuclides relevant to human body, structural materials, shielding materials, air and coolant materials, materials included in soil etc.

For a target design of accelerator-driven system, considered are not only target material itself but also materials of beam window, reflector, moderator, and coolant. The materials strongly depend on the concepts and options of target design: target for accelerator-driven transmutation of waste (ATW) or neutron source, solid or liquid target, subcritical assembly of fast reactor type or molten salt type, etc. Nuclear data needs for the target design of accelerator-driven system are double-differential neutron production cross section for neutron

transport calculation, activation cross section, charged-particle and gamma-ray production cross sections for heat calculation, etc. The elements considered for individual equipments can be summarized as following:

Target: Ta, W, Hg, Pb, Bi
Beam Window: V, Mo, W
Beam Window (HT-9): Cr, Mn, Fe, Ni, Mo
Beam Window (ceramics): C, O, Al, Si, Ti, Zn, Ba
Moderator: D
Reflector: Be, Ni, W, Pb
Coolant: H, O, He, Na, Hg, Pb, Bi

In addition, followings are included for ATW.

Fuel: Actinides, N-15, Cl (for molten salt)
Long-Lived FP: Tc-99, I-129

The concept and cost estimation of ATW Project at LANL has been summarized [1]. This report includes the needs of nuclear data library.

Nuclear data needs for radiation shielding are separated into two categories; transport calculation and activity estimation. They need double-differential particle emission cross sections and activation cross sections. The double-differential cross sections (DDX) for structural shielding materials are needed. Activation cross sections are needed for such as structural materials (beam window, beam tube, beam dump, magnet, moderator, reflector), coolant, air, soil, etc. in order to consider radiation shielding. Radioisotope production is one of the major radiological issues at hadron accelerators. For example, major radioactive isotopes created through activation processes in intermediate energy region are Na-24, Sc-46, Mn-52,54, Co-56,57,58 for structural materials and T, Be-7,11, C-10,11,14,15, N-13,16, O-14,15, Ar-37,41 for air [2]. From Japanese High Priority Request List, nuclear data for following materials are required for the accelerator related applications (Table 1).

Structural Material: V, Fe, Ni, Cu, Nb, Mo
Magnet: Na, Ca, Cr, Fe, N, Cu, Nb
Beam Window: V, Mo, W
Beam Window (HT-9): Cr, Mn, Fe, Ni, Mo
Beam Window (ceramics): C, O, Al, Si, Ti, Zn, Ba
Beam Tube: Na, Al, Ca, Cr, Fe, Ni
Beam Dump: C, Fe, Cu
Moderator: D
Reflector: Be, Ni, W, Pb
Coolant: O, Ga, Pb, Bi
Soil: C, Si
Air: N, O, Ar

In addition, photoneuton production data by high-energy photon are necessary for shielding of light source and free-electron-laser facilities. The photonuclear data for nuclides related to structural materials, beam tube, magnet, beam dump (C, Fe, heavy concrete), etc.

1.2 Important Gaps Remaining in the Data

Radioisotope (RI) production and particle transport are the major radiological issues at hadron accelerators for radiation protection and shielding. Radioisotope production cross sections are available only for some of the channels of interest and even for some certainly important ones data are difficult to find or do not exist. The transport cross sections (DDXs) are much less than activation cross sections and exist only for some structural and target materials. The required accuracies for each channel of nuclides are summarized in Table 1. If no experimental data exists for both differential and integral experiments, no one can confirm the reliability of the evaluated nuclear data.

Experimental data for activation cross sections exist only for the nuclides having residuals, whose half-life lengths are good to measure. These half-life lengths are one for the important RI in principle. RIs having longer half-lives are also important. The status of experimental data of activation cross sections for these nuclides and those of less interest than structural, target, beam dump materials is poor. For experimental data of the neutron DDXs, those of proton-induced case for structural and target materials are better situation. However nuclides, which have less (basic) physical interests, are poor. In addition, neutron induced cases are very rare for both activation cross sections and neutron DDXs. It is impossible to prepare reliable evaluated data satisfying requirements such as in Table 1 without reliable experimental data. For a lot of nuclides, the reliable experimental data are still insufficient. There is no data for photoneutron production data in high energy region.

1.3 Feasibility of Filling the Gaps with a Modest Research Effort

A lot of approaches from software side have been done. Computer codes have been developed by using physical models and systematics and model parameters have been corrected through many efforts such as IAEA/CRP on Reference Input Parameter Library. From experimental approaches for very important nuclides have been performed. However more efforts are needed from both code developments and measurements in order to reach the required accuracy. To fill the gaps between requirements and evaluation of nuclear data, differential and integral experiments must be performed. If those experimental data are available, the developments of physical models and codes can follow. So, it can be suggested for researches with the modest effort:

1) To collect the concepts of accelerator-driven system internationally for such as target, coolant, moderator, reflector and shielding materials,
2) To pick up nuclides needed for evaluation,
3) To determine those priorities,
4) To share and/or collaborate the experimental efforts for both differential and integral measurements concentrating high priority one according to the above determination,
5) To evaluate nuclear data and make an international comparison of both evaluated and experimental data, and
6) To discuss the difference between the evaluated results.

2 Medical Applications

2.1 Importance of Nuclear Data

Nuclear data needs for medical applications are well summarized by IAEA efforts [35].

Here, additional information from Japanese Nuclear Data Committee (JNDC) is described. In general, nuclear data for medical application can be categorized as;

1) Nuclear data for medical equipments for diagnosis and treatment,
2) Nuclear data related to RI researching chemical behavior in human body, and
3) Nuclear data for radiation interaction in human body.

2.2 Important Gaps Remaining in the Data

In the field between mediphysics and medical fields, recent requirements in categories 1) and 2) seems to be rather satisfied. On the other hand, microscopic data evaluating interaction processes at the final stage with human body such as absorbed dose are very poor. The data are needed for radiation interaction at the atomic and molecular levels, for example, electron (< 10 keV) interaction with DNA. From the point of view of radiation protection, decay data (decay mode, branching ratio, half-lives, gamma-rays, alpha and beta-rays, energies and intensities), cross sections and DDXs for radiation transport, KERMA factor etc. are necessary. The increase of accuracy is also important for beta- and gamma-ray yields of RI produced through high-energy neutron and proton interactions.

For recent radiation therapy, additional needs for photon (15 keV – 10 MeV), proton, neutron and high-energy electron induced data exist. Especially in the case of decay data, many decay data have been revising in ICRP Publication 38, whose old version used decay data from 1970s ENSDF! In the situation of difficulty for new reactor construction, securing low-energy neutron source by using electron (using (gamma, n) reaction) or proton (using (p, n) reaction) accelerator is important for Boron Neutron Capture Therapy (BNCT). For such purpose, nuclear data for target nuclei is needed (Table 2).

2.3 Feasibility of Filling the Gaps with a Modest Research Effort

Data needs for nuclear reactions seem to be similar as the accelerator-driven system. The big difference is that radiation protection field really needs the improvement of decay data including revision and addition. It looks rather on going project in the ENSDF group. However securing the manpower is the most urgent problem.

3 Miscellaneous Usage

3.1 Importance of Nuclear Data

Space and Astrophysical Applications: For cosmic-ray interaction study, reaction data for evaporation, spallation and fission cross sections are important [7,8]. For nucleosynthesis study, nuclear data related to capture cross section for mass chain in the keV energy region for S-process and mass yield data of fission reaction for R-process is necessary. The damage (ex. Single Event Up-set) of space ship equipments and radiology of astronauts require nuclear data for proton and heavy-ion induced reactions.

Radiation Damage Study: Recent microscopic approach of radiation damage study requires primary knock-on atom (PKA) spectra directly from nuclear reaction. For revising the reliable scale of amount of radiation damage, data of displacement per atom (DPA) cross section should be calculated also directly from nuclear reaction data. Nuclear heating and dose estimation of human body requires precise KERMA factor calculated from nuclear reaction.

Neutron Source for Neutron Calibration Fields: Development of monoenergetic neutron calibration fields [6] is necessary to calibrate neutron detectors. Nuclear data for proton and deuteron-induced neutron production data is required to nuclides for target (T, D, Li (metal, LiF), Sc), backing and structural materials (C, O, Cu, Fe, Al, Pt, Ti, Mo), source of neutron contamination (D(d,n) for D-T source, C(d,n), O-16(d,n) for D-D neutron), etc.

3.2 Important Gaps Remaining in the Data

Space and Astrophysical Applications: The reaction data for evaporation, spallation and fission cross sections for cosmic-ray are partly satisfied for proton-induced reactions by using the evaluated high energy library. Since the spectrum peak of cosmic-ray stands around several tens of MeV up to 100 MeV, only proton reaction data are available while much less heavy-ion reaction data. The neutron capture cross section data for mass chain in the keV energy region for nucleosynthesis are rather better for the nuclides near the beta-stable line, but not for those far from the line. The mass yield data of fission reaction are also available, however for wide energy region, the experimental data are poor and accuracy of evaluated data can not be confirmed, especially for minor actinides.

Radiation Damage Study: Big efforts were done by JAERI and LANL to calculate PKA spectra directly from evaluated nuclear data and during nuclear model calculation, respectively. However, because of the difficulty of direct measurement of PKA spectra, the reliability can not be confirmed. Only KERMA factor can be compared with measured data, and the PKA data can be checked indirectly. Therefore we can not recognize the accuracy of PKA data.

Neutron Source for Neutron Calibration Fields: The accuracy of generated monoenergetic neutron yield and spectra is not enough.

3.3 Feasibility of Filling the Gaps with a Modest Research Effort

The method to filling the gaps is similar to the high energy nuclear data evaluation mentioned at the section 1.3.

4 Concluding Remarks

Importance and needs of nuclear data were described for accelerator-driven system, high-energy shielding, medial application, space and astrophysical applications, radiation damage study, and neutron source for neutron calibration field. Many nuclear data such as activation, capture and the other cross sections, double differential cross sections for many of particle productions, PKA spectra, KERMA factors are required for various incident particles. Since we have to consider such a big amount of nuclear data by limited manpower, it is necessary to put the priorities to nuclear data development for each application field and share the manpower internationally. It is clear that the international collaboration is necessary. However, the individual evaluation efforts group by group like JENDL, ENDF, JEFF, etc. are also important, in order to keep the abilities and activities of nuclear data development in each group. We have to consider the balance about above two different directions. From this point of view, lots of international discussions are required.

Acknowledgements

The author would like to thank Drs. Y. Yamaguchi, T. Kobayashi, K. Shin and H. Yoshizawa for their helpful discussion and information to prepare this manuscript.

References

[1] F. Venneri, D. Crawford, H. Khalil, N. Li, T. Allen, M. Louthan, K. Woloshun, A. Brunsvold, C. Ehrman, D. Wade, M. Cappiello, M. Chadwick, P. Lisowski, S. Hayes, J. Herceg, P. MacDonald: "Rosdmap for the Development of Accelerator Transmutation of Waste: Target and Blanket System – Report of the Target and Blanket Technical Working Group", LA-UR-99-3022 (1999).
[2] M. Huhtinen: "Determination of Cross Sections for Assessments of Air Activation at LHC", CERN/TIS-RP/TM/96-29 (1997).
[3] "Nuclear Data for Neutron Therapy: Status and Future Needs", IAEA-TECDOC-992 (1997).
[4] "Status of Nuclear Data Needed for Radiation Therapy and Existing Data Development Activities in Member States", INDC(NDS)-365 (1997).
[5] "Summary Report of the Third Research Co-ordination Meeting on Development of Reference Charged-Particle Cross Section Database for Medical Radioisotope Production", INDC(NDS)-388 (1998).
[6] M. Baba et al.: Nucl. Instr. and Meth. **A376** (1996) 115.
[7] P. Silberberg and C.H. Tsao: Astrophy. J. Supp. Series No. 220(II), 25 (1973) 315.
[8] P. Silberberg and C.H. Tsao: Astrophy. J. Supp. Series No. 220(II), 25 (1973) 335.

Table 1 Summary of data needs for ADS target and shielding design from Japan

Reaction	Quantity	Energy Range [MeV]	Accuracy	Purpose
D(p,xnyp),(n,xnyp)	DDX	20 - 3000	50%	Moderator
D(p,n)	Cross Section & DDX		50%	
D(n,ela)	Cross Section & Ang. Dist.		20%	
Be(p,xnyp)(n,xnyp)	DDX	20 - 3000	50%	Target, Reflector
Be(p,x) Activation	Cross Section		30%	
C(p,xnyp)	DDX	800 - 1500	50%	Beam Window (ceramics)
C(p,x),(n,x) Activation	Cross Section	20 - 3000	30%	Beam Dump, Soil
C(p,xn)	DDX	20 - 3000	50%	Beam Dump
C(n,xn)	DDX	20 - 150	50%	Beam Dump
N(p,x) ,(n,x) Activation	Cross Section	20 - 3000	30%	Air
O(p,x) ,(n,x) Activation	Cross Section	20 - 3000	30%	Air & Cooling Water
O(p,xnyp)	DDX	800 - 1500	50%	Beam Window (ceramics)
Na(p,x) ,(n,x) Activation	Cross Section	20 - 3000	30%	Magnet & Beam Tube
Al(p,x) Activation	Cross Section	20 - 3000	30%	Beam Tube
Al(p,xnyp)	DDX	800 - 1500	50%	Beam Window (ceramics)
Si(p,xnyp)	DDX	800 - 1500	50%	Beam Window (ceramics)
Si(p,x) ,(n,x) Activation	Cross Section	20 - 3000	30%	Soil
Ca(p,x) ,(n,x) Activation	Cross Section	20 - 3000	30%	Magnet & Beam Tube
Ti(p,xnyp)	DDX	800 - 1500	50%	Beam Window (ceramics)
V(p,x) Activation	Cross Section	800 - 3000	30%	Beam Window
V(p,z)	DDX		50%	
Cr(p,x) ,(n,x) Activation	Cross Section	20 - 3000	30%	Magnet & Beam Tube
Cr(p,z)	DDX	800 - 3000	50%	Beam Window (HT-9)
Cr(p,xnyp)	DDX	800 - 1500	50%	Beam Window (HT-9)
Mn(p,xnyp)	DDX	800 - 1500	50%	Beam Window (HT-9)
Fe(p,xnyp)	DDX	20 - 3000	50%	Beam Window (HT-9) Beam Dump
Fe(p,x) ,(n,x) Activation	Cross Section	20 - 3000	30%	Magnet, Beam Tube, Beam Dump
Fe(n,xn)	DDX	20 - 150	50%	Beam Dump
Fe(p,z)	DDX	800 - 3000	50%	Beam Window
Ni(p,x) ,(n,x) Activation	Cross Section	20 - 3000	30%	Magnet, Beam Tube, Reflector
Ni(p,z)	DDX	800 - 3000	50%	Beam Window
Ni(n,xn)	DDX	20 - 150	50%	Reflector
Ni(p,xnyp)	DDX	800 - 1500	50%	Beam Window (HT-9)
Cu(p,xnyp),(n,xnyp)	DDX	20 - 3000	50%	Structural Material Beam Dump
Cu(p,x) Activation	Cross Section	20 - 3000	30%	Super Conductive Magnet, Beam Dump
Cu(n,x) Activation	Cross Section	20 - 150	30%	Beam Dump
Cu(p,z)	DDX	800 - 3000	50%	Beam Window
Zn(p,xnyp)	DDX	800 - 1500	50%	Beam Window (ceramics)
Ga(p,x) Activation	Cross Section	20 - 3000	30%	Coolant
Nb(p,xnyp),(n,xnyp)	DDX	20 - 1500	50%	Structural Material
Nb(p,x) Activation	Cross Section	20 - 3000	30%	Super Conductive Magnet
Mo(p,x) Activation	Cross Section	800 - 3000	30%	Beam Window
Mo(p,z)	DDX	800 - 3000	50%	Beam Window
Mo(p,xnyp),(n,xnyp)	DDX	20 - 1500	50%	Structural Material
Mo(p,xnyp)	DDX	800 - 1500	50%	Beam Window (HT-9)
Ba(p,xnyp)	DDX	800 - 1500	50%	Beam Window (ceramics)

Table 1 Summary of data needs for ADS target and shielding design (cont.)

Reaction	Quantity	Energy Range [MeV]	Accuracy	Purpose
Ta(p,xnyp),(n,xnyp)	DDX	20 – 3000	50%	Target
Ta(p,x) Activation	Cross Section	20 - 3000	30%	Target
Ta(n,x) Activation	Cross Section	20 - 150	30%	Target
Ta(p,xn)	DDX	20 - 3000	50%	Target
Ta(n,xn)	DDX	20 - 150	50%	Target
Ta(p,x+gamma)	Spectra	20 - 3000	50%	Target
Ta(n,x+gamma)	Spectra	20 - 150	50%	Target
W(p,x) Activation	Cross Section	20 – 3000	30%	Target, Reflector
W(n,x) Activation	Cross Section	20 - 150	30%	Target, Reflector
W(p,xn)	DDX	20 - 3000	50%	Target
W(n,xn)	DDX	20 - 150	50%	Target
W(p,z)	DDX	800 - 3000	50%	Beam Window
W(p,x+gamma)	Spectra	20 - 3000	50%	Target
W(n,x+gamma)	Spectra	20 - 150	50%	Target
W(p,xnyp),(n,xnyp)	DDX	20 - 1500	50%	Target
Hg(p,xnyp),(n,xnyp)	DDX	20 – 3000	50%	Target
Hg(p,non),(p,ela)	Cross Section	0.1 - 3000	30%	Target
Hg(n,tot),(n,ela)	Cross Section	0.1 - 3000	30%	Target
Hg(p,x) Activation	Cross Section	20 - 3000	30%	Target
Hg(n,x) Activation	Cross Section	0.1 - 150	30%	Target
Hg(p,xn)	DDX	20 - 3000	50%	Target
Hg(n,xn)	DDX	1 - 150	50%	Target
Hg(p,x+gamma)	Spectra	20 - 3000	50%	Target
Hg(n,x+gamma)	Spectra	1 - 150	50%	Target
Pb(p,x) Activation	Cross Section	20 – 3000	30%	Target
Pb(n,x) Activation	Cross Section	20 - 150	30%	Target
Pb(p,xn)	DDX	20 - 3000	50%	Target
Pb(n,xn)	DDX	20 - 150	50%	Target, Reflector
Pb(p,x+gamma)	Spectra	20 - 3000	50%	Target
Pb(n,x+gamma)	Spectra	20 - 150	50%	Target
Pb(p,nyp),(n,xnyp)	DDX	20 - 1500	50%	Target
Bi(p,xnyp),(n,xnyp)	DDX	20 – 1500	50%	Target
Bi(n,g0) Po-210 Prod.	Cross Section	thermal - 20	15%	Coolant
Bi(p,g) Po-210 Prod.	Cross Section	6 - 20	20%	Target
Bi(p,x) Activation	Cross Section	20 - 3000	30%	Target
Bi(n,x) Activation	Cross Section	20 - 150	30%	Target
Bi(p,xn)	DDX	20 - 3000	50%	Target
Bi(n,xn)	DDX	20 - 150	50%	Target
Bi(p,x+gamma)	Spectra	20 - 3000	50%	Target
Bi(n,x+gamma)	Spectra	20 - 150	50%	Target

Table 2 Nuclear data needs for neutron source for BNCT

Reaction	Quantity	Energy Range [MeV]	Accuracy	Purpose
Li-7(p,n),(p,xn)	Cross Section & Spectra	threshold - 3	10%	MEDICAL (N-Source for BNCT)
Be-9(p,n),(p,xn)	Cross Section & Spectra	threshold - 3	10%	MEDICAL (N-Source for BNCT)

HIGH ENERGY NUCLEAR DATA FOR SPALLATION TARGET DESIGN

SYLVIE LERAY

DAPNIA/SPhN, CEA/Saclay, 91191 Gif-sur-Yvette, Cedex, FRANCE

High energy nuclear data are required in the design of spallation targets for spallation neutron sources or accelerator-driven sub-critical reactors, but also in other fields such as rare isotope production, space or astrophysics. Recently measured experimental data, regarding neutron, charged particles and residue production allow to test the nuclear models used in high energy transport codes. Although gross features are correctly reproduced by the models, severe deficiencies are pointed out. This could be improved by completing the set of experimental data, performing more constraining experiments and, principally, dedicating more theoretical work to get better or new high energy models.

1 Needs for high energy nuclear data

With the growing interest for accelerator applications, the collection of high energy (i.e. above 150-200 MeV) nuclear data is becoming an important issue.

With a high intensity, high energy (around 1 GeV), proton beam bombarding a heavy material thick target it is possible to produce, through spallation reactions, an intense neutron flux. Because of the recent progress in high current accelerators, spallation sources can now be competitive with fission reactors in terms of neutron flux. Several spallation neutron sources, used for condensed matter, material structure and biology studies, are already in operation but more powerfull ones are under study (among which SNS to be built in the USA and the ESS project in Europe) and should provide fluxes more intense by one or two orders of magnitude.

Accelerator-driven reactors, combine a spallation neutron source and a sub-critical reactor that could be used to transmute long-lived nuclear waste and produce energy. The spallation neutrons, after being more or less moderated, are used to drive the sub-critical reactor in which the transmutation takes place. Various concepts of accelerator-driven systems (ADS) have been proposed with different goals: transmutation of fission products, incineration of minor actinides or military Pu, or energy production with

a minimized waste inventory. Sub-critical reactors could also be designed to serve as irradiation tools for structural materials and fuel studies for nuclear power industry.

Accelerator-driven systems could be also dedicated to the production of rare isotopes: tritium for military purposes, as in the former APT project in the USA, radioisotopes for medical or industrial applications or exotic radioactive beams for fundamental research in nuclear physics and astrophysics.

Finally, spallation reactions plays a role in space industry since cosmic rays, which can induced severe radiation damages, are composed mostly of protons and alpha particles with an energy spectrum presenting a maximum near 1 GeV/A. They are also important in astrophysics to explain part of the elemental abundance distribution in galactic cosmic rays.

2 Important parameters in spallation target design

For all these applications, specific high energy nuclear data are needed. In particular, the design and optimisation of spallation targets requires a precise knowledge of the production of all the particles produced by spallation reactions in the target and surrounding structures, as the window between the accelerator vacuum and the target.

• Neutron production

The main parameter characterizing the performance of a spallation target is the number of spallation produced neutrons. In an ADS, the average number of neutrons per incident proton and per GeV is directly related to the energy gain between the accelerator and the sub-critical reactor and depends on the beam nature and energy, and, on the target material and geometry. On the same time, it is important to control the back-scattered particles which would hit the window isolating the target from the accelerator vacuum and the escaping high energy neutrons which lead to problems of shielding and radiation damage in structural materials. Also, for safety requirements, the distribution of the neutrons along the target and their energy have to be precisely known when the target is inside a sub-critical reactor core.

• Induced radioactivity

Spallation reactions on a heavy material target lead to the production of residual nuclei covering a large part of the chart of nuclides. A lot of the spallation produced isotopes are radioactive. It is important to know their production rate, since, in a spallation neutron source, it will be necessary to keep the activity due to short-lived nuclei low enough to ensure access for maintenance or in case of failure. Furthermore, it will be essential to minimize the radiotoxicity caused by long-lived isotopes, the major part of which comes from high energy nuclear reactions. These considerations will have to be taken into account in choosing the target and surrounding materials.

• Material damage

In ADS, material damage will certainly be a major problem to solve, a particularly critical point being the window between the accelerator vacuum and the target. Radiation damage due to atom displacements (DPA) will arise not only from neutrons but also from secondary charged particles and, especially in the window, from the pro-

ton beam interactions and recoiling residual nuclei. A large number of light charged particles, mainly protons and alphas, are produced in spallation reactions leading to an amount of hydrogen and helium formed in ADS that could be one or two orders of magnitude larger than in fission reactors possibly inducing swelling (because of bubble formation) and embrittlement of structural materials. Also the transmutation of part of the elements into impureties, even in very low concentration, could be responsible for embrittlement of solid materials (window or target container for instance) and, in the case of liquid metals could enhanced chemical corrosion problems.

All this implies that it is crucial to know the production rate of all spallation produced particles with their characteristics such as velocities and direction of emission.

3 Models and codes at high energies

For most applications where spallation reactions play a role, it is generally necessary to simulate complex configurations in which the incoming and secondary particules undergo successive interactions and are slowed down. This is simulated with Monte Carlo transport codes that should be able to properly describe all the occurring nuclear reactions from the primary GeV proton beam down to the low energy neutron interactions. Below 20 MeV, there exists validated Monte Carlo codes, which can transport neutrons and photons, utilizing reliable evaluated data files. Above 20 MeV (and for charged particles), specific high energy transport codes are used, in which the production cross-sections and energy and angle distributions are generated by nuclear physics models. Actually, an important effort, both experimental and theoretical, is being carried out in order to extend the evaluated cross-section libraries up to intermediate energies around 150-200 MeV (see the paper by A. Koning). Because, above these energies, the number of open decay channels in a reaction becomes too large to allow the generation of data files, it is not foreseen to go beyond. Therefore, reliable physics models are necessary to compute interactions from the GeV region down to intermediate energies. In fact, 150-200 MeV also roughly corresponds to the limit of validity of the models describing high energy interaction mechanisms.

A spallation reaction is generally described by a two steps mechanism: first, successive hard collisions between the incident particle and the individual nucleons of the target nucleus, leading to the emission of a few fast nucleons, then, decay of the excited remnant nucleus by emission of low energy particles or, sometimes for heavy nuclei, by fission. The first step is generally described by Intra-Nuclear Cascade (INC) models while evaporation-fission models are used for the second one. Some authors, introduce a pre-equilibrium stage between INC and de-excitation. Several INC models are available, the most well-known one is the Bertini model [1] dating from 1963. More recently two other models have been developed, the Isabel [2] and Cugnon [3] ones, which have brought some improvements in the physics. The most widely used evaporation model in the domain of spallation reaction is the Dresner model [4], usually associated with the Atchison [5] fission model.

Several high energy transport codes are available, most of them originating from the same HETC [6] code from Oak Ridge and mainly use the Bertini-Dresner combination.

4 Examples of recently obtained data

During the last years, a rather large amount of new high energy experimental data have been collected (for a review see [7]), both fundamental nuclear elementary cross sections to probe basic nuclear models, and thick target data to test the transport part of the codes. A significative part of it was realized in the framework of the GEDEON program [8] in France in collaboration with other European laboratories and now under the HINDAS [9] European project. A few examples are displayed below.

4.1 Neutron production

An example of double-differential cross-sections measured at the SATURNE accelerator in Saclay is shown in fig.1 from ref. [10]. Measurements were actually performed with protons and deuterons between 0.8 et 1.6 GeV on various thin targets. Fig.1 presents neutron cross-section distributions in Pb(p,xn)X reactions at 1200 MeV. The histograms are numerical calculations, performed with the TIERCE [12] code system. Within TIERCE two different intranuclear cascade codes followed by the same evaporation model [4] have been used: Bertini [1] (full line) and Cugnon [3] (dotted line) codes. Whatever the angle, calculations with the Bertini cascade overestimate the production cross-sections below 20 MeV while the Cugnon model generally leads to a much better agreement. A similarly good agreement would have been obtained with the Isabel INC model [2]. This can be explained by the larger excitation energy left in the nucleus after the INC phase in the Bertini than in the Cugnon or Isabel model.

Neutron energy spectra originating from thick target were also measured at SATURNE, on various cylindrical lead, tungsten, iron and aluminium targets. The results were compared with calculations performed with the TIERCE code with either the Bertini or the Cugnon INC model. It was found that both models lead to an underestimation of backward emitted high-energy neutrons. As for thin targets, low energy neutrons are generally overpredicted by the Bertini model. In fact, the total number of neutrons per incident proton escaping the target, calculated with the Bertini model, is found to be larger by 20% compared to Cugnon model. This makes a discrepancy which is rather large for applications to the design of spallation targets. Results concerning neutron multiplicity distributions [13], both in thin and thick targets, led to rather similar conclusions.

4.2 Light charged particles

Production cross-sections for $p, d, t, ^3He$ and α have been measured by the NESSI collaboration [14] at several energies between 0.8 and 2.5 GeV and on different targets. Calculations performed with LAHET [16] and HERMES [17] codes, which both use Bertini INC and Dresner evaporation models, have been found to significantly disagree with experimental data. Comparisons to particle energy spectra led to the conclusions that the discrepancy very likely comes from an inadequate parameterisation of the Coulomb barriers in the Dresner model. On the other hand, calculations done with the Cugnon INC and GEMINI [15] evaporation-fission models lead to a more satisfying agreement.

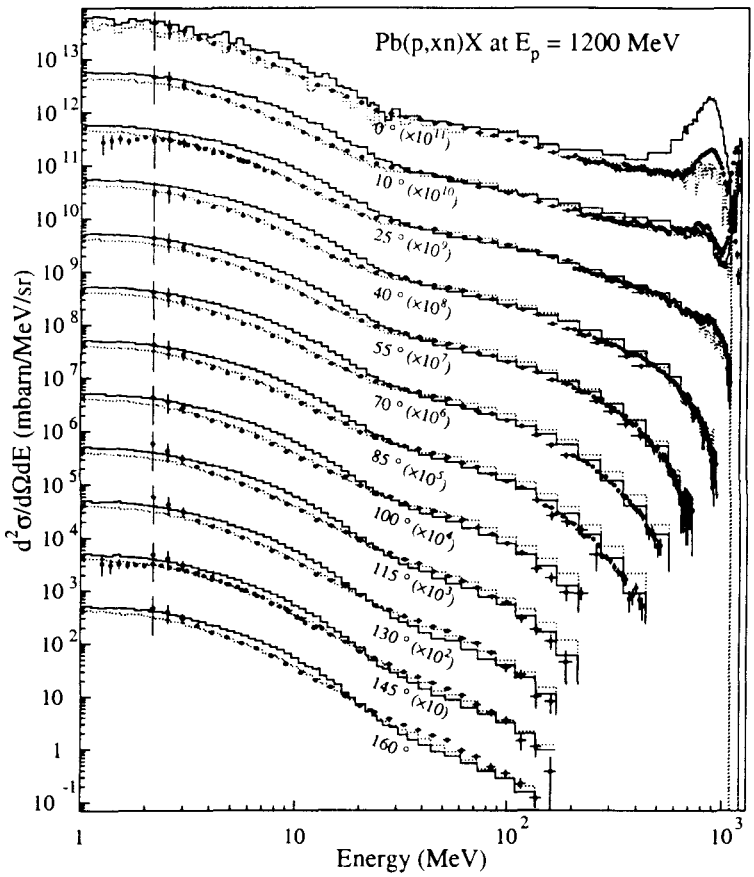


Figure 1: Neutron production double-differential cross-sections measured in proton induced reactions on a 2 *cm* thick Pb target at 1200. The histograms represent TIERCE calculations [12] using Bertini [1] (full line) or Cugnon [3] (dotted line) cascade model followed by the same evaporation model. From [10].

4.3 Residues

Up to recently, residues formed in spallation reactions were generally measured by radiochemical methods and γ-spectrometry (for instance [18]).

These kind of experiments suffers from the fact that the isotopes are identified after a radioactive decay chain. This makes the interpretation of the results rather difficult. A more direct measurement is possible using the heavy nucleus as a projectile on a hydrogen target in so-called reverse kinematic experiment. The various produced isotopes are then emitted in the forward direction with a velocity close to that of the beam and can thus be directly identified with a magnetic spectrometer. This technique, together with the fragment separator FRS, is being used at SIS accelerator in Darmstadt to identify isotopically all spallation residues, with production cross-sections larger than 0.1 mb, ranging from fission fragments to beam isotopes. So far, experiments with ^{197}Au [20], ^{238}U, ^{208}Pb [21] and ^{56}Fe beams at several energies on liquid hydrogen and deuterium targets have been carried out. Isotopic distributions of fragmentation residues in the case of the 1 GeV/A Pb on H_2 reaction are compared in fig.2 with three models: Bertini(+pre-equilibrium) and Isabel INC followed by the Dresner evaporation-fission and Cugnon INC followed by Schmidt [22] evaporation-fission model. None of

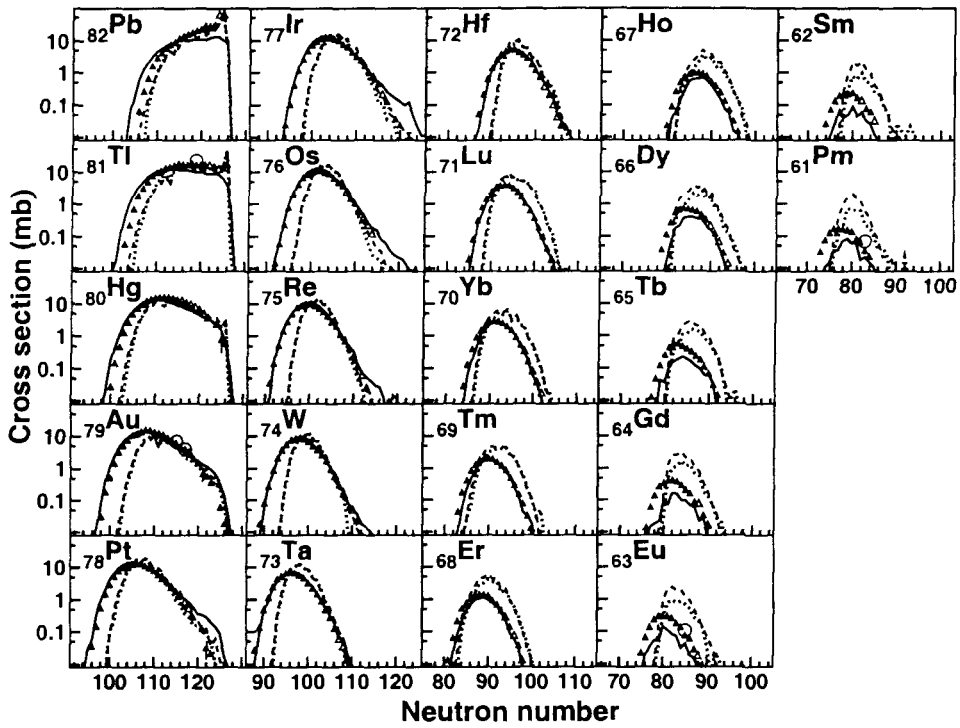


Figure 2: Isotopic production cross-sections of elements between Z=82 and 61, in the 1 GeV/A Pb on H_2 reaction, versus neutron number. Stable (resp. radioactive) isotopes are marked by open (resp. full) triangles. Gamma-spectroscopy data regarding shielded isotopes from [19] are plotted as open circles. The solid, dashed and dotted curves were calculated with the Cugnon-Schmidt, Bertini(+pre-equilibrium) and Isabel-Dresner models, respectively. From [21].

them is able to reproduce the whole set of experimental data and sometimes one order of magnitude discrepancy is observed. The shapes of the isotopic distributions obtained when using Dresner model are very similar whatever the INC and differ significantly from the experimental ones: they are shifted relative to the experimental ones towards the neutron-rich side. This could be ascribed to the fact that the prediction of the neutron-proton evaporation competition in the Dresner code is not satisfying.

An older similar experiment [23] had been performed at SATURNE for astrophysics purposes with an iron beam. From the elementary cross-sections, it was possible to calculate the production of impureties produced in an iron window supposed to have been irradiated by a proton beam for one year [24]. Results are shown in fig.3 together with simulations with the TIERCE code with either Bertini or Cugnon INC model. It can be seen that the code disagree by a factor two maximum with the rates obtained directly from the experimental data and that no clear conclusion on the best model can be drawn.

4.4 Coincidence experiments

The experiments performed so far have already allowed to rule out some of the most widely used spallation physics models and understand where they fail (too large excitation energy for the Bertini INC, wrong neutrons/charged particle competition in the

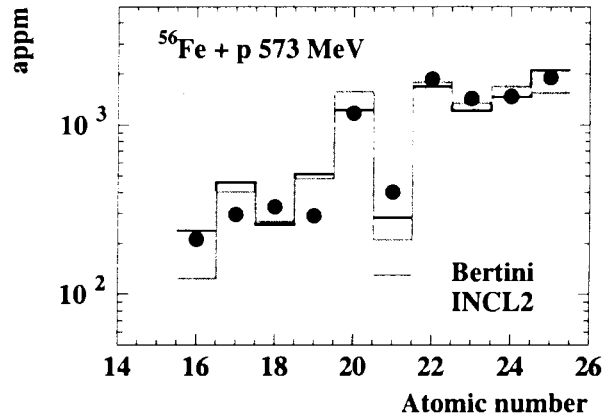


Figure 3: Production of spallation elements in an iron window after one year of irradiation by a $77\mu A/cm^2$, 573 MeV proton beam, deduced from data of ref [23] (black points) and calculated using TIERCE Code System with the Bertini (solid histogram) and the Cugnon (dashed histogram) INC. From ref. [24]

Dresner evaporation ...). Other models have been found to give promising results but are still not providing a general agreement with all the available data. To go further in the understanding of the reaction mechanism and then build better models, more constraining experimental data are needed.

A second generation of experiments using coincidence measurements of several observables is necessary. In particular, it is essential to be able to disentangle the two distinguished spallation stages (INC and de-excitation) which are most often mixed in the experimental observables. It is thus a challenge, both experimental and theoretical, to find observables of spallation reactions very sensitive to some aspects of the models and much less to others.

Coincident measurements of evaporation neutrons with light charged particles or fission fragments have already been performed by the NESSI collaboration [25, 14]. New projects to measure simultaneously identified residual isotopes together with neutrons and light charged particles are under study.

5 Conclusions

High energy data are important in a large variety of domains ranging from fundamental astrophysics to spallation sources for different applications. An important effort has been done recently for the collection of experimental spallation data, especially regarding neutron production in both thin and thick targets and residues. The reverse kinematics technique has allowed a break through in residue measurement. Since physics nechanisms evolve only slowly with the incident energy and mass of the target, it is not necessary to have experimental data for all nuclei at all energies. However, it is important to get complete sets of data (i.e. for all the possible decay channels) for a small number of nuclei chosen to be representative of the different part of the

periodic table of nuclei at some energies. This is the purpose of the HINDAS project funded by the EC. While neutron data seem to be sufficient, except maybe at energies around 300-400 MeV, still light charged particles data are missing and more residue measurement, for instance in the A=100 mass region, would be of high interest. Most of the recent experiments were done at energies above 800 MeV where very few data were existing but actually, not so many high quality data are available in the 200-600 MeV region. Should an ADS demonstrator be built which would use a proton beam in this energy range, more data would be needed.

Presently available high energy transport codes are able to describe gross features of a spallation driven system: for instance, the total number of neutrons produced in a spallation target per incident proton within 20% or the leakage of high energy neutrons (which is important for shielding estimation) within a factor 2. This is probably sufficient for a preliminary conceptual stage, but definitely not when a detailed engineering design will have to be achieved. A much higher precision will then be very likely required on certain quantities. Furthermore, even now, some quantities like the production of residues, which is of the highest importance for radiotoxicity, activity and chemical corrosion problems, are mispredicted with sometimes order of magnitude discrepancies. In particular, it was shown that the nuclear models most often used (i.e. the Bertini-Dresner combination) seem to have severe problems. New models, which are still under development, are promising but none of them is able to reproduce the whole set of available data. This means that further work has to be done, in particular to improve the models and validate them on the whole bulk of existing data. Since sometimes, it is difficult to disentangle the effects of the different ingredients of a code, more constraining experimental data should be collected. This can be achieved with a new generation of coincidence measurements of several types of particles simultaneously. Finally, an assessment of the impact of the new experimental data and improved models shoul be performed.

References

[1] H.W.Bertini et al., Phys. Rev. 131 (1963) 1801.

[2] Y.Yariv and Z.Fraenkel, Phys. Rev. C20 (1979) 2227: Phys. Rev. C24 (1981) 488.

[3] J.Cugnon, Nucl. Phys. A462 (1987) 751: J.Cugnon, C.Volant and S.Vuillier, Nucl. Phys. A620 (1997) 475.

[4] L.W.Dresner, Oak Ridge Report ORNL-TM-196 (1962).

[5] F.Atchison, Intermediate Energy Nuclear Data: Models and Codes, Proc. of a Specialists' Meeting, OECD/NEA, Issy-le-Moulineaux, France, May 30 - June 1 (1994) 199.

[6] T.W.Armstrong and K.C.Chandler, *HETC Monte-Carlo Nucleon-Meson Transport Code*, Report CCC-178, ORNL(1977) and Nucl. Sci. Eng. 49 (1972) 110.

[7] S.Leray, proceedings of the Workshop on Nuclear Reaction Data and Nuclear reactors: Physics, Design and Safety, ICTP Trieste, Italy, 13 March - 14 April 2000.

[8] GEDEON, *GEstion des DÉchets par des Options Nouvelles*, joint CNRS-CEA-EDF research program, France.

[9] HINDAS, *High and Intermediate energy Nuclear Data for Accelerator-driven Systems*, FIKW-CT-2000-00031 European programme.

[10] X.Ledoux et al., Phys. Rev. Lett. 82 (1999) 4412.

[11] S.Ménard, PhD Thesis, Orsay, January 1998; P.Casoli, Stage DEA, CEA/DAPNIA/SPhN, June 1999.

[12] O.Bersillon, 2nd Int. Conf. on Accelerator Driven Transmutation Technologies, Kalmar, Sweden, 3-7 Juin (1996).

[13] D.Hilscher et al., Nucl. Instr. and Meth. A414 (100) 1998.

[14] M. Enke et al., Nucl. Phys. A657 (1999) 317.

[15] R.J. Charity et al., Nucl. Phys. A483 (1988) 391.

[16] R.E.Prael and H.Liechtenstein, Report LA-UR-89-3014 Los Alamos National Laboratory (1989).

[17] P. Cloth et al., Report Jülich 2203 (1998).

[18] R.Michel et al., Nucl. Instr. and Meth. B113 (1996) 429 and 439.

[19] R.Michel et al., accepted for publication in Nucl. Instr. and Meth. (2000).

[20] J. Benlliure et al., Nucl. Phys. A683 (2001) 513; F. Rejmund et al., Nucl. Phys. A683 (2001) 540.

[21] W. Wlazlo et al., Phys. Rev. Lett. 84 (2000) 5736; T. Enqvist et al., Nucl. Phys. A686 (2001) 481.

[22] A. R. Junghans et al., Nucl. Phys. A 629 (1998) 635.

[23] W.R. Webber et al., Ap. J. 508 (1998) 940.

[24] A.Boudard et al., 3rd Int. Conf. on Accelerator Driven Transmutation Technologies and Applications, Praha, Czech Republic, June 7-11 1999.

[25] X.Ledoux et al., Phys. Rev. C57 (1998) 2375.

Nuclear Data Section
International Atomic Energy Agency
P.O. Box 100
A-1400 Vienna
Austria

e-mail: services@iaeand.iaea.org
fax: (43-1) 26007
cable: INATOM VIENNA
telex: 1-12645
telephone: (43-1) 2600-21710

Online: TELNET or FTP: iaeand.iaea.org
username: IAEANDS for interactive Nuclear Data Information System
usernames: ANONYMOUS for FTP file transfer;
FENDL2 for FTP file transfer of FENDL-2.0;
RIPL for FTP file transfer of RIPL.
NDSONL for FTP access to files sent to NDIS "open" area.
Web: http://www-nds.iaea.org