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PROGRESS REPORT ON NUCLEAR DATA RESEARCH IN THE FEDERAL REPUBLIC OF GERMANY

for the Period April 1, 2002 to March 31, 2003

July 2003

Edited by
S. M. Qaim
Forschungszentrum Jülich GmbH
Institut für Nuklearchemie
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FOREWORD

As in previous years, this report has been prepared to promote exchange of nuclear data research information between the Federal Republic of Germany and other member states of OECD/NEA and IAEA. It covers progress reports from the research centres at Karlsruhe and Jülich, the universities of Dresden, Hannover and Köln, as well as from the PTB Braunschweig. Each contribution is presented under the laboratory heading from where the work is reported. The names of other participating laboratories are also mentioned. The emphasis in the work reported here is on nuclear data for applied science programmes, such as those relevant to reactor technology and safety, transmutation concepts, accelerator shielding and development, astrophysics research, cosmogenic and meteoritic investigations, production of medically important radioisotopes, etc.

The coordination of nuclear data activities at the international level is done by two committees: the NEA-Nuclear Science Committee (NEA-NSC) and the IAEA-International Nuclear Data Committee (INDC). The present Editor has the privilege and the responsibility of representing Germany in both the committees. This report therefore also serves as a background information to some areas of work of those committees.

Jülich, July 2003 S.M. Qaim

This document contains information of a preliminary nature. Its contents should be used with discretion.

CONTENTS

FORSCHUNGSZENTRUM KARLSRUHE
INSTITUT FÜR KERNPHYSIK

1. Elastic α – ^{12}C Scattering and the ^{12}C(α,γ)^{16}O E2 S-factor*

P. Tischhauser[1], R.E. Azuma[2], L. Buchmann[3], R. Detwiler[1], U. Giesen[1,3], J. Görres[1], M. Heil, J. Hinnefeld[4], F. Käppeler, J.J. Kolata[1], H. Schatz[1,5], A. Shotter[6], E. Stech[1], S. Vouzoukas[1], M. Wiescher[1]

Angular distributions of ^{12}C(α,α)^{12}C have been measured for E_α = 2.6 - 8.2 MeV, at 32 angular positions from 24° to 166°. R-matrix analysis of the elastic scattering data below the proton threshold using an interaction radius of a = 5.5 fm yields a reduced width amplitude of $\gamma_{12}=0.30^{+0.03}_{-0.06}$ MeV$^{1/2}$ for the E_x = 6.917 MeV (2^+) subthreshold state in ^{16}O. Using this amplitude and incorporating radiative capture and β-delayed α-decay data, the E2 S-factor is calculated at E_{cm}=300 keV to be $S_{E2}=37^{+8}_{-18}$ keV b.

* Phys. Rev. Lett. 87 (2001) 251102 (4)

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[2] University of Toronto, Toronto, Canada
[3] TRIUMF, Vancouver, Canada
[4] Indiana University at South Bend, South Bend, IN, USA
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[6] University of Edinburgh, Edinburgh, UK

2. Alpha and Neutron Induced Reactions on Ruthenium Isotopes*

W. Rapp, M. Heil, D. Hentschel[2], F. Käppeler, R. Reifarth[1], H.J. Brede[2], H. Klein[2], T. Rauscher[3]

The uncertain origin of the proton-rich Mo and Ru isotopes has motivated cross section measurements of α- and neutron-induced reactions. The experiments were performed via the activation technique by irradiating thin layers of natural ruthenium with α-particle beams close to the Gamov window of the p process between 7.0 and 10.5 MeV. The cross sections of the reactions ^{96}Ru(α,γ), ^{96}Ru(α,n), ^{96}Ru(α,p) and ^{98}Ru(α,n) could be determined with uncertainties of typically 10%. On average, these results are about two to three times smaller than recent statistical model predictions. Additional activations in a quasi-stellar neutron spectrum corresponding to kT=25 keV allowed to obtain the complementary stellar (n,γ)-cross sections for ^{96}Ru, ^{102}Ru and ^{104}Ru. In these cases the agreement with model calculations is considerably better.

* Phys. Rev. C 66 (2002) 15803 (11)

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[2] Physikalisch-Technische Bundesanstalt, D-38116 Braunschweig, Germany
[3] Departement für Physik und Astronomie, Universität Basel, CH-4056 Basel, Switzerland

3. Neutron Capture Cross Sections for Stellar Cd Production *

K. Wisshak, F. Voss, F. Käppeler, L. Kazakov[1]

The neutron capture cross sections of ^{110}Cd, ^{111}Cd, ^{112}Cd, ^{113}Cd, ^{114}Cd and ^{116}Cd have been measured in the energy range from 3 to 225 keV at the Karlsruhe 3.75 MV Van de Graaff accelerator. Neutrons were produced via the ^{7}Li(p,n)^{7}Be reaction by bombarding metallic Li targets with a pulsed proton beam. The Karlsruhe 4π Barium Fluoride Detector was used for registration of capture events. The cross sections were determined relative to the gold standard using highly enriched metallic Cd samples. The respective ratios could be obtained with overall uncertainties between 0.8 and 1.6%, about an order of magnitude more accurate than previous data. Maxwellian averaged neutron capture cross sections were calculated for thermal energies between kT = 8 keV and 100 keV. Discrepancies of 30% to 40% were found with respect to previous data. The new cross sections provide a reliable definition of the s-abundance pattern of the Cd isotopes, thus improving the corresponding r-process residuals and, most importantly, the impact of s-only ^{110}Cd as a normalization point for the overall distribution of the s abundances.

* Phys. Rev. C 66 (2002) 15803 (11)
[1] IPPE, Obninsk, Kaluga Region, Russia

4. Neutron Activation Measurements on Natural Tellurium *

R. Reifarth, F. Käppeler

Natural tellurium has been activated with thermal neutrons (kT=22 meV) and with quasi-stellar neutrons (kT=25 keV). By analyzing the ground state decay of the odd isotopes as a function of time, information about the partial capture cross sections of the even isotopes to the isomeric states of the odd isotopes could be determined with small systematic uncertainties. Additionally, information about the decay of some isomers could be determined with better accuracy.

* Phys. Rev. C 66 (2002) 54605 (9)

5. The Stellar Neutron Capture Cross Sections of $^{128, 129, 130}$Xe *

R. Reifarth, M. Heil, F. Käppeler, F. Voss, K. Wisshak, F. Becvar[1], M. Krticka[1], R. Gallino[2]

The (n,γ) cross sections of the important s-process nuclei ^{128}Xe, ^{129}Xe and ^{130}Xe have been measured in the astrophysically relevant neutron energy range from 3 to 225 keV. Neutrons were produced via the ^{7}Li(p,n)^{7}Be reaction by bombarding metallic Li targets with the pulsed proton beam of the Karlsruhe 3.75 MV Van de Graaff accelerator. High pressure gas samples of isotopically enriched xenon were used in the experiment, and capture events were registered with the Karlsruhe 4π Barium Fluoride Detector. The cross sections were determined relative to the gold standard with overall uncertainties of 1.5 - 2.5% over most of the investigated energy range. From these results Maxwellian averaged stellar (n,γ) cross sections with typical uncertainties of 2% were calculated for thermal energies between kT=8 keV and 100 keV. In contrast to previous theoretical estimates, which were known to exhibit uncertainties of 30 to 50%, this work provides a reliable basis for quantitative astrophysical analyses and makes it possible to define the solar Xe abundance to 5.39±0.22 relative to 10^{6} Si atoms.

* Phys. Rev. C 66 (2002) 64603 (14)
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[4] Max-Planck Institut für Astronomie, Königstuhl 17, D-69117 Heidelberg, Germany

6. The Stellar Neutron Capture Rate of ^{147}Pm for Constraining the *s*-Process Neutron Density *

R. Reifarth, C. Arlandini, M. Heil, F. Käppeler, P.V. Sedyshev[1], A. Mengoni[2], M. Herman[3], T. Rauscher[4], R. Gallino[5], C. Travaglio[6]

The unstable isotope ^{147}Pm represents an important branch point in the s-process reaction path. This paper reports on the successful determination of the stellar (n,γ) cross section via the activation technique. The experiment was difficult because the relatively short ^{147}Pm half-life of 2.62 yr enforced the sample mass to be restricted to 28 ng or 10^{14} atoms only. By means of a modular, high-efficiency Ge-Clover array the low induced activity could be identified in spite of considerable background from various impurities. Both partial cross sections feeding the 5.37 d ground state and the 41.3 d isomer in ^{148}Pm were determined independently, yielding a total (n,γ) cross section of 709±100 mb at a thermal energy of kT=30 keV. The (n,γ) cross sections of the additional branch point isotopes ^{147}Nd and ^{148}Pm as well as the effect of thermally excited states were obtained by detailed statistical model calculations. The present results allowed considerably refined analyses of the s-process branchings at A=147/148 which are probing the neutron density in the He-burning shell of low-mass AGB stars.

* Ap. J. 582 (2003) 1251 - 1262
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[6] Max-Planck-Institut für Astrophysik, D-85748 Garching, Germany, and Istituto Nazionale di Astrofisica (INAF), Osservatorio Astronomico di Torino, I-10025 Pino Torinese (To), Italy

7. Neutron Capture Measurements on ^{171}Tm *

R. Reifarth[1], R.C. Haight[1], M. Heil, M.M. Fowler[1], F. Käppeler, G.G. Miller[1], R.S. Rundberg[1], J.L. Ullmann[1], J.B. Wilhelmy[1]

About 50 % of the heavy elements are produced during the slow neutron capture (s-) process. Of critical importance for understanding the dynamics of the s-process are the reactions on branch point nuclei. These are unstable isotopes that have half lives comparable to the neutron capture times during the s-process. While neutron capture cross sections have been measured for most stable species, very little information has been obtained on unstable species in the astrophysically interesting keV region.
Two experiments have been carried out. The first one using liquid scintillator detectors (C_6D_6) at the Los Alamos Neutron Science Center (LANSCE). A 2.5 mg ^{171}Tm target was made by irradiating ^{170}Er in a reactor at the Idaho National Engineering and Environmental Laboratory,

followed by chemical separation and processing in a hot cell at LANL. The ^{171}Tm was separated from the erbium target using high-performance liquid chromatography about 2 weeks before the experiment. The target was made by electroplating the ^{171}Tm on beryllium foil. Data were taken using two C_6D_6 detectors, each 12.5 cm in diameter and 7.5 cm in thickness, 8 m away from the neutron production target. Above 1 keV, the results show a significant deviation from a calculation with the statistical model code GNASH (Young, Arthur, and Chadwick, LANL Technical Report LA-12343-MS, 1992), indicating a not fully understood background contribution.
This discrepancy motivated a second experiment using the activation technique, carried out at the Institut für Kernphysik of the Forschungszentrum Karlsruhe. A 0.8 μg ^{171}Tm-sample, produced like the 2.5 mg sample, was irradiated for 4 d with quasistellar neutrons of kT = 25 keV. The integrated neutron flux was measured by simultaneous activation of Au foils before and behind the sample. After the irradiation the produced ^{172}Tm decays with a half-life of 63.6 d via β-decay to ^{172}Yb. The induced ^{172}Tm activity was measured for 5 d by detecting the emitted 1094 keV γ-rays with a 4π geometry of two face two face Ge-Clover detectors. The determined cross section for kT = 25 keV was 350 ± 30 mb which is in very good agreement with the GNASH prediction of 335 mb.
A new attempt resolving the systematic uncertainties of the previous experiments is the Device for Advanced Neutron Capture Experiments (DANCE) at LANL.

* Nucl. Phys. A718 (2003) 478c
[1] Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

8. The ^{208}Pb(n,γ) Cross Section *

H. Beer, W. Rochow[1], F. Käppeler, T. Rauscher[2]

At the Karlsruhe and Tübingen 3.75 MV Van de Graaff accelerators the ^{208}Pb(n,γ)^{209}Pb (3.25 h) cross section was measured by the activation technique via the β-spectrum of the ^{209}Pb-decay using a 4πβ Si(Li) spectrometer. Natural lead samples were irradiated back to back with gold foils which served as capture standard. Neutrons were generated using the ^{7}Li(p,n) and T(p,n) reactions. Maxwellian averaged cross sections were measured at thermal energies of kT=25 keV and 52 keV. In addition, average capture cross sections were obtained with neutron spectra of mean neutron energies 30, 104, 149, and 215 keV, respectively. The 30 keV cross section value represents a measurement of the important direct capture part of the cross section, because the first two resonances at 43.29 and 47.26 keV were clearly outside our 30 keV spectrum. The data are discussed with respect to resonance and direct capture. The stellar reaction rate is determined between 1 and 100 keV and nucleosynthesis at the termination of the s process is studied.

* Nucl. Phys. A718 (2003) 518c
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[2] Departement für Physik und Astronomie, Universität Basel, CH-4056 Basel, Switzerland

1. Validation Analyses for the simulation of D-Li Source Neutrons in Monte Carlo Transport Calculations

U. Fischer, S. P. Simakov

The Monte Carlo code McDeLicious was recently developed to simulate in transport calculations for the International Fusion Material Irradiation Facility (IFMIF) the generation of D-Li source neutrons on the basis of evaluated double-differential d + ^{6,7}Li cross-section data [1]. A complete set of evaluated cross-section data had been prepared to this end, including cross-sections for all reaction channels up to 50 MeV deuteron energy as well as energy-angle distributions for the neutrons emitted through the various ^{6,7}Li(d,xn)-reactions [2]. First tests against the then available experimental thick lithium target neutron yield data at 32 and 40 MeV deuteron energy showed that McDeLicious can predict both the neutron yield data and the angular energy spectra with considerably better accuracy than previous semi-empirical approaches.

New experimental thick lithium target data became available over the reporting period in the frame of the IFMIF project. Bem et al. [3] have measured the neutron yield spectra at 17 MeV incident deuteron energy using the U-120M cyclotron of the Nuclear Physics Institute at Rez. A liquid scintillator detector was used to record the pulse height spectra at five detector angles (0, 30, 60, 70 and 120°). Baba et al. [4] have measured the neutron yield spectra at 25 and 40 MeV deuteron incidence energy using the AVF cyclotron of the Tohoku University Cyclotron and Radioisotope Centre. The time-of-flight (TOF) technique was applied to obtain the neutron spectra at various angles between 0 and 110°.

A large number of experimental data is now available for the forward neutron yields. Fig. 1 shows a comparison of the measured and calculated yields as a function of the incident deuteron energy. Note that only the McDeLicious calculations can satisfactorily reproduce the experimental neutron yields over the whole deuteron energy range up to 40 MeV.

The recent Baba et al. neutron yield spectral data for 25 MeV and 40 MeV deuteron energy are compared in Figs. 2 and 3 with McDeLicious and MCNPX calculations. Again there is very good agreement between the McDeLicious calculations and the experimental neutron spectra, while MCNPX calculations tend to underestimate the neutron spectra at forward angles. This will result in a significant underestimation of the total neutron yield by MCNPX since neutrons are preferably emitted in forward direction. It is indicated by the Baba data, however, that McDeLicious overestimates the production of neutrons with energies less than 2 MeV. This is due to the fact that the experimental data of Lone et al. [5] have been used in evaluating the d + ^{6,7}Li cross-sections. Those data show a rather large neutron production in the low energy range below 2 MeV which is in contradiction to other measurements including the recent TOF data of Baba et al. It is thus concluded that the d + ^{6,7}Li data evaluation has to be updated for the compound nuclear reactions which populate the low energy region. Account must be taken of the recent measurements by Baba et al. for both thick and thin lithium targets.

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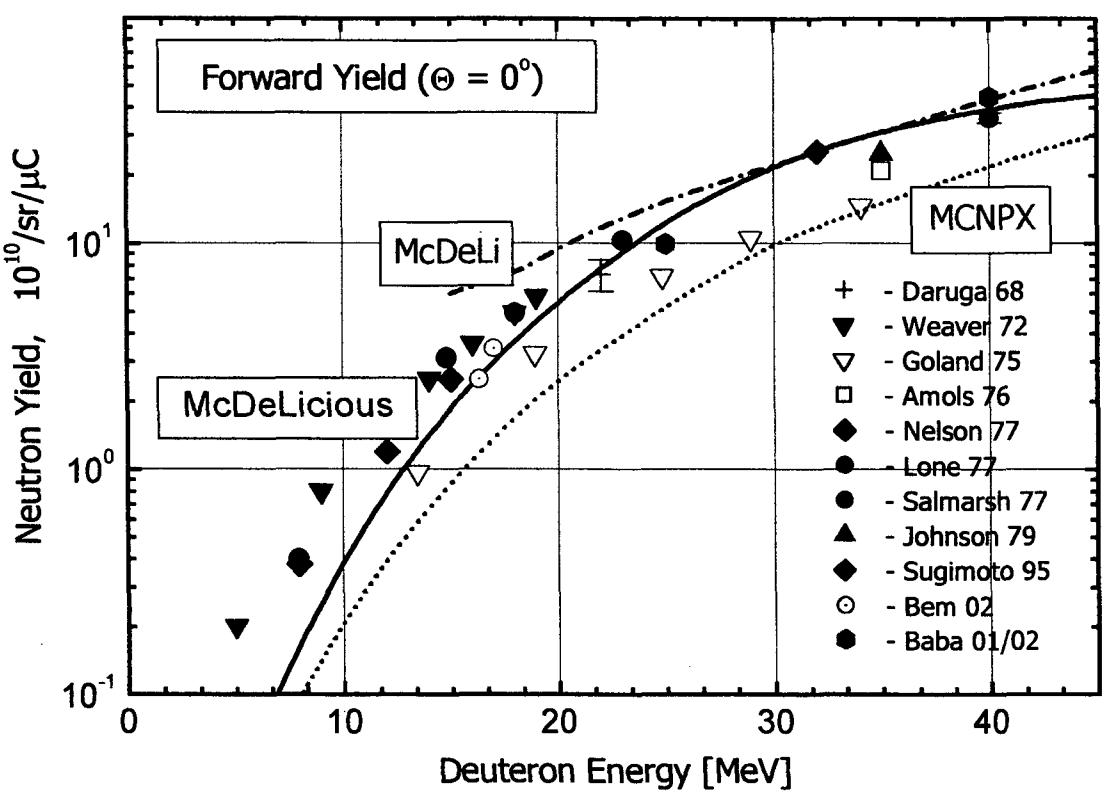

Fig. 1 Comparison of calculated and measured thick lithium target forward neutron yield data.

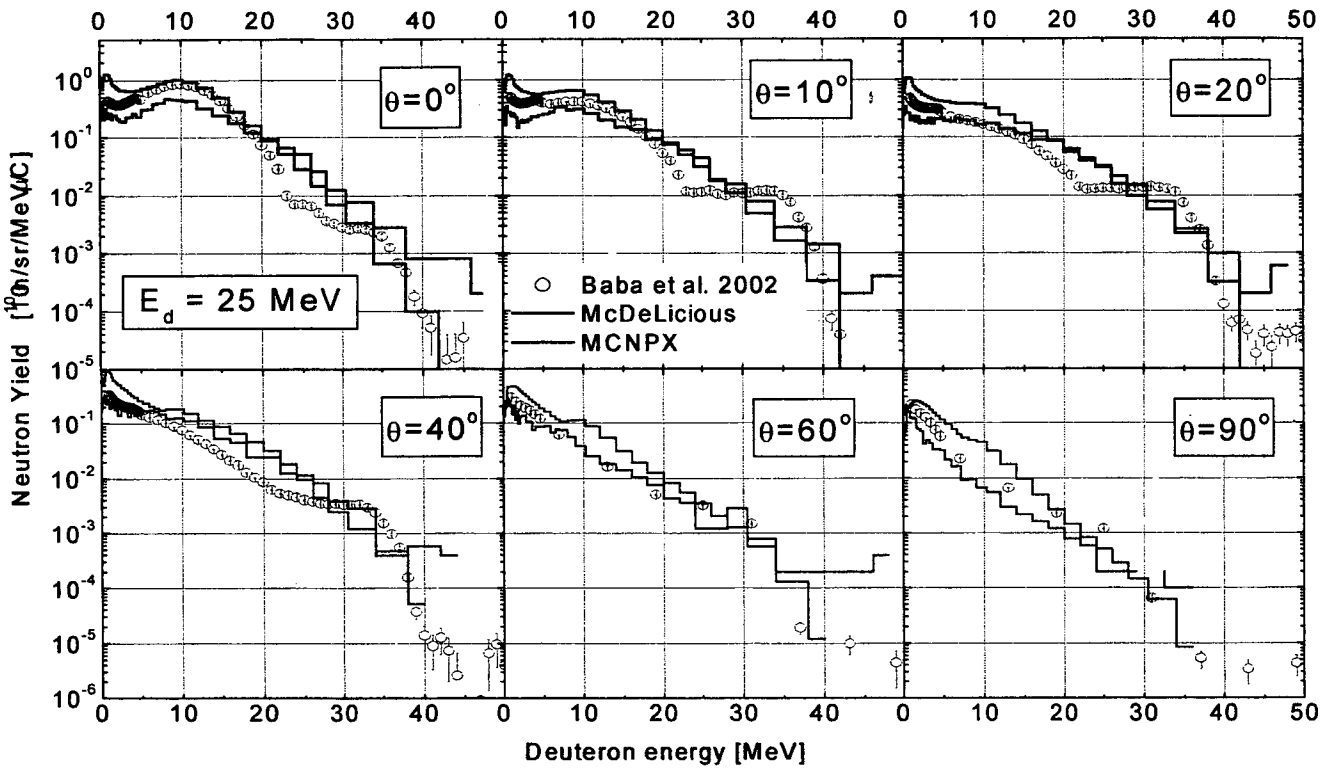

Fig. 2 Measured [4] and calculated thick target neutron yield angular spectra at an incident deuteron energy of 25 MeV.

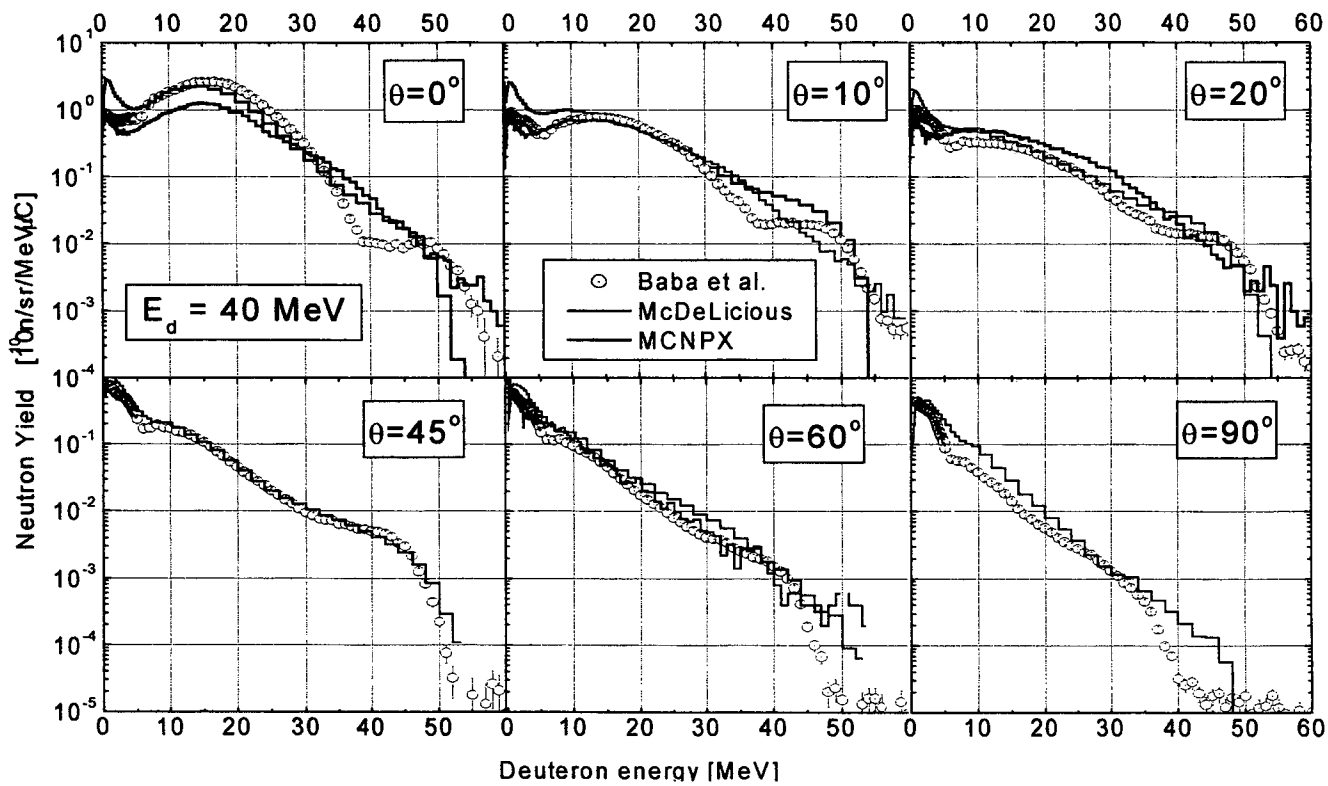

Fig. 3 Measured [4] and calculated thick target neutron yield angular spectra at an incident deuteron energy of 40 MeV.

2. Validation Analyses of IEAF-2001 Activation Cross-Section Data

U. Fischer, S. P. Simakov, U. v. Möllendorff, P. P. H. Wilson[1]

The Intermediate Energy Activation File IEAF-2001 [1] has been recently developed to enable activation and transmutation calculations for accelerator based neutron source facilities with neutron spectra extending to energies above 20 MeV. The IEAF-2001 data library contains neutron-induced activation and transmutation cross sections up to 150 MeV neutron energy for 679 target nuclides from hydrogen to polonium (Z = 84). Validation of the IEAF-2001 data library is required to ensure reliable results of the activation and transmutation calculations.

Over the reporting period, detailed IEAF-2001 validation analyses were started [2] on the basis of activation calculations with the ALARA code [3]. The validation procedure comprised several steps including various benchmark calculations and computational analyses of integral activation experiments. In the first step, ALARA activation calculations for the low activation steel Eurofer were benchmarked against FISPACT [4] calculations. Since FISPACT is not capable of using IEAF-2001 cross-section data, the benchmark was performed for a fusion reactor spectrum extending up to 20 MeV neutron energy, thus enabling FISPACT the use of the European Activation File EAF-99 [5] These data have been also adopted in the IEAF-2001 library for the energy range below 20 MeV. Satisfactory agreement was found for the calculated activity inventories, the decay heat production and the contact dose rate, thus showing consistency between ALARA/IEAF-2001 and FISPACT/EAF-99 activation calculations for the energy range below 20 MeV.

The second step was devoted to the testing of the IEAF-2001 data against results of an integral activation experiment performed previously at FZK [6]. Samples of SS-316 steel, F82H steel, pure vanadium, and V-4Ti-4Cr alloy were activated in a white neutron field produced by a 40-MeV deuteron beam on a thick lithium target. Results of the activation analyses for vanadium and vanadium alloy are shown in Fig. 1 in terms of C/E (calculation/experiment) ratios. Satisfactory agreement between the ALARA/IEAF-2001 activity results and the experiment was found for all product nuclides, except ^{47}Sc and ^{51}Cr. It was shown that the ^{51}V(n,nα) reaction cross-section needs to be updated to resolve the ^{47}Sc discrepancy. Sequential charged particle reactions (SCPR) such as ^{51}V(n,xp) $\Rightarrow$ ^{51}V(p,n)^{51}Cr play an important role in generating ^{51}Cr from ^{51}V. The ALARA code is currently being extended to include in the activation calculation the SCPR feature.

[1] University of Wisconsin-Madison, Fusion Technology Institute, WI 53706, USA

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[1] U. Fischer, D. Leichtle, U. v. Möllendorff , I. Schmuck, A. Yu. Konobeyev, Yu. A. Korovin, P.E. Pereslavtsev, Intermediate Energy Activation File IEAF-2001, NEA Data Bank Programme Library, package NEA-1656/01 (2001), RSICC Oak Ridge, DLC-217/IEAF-2001 (2002)

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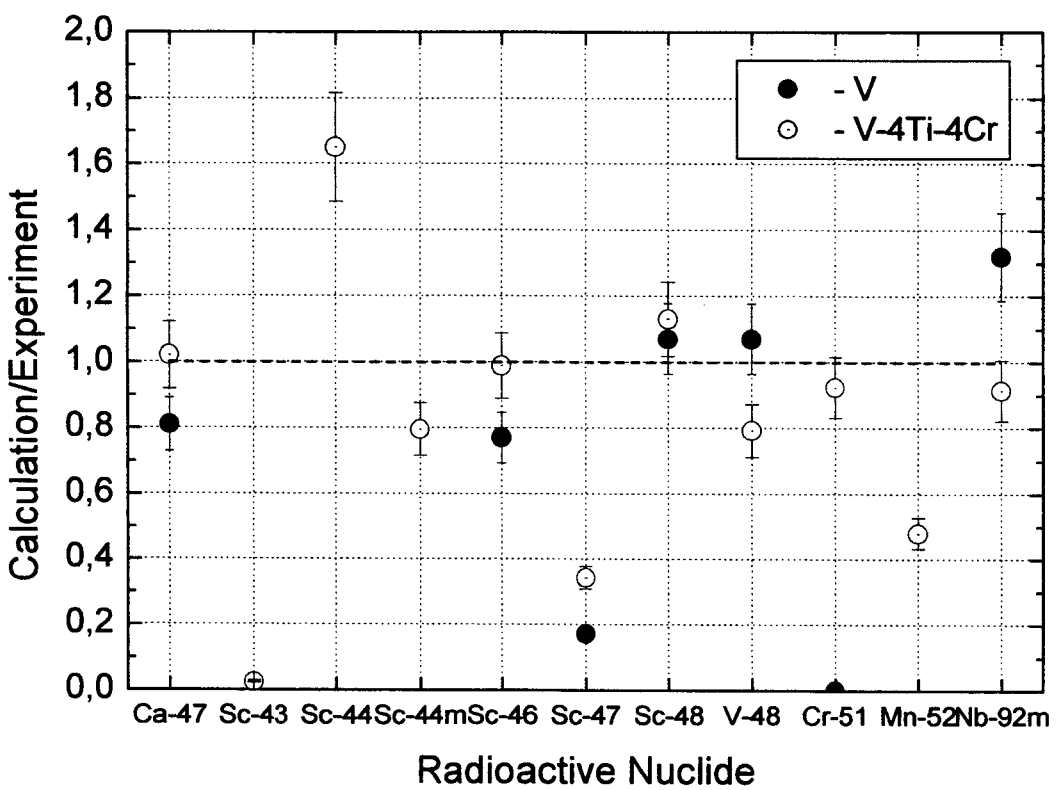

Fig.1 Comparison of calculated (ALARA/IEAF-2001) and measured [6] radioactivities induced by irradiating V and V-4Ti-4Cr alloy in a white D-Li neutron source spectrum.

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1. Neutron Activation Cross Sections

S. M. Qaim, I. Spahn, S. Sudár*, C. Nesaraja†

The cross section data of a few recently investigated nuclear reactions on isotopes of Zn, Ga and Ge in the neutron energy range of 5 to 12 MeV were critically analysed. The emphasis was on the formation of the isomeric pair ^{69m,g}Zn in the ^{70}Zn(n,2n)-, ^{69}Ga(n,p)- and ^{72}Ge(n,α)-reactions and the pair ^{71m,g}Zn in the ^{71}Ga(n,p)- and ^{74}Ge(n,α)-reactions. All the measured excitation functions and isomeric cross section ratios could be described by the statistical model code STAPRE, though within the extreme limits of experimental errors.

The measured isomeric cross section ratios for the formation of ^{69m,g}Zn in the three above mentioned reactions, viz. (n,2n), (n,p) and (n,α), are shown in Fig. 1. The ratio increases as a function of neutron energy, the increase being most pronounced in the (n,α) reaction. Thus, this reaction channel appears to have a strong effect on the isomer distribution. The results for the isomeric pair ^{71m,g}Zn are, however, somewhat different. In that case the (n,p) reaction channel has a strong effect.

A relatively new line of experimental activity deals with radiochemical measurements of (n,p) reaction cross sections averaged over a 14 MeV d(Be) break-up neutron spectrum. The spectrum characterization was done by irradiating a set of foils in which several nuclear reactions having different thresholds were induced. Using the different activities, the neutron spectrum was unfolded by means of the code SULSA. The emphasis in these integral cross section measurements lies on the reactions which lead to therapeutic radionuclides. During the period of this Progress Report three reactions, namely ^{64}Zn(n,p)^{64}Cu, ^{67}Zn(n,p)^{67}Cu and ^{89}Y(n,p)^{89}Sr were investigated. In each case a radiochemical separation of the product was

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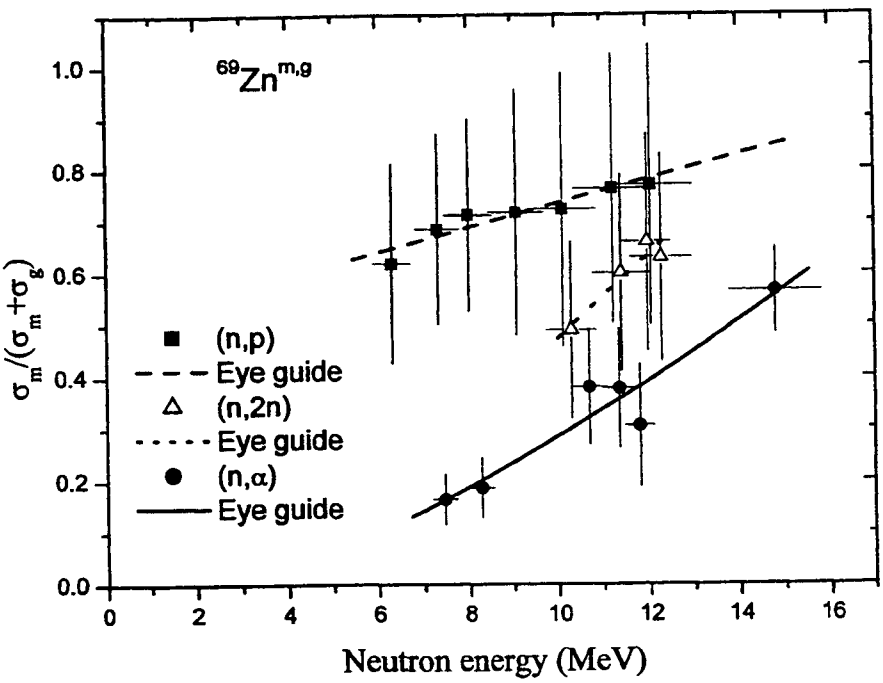

Fig. 1 Experimental isomeric cross section ratios for the formation of $^{69m,g}Zn$ in (n,α), (n,p) and (n,2n) reactions, shown as a function of incident neutron energy.

carried out and the radioactivity was determined via low-level β^- counting or high-resolution γ-ray spectrometry. The cross sections obtained are given in Table 1. For comparison, fission neutron spectrum averaged cross sections are also given. It is concluded that the three therapeutic radionuclides under consideration can be produced in higher yields using a fast neutron spectrum than in a fission reactor, provided a high-intensity fast neutron source is available.

Table 1 Comparison of spectrum averaged (n,p) reaction cross sections for a fission spectrum and a 14 MeV d(Be) neutron field

Nuclear reaction	$\langle\sigma\rangle$ d(Be) neutrons (mb)	$\langle\sigma\rangle$ fission neutron spectrum (mb)
$^{64}Zn(n,p)^{64}Cu$	132 ± 13	31 ± 2.3
$^{67}Zn(n,p)^{64}Cu$	5.1 ± 0.5	1.1 ± 0.04
$^{89}Y(n,p)^{89}Sr$	0.9 ± 0.14	0.3 ± 0.06

2. Charged Particle Induced Reaction Cross Sections

K. Hilgers, K. Kettern, H.E.Hassan*, Ye. Skakun†, S. Spellerberg, B. Scholten, D. Steyn‡, N. van der Walt‡, S.M. Qaim, H. H. Coenen .

As in previous years, systematic studies on charged particle induced reaction cross sections for medical applications were continued. During the period of the present Progress Report following investigations were carried out.

a) Excitation functions of nuclear reactions for production of longer-lived positron emitters (^{76}Br and ^{124}I)

The positron emitting radionuclide **^{76}Br** ($T_{1/2}$ = 16.0 h) and the γ-emitting radionuclide ^{77}Br ($T_{1/2}$ = 57.0 h) are of considerable medical interest. Using natural and highly enriched samples excitation functions of the following reactions were measured from their thresholds up to about 40 MeV: ^{76}Se(p,xn)^{75,76}Br, ^{77}Se(p,xn)^{76,77}Br and ^{nat}Se(p,xn)^{76,77,82}Br. Special emphasis was on the energy region below 15 MeV where the existing data base is rather weak. Analysis of the experimental data and nuclear model calculations to interpret the data are continuing.

Due to the increasing significance of the positron emitting radionuclide **^{124}I** ($T_{1/2}$ = 4.18 d) a search is under way for a high yield production process. Detailed investigations on the ^{126}Te(p,3n)^{124}I reaction, started last year, were continued. The yields of ^{124}I via the two intermediate energy reactions investigated, namely the ^{125}Te(p,2n)^{124}I and ^{126}Te(p,3n)^{124}I processes, appear to be comparable. The levels of the impurities are presently being analysed. They will play a crucial role in the choice of one of those two reactions for production purposes.

b) Excitation functions of nuclear reactions for production of therapy related radionuclides (^{64}Cu, ^{67}Cu, ^{103}Pd and ^{140}Nd)

The radionuclides **^{64}Cu** ($T_{1/2}$ = 12.7 h) and **^{67}Cu** ($T_{1/2}$ = 61.9 h) are of great interest in

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‡ Radionuclide Production, iThamba Laboratories, Somerset West, Cape Town, South Africa

systemic endoradiotherapy. The former is mostly produced via the $^{64}Ni(p,n)^{64}Cu$ reaction at a small-sized cyclotron. During the period of the present Progress Report two other processes, namely $^{66}Zn(d,\alpha)^{64}Cu$ and $^{68}Zn(p,\alpha n)^{64}Cu$, were investigated. In each case highly enriched target material (^{66}Zn or ^{68}Zn) was used. Since ^{64}Cu is almost a pure β^+ emitter, a clean radiochemical separation was invariably performed and a careful analysis of the decay curve of the annihilation radiation was carried out. The measured data for the $^{68}Zn(p,\alpha n)^{64}Cu$ process are shown in Fig. 2 as a function of proton energy. The integral yield of ^{64}Cu calculated from this excitation function is much lower than that obtained from the commonly used $^{64}Ni(p,n)^{64}Cu$ reaction. The only advantage of the $^{68}Zn(p,\alpha n)^{64}Cu$ reaction could be that ^{64}Cu is produced as a bye-product during the production of ^{67}Ga via the $^{68}Zn(p,2n)^{67}Ga$ process.

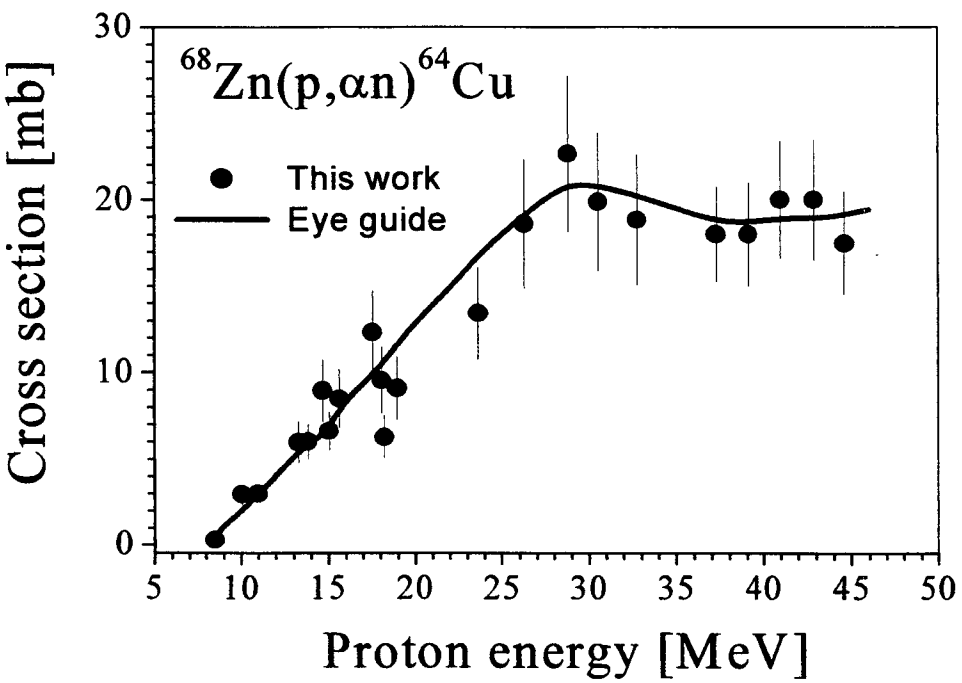

Fig. 2 Excitation function of the $^{68}Zn(p,\alpha n)^{64}Cu$ reaction.

The radionuclide ^{67}Cu is commonly produced via the $^{68}Zn(p,2p)^{67}Cu$ reaction at proton energies above 70 MeV. A low-energy reaction, viz. $^{70}Zn(p,\alpha)^{67}Cu$, has also been recently suggested but the yield is rather low. We now investigated the $^{64}Ni(\alpha,p)^{67}Cu$

reaction using a 78 % enriched target. The calculated yield of ^{67}Cu via this route is lower than via another low-energy reaction ^{70}Zn(p,α)^{67}Cu.

The radionuclide **^{103}Pd** ($T_{1/2}$ = 17.0 d) is of considerable significance in brachytherapy. It is produced routinely via the ^{103}Rh(p,n)^{103}Pd reaction. A detailed study on this reaction was reported last year. During the period of the present Progress Report two alternative routes, namely ^{102}Ru(^{3}He,2n)^{103}Pd and ^{100}Ru(α,n)^{103}Pd, were investigated thoroughly using highly enriched target isotopes. The measured excitation functions, and integral yields calculated therefrom, demonstrate the possibility of use of these two reactions for production of ^{103}Pd. The method of choice, however, remains the ^{103}Rh(p,n)^{103}Pd process since the yield is higher and a small-sized cyclotron is sufficient for production purposes.

The radionuclide **^{140}Nd** ($T_{1/2}$ = 3.4 d) is a potentially useful therapeutic isotope since it emits several Auger electrons per decay. It is generally produced via the ^{140}Ce(^{3}He,3n)^{140}Nd reaction. The reaction cross sections were, however, not known. Therefore, a detailed study was initiated last year and now it is reaching completion.

c) *Proton therapy related activation cross sections*

In proton therapy, the activation cross section data play only a secondary role. Nonetheless, two aspects need some consideration.

a) Formation of short-lived β^+ emitters in the human tissue.
b) Formation of long-lived activation products in collimator materials.

Rather extensive activation data exist in both the cases. Yet there are deficiencies and gaps for a few elements in several energy regions. During the period of this Progress Report measurements were performed on the formation of the β^+ emitters ^{11}C ($T_{1/2}$ = 20.3 min), ^{13}N ($T_{1/2}$ = 10.0 min) and ^{18}F ($T_{1/2}$ = 109.7 min) in interactions of protons of up to 200 MeV energy with the elements carbon, nitrogen and oxygen. The formation of ^{11}C from carbon and nitrogen is of special importance. The results for the ^{nat}N(p,x)^{11}C

process are given in Fig. 3. Evidently, several nuclear reactions on the two isotopes of nitrogen (^{14}N and ^{15}N) contribute to the formation of ^{11}C.

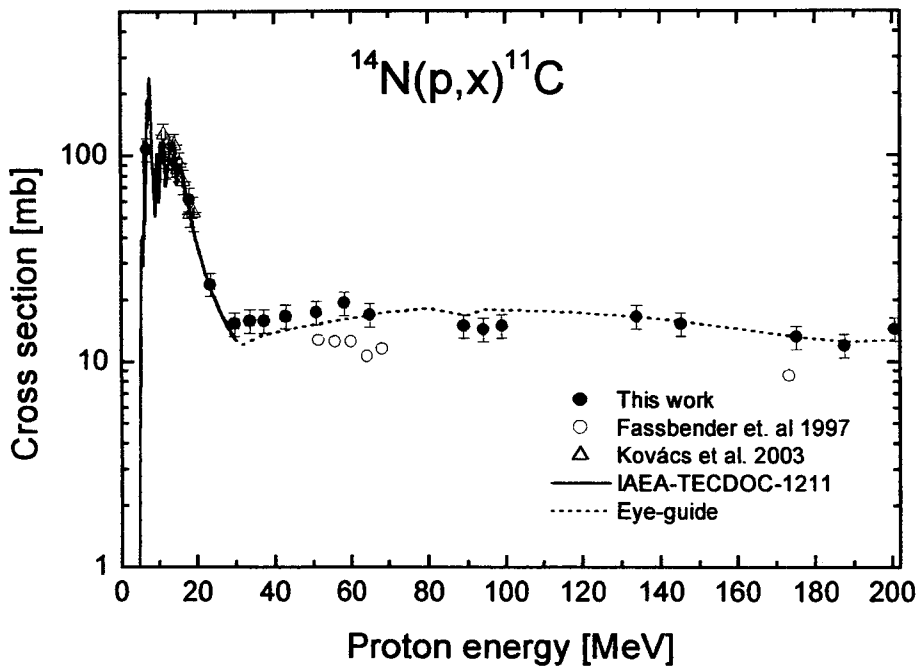

Fig. 3 Excitation function for the formation of ^{11}C in the interactions of protons with ^{nat}N.

As far as collimator activation is concerned, measurements were performed on Ti and Pb. The emphasis was on soft-radiation emitting longer-lived products like ^{45}Ca ($T_{1/2}$ = 163 d; I_{β^-} = 100 %; E_{β^-} = 258 keV), ^{49}V ($T_{1/2}$ = 330 d; EC = 100 %; 4.5 KeV X-rays) and ^{204}Tl ($T_{1/2}$ = 3.78 a; I_{β^-} = 97.4 %; E_{β^-} = 763 keV) which can only be studied radiochemically. In the case of ^{45}Ca and ^{204}Tl the radioactivity was determined via low-level β^- counting, and in the case of ^{49}V via high-resolution X-ray spectrometry. The analysis of data is in progress.

3. Other Activities

a) Chair of INDC

Syed M. Qaim continued to act as Chairman of the International Nuclear Data Committee (INDC) of the IAEA. The term of office will cover the period up to December 2005.

b) Participation in CRP

The Institute signed a Research Agreement with the IAEA to participate in the CRP on Therapeutic Radionuclides.

c) NRC-6

The Institute is organising the 6th International Conference on Nuclear and Radiochemistry (NRC-6) to be held in Aachen, Germany, from 29 August to 03 September 2004. Nuclear data activities will constitute an integral part of the programme of the conference.

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INSTITUT FÜR KERN- UND TEILCHENPHYSIK TECHNISCHE UNIVERSITÄT DRESDEN

1. Integral Activation Experiment with Tungsten*

R. Eichin, R. A. Forrest[1], H. Freiesleben, S. A. Goncharov[2], V. D. Kovalchuk[2], D. V. Markovskij[3], D. V. Maximov[2], K. Seidel, S. Unholzer

The radioactivity induced by neutrons in the materials of a fusion device represents a central topic of safety-related investigations. Radionuclides of a broad range of half-lives have to be included in the corresponding analyses. The short-term radioactivity is of interest with respect to heat production and shut-down dose rate whereas the long-term radioactivity determines the waste management. The radioactivity is mainly produced by the fusion neutrons of 14 MeV energy, where the number of open reaction channels is maximum, and at thermal neutron energy, where cross section values are large. In the present work, the radioactivity induced by 14 MeV neutrons in pure tungsten was investigated. Tungsten is the preferred material for the divertor plates of fusion devices and is a constituent of reduced activation structural materials.

In a calculation with the European Activation System [1], W was taken to be irradiated under reactor condition, i. e. with a flux density of 14 MeV neutrons corresponding to a power density of 1.0 MW/m^2, for a period of one year. The results obtained for the contact dose rate as a function of the decay time after irradiation, are shown in Fig. 1. After about 10 years the dose rate is expected to be below the recycling limit and after about 200 years below the hands-on limit. To investigate the activity of all nuclides, that dominate before the recycling limit is reached, two different irradiations were carried out. A short irradiation followed by activity measurements at decay times in the range labelled by "ts" in Fig.1 was directed to the short-term radioactivity; and after a longer irradiation with activity measurements at times "t1...t3", the production of the other nuclides was studied. Details of the irradiations performed at the high-intensity neutron generator SNEG-13 [2] at Sergiev Posad are described in Ref. [3].

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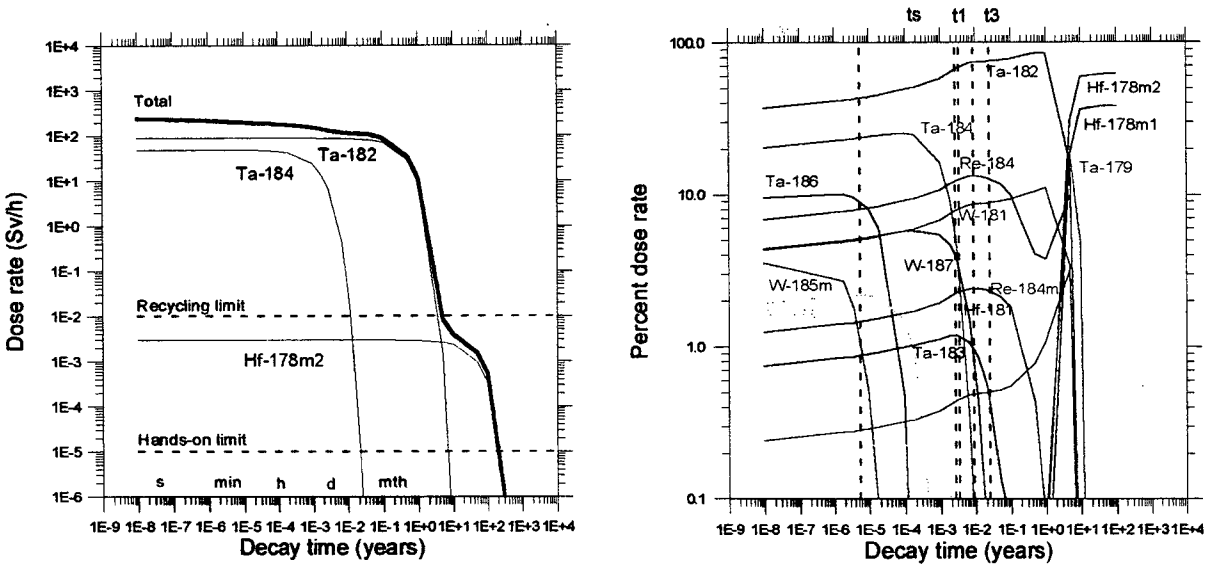

Fig. 1: Contact dose rate (left hand) and contribution of the different radionuclides to the total dose rate (right hand) after irradiation of W with 14 MeV neutrons of 1.0 MW/m^2 power density for one year as a function of decay time.

Gamma-ray spectra of the irradiated samples were measured with a Ge(Li)-spectrometer, and nuclide activities were derived with gamma-yield data from EASY. For each of the measured values the activity was calculated with EASY, and calculated-to-experimental values (C/E) were determined. The results are presented in Tables 1 and 2.

The C/E obtained for ^{187}W should not be used for validating the ^{186}W(n,γ) cross section at 14 MeV neutron energy, as the neutron field was contaminated by D-D neutrons [3].
The other activities investigated, typically show C/E of about 1.1 ... 1.8, resulting in an overestimation of the total dose rate of W under fusion reactor conditions by a factor of 1.4 for the first year after the operation. The reactions by which the activities were produced are of the type (n, charged particle). Their cross sections at 14 MeV are of the order of 1 mb due to the high Coulomb barrier of W, which means they are just above the reaction threshold. The representation of the cross section data in this energy region of the fusion neutron peak should be improved.

Table 1 Results from the short irradiation; radionuclides identified, their half-lives and the γ-rays used to determine the activities, the neutron reactions producing these radionuclides, the ratios of calculated-to-experimental activity, and the uncertainties of the calculated and of the experimental activities.

Radio-nuclide	Half-life	E_γ (keV)	Reaction contribution(%)	C/E	ΔC/C (%)	ΔE/E (%)
^{183}Hf	64 min	459 784	^{186}W(n,α) 100	1.33	18.	15.
^{184}Ta	8.7 h	253 318 414 921	^{184}W(n,p) 98.3 ^{186}W(n,t) 1.7	1.34	10.	6.5
^{186}Ta	10.5 min	738	^{186}W(n,p) 100	1.10	14.	16.
^{185m}W	1.67 min	132 174	^{186}W(n,2n) 100	0.88	61.	7.2
^{187}W	23.7 h	480 686 773	^{186}W(n,γ) 100	0.61	50.	5.9

Table 2 Results from the long irradiation

Radio-nuclide	Half-life	E_γ (keV)	Reaction contribution (%)	C/E	ΔC/C (%)	ΔE/E (%)
^{181}Hf	42.4 d	482	^{184}W(n,α) 100.	1.66	12.	7.5
^{182}Ta	114.7 d	1121 1189	^{182}W(n,p) 86.5 ^{183}W(n,d) 6.5	1.60	52.	6.8
^{183}Ta	5.09 d	354	^{183}W(n,p) 64.0 ^{184}W(n,d) 20.7 ^{186}W(n,α)β$^-$ 15.3	1.36	23.	7.8
^{184}Ta	8.7 h	318 414 921	^{184}W(n,p) 98.3 ^{186}W(n,t) 1.7	1.51	10.	11.
^{187}W	23.7 h	480 686 773	^{186}W(n,γ) 100	0.47	50	7.1

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2. Measurement and Analysis of Neutron and γ-ray Flux Spectra in Tungsten+

M. Angelone[1], P. Batistoni[1], U. Fischer[2], H. Freiesleben, D. Leichtle[2], C. Negoita, K. Seidel, S. Unholzer

Besides the integral activation experiment with W (report No 1, above), the transport data were tested in a benchmark experiment. A block of a tungsten alloy (density: 18.1 g/cm^3; elemental composition: 95 wt-% W, 1.6 wt-% Fe and 3.4 wt-% Ni) was irradiated at the Frascati Neutron Generator [1] with 14 MeV neutrons and the neutron and γ-ray flux spectra were measured at four positions inside the assembly.

The experimental set-up is shown in Fig. 1. The dimensions of the W block were 47 cm x 47 cm x 49 cm of length (z-axis). The neutron source was located at z = -5.3 cm. Flux spectra were measured in four positions (P 1,....P 4) at z = 5, 15, 25 and 35 cm.

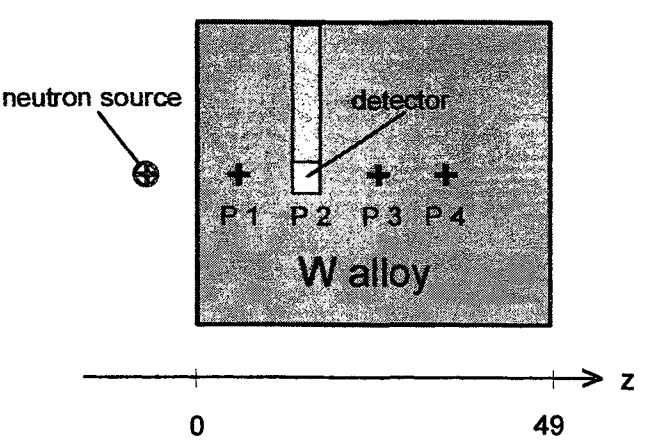

Fig. 1 Geometry of the benchmark assembly with the detector in position 2.

[1] Associazione ENEA-EURATOM, Fusion Divison, Frascati, Italy
[2] Forschungszentrum Karlsruhe, Institut für Reaktorsicherheit

Neutron and γ-ray flux spectra were measured simultaneously, using an NE 213 liquid-scintillation spectrometer. The dimensions of the cylindrical active volume of the detector were 38 mm in both height and diameter. The material had a mass density of 0.874 g/cm^3 and an elemental composition of 54.8 at-% H and 45.2 at-% C. The scintillator was coupled to a photomultiplier by means of a 50 cm long light guide. When the detector was located at one of the positions (P 2 in Fig. 1), the other ones were filled with rods of W.

The pulse-height response matrices of the detector were determined on the basis of detailed simulations of experimental distributions from mono-energetic neutron and γ-ray sources with Monte Carlo codes. The DIFBAS code [2] was employed for unfolding the measured pulse-height distributions. Neutron and γ-ray spectra were obtained as absolute fluxes, as the response matrices have been determined on an absolute scale. The flux spectra measured at P 2 are shown in Fig. 2 together with the calculated distributions.

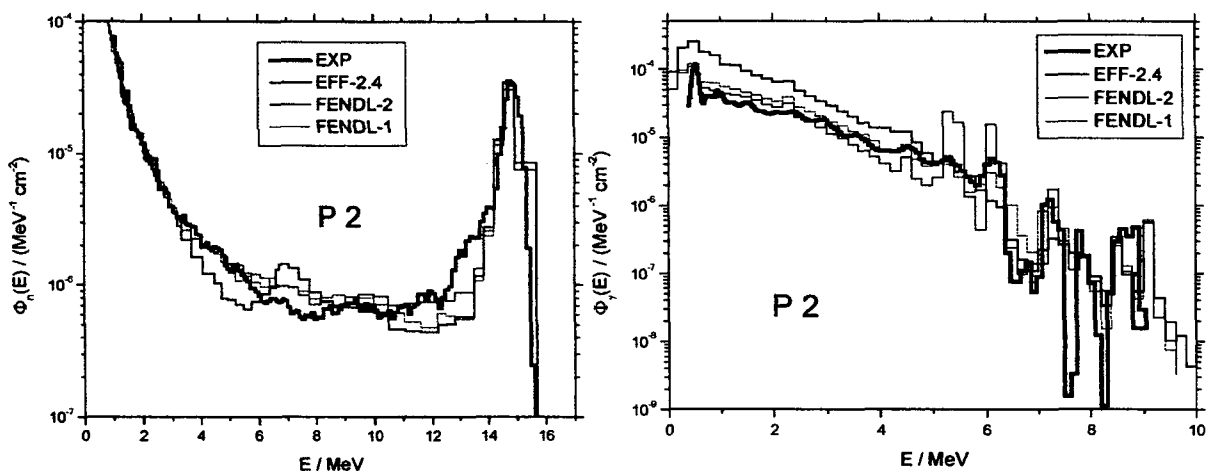

Fig. 2 Measured neutron (left hand) and γ-ray (right hand) flux spectra at detector position P 2 (normalised to one source neutron) in comparison to fluxes calculated with data from the libraries EFF-2.4, FENDL-1 and FENDL-2.

The calculations of the flux spectra were performed with the Monte Carlo code MCNP-4C [3] using a full 3D geometry model of the assembly, the neutron generator and the experimental hall. Nuclear data were taken from the Fusion Evaluated Nuclear Data Libraries FENDL/MC-2.0 [4] and FENDL-1 [5] and from the European Fusion File EFF-2.4 [6].

Comparisons of measured and calculated flux spectra for the other three detector positions and ratios of calculated-to-experimental fluxes (C/E) for energy ranges were published in Ref. [7].

These comparisons show that the fast (E > 1 MeV) neutron flux in a W assembly of a thickness corresponding to about 16 mean free path for 14 MeV neutrons can be reproduced by MCNP calculations within 20% using EFF-2.4, FENDL-1 or FENDL-2 data. FENDL-2 calculations give the best overall agreement with the measured spectra although the neutron flux for the 14 MeV peak range (E > 12.5 MeV) is underestimated by up to 30% at the deepest detector position.

The total γ-ray flux for E > 0.4 MeV is reproduced within 20% by FENDL-2 data. Overestimations with C/E = 1.7 – 2.9 were found for EFF-2.4 data. The flux in the capture peak region (E = 5 – 9 MeV) is underestimated by EFF-2.4 and FENDL-1 data, but overestimated by the FENDL-2 calculations up to a factor of 3 at deep detector positions. Especially the slow neutron induced γ-ray production data should be revised.

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3. Electron Transitions in Neon-like Xenon Ions investigated by Wavelength Dispersive x-ray Spectroscopy*

U. Kentsch[2], G. Zschornack[1], F. Großmann[2], V. P. Ovsyannikov[2], F. Ullmann[2], S. Fritzsche[3]

The room temperature Dresden Electron beam ion trap (EBIT) has proven its applicability in x-ray spectroscopy and extraction of highly charged ions in many investigations during the last years, where the production of ions such as Ar^{18+}, Fe^{26+}, Ni^{28+}, Xe^{44+} and Ir^{67+} has been shown [1 - 4]. The setup of the Dresden EBIT has already been described in detail elsewhere [5,6].

In the investigations described here xenon is injected as neutral gas and within 1 s ionized by an electron beam of 40 mA at an electron energy of 13 keV to the mean ion charge state of 44, that is neon-like xenon. L x-rays emitted from the xenon ions have been detected by a crystal diffraction spectrometer. The calibration of the spectrometer has been accomplished with the well known Ti^{20+} w line, using $TiCl_4$ as working gas.

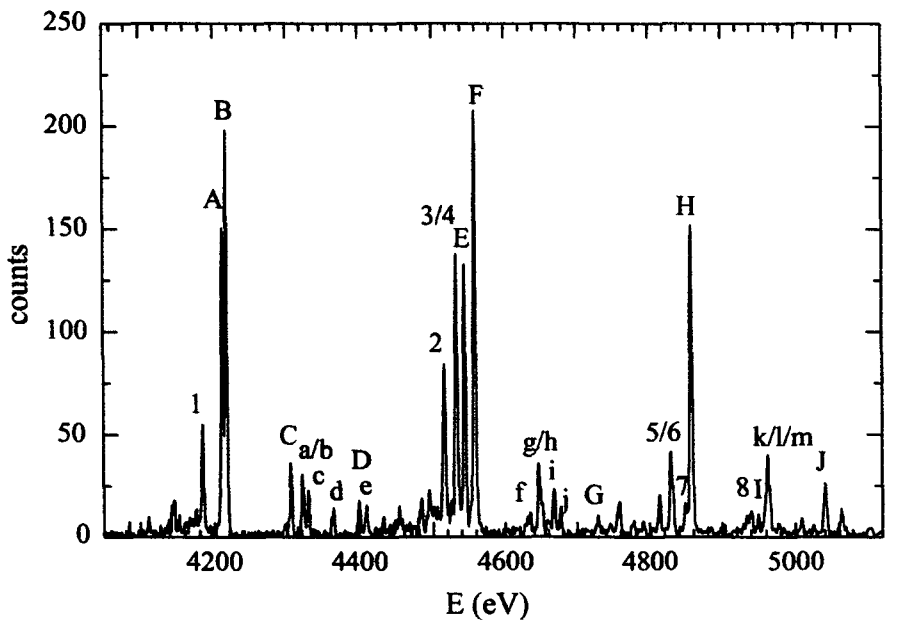

Fig. 1 Crystal diffraction spectrum of L x-ray transitions in $Xe^{43+...45+}$

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A complete neon-like Xe L x-ray spectrum with transition lines from adjacent charge states is shown in Fig. 1. Transition lines from Xe^{44+} are indicated by capital letters, transitions from Xe^{43+} by numbers and transitions from Xe^{45+} by small letters.

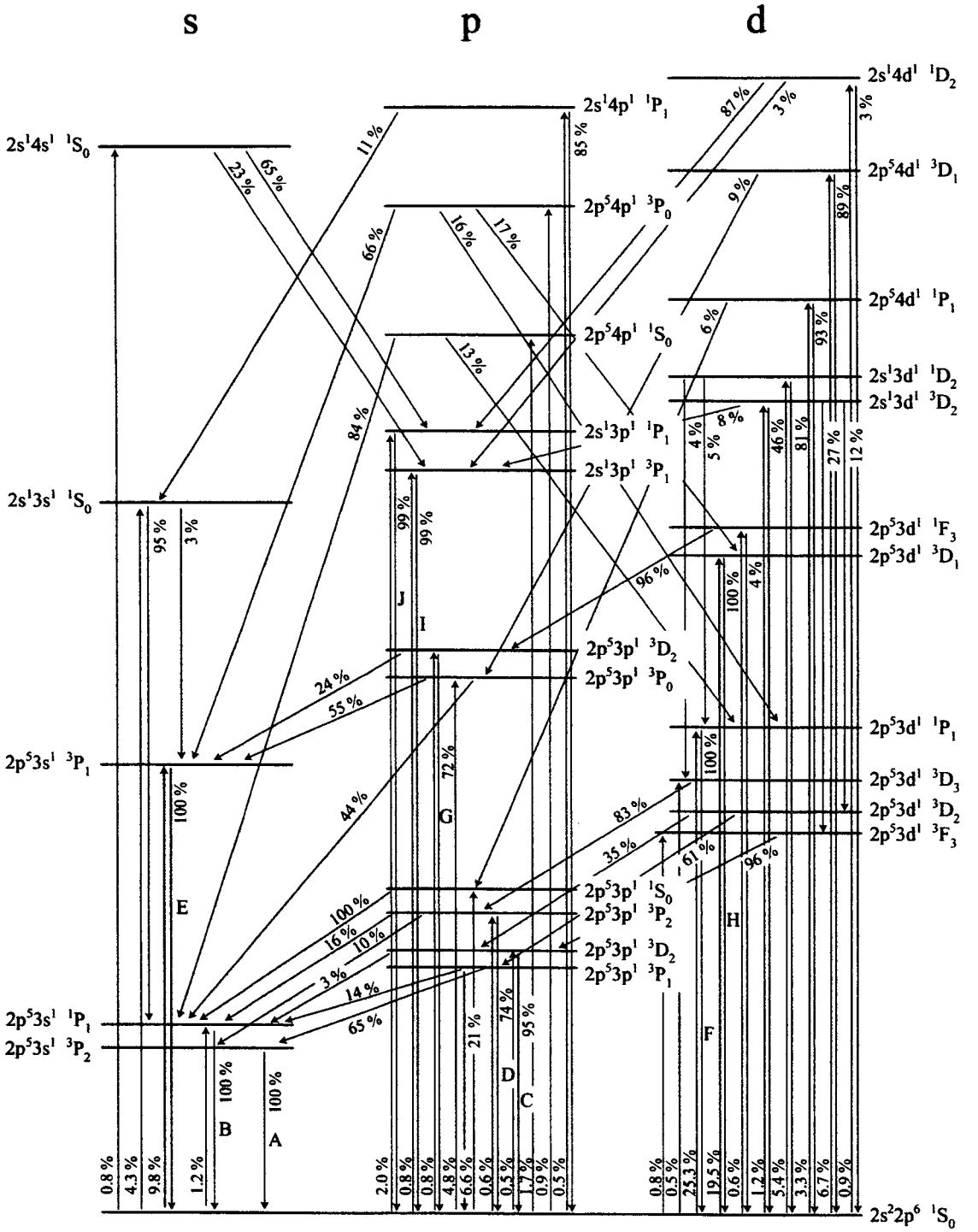

Fig. 2 Grotrian diagram of transitions in Xe^{44+} after 13 keV electron excitation

The energy of 13 keV is high enough to excite L electrons to all n=3, n=4 and higher levels leading to cascade feeding of the $2s^22p^53s^1$ configuration. This is shown in Fig. 2, where the

decay scheme of Xe^{44+} after excitation with an electron energy of 13 keV is given. The excitation probabilities are derived from calculated excitation cross sections and the decay probabilities are derived from calculated transition rates, where all 89 involved levels of the excitation from n=2 into the n=3 and n=4 shell are taken into account. It can be seen that line A contributes with less than 1% of the total spectrum intensity that follows the direct excitation. Indeed the observed intensity of line A is comparable to that of allowed transitions. There are two more processes which populate levels $2p^5 3s^1\ ^1P_1$ and $2p^5 3s^1\ ^3P_2$. One is the ionization of the L shell in Xe^{43+} where already an electron occupies the 3s shell and thus mainly contributes to the population of these levels. The other possibility is the capture of an electron in an outer shell of Xe^{45+} where a vacancy in the L shell already exists and the levels are filled by cascade decay.

From calculated cross sections it results that about 70% of the intensity of the spectrum in Fig. 1 originates from the decay after direct excitation and about 20% are decay transitions after recombination, that is electron capture into Xe^{45+}. The remaining 10% intensity is attributed to transitions after ionization of an L shell electron in Xe^{43+}.

Thus, with the Dresden EBIT a device is available that is excellently qualified for investigations of both excitation and recombination processes in highly charged ions.

We mention that now Xe^{44+} ions have also been extracted, and are available for ion surface interaction applications.

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ZENTRUM FÜR STRAHLENSCHUTZ UND RADIOÖKOLOGIE,
UNIVERSITÄT HANNOVER
AND
ABTEILUNG NUKLEARCHEMIE, UNIVERSITÄT ZU KÖLN

Production of Residual Nuclides at Medium Energies – Investigation of Proton- and Neutron-Induced Reactions within the HINDAS Project

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Introduction

Emerging from a European Concerted Action "Lead for Transmutation" [1] the project HINDAS "High- and Intermediate-Energy Data for Accelerator-Driven Systems" [2] was initiated in the European 5th Framework with the goals to establish a comprehensive and consistent data base covering all aspects of nuclear reaction data at intermediate and high energies relevant for accelerator-driven technologies and to develop two new code systems for the modeling of nuclear reactions. Including sub-contractors, nearly 20 European institutions collaborate within the HINDAS project. Three target elements, Fe, Pb and U, were chosen for which a complete experimental data base was and is being established by which the newly developed models shall be scrutinized. The HINDAS project made use of accelerators at UCL Louvain-La-Neuve, TSL University of Uppsala, PSI Villigen, KVI Groningen, GSI Darmstadt, FZJ Jülich, and the PSI/ETH Tandem AMS Facility at ETHZ Zürich to cover energies from 20 MeV up to 2 GeV. The tasks were formally divided in two energy regimes, one below and the other above 200 MeV.

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Though the HINDAS project will be completed only by the end of 2003, we can report here about some new results and upcoming developments of the HINDAS work-package 3. This work-package deals with measurements of cross sections for residual nuclide production by p- and n-induced reactions for energies between 30 MeV and 200 MeV, of cross sections for n-induced fission for energies between 30 and 180 MeV, and of excitation functions for the production of long-lived radionuclides such as Be-10, Al-26, Cl-36, and I-129, by p-induced reactions from thresholds up to 2.6 GeV. In detail, we report here the measurement of gamma-emitting residual nuclides produced in proton- and neutron-induced reactions. The investigations of fission cross sections performed by PTB and UCL and the measurements of cross-sections for the production of long-lived radionuclides performed in collaboration with PSI and ETHZ will be reported later.

Production of Residual Nuclides by Proton-Induced Reactions on Uranium up to 72 MeV

For proton-induced reactions, these investigations aim to further develop and complete the cross section database which was established by our collaboration in recent years [3], and references therein]. It is now extended to heavy target elements such as Ta, W, Pb and Bi [4-6]. Within the HINDAS project about 560 new cross sections for the production of 19 radionuclides from the target element iron have been measured from thresholds up to 2.6 GeV which will be reported in the HINDAS Final Report. For the target element lead a comprehensive set of excitation functions published recently [4] will be completed by cross sections for the production of long-lived radionuclides and for stable He-isotopes. Irradiation experiments with natural uranium were performed at the injector cyclotron at PSI using the stacked-foil technique and subsequent gamma-spectrometry. About 400 cross sections for the production of 18 radionuclides at energies 21 to 69 MeV were measured.

All these data allow stringent tests of nuclear models and codes when calculating cross sections for residual nuclide production from thresholds up to 2.6 GeV. The new codes are the TALYS code for energies up to about 200 MeV and the INCL4/GSI code for higher energies. Detailed reports of the new codes will be published in the HINDAS final report at the end of 2003. Comprehensive tests of the new codes, TALYS and INCL4/GSI, for Fe, W, Ta, Pb, Bi and U for energies from thresholds up to 2.6 GeV are underway.

Here we report new experimental results for proton-induced reactions on uranium and their description by TALYS calculations. The TALYS code is a new program system, which allows

calculation of all nuclear reaction data for a given reaction comprising a wide variety of features. The TALYS code combines among others a compound nucleus model in the form of Hauser Feshbach calculations [7] with a pre-equilibrium model according to the exciton model by Kalbach [8]. The transmission coefficients are calculated by an optical model making use of the recent parameterization by Koning and Delaroche [9]. The level densities are used in form of a back-shifted Fermi-gas model according to Gilbert and Cameron [10], taking into account the Ignatyuk formalism [11]. The competition between particle emission, gamma-de-excitation and fission is calculated at each energy step of the proceeding reaction. Fission is treated according to the Brosa model [12] as incorporated earlier into fission modeling by Duijvenstijn et al. [13].

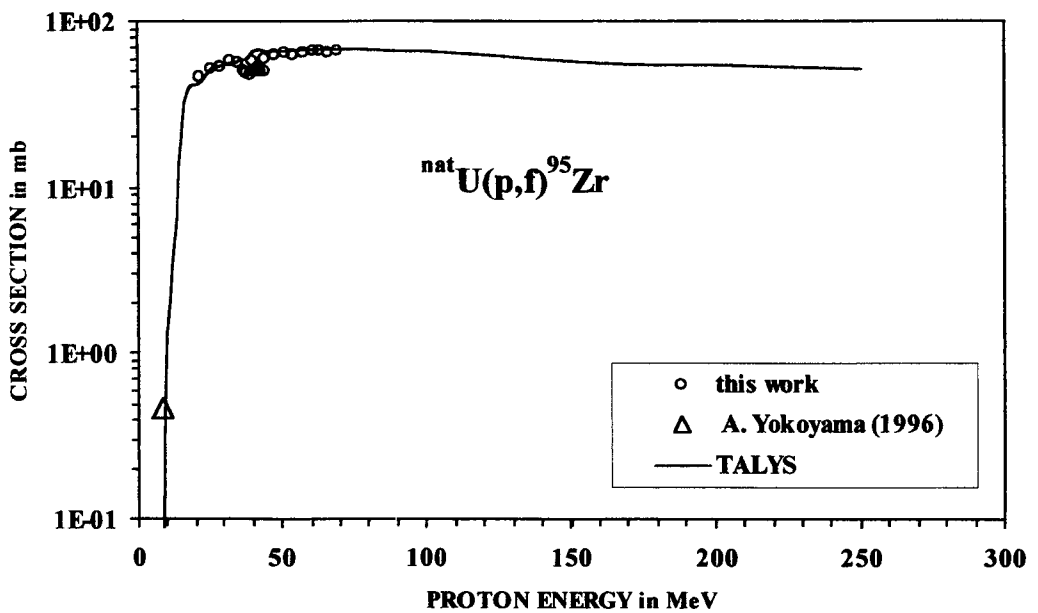

Fig. 1 Comparison of experimental cross sections from this work and from Yokoyama [14] with theoretical ones calculated by the TALYS code for the cumulative production of ^{95}Zr in proton-induced fission of natural uranium.

In our work, we measured cross sections for the formation of ^{91}Y, ^{95}Zr (Fig. 1), ^{95m}Nb (Fig. 2), ^{99}Mo, ^{103}Ru, ^{112}Pd, ^{115}Cd, ^{124}Sb, ^{126}Sb, ^{127}Sb, ^{132}Te, ^{131}I, ^{134}Cs (Fig. 3), ^{136}Cs, ^{137}Cs, ^{140}Ba, ^{141}Ce, ^{144}Ce, ^{147}Nd, and ^{238}Np from natural uranium at energies between 21 MeV and 69 MeV. Not many cross sections for proton-induced production of residual nuclides from uranium are available in literature. Moreover, they are mostly old and are neither systematic nor comprehensive with respect to the product nuclide and energy coverage. For example, for ^{95}Zr only one earlier measurement existed for energies up to 300 MeV. The TALYS calculations are in excellent agreement with our measurements and with the only earlier data point by Yokoyama [14] at lower energies. The agreement between theory and experiment seen in Fig. 1 is typical for all product nuclides with masses up to 115. Among the measured product nuclides there is just one isomer (^{95m}Nb), for

which theoretical data do not yet exist. For this nuclide there are also severe discrepancies between our new data and the only earlier measurement by Davies et al. [15] as seen in Fig. 2.

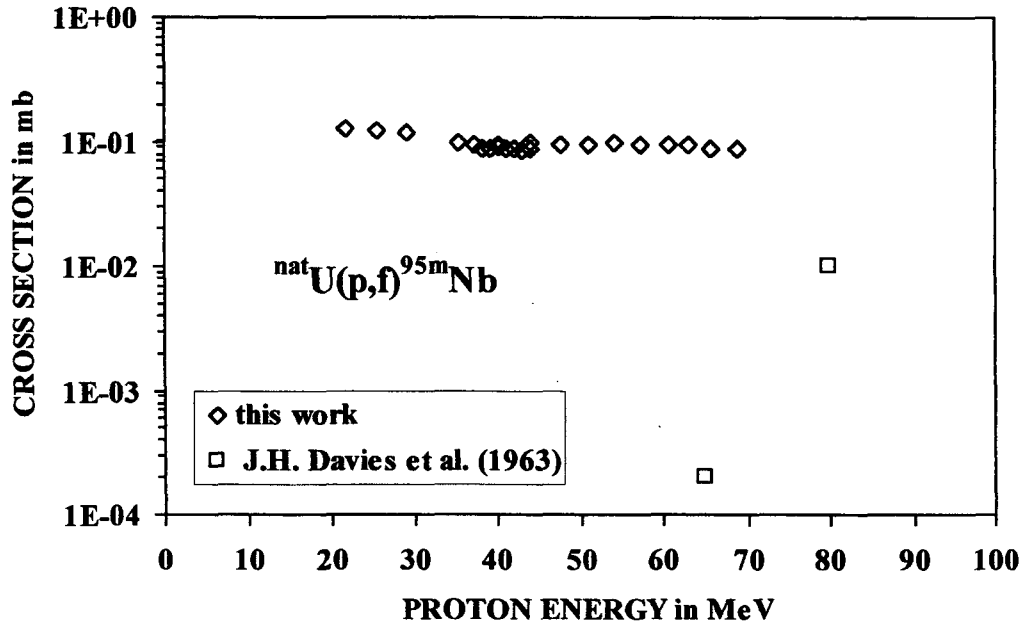

Fig. 2 Experimental cross sections from this work and from Davies et al. [15] for the production of ^{95m}Nb in proton-induced fission of natural uranium.

For higher mass fission products and for ^{238}Np there are still systematic deviations between theory and experiments. In Fig. 3 this is exemplified by the production of ^{134}Cs. For this nuclide, a number of earlier measurements exist for energies below 300 MeV which allow an estimation of excitation function fairly well. Above 100 MeV the TALYS calculations agree well with the experimental data. But at lower energies systematic deviations between theory and experiment are seen. Theory predicts a much steeper increase of the cross section with energy than the experiment reveals. Also, the maximum in the experimental excitation function is much less pronounced and located at higher energies than predicted by theory. These deviations are presently discussed as deficits of the fission model in the heavy part of the fission product distribution. The systematics of the new experimental data will provide here a basis to describe these deficits and to improve the fission models.

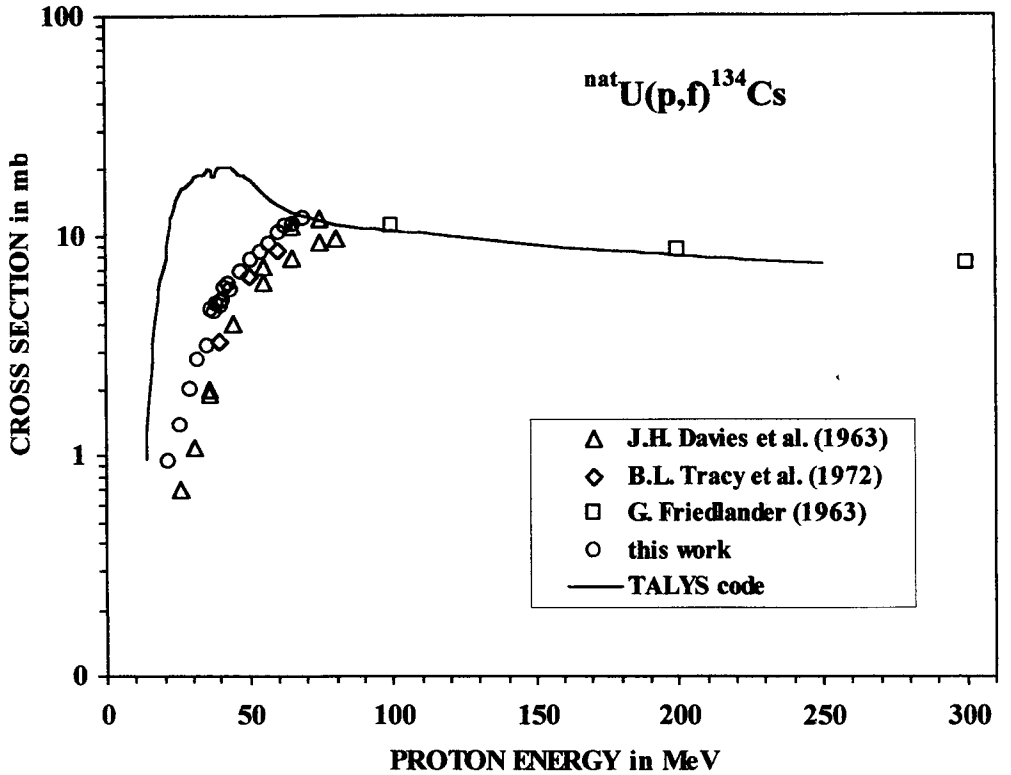

Fig. 3 Comparison of experimental cross sections from this work and from Davies et al. [15], Tracy et al. [16] and Friedlander [17] with theoretical ones calculated by the TALYS code for the independent production of ^{134}Cs in proton-induced fission of natural uranium.

Production of Residual Nuclides by Neutron-Induced Reactions up to 175 MeV

Within the HINDAS project, activation experiments with quasi mono-energetic neutrons produced by the $^{7}Li(p,n)^{7}Be$ reaction were performed at the neutron beam lines at TSL/Uppsala [18, 19] and UCL/ Louvain La Neuve [20] in order to determine excitation functions for the production of residual radionuclides from a variety of target elements up to 175 MeV. Last year, we described in some detail the experimental techniques, in particular the neutron monitoring techniques applied [21, 22].

A total of 10 activation experiments covered proton energies of 36.4, 48.5 and 62.9 MeV at the UCL and of 69.1, 76.4, 98.5, 136.7, 148.4, 162.7 and 178.8 MeV at TSL. The target elements C, N, O, Mg, Al, Si, Fe, Co, Ni, Cu, Ag, Te, Pb and U were irradiated with the highest beam currents available. Residual radionuclides with half-lives between 20 min and 5 years were measured by off-line γ-spectrometry. In spite of the long irradiation times and high beam currents applied, the measurement of the irradiated targets is a low-count-rate problem and requires close-to-detector

geometries. This results in particular problems with respect to efficiency determination and to necessary corrections for systematic coincidence. In addition, γ-self-absorption in the targets has to be corrected for. Therefore, a new method for the determination of detector efficiency was developed which takes into account all these effects simultaneously.

The γ-spectrometry of the activated target foils yields activities proportional to production rates P in $s^{-1}\ g^{-1}$. A production rate P of a nuclide produced is given by:

$$P = \frac{N_L}{A_T} \int \sigma(E) \cdot J(E)\, dE \quad \text{with} \quad J(E) = \frac{d^2\Phi(E)}{dE\, dt} \quad (1)$$

with N_L being Avogadro's number and A_T the atomic mass of target element. $\sigma(E)$ are the neutron cross sections, $J(E)$ the spectral neutron flux densities and $\Phi(E)$ the neutron fluences. The integral is taken over all neutron energies E.

Information on the energy dependence of the neutron spectra in the targets was obtained by modeling the neutron spectra by Monte Carlo techniques using the LAHET/MCNP code system [23, 24]. These transport calculations started either from the experimentally determined neutron spectra (at UCL) or from the systematics of experimentally measured neutron emission spectra of the ^{7}Li(p,n)-reaction [25] (at TSL). The calculations described the transport of the neutrons into the target stacks and into the individual targets as well the production and transport of secondary particles inside the massive target stacks which cannot be neglected.

Cross sections cannot be directly calculated from these response integrals since the neutrons used are just "quasi mono-energetic" with only about 30 to 50% of the neutrons in the high-energy peak with a width of a few MeV. The neutron cross sections $\sigma(E)$ have to be extracted from production rates P_i (i = 1,..., n) determined in a series of n irradiation experiments with different neutron energies by iterative procedures [26] or by unfolding using the STAYSL formalism [27, 28]. The feasibility of the unfolding method was successfully demonstrated in the analysis of thin-target and thick-target data [29 – 31].

In our earlier work [22], the determination of cross sections by unfolding turned out to be unfeasible, leading to unphysical shapes of the excitation functions. This was due to the fact that the information about the cross sections is not homogeneously distributed on the neutron energy axis but rather is concentrated at the peak neutron energies of the different experiments. Therefore, the iterative approach of Kim et al. [26] was used which does not give entire excitation functions but

results in individual cross sections at the peak neutron energies of the different experiments, only. This was presented for $^{nat}Pb(n,2pxn)^{203}Hg$ as an example [22].

In the mean time this problem was overcome by properly adjusting the unfolding procedure, the width of energies considered as relevant for each irradiation experiment. This means that the unfolding is performed iteratively, starting with the experiment with the lowest proton energy and proceeding to higher ones. Then, in each unfolding step all energies below the highest one are taken into account. This procedure results then in a complete excitation function up to the highest covered energy and gives the respective uncertainty of the cross sections for each energy point. In Fig. 4 this is exemplified for the reaction $^{nat}Ag(n,xn)^{106}Ag$. In Fig. 4 also the location and widths or the high energy peaks in the individual irradiation experiments are indicated. It is obvious that the uncertainties of the excitation function increase with increasing energies since less information is available for higher than for lower energies.

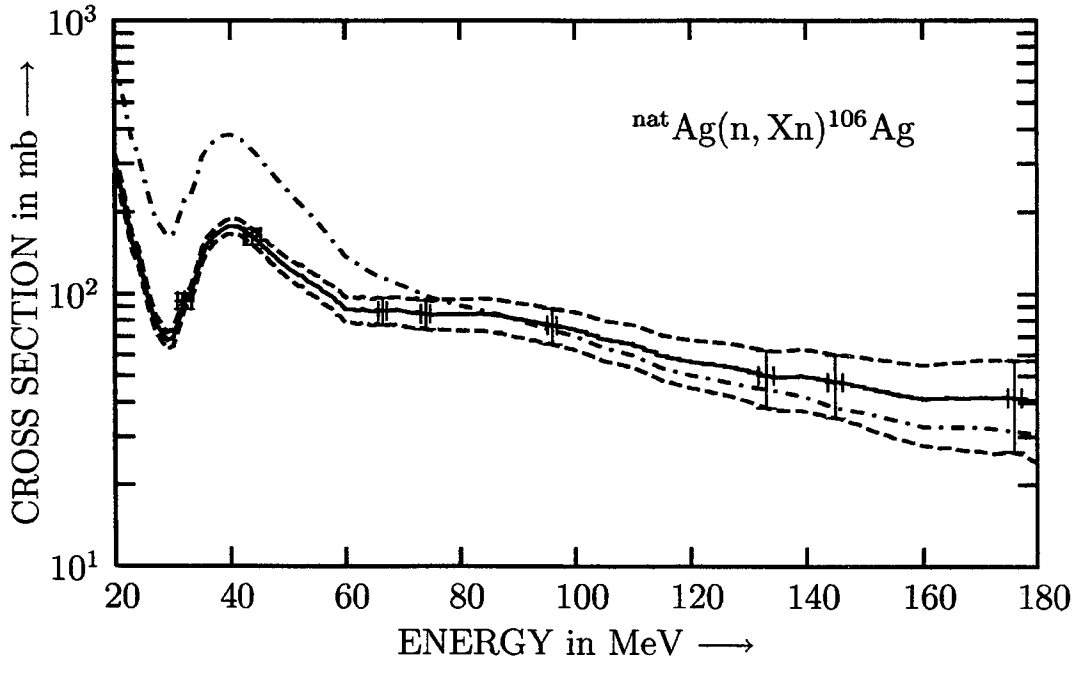

Fig. 4 Guess excitation function (dashed-dotted line) and final cross sections (solid line) with their uncertainties (dashed lines) for the reaction $^{nat}Ag(n,xn)^{106}Ag$. The crosses are the cross sections at the peak energies of the neutron irradiation experiments. The uncertainties in energy given for the cross sections represent the half-width of the peak in the neutron spectrum.

The unfolding needs a guess function to start with and which in our case was calculated using the ALICE-IPPE code [32]. But, the unfolding procedure is rather independent of the guess functions. This is exemplified in Fig. 5 for the reaction $^{nat}Cu(n,2pxn)^{56}Co$. There, two guess functions are

given calculated by the ALICE-IPPE code. The calculations of the guess functions differ just in the way the nuclear masses are calculated. But, independent of the guess functions used we end up with the same unfolded excitation function. This reaction is one of the rare cases where we can

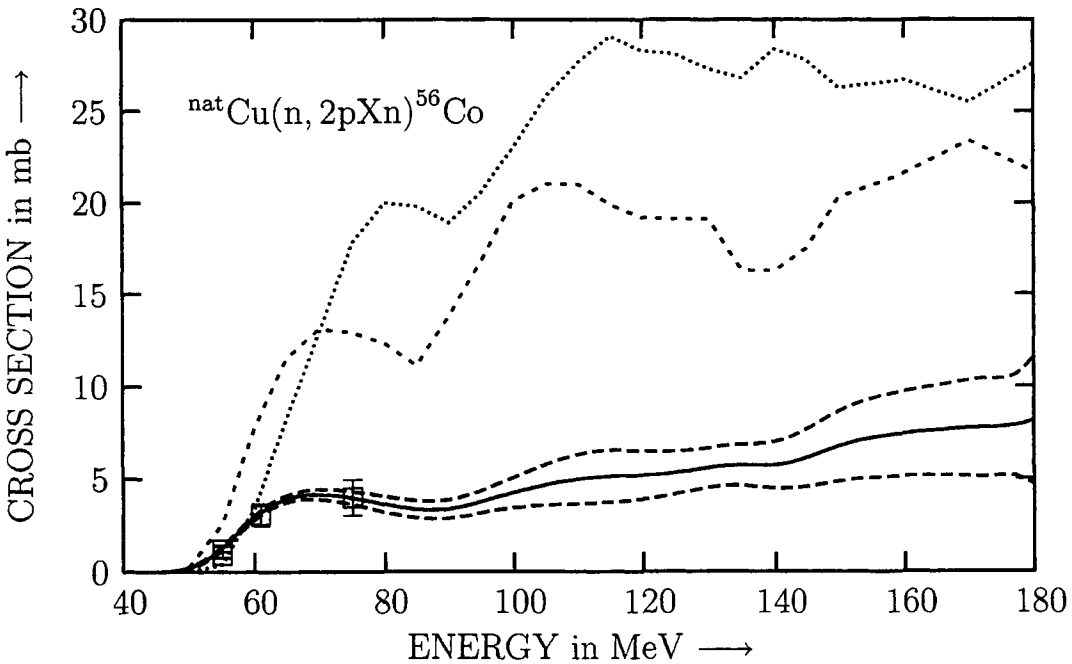

Fig. 5 Unfolded cross sections (solid line) with their uncertainties (dashed lines) for the reaction $^{nat}Cu(n,2pxn)^{56}Co$. The squares are cross sections reported earlier by Kim et al. [26]. The dotted and short-dashed lines represent two guess functions calculated by the ALICE-IPPE code using the Myers and Swiatecki mass formula [33] without and with shell corrections, respectively.

compare our results with those of other authors. Our excitation function is in excellent agreement with the data reported by Kim et al. [26].

The importance of distinguishing residual nuclide production by proton-induced reactions from that by neutron-induced ones is emphasized in Fig. 6, again for the reaction $^{nat}Cu(n,2pxn)^{56}Co$. The excitation functions for the proton- and neutron-induced production differ strongly with respect to shapes and absolute values. The total results of this study will comprise excitation functions for about 120 reactions and will provide a basis to scrutinize the capabilities of models and codes to describe adequately the differences in neutron- and proton-induced reactions.

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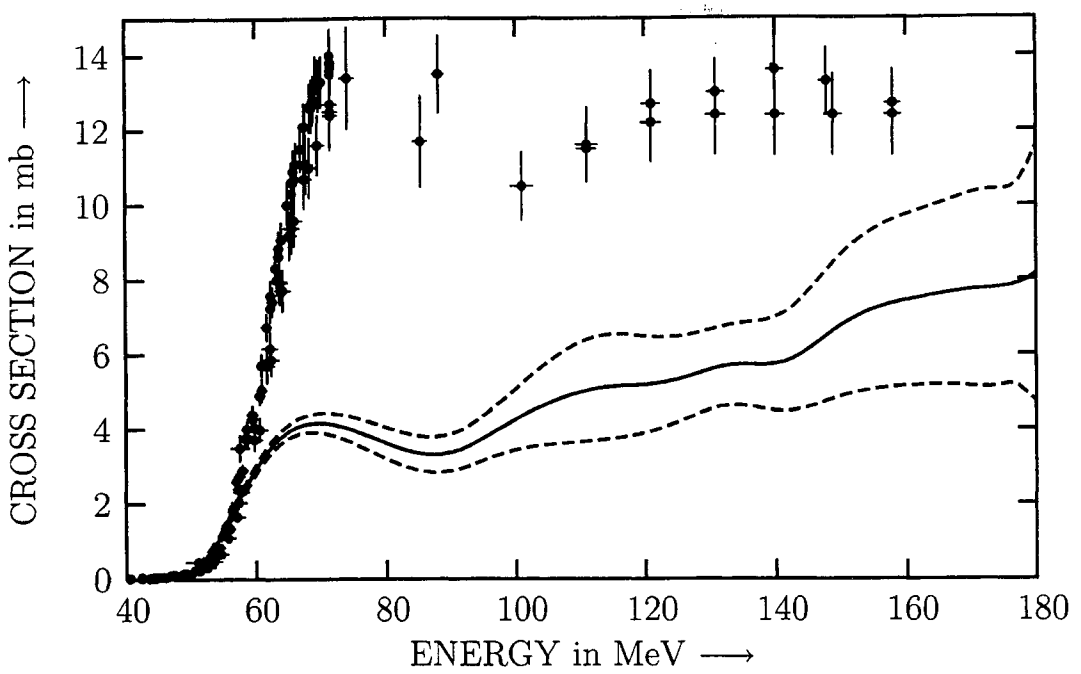

Fig. 6 Unfolded cross sections (solid line) with their uncertainties (dashed lines) for the reaction $^{nat}Cu(n,2pxn)^{56}Co$ compared with the cross sections from ref. [3] for the production of ^{56}Co via the $^{nat}Cu(p,3pxn)^{56}Co$ process (dots with error bars).

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PHYSIKALISCH-TECHNISCHE BUNDESANSTATLT
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Validation of Evaluated Cross Section Data in the Neutron Field of Spontaneous Fission of ^{252}Cf

W. Mannhart

Within the framework of an IAEA Data Development Project, it is envisaged to create a new version of the International Reactor Dosimetry File (IRDF-2002) which is primarily intended for reactor pressure vessel surveillance purposes and lifetime assessment. In this context, the individual reaction cross sections, $\sigma(E)$, taken from various cross section libraries were tested in a typical fission neutron field.

The cross section data investigated stem from the previous version of the International Reactor Dosimetry File (IRDF-90.2) [1], the JENDL Dosimetry File (JENDL/D-99) [2] and the Russian Reactor Dosimetry File (RRDF-98) [3].

The reference neutron field finally used for the validation was the neutron field of spontaneous fission of ^{252}Cf. Various aspects guided this decision, among them the availability of a sufficiently large and accurate database of experimental spectrum-averaged cross sections $\langle\sigma\rangle$ and a well-defined and documented spectral distribution N(E) of the relevant neutron field. Especially the latter criterion limited the number of suitable reference neutron fields. For example, the available representations of the reactor-based fission neutron field of $^{235}U + n(thermal)$ seriously fail in describing the high-energy portion (> 10 MeV) of the neutron spectrum and are lacking in the description of the spectral uncertainties.

The spectral distribution N(E) of the fission neutrons of ^{252}Cf is the result of an evaluation based on modern TOF measurements of the neutron spectrum [4]. The numerical figures and the covariance matrix are given in Ref. [5].

Calculated spectrum-averaged cross sections of $\int \sigma(E)\, N(E)\, dE \,/\, \int N(E)\, dE$ were determined for the various $\sigma(E)$ data. The associated uncertainties were obtained from the propagated uncertainties of $\sigma(E)$ and N(E). The calculated data were compared with the experimental

Table 1 C/E-values in the ^{252}Cf neutron field, calculated with σ(E) from the cross section libraries IRDF-90.2, JENDL/D-99 and RRDF-98.

Cf-252(sf)						
Reaction	E(50%)	Exp.		C/E		
	MeV	<σ> (mb)	%	IRDF-90.2	JENDL/D-99	RRDF-98 (Update)
Au-197(n,γ)Au-198m+g	0.75	7.679E+1	1.59	0.966 ± 0.021	0.977 ± 0.086	
Cu-63(n,γ)Cu-64	0.93	1.044E+1	3.24	0.996 ± 0.091	1.005 ± 0.196	
In-115(n,γ)In-116m1+m2	1.06	1.256E+1	2.23	0.969 ± 0.047	1.003 ± 0.047	
U-235(n,f)	1.70	1.210E+3	1.20	1.007 ± 0.012	1.021 ± 0.024	
Pu-239(n,f)	1.78	1.812E+3	1.37	0.980 ± 0.014	0.996 ± 0.025	
Np-237(n,f)	2.07	1.361E+3	1.59	0.999 ± 0.093	0.983 ± 0.016	
In-115(n,n')In-115m	2.68	1.974E+2	1.37	0.961 ± 0.025	0.961 ± 0.025	0.959 ± 0.020
U-238(n,f)	2.78	3.257E+2	1.64	0.969 ± 0.017	0.980 ± 0.026	
Hg-199(n,n')Hg-199m	3.10	2.984E+2	1.81		0.833 ± 0.067	
Ti-47(n,p)Sc-47	3.84	1.927E+1	1.66	1.006 ± 0.042	0.962 ± 0.021	
S-32(n,p)P-32	4.06	7.254E+1	3.49	0.969 ± 0.049	1.033 ± 0.090	
Ni-58(n,p)Co-58m+g	4.17	1.175E+2	1.30	0.982 ± 0.026	0.975 ± 0.016	1.000 ± 0.023
Zn-64(n,p)Cu-64	4.26	4.059E+1	1.65	1.037 ± 0.054	0.942 ± 0.023	
Fe-54(n,p)Mn-54	4.32	8.684E+1	1.34	1.015 ± 0.026	1.027 ± 0.019	
Co-59(n,p)Fe-59	5.76	1.690E+0	2.48			
Al-27(n,p)Mg-27	5.87	4.880E+0	2.14	0.958 ± 0.039	1.058 ± 0.027	
Ti-46(n,p)Sc-46m+g	6.08	1.407E+1	1.77	0.876 ± 0.029	0.984 ± 0.030	0.983 ± 0.037
V-51(n,p)Ti-51	6.44	6.488E-1	1.97			
Cu-63(n,α)Co-60m+g	7.28	6.887E-1	1.96	0.986 ± 0.033	1.059 ± 0.029	1.007 ± 0.037
Fe-56(n,p)Mn-56	7.56	1.465E+0	1.77	0.936 ± 0.030	0.962 ± 0.048	1.007 ± 0.035
Mg-24(n,p)Na-24	8.25	1.996E+0	2.44	1.082 ± 0.040	1.092 ± 0.034	1.073 ± 0.034
Co-59(n,α)Mn-56	8.36	2.218E-1	1.88	0.975 ± 0.036	1.040 ± 0.050	0.997 ± 0.043
Ti-48(n,p)Sc-48	8.38	4.247E-1	1.89	0.912 ± 0.032	0.931 ± 0.028	1.005 ± 0.057
Al-27(n,α)Na-24	8.66	1.016E+0	1.28	1.022 ± 0.026	1.022 ± 0.026	
V-51(n,α)Sc-48	9.97	3.900E-2	2.21	0.995 ± 0.044		0.989 ± 0.041
Tm-169(n,2n)Tm-168	10.34	[6.690E+0]	6.28		0.932 ± 0.065	
Au-197(n,2n)Au-196m+g	10.61	5.506E+0	1.83	1.044 ± 0.052	1.049 ± 0.031	
Nb-93(n,2n)Nb-92m	11.47	[7.490E-1]	5.07	1.041 ± 0.064	1.011 ± 0.070	1.030 ± 0.058
I-127(n,2n)I-126	11.75	2.069E+0	2.73	1.062 ± 0.045	1.096 ± 0.051	
Cu-65(n,2n)Cu-64	12.64	6.582E-1	2.22	1.030 ± 0.042	1.061 ± 0.039	
Mn-55(n,2n)Mn-54	12.84	4.075E-1	2.33	1.181 ± 0.115	1.237 ± 0.111	
Co-59(n,2n)Co-58m+g	13.06	4.051E-1	2.51	1.044 ± 0.051	1.030 ± 0.045	
Cu-63(n,2n)Cu-62	13.75	1.844E-1	3.98	1.134 ± 0.068	1.140 ± 0.066	
F-19(n,2n)F-18	14.02	1.612E-2	3.37	1.065 ± 0.063	1.151 ± 0.070	1.009 ± 0.064
Zr-90(n,2n)Zr-89m+g	14.41	2.210E-1	2.89	1.001 ± 0.061	0.979 ± 0.058	
Ni-58(n,2n)Ni-57	14.98	8.952E-3	3.57	1.033 ± 0.079	1.004 ± 0.072	

data and the C/E values were derived. The experimental data are from a detailed evaluation [6] of the available integral experiments.

The results are summarized in Table 1. The investigated neutron reactions are given in column 1. The reactions are listed with increasing energy response ranges. In column 2 the mean neutron energy E(50%) of the integrated response of each neutron reaction in the fission neutron field is given. The experimental spectrum-averaged cross sections and the uncertainties are given in columns 3 and 4. The two values given in brackets are from single experiments and were not part of the evaluation process. In columns 5 to 7 the C/E values obtained with the IRDF-90.2, the JENDL/D-99 and the RRDF-98 library are given. Most of the original σ(E) data of the RRDF-98 library were replaced by data of the recent updates [3].

The rigorous inclusion of all uncertainty components contributing to the given C/E values allows quantitative statements on the quality of the evaluated σ(E) data valid for the energy response range of the reaction. The calculated C/E values which show agreement with unity within the given uncertainties are printed in bold and indicate optimum agreement between integral and differential data. Besides such values, C/E values were also accepted which were within a band of ± 5 % around unity even if the calculated uncertainties were too small to overlap with unity. These values are shaded in the table in gray. For most of the investigated reactions a suitable data set of σ(E) is identified in one of the investigated libraries. Only for the reactions ^{199}Hg(n,n'), ^{24}Mg(n,p), ^{127}I(n,2n), ^{55}Mn(n,2n) and ^{63}Cu(n,2n) does this statement fail.

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APPENDIX

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