

Report

NPL REPORT IR 14

UKNSF(2008)P224 INDC(UK)0092 Distr. G

United Kingdom Nuclear Science Forum Progress Report Data Studies During 2007

Edited by N P Hawkes

NOT RESTRICTED

FEBRUARY 2009

United Kingdom Nuclear Science Forum

Progress Report

Data Studies During 2007

Edited by N P Hawkes Quality of Life Division

ABSTRACT

The United Kingdom Nuclear Science Forum (UKNSF) meets twice a year to discuss issues relating to the measurement and evaluation of nuclear data. Topics cover a wide range of applications in the UK nuclear industry. Links between members are maintained throughout the year, mainly through e-mail and the UKNSF website (www.uknsf.org.uk). Work of primary interest includes the measurement and evaluation of decay data (e.g. half-lives and gamma ray emission probabilities), fission yields, and neutron cross sections for fission and fusion. All known studies within the UK are summarised in this report. Specific applications and international links of relevance are also described.

© Crown copyright 2009 Reproduced with the permission of the Controller of HMSO and Queen's Printer for Scotland

ISSN 1754-2952

National Physical Laboratory Hampton Road, Teddington, Middlesex, TW11 0LW

Extracts from this report may be reproduced provided the source is acknowledged and the extract is not taken out of context.

Approved on behalf of the Managing Director, NPL, by Bajram Zeqiri, Knowledge Leader, Acoustics & Ionising Radiation Team, authorised by Martyn Sené, Director, Quality of Life Division

Contents

I	Inti	roduc	ction	1		
2	Measurements					
	2.1	Rad	lioactivity Metrology at NPL	2		
	2.1.	.1	Primary Radioactivity Standardisations	2		
2.1.2		.2	Equipment upgrades	2		
2.1.3		.3	Acknowledgement	2		
	2.2	Neu	ntron Cross Section Measurements for Hafnium Isotopes	2		
3 Nuclear Data Libraries and Data Evaluations						
3.1 European Activation File development			opean Activation File development	4		
	3.2	Val	idation and evaluation activities at Nexia Solutions	4		
	3.2.	.1	Validation of JEFF data	4		
3.2.2 Fission Product Yield evaluat		.2	Fission Product Yield evaluations	5		
3.2.3 Acknowledgement		.3	Acknowledgement	5		
	3.3	Dec	ay data evaluations at NPL	6		
	3.4	Dec	ay data evaluations at IAEA & Serco	6		
4	Analysis Codes					
	4.1	A st	tandardised version of the code REFIT	9		
5	Ref	feren	ces	10		
6	Ack	know	ledgements	11		
			Tables			
Ta	able 1.	Deca	ay data evaluated in 2006 / 07	7		
Т	able 2.	Deca	ay data evaluated in 2007 / 08	8		

NPL Report IR 14 UKNSF(2008)P224 INDC(UK)0092 Distr. G

1 Introduction

The United Kingdom Nuclear Science Forum (UKNSF) encourages technical discussions on the measurement and evaluation of nuclear data. Membership ranges across approximately 30 UK organisations. The Forum is funded by the Health & Safety Executive (HSE), and acts as the communication network for matters relating to the NEA Data Bank and the IAEA Nuclear Data Section.

During 2007, UKNSF members have assisted the NEA Nuclear Science Committee to formulate and progress programmes of work and to define priorities.

The Radioactivity and Neutrons Group at the National Physical Laboratory (NPL) provides the secretariat for the UKNSF, and the Forum's web site is at www.uknsf.org.uk. The Forum is chaired by R. A. Forrest of UKAEA Fusion, Culham, and the secretary is N. P. Hawkes of NPL.

The first UKNSF meeting of 2007 was held at the Durley Hall Hotel, Durley Chine Road, Bournemouth, on 24th May.

The second 2007 meeting was held at HSE London Office, Rose Court, on 23rd November.

2 Measurements

2.1 Radioactivity Metrology at NPL

Arzu Arinc, Chris Gilligan, Lena Johansson, John Keightley, Andy Pearce, John Sephton and Andy Stroak (Radioactivity & Neutrons Group, DQL, National Physical Laboratory)

2.1.1 Primary Radioactivity Standardisations

As in previous years, the National Physical Laboratory (NPL) has provided new primary measurements according to the needs of the user community. Primary standardisation activities are frequently coupled with nuclear decay data measurements as the attainment of precise activity standards coupled with high precision gamma spectrometry allows absolute emission probabilities to be determined.

Primary standards developed during 2007 include ⁵⁶Mn, ¹²⁴Sb, ²⁰¹Tl and ²¹⁰Pb. The standardisation of ⁵⁶Mn is of particular significance as it supports the UK's primary standard of neutron emission rate. The expected closure of the CONSORT reactor at Ascot has necessitated the investigation of techniques for standardising ⁵⁶Mn at much lower activities than were available from Ascot. In the final standardisation using material from Ascot, samples were counted by conventional techniques and also by liquid scintillation counting as proof of principle. Results were successfully submitted to the International Reference System (SIR) of the Bureau International des Poids et Mesures (BIPM), hence linking the UK standard into the international measurement system.

Primary standardisations of ¹⁰⁹Cd, ¹³⁴Cs and ^{166m}Ho are ongoing.

2.1.2 Equipment upgrades

The refurbishment of the counting facilities at NPL continued and 2007 saw the implementation of a new suite of data acquisition tools for coincidence counting. A new design of proportional counter amplifier has also been developed to reduce electronic noise and hence improve the performance of the proportional counter systems for low-energy electrons and X-rays.

2.1.3 Acknowledgement

Financial support from the National Measurement System Programme Unit of the U.K. Department for Innovation, Universities and Skills is acknowledged.

2.2 Neutron Cross Section Measurements for Hafnium Isotopes

T.C. Ware, M.C. Moxon (University of Birmingham); C.J. Dean (Serco)

The need for new neutron cross section measurement and evaluation is co-ordinated through the international High Priority Request List (HPRL) [1] maintained by the

OECD/NEA Working Party on International Nuclear Data Evaluation Co-operation (WPEC).

In the nuclear industry hafnium is used as a neutron absorbing material to regulate the fission process. The HPRL has identified that interpretations of critical experiments with UOx fuel, conducted by the CEA in the AZUR zero-power reactor, have shown systematic underestimation of the reactivity worth. This is attributed to an overestimated natural hafnium capture cross section in the epithermal energy range.

To rectify this, the UK has participated in time-of-flight measurements on natural hafnium samples at IRMM, Geel through the NUDAME project. The analysis of these measurements has begun with the objective of producing a new resonance range evaluation for the six natural hafnium nuclides. Measurements on oxide samples enriched in individual isotopes are planned. The evaluation is to be submitted to the JEFF project in 2009 and will form the PhD thesis of T. Ware.

A standardised version of Moxon's least squares fitting code REFIT (see §4.1) is to be used to derive the resonance parameters from the measurements.

3 Nuclear Data Libraries and Data Evaluations

3.1 European Activation File development

R. A. Forrest (UKAEA, Culham)

The Euratom/UKAEA Fusion Association has continued the development of the European Activation File (EAF) under the Nuclear Data Task of the EFDA Fusion Technology Programme. EAF covers the cross sections and decay data libraries that are required as input to the inventory code FISPACT.

The EAF-2007 libraries were released at the beginning of 2007 as part of the European Activation System (EASY-2007) [2]. The next task was validation of the library, which involved the comparison of library data with integral activation measurements in well-specified neutron spectra. The report for EASY-2007 has been released [3]. For each of 470 reactions, it gives graphs of differential data and diagrams showing C/E ratios (C being the effective cross section calculated from the library, and E the value deduced from experimental measurements). It is a large report covering 564 pages.

The use of experimental data, both integral and differential, to test EAF library data is very valuable. However, only ~1,800 reactions are supported by measurements, which is less than 3% of the entire library. The method of statistical analysis of cross sections (SACS) has therefore been used extensively to test the various reaction types in EAF-2007. A paper [4] gives examples of the findings for the important (n, p) and (n, α) reaction types.

Changes have been made to SAFEPAQ-II (the application used to produce the EAF libraries) to expand the various options available for the SACS tool. A major undertaking has been the updating of SAFEPAQ-II to use the modern versions of Visual Basic (VB.NET) and Access. It is expected that this rewriting will make the application easier to maintain.

The initial part of the work required to update the 'Activation Handbook' [5] has been completed. This involves the generation of importance diagrams for the stable elements using calculations made using EASY-2007. The lists of dominant nuclides and important reactions that can be extracted from these diagrams have been produced and will be used to define the future data needs for activation as regards neutron-induced cross sections and decay data. It is found that only about 3.5% of the reactions are classified as of 'major' importance and focussing on these both theoretically and experimentally will allow the next version of EAF to be produced efficiently.

3.2 Validation and evaluation activities at Nexia Solutions

R. W. Mills (Nexia Solutions Ltd.)

3.2.1 Validation of JEFF data

As part of NDA funded work within the Working Party on International Nuclear Data Evaluation Co-operation (WPEC) sub-group 25 on decay heat, a study [6] was

produced showing that it was possible to improve the results of JEFF-3.1 using recent Total Absorption Gamma-ray Spectrometry (TAGS) measurements. These new data have been incorporated by the CEA and IAEA into a new decay data library JEFF-3.1.1 released on 2nd July 2007 [7]. This updated library consists of decay data for the same 3852 nuclides as JEFF-3.1, but 59 nuclides have had their data updated with more recent and complete evaluations, 29 nuclides important for decay heat have had their mean gamma-ray and lepton energy release per decay adjusted to the Greenwood *et al.* TAGS measurements [8], and 4 errors reported on the JEFF project feedback website were corrected.

A review [9] has been produced which described checks on the consistency of the JEFF-3.1.1 file and repeated decay heat validation [10] using this new file and FISPIN. The validation has compared fission pulse decay heat measurements for 235 U, 238 U, 239 Pu and 241 Pu, and calorimetric measurements of 20 PWR fuel assemblies with cooling times between 2.4 and 8.2 years for irradiations between 25 and 40 GW d/t. The results of the consistency checks and the revalidation show that JEFF-3.1.1 is an improvement over JEFF-3.1 with the known major issues having been corrected. However, this review has shown some less important problems still remain. It is noted that results from the CEA [11] and Serco [12] show the same trends as this Nexia study.

A new study was also carried out using FISPIN with the JEFF-3.1 and the new JEFF-3.1.1 data, to compare calculated neutron emission with experimental measurements [13]. This showed good agreement between calculation and experiment, with little differences between JEFF-3.1 and JEFF-3.1.1. It was also shown that the new data increases the neutron emission for typical irradiated PWR fuel by ~3% from calculations using JEF-2.2 data. In addition, a review showed that only the ²³²U and ²³⁶Pu data differ significantly from the values currently in the Evaluated Nuclear Structure Data File (ENSDF) and these require a detailed review by a decay data evaluator.

3.2.2 Fission Product Yield evaluations

A review of ternary fission product yield measurements and new theoretical interpretations has shown that it should be possible to improve the yields in the JEFF-3.1 fission yield file, and it is hoped to produce an updated version of this file for the release of JEFF-3.2 expected in 2010.

Work has continued on improving the energy dependent fission yields in UKFY4.0 [14] and including gamma-ray induced fission and spontaneous fission yields. The new file UKFY4.1 is planned to be issued in 2008.

3.2.3 Acknowledgement

Financial support of the Nuclear Decommissioning Authority (NDA) is acknowledged.

NPL Report IR 14 UKNSF(2008)P224 INDC(UK)0092 Distr. G

3.3 Decay data evaluations at NPL

Arzu Arinc and Andy Pearce (Radioactivity and Neutrons Group, DQL, National Physical Laboratory)

NPL is participating in the International Atomic Energy Agency's Coordinated Research Programme *Updated Decay Data Library for Actinides*. Evaluations of ²³²Th and ²³²U are in progress with ²³¹Pa, ²²⁸Ac and ²²³Ra planned for the near future. The final Research Coordination Meeting is scheduled for October 2008 with publication shortly thereafter.

Evaluations of ¹⁰⁶Ru and ¹⁰⁶Rh for the Decay Data Evaluation Project (DDEP) are also being prepared by NPL.

Financial support from the National Measurement System Programme Unit of the U.K. Department for Innovation, Universities and Skills is acknowledged.

3.4 Decay data evaluations at IAEA & Serco

A. L. Nichols (IAEA), and R. J. Perry and C. J. Dean (Serco)

A continued programme of in-depth decay data evaluation work has been supported by British Nuclear Fuels (now Nexia Solutions) and the United Kingdom Atomic Energy Authority (UKAEA Fusion) at Culham. The aim is to improve the content of the Joint Evaluated Fission and Fusion File (JEFF) and European Activation File (EAF) for predicting composition and emissions from irradiated material in fission and fusion systems. The evaluated data improve and extend the contents of existing libraries, resulting in new versions of the UK Heavy Element (UKHEDD2.6 [15]) and Activation Product (UKPADD6.8 [16]) Decay Data files. Both databases have also been incorporated into the EVA database [17] that is held at the OECD NEA Data Bank.

The evaluated data are processed through the COGEND code [18] to yield complete and verified files in ENDF-6 format. Verification has included processing with the BNL codes CHECKR [19] and FIZCON [20], and confirming the overall consistency of the energy release by summing the emission by quanta and comparing with the total emission by decay mode. Modifications to UKHEDD have focussed only on formatting issues, rather than new decay data evaluations. Tables 1 and 2 are lists of the new evaluations for UKPADD together with any discrepancy in the energy balance, defined in terms of project years 2006 / 07 and 2007 / 08, respectively. The evaluated radionuclides are listed, together with their half-lives and the results of energy balance tests that compare the energy release summed over all modes of decay (calculated from Q-values and branching fractions) with the total energy of all individual emissions.

Table 1. Decay data evaluated in 2006 / 07 (See Ref. [21])

Radionuclide	Half-life	Consistency (% Deviation)
32-Ge-73m	0.50 (1) s	- 0.3557
33-As-73	80.30 (6) d	- 0.0344
33-As-76	26.17 (13) h	0.0406
34-Se-77m	17.55 (15) s	- 0.1025
38-Sr-87m	2.816 (11) h	- 0.0120
39-Y-87	79.8 (2) h	0.0077
39-Y-87m	13.37 (3) h	- 0.0408
45-Rh-101	3.2 (3) y	- 0.0854
45-Rh-101m	4.34 (1) d	- 0.0180
47-Ag-109m	39.7 (2) s	- 0.0051
66-Dy-166	81.6 (1) h	0.0847
68-Er-167m	2.270 (6) s	0.0743
69-Tm-167	9.246 (14) d	0.1193
69-Tm-168	90 (4) d	0.0759
75-Re-183	70.0 (14) d	0.2007
75-Re-183m	1.03(4) x 10 ⁻³ s	0.1037
75-Re-186	3.75 (3) d	0.0046
75-Re-186m	1.9 (2) x 10 ⁵ y	- 0.4061
76-Os-194	6.0 (2) y	- 0.1669
77-Ir-193m	10.54 (4) d	- 0.0699
78-Pt-195m	4.1 (1) d	0.2207
79-Au-195	186.094 (31) d	0.8595
79-Au-195m	30.5 (2) s	0.0121
80-Hg-194	444 (80) y	2.4780

Table 2. Decay data evaluated in 2007 / 08 (See Ref. [16])

Radionuclide	Half-life	Consistency (% deviation)
21-Sc- 45m	0.325 (4) s	-0.0903
31-Ga- 70	21.14 (5) m	-0.0002
32-Ge- 71	11.43 (2) d	-0.0402
32-Ge- 71m	0.02040 (18) s	-0.0361
32-Ge- 75	82.78 (4) m	-0.0284
32-Ge- 75m	48.0 (9) s	-0.0330
34-Se- 79	3.77 (40) x 10 ⁵ y	0.0000
39-Y - 90	64.00 (21) h	0.0089
39-Y – 90m	3.19 (6) h	-0.0020
40-Zr- 90m	0.8082 (20) s	-0.0421
41-Nb- 90	14.59 (5) h	-0.0108
41-Nb- 90m	18.81 (9) s	0.0185
41-Nb- 90n	0.00622 (8) s	0.0158
48-Cd-113	7.8 (3) x 10 ¹⁵ y	0.0000
48-Cd-113m	14.6 (1) y	0.0000
50-Sn-121	26.99 (10) h	0.0000
50-Sn-121m	43.9 (5) y	-0.0914
66-Dy-166	81.6 (2) h	0.0542
69-Tm-170	128.6 (3) d	0.0041
70-Yb-176m	11.4 (3) s	-0.0071
75-Re-184	35.4 (5) d	0.0468
76-Os-189m	5.81 (10) h	-0.2464
78-Pt-190	6.5 (3) x 10 ¹¹ y	-0.0157

4 Analysis Codes

4.1 A standardised version of the code REFIT

T.C. Ware, M.C. Moxon (University of Birmingham); C.J. Dean (Serco)

A standardised version of Moxon's least squares fitting code REFIT [22] is to be used to derive the resonance parameters from the hafnium measurements described in §2.2. This has been produced, together with associated documentation and test cases, and delivered to the NEA Data Bank for open release [23].

5 References

- 1. See web site http://www.nea.fr/html/dbdata/hprl/index.html
- 2. R. A. Forrest, *The European Activation System: EASY-2007 overview*, UKAEA FUS 533, March 2007.
- 3. R. A. Forrest, J. Kopecky, M. Pillon, A. Klix, S.P. Simakov, J.-Ch. Sublet, P. Bém, M. Honusek and E. Šimecková, *Validation of EASY-2007 using integral measurements*, UKAEA FUS 547, April 2008.
- 4. R. A. Forrest, J. Kopecky and A. J. Koning, *Detailed analysis of (n, p) and (n, α) cross sections in the EAF-2007 and TALYS-generated libraries*, Fus. Eng. Design **83** (2008) 634-643.
- 5. M. R. Gilbert and R. A. Forrest, *Handbook of activation data calculated using EASY-2003*, UKAEA FUS 509, July 2004.
- 6. R. J. Perry and C. J. Dean, Assessment of the Impact of TAGS on Gamma Decay Heat for JEFF3.1 ²³⁵U and ²³⁹Pu, JEF/DOC-1126 and UKNSF(2006)P199.
- 7. M. A. Kellet, *The JEFF-3.1.1 Decay Data Library*, JEF/DOC-1188, 2007.
- 8. R. C. Greenwood, R. G. Helmer, M. H. Putnam and K. D. Watts, *Measurement of β-decay intensity distributions of several fission-product isotopes using a total absorption γ-ray spectrometer*, Nucl. Instr. Methods in Phys. Res. **A390** (1997) 95 154. (Method described in R. C. Greenwood, R. G. Helmer, M. A. Lee, M. H. Putnam, M. A. Oates, D. A. Struttmann and K. D. Watts, *Total absorption gamma-ray spectrometer for measurement of beta-decay intensity distributions for fission product radionuclides*, Nucl. Instr. Methods in Phys. Res. **A314** (1992) 514 540).
- 9. R. Mills, Testing of the JEFF-3.1.1 decay data file for consistency and comparison with JEFF-3.1 results, JEF/DOC-1220 and UKNSF(2007)P222.
- 10. R. W. Mills, *Technical note on validation of JEFF-3.1 for decay heat and inventory calculations*, JEF/DOC-1147 and UKNSF(2006)P207.
- 11. T.-D. Huynh, *Comparisons between JEFF-3.1 and JEFF-3.1.1 with Decay Heat Calculations*, JEF/DOC-1219.
- 12. C. Dean, Gamma Decay Heat Prediction for JEFF3.1.1 ²³⁵U and ²³⁹Pu, JEF/DOC-1218 and UKNSF(2007)P223.
- 13. R. Mills and C. Shearer, *Technical note on using JEFF-3.1 and JEFF-3.1.1 data to calculate neutron emission from spontaneous fission and (α, n) reactions with FISPIN*, Nexia Solutions Report (07) 8088, Issue 2. Also JEF/DOC-1201 and UKNSF(2007)P221.
- 14. R. W. Mills, *Technical note on producing energy dependent fission product yield files*, UKNSF(2006)P210 and JEF/DOC-1157.

- 15. A. L. Nichols and R. J. Perry, *Heavy Element and Actinide Decay Data: UKHEDD2.6*, SERCO/TAS/000343/W002 Issue 1, UKNSF(2008)P225, and JEF/DOC-1224, February 2008.

 (See also: http://www.nea.fr/dbforms/data/eva/evatapes/ukhedd_26/)
- 16. A. L. Nichols and R. J. Perry, *Activation Product Decay Data: UKPADD6.8*, SERCO/TAS/000343/W003 Issue 1, UKNSF(2008)P226, and JEF/DOC-1227, March 2008.

 (See also: http://www.nea.fr/dbforms/data/eva/evatapes/ukpadd_68/)
- 17. See web site http://db.nea.fr/html/dbdata/eva/evaret.cgi
- 18. R. J. Perry and C. J. Dean (originally A. Tobias), COGEND: A Computer Code to Generate Nuclear Decay Scheme Data in ENDF-6 Format, UKNSF(2004)P190, JEF/DOC-1083.
- 19. C. L. Dunford, *ENDF Utility Codes Release 6.12*, International Atomic Energy Agency, 2001
- 20. R. J. Perry, M. J. Grimstone and C. J. Dean, *Specification and Maintenance Document for Decay Data Checking in FIZCON*, JEF/DOC-959 and UKNSF(2003)P160, March 2003.
- 21. A. L. Nichols and R. J. Perry, *Assessment and Evaluation of Decay Data for EAF* 2006/07, UKNSF(2007)P214 and JEF/DOC-1167, March 2007.
- 22. M. C. Moxon, T. C. Ware, C. J. Dean and others, *REFIT-2007 A Least-Square Fitting Program for Resonance Analysis of Neutron Transmission, Capture, Fission and Scattering Data. Users' Guide for REFIT-2007-08*, UKNSF(2007)P216, August 2007.
- 23. See web site http://www.nea.fr/abs/html/nea-0914.html

6 Acknowledgements

This report was produced with the support of the HSE Nuclear Directorate, and represents edited contributions from members of the UK Nuclear Science Forum and their co-workers.

The editor is grateful to David Thomas for his helpful comments on the manuscript.