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United Kingdom Nuclear Science Forum

Progress Report

Data Studies During 2005 and 2006

Edited by N P Hawkes

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ABSTRACT

The United Kingdom Nuclear Science Forum (UKNSF) meets one or more times 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.npl.co.uk/uknsf). 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.

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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 has the support of the Health & Safety Executive, and acts as the communication network for matters relating to the NEA Data Bank and the IAEA Nuclear Data Section.

UKNSF members have assisted the NEA Nuclear Science Committee during these two years to formulate and progress programmes of work and to define priorities.

The Radioactivity and Neutrons Group at the National Physical Laboratory provides the secretariat for the UKNSF, and the Forum's web site is at www.npl.co.uk/uknsf. The Forum is chaired by R. A. Forrest, UKAEA Fusion, Culham.

The 2005 meeting of the UKNSF was held at HSE London Office, Rose Court, on 16th June (Chairman, R. A. Forrest, UKAEA; Secretary, T. D. MacMahon, NPL).

The 2006 meeting was held at the same venue on 1st June (Chairman, R. A Forrest, UKAEA; Secretary, N. P. Hawkes, NPL).

2 Measurements

2.1 Radioactivity Metrology and Radionuclide Decay Data

Arzu Arinc, Seán Collins, Lena Johansson, Andy Pearce and Andy Stroak (Radioactivity & Neutrons Group, DQL, National Physical Laboratory)

2.1.1 International Comparison

An international comparison of ^{55}Fe activity concentration has been coordinated by the National Physical Laboratory (NPL) under the auspices of the Bureau International des Poids et Mesures (BIPM). ^{55}Fe is significant in nuclear site decommissioning, and presents difficulties in standardisation due to the low energy of the emissions. The samples have been dispatched to the participants and responsibility for the project has reverted to the BIPM for data collection and publication.

2.1.2 Euromet projects

Euromet project 721 “Activity Measurements and Gamma Emission Intensities Determination in the Decay of ^{65}Zn” has been completed. The specific activity and X- and γ- emission probabilities were measured by nine participating laboratories led by CEA Saclay and including NPL. New emission probabilities have been published [1], and have helped to resolve discrepancies observed by the BIPM in the Système International de Référence (SIR) [2].

Euromet project 591 “Alpha Particle Emission Probabilities in the Decay of ^{235}U” has been completed and was led by the Research Centre for Energy, Environment and Technology (CIEMAT) of Spain. The alpha spectra were measured at CIEMAT and the Institute for Reference Materials and Measurements (IRMM) with the University of Extremadura, and NPL was involved in the data analysis. New data for alpha emission probabilities of ^{235}U have been published [3] with an order of magnitude improvement in precision compared with current evaluated data in ENSDF.

2.1.3 Primary Radioactivity Standardisations

As in previous years, 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 allows absolute emission probabilities to be determined.

Use of ^{99m}Tc accounts for more than 80 % of the diagnostic nuclear medical procedures delivered to patients in the UK. The primary standard of this radionuclide for the UK was replaced in 2005. A ^{99m}Tc solution was standardised at NPL and used to calibrate ionisation chambers that, in turn, are used to provide calibration services to nuclear medicine departments. A sample of the standardised solution was submitted to the BIPM for inclusion in the SIR.

During the period 2005-2006 primary standards were also prepared of ^{54}Mn, ^{56}Mn, ^{125}I, ^{210}Pb, ^{64}Cu, ^{89}Sr and ^{201}Tl. Standardised samples of ^{54}Mn, ^{125}I and ^{201}Tl were submitted to the BIPM for inclusion in the SIR.

2.1.4 Equipment upgrades

In addition to the normal programme of standardisations, considerable effort has been invested in updating some of the measurement systems at NPL. The detector mounts for two high resolution germanium spectrometers have been redesigned to improve the mechanical rigidity, allowing today's larger, heavier crystals to be used without compromising the reproducibility of the source-detector geometry.

Kinematic mounts and optical benches have been used to ensure samples can be positioned reproducibly within the calibration geometry. A Kinematic mount allows a sample to be removed and replaced whilst controlling movement in six degrees of freedom. A new calibration geometry for the thin-film sources used in coincidence counting has also been introduced. This has advantages in the measurement of gamma emission probabilities, because the same source assayed by coincidence counting to determine absolute activity can now also be assayed by high resolution gamma spectrometry to determine photon emission rates. Previously two different sources were used for the two assays, introducing uncertainties due to weighing, adsorption to glassware and self-absorption corrections.

2.1.5 Acknowledgement

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

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.

3.1.1 Developments during 2005

An important part of the work on EAF involves the preparation of a validation report for each library released. This involved the comparison of library data with integral activation measurements in well-specified neutron spectra. The report for EASY-2005 has been released [4]; this covers 453 reactions giving 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 535 pages.

A new method of testing large data libraries, especially those containing many reactions with no experimental support, has been developed. Statistical analysis of cross sections (SACS) [5] involves plotting quantities such as the maximum cross section of a reaction (smax) against mass number (A). These scatter plots show well defined trends and any deviations indicate reactions that merit further investigation.

Using initial results from the integral validation and SACS it was found that there were some reactions in EAF-2005 that contained errors. This lead to the production of the maintenance release EAF-2005.1 which improved data for 250 reactions. This release also gave an opportunity to distribute a library of data for deuteron-induced reactions. This new library was motivated by the need to do activation calculations for IFMIF, a planned accelerator-based facility that will be used to test materials under intense irradiation of neutrons.

Changes to FISPACT and SAFEPAQ-II (the application used to produce the EAF libraries), to treat data for deuteron-induced reactions, have been made. Further analysis of the large number of activation calculations (using EASY-2003) presented in the ‘Activation Handbook’ has resulted in two papers [6, 7] presenting the data needs for activation as regards neutron-induced cross sections and decay data. It is hoped that these will allow attention, both theoretical and experimental, to be focused on the data most relevant for fusion applications.

3.1.2 Developments during 2006

Work on the development of the EAF-2007 libraries has been completed [8]. In addition to the neutron- and deuteron-induced cross section libraries released as EAF-2005.1, the new version also contains a proton-induced data library [9]. The charged particle data are needed for IFMIF, as mentioned above. IFMIF will use deuteron beams (protons may also be used during testing) which can cause activation in the accelerator and the target regions. The deuteron library in EAF-2007 has been improved by renormalizing some reactions to experiments and using new data sources.

The neutron-induced library has been improved using results from the EASY-2005 validation report [4] and the Statistical Analysis of Cross Sections (SACS) [5] method. Some additional targets are included so that all nuclides with $T_{½} > 6$ hours have cross section data. All cross section data extend to 60 MeV; this energy is chosen to allow activation calculations on IFMIF. In addition to the cross sections the decay library has been significantly improved with more nuclei (2,231 compared to 2,192 in EAF-2005) and use of the new JEFF-3.1 decay data.

The EASY User Interface has been extended so that cross section data from all three incident particle libraries can be viewed. Changes to FISPACT and SAFEPAQ-II (the application used to produce the EAF libraries) have been made; one extension allows data for proton-induced reactions to be treated. Work has started on the collection of data for the integral validation of EASY-2007 which will be carried out next year.

3.2 Fission Product yield evaluation, 2005 - 2006

R. W. Mills (Nexia Solutions Ltd.)

3.2.1 UKFY3.6 fission product yield file

In February 2005, the UKFY3.6 fission product yield file was issued. This is a modification of the UKFY3.5 file produced in 2004, which was developed from UKFY3 [10]. It included the Serot and Wagemans tritium and alpha yields [11], data from several newly available results supplied by JEFF members [12, 13, 14], and a detailed review of data for important nuclides and models (resulting in improved yields and charge balance). The cumulative yields were generated with an improved method to estimate the cumulative yield errors using the February 2005 decay data produced by the CEA for JEFF-3.1. It should be noted that the only change to this decay data before being released as part of JEFF-3.1 did not affect the fission product decay chains and thus did not require a repeated calculation of the fission yields.

Following a review of the new fission product yields in March by the CEA, several issues were raised. These issues were reviewed within the JEFF committees and two agreed changes were made to the file (the U235T Cs137 and the Pu239T Nd148 cumulative yields and their immediate precursors were adjusted). The revised file was issued in April 2005 as UKFY3.6A. This file was subsequently released by the Nuclear Energy Agency for testing purposes as the fission product yield component of JEFF-3.1.

3.2.2 Validation of JEFF-3.1 decay and fission product yield files

The new decay and fission product yield files of JEFF-3.1 were used in validation exercises with fission pulse decay heat measurements, PWR assembly heat measurements and post-irradiation examination analysis for four Gösgen samples [15, 16]. In addition, at the 2005 UKNSF meeting it was decided that to produce consistent results, the cross-sections generated via the WIMS reactor physics code for the FISPIN inventory calculations should be based upon JEFF-3.1 data. Serco Assurance was subsequently contracted to produce a WIMS9A library using the major nuclides (those carried through to the FISPIN inventory code) from JEFF-3.1, with funding from the Nuclear Decommissioning Authority (NDA).

In 2006, with the new WIMS9A library now available, the validation exercises were repeated. The results [17] were presented to the UKNSF and the JEFF Scientific Co-ordination Group. The new validation showed an improvement for total assembly decay heat but still showed differences against the beta and gamma-ray heating from the work of Tobias and others [18, 19, 20, 21]. The same differences were found by calculations from the CEA and others [22, 23]. As part of work with the WPEC sub-group 25 on decay heat, Serco Assurance presented an NDA-funded study [24] that showed it was possible to improve the results using the recent Total Gamma-ray Absorption Spectrometry measurements (TAGS). It is planned to include these new data in a new release of JEFF-3.1 (provisionally identified as JEFF-3.1.1) early in 2007, after which it is planned to repeat this validation.

3.2.3 Tritium production

In generating the new JEFF-3.1 libraries, it was found that new fission yield data for ^{6}Li production combined with the cross-sections lead to an increase in the production of tritium. A detailed study was carried out [25] that showed that the new data increased the calculated tritium production from spent fuel by about 20%, although with large uncertainties. Additionally using a simple model to estimate remaining missing data could lead to a further 10% increase. It is hoped to carry out a study to try to improve the uncertainties on the existing ^{6}Li yields and consider better models to estimate missing data next year.

3.2.4 Energy-dependent fission yields

The UK has continued international collaboration with colleagues in the USA, China, France, Germany, Japan, Netherlands and Russia, through the forum of the IAEA Coordinated Research Programme on *Fission product yield data required for transmutation of minor actinide nuclear waste*. Work by a US participant, A.C. Wahl [26], allows energy dependent effects on fission yields to be approximated, and in 2006 a set of yield files was produced [27] in ENDF/B format using Wahl's CYFP code. The scope of this work was all 21 fissioning systems in JEF-2.2 and JEFF-3.1 (^{232}Th, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm and ^{252}Cf) for fissions induced by neutrons, protons, deuterons and alpha particles between 10^{-5} eV and 150 MeV. These preliminary files have been identified by the name UKFY4.0 (Issue 1) and are intended as a proof of principle. It is hoped next year to extend the results to gamma-induced fission and for a wide range of spontaneously fissioning nuclides.

3.2.5 Acknowledgement

Financial support of the NDA is acknowledged.

3.3 Decay data evaluations at NPL

Arzu Arinc, Desmond MacMahon, 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*. At the 1st Research Coordination Meeting in October 2005, 94 radionuclides were identified as being of interest, of which 58 were selected as the highest priorities for evaluation and assigned to participants. NPL will provide evaluations for ^{232}U, ^{232}Th, ^{231}Pa, ^{228}Ac and ^{223}Ra. It was agreed that the analysis and checking procedure would be that of the Decay Data Evaluation Project (DDEP), and that the new results would be included in the DDEP as they become available.

Evaluations of three of these radionuclides were progressed in 2006 although none have yet reached the stage where they can be submitted for review. The final Research Coordination Meeting is scheduled for October 2008 with publication shortly thereafter.

In March 2006 a workshop was held by CEA Saclay (France) with the aim of providing training for a new generation of decay data evaluators. Experienced evaluators gave lectures on evaluation and statistical analysis of data. There were also practical sessions and presentations from students. Three NPL staff attended, one as a lecturer and two as students. The full report of the workshop can be found at the DDEP web page, www.nucleide.org/DDEP.htm.

Evaluations of ^{106}Ru and ^{106}Rh for the 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 Vienna & Serco Winfrith

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

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) 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 extend existing libraries resulting in new versions of the United Kingdom Heavy Element and Activation Product Decay Data files (UKHEDD2.4 and UKPADD6.6). These have been installed within the EVA database at the OECD NEA Data Bank (http://db.nea.fr/html/dbdata/eva/evaret.cgi).

The evaluated data are processed through the COGEND code [28] to yield complete and verified ENDF6 format files. The verification includes processing with the BNL codes CHECKR [29] and FIZCON [30] and includes confirming the overall consistency of the energy release by summing the emission by quanta and comparing with total emission by decay mode. The following tables list the new evaluations together with any discrepancy in the energy balance. The first table lists the nuclides developed in the UKHEDD library. The remaining tables show UKPADD developments by year.

Table 1. Actinide evaluated decay data

Radionuclide	Half-life	Consistency (% Deviation)
80-Hg-206	8.31m	0.061
81-Tl-206	4.202m	-0.005
81-Tl-208	3.060m	0.0088
81-Tl-210	1.3m	0.0191
82-Pb-210	22.160y	-0.4568
82-Pb-212	10.640h	0.0931
82-Pb-214	26.8m	0.0263
83-Bi-210	5.012d	0
83-Bi-212	1.009h	-0.0021
83-Bi-212m	25.0m	-0.0437
83-Bi-212n	9.0m	0
84-Po-210	138.388d	-0.0014
84-Po-212	3.0E-07s	-0.0013
84-Po-212m	45.1s	-.0.0013
84-Po-214	1.637E-04s	-0.0013
84-Po-216	0.150s	-0.0009
84-Po-218	3.098m	-0.0014
85-At-218	1.5s	-0.0012
86-Rn-218	0.035s	-0.0006
86-Rn-220	55.8s	-0.0011
86-Rn-222	3.823d	-0.002
88-Ra-224	3.640d	0.001
88-Ra-226	1.60496E+03y	0.0013
90-Th-228	1.913y	-0.0175
90-Th-230	7.54015E+04y	0.199
91-Pa-232	1.310d	-0.3363
92-U-235m	26.0m	0
93-Np-236	1.52003E+05y	0.0884
93-Np-240m	7.4m	-0.4742
94-Pu-238	87.7y	0.0130
94-Pu-239	2.41135E+04y	-0.0376
94-Pu-240	6.563E+03y	0.0077
94-Pu-241	14.330y	-0.0012
97-Bk-249	320d	-0.4985

Table 2. Radionuclides evaluated in 2002 - 04

Nuclide	Half-life	Consistency (% deviation)
34-Se-81	18.39 m	0.0027
34-Se-81m	57.28 m	0.1410
35-Br-72	78.6 s	-0.2821
35-Br-72m	10.6 s	0.2164
38-Sr-94	75.3 s	0.0259
40-Zr-99	2.2 s	-0.0510
45-Rh-110	28.5 s	0.0172
45-Rh-110m	3.2 s	0.1721
74-W-176	2.5 h	0.1347
76-Os-180	21.5 m	0.0346
76-Os-196	34.9 m	-0.0177
77-Ir-192	73.822 d	0.1323
77-Ir-192m	1.44 m	-0.0780
77-Ir-192n	241 y	0.0069
78-Pt-202	44 h	0.0000
79-Au-192m[#]	0.160 s	0.1114

#The recognised decay data for the 29 ms nuclear level of Au-192 have been included as part of the decay data for Au-192m.

Table 3. Radionuclides evaluated in 2004 - 05

Nuclide	Half-life	Consistency (% Deviation)
32-Ge- 80	27.0 s	-0.0204
39-Y - 97	3.75 s	0.0105
39-Y - 97m	1.17 s	-0.0227
39-Y - 97n	0.142 s	-0.0264
45-Rh-111	12.0 s	0.0125
46-Pd-113	91.0 s	-0.1194
46-Pd-113m	0.3 s	0.3404
50-Sn-129	2.23 m	-0.0200
50-Sn-129m	7.2 m	-0.0530
50-Sn-130	3.73 m	-0.0085
50-Sn-130m	1.7 m	0.0458
55-Cs-123	5.91 m	-0.0424
55-Cs-123m	1.7 s	-0.0020
58-Ce-149	5.3 s	0.0777
61-Pm-155	41.5 s	incomplete
64-Gd-163	68.0 s	0.0550
65-Tb-146	8.0 s	-0.0027
65-Tb-146m	24.0 s	-0.1118
68-Er-167m	2.269 s	-0.0144

Table 4. Radionuclides evaluated in 2005 - 06

Nuclide	Half-life	Consistency (% Deviation)
9-F-21	4.158(20) s	– 0.0153
17-Cl-39	55.6(2) m	0.0666
30-Zn-69	56.4(7) m	0.0000
30-Zn-69m	13.78(5) h	– 0.0071
41-Nb-96	23.35(5) h	0.0239
65-Tb-158	180(11) y	– 0.0411
65-Tb-158m	10.8(2) s	0.1545
65-Tb-160	72.3(2) d	0.0185
69-Tm-171	700(5) d	0.0017
69-Tm-172	63.6(2) h	0.1605
72-Hf-179m	18.67(3) s	0.0568
72-Hf-179n	25.1(2) d	0.0382
74-W-188	69.78(5) d	0.0013
75-Re-184	37.9(5) d	0.0468
75-Re-184m	168(8) d	0.1530
76-Os-191	15.3(3) d	0.0000
76-Os-191m	13.10(5) h	– 0.0030
77-Ir-191m	4.90(3) s	0.0772

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