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UK Chemical Nuclear Data Committee Progress Report: Data Studies During 1990

Edited by

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AEA TECHNOLOGY

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UK CHEMICAL NUCLEAR DATA COMMITTEE: DATA STUDIES DURING 1990

Edited by

A L NICHOLS

SUMMARY

Basic nuclear data requirements for commercial and industrial application are monitored by the UK Chemical Nuclear Data Committee (UKCNDC), including half-lives, decay data and fission yields. Summary reports of the work undertaken within this technical area are included in this document for information. Funding problems continue to plague the experimental and evaluation studies, and are seriously jeopardising the viability of radiochemistry/nuclear physics programmes within the UK.

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1. INTRODUCTION

The UK Chemical Nuclear Data Committee provides a suitable forum within the UK to monitor the measurement and evaluation of radionuclide decay data and fission product yields for reactor applications. Two meetings of the UKCNDC were held in 1990 at the beginning of July and December (Chairman, A L Nichols (WTC) and Secretary, P Robb (WTC)). The UK Data Library Sub-Committee (DLSC) also met twice over the same time period (Chairman, M F James (WTC) and Secretary, P Robb (WTC)). Members of the DLSC are responsible to the UKCNDC for the maintenance of the following computerised libraries:

(a) fission product yields,
(b) fission product decay data,
(c) activation product decay data,
(d) heavy element and actinide decay data.

Much of the progress during the year has involved the evaluation of fission yields and activation product decay data based on partial funding from Nuclear Electric and BNFL plc. All of this evaluation effort is linked to the international Joint Evaluation File (JEF).

The requirements and priorities for nuclear data measurements and evaluations are regularly reviewed within the UKCNDC. This exercise was undertaken in 1989, and the resulting UKCNDC Request List was published in March 1990 (AEEW-M 2613).

2. MEASUREMENTS

2.1 Measurement and Evaluation of Decay Data (P Christmas, D Smith, M J Woods, J L Makepeace, J P Sephton, D H Woods, S A Woods, G R Worthington, J C J Dean, S M Jerome, S E M Lucas, C W A Downey and M T Curry (National Physical Laboratory, Teddington), and R D Daniels (University of Manchester)).

Specific measurements have been undertaken or are underway:

(a) Measurement of the half-life of ^{90}Sr continues.

(b) Measurements of half-lives are in progress for radionuclides that are not known with sufficient accuracy, viz ^{3}H, ^{54}Mn, ^{57}Co, ^{65}Zn and ^{75}Se; measurements for ^{56}Co and ^{58}Co have been completed, and related work on ^{125}I and ^{137}Cs has been published (1, 2).

(c) New measurements are underway to relate activity, ion-chamber current and air-kerma rate of ^{192}Ir brachytherapy sources.

(d) ^{154}Eu samples of >98% purity have been produced from ^{153}Eu separated on the electromagnetic isotope-separator and standardised. Independent measurements of the emission probabilities of the principal gamma-rays have been made.

(e) A chemical method has been devised to separate ^{125}Sb from daughter ^{125}Te. The ^{125}Sb has been standardised and independent measurements of the emission probabilities of the principal gamma-rays have been made.

(f) The SERC-supported collaboration with the University of Manchester has continued in conjunction with Argonne National Laboratory, USA. Measurements of the internal conversion electron spectrum following the decay of ^{245}Cm are progressing.

(g) Measurements of tritium-in-nitrogen using the new gas counting equipment are continuing; an international intercomparison of tritium-in-nitrogen is planned.

(h) A primary standard of ^{222}Rn has been established and verified by intercomparison with the standard held at NIST.

Various papers have been published during 1990 based on work undertaken in previous years, including ^{201}Tl, ^{237}Np and ^{244}Cm (3, 4, 5). Draft reports have also been prepared on the decay schemes of ^{124}I and ^{241}Am (6, 7).

2.2 <u>Nuclear Data Measurements</u> (A J Fudge, M F Banham, R A P Wiltshire and I Jackson (Harwell Laboratory))

Efforts have continued to improve the spectroscopic measurement facilities (which include a full range of alpha-particle and gamma-ray counters and spectrometers) and the codes for the analysis of spectral data. Accreditation of the laboratory under the National Physical Laboratory NAMAS scheme has continued for alpha particle measurements, and several gamma-ray spectrometry systems are subject to strict quality control procedures.

Work has continued in the following areas:

(a) <u>Calibration of ^{93m}Nb</u>

A solution of ^{93m}Nb has been prepared from pure niobium metal irradiated for a very long time in the Dounreay Fast Reactor (DFR). The calibration of this solution has been carried out simultaneously at Harwell, NPL and PTB, Braunschweig, Germany. The results are currently being evaluated prior to the solution being made generally available for use as a standard for fast neutron dosimetry.

(b) <u>Source preparation for nuclear data measurements</u>

The production of sources has continued for a range of actinide and transactinide nuclides for use in other laboratories. This work included the preparation of ^{237}Np sources for alpha-particle studies (8).

(c) <u>Decay scheme and half-life of ^{237}Np</u>

Work was completed at Imperial College and Harwell by a CASE student (funded and partially supervised by Harwell) on the low-energy gamma-ray spectum and half-life of ^{237}Np. A report of these studies is now being prepared.

(d) <u>Decay scheme of ^{239}Np</u>

Measurements of the gamma-ray spectrum of ^{239}Np (derived from ^{234}Am) have been completed by the Harwell-funded CASE student at Imperial College. The results of these measurements are being evaluated.

2.3 **Measurements and Evaluations of Half-Lives and Gamma-Ray Emission Probabilities** (M Hammed, I M Lowles, T D MacMahon and M U Rajput (Imperial College Reactor Centre, Ascot))

(a) ^{237}Np half-life (in collaboration with AEA Technology, Harwell Laboratories): the specific activity of ^{237}Np has been determined as (26.03 ± 0.20) Bq/µg, which corresponds to a half-life of (2.143 ± 0.016) x 10^6 years, in excellent agreement with the 1960 value of Bauer et al [9].

(b) The emission probabilities of low-energy gamma-rays from ^{237}Np have been determined, although, these new data still do not permit a consistent ^{237}Np decay scheme to be constructed from the available alpha-particle, conversion electron and gamma-ray data.

(c) Gamma-ray emission probabilities in the decay of ^{239}Np have been determined with sources originating from both the alpha decay of ^{243}Am and the beta decay of ^{239}U.

(d) Absolute gamma-ray emission probabilities have been measured for the 411.8, 675.9 and 1087.7 keV transitions in ^{198}Au, and are in good agreement with the evaluation of Nichols and Robb [10]. Using available internal conversion coefficients, the beta branching ratios to the two excited states of ^{198}Hg at 411.8 and 1087.7 keV have been deduced.

(e) Relative gamma-ray emission probabilities have been determined in the decay of ^{154}Eu. These data have been used with previously published gamma-ray, conversion electron and beta branching ratio data to produce a self-consistent ^{154}Eu decay scheme, leading to an absolute gamma-ray emission probability of 0.3479 ± 0.0014 for the 1274.5 keV transition.

(f) A new technique for evaluating discrepant data has been developed and applied to the 'problem' half-lives of ^{90}Sr, ^{137}Cs and ^{252}Cf.

3. UKCNDC DATA LIBRARY SUB-COMMITTEE

Membership during 1990:

M F James	WTC (Chairman)
P Robb	WTC (Secretary)
A J Fudge	Harwell Laboratory
T D MacMahon	Imperial College, Ascot
R W Mills	BNFL plc/University of Birmingham/WTC
A Whittaker	BNFL plc
D R Weaver	University of Birmingham

3.1 Data Library Development

The current status of the UKCNDC Data Libraries is summarised in Table 1. Significant progress has been made in the evaluation of fission yield and decay data. Efforts have also been made during the year to incorporate these data into the UK and JEF files. ENDF/B-VI format has been identified as the most appropriate form for the data files in the 1990s, and the COGEND data-processing code has been modified to generate such a format for the radionuclide decay data.

(a) Fission Product Decay Data (A L Nichols (WTC))

No progress has been made in producing an updated version of the UKCNDC fission product decay data files.

(b) Activation Product Decay Data (A L Nichols and P Robb (WTC))

Considerable progress has been made in evaluating the decay data for a wide range of activation products and generating the UKCNDC data files (10). The contents of this library are based on a subset of the listing in reference 11. Decay data have been evaluated for 113 radionuclides, and these data sets have been generated in ENDF/B format (Table 2). Some of the measured data for specific radionuclides proved insufficient to produce a consistent decay scheme, and additional parameters had to be derived (eg beta-branching fractions and theoretical internal conversion coefficients).

(c) Heavy Element and Actinide Decay Data (A L Nichols (WTC))

COGEND output data in ENDF/B format have been generated for the 4n series of radionuclides. Other specific actinides were also included in this exercise (eg Pa-321, U-234 and U-239) to improve the resulting data library and produce UKHEDD-2. Mean alpha, beta and gamma energies were derived for the 4n series (Table 3); the recommended data were rigorously tested for completeness, and excellent consistency was achieved (Table 4).

TABLE 1

UKCNDC DATA LIBRARIES: STATUS TABLE - DECEMBER 1990

Data	Present Studies	File Development
1 Fission Product Decay Data	Part of JEF-1 Decay Data Files, which include UK evaluations (ENDF/B-V format). UKFPDD-2 library (ENDF/B-IV format) superseded, but largely included in JEF-1.	P_n values for delayed neutron precursors from Swedish evaluation. Delayed neutron precursor spectrum from Kratz et al and Rudstam et al.
2 Activation Product Decay Data	UKPADD-1 part of JEF-1 Decay Data Files, which include UK evaluations (ENDF/B-V format).	113 nuclides have been evaluated for UKPADD-2 in ENDF/B-VI format. ^{51}Cr, ^{65}Zn, ^{75}Se and ^{198}Au have also been re-evaluated as part of an IAEA programme and they are also available in ENDF/B-VI format. All evaluations will be submitted to JEF.
3 Heavy Element and Actinide Decay Data	Part of JEF-1 Decay Data Files, which include UKHEDD-1 evaluations (ENDF/B-V format).	^{236}Np, ^{236m}Np, ^{236}Pu, ^{232}U, ^{228}Th, ^{224}Ra, ^{220}Rn, ^{216}Po, ^{212}Pb, ^{212}Bi, ^{212m}Bi, ^{212n}Bi, ^{212}Po, ^{212m}Po ^{212n}Po and ^{208}Tl evaluations completed. ^{231}Pa, ^{234}U, ^{239}U and ^{243}Am decay data and ^{242m}Am half-life have been evaluated.
4 Fission Yields	JEF-1 fission yields based on UKIFY-1 evaluation. UKIFY-2 (Mills and James) now complete and submitted to JEF-2: 39 sets of evaluated independent yields for 21 fissile nuclides adjusted to satisfy conservation of nucleons and charge	Major report on evaluations leading to UKIFY-2 in preparation by James, Weaver and Mills. Weaver and James have recommended integral yields, group data and spectra for delayed neutrons.

Spectral data from the decay data files may be accessed via the retrieval system described by Tobias (RD/B/5170N81, 1981).

TABLE 2

ACTIVATION PRODUCTS FOR UKPADD-2

Status: E, evaluated
NE, to be evaluated

File No.	Radionuclide	Status	File No.	Radionuclide	Status
7000	3-H	E	7023	26-Na	E
7001	6-He	E	7024	27-Mg	E
7002	8-He	E	7025	28-Mg	E
7003	8-Li	NE	7026	26-Al	E
7004	9-Li	NE	7027	26m-Al	E
7005	7-Be	E	7028	28-Al	E
7006	8-Be	NE	7029	29-Al	E
7007	10-Be	E	7030	30-Al	E
7008	11-Be	E	7031	31-Si	E
7009	12-B	E	7032	32-Si	E
7010	13-B	E	7033	32-P	E
7011	14-C	E	7034	33-P	E
7012	15-C	E	7035	34-P	E
7013	13-N	E	7036	35-S	E
7014	16-N	E	7037	37-S	E
7015	19-O	E	7038	34-Cl	E
7016	18-F	E	7039	34m-Cl	E
7017	20-F	E	7040	36-Cl	E
7018	23-Ne	E	7041	38-Cl	E
7019	22-Na	E	7042	38m-Cl	E
7020	24-Na	E	7043	37-Ar	E
7021	24m-Na	E	7044	39-Ar	E
7022	25-Na	E	7045	41-Ar	E

File No.	Radionuclide	Status	File No.	Radionuclide	Status
7046	42-Ar	E	7071	53-V	NE
7047	38-K	E	7072	54-V	NE
7048	38m-K	E	7073	49-Cr	E
7049	40-K	E	7074	51-Cr	E
7050	42-K	E	7075	55-Cr	E
7051	43-K	E	7076	54-Mn	NE
7052	44-K	E	7077	56-Mn	NE
7053	41-Ca	E	7078	53-Fe	NE
7054	45-Ca	E	7079	53m-Fe	NE
7055	47-Ca	NE	7080	55-Fe	NE
7056	49-Ca	E	7081	59-Fe	NE
7057	44-Sc	E	7082	55-Co	NE
7058	44m-Sc	E	7083	56-Co	NE
7059	46-Sc	E	7084	57-Co	NE
7060	46m-Sc	E	7085	58-Co	NE
7061	47-Sc	NE	7086	58m-Co	NE
7062	48-Sc	E	7087	60-Co	NE
7063	49-Sc	E	7088	60m-Co	NE
7064	50-Sc	NE	7089	57-Ni	NE
7065	50m-Sc	NE	7090	59-Ni	NE
7066	45-Ti	NE	7091	63-Ni	E
7067	51-Ti	NE	7092	65-Ni	E
7068	48-V	E	7093	62-Cu	NE
7069	49-V	E	7094	64-Cu	NE
7070	52-V	NE	7095	66-Cu	NE

File No.	Radionuclide	Status	File No.	Radionuclide	Status
7096	63-Zn	NE	7121	95-Nb	E
7097	65-Zn	E	7122	95m-Nb	E
7098	74-As	E	7123	93-Mo	E
7099	75-Se	E	7124	93m-Mo	E
7100	79m-Br	E	7125	99-Mo	E
7101	80-Br	E	7126	99-Tc	E
7102	80m-Br	E	7127	99m-Tc	E
7103	82-Br	E	7128	103-Ru	E
7104	82m-Br	E	7129	102-Rh	E
7105	79-Kr	E	7130	102m-Rh	E
7106	79m-Kr	E	7131	103m-Rh	E
7107	85-Sr	NE	7132	104-Rh	E
7108	85m-Sr	NE	7133	104m-Rh	E
7109	89-Sr	NE	7134	107m-Ag	E
7110	88-Y*	E	7135	108-Ag	E
7111	89m-Y	NE	7136	108m-Ag	E
7112	90-Y	NE	7137	109m-Ag	E
7113	90m-Y	NE	7138	110-Ag	NE
7114	89-Zr	NE	7139	110m-Ag	NE
7115	89m-Zr	NE	7140	109-Cd	E
7116	93-Zr	E	7141	111m-Cd	NE
7117	95-Zr	E	7142	113-Cd	NE
7118	93m-Nb	E	7143	113m-Cd	NE
7119	94-Nb	E	7144	111-In	NE
7120	94m-Nb	E	7145	111m-In	NE

File No.	Radionuclide	Status	File No.	Radionuclide	Status
7146	113m-In	NE	7171	125m-Te	NE
7147	114-In	NE	7172	125-I	NE
7148	114m-In	NE	7173	126-I	E
7149	115-In	E	7174	125-Xe	NE
7150	115m-In	E	7175	125m-Xe	NE
7151	116-In	NE	7176	127-Xe	NE
7152	116m-In	NE	7177	127m-Xe	NE
7153	116n-In	NE	7178	134-Cs	NE
7154	113-Sn	NE	7179	135-Cs	NE
7155	113m-Sn	NE	7180	135m-Cs	NE
7156	117m-Sn	E	7181	136-Cs	NE
7157	119m-Sn	NE	7182	136m-Cs	NE
7158	121-Sn	NE	7183	137-Cs	E
7159	121m-Sn	NE	7184	133-Ba	NE
7160	123-Sn	NE	7185	133m-Ba	NE
7161	123m-Sn	NE	7186	137m-Ba	E
7162	125-Sn	NE	7187	139-Ce	NE
7163	125m-Sn	NE	7188	145-Pm	NE
7164	126-Sn	E	7189	145-Sm	NE
7165	122-Sb	NE	7190	146-Sm	E
7166	122m-Sb	NE	7191	152-Eu	NE
7167	124-Sb	E	7192	154-Eu	NE
7168	124m-Sb	E	7193	155-Eu	NE
7169	124n-Sb	E	7194	157-Tb	NE
7170	125-Sb	NE	7195	157-Dy	NE

File No.	Radionuclide	Status	File No.	Radionuclide	Status
7196	159-Dy	NE	7216	204m-Pb	NE
7197	174-Hf	NE	7217	231-Th	NE
7198	175-Hf	NE	7218	228-Th	NE
7199	181-Hf	NE	7219	239-Np	NE
7200	179-Ta	NE	7220	241-Am	NE
7201	180m-Ta	NE	7221	243-Am	E
7202	182-Ta	NE			
7203	182m-Ta	NE			
7204	182n-Ta	NE			
7205	181-W	NE			
7206	185-W	NE			
7207	185m-W	NE			
7208	187-W	NE			
7209	198-Au	E			
7210	197-Hg	NE			
7211	197m-Hg	NE			
7212	203-Hg	NE			
7213	204-Tl	NE			
7214	207-Bi	NE			
7215	204-Pb	NE			

TABLE 3: 4n SERIES
MEAN ALPHA, BETA AND GAMMA ENERGIES FOR DECAY

Nuclide	Mean Energy Per Decay (keV)		
	Alpha	Beta	Gamma
^{236m}Np	0	91.4 ± 3.3	49.1 ± 4.3
^{236g}Np	8 ± 2	240 ± 20	153 ± 9
^{236}Pu	5851 ± 24	13.3 ± 1.1	1.6 ± 0.2
^{232}U	5397 ± 67	16.8 ± 1.4	1.7 ± 0.1
^{228}Th	5495 ± 69	21.7 ± 2.0	3.2 ± 0.1
^{224}Ra	5776.5 ± 3.2	2.2 ± 0.2	10.1 ± 0.1
^{220}Rn	6404.1 ± 1.0	0	0.69 ± 0.07
^{216}Po	6906.5 ± 0.5	0	0.015 ± 0.002
^{212}Pb	0	174.6 ± 8.2	144.5 ± 0.3
^{212n}Bi	0	1260 ± 400	5.1 ± 0.5
^{212m}Bi	5800 ± 135	49 ± 11	1.8 ± 0.1
^{212g}Bi	2218 ± 135	499.5 ± 2.6	108.5 ± 1.3
^{212n}Po	11783 ± 19	0.38 ± 0.04	91.2 ± 3.4
^{212m}Po	1340 ± 140	123 ± 11	1121 ± 53
^{212g}Po	8953.7 ± 8.8	0	0
^{208}Tl	0	594 ± 15	3385 ± 12

TABLE 4: DATA CONSISTENCY - PERCENTAGE DEVIATION BETWEEN EFFECTIVE Q-VALUE AND CALCULATED Q-VALUE

Nuclide	Percentage Deviation
^{236m}Np	0.0735
^{236g}Np	0.0486
^{236}Pu	0.0153
^{232}U	0.0360
^{228}Th	0.0122
^{224}Ra	0.0020
^{220}Rn	0.0011
^{216}Po	0.0012
^{212}Pb	0.1244
^{212n}Bi	0.0000
^{212m}Bi	0.0052
^{212g}Bi	0.0049
^{212n}Po	0.0010
^{212m}Po	0.0247
^{212g}Po	0.0008
^{208}Tl	0.1086

(d) <u>Fission Yields: Evaluations for the European JEF-2 Library</u> (M F James (WTC), R W Mills (BNFL plc) and D R Weaver (University of Birmingham))

The UKFY2 experimental database was used to generate adjusted independent and cumulative yields (ENDF/B-VI format) in February 1990. The library included neutron fission from ^{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 ^{243}Cm, ^{244}Cm and ^{245}Cm, and from the spontaneous fission of ^{242}Cm, ^{244}Cm and ^{252}Cf. This library has been supplied to the NEA Databank, and included in the preliminary release of JEF-2.

Following a query about some low standard deviations for the cumulative yields, an error was found in the program. This problem was corrected and the revised library dispatched to the NEA Databank for inclusion in the final version of JEF-2, planned to be available late September 1991.

The fission yield evaluations are described in three reports (AEA-TRS-1015, 1018 and 1019). The first document outlines the evaluation methods and summarises the results, the second tabulates experimental yields and their weighted average, and the third lists those yields for which measurements are sparse or discrepant. It should be noted that there are still forty-five such chain yields for ^{239}Pu fast fission and sixty-five for ^{238}U fast fission (and thirty-three for ^{235}U thermal fission).

(e) <u>Delayed Neutron Yields</u> (M F James (WTC), R W Mills (BNFL plc) and D R Weaver (University of Birmingham))

The fission product yields have been used with the decay data in JEF-2 to calculate delayed neutron data; these calculated values for $\bar{\nu}_d$ agree well with measurements except for ^{238}U. Further studies are underway both to investigate the sensitivity of $\bar{\nu}_d$ calculations to input parameters (such as fission yields and delayed neutron emission probabilities) and to estimate the time dependence of delayed neutron emission. Decay heat calculations will also be made.

A review paper has been written in collaboration with colleagues in France and the United States of America on the status of delayed neutron data. This document will soon be published as an NEACRP committee paper (NEACRP-L-232).

4. Conclusions

Progress in the UK measurement and evaluation of nuclear data depends on a low level of effort and dedicated expertise. Suitable personnel to undertake this work are retiring or being redirected towards other studies within AEA Technology, undermining the ability of this organisation to undertake such measurements. It is hoped that relevant studies can be maintained within academia, but such an approach will prove increasingly ineffective if the UK nuclear industry is not more supportive.

While UK measurement programmes continue under extreme difficulties, the various evaluation efforts have been maintained at a feasible level. Significant progress has been made in preparing the appropriate fission yield data files and activation product decay data library. However, even for these areas, support within the UK will be difficult to maintain, underlining the importance of supporting multinational efforts in this field.

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