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Summary Report

Second Research Coordination Meeting on

Updated Decay Data Library for Actinides

IAEA Headquarters, Vienna, Austria
28 – 30 March 2007

Prepared by

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IAEA Nuclear Data Section
Vienna, Austria

December 2007

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Abstract

The second meeting of the Coordinated Research Project on "Updated Decay Data Library for Actinides" was held at the IAEA, Vienna on 28-30 March 2007. A summary of the presentations made by each participant is given, along with subsequent discussions. The evaluation procedure was reviewed and a short tutorial session on using the software of the Decay Data Evaluation Project (DDEP) was given. The list of radionuclides for evaluation was updated, and their allocation amongst the participants agreed.

December 2007

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1. Introduction

The 2nd Research Coordination Meeting (RCM) of the Coordinated Research Project (CRP) on an "Updated Decay Data Library for Actinides" was held at the IAEA, Vienna, 28-30 March 2007. This meeting followed the 1st Research Coordination Meeting, held 17-19 October 2005 and summarized in document INDC(NDS)-0479. The Agenda, as adopted at this meeting, and the list of participants are given in Appendix A and B respectively.

This CRP originated following the support of the International Nuclear Data Committee (INDC) which advises the Nuclear Data Section (NDS) on nuclear data issues, who noted the need for this work for a wide range of applications. The INDC recommended in the Summary Report of their meeting of May 2002, INDC/P(02)-23, that a CRP on "Updated Decay Data Library for Actinides" be initiated in 2005, re-emphasising their support in May 2004.

A. L. Nichols, Head of the IAEA-NDS, opened the meeting and M. A. Kellett (IAEA-NDS), the Project Officer, presented some initial comments reiterating the aim and scope of the CRP. F. G. Kondev (ANL, USA) was elected Chairman of the meeting and M. A. Kellett Rapporteur.

Following the adoption of the Agenda (see Appendix A) the Chairman invited each participant (see Appendix B) to present a summary of their relevant evaluation and/or measurement work being carried out under the auspices of the CRP.

Apologies for absence were received from A. Luca (Romania) who was unable to attend the meeting.

2. Presentations

2.1 M.-M. Bé: Status of Evaluations at LNHB

A list of the current radionuclides being evaluated by the group at the Laboratoire National Henri Becquerel (LNHB) was given. Currently two uranium decay schemes, ^{234}U and ^{238}U, have just been reviewed and the comments are being considered. A further ten radionuclides are in various different stages of the evaluation process, including three which were not originally allocated at the 1st RCM, i.e. ^{210}Tl, ^{218}Rn and ^{218}At.

The LNHB publish periodically evaluations carried out within the Decay Data Evaluation Project (DDEP) as a *Monographie* of the *Bureau International des Poids et Mésures* (BIPM), Sèvres, France. The latest publication, volume 3 of *Monographie BIPM-5*, contains details of 28 evaluations, ten of which were undertaken within this CRP.

The LNHB also administer the webpage of the DDEP (http://www.nucleide.org/NucData.htm), where all evaluated decay data files and comments can be viewed. The webpage has been recently enhanced to include the decay data files in the internationally accepted ENSDF format. This particular format is made use of worldwide by decay data evaluators, in particular, those carrying out mass chain evaluation work published in Nuclear Data Sheets under guidance from the IAEA Network of Decay Data Evaluators. Making data files available in this format, makes them more accessible to general users of such data and also allows their manipulation and testing with a variety of standard tools and applications. It can be reiterated here that one of the aims of this CRP is to make available all evaluations in this format.

In order to help with the coordination and communication within the DDEP, the LNHB have created a discussion forum for its evaluators and collaborators. This is available at the following web address: http://laraweb.free.fr/DDEP_forum/ and interested parties may register in order to have access.

Finally, details of the experimental determination of the ^{233}U half-life were given, which involved members from the LNHB group, as well as three other European laboratories. The same team also plan to measure the half-lives of ^{235}U and ^{238}U, after this initial proving experiment.

2.2 V. Chechev: Production of Recommended Decay Data Files for a number of Actinides

The progress on the recent evaluation of eight actinides was presented. Most are nearing completion or have already been reviewed. A further eight actinides have also been fully evaluated and are completed. However, all sixteen of these actinides will continue to be monitored and minor updates produced, if necessary, within the framework of the CRP.

A variety of difficulties occur where there are limited experimental data available for the evaluations and these were highlighted. In particular, it was noted that no measurements for the half-life of ^{237}U have been made since 1958 and that the evaluated value is dominated by one very accurate measurement from the three available. It was stated that this is not an isolated case where data are scarce. However, for many of the evaluations carried out sufficient data are available and consistent values amongst different experimental datasets were obtained.

Finally, a list of experimental needs, relating to the work carried out, was presented (see section 4).

2.3 X. Huang: Evaluation of Decay Data for ^{225}Ac and ^{213}Bi

Details of the evaluations of two radionuclides were presented.

For ^{225}Ac large experimental datasets exist for the γ-ray energies and emission probabilities and from these a comprehensive decay scheme was derived, allowing the α-particle energies and emission probabilities to be determined. The subsequent evaluated values agree well with available experimental data. It was noted that a further measurement of the half-life was desirable, owing to only two measurements having been made, the latest being in 1950. Adjustment was made to the measured intensity of the 150.04-keV γ ray, owing to interference in measured spectra from two lines of similar energy from two radionuclides in the same α-decay chain. Three new levels were suggested; however, there still remain nine measured γ rays which are still unplaced in the final proposed decay scheme.

For ^{213}Bi the branching fractions for the two decay modes, α and β^-, were calculated based on the absolute emission probability of the 465-keV γ ray associated with the subsequent β^--decay of ^{209}Tl, i.e. the α-decay daughter. A new decay scheme has been proposed which includes four new levels in the β^- branch and only one level, instead of two, in the α branch. These newly defined level assignments allowed for the placing of an additional eight measured γ rays compared to the previously accepted decay scheme, but a further four still remained unplaced.

2.4 A.L. Nichols: Status of the Decay Data Evaluations for $^{242, 242m, 244, 244m}$Am

The progress with the evaluations of the four americium isotopes $^{242, 242m, 244, 244m}$Am was presented.

In the case of ^{242}Am only limited data are available for both the half-life and the γ-ray energies and emission probabilities, owing to their low energy (~ 40 keV) and intensity. Further measurements would be required to confirm the validity of the proposed simple decay scheme. For ^{242m}Am, the evaluation process was at the reference collection stage, but it already appeared that improved internal transition data were required.

For the ^{244}Am ground state the data are scarce, there being only one half-life measurement in 1962 and of the four measurements of the γ-ray emission probabilities, only the latest set from 1984 contained any estimate of the uncertainties. Nonetheless, three of the datasets were in reasonably good agreement. A final decay scheme with a single β^--decay and seven γ rays was presented. In the case of the associated meta-stable state, ^{244m}Am, data are also scarce for the half-life (only two measurements

without uncertainties from the 1950s) and the γ-ray emission probabilities, where only one known dataset existed. Two measurements of the EC/β^- branching ratio were found, from which the evaluated value was determined. A total of eight γ rays were placed into the final proposed decay scheme. For both the ground and meta-stable states, further half-life and γ-ray emission probabilities measurements are required.

2.5 A. Pearce: Status of the Decay Data Evaluations for ^{232}U and ^{228}Ac

A report was given on the progress of the evaluation work for the two radionuclides ^{232}U (α-decay) and ^{228}Ac (β^--decay), both of which decay to the same daughter ^{228}Th. Only one reference for each nuclide was obtainable which reported a measurement of the half-life; other reported references are still to be obtained. Reasonably good and complete γ-ray energy and emission probability measurements exist for ^{228}Ac, but at this early stage it was not clear how good equivalent data are for ^{232}U; also problems occur with the α-decay data.

The evaluations for ^{232}Th and ^{231}Pa are planned to start once these first two evaluations are completed.

Although the National Physical Laboratory (NPL) has excellent measurement capabilities, it was reported that due to funding restrictions, it was unlikely that any measurements relevant to the CRP could be carried out. However, such measurements could be proposed to the UK Nuclear Science Forum (UKNSF) to see whether interest and/or funding were available elsewhere in the UK.

2.6 G. Mukherjee: Status of the Decay Data Evaluations for ^{233}U and ^{229}Th

Progress on the early stages of the ^{233}U evaluation was given. Many experimental measurements have been made for this radionuclide, which undergoes 100% α-decay to ^{229}Th. In particular, there have been recently published γ-ray studies by Helmer *et al.* (1994) and Barci *et al.* (2003) leading to the need for new levels to be introduced compared to the most recent ENSDF evaluation of 1990. The very low-lying level at ~3.4 eV has been closely re-examined.

2.7 F.G. Kondev: Status of Decay Data Measurements at ANL

Details of the experimental facilities available at Argonne National Laboratory (ANL) were presented. These include the GAMMASPHERE hyper-pure germanium detector array, as well as other more standard germanium detectors for γ-ray measurements. ANL also has a variety of silicon based detectors for conversion electron and β^--decay experiments, which are also used for α-decay studies. It was recalled that a list of the available sources at ANL was presented at the 1st RCM, and is available as Appendix D in the summary report of that meeting (INDC(NDS)-0479).

An ambitious experimental programme is well underway and results have already been published for the half-life and/or α-particle energies and emission probabilities of ^{250}Cf, ^{246}Cm, ^{245}Cm and ^{240}Pu; work is still ongoing for ^{243}Cm.

Analysis of the experimental technique required in order to measure the currently discrepant γ-ray emission probability for the ~28.6-keV line following ^{233}Pa β^--decay, have showed that studies measuring this quantity with the ^{233}Pa daughter in equilibrium with the ^{237}Np parent, have resulted in great difficulties in resolving this particular line in the γ-ray spectra owing to the proximity of a much more intense 29.37-keV line following ^{237}Np α-decay. Hence, it is necessary to "milk" the ^{233}Pa daughter and use chemical separation to obtain a "pure" source, prior to carrying out the γ-ray emission probability measurements. However, further conversion electron measurements are also probably required in order to help resolve this problem completely.

Finally, discrepancies were found in reported α-particle emission probabilities measured using different detector types (i.e. semiconductor detectors and magnetic spectrographs) and so a systematic investigation is underway at ANL to better understand and quantify this problem.

2.8 F.G. Kondev: Status of Decay Data Evaluations at ANL – ^{246}Cm, ^{206}Tl and ^{206}Hg

A very detailed presentation was given on the two completed evaluations for ^{206}Tl and ^{246}Cm. These have both been reviewed within the DDEP system and have been published in two ANL reports of the Nuclear Data and Measurements Series, ANL/NDM-162 and -164 respectively, copies of which were made available. The evaluation for ^{206}Hg is being finalised and the associated report ANL/NDM-166 is in preparation.

In the case of ^{246}Cm (~100% α-decay), measurements were also performed at ANL for the α-decay half-life and the α-particle energies and emission probabilities. These yielded consistent results which were appropriately incorporated into the evaluation. The two γ-ray emission probabilities, having not being directly measured, were deduced from the relative α-particle transition probabilities, taking into account any conversion electrons.

For ^{206}Tl (100% β^--decay) a consistent set of data existed for the half-life, allowing an accurate evaluated value to be determined. In the case of the three γ-ray transitions, one of these is an E0 transition and details were given on how this was treated, involving the calculation of the associated X-rays and that no conversion electrons are produced from this transition. A warning was given that caution should be taken in the automatic use of any auxiliary code for calculating internal conversion coefficients as this approach does not apply to E0 transitions.

3. Review of the Evaluation Procedure and Rules

Following discussion amongst the participants, the exact procedure to be followed once an evaluation has been produced was clarified.

The basic procedure is outlined as follows:

- All evaluated data are entered into the SAISINUC program, supplied by M.-M. Bé, which produces a MS-Access database file called *donnees.mdb*
- A file containing the evaluator's comments on the evaluation should be produced in MS-Word, i.e. *comments.doc*
- These two files are then sent to M.-M. Bé who will return a PDF of generated tables, i.e. *tables.pdf*
- The evaluator should then check the contents of the three files *donnees.mdb*, *comments.doc* and *tables.pdf*, prior to sending them to E. Browne, the DDEP co-ordinator
- E. Browne will organise for an independent review of the submitted evaluation to be made and will return appropriate corrections/suggestions to the original evaluator
- Once these corrections/suggestions have been incorporated into the evaluation and SAISINUC, the evaluator should send updated *donnees.mdb* and *comments.doc* files to M.-M. Bé for final acceptance and publishing on the DDEP website

One point was noted that the review process is likely to mean that some participants will be expected to review evaluations of their fellow participants, owing to the limited overall membership of the DDEP. Therefore it was emphasised that participants should allocate an appropriate amount of time to this task in order to ensure the final quality of the evaluations.

A very useful demonstration session using the SAISINUC program was held, which involved entering actual data from one of the participant's evaluations. All participants found this most informative and M.-M. Bé was thanked for her efforts and also for the continuing enhancement of the SAISINUC program and its interfacing with associated programs.

4. Review of Identified Measurement Requirements

During the various evaluations carried out to date by the participants a variety of experimental needs were identified, as shown in Table 1. It is possible that some of these might be addressed within the experimental programmes of participants, but certainly not all.

Table 1 should not be treated as an exhaustive list of the required measurements for all of the radionuclides being considered, but merely as a subset for which specific needs have so far been identified.

TABLE 1. MEASUREMENT REQUIREMENTS SO FAR IDENTIFIED

Radionuclide	Problem
^{224}Ra	Data for the α-particle and γ-ray emission probabilities are inconsistent and further spectroscopic studies are required.
^{226}Ra	Only one set of data for the α-particle emission probabilities with no uncertainties - requires further measurement.
^{225}Ac	Only two measurements of the half-life, the latest in 1950 – further measurements desirable.
^{233}Th	All the emission probabilities are reported without uncertainties – accurate measurements are required.
^{233}Pa	New precise measurements of the low-energy γ rays and LX-rays with a pure ^{233}Pa source would prove beneficial.
^{237}U	Very poor and early experimental half-life data – further measurements required.
^{239}U	A number of γ rays observed in the reference 2006Wo03 were not placed in the decay scheme, hence further measurements required. However the relative intensity of these unplaced γ rays is ~0.5% of the total.
^{236,236m}Np	Poor experimental data with two conflicting measurements of the EC/β^- branching ratio – further measurements required.
^{239}Pu	Measurements of the multipolarities of the low-energy γ rays would prove beneficial.
^{241}Am	A number of γ-ray transitions (27.03-, 54.1- and 95.0-keV) require more detailed measurement, including the associated conversion electron emission probabilities.
^{242}Am	For half-life determination have only three sets of data – requires further measurement. Spectroscopic γ-ray study also required as no emission probability measurements exist. Currently γ-ray energies constructed from level scheme (Akovali 2002) and emission probabilities from $P_{ce}/P_{\beta-}$ data.
^{242m}Am	Only limited internal transition data available – further measurements highly desirable.
^{244}Am	Only one half-life measurement known - requires further measurement. Spectroscopic γ-ray study also required. Currently γ-ray energies constructed from level scheme (Akovali 2002) and emission probabilities adjusted as necessary from 1984Ho02 as this is the only reference providing uncertainties.
^{244m}Am	Only two half-life measurements from the 1950s, neither of which quote uncertainties – further measurements required. Spectroscopic γ-ray study also required as currently have only one set of data for the emission probabilities.
^{242}Cm	Accurate measurements of the 44-, 102-, 157-, 210-keV γ rays required.

5. Review of Allocated Nuclides

Following the various presentations a review of the currently allocated nuclides was made.

Table 2 shows the updated list of nuclides allocated to each participant following discussion at this meeting.

TABLE 2. ALLOCATION OF NUCLIDES

Participant	Actinides	Decay daughters
A. Luca	^{234}Th, ^{236}U	^{228}Ra
A. L. Nichols	^{228}Th, ^{242}, ^{242m}, ^{244}, ^{244m}Am	^{208}Tl, ^{212}Pb, ^{212}, ^{215}Bi, ^{212}, ^{216}Po, ^{211}, ^{219}At, ^{219},^{220}Rn, ^{224}Ra
A. Pearce	^{232}Th, ^{231}Pa, ^{232}U	^{228}Ac, ^{223}Ra
F. G. Kondev	^{243}, ^{245}, ^{246}Cm	^{206}Hg, ^{206}, ^{207}, ^{209}Tl, ^{209}, ^{211}Pb
G. Mukherjee	^{229}Th, ^{233}U	
M.-M. Bé	^{243}Am, ^{234}, ^{238}U, ^{252}Cf	^{210}Tl, ^{210}, ^{214}Pb, ^{210}, ^{214}Bi, ^{210}, ^{214}, ^{218}Po, ^{218}At, ^{218},^{222}Rn, ^{226}Ra
V. P. Chechev	^{233}Th, ^{233}Pa, ^{237}, ^{239}U, ^{236}, ^{236m}, ^{237}, ^{238}, ^{239}Np, ^{238}, ^{239}, ^{240}, ^{241}, ^{242}Pu, ^{241}Am, ^{242}, ^{244}Cm	^{227}Ac
Huang Xiaolong	^{231}Th, ^{235}U	^{221}, ^{223}Fr, ^{217}At, ^{217}Rn, ^{213}Bi, ^{213}Po, ^{225}Ra, ^{225}Ac
Unallocated		^{211}Bi, ^{211}, ^{215}Po, ^{215}At

Key to colours:
Red = done
Blue = in progress
Green = to be monitored/updated
Black = to be done

6. Review of Actions

Throughout the meeting a number of actions were generated on all or particular participants. A complete list of these actions is given below.

1) **ALL:** Check the Q_{eff} calculated from the level scheme and compare with the AME-2003 value (Audi *et al.*). Add a paragraph into the comments file stating this balance. Could lead the evaluator to see where the level feeding might be inconsistent.
2) **ALL:** Check the calculated X-ray emissions with any available experimental data, as differences can be quite significant. Add a paragraph into the comments file stating this balance.
3) **Bé:** Check the ability of the SAISINUC program to add the EMISSION program output data correctly.
4) **Bé:** Investigate adding the capability for the SAISINUC program to read the level energy information from an ENSDF (or equivalent) file.
5) **Kondev:** Contact E. Browne to see if the EC-CAPTURE program can be modified to output the P_{L1}, P_{L2} and P_{L3} values.

6) **Bé:** Contact the ^{240}Pu measurers (EUROMET project - G. Sibbens, IRMM) to obtain any information as soon as possible.

7) **Pearce/Mukherjee:** Investigate ^{233}Th (and ^{233}Pa) emission probability measurements (produced from n-capture on ^{232}Th, hence probably need a reactor source).

8) **Pearce/Mukherjee:** Investigate ^{237}U emission probability and half-life measurements. There is a large δQ discrepancy.

9) **Chechev:** Calculate the total relative intensity of the unplaced γ rays of ^{239}U (2006Wo03) compared to the placed γ rays, in order to quantify the unplaced intensity.

10) **Kondev:** Investigate the EC/β^- branching fraction problem in ^{236,236m}Np, as the measurements were originally made at Argonne National Laboratory.

11) **Kondev/Bé:** Investigate the possibility of undertaking α-particle emission probability measurements for ^{224,226}Ra (should sources be available).

7. Final Remarks

During the meeting various discussions were held on the appropriate outcomes required from this CRP. The final report will include tabulations of the most important decay parameters and data files will be produced. Using the SAISINUC program, files in the ENSDF format can be directly produced. The participants also felt that production of data files in the ENDF format was important and this point was noted. No expertise within the group exists for the conversion of files from the ENSDF format to the ENDF format, but at a time when the final files are available, the IAEA will explore the possibility of employing a consultant to undertake this task.

The next, and final, RCM is planned for approximately eighteen months time. Note: the date has now been confirmed for **Wednesday 8th to Friday 10th October 2008**.

The meeting participants felt that the meeting was a success and that all necessary issues were appropriately covered.

The Chairman closed the meeting and was thanked for his excellent running of the meeting.

2nd Research Coordination Meeting on

"Updated Decay Data Library for Actinides"

IAEA Headquarters, Vienna, Austria
28 – 30 March 2007
Meeting Room A2313

AGENDA

(As adopted at the meeting)

Wednesday, 28 March

08:30 - 09:30	**Registration** (IAEA Registration desk, Gate 1)
09:30 - 10:15	**Opening Session**
	Welcoming address – A.L. Nichols
	Introductory Remarks – M.A. Kellett
	Election of Chairman and Rapporteur
	Discussion and Adoption of Agenda (Chairman)
	Apologies for Absence
10:15 - 11:00	**Administrative and Financial Matters related to participants**
	Coffee break
11:00 - 12:30	**Session 1: Presentations by participants**
	(15 minutes for each presentation and 5 minutes for discussion)
12:30 – 14:00	*Lunch*
14:00 - 15:30	**Session 1 (cont'd): Presentations by participants**
	(15 minutes for each presentation and 5 minutes for discussion)
	General Discussion
15:30 - 16:00	*Coffee break*
16:00 - 17:30	**Session 2: Review of the Selected Nuclei**
	Importance/priority of selected nuclei
	Availability of data

Thursday, 29 March

09:00 - 10:30	**Session 3: Status**
	Measurements
	Evaluations
10:30 – 11:00	*Coffee break*
11:00 – 12:30	**Session 3 (cont'd): Status**
12:30 - 14:00	*Lunch*
14:00 – 15:30	**Session 4: Review of Evaluation Procedure and Rules**
	Evaluation procedure and reference coverage
	Available software – comments/criticisms
15:30 – 16:00	*Coffee break*
16:00 – 17:30	**Session 5: Output**
	Evaluation Report – style and content
	Data Table format
	Data-file formats, e.g. ENSDF, ENDF
19:00	**Dinner at a Restaurant in the Centre of Vienna**

Friday, 30 March

09:00 - 10:30	**Session 6: Allocated and Re-allocation (?) of Nuclei**
	Nuclei being measured
	Nuclei being evaluated
10:30 – 11:00	*Coffee break*
11:00 – 12:30	**Session 7: Summary Report**
	Drafting of the 2nd RCM Summary Report
12:30 - 14:00	*Lunch*
14:00 – 15:30	**Session 8: Final Remarks and Close of the Meeting**
	Any other business and time schedule for the 3rd RCM

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