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Improving Nuclear Data Accuracy of the ^{241}Am Capture Cross-section

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Foreword

Nuclear energy applications require knowledge of fundamental nuclear physics in order to design and operate facilities. Novel systems including Generation IV nuclear designs have target accuracies in essential operating quantities that are beyond the current state of the art in modelling and simulation. Improved understanding of the relevant physics and/or uncertainties is necessary to achieve these accuracies, requiring new measurements and/or data evaluations.

The Nuclear Energy Agency (NEA) Working Party on International Nuclear Data Evaluation Co-operation (WPEC) was established under the NEA Nuclear Science Committee (NSC) in 1989 to promote the exchange of information on nuclear data. Following the recommendations of NEA WPEC Subgroup 26 on Uncertainty and Target Accuracy Assessment for Innovative Systems Using Recent Covariance Data Evaluations, the WPEC Expert Group on the High Priority Request List for Nuclear Data (EGHPRL) reviewed and accepted requests for improved knowledge of americium neutron capture.

Following several international experimental campaigns, WPEC agreed that co-ordinated analysis of the measurements would yield substantial improvements in terms of data accuracy and uncertainty reduction. NEA WPEC Subgroup 41 on Improving Nuclear Data Accuracy of the ^{241}Am Capture Cross-section was launched to review all of the available data, address discrepancies and produce recommendations for ^{241}Am nuclear data.

The present report provides a summary of the experiments analysed and consensuses reached by NEA WPEC Subgroup 41, including final recommendations based on all experimental data available at the time. These data have already been incorporated into new evaluations that have been or will be included in the most recent nuclear data library releases.

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List of abbreviations and acronyms

ADS	Accelerator-driven system
AGS	Analysis of Geel Spectra (JRC Geel, Belgium)
ANNRI	Accurate Neutron Nuclear Reaction Measurement Instrument (Japan)
APOLLO	Multi-group transport code (CEA, France)
BNL	Brookhaven National Laboratory (United States)
BGO	Bismuth germanium oxide
BRR	Budapest Research Reactor (Hungary)
BWR	Boiling water reactor
CASMO	Lattice physics code for reactor modelling
C/E	Calculation and experimental values
CEA	Commissariat à l'énergie atomique et aux énergies renouvelables (Alternative Energies and Atomic Energy Commission, France)
CERN	European Organisation for Nuclear Research
CHANDA	CHAllenges in Nuclear DAta project (European Union)
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Spain)
DANCE	Detector for Advanced Neutron Capture Experiments (LANL, United States)
DDEP	Decay Data Evaluation Project
EC	Electron capture
EGHPRL	Expert Group on the High Priority Request List for Nuclear Data (NEA)
ENDF	Evaluated Nuclear Data File (United States)
ENSDF	Evaluated Nuclear Structure Data File
EOLE	Heavy water research reactor (CEA, France)
ERANOS	European Reactor ANalysis Optimised calculation System
EXFOR	Exchange FORmat
FRM II	Forschungsreaktor München II (research reactor Munich II, Germany)
FUBILA	BWR Full MOX Core Physics Experiments (Japan)
FZJ	Forschungszentrum Jülich (Jülich Research Centre, Germany)
GEANT	Geometry And Tracking code
GELINA	Geel Linear Electron Accelerator (JRC Geel, Belgium)

HPGe	High-purity germanium
HPRL	High Priority Request List for Nuclear Data (NEA)
IAEA	International Atomic Energy Agency
ICC	Internal Conversion Coefficient
INDA	Improvement of Nuclear DAta project
JAEA	Japan Atomic Energy Agency (Japan)
JEFF	Joint Evaluation Fission and Fusion File
JENDL	Japanese Evaluated Nuclear Data Library (Japan)
J-PARC	Japan Proton Accelerator Research Complex (Japan)
JRC	Joint Research Centre (European Commission)
JSI	Jožef Stefan Institute (Slovenia)
KUR	Kyoto University Research Reactor (Japan)
LANL	Los Alamos National Laboratory (United States)
LNHB	Laboratoire National Henri Becquerel (France)
LWR	Light water reactor
MA	Minor actinide
MC	Monte Carlo
MCNP	Monte Carlo N-Particle Transport Code (LANL, United States)
MELUSINE	Light water moderated research reactor (CEA, France)
MINERVE	Low power research reactor (CEA, France)
MISTRAL	MOX core physics experiments (EOLE, France)
MLF	Materials and Life Sciences Experimental Facility (J-PARC, Japan)
MOX	Mixed oxide fuel
MTR	Materials test reactor
NEA	Nuclear Energy Agency (OECD)
NSC	Nuclear Science Committee (NEA)
n_TOF	Neutron Time-of-Flight Facility (CERN)
OECD	Organisation for Economic Co-operation and Development
OSMOSE	Experiments quantifying integral cross-sections (MINERVE, CEA, France)
PGA	Prompt gamma-ray analysis
PHÉNIX	Prototype fast breeder reactor (France)
PROFIL	Experiments performed at the PHÉNIX reactor
PTB	Physikalisch-Technische Bundesanstalt (Germany)
PWR	Pressurised water reactor

TOF	Time of flight
TRIGA	Nuclear research reactors (General Atomics, United States)
TRIPOLI	Monte Carlo particle transport code (CEA, France)
WIMS	Winfrith Improved Multi-group Scheme
WPEC	Working Party on International Nuclear Data Evaluation Co-operation

Executive summary

Knowledge of basic nuclear physics of the constituents of nuclear systems is fundamental to our ability to model, simulate and understand their operation. State-of-the-art databases of these physics contain uncertainties that reflect those of the experiments they are based on. These uncertainties may introduce significant conservatisms, particularly for advanced reactor designs and fuel cycles. Identifying the priorities for new nuclear physics measurements has always been a key objective of the Nuclear Energy Agency (NEA) Working Party on International Nuclear Data Evaluation Co-operation (WPEC) mandates.

The WPEC Expert Group on the High Priority Request List for Nuclear Data (EGHPRL) organises the unique worldwide reference that assists national governments in prioritising measurements that use expensive and complex research infrastructure. Proposing successful, detailed entries to this list is a challenging task and WPEC periodically raises subgroups to collaborate on the analyses that are required to rigorously analyse and quantify the potential impact of new measurements. This work includes a combination of application accuracy requirements, state-of-the-art simulation, sensitivity-uncertainty analyses and a review of the feasibility of potential experiments.

Following the recommendations of a previous WPEC Subgroup that identified ^{241}Am as the isotope with the greatest opportunity for uncertainty reduction in advanced systems, the EGHPRL accepted two requests related to ^{241}Am and laboratories from around the world made measurements on ^{241}Am neutron-induced reactions. WPEC launched Subgroup 41 on Improving Nuclear Data Accuracy of the ^{241}Am Capture Cross-section to bring together experimentalists and theorists to evaluate all of the recent data and generate a state-of-the-art recommendation for the essential ^{241}Am physics for advanced nuclear system operation.

Subgroup 41 reviewed all recent and previous measurements from numerous techniques, critically evaluating the uncertainties from each experimental methodology and analysing discrepancies between different datasets. These reviews resulted in a consensus expert position on the set and hierarchy of experiments for data evaluation and, through a detailed statistical analysis of the available information, Subgroup 41 made recommendations for the essential ^{241}Am physics with uncertainty that is one-third the value from previous state-of-the-art evaluations.

1. Introduction

The need to improve accuracy of nuclear data has been quantified by the Nuclear Energy Agency (NEA) Working Party on International Nuclear Data Evaluation Co-operation (WPEC) Subgroup 26 [1] on Uncertainty and Target Accuracy Assessment for Innovative Systems Using Recent Covariance Data Evaluations for the development of innovative nuclear reactor systems. The work of Subgroup 26 identified a wide range of needs for advanced reactors, including those being considered within the Generation IV International Forum (GIF). While different concepts have their own parameters and accuracy requirements, many of the constituent materials and their nuclear reaction physics are shared. According to the analyses, capture cross-sections of MAs were shown to be one of the most important data. The analyses showed the need for improving the accuracy of capture cross-sections quantitatively by a factor of approximately two to three times. As one of the most innovative nuclear reactor systems, the accelerator-driven system (ADS) has received worldwide attention, in part because of its ability to incinerate minor actinides (MAs). Recently, sensitivity and uncertainty analyses have been performed [2-3] on ADS concepts using the covariance of the Japanese Evaluated Nuclear Data Library (JENDL-4.0) [4].

To meet these needs, measurements on capture cross-sections of MAs have been actively performed in this decade at various facilities, including Los Alamos National Laboratory/Detector for Advanced Neutron Capture Experiments (LANL/DANCE) in the United States [5-6]; the Japan Proton Accelerator Research Complex/Accurate Neutron Nuclear Reaction Measurement Instrument (J-PARC/ANNRI) in Japan [7-9]; the European Organisation for Nuclear Research/Neutron Time-of-Flight Facility (CERN/n_TOF) in Switzerland [10-11]; the Joint Research Centre/Geel Linear Electron Accelerator (JRC/GELINA) in Belgium [12]; the Forschungsreaktor München II (FRM II) in Germany [13]; the Budapest Research Reactor (BRR) in Hungary [14]; the light water moderated research reactor (MELUSINE) of the Commissariat à l'énergie atomique et aux énergies renouvelables (French Alternative Energies and Atomic Energy Commission [CEA]) in Grenoble, France [15]; the low power research reactor (MINERVE) of CEA Cadarache in France [16]; and Kyoto University Research Reactor (KUR) in Japan [17]. Different measurement methods have been utilised, including neutron time-of-flight methods using pulsed neutrons, activation methods using reactor neutrons, pile-oscillation methods and plutonium ageing (Pu-ageing) methods. There have been noticeable advancements on capture cross-section measurement techniques in this decade that enhanced the precision of data. However, there are still serious discrepancies between different measurements on the absolute value. This discrepancy has made the accuracy improvement a difficult exercise.

To achieve the requested accuracy, it is indispensable to ascertain main physical reasons behind the discrepancies as identified by the WPEC Subgroup 31 [18]. It was also considered necessary to integrate the knowledge of different measurement specialists together with evaluation specialists to solve the discrepancy problems. Reflecting this consideration, the first WPEC Subgroup 41 Workshop on Improving Nuclear Data Accuracy of ^{241}Am and ^{237}Np Capture Cross-sections was held in May 2015 at the NEA.

At this workshop, it was decided that the items of investigation were to be most concentrated on the capture cross-section of ^{241}Am at thermal-neutron energy, since there are various experimental data using state-of-the-art techniques for ^{241}Am, while comparatively few are available for ^{237}Np. At the second workshop of Subgroup 41, again held at the NEA in May 2016, details of the experiments and their analyses were exchanged and discussed by different measurement and evaluation specialists.

The members of Subgroup 41 are composed of specialists on energy-dependent cross-section measurements, spectrum-averaged experiments, relevant nuclear structure data and evaluations. The nuclear structure data, especially gamma-ray and X-ray emission probabilities, were invaluable for the work of Subgroup 41, since specialists on cross-section measurements often do not have enough information on their uncertainties – even though it plays an important role in determining the capture cross-section by activation methods.

In Chapter 2, energy-dependent cross-section measurements were reviewed including recent data at GELINA, DANCE, n_TOF, and ANNRI measured before 2015, and the ANNRI data published in 2018 [19] were added for evaluating the averaged value.

The origins of systematic bias have been discussed, including the effect of sample inhomogeneity, sample measurements, neutron flux determination, neutron self-shielding, multiple-scattering effects and normalisation independence. The averaged capture cross-section at thermal energy was deduced using the energy-dependent experimental data, where the data were selected based on the discussions presented in this report. It should be noted that some unselected data for deducing the averaged value play an important rule for cross-checking and even more for extracting a physical quantity such as a Westcott g-factor, which is needed in the evaluation of spectrum-averaged data. Some key bias effects were also identified for the unselected data for future improvement. Since the energy region covered by each facility is complementary, there is great value if these identified corrections are performed and the knowledge is integrated into the analysis.

In Chapter 3, recent spectrum-averaged cross-section experiments were reviewed, including the FRM II, KUR, MELUSINE and MINERVE data measured before 2015. The experiments include activation methods, pile-oscillation methods and Pu-ageing methods. For the activation method, two types of neutron sources have been utilised; one is the reactor neutrons with thermal Maxwellian and slowing-down type spectrum and the other is the cold neutron beam extracted from the research reactor. The advantage of utilising cold neutron beams at the FRM II and the BRR was shown due to the low background characteristics in their beamlines. The need for accurate energy-dependent cross-sections and X-ray emission probabilities was also identified as essential. Some of the identified bias effects have been evaluated in Section 3.1. Activation experiments using reactor neutrons with thermal Maxwellian and slowing-down type spectrum have been re-evaluated by taking into accounts the identified systematic bias effects as summarised in Section 3.2. Before these corrections, there were severe fragmentations between the activation data, which generated a large external uncertainty. The re-evaluation solved the fragmentation problem significantly, and gave a cross-section value with a comparatively small uncertainty. The measurements of pile-oscillation and plutonium ageing methods also need the energy-dependent information on both cross-section data and neutron spectrum to be used for validation of the thermal capture cross-section. The current status of these methods was reviewed in Section 3.3 and Section 3.4, respectively.

In Chapter 4, the relevant nuclear structure data and candidates for the appropriate emissions suitable for activation experiments are summarised. Uncertainties of the current

emission probabilities are identified and quantified. The difficulty due to the contaminated emissions is discussed, which will be a significant candidate of unrecognised systematic error.

In Chapter 5, the capture cross-section of ^{241}Am, together with its uncertainty for thermal neutrons, are derived based on the discussions in WPEC Subgroup 41 and integrated information including both energy-dependent and energy integral measurements. The weighted mean of the cross-section for ^{241}Am at 25.3 meV is determined to be 717 (13) b.

Recommendations of future actions are included in Chapter 6 to improve the accuracy of nuclear data, based on the discussions of Subgroup 41.

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2. Energy-dependent cross-section measurements

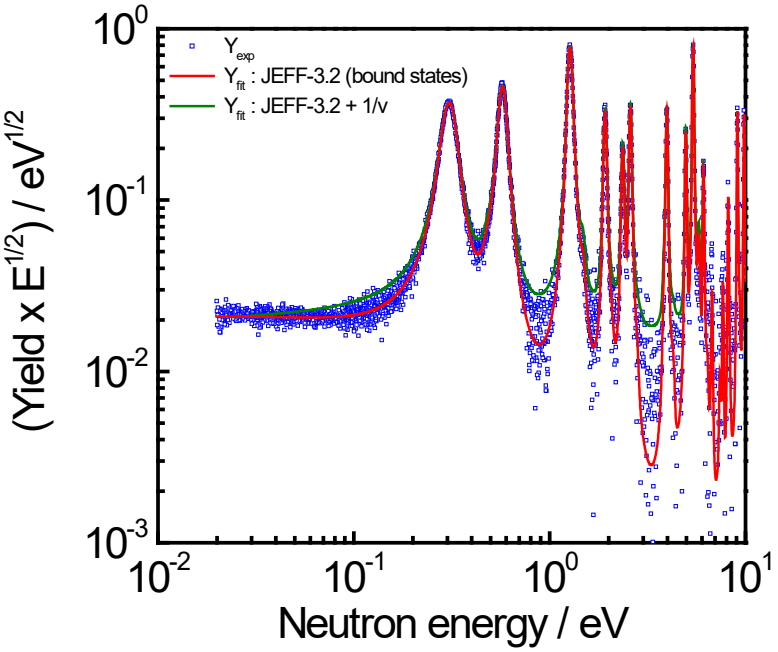
An overview of time-of-flight cross-section data that can be used to re-evaluate the cross-section for ^{241}Am neutron capture in the thermal and resolved resonance region is given by Lampoudis et al. [1] and Noguère et al. [2]. These lists can be complemented using results of capture experiments at the ANNRI facility of J-PARC [3] and at the n_TOF facility at CERN [4]. Noguère et al. [2] performed a re-evaluation of the total and capture cross-sections in the resolved and unresolved resonance region. This evaluation was adopted in the Joint Evaluation Fission and Fusion File (JEFF)-3.2 evaluated data file. The evaluation of Noguère et al. [2] results in a neutron capture cross-section for neutrons at 2 200 m/s of 747.8 b and a Westcott factor of 1.006 for a pure Maxwellian neutron spectrum with kT = 25.3 meV. The results of Noguère et al. [2] are fully consistent with those of Lampoudis et al. [1]. This agreement is not surprising. Noguère et al. [2] used the data of Lampoudis et al. [1] as a basis to renormalise the other capture data used in the evaluation, i.e. those of Jandel et al. [5] and Vanpraet et al. [6], and to adjust the sample properties for the measurements reported by Derrien and Lucas [7].

The data of Lampoudis et al. [1] result from transmission and capture measurements at the time-of-flight facility GELINA. The capture experiments were carried out at a 12.5 m flight path applying the total energy detection principle using deuterated benzene (C_6D_6) detectors. The results of the transmission measurements at a 25 m station were used to normalise the capture data. To avoid bias effects due to the sample properties, a special procedure was applied to produce a homogeneous sample dedicated to determine the parameters of the low energy resonances by transmission and the capture cross-section at thermal energy by capture measurements. A matrix of 3.0 g of Y_2O_3 was prepared by the so-called "sol-gel" method using porous granules into which approximately 320 mg of AmO_2 powder was infiltrated. After drying and calcination, the powder was pressed into a pellet. Compared to commonly used oxide powder samples, a much higher degree of homogeneity can be reached in such a sample and problems related to the powder grain size distribution are avoided. For the analysis of transmission data obtained with powder samples the Lambert-Beer attenuation low cannot directly be applied as discussed in detail by Becker et al. [8]. It requires a special model to account for the particle size distribution in the sample. Becker et al. [8] demonstrated that, when not accounting for the particle size distribution, the resonance strength ($g\Gamma_n$) will be underestimated. The underestimation depends on the sample properties, increases with increasing total cross-section and coincides with an overestimation of the total resonance width.

The same method was applied to prepare the sample that was used by Mendoza et al. [4] and Fraval et al. [9] for the capture experiments at the n_TOF facility. For this sample, an Al_2O_3 matrix was used. These experiments were carried out at a 180 m flight path. The data of Fraval et al. [9] are based on the total energy detection principle applying the pulse height weighting technique using C_6D_6 detectors. The data of Mendoza et al. [4] result from measurements with a BaF_2 total absorption detector. The two data sets were normalised fully independent of any other capture or transmission data for ^{241}Am.

To verify the procedure applied by Noguère et al. [2] only a limited number of independent data sets are available. Some of the data suffer from systematic bias effects, i.e. the capture data of Jandel et al. [5] and the transmission data of Derrien and Lucas [7] and Kalebin et al. [10]. In addition, the capture data of Harada et al. [3] and Weston and Todd [11] provide only information on the energy dependence of the ^{241}Am(n,γ) cross-section. The capture yields obtained by Jandel et al. [5] have to be reviewed because of an error in the determination of neutron flux [12]. Unfortunately, no details about this bias effect have been reported. Therefore, these data will not be included in the present discussion. The transmission data of Derrien and Lucas [7] and Kalebin et al. [10] suffer from a systematic effect due to sample characteristics, as discussed in [1] and in more detail in the following paragraphs. In this chapter the energy dependence of the cross-section in the low energy and the cross-section at thermal energy resulting from the time-of-flight cross-section data are investigated.

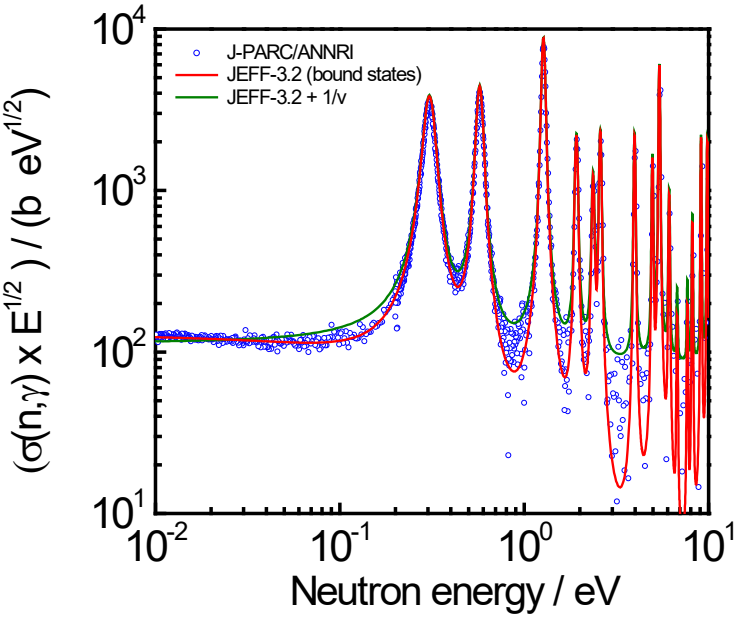
Figure 2.1. Yield multiplied by the square root of the energy as a function of neutron energy



Note: The experimental yield obtained by Lampoudis et al. [1] is compared with the theoretical yield derived from the resonance parameters in JEFF-3.2. The theoretical yield based on the unbound states in JEFF-3.2 combined with a 1/v contribution is also shown.

Source: JRC-Geel, 2019.

Figure 2.2. Capture cross-section of ^{241}Am multiplied by the square root of the energy as a function of neutron energy



Note: The experimental data obtained by Harada et al. [3] are compared with the cross-section derived from the resonance parameters in JEFF-3.2. The cross-section based on the unbound states in JEFF-3.2 combined with a 1/v contribution is also shown. The data are normalised at thermal energy using a cross-section σ_v = 749 b.

Source: JRC-Geel, 2019.

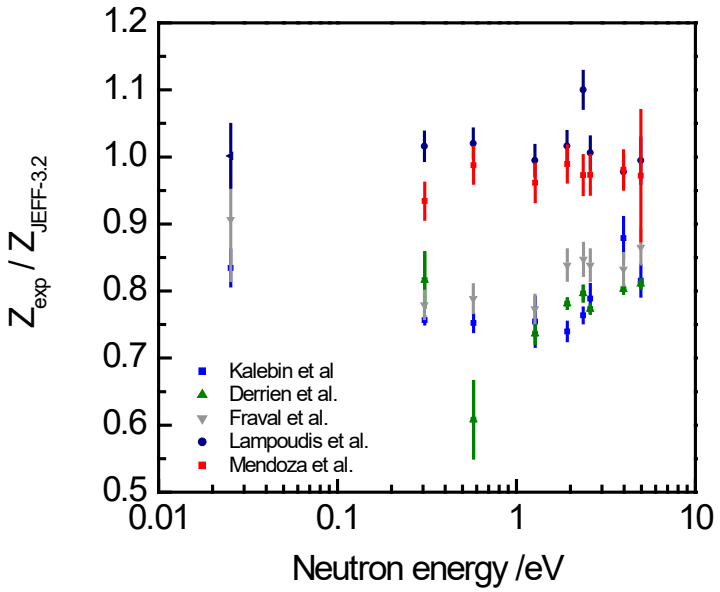
The observed resonances or unbound states contribute for only 30% to the capture cross-section for ^{241}Am at thermal energy, as shown in [13]. Therefore, a contribution of at least one bound state or of an additional 1/v component is required to describe the cross-section in the thermal energy region. In the JEFF-3.2 file, two bound states close to the neutron separation energy were added. The effect of adding these bound states is shown in Figure 2.1. In this figure, the experimental capture yield multiplied by the square root of the energy is plotted as a function of incident neutron energy. The data of Lampoudis et al. [1] are compared with the theoretical yield derived from the parameters in JEFF-3.2 and with a theoretical yield obtained from the unbound states in JEFF-3.2 combined with a pure 1/v additional contribution to match the cross-section at 2200 m/s. This figure clearly shows that the experimental data of Lampoudis et al. [1] require a contribution of bound states with an energy close to the neutron separation energy and not a 1/v additional contribution due to a direct capture component.

This is confirmed by the data of Harada et al. [3] in Figure 2.2. The experiments were carried out using the germanium spectrometer of ANNRI installed at the J-PARC/MLF facility. The data were obtained with a relatively thin sample applying the total energy detection principle. In addition, they were corrected for self-shielding and multiple interaction events. Hence, they can be considered as an estimate of the Doppler broadened cross-section. The cross-section multiplied by the square root of the energy and normalised to a cross-section of σ_γ = 749 b at 25.3 meV is plotted as a function of neutron energy in Figure 2.2. The experimental data of Harada et al. [3] are compared with the Doppler broadened cross-section derived from the parameters in JEFF-3.2 and with the one derived from the unbound states in JEFF-3.2 combined with an additional 1/v component. This

figure reveals that the energy dependence of the cross-section derived by Harada et al. [3] is consistent with the energy dependence of the cross-section recommended in JEFF-3.2. This can also be concluded by comparing the Westcott factor g_γ for a pure Maxwellian neutron spectrum at room temperature with kT = 25.3 meV. The Westcott factor derived from the resonance parameters in JEFF-3.2 and from the data of Harada et al. [3] are g_γ = 1.006 and 1.005, respectively. The cross-section data of Harada et al. [3] have been complemented below 0.01 eV with extrapolated data based on a 1/v shape. The amplitude of the 1/v component was derived from the experimental data in the region between 10 meV and 20 meV. If the contribution of the bound states is replaced by a pure 1/v contribution, a Westcott factor of g_γ = 1.04 is found. This suggests that the value g_γ = 1.05 recommended by Mughabghab [14] was derived by adding a 1/v contribution without verifying the consistency of this assumption with energy-dependent experimental data.

The cross-section data in the low energy region can also be verified by comparing the thermal capture cross-section and the resonance strengths ($g\Gamma_n$). Such a comparison can be applied when only the results of a resonance shape analysis are available. To verify the JEFF-3.2 data the ratios of the thermal capture cross-section and resonance strength of resonances with energy below 5 eV derived from experimental data and the values recommended in JEFF-3.2 were computed. These ratios derived from data of Lampoudis et al. [1] (transmission and capture), Derrien and Lucas [7] (transmission), Kalebin et al. [10] (transmission), Mendoza et al. [4] (capture) and Fraval et al. [9] (capture) are plotted as a function of neutron energy in Figure 2.3.

Figure 2.3. Ratio of the experimental thermal capture cross-section and the resonance strength as a function of energy



Note: Resonance strength is represented by Z_{exp} and the corresponding values recommended in JEFF-3.2 as $Z_{JEFF-3.2}$. The ratio is shown for the experimental data reported by Lampoudis et al. [1], Mendoza et al. [4], Derrien and Lucas [7], Kalebin et al. [10] and Fravel et al [9].

Source: JRC-Geel, 2019.

Evidently, the data of Lampoudis et al. [1] are in good agreement with the values recommended in the JEFF-3.2 evaluation. The resonance strengths for the resonances at 0.306 eV, 0.574 eV, 1.271 eV, 2.362 eV, 2.586 eV, 3.964 eV and 4.957 eV derived from the data of Mendoza et al. [4], which have not been included in the evaluation of Noguère et al. [2], are consistent with those in JEFF-3.2 and Lampoudis et al. [1]. The strengths derived by Mendoza et al. [4] are on average about 2.5% lower. This systematic difference is within the normalisation uncertainty of 2.0% and 2.6 % of the data of Lampoudis et al. [1] and Mendoza et al. [4], respectively. The data of Mendoza et al. [4] were normalised independently of any transmission or capture data for ^{241}Am. Hence, they confirm the resonance strengths derived by Lampoudis et al. [1] and support the normalisation procedure applied by Noguère et al. [2]. Unfortunately, the data are limited to energies above 0.2 eV and do not provide information about the cross-section at thermal energy.

The capture data of Fraval et al. [9] and the transmission data of Derrien and Lucas [7] and Kalebin et al. [10] show a substantial reduction in thermal capture cross-section and resonance strengths compared to the values recommended in JEFF-3.2. The data of Derrien and Lucas [7] and Kalebin et al. [10] resulted from transmission measurements with powder samples. In the papers of Derrien and Lucas [7] and Kalebin et al. [9], no special analysis procedure to account for the particle size distribution is mentioned. Therefore, their data suffer from a systematic underestimation of the cross-section and resonance strength due to the use of powder samples. A comparison of the total widths in [1] confirms that Derrien and Lucas [7] and Kalebin et al. [10] deduced larger total widths from their data compared to the widths of Lampoudis et al. [1]. For the systematic difference between the thermal capture cross-section and resonance strengths derived by Fraval et al. [9] and those of JEFF-3.2 no direct explanation can be given.

From this discussion it can be concluded that only the data of Lampoudis et al. [1] and Fraval et al. [9] can be used without any additional correction to derive the capture cross-section at thermal energy. From their data (σ_γ = 749 (35) b [1] and σ_γ = 678 (68) b [9]), a weighted average σ_γ = 734 (31) b is derived and listed in Table 2.1. This value together with the resonance parameters can still be improved by performing a resonance shape analysis including all time-of-flight cross-section data, which do not suffer from systematic effects or which can be corrected for identified bias effects. For example, the data of Kalebin et al. [10] can be included by performing a detailed analysis of the resonance profiles using the model applied by Becker et al. [8] to obtain information about the sample properties. The data of Jandel et al. [5] can be included once they are corrected for the bias related to the flux determination.

Table 2.1. Thermal capture cross-section of ^{241}Am based on TOF measurements

TOF methods	σ_0 [b]	Comments
Lampoudis et al. [1]*	749 (35)	
Fraval et al. [9]**	678 (68)	
Terada et al. [15]***	707 (29)	
Weighted mean of references [1,9] (δ)	734 (31)	δ = max(δ1, δ2), δ1 = 31 b, δ2 = 29 b
Weighted mean of references [1,9,15] (δ)	720 (21)	δ = max(δ1, δ2), δ1 = 21 b, δ2 = 17 b

Note: The definitions of δ, δ1 (an internal uncertainty of measurements), and δ2 (an external uncertainty of measurements) are given in Appendix A.

Source: *Lampoudis et al., 2013; **Fraval et al., 2014; ***Terada et al., 2018.

After closing the Subgroup 41, an absolute measurement of the cross-section for ^{241}Am(n,γ) has been published [15], which was measured at the ANNRI facility of J-PARC. The thermal cross-section is σ_γ = 707 (29) b, which is consistent with the weighted mean value σ_γ = 734 (31) b within their uncertainties. The weighted average including the result of [15] contributes to reduce the uncertainty, and the mean value is σ_γ = 720 (21) b. Although for the experiments in [15], a powder sample was used, the effect discussed in [8] can be neglected because the thickness of the sample is small enough, and furthermore the absolute value of the capture cross-section was determined without using their transmission data that determined the total cross-section. For the evaluation of the total cross-section using transmission data, a careful study on the effect should be encouraged.

References

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3. Spectrum-averaged data

In contrast to energy-dependent data obtained by neutron time-of-flight measurements, spectrum-averaged data are derived from integral measurements with neutrons of energies within a certain band width. In order to deduce the cross-section at 25.3 meV from the spectrum-averaged data, the information on both the energy-dependent cross-section and the neutron energy spectrum are required; the absolute value of the cross-section is not necessarily required. If the thermal cross-section value is determined accurately, it serves as a point of anchorage in energy-dependent data evaluation.

Various techniques have been developed to deduce thermal capture cross-sections, such as the neutron activation method with Westcott convention and the pile-oscillation method. The Westcott convention was developed to approximate the well-moderated reactor neutron spectrum. It is widely utilised to deduce capture cross-sections at thermal energy and resonance integrals from activation measurements. One of the advantages of activation methods using reactor neutrons is the availability of high neutron flux, which enables the cross-section measurement for highly radioactive samples since only small amounts of the measured material in samples are needed. The other advantage of activation methods is the availability of precisely known capture cross-section at 25.3 meV for some standard reactions, such as ^{197}Au(n,γ)^{198}Au (98.65 (9) b), ^{59}Co(n,γ)^{60}Co (37.18 (6) b).

Recently, cold neutron beams have also been utilised for activation measurements. Although energy-dependent data are required to bridge between cold neutron energy and thermal energy, the potential advantage of utilising cold neutron beams is the extremely low epithermal neutron background in their beamlines. The measurements utilising cold neutron beams are reviewed in Section 3.1. Some of the identified bias effects have been indicated and the data were re-evaluated.

In Section 3.2, activation experiments using reactor neutrons with the Westcott approximation analysis were re-evaluated by taking into account the identified systematic bias effects such as a strong deviation of the ^{241}Am capture cross-section from a 1/v dependence due to low-energy resonances and bound states with an energy close to the neutron separation energy. Before these corrections were done, there had been a severe fragmentation between the data, which generated a large external uncertainty. The re-evaluation solved the fragmentation problem effectively and resulted in a cross-section value with much smaller uncertainty.

The measurements by utilising pile-oscillation and Pu-ageing methods also need the energy-dependent information on both cross-section data and neutron spectrum to be used for validation of the thermal capture cross-section. The current status of these methods was reviewed in Section 3.3 and Section 3.4, and possible bias effects are discussed.

3.1 Activation measurements in a cold neutron beam

Many research reactors have integrated cold neutron beams typically filled with liquid hydrogen or deuterium at ~20 K in their moderator tanks enabling the extraction of 10^7 to 10^{10} cm^{-2} s^{-1} of ~0.7 nm to ~0.4 nm (~1.7 meV to ~5.1 meV) neutrons. The advantage of cold compared neutrons to thermal neutrons is their high performance on transportation achieved by ^{58}Ni-coated beam tubes due to enhanced total reflection performance. Hence, cold neutrons can be extracted from the reactor and transported to the experimental area with a low background condition. Cold neutrons can also be focused with lenses and super mirror guides, which allow experimentalists to enhance the flux further.

Cold neutron beams at research reactors have different characteristics concerning their intensity and energy distribution. In Figure 3.1, cold beams at the Budapest Research Reactor (BRR) in Hungary and at the Forschungsreaktor München II (FRM II) in Garching are displayed [1]. Whereas the beam in Budapest peaks at ~3.5 meV, the beam at the FRM II has a maximum at ~1.8 meV. An integral neutron capture rate can be related to the thermal capture cross-section by considering the energy dependence for an energy range from cold (1-5 meV) to thermal (25.3 meV) energy. The cross-section is deduced from reaction rate ratios to irradiated samples containing materials such as hydrogen, nitrogen, chlorine or gold, for which the thermal capture cross-sections are very well known and are considered as standards.

In order to deduce the capture cross-section using a cold neutron beam, a prompt gamma-ray analysis (PGA) method and an activation method have been utilised [2,3].

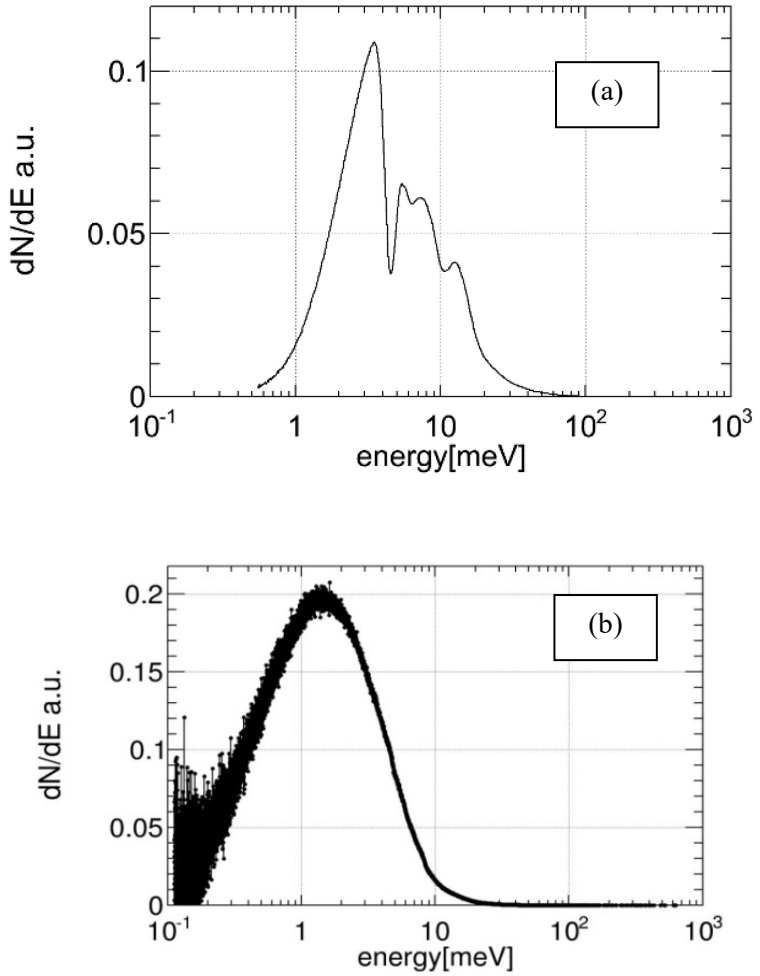
Sample configuration performed in a directed beam of cold neutrons using PGA or activation method needs careful consideration. As low-energy neutrons are scattered and absorbed in dense materials, samples should be sufficiently thin to enable accurate estimation of self-shielding and multiple-scattering correction factors in the samples. This is also crucial for the efficient detection of the resulting prompt gamma-rays with semi-conductor detectors, including high-purity germanium (HPGe) detectors, possibly Compton-shielded by bismuth germanium oxide (BGO) during irradiation. Detectors must be well energy and efficiency calibrated.

To ensure complete illumination of the material, samples should be smaller than the diameter of the beam. A comparator foil, mad of materials such as gold, of the same size should be co-irradiated to use the well-known thermal capture cross-section for flux determination.

Since the PGA method is difficult to utilise with heavy nuclei such as ^{241}Am, where prompt gamma-ray spectrum is expected to be complex, only activation measurements are discussed hereafter in this report.

If the energy-dependent cross-section does not follow the 1/v-dependence as a result of bound states with an energy close to the neutron separation energy, as existing in ^{241}Am (see Figure 2.2), the correction on energy dependence might become a significant systematic factor. The correction must be investigated for both target and monitor samples. In order to calculate the correction factor, the energy-dependent shape on cross-section is relevant, but the absolute value is not required.

Figure 3.1. Characteristics of cold neutron beams at (a) the Budapest Research Reactor (BRR) and (b) the Forschungsreaktor München II (FRM II)

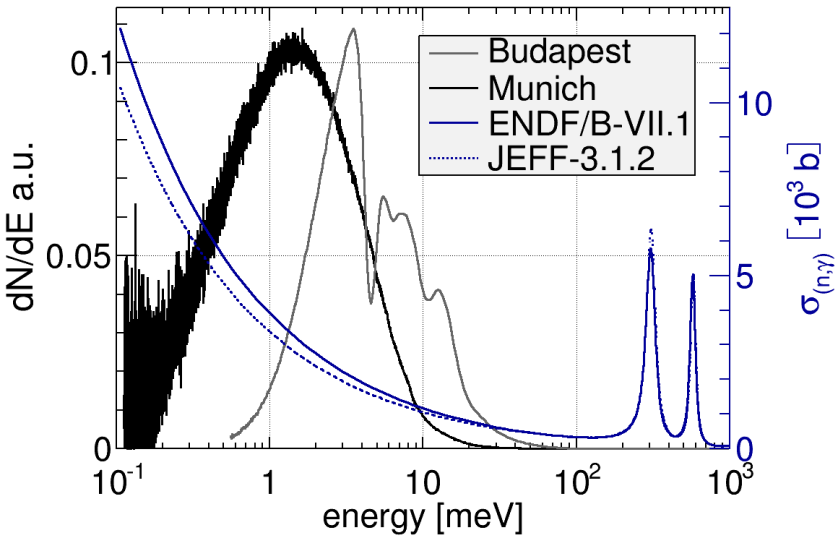


Note: The y-axes are in arbitrary units, as measured with compact chopper setups enabling time-of-flight measurements.

Source: FZJ, 2019.

As is shown in Figure 3.2, there is a large discrepancy between the Evaluated Nuclear Data File (ENDF)/B-VII.1 and JEFF-3.1.2 on the cross-section, which is prominent in the cold neutron region. Furthermore, it is difficult to evaluate the uncertainty of the cross-section at the cold neutron region since the energy region covered by time-of-flight measurements at accelerator facilities is limited to the region higher than 10 meV, even in the case of measurements performed at ANNRI. It should be noted that the large correction due to the energy-dependent shape on cross-section is needed and its uncertainty is not evaluated in detail. More discussions related to the bias effects are provided on the cross-section measurement using cold neutron beams at the BRR and the FRM II below.

Figure 3.2. Energy distribution of the cold beams at the Budapest Research Reactor (BRR) and the Forschungsreaktor München II (FRM II)



Note: The energy-dependent capture cross-section of ^{241}Am is also shown. Note that there is a discrepancy between ENDF/B-VII.1 and JEFF-3.1.2 on the cross-section in the cold neutron region.

Source: FZJ, 2019.

Measurement, analyses and results obtained at the Budapest Research Reactor (BRR) and the Forschungsreaktor München II (FRM II) for ^{241}Am

Two samples from ^{241}Am nitric solution were created by drying the solution on a circular 3 µm thick gold foil of 3 mm in diameter. The ^{241}Am activities were certified as 4.66 (4) MBq and 3.87 (3) MBq at the Physikalisch-Technische Bundesanstalt (PTB). The covered gold foils were hence sealed between two 4×4 cm Suprasil© Quartz sheets. In addition, blank samples were produced for both sample types. Due to the low mass, the ^{241}Am measurements were carried out at the FRM II. Blank sample irradiations were carried out for background characterisation.

From decay measurements after the irradiations, the thermal capture cross-section of ^{241}Am was deduced. An accurate assessment was performed by using MCNP5 and Geant4 simulation codes to perform neutron self-shielding and photon self-absorption calculations. The uncertainties in the sample dimensions were propagated through the simulations.

Evaluated neutron-induced nuclear data were used to calculate the simulated reaction rate within the samples via integration over the energy-dependent neutron capture cross-section. By comparing the simulated reaction rates in the actinide and the gold foil, a correction factor for the measured reaction rate ratio was derived.

For the ^{241}Am samples, the photon corrections were negligible. Although the neutron attenuation was below 1%, the significantly different shape of the cross-section between the JEFF-3.2 and the ENDF/B-VII.1 data set caused very different corrections for the reaction rate ratio, that is, 0.952 (4) using JEFF-3.2 data and 0.891 (4) using ENDF/B-VII.1 data. This discrepancy, at least about 7%, causes a significant systematic uncertainty on the overall resulting thermal radiative capture cross-section of ^{241}Am.

The thermal radiative capture cross-section of ^{241}Am was derived using the K_{α}, and $K_{\beta 1}$, $K_{\beta 2}$ X-rays following the decay of ^{242g}Am into ^{242}Pu, as no emission data on the few low energetic γ-rays of the decay exist. The emission probabilities of these X-rays were taken from the Decay Data Evaluation Project (DDEP); it should be noted that the uncertainty of these data is as large as about 5%, since these are based on calculations because of lack of measurements. The other difficulty of this measurement is the background subtraction. The K_{α} X-rays from the decay of ^{242g}Am are lying near the energies of ^{241}Am γ-rays and their contribution had to be subtracted. However, the measurements at the BRR and the FRM II were consistent within one standard deviation. The average of both measurements was 571 (26) b using the JEFF-3.2 based reaction rate correction and 610 (28) b using the ENDF/B-VII.1 based correction. Starting from this ground state production cross-section, the thermal capture cross-section of ^{241}Am was derived using the isomeric production ratio of 0.914 (7) measured by Fioni et al. [4] at thermal energies. This results in 626 (29) b using the JEFF-3.2 based reaction rate correction and 668 (31) b using the ENDF/B-VII.1 based correction. The correction factors are 0.891 for JEFF-3.2 and 0.952 for ENDF/VII.1, respectively.

It should be noted that there is no direct energy-dependent measurement covering the energy range from 1 meV to 25.3 meV. Therefore, a relatively large uncertainty on the correction factor needs to be considered to use the cold neutron activation data to deduce the thermal capture cross-section. The uncertainties shown in parentheses do not include this uncertainty. The utilisation of the isomeric production ratio measured at thermal energies might give an additional bias. The uncertainty due to this utilisation for cold neutron region should also be evaluated and added to the total uncertainty.

3.2 Neutron activation experiments

Irradiation experiments by reactor neutrons at MELUSINE and PHÉNIX

Irradiation experiments performed with reactor neutrons are often based on activation techniques. A sample is irradiated and the number of nuclei produced is quantified to evaluate the capture cross-section. This is similar to those using cold neutron beams described in Section 3.1. However, irradiation experiments differ from those in Section 3.1 in that a more intense neutron flux with longer irradiation time is used, which may significantly change the sample composition. Irradiation experiments by reactor neutrons are often associated with destructive measurements (e.g. nitric acid dissolution followed by a mass spectrometry analysis), while those using cold neutron beams mostly rely on non-destructive measurements (e.g. X-ray or gamma-ray spectrometry). This is why irradiation experiments are more complex to settle as they require hot cells and dedicated nuclear material transportation. On the other hand, as these experiments produce several decay products in the nuclide chain, they can give access to cross-section data for radioactive materials (e.g. ^{242m}Am) that would be very difficult to prepare for an activation experiment. Moreover, the use of destructive analyses avoids the use of radioactive decay data that may be the limiting factor in the uncertainty budget of neutron activation experiments.

The normalisation is often made by using not only standard activation foils, as described in Section 3.1, but also by using the build-up of one or several reference fission products. ^{145}Nd and/or ^{148}Nd are good candidates as their cumulative fission yield is known at better than 1.5%, they are stable and they have very small capture cross-sections, which avoids any additional uncertainty due to their disappearance.

More discussion related to the bias effects can be found on the cross-section measurement using activation method at MELUSINE and PHÉNIX below.

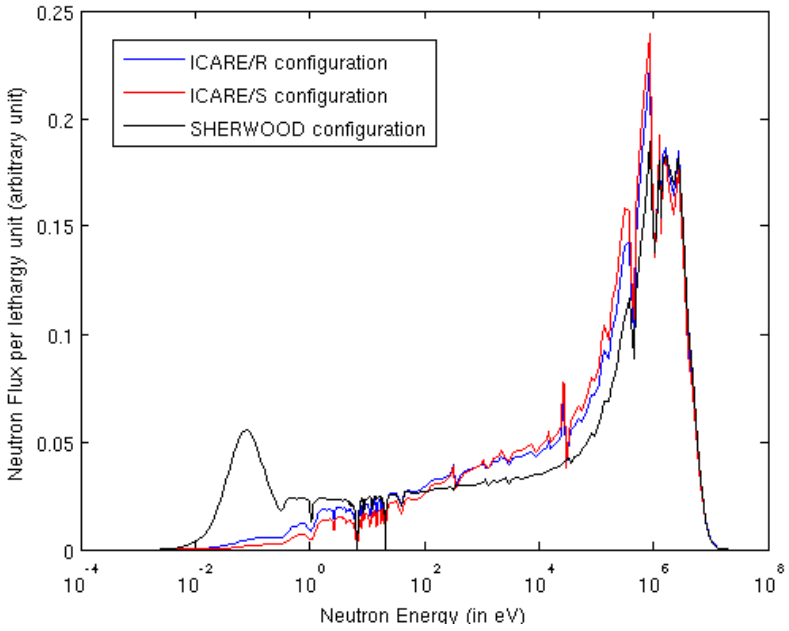
Results obtained at MELUSINE for ^{241}Am

MELUSINE is the 8 MW pool-type light water moderated reactor located in Grenoble, made with materials test reactor (MTR) highly enriched uranium assemblies. In the years 1980s, it was extensively used to perform irradiation experiments on various actinides and fission products relevant to light water reactor (LWR) applications. Three core configurations, named SHERWOOD, ICARE/R and ICARE/S, were considered in this analysis.

The SHERWOOD assembly consisted of a square lattice of 5×5 uranium rods; the central one being comprised of a stack of UO_2 depleted pellets. Some of these rods were doped with specific isotopes (^{239,240,242}Pu, ^{241,243}Am and ^{244}Cm). The ICARE/R and ICARE/S square assemblies were constructed with mixed oxide (MOX) fuel pins. They were loaded in a hexagonal lattice surrounded by natural cadmium screens. The volumetric moderation ratio was respectively ~0.9 (ICARE/R) and 0.5 (ICARES), providing an epithermal neutron flux with an increasing hardness from one configuration to the other one. Two central pins were made of a stack of UO_2 pellets and 34 of them were doped with the isotopes of interest (^{236}U, ^{237}Np, ^{239,240,242}Pu, ^{241}Am, ^{244}Cm, ^{149}Sm and ^{153}Eu).

An accurate modelling of the irradiation was made based on the APOLLO2 calculation code. Three dimensional correction factors were evaluated with TRIPOLI4 Monte Carlo calculations to account for shadow effects between the different doped pellets. A detailed uncertainty analysis was also performed to account for the different assumptions in the temperature profile, irradiation history and material compositions. More details on the experiments and the calculation methodology can be found in [5]. The corresponding neutron flux spectra, as calculated by the APOLLO2.8 code, are presented in Figure 3.3. Table 3.1 shows the ratio of calculation and experimental values (C/E) that were described in [5,6].

Figure 3.3. Neutron flux spectra in the various MELUSINE core configurations



Source: CEA, 2019.

Table 3.1. Integral trends for the ^{241}Am capture cross-section deduced from the MELUSINE irradiation experiments

	C/E-1 (%)					
	JEFF-3.1.1			JEFF-3.2		
Isotope	SHERWOOD	ICARE/R	ICARE/S	SHERWOOD	ICARE/R	ICARE/S
^{241}Am	-17.6 (22)	-21.4 (48)	-16.3 (25)	-3.0 (21)	-9.0 (48)	-4.0 (27)

Note: The C/E-1 quantities correspond to the reaction rates weighted with the spectra shown in Figure 3.3.

Source: CEA, 2019.

The measurements on ^{241}Am are consistent between the different irradiation experiments considered. The differences between the calculated and experimentally-determined values show an underestimation of about 16 (2) % on the energy-integrated capture cross-section in the case of JEFF-3.1.1. It should be noted that SHERWOOD and ICARE/S have completely different sensitivity profiles, the first one being related mostly to the thermal value and the first three resonances, while the second one is only focused on the energy range above 1 eV. The JEFF-3.2 significantly improves the previous results with a consistency of calculated values with the experiment within 2σ uncertainty.

Results obtained at PHÉNIX for ^{241}Am

The PROFIL and PROFIL-2 experiments, performed in the PHÉNIX sodium-cooled fast reactor, consist in the irradiation of 130 small separate samples containing almost pure isotopes of major and minor actinides and several fission products. Their analysis was carried out using the ERANOS-2.1 code system and various versions of the JEFF-3 library. More details on the experiment and calculation procedure can be found in [7]. The spectrum, as calculated by the ERANOS code, is presented in Figure 3.4.

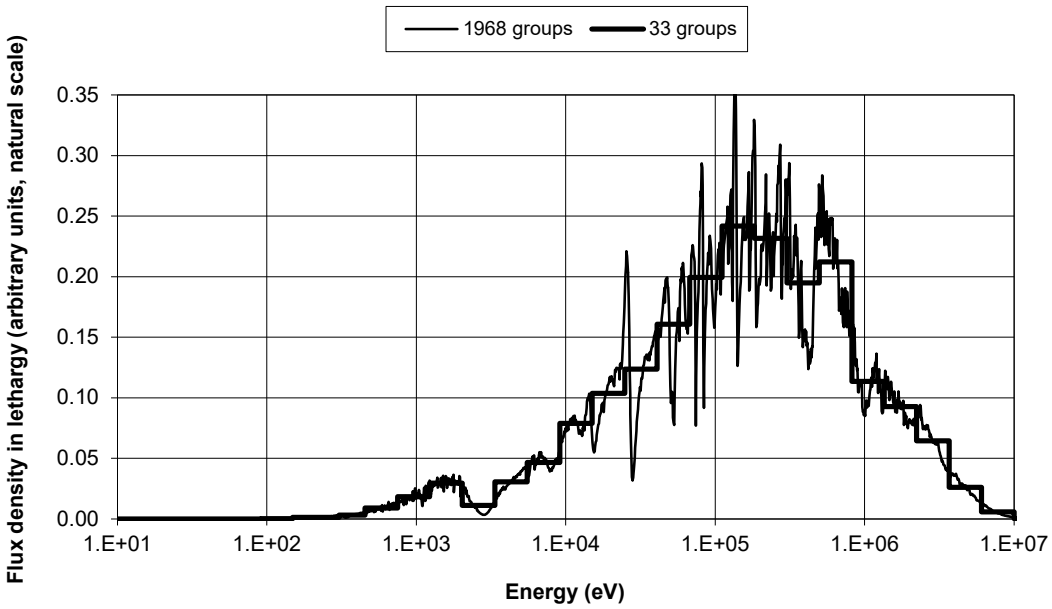
The JEFF-3.1 evaluation provided better agreement with the experiment for the ^{241}Am capture cross-section, compared with JEFF-3.0. These fast neutron irradiation experiments are relevant for validation purpose as well. Results for the ^{241}Am capture cross-section are shown in Table 3.2.

Table 3.2. Integral trends for the ^{241}Am capture cross-section deduced from the PHÉNIX irradiation experiments

Isotope	C/E-1	
	JEFF-3.0	JEFF-3.1
^{241}Am	-7.6 (5) %	-0.8 (5) %

Source: CEA, 2019.

Figure 3.4. Neutron flux spectra in the PHÉNIX reactor



Source: CEA, 2019.

3.3 Pile-oscillation experiments

The pile-oscillation technique is one of the oldest experimental techniques to measure integral cross-sections [8]. The basic principle is to measure the neutron flux modulation due to a periodic insertion of a sample inside a critical reactor. The standard perturbation theory, an efficient tool used by reactor physicist for uncertainty/sensitivity analysis, provides also an exact formalism to analyse the data by relating the reactivity change to the variation of Boltzmann operator H, which includes the different partial cross-sections:

$$\Delta\rho = \frac{<\phi_1^*, \delta H \phi_2>}{<\phi_1^*, P_2 \ \phi_2>}, \quad (1)$$

where ϕ_1^* is the adjoint flux in the situation where the sample is out of the core, (index '1'), ϕ_2 is the forward flux in the situation where the sample is inside the core (index '2'), P_2 is the production operator of the Boltzmann equation, which is related to the total production of neutron by fission.

In specifically defined conditions (flat flux at the oscillation position, energy independent adjoint flux, etc.), the reactivity change can be attributed to only the neutron capture cross-section.

As most other techniques, this method is essentially relative, which means that it has to be normalised to a sample with reference nuclear data. This is preferred to absolute reactivity measurements because the determination of the denominator of the equation (1) would introduce larger uncertainties than considering a ratio of reactivity worth where this term will be cancelled. This is due to the fact that the sample insertion/withdrawal does not change the total neutron production rate by more than 0.1%. The normalisation is

commonly made using samples of ^{235}U, ^{10}B or ^{6}Li, because their fission and absorption cross-sections are well-known and referred to as neutron cross-section standards.

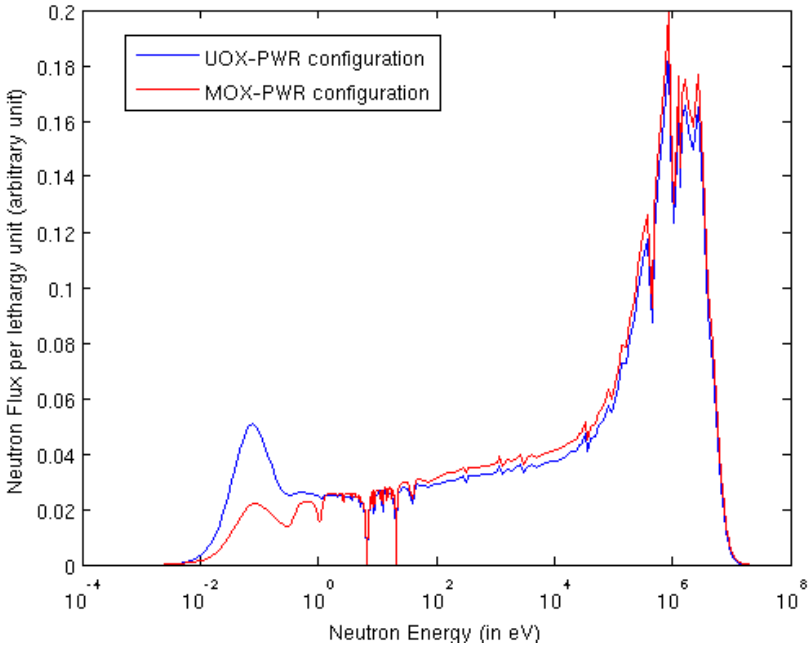
Very careful attention should be paid to the material balance of the sample, especially the number of atoms and the sample homogeneity, which has the same kind of impact as in neutron activation experiments. These uncertainties are the limiting factor for the experiment, and usually provide a 1-3% bias (1σ). Compared to the measurement uncertainty (mainly linked to neutron and electronic noise) of less than 1.0%, and to the neutron cross-section standard uncertainty of less than 0.5%, the sample description is often the dominant contribution. The overall uncertainty on the energy-integrated cross-section is typically 3-4%.

Compared with neutron activation experiments, the pile-oscillation technique has the advantage of not relying on the knowledge of any radioactive decay data. Nevertheless, these experiments are often considered very complex to analyse due to very small reactivity worth of the oscillated sample (typically in the range of 1 pcm to 10 pcm, with 1 pcm = 10^{-5}), which precludes the use of Monte Carlo methods. Consequently, deterministic codes (e.g. APOLLO, WIMS, CASMO) are used for the analysis of these experiments, which involve simplifications in the geometrical modelling and in solving the Boltzmann equation (e.g. multigroup approach, Legendre polynomials for the scattering law). These simplifications introduce some numerical bias that should be evaluated and reduced as much as possible. This can be done by an extensive comparison with Monte Carlo methods on simplified benchmarks. This step is crucial to avoid the introduction of numerical bias in the calculation to experiment comparison, which is intended to provide feedback on nuclear data. This is why very old pile-oscillation experiments, such as those from Pomerance for ^{241}Am [9], must be considered carefully as the authors often did not rigorously document their modelling simplifications used to evaluate the related numerical errors.

Results obtained at MINERVE for ^{241}Am

A series of different experiments was performed in the MINERVE reactor from 2005 to 2015 to improve the knowledge of, and to provide realistic uncertainty on, the capture and fission cross-sections of major and minor actinides ranging from ^{232}Th to ^{245}Cm. These experiments are referred as the Experiments Quantifying Integral Cross-sections (OSMOSE) programme, which was firstly presented in [10] and with preliminary analysis presented in [11]. These experiments used natural UO_2 samples, which include separated actinide oxides. For ^{241}Am, two samples were manufactured with 600 mg of ^{241}Am included in the first one and 200 mg in the second one. Experiments were performed in two core configurations, with the one typical of a uranium oxide pressurised water reactor (PWR) spectrum and one typical of a MOX-PWR spectrum (including a harder neutron spectrum). The neutron spectra calculated by APOLLO2.8 are presented in Figure 3.5.

Figure 3.5. Neutron flux spectra in the two MINERVE core configurations



Source: CEA, 2019.

The most recent published results were presented at the NEA JEFF Meeting in April 2013 [12]. Since then, the analysis was slightly revised to account for a small correction due to adjoint flux calculations, which were performed before by the APOLLO2 deterministic code. This correction, which is linked to over-simplification of the geometrical model of the experiment, involves an increase of 1.5% of the calculated reactivity worth of ^{235}U samples, which are used to normalise the data. Moreover, thanks to the recent implementation of an exact perturbation method in the TRIPOLI4 code, any simplification was removed due to the use of deterministic methods compared with previous results.

Table 3.3 provides the recently updated data. The neutron activation results are also included, which have been performed with the same samples. For the pile-oscillation results, the C/E comparison is more than 98% related to the energy-integrated capture cross of ^{241}Am, due to the negligible effect of scattering (<0.1%) and fission cross-sections. It should be noted that the reference ^{235}U fission cross-section was taken from JEFF-3.1.1.

Table 3.3. Integral trends for ^{241}Am reactivity worth deduced from the OSMOSE pile-oscillation experiment

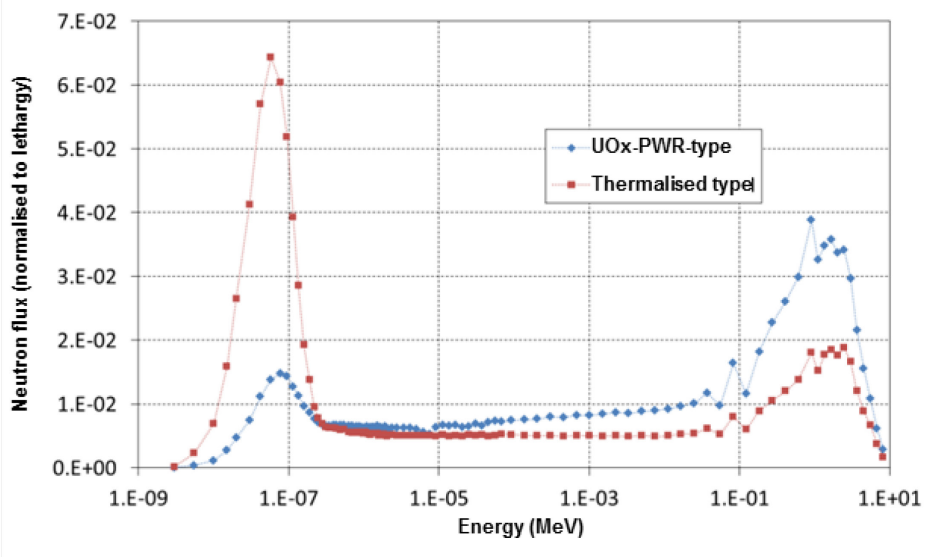
Isotope	C/E-1			
	JEFF-3.1.1		JEFF-3.2	
	UOx-PWR spectrum	MOX-PWR spectrum	UOx-PWR spectrum	MOX-PWR spectrum
^{241}Am	-7.0 (22) %	-6.4 (24) %	+5.7 (22) %	+5.8 (28) %

Source: CEA, 2019.

For ^{241}Am, the results show an average 6.7% underestimation with JEFF-3.1.1, and an average 5.7% overestimation with JEFF-3.2, as shown in Table 3.3. Considering that almost 50% of absorption occurs below 0.1 eV (more than 30% in the first three resonances), it would be unrealistic to derive a thermal capture cross-section for ^{241}Am from these integral experiments. However, these results tend to indicate that a thermal cross-section value of 700 b would better fit the experiment than the 650 b used in JEFF-3.1.1 or the 749 b used in JEFF-3.2.

It should be noted that these results are not consistent with the trends deduced from thermal-neutron irradiation in the MELUSINE facility. To overcome this disagreement, a new experiment, referred hereafter as the "AMSTRAMGRAM programme", was proposed in the framework of the CHAllenges in Nuclear DAta project (CHANDA) European Project on Nuclear Data, in collaboration between CEA, Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT) and JRC Geel. The starting point was to use ^{241}Am samples that were available at JRC Geel to perform pile-oscillation experiments. These samples, which are made of sintered pellets of Al_2O_3 mixed with ^{241}Am oxide (~40 mg of ^{241}Am per sample), were initially manufactured for (n,2n) measurements at the JRC Van De Graaff, but were also used for capture measurements at n_TOF at CERN. Having the same sample used in various experiments is an appropriate way to solve long-lasting inconsistencies between integral and energy-dependent experiments. Moreover, as previous MINERVE experiments performed in the UOx-PWR core configuration had a significant component due to the resonance integral, it was decided to define a dedicated core configuration to emphasise the neutron absorption of ^{241}Am below 0.1 eV due to an over-thermalised configuration (see Figure 3.6). This dedicated core configuration was obtained by removing several layers of UO_2 fuel pins from the reactor, which have been replaced by light water holes around the central guide tube for oscillations.

Figure 3.6. Neutron flux spectra in the OSMOSE (UOx-PWR) and AMSTRAMGRAM (thermalised) core configurations



Source: CEA, 2019.

In order to minimise any calculation bias due to the normalisation to a reference sample, it was decided that the use of metallic foils of gold and lithium with similar geometries as the ^{241}Am samples would be ideal to calibrate the pile-oscillation experiments. These standard materials have the same absorption behaviour as ^{241}Am and, compared to ^{235}U, ^{241}Am is less complicated to handle as ^{235}U reactivity worth involves significant contributions from capture and fission cross-sections, neutron total multiplicity and fission neutron spectra.

The experiments were performed from November 2015 to March 2016. Several neutron spectrum characterisation measurements (e.g. dosimetry, spectral index, cadmium ratio) were performed to assess the ratio of epithermal to thermal neutrons. The finalised C/E comparison is presented in Table 3.4.

Table 3.4. Integral trends for ^{241}Am reactivity worth in a well-thermalised neutron spectrum

Isotope	C/E-1	
	JEFF-3.1.1	JEFF-3.2
^{241}Am	-5.4 (22) %	+8.8 (22) %

Source: CEA, 2019.

These results confirm that an intermediate cross-section value ranging between the 650 b of JEFF-3.1.1 and the 750 b of JEFF-3.2 would be in better agreement with the overall set of experimental data.

Both OSMOSE and AMSTRAMGRAM experiments can be considered as “clean” integral measurements and can be used for validation purposes. AMSTRAMGRAM should be preferred to validate the thermal capture cross-section value as the experiment was specifically designed for that purpose.

3.4 Plutonium ageing experiments

Plutonium ageing (Pu-ageing) experiments are based on the measurement of the reactivity change of MOX fuel critical configurations due to the beta decay of ^{241}Pu to ^{241}Am. The increase in the criticality with the number of long time-elapsed MOX fuels gives useful information to improve the evaluation of cross-sections of ^{241}Am and ^{241}Pu.

Such experiments were performed in the MINERVE facility, based on the oscillation of a pure ^{241}Pu sample at various times between 2006 and 2017. Pu-ageing experiments were also performed in the EOLE critical mock-up with the MISTRAL2 and FUBILA 100% MOX core configurations, using critical size adjustment over the programme time period.

These experiments were analysed with TRIPOLI4 continuous energy calculations. All the results were presented in [5] and are gathered in Table 3.5.

Table 3.5. Integral trends for Pu-ageing experiments

Experiment	JEFF-3.1.1	JEFF-3.2
MINERVE/OSMOSE [2006->2009]	-10.0 (30) %	-5.1 (30) %
MINERVE/OSMOSE [2006->2017]	-7.3 (15) %	-1.2 (15) %
EOLE/FUBILA	-3.1 (40) %	5.3 (40) %

Source: CEA, 2019.

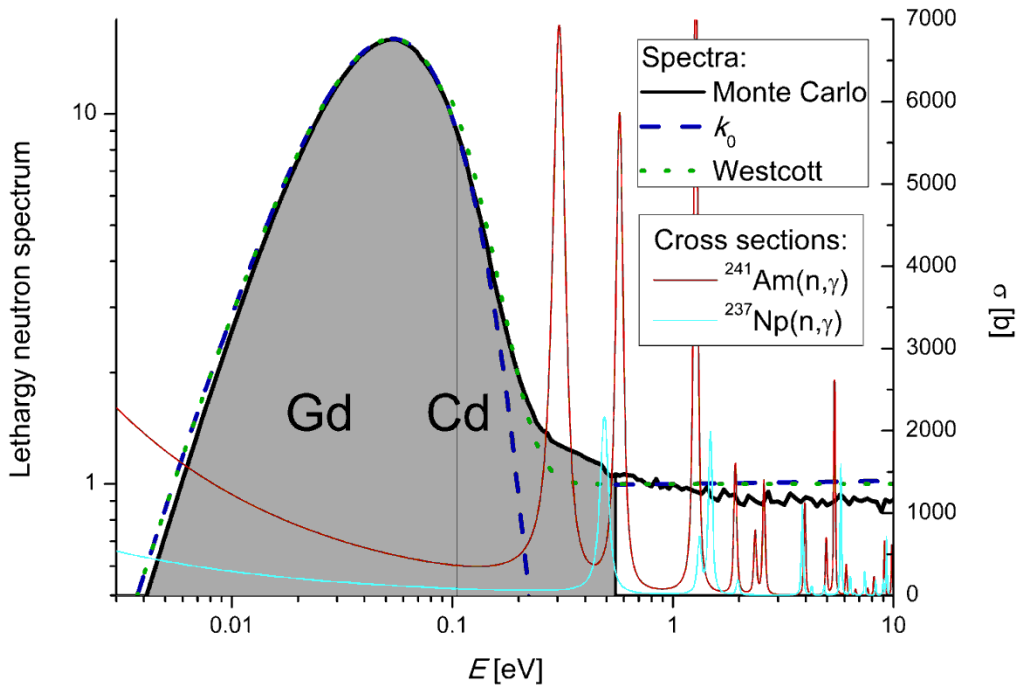
On average, the results obtained with JEFF-3.2 better match the experimental data than the results obtained with JEFF-3.1.1 data. The 30% contribution to reactivity due to ^{241}Pu was independently validated in JEFF-3.1.1 using the pile-oscillation of the ^{241}Pu sample, before any significant build-up of ^{241}Am. It was reported to be consistent with the experiment 0.6 (48) % [11].

These Pu-ageing experiments can also be considered as "clean" integral experiments, but with a larger uncertainty than the other integral experiment techniques that were presented before.

3.5 Systematic study to estimate bias effects and correct data

A systematic study was performed to estimate the possible biases in thermal capture cross-section values derived from neutron activation measurements using conventional methods such as the k_0 standardisation method [13] and the Westcott convention [14] as implemented in [15-16]. Both methods are relatively crude, since they approximate the neutron spectrum with only two free parameters (three in case variations in spectrum temperature are allowed). The approximation works with cadmium transmission filters for most materials following the 1/v dependence of the capture cross-section below the cadmium-cut-off energy at 0.55 eV. However, there are resonances in that energy region for ^{241}Am, which introduce biases in the reaction rates, as shown in Figure 3.7. The use of gadolinium filters with an effective cut-off energy at around 0.1 eV can effectively solve this problem. Except the activation measurements by Nakamura et al. [15], such experimental data are not available and correction methods for existing activation measurements were proposed [17,18].

Figure 3.7. Comparison of the Monte Carlo calculated spectrum and approximate 2-parameter spectra for a typical irradiation channel in a research light water reactor



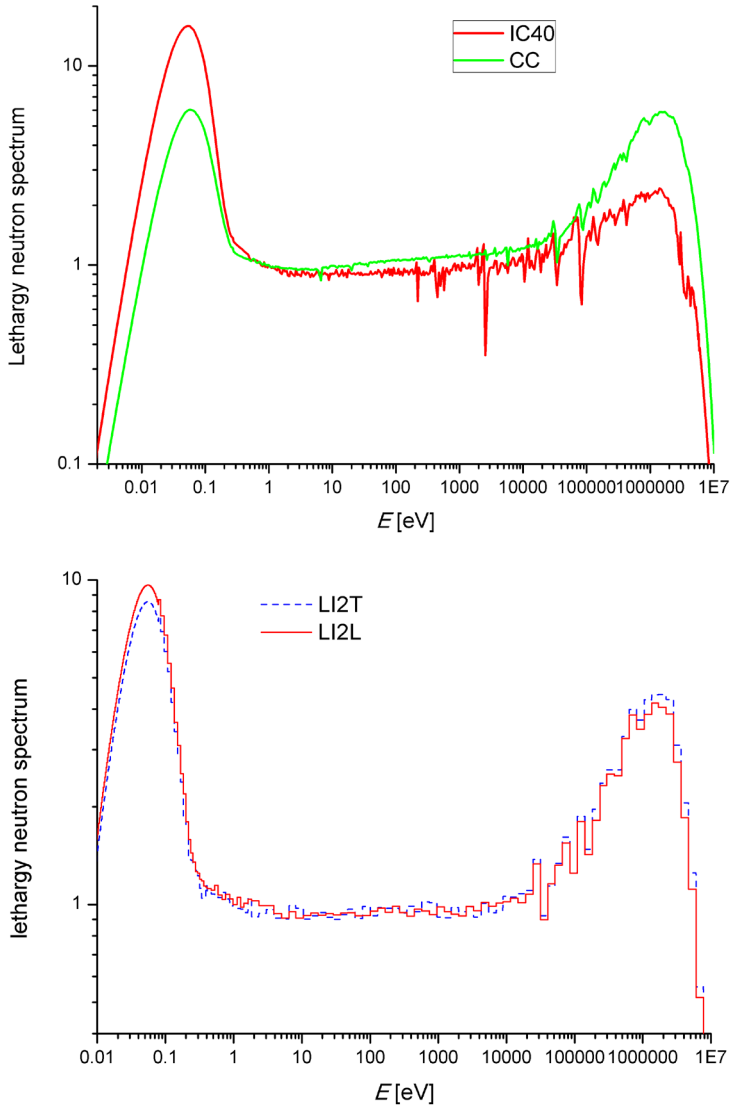
Note: Energy regions relevant for cadmium and gadolinium transmission filter methods are indicated. Capture cross-sections for ^{241}Am are also shown to emphasise the importance of the systematic error made by assumption of the analytical spectra.

Source: JRC-Geel, 2019.

A general method [18] based on Monte Carlo (MC) calculated reactor spectra (see Figure 3.8) and the JEFF-3.2 cross-section library was used to calculate reference reaction rates for measured reactions and standards (^{197}Au(n,γ), ^{59}Co(n,γ), etc.), irradiated bare and under cadmium and gadolinium filters. Even though the method is general, results for ^{241}Am only are presented here in Table 3.6. All spectra are normalised to 1 at 1 eV.

Adopting the reaction rates from Table 3.6 as reference "experimental" data, k_0 and Westcott methods have been used to derive capture cross-section at thermal energy (25.3 meV) σ_0. The biases are produced as a result of limitations in methodologies are presented in Table 3.7.

Figure 3.8. Monte Carlo calculated spectra in irradiation channels of the JSI TRIGA (top) and KUR (bottom) reactors



Source: JRC Geel, 2019.

Table 3.6. Reaction rates R from normalised MC spectra and assuming JEFF-3.2 cross-sections

Irradiation channel	R (^{241}Am(n, γ)) [b eV]		
	bare	1 mm Cd	35 µm Gd
IC40	21 900	1 510	5 000
CC	10 700	1 560	4 210
LI2T	14 100	1 570	4 670
LI2L	15 500	1 640	4 930

Source: JRC Geel, 2019.

Table 3.7. Relative biases in the derived σ0 due to approximations in methodologies

Irradiation channel	^{241}Am(n, γ) rel. bias in σ_0		
	k_0 Cd	Westc. Cd	Westc. Gd
IC40	8.9%	10.3%	0.0%
CC	21.1%	24.7%	1.1%
LI2T	8.1%	18.6%	0.7%
LI2L	15.0%	17.2%	0.6%

Note: The reference σ_0 values is 748 b for ^{241}Am.

Source: JRC Geel, 2019.

For ^{241}Am, a systematic trend of significant overestimation of σ_0 for activation method using cadmium filters is observed. The magnitude of overestimation mainly depends on the ratio of the thermal spectrum peak to the epithermal spectrum component. This is due to inability of the analytical methods to consider the contribution of the epithermal neutron spectrum component to the reaction rate between about 0.1 eV and 0.55 eV, which is significant for ^{241}Am due to the presence of resonances below 0.55 eV. The bias is larger if the epithermal component of the neutron spectrum is stronger (corresponding to a lower thermal Maxwellian peak). Use of a gadolinium filter with an effective cut-off energy around 0.1 eV, the problem of the bias is practically non-existent. This method was used by Nakamura [15].

An independent study [17], based on analytical corrections to the Westcott formalism due to deviation of the neutron spectrum from idealised shape, yielded similar correction factors for ^{241}Am(n,γ) cross-section at 25.3 meV as the above described numerical method. The results of both studies applied to activation measurements on ^{241}Am are summarised in Table 3.8. The results in the column "MC (Monte Carlo) corrected σ_0" were also corrected for the deviation of the transmission filter from ideal and the generalised Westcott factor. The correction relies on the JEFF-3.2 library, while the corrections in [17] rely on the JENDL-4.0 library. Differences between resulting cross-sections by [17] and [18] are about 5 b. This uncertainty can be neglected since it is much smaller than the uncertainty of the weighted mean average, which is 15 b. The averaged weighted mean of [17] and [18] gives

707 (15) b. A similar systematic study of spectrum-averaged experimental data was carried out by Žerovnik et al. [23]. A re-analysis of the data that provided enough information to apply the methodology proposed by Žerovnik et al. [23] resulted in an average capture cross-section of 716 (16) b. In the work of Žerovnik et al. [23], not only the effect of the bound states with energy close to the neutron separation energy but also a refined model including a joining function was included. Selecting only those data sets for which enough information is available to apply the methodology was proposed by Žerovnik et al. [23]. Therefore, this version of the re-analyses [23] is adopted as the weighted mean value for activation measurements.

Table 3.8. Original and corrected values for ^{241}Am(n,γ) cross-section at 25.3 meV from neutron activation measurements

Author(year)	Original σ_0 [b]	Corr. [17] σ_0 [b]	MC corr. [18] σ_0 [b]	MC corr. [23] σ_0 [b]
Bak (1967) [19]	740 (60)	691 (60)	672 (64)	
Harbour (1973) [20]	832 (18)	*	694 (73)	
Gavrilov (1976) [21]	853 (52)	725 (52)	725 (48)	708 (43)
Shinohara (1997) [16]	854 (58)	738 (52)	727 (60)	720 (50)
Bringer (2007) [22]	705 (23)	705 (23) **	710 (23)	714 (23)
Nakamura (2007) [15]	687 (25)	687 (25) **	718 (28)	721 (28)
Weighted mean (δ)	768 (69)	702 (15)	712 (15)	716 (16)
(δ1)	(12)	(15)	(15)	(16)
(δ2)	(69)	(15)	(12)	(3)
Averaged weighted mean of references. [17] [18]		707 (15)		
Adopted weighted mean value [23]				716 (16)

Note: The definitions of δ1 (a weighted uncertainty of measurements), and δ2 (a standard deviation of measurements) are given in Appendix A.

*: Not considered, **: Not corrected.

Sources: Nakamura et al., 2007; Shinohara et al., 1997; Mizuyama et al., 2017; Žerovnik et al., 2017; Bak et al., 1967; Harbour et al., 1973; Gavrilov et al., 1976; Bringer et al. 2007; Žerovnik et al., 2018.

3.6 Summary of spectrum-averaged data

Several types of integral measurements have been discussed by the Subgroup 41 participants:

- activation by cold neutron beams;
- neutron activation experiments with reactor neutrons;
- pile-oscillation experiments;
- plutonium ageing experiments.

Regarding activation measurements utilising cold neutron beams, it was pointed out that a large correction factor needs to be considered since the energy-dependent cross-section does not follow the 1/v-dependence as a result of bound states with energy close to the neutron separation energy in the case of ^{241}Am. For example, the correction factors are 0.95 in the case of JEFF-3.2 data but 0.89 in the case of ENDF/B-VII.1 data. Therefore, there is at least a 7% ambiguity due to this factor. It should be noted that there is no direct, energy-dependent measurement covering the energy range from 1 meV to 25.3 meV. Therefore, a careful assessment of the correction factor is required. The bias due to utilising the isomeric production ratio measured at thermal energies was also pointed to be included as an additional bias. The uncertainties of X-ray emission probabilities used to determine the amount of the decay product are also not negligible and amount to about 5%. Therefore, at least 9% uncertainty needs to be considered. Since this uncertainty is much larger than that of evaluated values from activation experiments in Section 3.5, the cold neutron data was not included in the evaluation of the spectrum-averaged data.

Utilisation of cold neutron beam has a potential to improve the accuracy of the capture cross-section because cold neutrons are easier to be shielded, giving high signal to noise ratios in neutron fields as described in Section 3.2. However, the energy-dependent data bridging the cold neutron region and thermal-neutron region is required in order to extrapolate the cold neutron data to thermal energy. Such an energy-dependent measurement is encouraged, covering the energy region from about 0.5-1 meV to 25 meV. Accuracy improvement of the relevant emission probabilities of X-rays should be also encouraged. The activation measurement using cold neutron beams together with these relevant data refinement activities on bound states with energy close to the neutron separation energy and decay data should also be considered for future experiments.

Between data reported from neutron activation experiments by reactor neutrons there were large discrepancies. However, Subgroup 41 and independent studies found that the physical reasons for the discrepancies could be identified and corrected by re-analysing the published data in detail. The discrepancies have been significantly reduced. A re-analysis of the data that provided enough information to apply the methodology proposed by Žerovnik et al. [23] resulted in an average capture cross-section of 716 (16) b, which was adopted as the weighted mean value for activation measurements as discussed in Section 3.5.

Pile-oscillation experiment supported the value of about 700 b to 750 b rather than 650 b in JEFF-3.1.1. Recent advancements to remove some numerical bias by utilising detailed Monte Carlo methods were reviewed, and pointed out shortcomings in the analysis of very old pile-oscillation experiments, such as the one from Pomerance for ^{241}Am [8]. The ongoing programme utilising the pile-oscillation method has a potential to validate the cross-sections with uncertainty of about 2-3% (1σ). The final results are expected to be published and cross-checked with the spectrum averaged data by Subgroup 41 participants.

The Pu-ageing experiments on average supported the JEFF-3.2 results more than the JEFF-3.1.1 evaluation. These Pu-ageing experiments can be considered as “clean” integral experiments, but with a larger uncertainty than the other integral experiment techniques that were presented in other sections. Therefore, the data was not included in the evaluation of the spectrum-averaged data.

The results of the spectrum-averaged data are summarised in Table 3.9. The average of re-evaluated activation data was adopted as the thermal capture cross-section of ^{241}Am based on the spectrum-averaged measurements and is 716 (16) b.

Table 3.9. Summary of the spectrum-averaged measurements

Methods	Ave. of σ_0 [b]	Comments
Activation by cold neutron beam		Large uncertainty due to the lack of energy-dependent data connecting cold neutron energy and thermal-neutron energy
Neutron activation experiments	716 (16)	
Pile-oscillation experiments		Support the value of about 700-750 b
Pu-ageing experiments		Support the value of about 700-750 b
Weighted mean (δ) (δ1) (δ2)	716 (16) (16) (3)	

Source: OECD/NEA, 2019.

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4. Relevant decay data

Nuclear structure data, especially gamma-ray and X-ray emission probabilities, play an important role in determining the neutron capture cross-section by an activation method as discussed in Section 3.2. On gamma-ray and X-ray emission probabilities, the table of isotopes [1] or nuclear data sheets [2] are standard reference resources. There are also several websites available, as summarised in Table 4.1, including the Decay Data Evaluation Project (DDEP) and Evaluated Nuclear Structure Data File (ENSDF), which are hosted by national laboratories and the International Atomic Energy Agency (IAEA).

Table 4.1. Websites on nuclear decay data

Name of library	Organisation	Websites
Nuclear Data Sheets	Elsevier	www.sciencedirect.com/science/journal/00903752
Chart of Nuclides	IAEA	www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html
ENSDF	BNL	www.nndc.bnl.gov/ensdf/ or www.nndc.bnl.gov/nudat2/
DDEP	LNHB	www.bipm.org/fr/publications/monographie-ri-5.html

Source: OECD/NEA, 2019.

The current status of decay data relevant to the capture cross-section of ^{241}Am determination by activation method is briefly summarised below.

Following neutron capture in ^{241}Am, ^{242g}Am is the primary product. It decays via 2 β-transitions to ^{242}Cm (83.1% or 82.7%) and 2 electron capture (EC) transitions to ^{242}Pu (16.9% or 17.3%). The two resulting γ-rays are of very low energy (42.13 keV and 44.54 keV, respectively) and are highly converted. The Internal Conversion Coefficients (ICC) are 1 155 and 748, respectively. The only radiation that can be sensibly measured are the K X-rays from ^{242}Pu. The $K_{\alpha 2}$ line is at 99.525 keV with an absolute intensity of 3.55 (17) % or 3.6 (3) % and the $K_{\alpha 1}$ line is at 103.374 keV with an absolute intensity of 5.6 (3) % or 5.7 (4) %. Both these lines coincide with γ-rays coming from ^{241}Am (98.97 keV and 102.98 keV) and are difficult to be de-convoluted. Therefore, these γ-rays coming from ^{241}Am were subtracted by measurement of γ-ray spectrum after long cooling time.

It should be noted that there is large difference between evaluated nuclear decay data on uncertainties on K X-rays from ^{242}Pu. The situation is summarised in Table 4.2.

Table 4.2. Evaluated nuclear decay intensity data on K X-rays from ^{242}Pu

Name of library	DDEP[3]*	ENSDF[4]**
$K_{\alpha 2}$ 99.525 keV	3.55 (17) %	3.6 (3) %
$K_{\alpha 1}$ 103.374 keV	5.6 (3) %	5.7 (4) %

Sources: *Helmer et al., 2002; **Bhat et al., 1992.

Although the intensities of relevant decay X-rays themselves agree well within 2%, the difference of uncertainty is significant. If the uncertainty in ENSDF is adopted for the 99.525 keV transition, about 8% uncertainty needs to be included as one of the systematic uncertainties to deduce the cross-section. If the DDEP evaluation is adopted, the uncertainty is about 5%. Nevertheless, the uncertainty of the decay data is too large to be used to deduce the cross-section with the accuracy of about 2-3% as requested by WPEC Subgroup 26.

The needs of standard intensity data were also identified for detector efficiency calibration. For detector calibration, the use of the IAEA X-ray and gamma-ray standards library, STI/PUB/1298, was recommended. It should be noted that there is no suitable standard in the energy range between 99 keV and 103 keV.

References

[1] Firestone, R.B. and V.S. Shirley (Eds) (1996), *Table of Isotopes*, 8th Edition, John Wiley & Sons, Inc.

[2] Akovali, Y.A. (2002), "Nuclear Data Sheets for A = 242", *Nuclear Data Sheets* 96, 177-240.

[3] Helmer, R.G., E. Browne, M.-M. Be (2002) "International Decay Data Evaluation Project", *Journal of Nuclear Science and Technology* 39, 455, DOI:10.1080/00223131.2002.10875138.

[4] Bhat, M.R., *Evaluated Nuclear Structure Data File* (ENSDF) (1992)," conference proceedings, 1991 International Conference on Nuclear Data for Science and Technology (S. M. Qaim, ed.), p. 817, Springer-Verlag, Berlin, Germany, DOI:10.1007/978-3-642-58113-7_227.

5. Re-evaluated thermal capture cross-section of ^{241}Am

Following the careful review of experimental data in the previous chapters, the Nuclear Energy Agency (NEA) Working Party on International Nuclear Data Evaluation Co-operation (WPEC) Subgroup 41 has recommended thermal capture cross-section values for neutron capture in ^{241}Am. The weighted mean value of energy-dependent data is 720 (21) b. The weighted mean value of spectrum average data is 716 (16) b. The weighted mean value of energy-dependent mean and spectrum average mean is 717 (13) b for the thermal capture cross-section of ^{241}Am. The definitions of the weighted mean value and the uncertainty are given in Appendix A. These values are summarised in Table 5.1.

Table 5.1. Re-evaluated thermal capture cross-section of ^{241}Am

Methods	σ_0 (δ) [b]	Comments
1) Energy-dependent data	720 (21)	δ1 = 21 b, δ2 = 17 b
2) Spectrum average data	716 (16)	δ1 = 16 b, δ2 = 3 b
Weighted mean of 1) and 2)	717 (13)	δ1 = 13 b, δ2 = 2 b

Source: OECD/NEA, 2019.

6. Conclusions and recommendations for future actions

To meet the needs of improving accuracy of nuclear data quantified by the Nuclear Energy Agency (NEA) Working Party on International Nuclear Data Evaluation Co-operation (WPEC) Subgroup 26 for the development of innovative nuclear reactor systems in the world, including ADS projects, measurements on capture cross-sections of minor actinides have been actively performed in the past decade at various facilities. There is an apparent advancement on the quality of energy-dependent data by these efforts, but there were still serious discrepancies on the absolute values − especially for ^{241}Am. The NEA WPEC Subgroup 41 intensively discussed the issues related to the absolute value of the capture cross-section by concentrating on thermal neutrons, which is a key data point for normalisation of the energy-dependent data. In the discussions of Subgroup 41, participants reviewed the status of recent various activities including energy-dependent and integral measurements, and cross-section and decay data evaluations. Participants attempted to identify and quantify the possible bias effects as much as possible. By integrating knowledge obtained in different specialised areas, the discrepancies identified were much improved. In particular, the activation data had essential value in correcting the overlooked bias effects, where the correction factors were quantitatively derived based on recent energy-dependent data. As a result, the uncertainty of the capture cross-section at thermal point was significantly reduced from about 6% in JENDL-4.0 to 2% in the Subgroup 41 recommendations.

Based on the experience and discussions from Subgroup 41, recommendations for future actions are summarised below:

- Regarding experimental samples, it is essential to not only quantify the total sample amount and impurity content, but also rigorously determine the dimensions and any inhomogeneity that may exist. However, it is difficult to characterise all of these quantities in one institute or laboratory. To overcome the sample characterisation issue, an inter-organisational and/or international framework for characterising and exchanging samples is recommended. A close working network would be also valuable to improve the versatility of new samples, which can be utilised for both integral and energy-dependent measurements, at the early stage of their design.
- For energy-dependent data, the currently limited energy region by time-of-flight (TOF) measurements should be extended to cover the regions in question. As discussed in Subgroup 41, the region should cover the main energy domain of Maxwellian neutron spectra in order to use the large number of activation data using reactor neutrons for normalisation. If the energy region is extended to cold neutron energy, the cold neutron data will become very valuable for normalisation of the cross-section at the thermal point.
- The other approach for improving the accuracy is to increase the number of TOF measurements, where the absolute value is determined independently. New data has been published after the closure of Subgroup 41. Evaluation including these data is recommended to be performed in the near future. Ongoing advanced

analyses of integral measurements are also expected to be useful for validating the evaluated data.

- The method of cross-section re-evaluation by integrating energy-dependent and integral data has proven effective and it is recommended to be utilised for other important isotopes such as ^{237}Np and ^{243}Am in the future framework of WPEC.
- The method of cross-section re-evaluation is expected to be utilised for the fast neutron region for important isotopes such as ^{237}Np and ^{241,243}Am. It is recommended that the energy region of TOF measurements be extended to cover the fast neutron region. Integral measurements for validation should also be performed. Some of the activities related to fast neutron region are ongoing and some of these were discussed in Subgroup 41. Evaluation including these data is recommended as soon as these data get an open status.
- The results of this exercise demonstrate the importance of documenting the experimental conditions including sample characteristics. It is necessary to provide detailed information about the uncertainty of all components. Recommendations for the reporting of time-of-flight cross-section data, including full covariance information, are described in [1]. The reporting of the experimental data is based on the Analysis of Geel Spectra (AGS) concept described in [2,3].

References

[1] Gunsing, F., P. Schillebeeeckx, V. Semkova (2013), *Summary Report of the Consultants' Meeting on EXFOR Data in Resonance Region and Spectrometer Response Function*, IAEA Headquarters, Vienna, Austria, 8-10 October 2013, INDC(NDS)-0647.

[2] Becker, B. et al. (2012), "Data reduction and uncertainty propagation of time-of-flight spectra with AGS", *Journal of Instrumentation*, Vol. 7 (2012) P11002, DOI: 10.1088/1748-0221/7/11/P11002.

[3] Becker, B., C. Bastian, J. Heyse, S. Kopecky, P. Schillebeeckx (2014), *AGS – Analysis of Geel Spectra User's Manual,* NEA/DB/DOC(2014)4.

Appendix A. Weighted mean value, internal and external uncertainties

The weighted average $\bar{A}$ of n independent measurements A_i is given by:

$$\bar{A} = \frac{\sum_{i=1}^{n} \omega_i A_i}{\sum_{i=1}^{n} \omega_i}$$

where A_i is each measurement value and ω_i is a square of inverse of each measurement uncertainty ($1/\delta A_i^2$). The internal ($\delta 1$) and external ($\delta 2$) uncertainties are defined as:

$$\delta 1 = \frac{1}{\sqrt{\sum_{i=1}^{n} \omega_i}}$$

$$\delta 2 = \frac{\sqrt{\sum_{i=1}^{n} \omega_i (A_i - \bar{A})^2}}{\sqrt{(n-1)\sum_{i=1}^{n} \omega_i}}$$

The uncertainty $\delta\bar{A}$ associated to $\bar{A}$ is the larger of the internal ($\delta 1$) and external ($\delta 2$) uncertainties.