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P. Schillebeeckx
B. Becker
S. Kopecky

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Institute for Reference Materials and Measurements

Contact information
Peter Schillebeeckx
Address: Joint Research Centre, Retieseweg 111, B-2440 Geel, Belgium
E-mail: peter.schillebeeckx@ec.europa.eu
Tel.: +32 14 57 1475
Fax: +32 14 57 1862

http://irmm.jrc.ec.europa.eu/
http://www.jrc.ec.europa.eu/

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P. Schillebeeckx, B. Becker and S. Kopecky

European Commission, Joint Research Centre - IRMM,
Retieseweg 111, B - 2440 Geel, Belgium

Abstract

This report is the result of the EC-JRC-IRMM support to the OECD-NEA WPEC Subgroup 31, " Meeting Nuclear Data Needs for Advanced Reactor Systems *". It summarizes the contribution that is related to capture cross section data. The status of capture cross section measurements based on the detection of prompt γ-rays is discussed and experimental data that are available to evaluate the capture cross section for ^{28}Si, ^{206}Pb, ^{238}U and ^{241}Am for advanced reactor systems are reviewed.*

1. Introduction

Nuclear data needs for advanced reactor systems have been identified by the Subgroup 26 “Uncertainty and Target Accuracy Assessment for Innovative Systems Using Recent Covariance Data Evaluations” of WPEC (Working Party on International Nuclear Data Evaluation Co-operation). Within this subgroup a target accuracy assessment was performed and a list of nuclear data requiring improvements was defined. It was shown that there are significant gaps between the current uncertainties and the target accuracies. To reduce these gaps the WPEC Subgroup 31 “Meeting Nuclear Data Needs for Advanced Reactor Systems” was organized. The subgroup consisted of measurement experts from each of the international data projects.

2. Status of capture cross section measurements

Neutron induced capture cross section measurements rely either on post-irradiation activation analysis or on the detection of prompt γ-rays emitted in the (n,γ) reaction. The choice of the principle and related detection system depends on the reaction to be studied, the energy region of interest, the amount of available sample material and the required accuracy and resolution. Capture cross sections in the resonance region are best derived from results of experiments with a prompt γ-ray detection system that is optimized for time-of-flight (TOF) measurements [1]. Post-irradiation activation analysis is suited to determine capture cross sections at thermal energies and in the continuum region, and to derive experimental resonance integrals. Recently, the activation method has also been applied to determine cross sections using neutrons from a fast reactor.

A prompt γ-ray detection system optimized for TOF-measurements fullfills the following requirements [1]:

- the detection efficiency for a capture event is independent of the γ-ray cascade, i.e. independent of the multiplicity of the γ-ray spectrum and the γ-ray energy distribution;
- the sensitivity to neutrons scattered by the sample is low compared to the sensitivity to γ-rays produced by the capture reaction in the sample;
- the detector has a good time resolution;
- for the study of a fissioning nucleus, the γ-rays from neutron capture can be separated from those resulting from fission; and
- in case of a radioactive sample, the prompt γ-rays can be separated from the γ-rays emitted due to the radioactive decay.

Three different principles based on the direct detection of prompt γ-rays can be distinguished [1]: (1) γ-ray spectroscopic (GS), (2) total γ-ray absorption (TA) and (3) total energy detection principle (TE). The main uncertainty for the three principles is related to the normalization of the data and the determination of the background [1].

2.1 Spectroscopic measurements

Capture cross sections based on γ-ray spectroscopic measurements with high resolution γ-ray detectors can be derived from [1-3]:

- the sum of all the partial cross sections of primary transitions depopulating the capture state (GS1);
- the sum of the partial capture cross sections of the transitions feeding the ground state (GS2); or
- the sum of all the observed partial cross sections weighted with the energy of the transition divided by the total γ-ray energy liberated in the capture event (GS3).

The accuracy strongly depends on the complexity of the level scheme of the compound nucleus. Cross sections can be determined accurately when the γ-ray transitions of the cascade are well known. Therefore, γ-ray spectroscopic methods are very powerful to determine capture cross section data for light nuclei or for nuclei with a proton and neutron number close to a magic shell [2, 3]. When not all γ-ray transitions can be determined the results are biased and only lower limits can be derived [4]. To verify the impact of missing transitions, the principle of γ-ray intensity balance [5] or crossing intensity sum [6] can be applied. The missing contributions can also be based on statistical models to simulate the full γ-ray cascade. Codes that can be used are e.g. DICEBOX [7], DECAYGEN [8] and γDEX [9]. The γ-ray cascade simulations rely on nuclear level statistical models and nuclear data input (low-lying level scheme, average radiation widths, level densities). Using spectroscopic measurements the accuracy of the cross section depends on the statistical nature of the γ-ray cascade.

2.2 Total absorption principle

The total γ-ray absorption principle relies on the detection of the energy sum of the γ-rays emitted in a capture event. An ideal detector has a 4π geometry and a 100 % absolute detection efficiency allowing for the detection of the entire electromagnetic cascade. Thus, the energy deposited in the detector is directly proportional to the total energy available in the capture event and independent of the γ-ray cascade.

The first total absorption detectors were large liquid organic scintillation tanks [10, 11]. The uncertainty of these systems is limited to 5 % - 10 % and depends on the reaction under study. The limitation is primarily due to corrections that are required to estimate the efficiency to detect a capture event [1, 11]. Organic liquid scintillators (OLS) have extensively been used to determine capture cross sections of fissile material. To separate capture events from fission events different methods have been applied. Some of them use an additional fission chamber in parallel or as an additional measurement to determine correction factors. An extensive list of capture-to-fission ratio measurements for $^{233, 235}$U and ^{239}Pu is given in Ref. [1].

Nowadays in-organic detectors are used. An overview of systems that are in use is given in Ref. [1]. In-organic scintillators are smaller in size and have a better detection efficiency compared to OLS. Therefore their sensitivity to the ambient background is reduced. However, they still suffer from neutron sensitivity due to (n,γ) reactions in the detection material. Therefore, they are limited to measurements in the resolved resonance region and for nuclei with small scattering to capture ratios. The final accuracy of such systems depends strongly on the reaction under study. Since an ideal detector with a 100 % γ-ray detection efficiency does not exist, a correction is needed when the normalization is performed using a capture reaction which has a different γ-ray cascade from the reaction under study. Such a correction becomes even more important when a constraint is imposed on the multiplicity and energy deposition to reduce the background and when the γ-ray cascade changes from resonance to resonance. Due to an improved understanding of the measurement equipment and techniques through Monte Carlo simulations the detection efficiencies can be determined with better accuracies. However, the final accuracy depends strongly on the statistical nature of the γ-ray cascade, as in the case of organic scintillators. These detectors can also be used to derive capture-to-fission ratio for fissile materials [12,13].

2.3 Total energy detection principle

When the contribution of the fission channel can be neglected, the most accurate capture cross section data can be measured by applying the total energy detection principle (TE) using C_6D_6 detectors combined with the pulse height weighting technique (PHWT). The application of the total energy detection principle requires a γ-ray detector with a relatively low γ-ray detection efficiency which is proportional to the γ-ray energy. Under these conditions the efficiency to detect a capture event is directly proportional to the sum of the energies of the γ-rays emitted in the cascade. This makes the efficiency in first approximation independent of the γ-ray cascade.

The Moxon-Rae detector achieves approximately the proportionality between the γ-ray energy and detection efficiency by a special design of the detector [14]. However, uncertainties due to imperfect linearity between the detection efficiency and the γ-ray energy are at least 5 % [15,16].

Correction factors in case of the total energy detection principle combined with PHWT are limited compared with all the other principles (GS or TA). This has a strong impact on the accuracy that can be reached. An experimental validation of the total energy detection principle combined with the PHWT for C_6F_6 detectors was performed by Yamamuro et al [17]. Normalization factors derived from the saturated resonances 4.3 eV in ^{181}Ta, 4.9 eV in ^{197}Au and 5.2 eV in ^{109}Ag, were consistent within 2 % [17]. A more extensive performance assessment for a C_6D_6 based system has been carried at the GELINA facility of the EC-JRC-IRMM [1,18,19]. The results in [1,18] demonstrate that capture yields with uncertainties better than 2 % can be deduced from thermal energy up the URR when the total energy detection principle in combination with the PHWT is applied. However, such a low uncertainty can only be reached under specific constraints, as discussed in Ref. [1].

3. Review of cross section data for the capture reaction of ^{28}Si, ^{206}Pb, ^{238}U and ^{241}Am

In this section documented cross section data that can be used to evaluate the capture cross sections for ^{28}Si, ^{206}Pb, ^{238}U and ^{241}Am from thermal up to the URR are discussed. In the RRR reliable resonance parameters can only be derived when transmission data are available [1,20]. Total cross section data provide also important prior information to improve the accuracy of capture cross sections in the URR, as shown by Sirakov et al. [21]. Hence, transmission data are needed to perform a consistent evaluation of the capture cross section in the resonance region with uncertainties that are requested in the conclusions of SG-26. Therefore, in the discussion of available experimental data also results of transmission measurements have been considered.

3.1 Cross section data for ^{28}Si(n,γ)

Experimental data that can be used for an evaluation of the thermal capture cross section of ^{28}Si are summarized in Table 1. The reference cross section that was used is also given. The value recommended by Raman et al. [3] is fully consistent with the cross section derived from the three γ-spectroscopic methods mentioned in the introduction of section 2.1. This cross section is relative to the (332.6 ± 0.6) mb capture cross section for ^{1}H determined by Cokinos and Melkonian [25]. The capture cross section reported by Islam et al. [24] deviates by more than 20 %. This is partly due to the reference value for ^{14}N(n,γ) used in Ref. [24], which is 15% higher compared to the value 68.77 (0.56) mb reported by Belgya [6]. For a full consistent evaluation the coherent scattering length b_c = 4.106 (0.006) fm recommended by Koester et al . [26] can also be used.

		^{28}Si(n,γ)	Reference cross section	
Pomerance	[22]	81 (24) mb	^{197}Au(n, γ)	95 b
Spits and De Boer	[23]	156 (23) mb	Al(n, γ)	239 (3.0) mb
Spits and De Boer	[23]	163 (57) mb	Na(n, γ)	534 (5.0) mb
Spits and De Boer	[23]	166 (33) mb	Mn(n, γ)	13.3 (0.2) b
Islam et al.	[24]	207 (4) mb	^{14}N(n,γ)	79.8 (1.4) mb
Raman et al.	[3]	169 (4) mb	^{1}H(n,γ)	332.6 (0.6) mb
GS1		169.4 (3.9) mb		
GS2		168.0 (3.7) mb		
GS3		168.1 (3.9) mb		

Table 1 Results of capture cross section measurements for ^{28}Si at 0.0253 eV.

Capture and transmission measurements for ^{28}Si+n have been carried out at GELINA at a 130-m and 400-m flight path, respectively [27]. For the capture measurements a γ-spectroscopic detection system based on BGO-detectors was used. The capture data were normalized to the 1.15 keV resonance of ^{56}Fe. However, no further details about the normalization procedure, e.g. the partial radiation width used for the normalization, were specified. Partial capture cross sections for transitions to the ground state and the first and third excited state have been determined. Resonance areas for ^{28}Si are given for resonance energies up to 4638 keV. Unfortunately the experimental transmission and yields are not available in numerical data. Documented TOF-data resulting from total and capture measurements at ORELA are available in numerical form [28-32]. Transmission measurements on natural samples with different thickness have been carried out at a 47-m, 80-m and 200-m flight path [28-31]. Results of capture measurements at a 40-m station have been reported by Guber et al [32]. The total energy detection principle in combination with the PHWT using C_6D_6 detectors was applied. From these data capture areas and peak cross sections for neutron energies < 700 keV can be deduced with an accuracy < 5 %. However, this level of accuracy can not be reached for the capture cross section between resonances. This is due to the contribution of direct capture and/or possible interference effects.

The ORELA data together with the transmission data of Adib et al. [33] have been used in a resonance shape analysis by Derrien et al. [34]. In the analysis the impact of a direct capture contribution was also considered. The analysis was based on a Reich-Moore approximation of the R-matrix theory. Hence, interference effects for the capture channel have been neglected. The resulting thermal capture cross section is consistent with the one recommended by Raman et al. [3]. However, the coherent scattering length is about 5 % smaller compared to the one recommended by Koester et al. [26]. Although the above mentioned data on ^{28}Si and the evaluation of Derrien et al. [34] do not cover the energy region of interest in Table 32 of SG-26 (i.e. 6 MeV – 20 MeV), they provide essential data to produce consistent capture cross sections in the high energy region.

3.2 Cross section data for ^{206}Pb(n,γ)

Experimental capture cross sections at thermal energy that have been reported in the literature are: $\sigma(n_{th},\gamma)$ = 25.5 (5.0) mb [35], 30.5 (0.7) mb [36], 26.6 (1.2) mb [37] and 29^{+2}_{-1} mb [2]. These values together with the coherent scattering lengths b_c = 9.22 (0.07) fm determined by Ioffe et al. [38] and b_c = 9.23 (0.05) by Koester and Knopf [39] can be used to derive consistent capture and scattering cross sections at thermal energy.

Transmission measurements for neutron energies between 1 keV and 900 keV have been carried out at a 78-m and 200-m station of ORELA using radiogenic lead samples enriched to 88.4 % in ^{206}Pb [40-42]. In Ref. [40] capture cross section measurements using Ge-detectors at a 40-m station of ORELA are reported. Capture and transmission measurements on a pure ^{206}Pb sample at a 60-m and 25-m station, respectively, have been performed at GELINA by Borella et al. [43]. Capture yields were obtained by applying the total energy detection principle combined with the PHWT using C_6D_6 detectors. The neutron sensitivity of the set-up was determined by Monte Carlo simulations and verified by experiment. The experiments, data reduction and analysis procedures were carried out following the recommendations in Ref. [1]. In Ref. [43] results of a simultaneous analysis of capture and transmission data for neutron energies < 80 keV are given. For neutron energies between 80 keV and 625 keV the experimental yield was analyzed by fixing the neutron widths reported by Horen et al. [41,42]. From these data capture areas with an accuracy < 5% have been deduced. The thermal data of Refs. [2,35 – 39] and the TOF-data of Refs [40 - 43], which are available in numerical form in the EXFOR library, can be used to improve the total and capture cross section for neutron induced reactions in ^{206}Pb below 650 keV. From such an evaluation capture areas and peak cross sections for neutron energies < 200 keV can be deduced with an accuracy < 5 %. However, this level of accuracy cannot be reached for the cross section between resonances. Since Borella et al. [43] and Mizumoto et al. [40] have demonstrated that the γ-ray emission spectra are limited to a few cascades, the contribution of direct capture and/or interference effects cannot be excluded.

Capture measurements using C_6D_6 detectors on a pure ^{206}Pb sample have been reported in Ref. [44]. However, the experimental yield is not available in numerical form. In addition, a weighting for an ideal detection system was applied, i.e. for a 0 MeV discrimination level on the observed energy deposited in the C_6D_6 detector. Therefore, correction factors to account for the finite discrimination level are required, which are very difficult to be determined accurately for neutron capture on ^{206}Pb as discussed in Ref. [1].

3.3 Cross section data for ^{238}U(n,γ)

The status of the thermal capture cross section has been reviewed by Trkov et al. [45]. A list of measured coherent scattering lengths is given in Ref. [46]. These scattering lengths together with results of transmission and capture measurements at ORELA have been used by Derrien et al. [47] to determine parameters of individual resonances for ^{238}U below 20 keV. The transmission experiments were carried out at a 40-m, 150-m and 200-m station and the capture measurements at a 40-m and 150-m station using an OLS. The capture data of Corvi et al. [48] obtained at GELINA together with the thermal capture cross section of Poenitz et al. [49] have been used to adjust the parameters of the bound state(s). The resulting capture cross section at thermal $\sigma(n_{th},\gamma)$ = 2.7 b is close to the value $\sigma(n_{th},\gamma)$ = 2.683 (0.012) b recommended by Trkov et al. [45].

A list of capture cross section data (including absolute and shape data) for ^{238}U that can be used to determine the average capture cross section in the URR and higher is given in Ref. [50]. These data have been used to evaluated the average capture cross section up to 2.2 MeV based on a least squares adjustment using the GMA code developed by Poenitz [51]. In Ref. [50] average capture cross sections below 200 keV are recommended with uncertainties between 0.5% and 3.3%. Other evaluations of the capture cross section for ^{238}U below 200 keV are reported by Fröhner [52], Maslov et al. [53] and Courcelle et al. [54]. These evaluations result from a parameterization of the cross section data by the Hauser-Feshbach formalism including width fluctuation corrections. The evaluation process includes results of total cross section and in-elastic cross section data.

Unfortunately, only one set of capture cross section data in the resonance region, the one reported by Yamamuro et al. [55], was based on the total energy detection principle using C_6D_6 detectors. In addition, the data suffer from a rather large 7.7% normalization uncertainty. Recently, capture cross section experiments in the resonance region have been carried out at GELINA and n_TOF as part of the ANDES project [56]. Measurements using C_6D_6 detectors combined with the PHWT have been carried out at GELINA [57] and n_TOF [58]. Measurements with a total absorption detector were also carried out at the n_TOF facility [59]. A re-evaluation of the cross section data, including the results of these measurements, should result in a capture cross section for ^{238}U with uncertainties around 2% for neutron energies < 200 keV.

3.4 Cross section data for ^{241}Am(n,γ)

A list of experimental data that can be used to re-evaluate the capture cross section of ^{241}Am is given in Refs. [60,61]. The data include thermal capture cross sections, integral measurements and transmission and capture TOF-data. Transmission measurements in the resonance region have been performed by Adamchuk et al. [62], Slaughter et al. [63], Derrien and Lucas [64], Belanova et al. [65], Kalebin et al. [66] and Lampoudis et al. [61]. Results of capture experiments have been reported by Weston and Todd [67], Gayther and Thomas [68], Wisshak and Käppeler [69], Vanpraet and Cornelis [70], Jandel et al. [71] and Lampoudis et al. [61].

Reference			σ_{m+g}	σ_g	σ_m
Pomerance	[72]	P	628. 5(35)		
Bak et al.	[73]	A	740 (60)	670 (6)	70 (5)
Dovbenko et al.	[74]	A		573 (103)	74 (15)
Harbour et al.	[75]	T	612 (25)		
Kalebin et al.	[66]	A	624 (20)		
Gavrilov et al.	[76]	A	853 (52)	780 (50)	73 (14)
Shinohara et al.	[77]	A	854 (58)	768 (58)	85.7 (6.3)
Maidana et al.	[78]	A	673 (10)	602 (9)	
Fioni et al.	[79]	A	696 (46)	636 (46)	60 (4)
Bringer et al.	[80]	A	705 (23)		
Nakamura et a.	[81]	A	702 (25)	628 (22)	
Jandel et al.	[71]	T	665 (33)		
Lampoudis et al.	[61]	T	603 (36)	540 (32)	
ENDF/B-VI.8			620 (13)		
JEFF-3.1.2			647 (34)		
JENDL-3.3			639.5		
JENDL-4.0			684 (15)	620.1 (7.8)	64.8 (7.8)

Table 2 Experimental data for the thermal capture cross section of ^{241}Am. The symbols (σ_{m+g}, σ_g, σ_m) are explained in the text. The experimental method that was used is indicated by P (pile oscillation), A (activation) and T (time-of-flight).

In Table 2 experimental cross section data for the capture reaction at 0.0253 eV are listed. Data reported in the literature for the total capture cross section (σ_{m+g}), for the capture cross section to the 1^- ground state in ^{242}Am (σ_g) and for the reaction to the 5^- isomeric state at 49 keV of ^{242}Am (σ_m) are given. The second column specifies the measurement technique that was applied, i.e. neutron activation analysis (A), pile oscillator measurements (P) or time-of-flight experiments (T). The most recent thermal capture cross section derived by Lampoudis et al. [61] (σ (n_{th}, γ) = 749 (35) b) is 19% and 12% larger than the one derived from the TOF-measurements of Kalebin et al. [66] and Jandel et al. [71], respectively. This systematic difference is reflected in differences with recommended values in the evaluated data libraries, which are also listed in Table 2. The value of Lampoudis et al. [61] is 10% larger than the latest evaluated value (JENDL-4.0), but is within the quoted uncertainties in agreement with results of measurements at ILL [79,80] and at KURRI [81]. As noted in Ref. [82], the cross section derived from the data in Ref. [81] is even underestimated due to the Westcott factor g = 1.05 that was used. The overestimation of the Westcott factor g = 1.05 recommended by Mughabghab [83] is also confirmed by results from measurements at J-PARC [84].

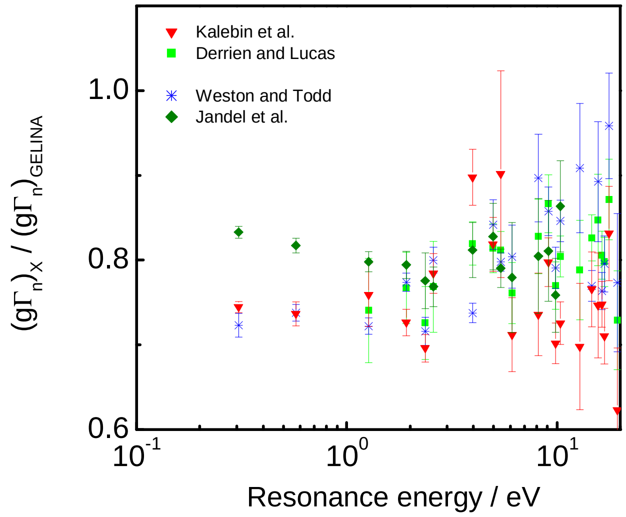

Fig. 1 Ratio of resonance strengths ($g\Gamma_n$) derived by Derrien and Lucas [64], Kalebin et al. [66], Weston and Todd [67] and Jandel et al. [71] and the one of Lampoudis et al. [61]. The latter was obtained from measurements at GELINA.

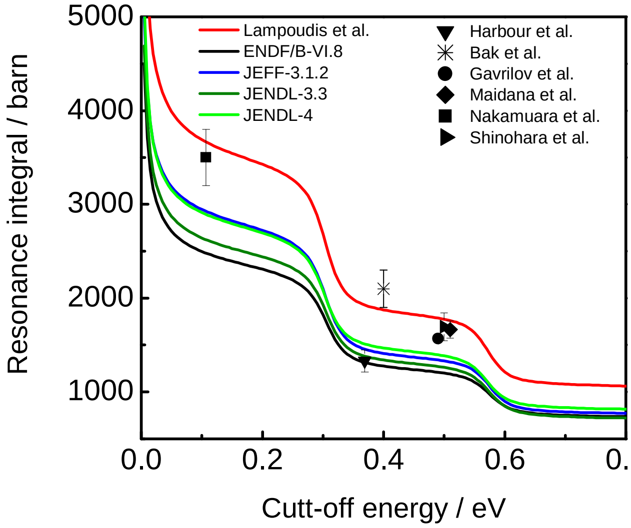

Fig. 2 The resonance integral for the ^{241}Am(n,γ) as a function of the cut-off energy. The values resulting from direct measurements are compared with the integral calculated from resonance parameters.

Fig. 1 reveals that the resonance strength of the resonances at 0.306 eV, 0.574 eV and 1.272 eV derived by Derrien and Lucas [64], Kalebin et al. [66], Weston and Todd [67] and Jandel et al. [71] are systematically lower compared to those of Lampoudis et al. [61]. On the other hand the resonance integrals calculated with the parameters of Lampoudis et al. [61] are fully consistent with the results of direct measurements [73,76-78,81] as illustrated in Fig. 2. This suggests that the resonance strengths reported by Derrien and Lucas [64], Kalebin et al. [66], Weston and Todd [67] and Jandel et al. [72] are underestimated. Such an underestimation might occur when powder samples are used and no correction for the particle size is applied. As discussed in Ref. [1], an underestimation of the neutron width when using powder samples will coincide with an overestimation of the radiation width. A comparison of the data in Table 2 confirms that Derrien and Lucas [64] and Weston and Todd [67] deduced larger radiation widths from their data. However, this does not explain the smaller resonance strengths from Kalebin et al. [66] and Jandel et al. [71]. Lampoudis et al. [61] suggest that the resonance strengths and thermal value of Jandel et al. [71] are underestimated due the normalization procedure that is applied.

		Average radiation width
Derrien and Lucas	[64]	44.2 (0.1) meV
Kalebin et al.	[66]	42.9 (0.3) meV
Weston and Todd	[67]	47.6 (0.2) meV
Jandel et al.	[71]	43.8 (1.3) meV
Lampoudis et al.	[61]	42.1 (0.3) meV

Table 2 Comparison of the average radiation width for ^{241}*Am + n reported by Derrien and Lucas [64], Kalebin et al. [66], Weston and Todd [67], Jandel et al. [71] and Lampoudis et al. [61]*

The accuracy of both the resonance parameters and thermal capture cross section can be improved by re-analyzing the above mentioned TOF-data together with results of capture measurements at J-PARC [84] and n_TOF [85] and a neutron activation experiment at FRM-II in Garching [86]. Although such a re-evaluation will not directly contribute to an improved capture cross section in the keV region these data, in particular the transmission data of GELINA, can be used to correct for systematic effects due to sample characteristics and improve the normalization of the capture data of Weston and Todd [67], Gayther and Thomas [68], Wisshak and Käppeler [69], Vanpraet and Cornelis [70] and Jandel et al. [71].

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Abstract

This report is the result of the EC-JRC-IRMM support to the OECD-NEA WPEC Subgroup 31, “Meeting Nuclear Data Needs for Advanced Reactor Systems”. It summarizes the contribution that is related to capture cross section data. The status of capture cross section measurements based on the detection of prompt γ-rays is discussed and experimental data that are available to evaluate the capture cross section for ^{28}Si, ^{206}Pb, ^{238}U and ^{241}Am for advanced reactor systems are reviewed.

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