Improving nuclear data accuracy of ^{241}Am and ^{237}Np capture cross sections

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Abstract. In the framework of the OECD/NEA WPEC subgroup 41, ways to improve neutron induced capture cross sections for ^{241}Am and ^{237}Np are being sought. Decay data, energy dependent cross section data and neutron spectrum averaged data are important for that purpose and were investigated. New time-of-flight measurements were performed and analyzed, and considerable effort was put into development of methods for analysis of spectrum averaged data and re-analysis of existing experimental data.

1. Introduction

There is a serious gap between required accuracy and current accuracy on nuclear data for developing innovative nuclear reactor systems. To bridge this gap, an international joint activity entitled "Improving Nuclear Data Accuracy of ^{241}Am and ^{237}Np capture cross sections (INDA)" is performed under WPEC. In this joint study, the forefront knowledge of energy dependent data, spectrum averaged data, relevant decay data, and evaluations are intended to be integrated on the capture cross sections of ^{237}Np and ^{241}Am. Each kind of data by state-of-art technique has been reviewed at first, and key issues on systematic errors have been identified for each kind of data to be solved to bridge the gap.

It was recognized that the forefront knowledge of each kind of data are valuable each other for improving data accuracy. Furthermore, key issues have been identified for each kind of data toward further improvement of nuclear data. In chapter 2-4, the forefront knowledge of each kind of data and key issues on systematic errors are summarized. In chapter 5, an example of successful achievement is described obtained by integrating the forefront knowledge. In chapter 6, the benefit of these collaborative works is discussed from the view point of evaluating nuclear data.

2. Current status of decay data

The relevant nuclear structure data for ^{237}Np and ^{241}Am measurements, are decay data for ^{233}Pa, ^{237,238}Np, ^{238}Pu, and ^{241,242g,242m}Am, ^{242}Cm, respectively. The main databases for radioactive decay data are ENSDF [1] and DDEP [2]. In Tables 1 and 2, most prominent X-, γ-ray, and α lines are summarized. Candidates for the emissions for activation experiments were summarized, and uncertainties of the current emission probabilities were explained in the framework of the WPEC subgroup 41. The difficulty due to the contaminated emissions will be a candidate for unrecognized systematic error.

3. Current status of energy dependent cross section measurements

Recent energy dependent cross section measurements were reviewed from GELINA, DANCE, n_TOF, and ANNRI (Fig. 1 for ^{241}Am(n,γ)). Origins of systematic errors and important correction factors have been systematically identified, for example, sample impurity, sample amount, flux, detection efficiency, neutron self-shielding & multiple-scattering factors, and normalization. The importance of deducing the Westcott g-factor from energy dependent data was also pointed out, which is reflected in the spectrum averaged data.

The derived values for thermal (0.0253 eV) capture cross section for ^{241}Am are 749 b ± 34 b, 710 b ± 35 b

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Table 1. Main decay data, important for ^{237}Np measurements.

Nuclide	Energy [keV]	Intensity [%]		Type
		DDEP	ENSDF	
^{233}Pa	311.904(5)	38.3(5)	38.5(4)	γ
^{237}Np	29.374(20)	14.3(6)	14.12(15)	γ
	86.477(10)	12.26(12)	12.4(3)	γ
	95.869(-)	2.906(20)	2.70(14)	XKα1
	4771.4(8)	23.0(3)	23.2(3)	α
	4788.0(9)	47.64(6)	47.64(6)	α
^{238}Np	984.45(2)	25.18(13)	25.2(3)	γ
	1028.54(2)	18.25(13)	18.23(12)	γ
^{238}Pu	5456.3(2)	28.85(6)	28.98(10)	α
	5499.03(20)	71.04(6)	70.91(10)	α

Table 2. Main decay data, important for ^{241}Am measurements.

Nuclide	Energy [keV]	Intensity [%]		Type
		DDEP	ENSDF	
^{241}Am	16.96(-)	18.58(13)	–	XLβ
	21.16(-)	37.66(17)	36(4)	XL
	59.5409(1)	35.92(17)	35.9(4)	γ
	5442.86(12)	13.23(10)	13.1(3)	α
	5485.56(12)	84.45(10)	84.8(5)	α
^{242g}Am	17.1385(-)	10.8(5)	11.0(7)	XL
	18.08(-)	18.0(11)	17.9(25)	XL
	42.13(5)	0.040(2)	0.039(4)	γ
	103.734(-)	5.6(3)	4.7(4)	XKα1
^{242m}Am	16.681(-)	0.37(4)	–	XL
	17.6065(-)	25.0(11)	–	XL
	49.371(3)	0.134(4)	0.134(5)	γ
	5207.15(25)	0.409(9)	0.409(11)	α
^{242}Cm	17.595(-)	9.92(23)	–	XL
	6069.37(9)	25.94(7)	25.94(6)	α
	6112.72(8)	74.06(7)	74.08(7)	α

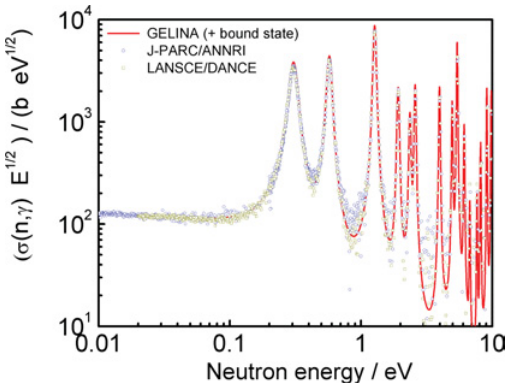

Figure 1. Comparison of recent measurements of neutron capture cross sections for ^{241}Am, normalized to 749 b at 0.0253 eV.

and 678 b ± 68 b for GELINA (Lampoudis et al., Ref. [3]), DANCE (Jandel et al., Ref. [4] + modifications[1]) and n_TOF (Fraval et al., Ref. [5]). The J-PARC/ANNRI measurement by Harada et al. [6] is relative, but covers a wide incident neutron energy region from cold to epithermal.

[1] M. Jandel, private communication, May 2016.

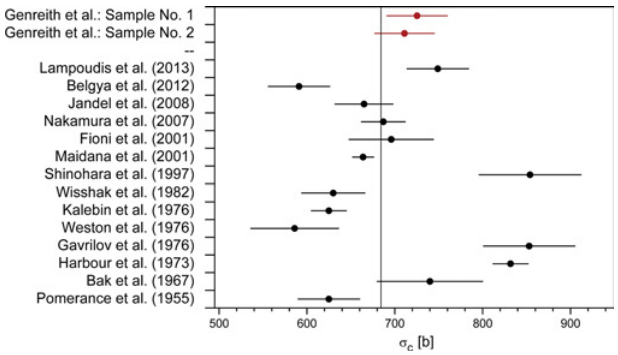

Figure 2. ^{241}Am thermal neutron capture cross sections as calculated from measurements performed at the cold PGAA beam in Garching, FRM II, along with other values from literature. The ENDF/B-VII.1 value is given for comparison (line at 684.3 b).

Table 3. Reaction rates R for the ^{237}Np(n,γ) reaction from normalized MC spectra and assuming JEFF-3.2 cross sections.

Irradiation channel	$R(^{237}$Np(n,γ)) [b]		
	bare	1 mm Cd	35 μm Gd
IC40	5230	632	1180
CC	2590	670	1057
LI2T	3360	661	1120
LI2L	3690	675	1170

4. Current status of spectrum averaged experiments

Reported results from integral measurements of ^{241}Am neutron capture cross sections over the last 60 years scatter by >40% as can be seen from Fig. 2. A source of systematic bias could not be allocated, hence a concerted action for improving of nuclear data has been initiated with the scope to augment accuracy and lower uncertainty substantially. The major underlying reasons for bias in ^{241}Am capture cross sections are low energy resonances and bound levels with variable influence to the measurement technique applied.

5. Feedback to spectrum averaged experiments

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 standardization method [7] and the Westcott convention [8] as implemented e.g., in Refs. [9,10]. Both methods are crude since they approximate the neutron spectrum $\varphi(E)$ with only two free parameters (three in case variations in spectrum temperature are allowed). A general method based on Monte Carlo (MC) calculated reactor spectra (Fig. 3) and JEFF-3.2 cross section library was used to calculate reference reaction rates

$$R = \int \varphi(E)\sigma(E)dE \qquad (1)$$

for the measured reactions and standards (^{197}Au(n,γ), ^{59}Co(n,γ), etc.), irradiated bare and under Cd/Gd filters. Due to limited space, results for ^{237}Np and ^{241}Am only are presented here (Table 3 and Table 4, respectively).

Table 4. Reaction rates R for the ^{241}Am(n,γ) reaction from normalized MC spectra and assuming JEFF-3.2 cross sections.

Irradiation channel	$R(^{241}$Am(n,γ)) [b]		
	bare	1 mm Cd	35 μm Gd
IC40	21900	1510	5000
CC	10700	1560	4210
LI2T	14100	1570	4670
LI2L	15500	1640	4930

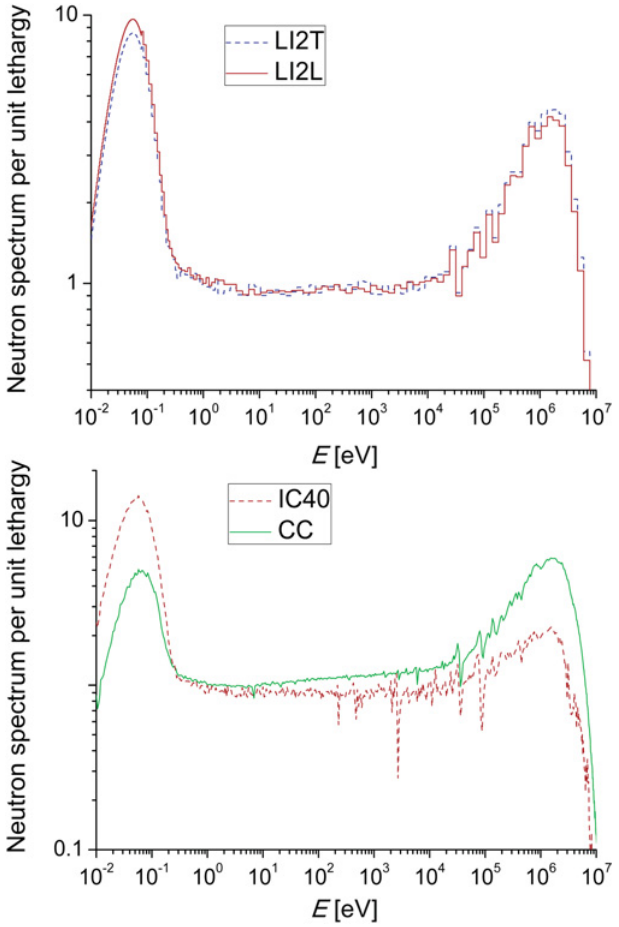

Figure 3. Monte Carlo calculated spectra in irradiation channels of JSI TRIGA (below) and KUR (above) reactors.

Table 5. Biases in the derived σ_0 for ^{237}Np(n,γ), relative to the thermal reference value 181 b due to approximations in methodologies.

Irradiation channel	$\sigma_0(^{237}$Np(n,γ)) relative bias		
	k_0 Cd	Westcott Cd	Westcott Gd
IC40	5.1%	3.8%	0.2%
CC	13.3%	9.4%	0.3%
LI2T	1.7%	6.7%	0.2%
LI2L	8.6%	6.1%	0.2%

Adopting the reaction rates from Table 3 and Table 4 as reference "experimental" data, k_0 and Westcott methods have been used to derive the capture cross section σ_0 at thermal energy (25.3 meV). The biases produced due to the limitations in methodologies are presented in Table 5 and Table 6.

For ^{241}Am, a systematic trend of significant overestimation of σ_0 for activation method using Cd filters with an effective cut-off energy of around 0.55 eV can be

Table 6. Biases in the derived σ_0 for ^{241}Am(n,γ), relative to the thermal reference value 748 b due to approximations in methodologies.

Irradiation channel	$\sigma_0(^{241}$Am(n,γ)) relative bias		
	k_0 Cd	Westcott Cd	Westcott 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%

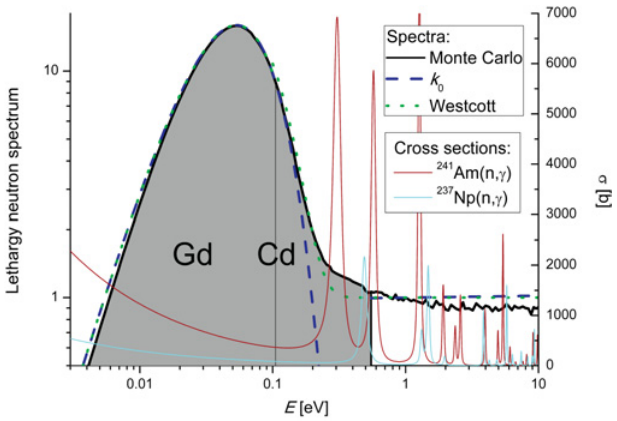

Figure 4. Comparison of Monte Carlo calculated (reference) and approximate (k_0 and Westcott convention) neutron spectra for a typical thermal research reactor irradiation channel (JSI TRIGA IC40). The differences in spectra clearly overlap with the low energy resonances in ^{241}Am and ^{237}Np neutron induced cross sections inducing biases in the derivation of the thermal cross section from the reaction rates.

observed. The magnitude 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 take into account the contribution of the epithermal neutron spectrum component to the reaction rate between about 0.1 eV and 0.55 eV, which is for most nuclides insignificant. However the latter for ^{237}Np and especially ^{241}Am it is very important due to low-energy resonances below and around 0.5 eV (Fig. 4). Expectedly, the bias is larger if the epithermal component is stronger (corresponding to a lower thermal Maxwellian peak). With the use of Gd filter (effective cut-off energy around 0.1 eV), the problem of the bias is practically non-existent. This method was used e.g., by Nakamura [9].

An independent study [11], based on analytical corrections to the Westcott formalism due to deviation of the neutron spectrum from idealized shape, yields 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 summarized in Table 7. The results in the column "MC corrected σ_0" were also corrected for the deviation of the transmission filter from ideal and the generalized Westcott factor. The MC correction relies on JEFF-3.2 library, while the corrections in Ref. [11] rely on JENDL-4.0 library.

6. Feedback to evaluation

From the joint study, it was shown that integration of knowledge from the independent specialities enables us

Table 7. Original and corrected values for ^{241}Am(n,γ) cross section at 25.3 meV from neutron activation measurement.

Author(year)	σ_0(unc.) [b] original	σ_0 [b] corr. MC	σ_0 [b] corr. Ref. [11]	Meth.
Bak (1967)	740(60)	672(64)	691(60)	Cd, α
Harbour (1973)	832(18)	694(73)	/*	Cd, α
Gavrilov (1976)	853(52)	725(48)	725(52)	Cd, α
Shinohara (1997)	854(58)	727(60)	738(52)	Cd, α
Bringer(2007)	705(23)	710(23)	705(23) **	Cd, mass s
Nakamura(2007)	687(25)	718(28)	687(25) **	Gd, α

* Not considered.
** Not corrected.

not only to crosscheck between data obtained by different techniques but also to improve the measurement accuracy of each other.

The main outcome of the study in terms of feedback to evaluations will be to give recommendations to JEFF, JENDL and ENDF projects in order to take into account recent findings of the WPEC SG-41 and implement them into the new releases of the nuclear data libraries.

7. Conclusions

Neutron induced capture cross sections of ^{237}Np and ^{241}Am were studied in order to improve the quality of nuclear data. A detailed look into available experimental data was performed, including time-of-flight, reactor activation data, integral experiments and measurement in cold neutron beams. For a consistent analysis of experimental data, reliable and accurate decay data are also required.

Re-analysis of the existing activation data in reactor spectra is currently under way in order to revise the value of the neutron induced cross section of ^{241}Am at thermal energy. Combined with TOF measurements and measurements with cold neutrons, a new recommended value will be presented as one of the main results of the WPEC SG-41 activities.

This contribution presents the outline on the international joint activity coordinated via the OECD/NEA Working Party on Evaluation Cooperation (WPEC) Subgroup 41.

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