^{232}Th, ^{238}U, ^{237}Np, AND ^{239}Pu (γ, xn), (γ, sn), (γ, n), (γ, 2n), AND (γ, f) REACTIONS CROSS SECTIONS EVALUATION BASED ON DATA OBTAINED USING QUASIMONOENERGETIC ANNIHILATION AND BREMSSTRAHLUNG PHOTONS

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The detailed systematical analysis of the (γ, xn), (γ, sn), (γ, n), (γ, 2n) and (γ, f) reaction cross section data obtained by using quasimonoenergetic annihilation photon beams at Livermore (USA) and Saclay (France) was carried out for 4 actinides nuclei ^{232}Th, ^{238}U, ^{237}Np and ^{239}Pu. For overcoming of significant disagreements between the data and moving them into consistence the special method proposed before for taking into account both laboratories neutron multiplicity sorting procedure features was applied. The results of experiments used bremsstrahlung were also used. For all 4 nuclei the jointly corrected reaction cross sections were evaluated.

1. Introduction

Photonuclear data are widely used in both basic and applied research and in variety of applications. At the last time there is a renewed interest in photonuclear reactions especially for heavy fission nuclei, first of all to actinides. The importance of those data was reflecting by the existence of the special IAEA coordinated research program [1] and by discussing in many international Conferences, for example [2].

The situation with such kind data is not simple and clear. From one side, there are many data published, but, at the same time, there are many significant disagreements between data obtained using different methods and/or at different laboratories. The majority of experiments devoted to determination of partial photoneutron and photofission reaction cross sections have been carried out at USA National Lawrence Livermore Laboratory and at Centre d'Etudes Nucleaires de Saclay (France) using quasimonoenergetic annihilation photons. A detailed systematic analysis [3] of the (γ, xn), (γ, n) and (γ, 2n) reaction cross-section data obtained at Livermore and Saclay was carried out for 19 nuclei (7 initially): ^{51}V, ^{75}As, ^{89}Y, $_{90}$Zr, ^{115}In, ^{116,117,118,120,124}Sn, ^{127}I, ^{133}Cs, ^{159}Tb, ^{165}Ho, ^{181}Ta, ^{197}Au, ^{208}Pb, ^{232}Th, ^{238}U. It was found out that the (γ,xn) reaction cross-section data obtained at both laboratories without using a neutron multiplicity determination procedure disagreed in absolute value by ~ 10 - 15 % (Saclay data are higher), but the disagreement of the (γ, n) and (γ, 2n) partial reaction cross-sections obtained at both laboratories using neutron multiplicity determination procedure was significantly greater (up to 30 - 40 %), and as a rule in different directions. Those disagreements were interpreted as being the result of differences in the neutron multiplicity determination procedures used in both laboratories: the procedure at Saclay was incorrect, resulting in the incorrect attribution of part of the (γ, 2n) reaction cross- section to the (γ, n) reaction: Saclay data for (γ, 2n) reaction were underestimated (some of those data were interpreted as (γ, n) events) and correspondingly that for (γ, n) reaction – vise versa overestimated. A special method was used to make the data consistent.

Unfortunately for fission nuclei ^{232}Th and ^{238}U the possible contributions of photofission reaction (γ, f) cross sections playing important role in all energy region have not been taken into account [3]. Therefore the new overview and analysis for actinides nuclei ^{232}Th and ^{238}U photonuclear reaction cross sections (γ, xn), (γ, sn), (γ, n), (γ, 2n) and (γ, f) data obtained in both laboratories have been carried out [4]. ^{237}Np for which also data were obtained in both laboratories has to be added to group of fission nuclei and after that new evaluation recommendations have been done for ^{239}Pu investigated only at Livermore.

2. Systematical overview of total and partial photoneutron reactions and photofission reactions cross sections

Complete systematic [5] of the integrated cross sections data ratio $R^{int}_{syst} = \sigma^{int}_{various}(\gamma, xn)/\sigma^{int}_{Livermore}(\gamma, xn)$ for more than 500 of (γ, xn) reaction cross section data for nuclei from ^{3}H to ^{238}U confirms clearly that systematical disagreements exist definitely. The data of Livermore are lower than those of other: ratio values obtained in various laboratories are clearly concentrated near the value $\langle R^{int}_{syst}\rangle = 1.12 \pm 0.24$. Moreover (γ, xn) Saclay reaction cross section data in absolute values are more consistent with data of other laboratories obtained using both quasiumonoenergetic photons and bremsstrahlung than with Livermore data. Such divergences could be caused by "… an Livermore experiments error either in the photon flux determination or in the neutron detection efficiency or in both" [6]. For actinides nuclei under discussion ratios R^{int}_{syst} are to ~ 0.8. That means that (γ, xn) cross sections obtained at Livermore are not smaller but larger than those obtained at Saclay and other laboratories.

The systematics of the values obtained for the coefficients $R(n) = \sigma^{int}_{S}(\gamma, n)/\sigma^{int}_{L}(\gamma, n)$ and $R(2n) = \sigma^{int}_{S}(\gamma, 2n)/\sigma^{int}_{L}(\gamma, 2n)$ for all 19 nuclei mentioned were obtained and analysed in [3] for cross-sections of the single- and two-neutron reactions determined at Saclay (S) and Livermore (L). They clearly confirm: R(n) values are appreciably higher than R(2n); R(n) coefficients mostly have values greater than 1 (roughly 10 - 25 % more); overall, this agrees with the systematics of the R^{int} syst values; R(2n) values are mostly appreciably lower than 1.

3. Comparison of ^{232}Th, ^{238}U and ^{237}Np photoneutron and photofission reaction cross sections obtained both at Saclay and Livermore

All initial data obtained at Livermore and Saclay for ^{232}Th [7, 8], ^{238}U [7, 8] and ^{237}Np [8, 9] were analyzed. ^{232}Th initial data are presented on the left side ("Before") of Fig. 1. The situation is very typical for all three nuclei mentioned: almost all Saclay cross sections under discussion have absolute values clearly smaller than Livermore ones. It is very important that though all reaction cross sections must be in consistency to each other the correspondent ratios S/L are differing individually for each reaction. That is very clearly seen from integrated cross sections and ratios data (Table 1, columns "Before"): 0.62 - 1.02 for ^{232}Th, 0.80 - 0.88 for ^{238}U and 0.41 - 0.93 for ^{237}Np. It must be pointed out that for each of three nuclei under investigation the situations are individual: for ^{238}U Saclay and Livermore data are near consistency, for ^{232}Th the most inconsistency exists for (γ, f) reaction, but for ^{237}Np – for (γ, 2n) reaction. For all three nuclei data confirm the conclusions [3] that at Saclay compare to Livermore σ(γ, n) reaction cross sections are overestimated and those for σ(γ, 2n) reaction underestimated. From the [3] point of view it is resulted from the different procedures of neutron multiplicity sorting.

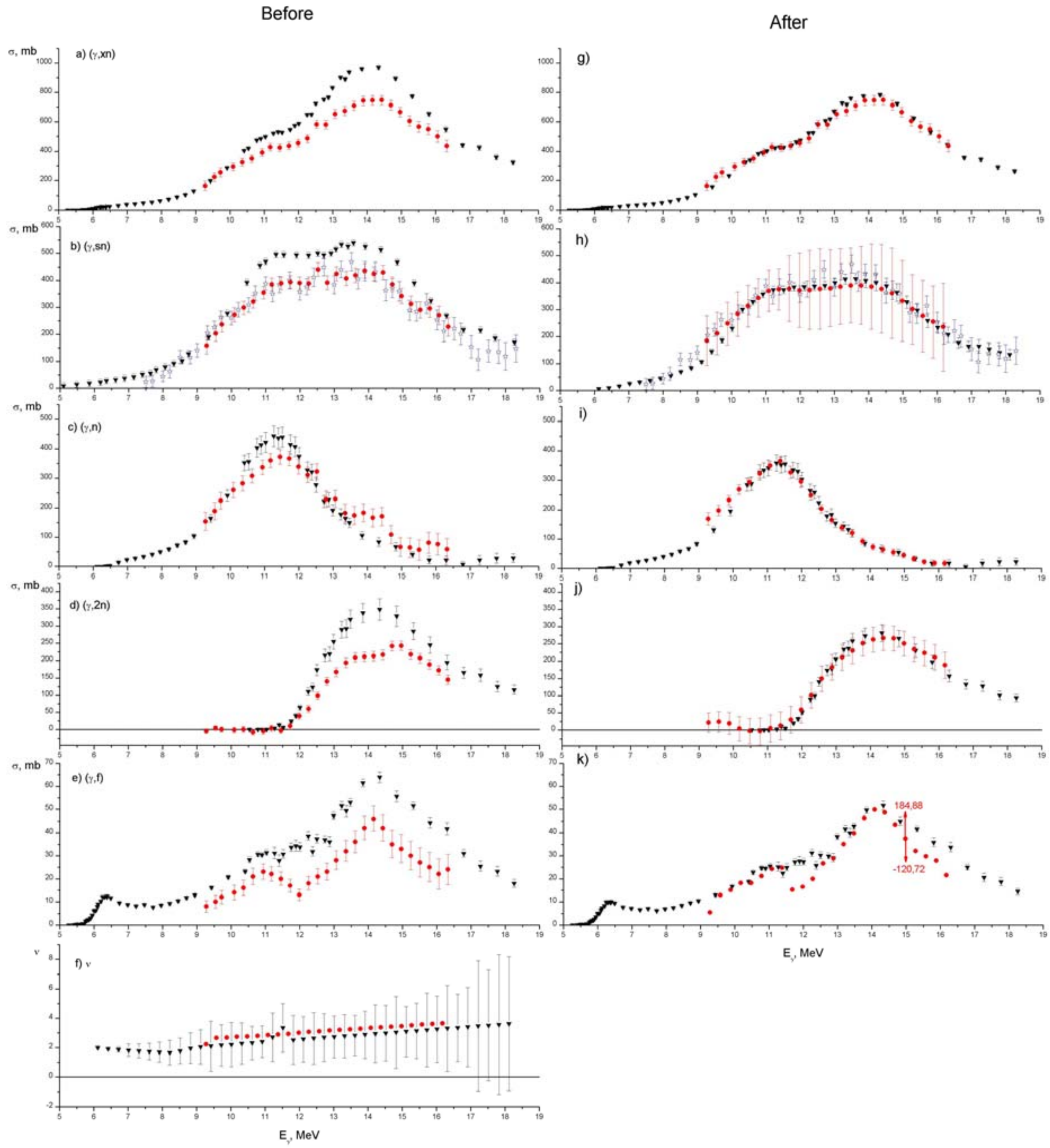

Fig 1. All published initial ("Before", left) and evaluated ("After", right) data for ^{232}Th under discussion (Livermore [7] – triangles, Saclay [8] – circles) data: *a*, *g* – total photonuclear reaction (γ,xn) cross section; *b*, *h* – (γ, sn) reaction cross section; stars – (γ, abs) cross section [10]; *c*, *i* – (γ, n) reaction cross section; *d*, *j* – (γ, 2n) reaction cross section; *e*, *k* – (γ, f) cross section; *f* – neutron multiplicity.

Table 1. **Various reactions integrated cross section Saclay (S) and Livermore (L) data and ratios (S/L) calculated for joint energy ranges before and after joint evaluation**

Reaction	Before			After		
	S, MeV·mb	L, MeV·mb	Ratio S/L	S, MeV·mb	L, MeV·mb	Ratio S/L
^{232}Th S[8] and L [7] data and S/L ratios						
(γ, xn)	3637 ± 82	4594 ± 27	0.79	3636 ± 82	3711 ± 22	0.98
(γ, sn)	2468 ± 72	3046 ± 31	0.81	2304 ± 334	2322 ± 53	0.99
(γ, n)	1510 ± 78	1482 ± 59	1.02	1205 ± 37	1194 ± 48	1.00
(γ, 2n)	784 ± 29	1160 ± 51	0.68	912 ± 88	913 ± 40	1.00
(γ, f)	175 ± 12	284 ± 5	0.62	194 ± 413	225 ± 4	0.86
^{238}U S [8] and L [7] data and S/L ratios						
(γ, xn)	6054 ± 165	7283 ± 40	0.83	6054 ± 165	5823 ± 32	1.04
(γ, sn)	2945 ± 22	3806 ± 37	0.77	2837 ± 157	2740 ± 54	1.04
(γ, n)	1161 ± 30	1320 ± 55	0.88	1070 ± 30	1052 ± 44	1.02
(γ, 2n)	906 ± 20	1129 ± 51	0.80	929 ± 57	892 ± 40	1.04
(γ, f)	895 ± 16	1065 ± 8	0.84	869 ± 120	822 ± 6	1.06
^{237}Np S [8] and L [10] data and S/L ratios						
(γ, xn)	7242 ± 133	9056 ± 109	0.80	7242 ± 133	7104 ± 86	1.02
(γ, sn)	2529 ± 86	3159 ± 110	0.80	2462 ± 203	2404 ± 84	1.02
(γ, n)	937 ± 121	1016 ± 77	0.93	876 ± 62	788 ± 60	1.11
(γ, 2n)	120 ± 73	291 ± 50	0.41	134 ± 144	222 ± 38	0.60
(γ, f)	1520 ± 35	1890 ± 18	0.81	1467 ± 119	1420 ± 13	1.03

As was mentioned above because of very low thresholds of (γ, f) reaction instead of equation σ(γ, xn) = σ(γ, n) + + 2σ(γ, 2n) has been used in [3] the following one σ(γ, xn) = σ(γ, n) + 2σ(γ, 2n) + νσ(γ, f) must be used for joint evaluation where ν is the averaged prompt photofission neutron multiplicity. Using the last equation one can obtain (Fig. 1, *f*) real values for ν from the experimental photoneutron and photofission reaction cross sections

$$\nu = [\sigma(\gamma, xn) - \sigma(\gamma, n) - 2\sigma(\gamma, 2n)]/\sigma(\gamma, f). \quad (1)$$

For nuclei under discussion ν data have been obtained for Livermore cross sections are in good agreement with Livermore data [9, 11]. Data for ν obtained using equation (3) for Saclay cross sections are close to Livermore data for ^{238}U, systematically higher in comparison with Livermore data for ^{232}Th (Fig. 1, *f*) and have clear differed energy dependence for ^{237}Np. Because of that later on the Livermore ν data will be used for correction of Saclay reaction cross sections.

4. Photoneutron and photofission reaction cross sections joint evaluation

4.1. ^{232}Th and ^{238}U photoneutron and photofission reaction cross sections

Because of unusual balance of total photonuclear reaction (γ, xn) cross sections – those from Livemore are not smaller but larger that those from Saclay – it is very important to compare both of them with another kind data, first of all obtained using bremsstrahlung.

Unfortunately there are not such kind data, but for both nuclei the total photoabsorption cross sections

$$\sigma(\gamma, abs) = \sigma(\gamma, n) + \sigma(\gamma, 2n) + \sigma(\gamma, p) + \sigma(\gamma, f) \quad (2)$$

obtained using bremsstrahlung have been published [10].

It is well known that σ(γ, p) for heavy nuclei is very small in comparison of others mentioned. For example for ^{208}Pb the amplitude of σ(γ, p) is equal to ~ 2 mb, σ(γ, 2n) - ~ 140 mb, σ(γ, n) - ~ 700 mb. Therefore one can describe the total photoabsorption reaction cross section as

$$\sigma(\gamma, abs) \approx \sigma(\gamma, n) + \sigma(\gamma, 2n) + \sigma(\gamma, f) = \sigma(\gamma, sn). \quad (3)$$

The experimental total photoabsorption reaction cross section [10] for ^{232}Th is presented on Fig. 1, *b*. The situation is typical for ^{232}Th, ^{238}U and ^{237}Np. One can see that the shape of σ(γ, abs) [10] is in very good agreement with the shapes of both Saclay σ(γ, sn) [8] and Livermore σ(γ, sn) [7]. At the same time absolute value of σ(γ, abs) [10] is in agreement to that of Saclay σ(γ, sn) [8] and contradicts to that of Livermore σ(γ, sn) [7]. That confirms again the conclusions [3, 4] about reliability of total photoneutron reaction cross section data obtained at Saclay and about many doubts in reliability of those obtained at Livermore.

Livermore data evaluation. Therefore the way to evaluate the most reliable data for all σ(γ, xn), σ(γ, sn), σ(γ, n), σ(γ, 2n) and σ(γ, f) reaction cross sections is normalization of Livermore experimental cross sections using ratio

$$K = \sigma^{int}_{[10]}(\gamma, abs)/\sigma^{int}_{[8]}(\gamma, sn). \quad (4)$$

For ^{232}Th (Fig. 1, *b*) K(Th) = 0.84, for ^{238}U (Fig. 2, *b*) K(U) = 0.83. The data obtained for ^{232}Th for all reaction cross sections by this way – normalization of Livermore data using K(Th) and K(U) calculated values are presented in Fig. 1, *g* - *k*.

Saclay data evaluation. Because of Saclay incorrect procedure of neutron multiplicity sorting [3] the reciprocal correction method proposed for joint Saclay and Livermore data evaluation must be used:

$$R = \sigma(\gamma, xn)_S/\sigma(\gamma, xn)_L = (\sigma(\gamma, n)_S + 2\sigma(\gamma, 2n)_S)/(\sigma(\gamma, n)_L + 2\sigma(\gamma, 2n)\sigma_L), \quad (5)$$

$$\sigma(\gamma, xn)_S = (\sigma(\gamma, n)_S + 2\sigma(\gamma, 2n)_S) = R\sigma(\gamma, xn)_L = R(\sigma(\gamma, n)_L + 2\sigma(\gamma, 2n)\sigma_L), \quad (6)$$

$$R\sigma(\gamma, 2n)_L = \sigma(\gamma, 2n)_S^{eval} = \sigma(\gamma, 2n)\sigma_S + 1/2(\sigma(\gamma, n)_S - R\sigma(\gamma, n)_L), \quad (7)$$

$$R\sigma(\gamma, n)\sigma_L = \sigma(\gamma, n)_S^{eval} = \sigma(\gamma, n)\sigma_S - (\sigma(\gamma, n)_S - R\sigma(\gamma, n)_L). \quad (8)$$

In accordance with the method described the way for Saclay data evaluation:

- after appropriate correction of the energy scales of the cross sections to be compared [3] the ratio $R = \sigma^{int}(\gamma, xn)_{[8]}/\sigma^{int}(\gamma, xn)_{[6]}$ is calculated; R(Th) = 0.93, R(U) = 1.00;
- in the energy region below the reaction (γ, 2n) threshold $\sigma^{eval}(\gamma, n)_S = \sigma^{exp}(\gamma, n)_S$;
- in the energy region behind $\sigma^{eval}(\gamma, n)_S = R\,\sigma^{exp}(\gamma, n)_S$;
- $\sigma^{eval}(\gamma, 2n)_S = \sigma^{exp}(\gamma, 2n)_S + 1/2\,[\sigma^{exp}(\gamma, n)_S - R\,\sigma^{exp}(\gamma, n)_L]$;
- $\sigma^{eval}(\gamma, f)_S = [\sigma^{exp}(\gamma, xn)_S - \sigma^{exp}(\gamma, n)_S - 2\,\sigma^{eval}(\gamma, 2n)_S]/\nu_L$, where ν is the averaged prompt photofission neutron multiplicity obtained for correspondent Livermore data (Fig. 1, *f*).

Saclay data evaluated by the way described are also presented on Fig. 1, *g* - *k* (circles). From Fig. 1 and Table 1 one can see that on the whole these data are in good agreement with correspondent evaluated Livermore data. Arbitrarily poor agreement for reaction ^{232}Th(γ, f) could be because that the shapes of initial Livermore and Saclay cross sections differ significantly.

4.2. ^{237}Np photoneutron and photofission reaction cross sections

Unfortunately there are no appropriate data for extra normalization of data for ^{237}Np. But total photoneutron reaction (γ, xn) cross sections obtained at Saclay unlike to those obtained at Livermore are in consistency with many data obtained at various other laboratories using various photon beams. Therefore one can use normalization of Livermore (γ, xn) reaction cross sections data to Saclay ones. All other ideas described can be used for joint evaluatuion of the all discussed reaction cross sections obtained at Livermore [9] and Saclay [8].

Livermore data evaluation. The way to evaluate data for all σ(γ, xn), σ(γ, sn), σ(γ, n), σ(γ, 2n) and σ(γ, f) reaction cross sections is normalization of Livermore experimental cross sections using coefficient $R = \sigma(\gamma, xn)_S/\sigma(\gamma, xn)_L = \sigma^{int}(\gamma, xn)_{[8]}/\sigma^{int}(\gamma, n)_{[7]} = 0.78$.

Saclay data evaluation. For Saclay data the method described above (5) - (8) must be applied. The integral cross section ratio data obtained for all reaction cross sections are presented in Table 1. One can see that similar to two other nuclei under discussion on the whole evaluated Saclay data are in good agreement with correspondent evaluated Livermore data. Arbitrarily poor agreement is achieved for reaction ^{237}Np(γ, 2n). The possible reason could be that data obtained at both laboratories have very different shapes and at the same time very poor accuracy.

Table 2. **Final evaluated integrated cross section data (renormalized Livermore data)**

Nucleus	Reaction	E^{int}_{max}, MeV	σ^{int}, MeV·mb	EXFOR SUBENT
^{232}Th	(γ, xn)	18.3	4534.6	002
	(γ, sn)	18.2	2795.3	003
	(γ, n)	18.3	1344.0	004
	(γ, 2n)	18.3	1175.1	005
	(γ, f)	18.3	300.2	006
^{238}U	(γ, xn)	18.3	5983.9	007
	(γ, sn)	18.2	2854.1	008
	(γ, n)	18.3	1090.0	009
	(γ, 2n)	18.3	908.5	010
	(γ, f)	18.3	870.3	011
^{237}Np	(γ, xn)	17.8	8227.9	012
	(γ, sn)	18.0	2942.2	013
	(γ, n)	18.2	917.4	014
	(γ, 2n)	18.2	271.8	015
	(γ, f)	18.2	1782.7	016
^{239}Pu	(γ, xn)	17.7	9039.8	017
	(γ, sn)	17.7	2730.1	018
	(γ, n)	17.7	580.6	019
	(γ, 2n)	17.7	(130.0)	020
	(γ, f)	17.7	1976.3	021

4.3. ^{239}Pu photoneutron and photofission reaction cross sections

The situation with data published for ^{239}Pu (Fig. 2) can be treated as intermediate between two situations studied before – there are only Livermore data [9] and no Saclay data. But there are photoabsorption reaction data obtained bremsstrahlung [10]. Unfortunately for ^{239}Pu in difference to ^{232}Th and ^{238}U discussed above the σ(γ, abs) [10] and Livermore σ(γ, sn) [7] are in contradiction not only in absolute value but in shape also. Moreover the very strange shapes (negative values) of (γ, sn), (γ, n), (γ, 2n) reactions cross sections in energy range near ~ 15 - 17 MeV are the reasons to suspect that in difference to all cases analyzed before in the case of ^{239}Pu there are some additional photoneutron multiplicity errors in Livermore data.

But nevertheless formally all ideas described give to one possibility to use simple way for obtaining the evaluated (slightly improved) data – normalization of Livermore ^{239}Pu(γ, sn) data to bremsstrahlung photoabsorption data using coefficient $K^*(Pu) = \sigma^{int}_{[9]}(\gamma, abs)/\sigma^{int}_{[8]}(\gamma, sn) = 0.92$, where * means that K was calculated for energy range up to B(2n) - threshold of (γ, 2n) reaction.

Evaluated cross sections are included into the IAEA EXFOR fund as M0722 ENTRY, correspondent integrated cross section data are presented in the Table 2.

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9th International Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation NEA/OCDE. Nimes (France), September. Simulation is still and will continue to be for few years, the basis of the evaluation and extrapolation of performance, viability, cost and safety of the proposed transmutation devices. Improvements on the simulation tools, programs and evaluated nuclear data libraries, are key elements to base our benefit/cost evaluations and to prepare a solid base for the definition of DEMO reactors that can provide the required validation before industrial deployment of tr...Â Activation reaction Cross section âŠ— (n,Î³), (n,Î³)* of actinides with (n,2n) +â€¦ half-live > 100d. Neutron flux Spectrum elastic, inelastic,(n,2n),â€¦ fuel matrix, Struct. Figure 1. 237Np(n,f) cross-section in the vicinity of the rst resonance cluster around En = 40 eV (Fig. 6 [12]) and in the neutron energy range between 1.5 MeV and 9 MeV (Fig. 7 [14]); the depicted data are from dierent experiments, as well as from dierent evaluated nuclear data libraries (see the references in [12â€“14]).Â For example, several experiments based on the time-of-ight (TOF) technique were dedicated to the measurement of ssion cross section data in a wide neutron energy range (0.7 eVâ€“1 GeV), using dierent ssion-fragment detectors [12â€“14].Â That is why, to study the 237Np(n,f) and 238U(n,f) cross sections in a comprehensive manner, two experiments were performed at two dierent Van de Graaf (VDG) facilities: IRMM and NPL. Analysis of both experiments is in progress.