THE NEUTRON CAPTURE AND TOTAL CROSS SECTION OF ^{99}TC IN THE RESONANCE REGION

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Abstract

In order to improve nuclear data for nuclear waste transmutation purposes, measurements of the neutron radiative capture and total cross sections of ^{99}Tc in the resonance region have been performed, using the time-of-flight method at the pulsed white neutron source GELINA of the Institute for Reference Materials and Measurements (IRMM) in Geel, Belgium. A neutron capture experiment has been set up and samples of thickness 0.10, 0.26 and 1.0 g/cm^2 have been measured. For the total cross section, a transmission experiment was carried out on samples of thickness 0.16, 0.44 and 4.0 g/cm^2. For both measurements, the total energy range covered was from 3 eV to 100 keV. The neutron resonance shape fitting programme REFIT was used to derive the neutron resonance parameters. The results up to 2 keV are presented here.

Introduction

During the period of irradiation of the fuel assemblies in the core of a nuclear reactor, noxious waste is produced by fission and activation. In order to reduce the amount of the fission products and minor actinides which could induce very long-term radioactivity, transmutation techniques using reactors (fast or thermal) or hybrid systems are under study. One of the most harmful fission products is ^{99}Tc. Produced in very large quantities in the Pressurized Water Reactors (about 1 ton is extracted per year from spent fuel in France), ^{99}Tc has a very long β^- half-life of 2×10^5 years. It also has the ability to migrate in any kind of material, including the storage glasses, which could, in a worst-case scenario, lead to its release in the environment during long-term storage. But, after neutron capture, ^{99}Tc becomes ^{100}Tc, decaying to the stable isotope ^{100}Ru with a half-life of 16 s. Therefore, ^{99}Tc is a very good candidate for transmutation.

Several measurements of the neutron cross sections of ^{99}Tc have been performed in the past [1-5] concerning limited energy ranges and providing a reduced number of resonance parameters. Two existing evaluations, JEF2.2 [6] and ENDF/B6-r3 [7] have also been considered, containing respectively parameters for 89 resonances up to 1.114 keV and 68 resonances up to 800 eV. As more precise parameters were needed over a wider energy range, new measurements of the total and capture cross sections of ^{99}Tc have been required in the High Priority Nuclear Data Request List [8] of the OECD, from 1 eV to 100 keV, with an accuracy of 5%.

Apart from the transmutation purposes, the improvement of the cross sections of ^{99}Tc is of great interest for the reactivity calculations of power reactors. According to the large quantities produced, its absorption near the end of the fuel cycle is noticeable, specially in the epithermal range.

Experimental details

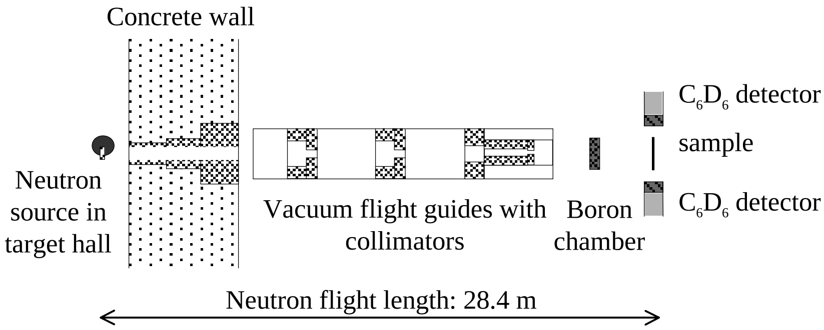
In the framework of a collaboration between the Commissariat à l'Energie Atomique (CEA, Saclay, France) and the Institute for Reference Materials and Measurements (IRMM, Geel, Belgium), radiative capture and transmission measurements have been performed at the pulsed white neutron source GELINA (Geel Linear Accelerator) of the IRMM. The samples have been made at the sample preparation group of the IRMM. Two sets of measurements have been carried out and the experimental details are given in Table 1.

Table 1. **Experimental details**

Energy range	Low energy measurements 3 → 800 eV		High energy measurements 50 eV → 100 keV	
	Capture	Transmission	Capture	Transmission
Sample thickness (g/cm^2)	0.10	0.16	1.0	4.0
	0.26	0.44	2.0	
Total acquisition time (h)	250	230	380	450
Mean energy of e^- (MeV)	100		100	
Pulse width (ns)	15		2	
Average beam current (μA)	20		60	
Repetition rate (Hz)	200		800	
Sample composition	$^{99}Tc_4Al_{11}$		metallic Tc	

Capture Set-up

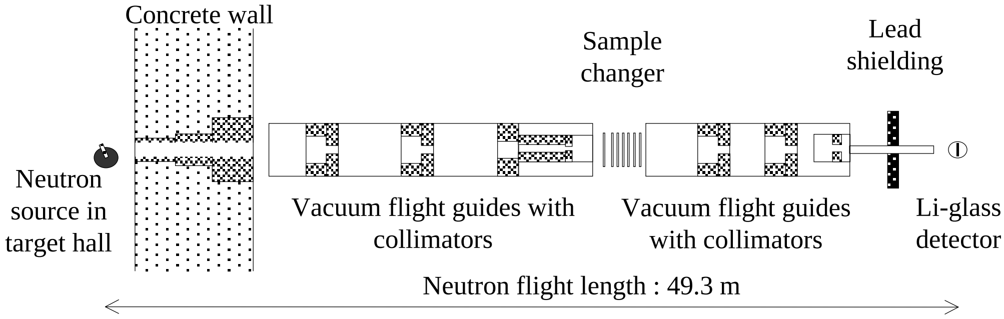
Figure 1. **Schematic view of the capture set-up**



The neutron beam produced by the neutron source was collimated in the evacuated flight tubes before reaching the sample. The prompt gamma rays emitted after the $^{99}Tc(n,\gamma)^{100}Tc$ reaction were detected by two total energy C_6D_6 detectors, placed at 90° on both sides of the sample. At the same time, the incoming neutron flux was measured with a boron chamber placed in the beam before the sample. The efficiency of the capture detectors was made independent of the gamma decay mode using a pulse height weighting method: the experimental weighting function used was determined experimentally at Geel [9,10]. The background on the incoming neutron flux was measured using the black resonance method, placing filters at 10 m from the neutron source. The background on the capture detectors was determined by iterations between the reduction and analysis programmes. The acquisition of the capture data was made in list-mode, using a dedicated package [11] developed at IRMM and complemented with a set of post-acquisition programmes. This post-treatment of the data for each cycle of measurement allows to check possible modifications of the experimental conditions, which could induce systematic uncertainties on the results. It also reconstructs the summed histogrammes from the lists of all the cycles of the whole measurement.

Transmission set-up

Figure 2. **Schematic view of the transmission set-up**



The collimated neutron beam was attenuated by the sample placed at a distance of 23 m from the neutron source. The neutrons which did not react in the sample were further collimated before being detected by a lithium glass, placed at a flight distance of 49.3 m. The transmission set-up is provided with a sample changer, driven by the acquisition system. Each measurement cycle was composed of four sequences, recorded alternatively in order to reduce systematic uncertainties. In the first two sequences, data were recorded with the sample in and out of the beam. In the two last sequences, filters were added in the beam with and without the technetium sample, in order to derive the background. The acquisition system, from the trademark FAST [12], was used in histogramme-mode.

Data reduction

The experimental data coming from the capture and transmission acquisition systems, composed of different histogrammes and scalers, were combined and corrected for experimental effects. The data reduction package [13,14] used has been developed at IRMM, in collaboration with CEA. It calculates propagation of the uncertainties [15], statistical and correlated induced by the different parameters used at each stage of the data reduction. The spectra, the associated uncertainties and a description of the action performed at each step are gathered in a unique file. The results of the data reduction are the experimental transmission and capture yield, and the total associated covariance matrices.

The dead time correction was applied to the transmission experimental spectra. The backgrounds were determined and subtracted from the spectra with and without the technetium sample in the beam. The transmission is obtained as the ratio of these two spectra. The transmission factor was normalised the flux monitors located in the uranium target hall.

Concerning the capture measurements, the pulse height weighting function was applied to the bi-dimensional C_6D_6 spectra. The histogrammes, after dead time correction and background subtraction, were normalised at present to the capture area determined for the resonances from the transmission experiment.

The results of the capture and transmission measurements are presented in Figure 3 at the end of the data reduction. The capture yield is represented in the upper parts of the graphs for the sample of 1.0 g/cm^2 thickness and the transmission in the lower part for the sample of 4.0 g/cm^2 thickness.

Analysis

The analysis of the data was made with the neutron resonance shape analysis programme REFIT [16], using the least squares method.

Preliminary resonance parameters obtained from the analysis of the transmission data have been used as a starter file for the capture yield analysis and until now, capture and transmission have been processed separately. For each measurement, the different thicknesses were first analysed separately before being processed simultaneously in the common energy ranges.

The systematic uncertainties are still under study. The errors presented in all the tables here are calculated by the data reduction programme, as the result of the propagation of the contribution of the statistical uncertainties and the contribution of the correlated uncertainties induced by the parameters used during the reduction.

Figure 3. **The capture yield and transmission spectra derived from the measurements up to 90 keV, at the end of the data reduction**

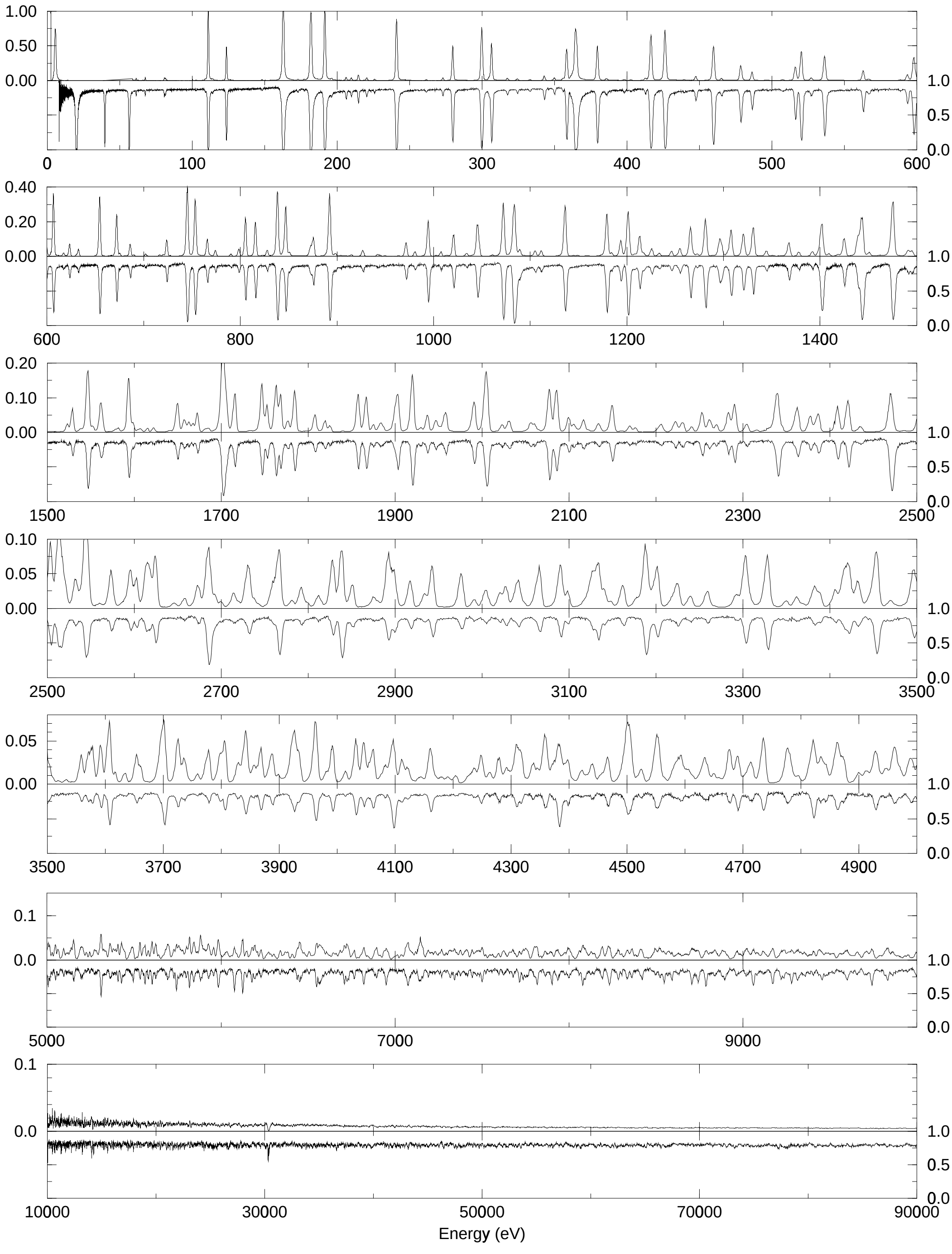
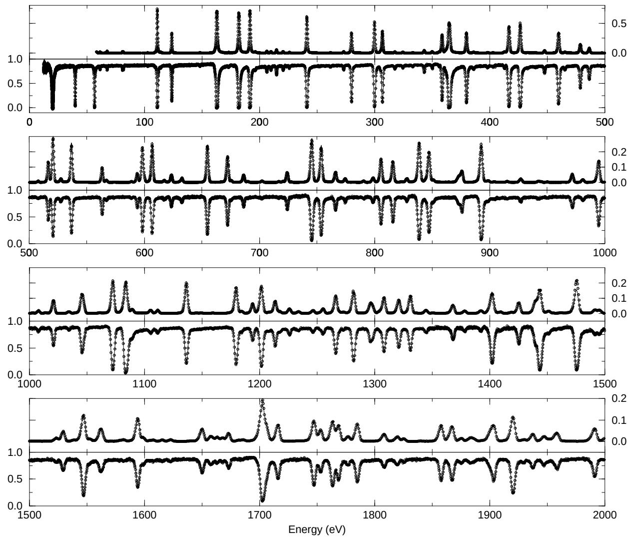


Figure 4. **The capture yield and transmission resulting from the analysis up to 2 keV on the same energy scale**

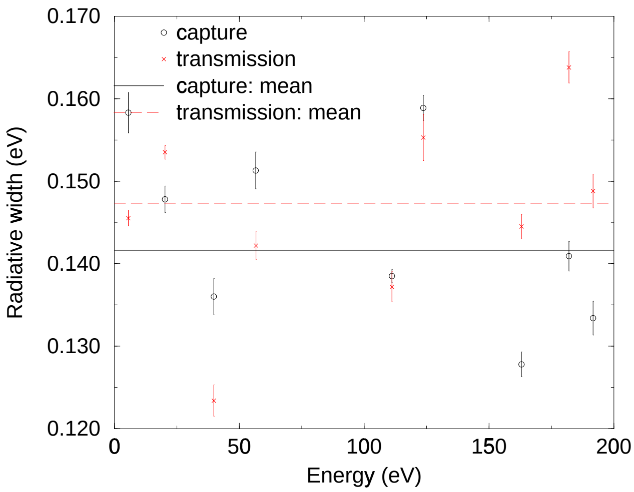


In addition to the capture and transmission study, a measurement of the prompt gamma capture decay spectra was done with a Ge-detector at IRMM, and the spin of 51 *s*-resonances was determined. A parity assignment was also performed to separate ℓ=0 and ℓ=1 resonances up to 1 keV (assuming that higher total angular momentum resonances are not detected in our energy range). The results of these studies [17] have been introduced in the analysis.

Up to 2 keV, capture yield and transmission resulting from the adjustments are shown in Figure 4, for the sample of thickness 1.0 g/cm^2 and 4.0 g/cm^2, for capture and transmission, respectively. The experimental points are represented by small circles with the uncertainty resulting from the data reduction, the result of the adjustment by the plain line.

Figure 5. **Distribution of the adjusted G_g for capture and transmission on *s*-resonances below 200 eV and comparison of the values given in references [1,2,6,7,18] and obtainedfrom the present study**

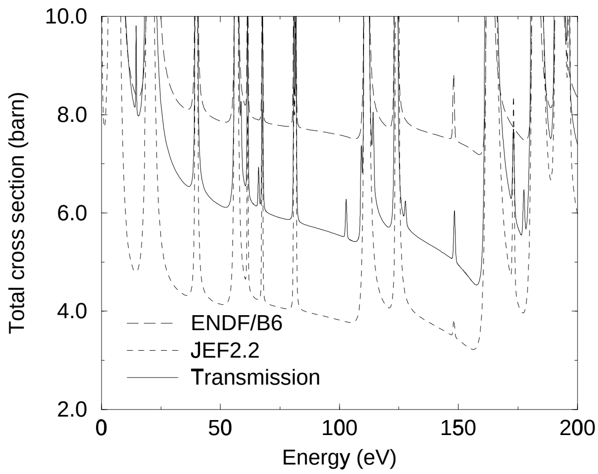
	$\overline{\Gamma}_\gamma$ (meV)
Watanabe et Reeder [1]	112
Adamchuck et al. [2]	174 ± 8
Mughabghab [18]	160
ENDF/B6-r3 [7]	130
JEF2.2 [6]	160.4
Present study: capture	141.6 ± 11
Present study: transmission	147.4 ± 12



The value of the resolution function, due mainly to the Doppler effect and in a minor part to the experimental set-ups (machine + detectors), allowed us to deduce the radiative width Γ_γ for both measurements, on 9 *s*-resonances below 200 eV. As the distribution of Γ_γ shows little variations in energy, and as the adjustment was not possible for *p*-resonances, which are much smaller than *s*-resonances, the means of the distributions have been assigned to all other *s*- and *p*-resonances. Figure 5 shows the distribution of the adjusted Γ_γ and the mean values for capture and transmission. For both measurements, the dispersion of the distribution is 8%.

Figure 6. **Comparison of the potential scattering radius, for references [6,7,18] and the present study and comparison of the corresponding total cross sections of ^{99}Tc at 300 K**

	R (fm)
JEF2.2	6.00
ENDF/B6-r3	7.91
Mughabghab	6.00 ± 0.50
Present study	7.20 ± 0.30



The potential scattering radius was deduced from the transmission measurement of the thickest sample (4.0 g/cm^2). Figure 6 gives a comparison between the values given in the evaluations JEF2.2 [6] and ENDF/B6-r3 [7], and the present study. Our result is situated between the two evaluations. It can be seen on the graph, which shows the total cross sections calculated at 300 K with NJOY for the evaluations and with REFIT for the experimental results. The value of the total cross section between the resonances is very sensitive to the potential scattering.

The high resolution of GELINA allowed to detect small resonances, unreported before. A total of 224 resonances have been fully analysed from the transmission results up to 2 keV. As the capture measurements are more sensitive to small resonances, 245 resonances are reported from the capture measurements in the same energy range. An example of new resonances is given in Figure 7, as a comparison between the capture cross sections calculated with NJOY [19] for the two evaluations [6,7] and from the present results of the transmission measurements.

Figure 7. **Comparisons of the capture cross sections of ^{99}Tc, calculated at 300 K with NJOY [19] for the two evaluations [6,7] and from our results. In the left graph, capture and transmission coincide.**

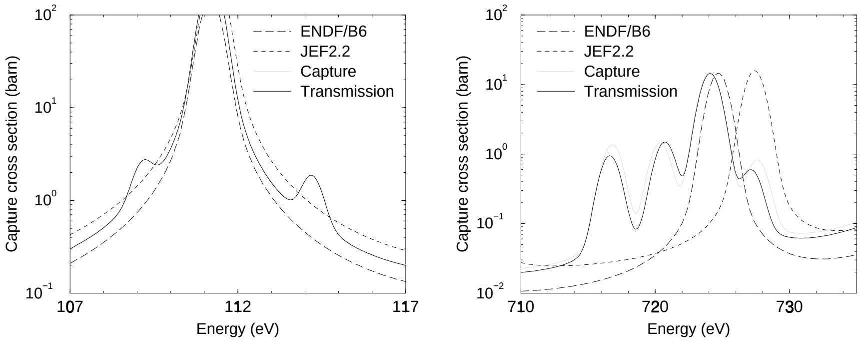
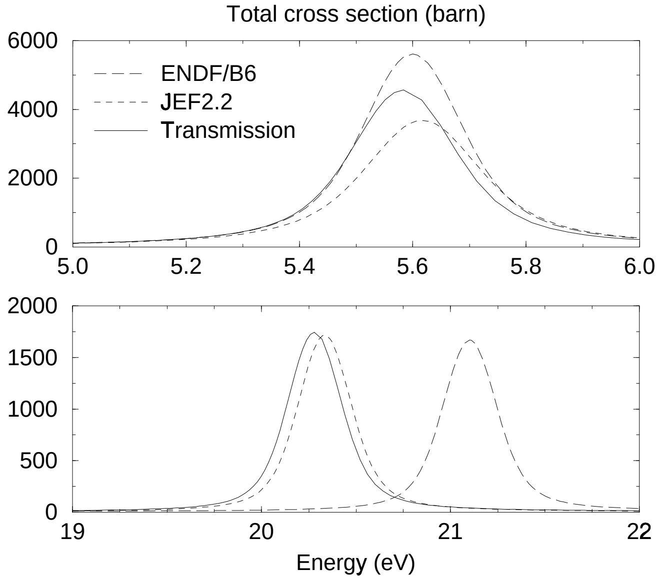


Table 2. **Resonance parameters of the first and second *s*-resonances of ^{99}Tc, from the present work, the evaluations [6,7] and previous measurements [1,2,3,4]**

	First *s*-resonance			Second *s*-resonance		
	E (eV)	$2gG_n^0$ (meV)	G_g (meV)	E (eV)	$2gG_n^0$ (meV)	G_a (meV)
Watanabe et Reeder [1]	5.6	1.84 ± 0.15	134 ± 4	20.3	1.66 ± 0.03	140 ±7
Adamchuck *et al*.[2]	5.6	-	-	21.1 ± 0.02	1.61 ± 0.26	150 ± 30
Chou *et al*.[3]	5.65	1.29 ± 0.36	263 ± 6.5	20.3	1.58 ± 0.41	263.9 ± 9.8
Fischer *et al*.[4]	5.59 ± 0.01	1.45 ± 0.07	171.5 ± 4.5	20.32 ± 0.01	1.49 ± 0.015	176.0 ± 3.9
JEF2.2[6]	5.6175	1.481	177	20.333	1.599	147
ENDF/B6-r3[7]	5.60	1.902	134	21.100	1.611	150
Present analysis: capture	5.5837 ± 0.0007	1.6186 ± 0.02	158.3 ± 2.4	20.272 ± 0.0007	1.7303 ± 0.02	147.8 ± 8.7
Present analysis: transmission	5.5835 ± 0.0003	1.6098 ± 0.007	145.5 ± 0.91	20.273 ± 0.0005	1.6719 ± 0.005	153.5 ± 0.82

The first *s*-resonances of ^{99}Tc, of energies 5.6 and 20.3 eV, are the most important ones. Our resonance parameters are compared to the values of different references [1-4,6-7] in Table 2, and the total cross sections are plotted in Figure 8, for the transmission measurements and two evaluations [6,7]. Concerning the first resonance, the present study is situated between the two evaluations, which presents a large difference in size. Concerning the second one, at 20.3 eV, there was an important discrepancy in the energy and our results seem to be in good agreement with the JEF2.2 data.

Figure 8. **Total cross section of ^{99}Tc at 300 K, obtained with NJOY [19] for the evaluations [6,7] and calculated with the results of the present study**



Conclusion and perspectives

The present work adds a considerable amount of consistent information to the existing one. Transmission and capture measurements have been performed on a large energy range and with different sample thickness. The analysis of the experimental data has been made up to 2 keV at the present time and adds a noticeable number of new information concerning the resonance region of ^{99}Tc.

The analysis of both measurements is still going on. Resonances will be resolved for both measurements at least up to 5 keV, probably more. A simultaneous adjustment of capture and transmission measurements will be done, and a new parity assignment will be performed on the resulting resonance parameters. From theses parameters and the results of the measurements, average parameters will be derived in the unresolved range up to 100 keV. At the present time, the systematic uncertainty has not been determined and is not taken into account in the results presented here. A first evaluation shows that, for both measurements, it will be situated between 4% and 5%.

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