Preliminary results of new GELINA data for natural cadmium

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Abstract. The total neutron cross section of cadmium (Cd) is very large in the thermal neutron energy region which determines its special role in reactor technology. The first resonance of $^{113}$Cd at 0.178 eV is of high importance in the interpretation of Cd neutron cross section structure in the thermal region. The evaluated data files for Cd isotopes and particularly the resonance parameters do not correspond to the requirements of the advanced reactor technologies. To provide data for a new evaluation, a set of top quality transmission experiments has been designed and performed at the neutron time-of-flight facility GELINA at the Institute for Reference Materials and Measurements (IRMM) in Geel, Belgium. The measurements have been carried out at 26.45 m and 50 m flight paths of GELINA at 50 Hz, 400 Hz and 800 Hz working frequencies of the LINAC, using natural Cd samples with various thicknesses (purity 99.99% in Cd). The experiments at 50 Hz provide high quality data in the thermal energy region, from which we could extract the resonance parameters for the first resonance of $^{113}$Cd at 0.178 eV with high accuracy. Considering the transmission data obtained with various sample thicknesses as a source of additional information for low level cross section we validate a new approach for cross section description in this energy region.

1. Introduction:
Cadmium (Cd) is one of the elements with a large total neutron cross-section in the thermal neutron energy region, which determines its special role in reactor technology. The total neutron cross-section of cadmium is crucial either for the design and development of nuclear reactors and also for the basic study of neutron interaction with nuclei. The first resonance of $^{113}$Cd at 0.178 eV is of high importance for the interpretation of Cd neutron cross section structure in the thermal region.

The evaluated data files for Cd isotopes and particularly the resonance parameters do not correspond to the requirements of the advanced reactor technologies. For a variety of nuclides differences between the resonance integral as measured by integral experiments and the values derived from resonance parameters are observed. It has been suggested that the interpretation of the integral measurements could be the source for some of the observed discrepancies. Many of these integral experiments use cadmium foils as shielding against thermal neutrons and it was proposed that these foils are the sources of the observed discrepancies [1]. To clarify the situation and provide data for a new evaluation, a set of top quality transmission experiments has been designed and performed at the neutron time-of-flight facility GELINA, at IRMM in Geel, Belgium.
2. Experimental details
To improve the experimental data base for the determination of the resonance parameters for \(^{113}\)Cd, high resolution transmission and capture measurements were performed at GELINA [2, 3]. It is a pulsed neutron source providing neutrons in the energy range between 1 meV and 20 MeV. Intense pulsed electron beams with repetition rates up to 800 Hz are accelerated to an energy up to 150 MeV in a linear electron accelerator. The electron pulses are compressed using a specially designed post-acceleration compression magnet to a duration of less than 1 ns [4]. After the compression, these high-energy electrons hit the rotating mercury-cooled uranium target [5]. Colliding with the target, electrons produce Bremsstrahlung in it, and subsequently neutrons by \((\gamma,n)\) and \((\gamma,f)\) reactions. To enhance the neutron intensity in the low energy region, two water-filled beryllium containers are used as moderators.

The determination of the neutron energy is based on the Time-Of-Flight (TOF) method. The very short pulse duration of less than 1 ns in combination with the very long flight paths of up to 400 m results in the extremely high energy resolution of the GELINA TOF facility.

Cadmium transmission measurements were built up at 26.45 m and 50 m flight paths of GELINA, which form an angle of +9° and -9°, respectively, with the normal to the moderator. The experiments were carried out at 50 Hz, 400 Hz and 800 Hz working frequencies of the LINAC, using natural Cd samples enriched to 99.99% in Cd.

A detailed description of the experimental arrangement of the Cd transmission measurements at the 26.45 m flight-path is shown in figure 1. The Cd samples were placed at 9.1 m distance from the neutron producing target in an automatic sample changer, operated by the data acquisition system. Black resonance filters were also mounted on the sample changer. The count rate in the minima of the black resonances was used to determine the time dependence of the background.

After the filters and the sample, the neutron beam was further collimated and finally detected by a Li-glass scintillator (NE 905), enriched to 95% in \(^{6}\)Li, placed at 26.45 m from the neutron producing target. This scintillator, with an effective diameter of 110 mm and a thickness of 12.7 mm, was placed in an aluminium canning and viewed by two EMI 9823 KQB PhotoMultiplier (PMT), placed outside the neutron beam and perpendicularly to the neutron beam axis. The aluminium canning was covered with a thin Teflon foil to enhance the light reflection. The temperature at the sample position was continuously monitored. The average temperature is used in the resonance shape analysis program to calculate the Doppler broadening of the resonances.

The anode pulses of both PMT’s are transmitted to a constant fraction discriminator to create fast logic signals for a coincidence unit. Whenever a signal from each PMT was present within a 30 ns coincidence window, a coincidence pulse was sent to the Time to Digital Converter (TDC), which determines the TOF of the detected neutron from the time difference between the start signal, given at
each electron burst, and the stop signal from the coincidence unit. Selecting only coincident pulses, a
significant improvement of the signal-to-noise ratio in the TOF-spectrum was obtained. The time
spectra from the BF$_3$ proportional counters, neutron flux monitors located in the ceiling of the target
hall, was also recorded to verify the stability of the accelerator and their count rate used to normalize
the sample-in and sample-out measurements to the same total neutron fluence.
The transmission experiments at the 26.45 m flight path were performed at three different working
frequencies of GELINA 400 Hz, 800 Hz and 50 Hz.
The measurements at 400 Hz and 800 Hz were carried out with a thick natural Cd sample consisting of
plates with a nominal diameter of 50 mm and with a total mass of (441.6460 ± 0.0001) g. The sample
thickness was 25.818 mm.
To obtain a quality data in the thermal energy region, four additional Cd transmission measurements
were performed at the 26.45 m flight-path at 50 Hz working frequency of GELINA. In the
experiments were used four samples. Three were thin natural cadmium metallic foils with nominal
diameter of 80 mm, their respective nominal thicknesses were 2.03 mm, 30 µm and 50 µm, and their
weights (87.6300 ± 0.0001) g, (1.2814 ± 0.0001) g and (2.0993 ± 0.0001) g respectively. The fourth
sample was a solution sample. This sample consisted of Quartz Cell with an internal path-length of
4.98 mm, filled with (1.1918 ± 0.0001) g Cd (NO$_3$)$_2$ dissolved in H$_2$O. For the sample-out
measurements a similar Quartz Cell with an internal pathlenght of 5.21 mm filled only with H$_2$O was
used.
A number of Cd transmission experiments were performed at the 50 m flight-path number 4 of
GELINA. The experimental setup for the Cd measurements was the same as the one shown in figure
1. The samples were placed at a 25-m distance from the neutron source. A 6 mm thick NE912 Li-glass
scintillator, placed in an aluminium sphere and viewed by a single 125 mm EMI9823-QKB
photomultiplier, placed outside the neutron beam, was used to detect the neutrons. A constant fraction
discriminator was used to determine time of an event, and only signals lying within the (n,α) peak in
the pulse height spectrum were sent to the Time to Digital Converter (TDC).
The Cd transmission measurements at the 50 m flight path were performed at 400 Hz and 50 Hz
working frequency of GELINA.
The 400 Hz transmission experiments were performed with three natCd samples. The first experiment
was carried out with a thin natural Cd disk sample with a nominal diameter of 50.00 mm, a thickness
of 0.125 mm and weight (1.9878 ± 0.0001) g. In the second experiment it was measured a natural Cd
foil sample with a nominal diameter of 100.07mm and a thickness of 2.00 mm. The weight of the
sample was (174.2655 ± 0.0001) g. The last measurement at 400 Hz was carried out with a sample
consisting of two Cd foils one of which was already used in the previous experiment. The other Cd
foil had a nominal diameter of 101.93mm, a thickness of 3.00mm, and weight (272.9500 ± 0.0001) g.
Four more transmission experiments were carried out at the 50 Hz repetition rate of the LINAC. In
three of them the same 25.818 mm Cd sample was used as the one already measured at 400 Hz
and 800 Hz working frequency at the 26.45 m flight path. The last measurement was done with the 0.125
mm Cd sample, which was already measured at 400 Hz.

3. Results and discussion
In the transmission experiments, the neutron time-of-flight (TOF) spectra were measured alternating
the sample in and out of the neutron beam, using a sample changer. To avoid systematic uncertainties
due to possible slow variations of the beam profile or detector efficiency as a function of time, the
alternating sequence of measurements was repeated many times in order to approach an identical
incoming average neutron fluence for the “in” and “out” cycles. The dead-time of the whole detection
system was measured by registering the time-interval between two events (see table 1). The dead-time
correction was well below 1% in the energy region of interest.
The transmission factor T(t_n), as a function of the neutron time-of-flight t_n, was obtained as the ratio of
the time-integrated spectra C_{in}(t_n) and C_{out}(t_n), the sample in and out spectra respectively, which have
been corrected for the incident neutron flux and the dead-time effects.
The background spectra $B_{in}(t_n)$ and $B_{out}(t_n)$ were described as a function of the neutron time-of-flight by the formula:

$$B(t_n) = a_1 + a_2 t_n + a_3 e^{-a_4 t_n} + a_4 e^{-a_5 t_n}.$$  

(2)

The parameters were obtained by fitting them to background points determined by the black resonance technique. The background contributions were approximately 1% (see figure 2).

Table 1. Dead-time data obtained in different experiments.

| Measurement       | Dead time |
|-------------------|-----------|
| 26.45 m flight path |           |
| 400 Hz            | 90 ns     |
| 800 Hz            | 2015 ns   |
| 50 Hz             | 93.5 ns   |
| 50 m flight path  |           |
| 400 Hz            | 2050 ns   |
| 50 Hz             | 2050 ns   |

Figure 2. Dead time corrected cadmium TOF spectra and the shape of the background for sample-in and sample–out measurements of the metallic sample with 50 µm thickness.

The transmission measurements at 50 Hz working frequency of GELINA of the two Cd foil samples - 0.030 mm and 0.050 mm, and the 0.5 cm Cd (NO$_3$)$_2$.4H$_2$O solution sample were performed in order to obtain a quality data in the thermal energy region, from which we could extract the resonance parameters for the first well-known resonance in $^{113}$Cd at 0.178 eV. In the region of the lowest resonance the full width half maximum of the energy resolution of the experimental setup was of the order of 3 meV, and of the Doppler broadening effect was approximately 30 meV. The natural width for the 0.178 eV resonance is approximately 115 meV, this value is almost completely determined by the capture width. As the broadening is much smaller than the natural line width, the capture width can be extracted from the total width. The resonance area will then give the neutron width. Therefore, by using a shape analysis program both the neutron and the radiation width could be determined from a suitable transmission measurements alone. The free gas model with an effective temperature $T_{eff}$ was used to describe the Doppler effect. The effective temperature is a function of the Debye $T_{debye}$ and room temperature $T_{room}$:

$$T_{eff} = \frac{3T_{debye}}{8} \coth \left( \frac{3T_{debye}}{8T_{room}} \right).$$  

(3)
The Debye temperature was taken from the literature as 186 K [6]. The resonance parameters were determined using the program REFIT [7]. This code uses the Reich–Moore approximation of the R-matrix to calculate the nuclear cross sections.

The three sets of experiments were fitted separately and their results compared to each other and to a simultaneous fit to all three data sets (see figure 3). The results are given in table 3. The uncertainties given in table 3 take into account the uncertainties due to the counting statistics and the uncertainty of the sample thickness.

Figure 3. The REFIT fitting results of the transmission data obtained in 0.030 mm and 0.050 mm sample measurements respectively.

The resonance parameters determined in the present measurement have a greater accuracy than the published values (see table 2). The previously published resonance energy and capture width are in reasonable agreement within their quoted uncertainties with the values given in this paper.

Table 2. Comparison of the resonance parameters of the 0.178 eV $^{113}\text{Cd}$ resonance found in the literature.

| Experiment: sample type | Eres (eV)     | $\Gamma_\gamma$ (meV) | $\Gamma_n$ (meV) |
|-------------------------|---------------|-----------------------|------------------|
| R. O. Akyuz et al [8]   | metal 0.181 ± 0.003 | 109 ± 3               | 0.645 ± 0.025    |
| L. J. Rainwater et al [9]| metal 0.176 ± 0.002 | 115 ± 2               | 0.620 ± 0.020    |
| C. N. Brockhouse [10]  | metal 0.180 ± 0.003 | 113 ± 2               | 0.680 ± 0.020    |
| U. Harz et al [11]     | various 0.1783 ± 0.0002 | 113.5 ± 0.5           | 0.650 ± 0.005    |
| F. Widder et al [12]   | powder 0.1776 ± 0.0006 | 114.3 ± 0.6           | 0.618 ± 0.003    |

Table 3. Resonance parameters fitted to our experimental data.

| Sample type | Sample thickness (at/b) | Eres (meV) | $\Gamma_\gamma$ (meV) | $\Gamma_n$ (meV) |
|-------------|-------------------------|------------|-----------------------|------------------|
| metal       | 1.36 × 10^{-4}          | 178.8 ± 0.2 | 113.6 ± 0.3           | 0.642 ± 0.003    |
| metal       | 2.24 × 10^{-4}          | 178.6 ± 0.2 | 113.4 ± 0.2           | 0.638 ± 0.003    |
| solution    | 1.40 × 10^{-4}          | 178.5 ± 0.3 | 114.5 ± 1.0           | 0.643 ± 0.006    |
| weighted average | 178.7 ± 0.1 | 113.5 ± 0.1 | 0.640 ± 0.002          |
| Simultaneous fit results | 178.6 ± 0.1 | 113.4 ± 0.2 | 0.639 ± 0.002          |
| Final results           | 178.7 ± 0.1 | 113.5 ± 0.2 | 0.640 ± 0.004          |

The derived resonance parameters for the 0.178 eV resonance in $^{113}\text{Cd}$ were validated by integral experiments [13, 14]. The impact of the change of the first resonance is quite significant, with changes in the $k_{eff}$ between 0.02 and 0.05, and the agreement between calculations and measurements is clearly improved [1]. The changes are significant but not quite big enough to bring calculations and
experiments in agreement. This might be an indication that higher energy cadmium resonances or the thermal cross sections for other cadmium isotopes should be investigated.

Considering the transmission data obtained with various sample thicknesses as a source of additional information for low-level cross section, we validate a new approach for cross section description in this energy region. The method was proposed by Prof. A. A. Lukyanov [15]. We assume that the application of this method will let us obtain a better description of the cross section in the low energy region \( E \leq 1 \text{ eV} \) and the negative levels. In this energy region it is necessary to take into account a number of effects that otherwise would be negligible for energies above 1 eV. For instance, for most of the heavy nuclei the main energy dependence \( 1/E \) could be changed accounting of the low-level resonances and the effects of the negative level tails. We take into account the energy dependence of the total resonance width. According to this approach in the analysis of the negative levels, the shift factor \( \Delta \) has to be proportional to \( \sqrt{E} \). In the low energy region, it is very important to study the Doppler Effect with thermal corrections as applying the Exact Free Gas Model with Maxwell-Boltzmann distribution of the thermal motion velocities of the atoms of the target \( \bar{u} \):

\[
B(u)du = \frac{1}{(\pi u_T^2)^{3/2}} e^{-u^2/u_T^2} u^2 du d\Omega
\]

(4)

where \( u_T^2 = 2kT / M \), \( M \) is the mass of the sample (target) nucleus, \( k \) is the Boltzmann’s constant, and \( T \) is the effective temperature of the sample. For neutrons with velocities \( \bar{w} \) in the Lab system, reaction cross section is result of averaging the relative motion velocities of the neutrons \( \bar{w} = \bar{v} - \bar{u} \) \( (w^2 = v^2 + u^2 - 2uv\cos\theta) \) having the same distribution as the one in equation (4). The reaction rate \( w\sigma_c(w) \) is presented as:

\[
v\sigma_c(v,u_T) = \int d\bar{u} \sigma_c(w) wB(u)
\]

(5)

We obtain as a result:

\[
v\sigma_c(v,u_T) = \frac{1}{v\sqrt{\pi u_T^2}} \int_0^\infty w\sigma_c(w) w dw \left[ e^{-(v-w)^2/u_T^2} - e^{-(v+w)^2/u_T^2} \right]
\]

(6)

We will have for the corresponding energies \((2E = m_n v^2, 2E' = m_n w^2)\) in the center of mass system:

\[
\sqrt{E} \sigma_c(E,\delta) = \int_0^\infty \sqrt{E'} \sigma_c(E') S(E',E,\delta) dE'
\]

(7)

where

\[
S(E',E,\delta) = \frac{1}{2\delta \sqrt{\pi E}} \left[ e^{-\sqrt{E}-\sqrt{E'}} / \delta^2 - e^{-\sqrt{E}+\sqrt{E'}} / \delta^2 \right]
\]

\[
\delta^2 = kT / A
\]

\[
\int_0^\infty S(E',E,\delta) dE' = 1
\]

This function is normalized to unity, and in the limit of the low energies, where \( \sqrt{E} \sigma_c(E') = \text{const} \) it keeps the reaction rate; the cross section does not depend on the temperature.

The analytical method for determination of the integral shown in the equation (7) depends on the choice of the form of the energy dependence of the evaluated resonance cross section \( \sigma_c(E) \). It could be presented in various functional forms deduced from the reaction theory: Single Level Breit Wigner (SLBW), Multi Level Breit Wigner (MLBW), Adler Scheme, Reich–Moore approximation of the R –
Matrix theory with a set of evaluated relevant resonance parameters. According to Prof. Hwang’s last works [16], all these forms can be combined in one general, universal method of parameterization - Rigorous pole representation of the collision matrix in the momentum domain (as a function of $\sqrt{E}$):

$$\sqrt{E}\sigma_c(E) = \sum_k \operatorname{Re} \left( \frac{(-i)\rho_{kc}}{p_k - \sqrt{E}} \right) = \frac{1}{\sqrt{E}} \sum_k \frac{n_k\alpha_{kc} + \left( m_k - \sqrt{E} \right)\beta_{kc}}{\left( m_k - \sqrt{E} \right)^2 + n_k^2}$$  \hspace{1cm} (8)

where $p_k = m_k - in_k$ and $\rho_{kc} = \alpha_{kc} + i\beta_{kc}$ are energy independent complex constants, which could be determined using evaluated resonance parameters from standard formalisms.

This method of parameterization is very useful for account of the Doppler Effect. In general case, the averaged multi level formulas could be presented as a sum of the same type of integrals for each term presented in equation (8):

As part of our analysis, we determined the nuclear radii for the individual isotopes of Cd. They were defined in the fits from the resonance-potential interference effects in the regions of the larger s-wave resonances. Table 4 contains our values compared to the ones in different evaluated data files.

| Cadmium Isotope | Potential Scattering Radius [fm] |
|-----------------|----------------------------------|
| Cd 110          | 5.790 6.418 6.200               |
| Cd 111          | 5.900 5.797 6.200               |
| Cd 112          | 5.860 5.814 6.200               |
| Cd 113          | 6.300 6.737 6.200               |
| Cd 114          | 6.310 6.310 6.200               |
| Cd 116          | 6.100 6.500 6.200               |

Table 4. Comparison between the nuclear radii of the individual isotopes of cadmium determined from fitting our data with the ones in the evaluated data files.

Our analysis is in progress. Cadmium transmission data obtained in the measurements at GELINA have been fitted and analyzed in the energy region from thermal up to about 3 keV. In figure 4 are presented part of the preliminary results.

Figure 4. Preliminary results of the analysis of the transmission data of 25 mm thick Cd sample obtained in the experiment at 26.45 m flight path.

4. Conclusions
Cd transmission data have been measured between thermal and 3 keV neutron energy.
We have presented the resonance parameters of the lowest lying resonance of $^{113}$Cd as: $E_{\text{res}} = 178.7 \pm 0.1$ meV; $\Gamma_\gamma = 113.5 \pm 0.2$ meV and $\Gamma_\text{n} = 0.640 \pm 0.004$ meV. They are in general agreement with the available in the literature parameters of this resonance within the given uncertainties. For the neutron width, the values from evaluated data files are 1–2% larger than our result. For the resonance energy and $\Gamma_\gamma$ the observed differences are below 1%. Using the parameters recommended in this work gives a better agreement between calculated $k_{\text{eff}}$ and measured $k_{\text{eff}}$ for the integral experiment by Lloyd et al. [17, 18].

We have reported the determined from us values of nuclear radii for the individual isotopes of Cd and the comparison to the ones in the evaluated data files in the ENDF/B – VI.8, ENDF/B – VII, JEFF – 3.1, JENDL - 3.3 libraries.

In this paper, a scheme of a new approach for cross section description in the low energy region is presented.

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