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Low energy transmission measurements of ^{240,242}Pu at GELINA and their impact on the capture width

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Abstract. We present an approach to account for sample inhomogeneities as observed in thin powder samples. To test this model a series of transmission measurements on enriched ^{240}Pu and ^{242}Pu oxide samples was carried out. These measurements were performed at GELINA, the pulsed, white neutron source at the Institute for Reference Materials and Measurements in Geel. For all samples a mixture of PuO_2 and graphite powder was used. To reduce the influence of the models for Doppler broadening on the extracted resonance parameters, measurements were performed with sample temperatures of approximately 12 K, 77 K and 300 K. To describe the variation in the sample thickness, caused by the particle distribution in a thin powder sample, a Monte Carlo and an analytical description were used in the resonance shape analysis. The values and uncertainties of the extracted resonance parameters are discussed, and suggestion for future measurements are given.

1 Introduction

Accurate resonance parameters for all of the plutonium isotopes are required for some of the designs suggested in the Generation IV International Forum. For some of the plutonium isotopes it can be observed that microscopic data have to be adjusted to fit the results of integral measurements. As only a small number of experiments can be found in the EXFOR[1] database, more and precise measurements of the plutonium cross section are therefore needed. To facilitate a reliable extraction of resonance parameters the influence of experimental properties, e.g., Doppler broadening and sample inhomogeneities, has to be calculated with sufficient accuracy. It seemed therefore essential to perform a systematic study of the effects of sample inhomogeneities of thin powder samples, as such samples are commonly used for cross section measurements of the actinides.

The first part of this paper will try to develop a method to describe the variation of sample thickness in a thin powder sample correctly. The second part of the paper is dedicated to experiments performed at GELINA to test these models.

2 Transmission through a thin powder sample

When trying to extract accurate resonance parameters from transmission data, the treatment of the sample properties can be of big importance. To illustrate such effects, calculations for the transmission of the 1.05 eV resonance of ^{240}Pu assuming different sample inhomogeneities were performed. In these calculations we assumed a log-normal distribution with an expectation value of one and a variance of σ. This parameter σ was then varied between 0 and 0.6 with steps of 0.1. The expectation value was then renormalized to agree with the nominal sample thickness, which for this calculations was assumed to be 2×10^{-5} atoms/barn. As can be seen from figure 1 for a variance of 10% the influence on the transmission

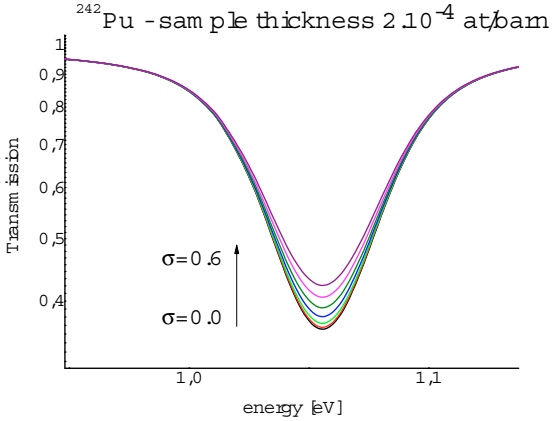

Fig. 1. Effect of different parameters for the variation in the sample thickness on the calculated transmission. The average sample thickness is kept the same for all cases.

is almost negligable. With the here applied parameters the maximum change of the transmission is of the order of 0.2%, which is for most experiments smaller than the systematic uncertainties. But for larger variances the effect is no longer negligible, and a significant reduction of the transmission minimum will be observed. Therefore, in a resonance shape analysis ignoring the sample inhomogeneities will lead to an underestimation of Γ_n and an overestimation of Γ_γ. The induced errors easily can reach 10–30%. Therefore a model has to be developed to estimate the magnitude of the variation for sample thickness of a powder sample.

As a first step a Monte Carlo approach was used to obtain information on the shape and magnitude of the distribution. The basic parameters and principles of this calculation were as follows. At first a mean layer thickness was assumed, i.e., the average number of particles that will be in "line-of-sight" for a neutron that passes through the samples. Knowing the mean layer thickness a mean particle radius can be calculated. As a next step a particle size distribution has to be assumed.

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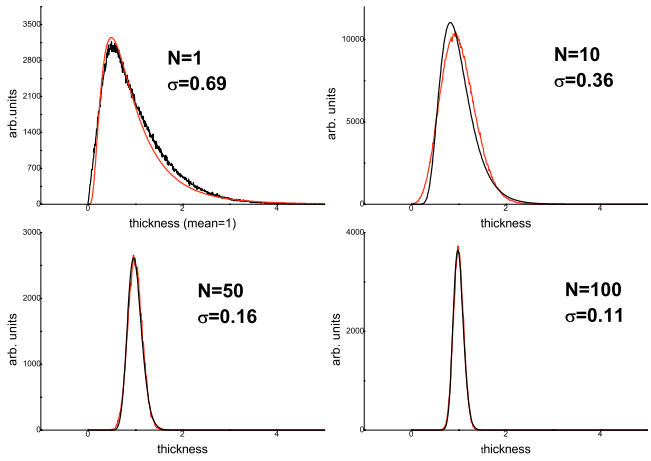

Fig. 2. Variation of sample thickness as calculated by the Monte Carlo model (black lines) and compared to a log-normal distribution (red lines), N is the average layer thickness, σ is the variance of the fitted log-normal distribution.

In the presented calculations a log-normal distribution was chosen. This distribution is defined such, that the expectation value coincides with the mean particle size. The variance of the log-normal distribution can be varied to fit with an observed distribution. For simplicity reasons spherical particles were assumed. The number of particles a neutron encounters within the sample is given by a Poisson-distribution, using the layer thickness as scaling parameter. To derive the thickness distribution the sampling has to be performed over all the possible layer thicknesses and particle sizes. In figure 2 such simulations are shown for different layer thicknesses N. In the same plots log-normal distribution are fitted to the distribution derived by the simulation. In these calculations the log-normal distribution was rescaled to give a mean-value of one, leaving the variance σ as the only free parameter for this distribution.

$$f(x,\sigma) \propto \exp\left\{-\frac{ln(x)+\frac{\sigma^2}{2}}{2\sigma^2}\right\}. \quad (1)$$

Not displayed in this plot is the number of holes in the sample, i.e., the percentage of cases that a neutron does not encounter a particle in the sample, a value given by the Poisson-distribution. This percentage can be significant for samples with a small number of layers, cases that might be encountered quite frequently. For instance in the case of ^{240}Pu the ideal sample thickness for a transmission measurements of an oxide sample is approximately 10 μm. The mean particle size for plutonium oxide, depending on the production method, is between 1 and 10 μm. Therefore the percentage of holes could be as high as 36% in this case.

For thin samples the observed thickness distribution is strongly depending on the underlying particle size distribution. With higher layer number this dependence decreases, and the thickness distribution becomes more symmetric. The variance is then strongly depending on the layer number. Only after a layer number of 50–100, the resulting variation of the sample thickness can be described by a distribution with a variance of approximately 10%. As mentioned before, that number is low enough to ensure that the effect of sample inhomogeneity can be neglected in a resonance shape analysis. It has to be emphasized that diluting the "active powder" in a matrix to facilitate easier sample preparation, e.g., graphite or sulfur, does not improve the inherent problem related to the particle size distribution and layer thickness.

To be able to better estimate the uncertainties related to the inhomogeneity both methods of assessing the thickness variation – the Monte Carlo approach and the log-normal description – were included in the resonance shape analysis code REFIT [2].

3 Experiments

The experiments were performed at the 25 m flight station of flight path #2 at GELINA, the pulsed, white neutron source at IRMM. GELINA is a 150 MeV electron linac, which was operated with a repetition rate of 800 Hz and an electron pulse width of 1ns. This narrow pulse-width is achieved by post-acceleration bunching [3]. The electrons imping on a mercury cooled, rotating U-target, where they produce bremsstrahlung and consequently neutrons. The neutrons are moderated in two 4 cm beryllium canned water containers mounted on top and bottom of the neutron production target. At approximately 10 m from the target is the sample-station located. At the same position neutron overlap-filters, Pb-filters for suppressing the gamma-flash and black-resonance filters (Au, Na, Co) for estimating the background can be inserted in the beam. To allow for low-temperature measurements both the sample and the dummy were put into the cryostat, which is based on the Gifford-McMahon cycle. The temperature was monitored and stabilized with two temperature sensors, which regulated the heating of the sample. For the temperature range between 20 K and 350 K the stability of the system is better than 0.5 K.

At 26.45 m from the neutron production target the neutron detectors is situated. It consists of a 1.25 cm thick ^{6}Li-glass scintillator which is viewed by two photomultiplier tubes. To register a signal it is required that both photomultipliers give a signal within a narrow coincidence window. This procedure allows to reduce the electronic background significantly, compared to a single photomultiplier setup. The so derived timing signals are registered with a Fast-Timing-Digitizer, which has a time-resolution of 0.5 ns. The total dead-time interval was monitored electronically and measured as 1130 ns.

The samples were cycled in and out of the neutron beam approximately every 10 minutes, therefore reducing the measurement uncertainties caused by short term fluctuations of the neutron beam intensity, which is monitored by BF_3 detectors mounted in the ceiling of the target hall. It is estimated that the uncertainty on the normalization is less than 0.5%.

The samples themselves were PuO_2 powder, that was mixed with graphite powder and then canned in copper containers. The dummies consisted of the same amount of graphite powder. The enrichment of the samples was 98.48% for ^{240}Pu and 99.93% for the ^{242}Pu sample. The thicknesses can be given as $8.57 \cdot 10^{-5}$ atoms/barn for ^{240}Pu and $2.51 \cdot 10^{-5}$ atoms/barn for the ^{242}Pu. With these thicknesses the minimum transmission of the ^{242}Pu sample for the 2.67 eV resonance should be around 0.3, whereas the 1.05 eV resonance in ^{240}Pu should be black.

4 Results and discussion

To check then influence of the different models, the resonance shape analysis program REFIT was used with different option, i.e., for the Doppler broadening the free gas model (FGM) and the crystal lattice model (CLM), as implemented in the code DOPUSH [4], were used. Concerning Doppler broadening, the best simultaneous fits, i.e., the data for one isotope for all temperatures were fitted at the same time, were achieved when using the CLM. With the FGM only data at room-temperature could be described satisfactory. At low temperatures this model failed to describe the data. The chi-squared per degrees of freedom became larger, whereas the extracted resonance parameters did not differ significantly for both models of Doppler broadening.

Bigger differences could be observed when comparing the effects of the sample inhomogeneities. For both tested isotopes, the fits using a model for the thickness variation were superior to the calculations using only a constant sample thickness. Of the two applied methods – Monte Carlo and log-normal distribution – the slightly better results were obtained with the analytical log-normal description. The Monte Carlo approach, especially in the case of ^{240}Pu, was very slow in finding the chi-squared minimum. And in most cases it did not quite reach the same level of accuracy as the analytical function. For the ^{242}Pu-sample both models returned a value of approximately 1.1 for the layer thickness, and the parameter σ for the particle size variation can be given as approximately 0.25. In case of ^{240}Pu the variance parameter was similar (the fitting uncertainties for these parameters were of order 10%) and the mean layer thickness obtained was approximately 4 (3.9 for the analytical function and 4.3 for the numerical model). As the ^{240}Pu sample was approximately 4 times thicker than the ^{242}Pu sample one obtains the same average particle size and a particle distribution for both samples. The extracted resonance parameters were in agreement with the values given in the literature [5]. It has to be estimated that for the here presented cases the uncertainty on the resonance parameters is between 5 and 10%, the major part is caused by uncertainties in correcting for the thickness variation.

The measured transmission factors were also fitted using a constant sample thickness. The obtained fits were reasonable, i.e., the values for the chi-squared degrees of freedom were only slightly, but statistically significant, higher. Furthermore the residuals were not distributed randomly, indicating some problems in the description of the shape of the resonance. The extracted values for Γ_γ were approximately 40% larger and those of the Γ_n by almost 40% smaller than the literature values. Therefore one has to emphasize the importance of accounting correctly the thickness variation caused by the powder structure of thin samples.

Most of the transmission measurements that can be found in the EXFOR data bank for ^{242}Pu have been measured with powder samples [6–8]. In general not much information with respect to the sample preparation is given in those papers. Therefore it is hard to judge what the particle size and distribution for those samples are. An evaluation of the uncertainties connected to sample inhomogeneities is absolutely impossible. Measurements on plutonium metal samples, e.g., measurements for ^{241}Pu [9, 10] that have a few percent "impurity"

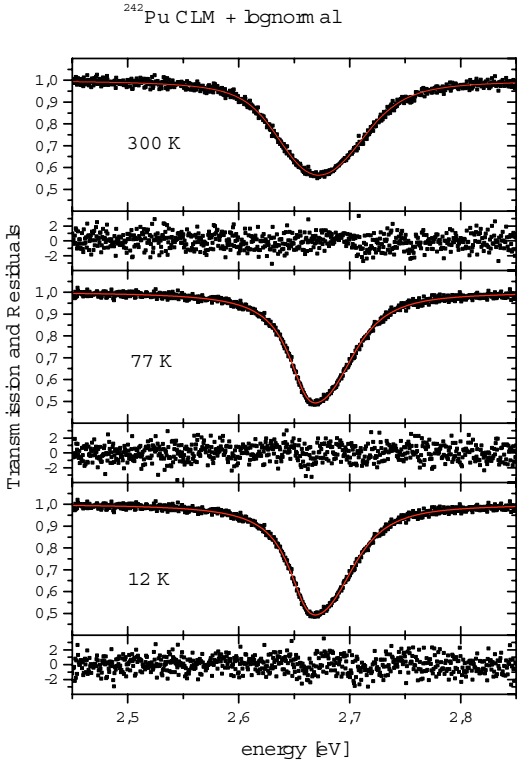

Fig. 3. Fitted transmission data and their residuals for the 2.67 eV resonance in ^{242}Pu at 12 K, 77 K and 300 K, using the crystal lattice model and the log-normal description for the sample inhomogeneity.

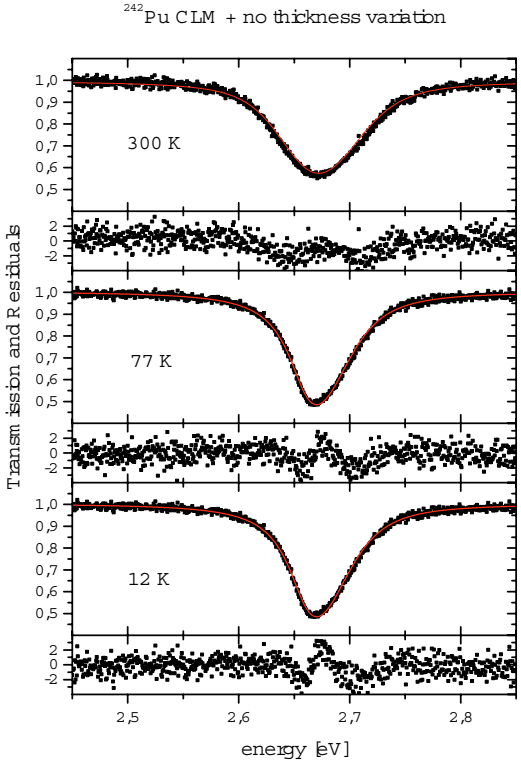

Fig. 4. Fitted transmission data and their residuals for the 2.67 eV resonance in ^{242}Pu at 12 K, 77 K and 300 K, using the crystal lattice model – no correction for sample inhomogeneity was applied.

of ^{242}Pu, can provide a source for obtaining ^{242}Pu resonance parameters. For future measurements using enriched samples, the most promising solution for deriving accurate resonance parameters is the use of solution sample for transmission – and capture – measurements. In collaboration with the EC Joint Research Centre in Karlsruhe a project for measuring the cross section of actinides using sol-gel based samples has been started.

5 Summary

We have presented an model to describe the sample inhomogeneities encountered in powder samples. This model has been applied in the analysis of experiments at GELINA. The importance of these correction could be shown for the cases of ^{240}Pu- and ^{242}Pu-oxides. Considering the importance of the cross section of the actinides, future measurements should avoid using powders as sample materials. The use of solutions, either in the form of very low enriched materials or as sol-gel would be advisable.

References

1. EXFOR, Nuclear reaction data, EXFOR was accessed at http://www.nndc.bnl.gov/, http://www.nea.fr/, or http://www-nds.iaea.org/.
2. M.C. Moxon, REFIT – A least square fitting program for resonance analysis of neutron transmission and capture data, AEA-InTec-0470, 1991.
3. D. Tronc, J.M. Salomé, Böckhoff, Nucl. Instrum. Meth. Phys. Res. **155**, 11 (1978), .
4. D. Naberejnev, C. Mounier, R. Sanchez, Nucl. Sci. Eng. **131**, 222 (1999).
5. S.F. Mughabghab, *Atlas of Neutron Resonances* (Elsevier, Amsterdam, 2006).
6. R.E. Cote, L.M. Bollinger, R.F. Barnes, H. Diamond, Phys. Rev. **114**, 505 (1959).
7. G.F. Auchampaugh, C.D. Bowman, M.S. Coops, S.C. Fultz, Phys. Rev. **146**, 840 (1966).
8. T.E. Young, S.D. Reeder, Nucl. Sci. Eng. **40**, 389 (1970).
9. J.A. Harvey, O.D. Simpson, EXFOR entry 13767.
10. W. Kolar et al., EXFOR entry 20116.