Determination of the thermal neutron capture cross section of trans-uranium actinides

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The reliability of available nuclear data for trans-uranium actinides falls far short of the requirements in many cases. Our goal is to determine the thermal neutron capture cross-sections for these nuclides with high accuracy. Actinides are radioactive, thus the neutron induced gamma radiation can be properly detected only in a high-flux neutron beam. The PGAA facility offers unique opportunities for experiments of this type. So far, Np-237 and Pu-242 nuclides have been measured in the high-flux cold neutron beam. The measurements were performed within the framework of extensive international collaboration, also supported by the BMBF.

Importance of nuclear data

Nuclear data are of great importance in nuclear research and industry. While there are many compilations and databases available worldwide, the quality of these data falls far short of the requirements of the users. The only way to improve the reliability of these values is to redetermine them experimentally.

Figure 1: High-purity germanium detector with Compton-suppression system in an open lead shielding.

Trans-uranium actinides are important components of nuclear waste. The available capture cross-section data are highly unreliable (see table 1). Our main goal is to measure the thermal neutron capture cross-sections of trans-uranium actinides with high accuracy. The PGAA facility at Garching uses the strongest neutron beam in the world, which offers unique opportunities for experiments of this type.

The nuclides with long half-lives (also important components of nuclear waste) can be activated in a neutron beam with a good signal-to-background ratio, as the contribution from decay activity is relatively small. At least 1 to 10 mg per nuclide has to be irradiated depending on the respective capture cross-section. So far, a demonstration experiment has been performed in the medium-flux cold neutron beam for ^{237}Np and ^{242}Pu. Currently, the main emphasis is on optimization of the preparation process for reliable targets.

Determination of cross-sections

The most accurate measurement can be performed with internal standardization, when the comparator nuclide is homogeneously distributed in the sample, since this takes into account the neutron self-shielding inside the sample. This can be achieved using e.g. stoichiometric compounds such as metal chlorides or nitrates. Currently, we are using metallic gold as a very thin foil to cover the sample.

Nuclide	σ_n (barn) ENDF	σ_n (barn) JENDL	σ_n (barn) JEFF	Half-life (years)	Specific activity (Bq/mg)
^{237}Np	180	178.1	195.8	2.144×10^6	2.61×10^4
^{239}Pu	289	271.5	272.4	2.411×10^4	2.30×10^6
^{241}Pu	360	363	363	14.290	3.84×10^9
^{242}Pu	19	19.88	18.8	3.73×10^5	1.47×10^5
^{241}Am	550	684.3	647	432.6	1.27×10^8
^{243}Am	78	79.26	76.7	7.370×10^3	7.39×10^6
^{243}Cm	130	131.4	130.2	29.1	1.87×10^9
^{244}Cm	15	15.24	10.4	18.11	3.00×10^9
^{245}Cm	360	347	359.1	8.5×10^4	6.36×10^5

Table 1: Thermal neutron capture cross-sections of a few trans-uranium actinides from different sources.

First, the detector was calibrated (see figure 1) using standard calibration sources. Secondly, the beam flux was determined using a known-mass gold foil. Then, about 10 mg of the nuclides investigated were sealed in quartz tubes. A similar set of samples was irradiated using a powder mixture of gold and the actinide oxide. All samples were irradiated in the medium- and high-flux beam of the PGAA instrument at the Forschungs-Neutronenquelle Heinz Maier-Leibnitz, FRM II in Garching.

From the detected areas of the characteristic peaks, the activities and partial cross-sections for individual gamma lines were determined after appropriate self-shielding corrections. From the decay measurement of the (n,γ) product nuclides, the total capture cross-sections were also determined.

Understanding PGAA spectra of actinides

The prompt gamma ray spectra of the $^{242}PuO_2$ and $^{237}NpO_2$ samples were evaluated using Hypermet-PC. In the gamma ray spectrum of the $^{242}PuO_2$ sample, more than 600 peaks were identified. Following subtraction of the background peaks, X-rays of Pu, the escape peaks and decay gamma rays, about 80 prompt gamma rays from ^{242}Pu were identified. The ^{241}Am isotope originates from the decay of ^{241}Pu as trace impurities in the $^{242}PuO_2$. Among the 300 peaks identified in the gamma ray spectrum of the $^{237}NpO_2$ sample, X-rays of Np and decay radiation of ^{237}Np, ^{238}Np (activation product) and ^{233}Pa (decay product of ^{237}Np), more than 100 prompt gamma ray peaks of ^{237}Np were found.

The cross-sections were determined relative to that of 198Au. The thermal neutron capture cross-section value of 19.9 ± 4.0 b deduced by the 287.60 keV prompt gamma ray fits well with the existing data sets. For the ^{237}Np thermal neutron capture cross-section a value of 178.3 ± 8.0 b derived from the 1028.54 keV decay gamma ray is in agreement with existing values, with the exception of the very low value of 141.7 ± 5.4 b reported by Katoh et al. [1].

Conclusions and future plans

It can be noted that the uncertainties of the thermal capture cross-sections evaluated in this work are quite high which is basically due to uncertainties in the gamma ray emission probabilities taken from the literature. In the case of ^{237}Np, using the decay of its activation product ^{238}Np, the uncertainty of the mass determination becomes dominant and is about about 5 %. Therefore, the sample preparation procedure is under development and we hope to repeat our experiments in the near future with improved samples and deliver accurate results with uncertainties of < 5 %.

[1] T. Katoh et al., J. Nucl. Sci. Technol., 40, 559 (2003).