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ORIGINAL PAPER

First results of the prompt gamma characterization of ^{237}Np

Christoph Genreith, Matthias Rossbach, Eric Mauerhofer, Támas Belgya, Guido Caspary

Abstract. A sample of ^{237}Np was irradiated in the guided cold neutron beam of the PGAA-NIPS facility at the Budapest Research Reactor to acquire data on its characteristic prompt γ-rays such as energies and relative intensities. A first evaluation of the obtained data showed significant differences from data reported in the evaluated nuclear structure data file (ENSDF) catalog.

Key words: ^{237}Np • actinides • nuclear data • nuclear waste management • prompt gamma activation analysis (PGAA)

C. Genreith✉, M. Rossbach, E. Mauerhofer
Institute of Energy and Climate Research, IEK-6,
Forschungszentrum Jülich GmbH,
D-52425 Jülich, Germany,
Tel.: +49 2461 61 9351, Fax: +49 2461 61 2450,
E-mail: c.genreith@fz-juelich.de

T. Belgya
Institute of Isotopes Hungarian Academy of Sciences,
Department of Nuclear Research,
H-1525 Budapest, P. O. Box 77, Hungary

G. Caspary
Nuclear Services,
Decontamination and Waste Management, N-D,
Forschungszentrum Jülich GmbH,
D-52425 Jülich, Germany

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Introduction

Due to their importance in nuclear waste management, transuranic actinides are of particular interest in research and technology. Radiotoxicity, longevity and decay radiation of these elements require the development of innovative non-destructive methods for the characterization of nuclear waste.

Prompt gamma activation analysis (PGAA) offers an alternative to standard gamma spectroscopic techniques for the identification and quantification of transuranic actinides in complex waste matrices. PGAA is based on prompt gamma emission of excited nuclei after neutron capture. The emitted prompt gamma radiation covers the energy region from a few keV up to several MeV, and can therefore pass through strongly absorbing waste matrices and shielding structures with a high transmission rate.

The PGAA catalog, developed in Budapest [8] and evaluated by the IAEA [5], contains prompt gamma data of elements up to uranium only.

Energies and relative intensities of prompt γ-rays of higher actinides like Np or Pu can be found partially in nuclear data reported in the ENSDF database [1, 3]. In this database the neutron capture data for some nuclides consists of compilations of data from several individual experiments in separate energy regions. Experimental data based on direct measurements over a large energy range from 40 keV to 12 MeV are not yet available.

Besides the energies and relative intensities, neutron capture cross sections have to be known for the quantification of actinide content in nuclear waste.

The available literature on neutron capture cross sections for neptunium shows inconsistencies [7].

Thus, experiments are performed to study the basic neutron capture data of selected actinides. For this purpose, prepared actinide samples are irradiated in a guided cold neutron beam at the Budapest Research Reactor. In addition, the acquired data can be transferred into a database for numerical simulation tools (e.g. MCNP5, Geant4), which can be used to develop analytical instruments for actinide quantification in waste matrices.

In this work, the first evaluation of the PGAA spectra of ^{237}Np is presented.

Description of the experiment

Sample preparation

$^{237}NpO_2$ powder was obtained from Oak Ridge National Laboratory (USA) via TU München (Germany). The preparation was performed in a glove box at Forschungszentrum Jülich. Since the long-lived ^{237}Np (half-life 2.144×10^6 y) was produced about forty years ago, it is in equilibrium with its decay product ^{233}Pa (half-life 26.967 d). About 8.6 mg of the powder was pressed to form a pellet of 3 mm in diameter using a pellet press with a maximum pressure of 1 t. The pellet was mounted between two circular foils of 99.99% pure aluminum with a thickness of $d = 0.25$ mm each held in a screw cap frame made from industrial aluminum. Both the foils had a diameter of 5 cm with a deepening in one of them where the $^{237}NpO_2$ pellet was mounted. In addition, a blank sample without $^{237}NpO_2$ was prepared.

Measurement

The irradiation of the samples was performed at the PGAA facility of the Budapest Research Reactor using a guided cold neutron beam with a thermal equivalent flux of $\Phi_n \approx 1.2 \times 10^8$ cm^{-2}·s^{-1} [10]. The neutron beam was collimated to 2 times 2 cm.

The sample was placed within a neutron shielded sample chamber 23.5 cm in the front of a collimated HPGe/BGO Compton suppression spectrometer, located perpendicular to the beam axis. The collimator is 10 cm long with a beam hole of 2 cm in diameter. The placement was made so that the $^{237}NpO_2$ pellet was located at the intersection of the neutron beam axis and center symmetry axis of the detector. The $^{237}NpO_2$ pellet as well as a part of the aluminum foils were irradiated. The projection of the neutron beam (area inside the black rectangle) on the sample is shown in Fig. 1. A more detailed description of the facility can be found in [11]. The sample was tilted at an angle of $\theta = 30°$ to the beam axis.

First, the $^{237}NpO_2$ sample has been measured for about 15 h without neutron irradiation to assess the background radiation spectrum from the decay of ^{237}Np and ^{233}Pa. Afterwards, the sample has been irradiated with neutrons for approximately 20 h to obtain the combined γ-ray spectrum of prompt γ-emission from the reactions ^{237}Np(n,γ)^{238}Np, ^{27}Al(n,γ)^{28}Al (sample

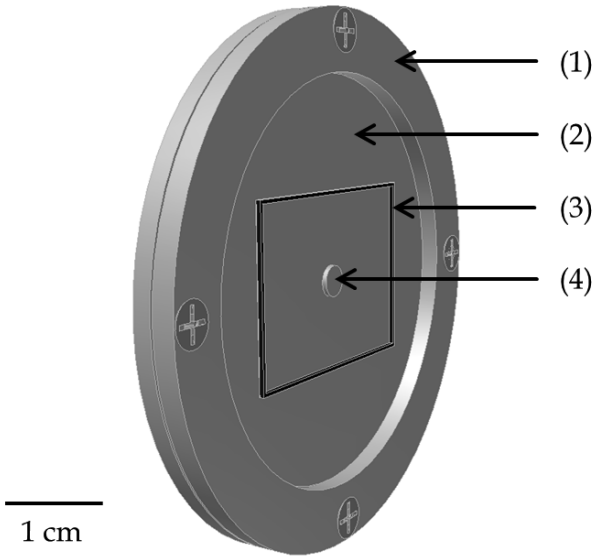

Fig. 1. Drawing of the sample capsule. 1 – screw cap frame; 2 – aluminum foils; 3 – projection of the boundary of the neutron beam; 4 – the deepening in which the $^{237}NpO_2$ pellet is mounted.

housing), ^{14}N(n,γ)^{15}N (air) and the decay γ-rays of ^{238}Np, ^{28}Al, ^{237}Np and ^{233}Pa.

The blank sample was irradiated for about 40 h to assess the background from the irradiation of the aluminum housing and the air in the sample chamber.

The obtained spectra are shown in Fig. 2 where both background spectra have been weighted to the same lifetime as the $^{237}NpO_2$ sample.

The sample spectrum is dominated by the prompt gamma emission from the aluminum housing in the medium and high energy region and by the ^{233}Pa decay in the lower energetic region. The most intense prompt ^{237}Np peak at 182.84 keV can be identified in between the decay γ-rays as well as several ^{237}Np peaks around 5 MeV in between prompt ^{27}Al γ-rays. In Figure 3 a part of the γ-ray spectrum is shown which contains 4 prompt ^{27}Al γ-rays, that are also visible in the lifetime weighted prompt γ-ray spectrum of the blank sample irradiation, and the ^{237}Np γ-ray at 5352.03 ± 0.06 keV with a maximum relative intensity in the high energy region.

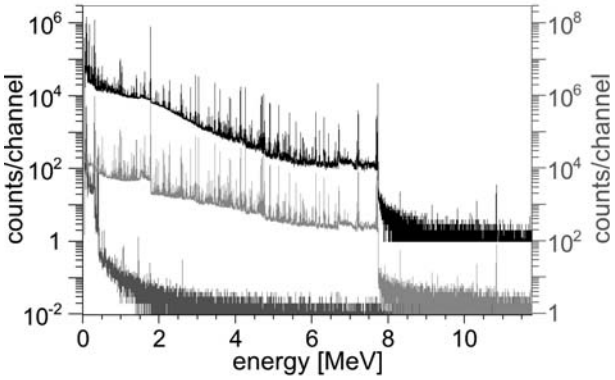

Fig. 2. The upper spectrum (black) corresponds to γ-ray spectrum obtained during the 20 h measurement of the $^{237}NpO_2$ sample. The lower spectrum (dark gray) corresponds to the ^{237}Np/^{233}Pa decay and the middle one (light gray) to the blank sample irradiation. Both background spectra have been shifted by 2 decades, the axis valid for them is on the right hand side (gray).

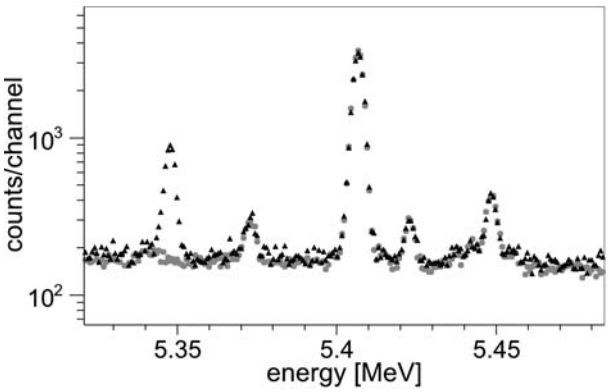

Fig. 3. Part of the γ-ray spectra obtained during irradiation of the $^{237}NpO_2$ sample (black triangles) and of an aluminum housing without sample (light gray dots). The background spectrum is lifetime weighted to match the lifetime of the measurement of the sample containing $^{237}NpO_2$.

Evaluation

The evaluation of peaks in the obtained γ-ray spectrum was performed using Hypermet-PC [12].

The detector efficiency and non-linearity effects in the channel-energy-mapping were taken into account by measurements of calibration sources (^{133}Ba, ^{152}Eu, ^{226}Ra, ^{241}Am, ^{207}Bi, ^{60}Co and prompt gammas of ^{35}Cl(n,γ)^{36}Cl, ^{14}N(n,γ)^{15}N.

The path length x of direct propagation from the sample to the detector was taken into account (Fig. 4) in order to correct the acquired peak areas for the absorption in the aluminum housing. In this way x is calculated to 0.29 ± 0.03 mm.

In addition, self-absorption in the $^{237}NpO_2$ sample was taken into account by assuming that the sample can be approximated as a cuboid [9]. The effective thickness y of the pellet is calculated with the known pellet diameter and the density ρ_{NpO_2} = 11.1 g/cm^3 of $^{237}NpO_2$ and the tilting angle to y = 0.13 ± 0.01 mm.

Because of these arguments, the corrected peak area P_r is calculated by

$$(1)\quad P_r(E_\gamma) = P_m \exp(\mu_{Al}(E_\gamma)x) \times \frac{\mu_{NpO_2}(E_\gamma)y}{1-\exp(-\mu_{NpO_2}(E_\gamma)y)}$$

Here, P_m denotes the peak area obtained from Hypermet-PC, $\mu_{Al}(E_\gamma)$ denotes the linear attenuation coefficient in aluminum at an energy E_γ and $\mu_{NpO2}(E_\gamma)$ respectively the linear attenuation coefficient in neptunium dioxide. The values of $\mu_{Al}(E_\gamma)$ and $\mu_{NpO_2}(E_\gamma)$ were obtained from the NIST XCom database [2].

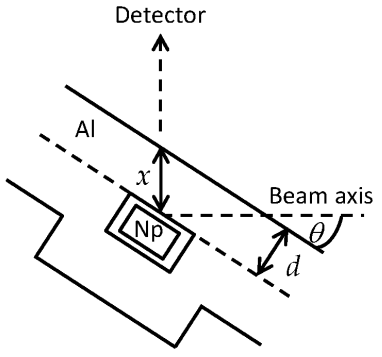

Fig. 4. Sketch of the neptunium sample in the aluminum housing showing the quantities for the peak correction. This sketch is not true to scale.

The peaks in the prompt γ-ray spectrum of ^{237}Np were checked for being a single or double escape peak or a Compton edge.

The relative intensities of the identified peaks are calculated by dividing $P_r(E_\gamma)$ by the area of the most intense ^{237}Np peak at P_r (182.84 keV).

The acquired data were compared with data from the ENSDF database [1, 3]. The ENSDF data consists of two data sets from independent experiments.

One set was measured in the energy region of 2.6 to 5.5 MeV with a pair spectrometer [6]. The other set was measured with a curved-crystal spectrometer in the energy region of 20 to 650 keV [4].

The detection limit in this measurement setup for $^{237}NpO_2$ was estimated to 0.07 mg with three standard deviations of background at the most intense line of ^{237}Np.

Results and discussion

Among 800 detected peaks, 97 prompt gamma lines of ^{237}Np have been identified. These can be divided into two sets of different energy regions. The first energy region below 700 keV contains 62 lines. The remaining 35 lines can be found in the second energy region between 3.9 and 5.5 MeV.

A comparison of the obtained values with the literature values showed a reasonable agreement with respect to the energies of the identified lines, but significant differences with respect to the relative intensities.

In Fig. 5 this is demonstrated by a histogram of the uncertainty weighted residuals between the literature data and the measured data.

In the case of good agreement, the histogram is expected to show a Gaussian shape centered at 0 with more than 68% of the distributed values between –1 and 1 corresponding to a root mean square (RMS) of 1.

The shape of the histogram containing the lower energetic lines is approximately Gaussian with an RMS of 2.41. Even 3 lines deviate with more than 5 standard deviations which means that they are with less than 10^{-5} compatible with the corresponding literature values.

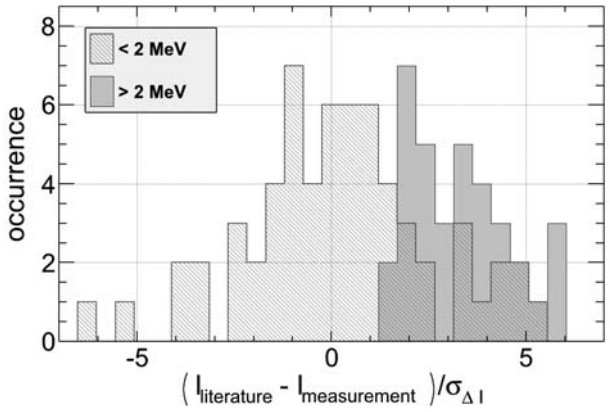

Fig. 5. Histogram of the uncertainty weighted residuals of the measured and literature intensities for different energy regions.

The histogram of the higher energetic lines has a more reasonable RMS of 1.26, but it is systematically shifted as the mean is 3.32.

Possible explanations like a wrong efficiency correction or a bad fit of the reference peak 182.84 keV have been rechecked. Furthermore, the fits of all peaks have carefully been rechecked taking into account the residuals and χ^2's of the fits.

In the ENSDF database two measurement sets were merged which may account for the observed systematic shift in the relative intensities of the higher energetic prompt gamma lines.

These results indicate discrepancies with the literature data and require another measurement with a different sample to be performed to exclude the presence of systematic effects.

Conclusion

A $^{237}NpO_2$ sample enclosed in aluminum has been irradiated with a guided cold neutron beam at the PGAA--NIPS facility. A direct measurement of the sample's prompt γ-ray spectrum has been performed during neutron irradiation with a collimated HPGe detector surrounded by a Compton suppressor BGO. The first evaluation of the spectrum resulted in the identification of 97 prompt γ-ray lines and corresponding relative intensities of ^{237}Np. In the high energy region between 3.9 and 5.5 MeV there are 35 lines, whereas the others are below 700 keV.

Comparison of relative intensities of the obtained data with the ENSDF database has shown significant discrepancies.

For the purpose of identifying ^{237}Np in the shielded or high density nuclear waste samples the higher energetic 34 lines are of particular interest.

Especially, the γ-ray at 5352.03 ± 0.06 keV with a maximum relative intensity in the high energy region seems to be a promising indicator for the presence of ^{237}Np in nuclear waste packages.

Other direct measurements of the ^{237}Np(n,γ)^{238}Np reaction will be performed to investigate the discovered discrepancies.

Further investigations on the prompt γ-ray spectra of ^{237}Np and other actinides will be conducted to determine the partial capture cross sections.

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