Radiochemical Analysis of a Sample of Mixed Uranium–Plutonium Nitride Fuel


*Research Institute of Atomic Reactors, Zapadnoe sh. 9, Dimitrovgrad, Ulyanovsk oblast, 433510 Russia
**Innovation and Technological Center of Proryv (Breakthrough) Project, Varshavskoe sh. 46, Moscow, 115230 Russia

e-mail: *niiar@niiar.ru, **info@proryv2020.ru

Received December 5, 2016

Abstract—Radiochemical analysis of a mixed uranium–plutonium nitride (MUPN) fuel sample irradiated in a BOR-60 reactor was performed. The study was made using the set of procedures developed at the Research Institute of Nuclear Reactors for determining the nuclide composition and gravimetric content of U, Pu, Am, Cm, Nd and other fission products, platinum group metals, and transition metals, including nuclear physical methods, atomic emission spectrum analysis, mass spectrometry for determining the nuclide composition, and isotope dilution mass spectrometry for determining the gravimetric content of nuclides with preliminary radiochemical separation of fractions of elements by ion-exchange, extraction-chromatographic, precipitation, and distillation methods. The MUPN fuel burn-up was determined from the ratio of the number of atoms of the fission product selected as a burn-up monitor to the number of heavy atoms in the dissolved fuel sample (method of fission product accumulation, MFP). The \(^{145}\text{Nd} + {^{146}\text{Nd}}\) sum and \(^{148}\text{Nd}\) were used as burn-up monitors.

Keywords: spent mixed uranium–plutonium nitride fuel, \(\alpha, \gamma\)-ray spectrometry, liquid scintillation spectrometry, mass spectrometry, isotope dilution, ion exchange, extraction chromatography, distillation

DOI: 10.1134/S1066362217040099

Information on the nuclide composition of spent mixed uranium–plutonium nitride nuclear fuel (MUPN SNF) at definite irradiation parameters is necessary for verifying the fuel and transmutation codes being developed, for determining the balance of fissile materials and fission products, the minor actinide production cross sections, etc. These data determine the physical processes occurring in the reactor, nuclear and environmental safety and economic characteristics of the nuclear fuel cycle, and the strategy of its development.

Published data on destructive radiochemical analysis of MUPN SNF, including determination of the gravimetric content of nuclides of uranium, plutonium, minor actinides, fission products, platinum group metals, and transition metals and of the burn-up of heavy atoms, are lacking. Data on the gravimetric content of tritium and \(^{14}\text{C}\) are of particular interest.

Here we report the results of radiochemical analysis of a MUPN fuel sample irradiated in a BOR-60 reactor as a part of 160 E spent fuel assembly in the period from December 5, 2006 to May 4, 2008 with a fluence of \(6.9 \times 10^{22} \text{ n cm}^{-2}\).

EXPERIMENTAL

To study the nuclide composition of MUPN SNF and the gravimetric content of fissile materials and fission products in it, we developed a general scheme of MUPN fuel analysis, shown in Fig. 1. The scheme includes the following procedures: (1) weighing of the fuel sample; (2) dissolution of the fuel sample with a reflux condenser; (3) replacement of the reflux condenser by a descending condenser and partial distillation of the acid, followed by analysis of the condensate for the tritium content; (4) filtration of the initial solution through a double cellulose filter and analysis of the undissolved residue for the content of transition metals (TMs) and platinum group metals (PGMs); (5) sampling of the initial solution and analysis of
the aliquot for the TM and PGM content; (6) preparation of the working solution by hundredfold dilution of an aliquot of the initial solution; (7) estimation of the optimum volume of the working solution aliquot for chromatographic separation, based on results of \(\alpha,\gamma\)-ray spectrometric analyses; (8) sorption separation of nuclides of cesium, uranium, plutonium, and americium–curium–rare earth fraction for mass-spectrometric determination of the isotope composition; (9) repeated sorption separation of the same nuclides in the presence of a combined spike for determination of the gravimetric content.

To determine the fuel weight \(m\), we first weighed a container with a fuel sample \(m_1\). After unloading the sample, we weighed an empty container \(m_2\). After completion of the fuel dissolution, the clad was taken off, washed, dried, and weighed \(m_3\). Additionally, we determined from the results of the atomic emission spectrum analysis of the initial MUPN SNF solution the weight of lead used in the fuel element as heat-transfer sublayer \(m_4\). The value of \(m\) was found from the difference:

\[
m = m_1 - (m_2 + m_3 + m_4).
\]

The weights of the container with the sample and empty container were determined with a UVS-1 kg technical balance with ±0.1 g uncertainty, and the clad weight, with an AX 205 analytical balance (Mettler Toledo) with ±0.25 mg uncertainty.

The fuel sample was dissolved in a 500-mL Erlenmeyer flask equipped with a reflux condenser, which, in turn, was connected with a trap in the form of a 100-mL Erlenmeyer flask containing 50 mL of a 1 M NaOH solution. The dissolution was performed in 250 mL of an 8 M HNO₃ solution.

To develop a procedure for off-gas trapping with the aim of developing a procedure for \(^{14}\)C determination in MUPN SNF, the fuel dissolution was performed stepwise: first for 2.5 h at 50°C and then, after temperature elevation, for 4.5 h at 80°C.

After the dissolution completion, the nitric acid and alkali solutions were analyzed by \(\alpha,\gamma\)-ray spectrometry. The energies of the analytical lines of the radionuclides, their relative intensities, and half-lives for calculating the specific activities of the radionuclides in conversion of the activity concentrations to gravimetric concentrations were taken from handbooks [1–3].

The distribution of \(\alpha\)-emitting radionuclides was monitored using an installation based on an SU-05 spectrometric device and an ATsP-8K-42 amplitude-to-digital converter produced by Aspekt Research and Production Association (Dubna, Russia). The \(\alpha\)-particles were recorded with an ion-implanted silicon detector produced by SNIP-Plyus (Moscow, Russia) with a window area of 450 mm² and energy resolution on the level of 20 keV. The uncertainty of the \(\alpha\)-ray spectrometric determination of the radionuclide content in the samples was 4–6%.

\(\gamma\)-Ray spectrometric measurements were performed with a spectrometer consisting of a DGDK detector, a PU-G-1K2 preamplifier, and an SBS-50M spectrometric printed-circuit board produced by Green Star Technologies (Moscow, Russia). Samples for \(\gamma\)-ray analysis were solutions (1 cm³) placed in standard glass test tubes 12 mm in diameter. The \(\gamma\)-ray spectra were processed using an integrated package of applied programs. The uncertainty of the \(\gamma\)-ray spectrometric determination of the radionuclide content in the samples was 4–10% depending on the quality of the analytical signal.