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Research Article

Capabilities of gamma-spectrometry methods for on-line monitoring of nitride SNF pyrochemical reprocessing*

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Abstract

The paper reports the first test results for detectors of various types and equipment of gamma-spectrometry channels under external radiation originating from pyrochemical reprocessing of spent mixed nitride uranium-plutonium (MNUP) fuel. Testing was carried out on a solid-state detector with a CdZnTe crystal, a scintillation detector with a $LaBr_3$crystal, and an ionization chamber based on compressed xenon. Simulated external gamma-radiation was created by means of a Co-based scattered gamma-radiation source. The paper also describes an experimental facility and a measurement technique, and presents the facility testing results for the above three detectors. The solid-state detector was proved to have the best performance. However, achieving the design characteristics of the gamma-spectrometry channel requires new solutions for protection and collimation of gamma-radiation produced by a real MNUP SNF reprocessing facility. What is meant here is the influence of the detectors' geometry on the configuration of the protective collimator which is proposed to be used in real conditions. Thus, if a Xe-based detector is used, the calculated mass of the protective collimator is 900 kg, while it is possible to use less massive protection for the other detectors. In addition, when manufacturing neutron shielding for detectors based on CdZnTe and $LaBr_3$, it is necessary to consider the neutron radiation factor in MNUP SNF processing. It is possible to surround the collimator with a moderating layer (for example, polyethylene) and create inside it a skin from a thermal neutron absorber (for example, based on cadmium).

Keywords

Pyrochemical process; mixed nitride uranium-plutonium spent nuclear fuel (MUPN SNF); pyroelectrochemical refining; fission products; on-line monitoring of nuclear materials; gamma-spectrometry; experimental facility

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Problem definition

The possibility of on-line monitoring of nuclear material (NM) movements in the oxide SNF pyrochemical regeneration processes using neutron radiometry methods was demonstrated in (Skiba et al. 2012). A similar problem is associated with on-line monitoring of available fission products (FP) in the molten salt during the electrochemical reduction of U and Pu from nitrides. Monitoring the FP content is important for determining and controlling process parameters as well as for assessing the final product quality. During processing, it is necessary to dispose, first of all, of the elements most "harmful" to the neutron-physical characteristics of the fast-neutron reactor core, such as: Zr, Mo, Tc, Ru, Rh, Pd, Ag, I, Cs, Pr, Nd, Pm, Sm, and Eu. To restore the nuclear physical properties of the regenerate, it is necessary that the cleanup factor for these elements should be at least 30. When working with MNUP SNF, it is necessary to take into account that some isotopes of Nb, Sb, Ba, Ce, La as well as steel activation products Cr, Mn, Fe, Ni, Co are radiation hazardous.

The paper analyzes the capabilities of gamma-spectrometry methods for on-line FP monitoring as exemplified by detecting radiation emitted from 1 kg of MNUP SNF (BREST-OD-300) placed in the molten salt the pyrochemical SNF reprocessing chamber.

The method of g-spectrometry is probably the only method for on-line monitoring of FPs which are mainly g-emitters and are present in the electrolyte as compounds. Of these, cesium-137 has the most active line (662 keV). However, in general, the SNF spectrum consists of several hundred lines of gamma-quanta with different energies; for example, europium-152 alone contains 128 lines. The intensity rate of the g-lines depends on the fuel burnout and cooling time to an extent that the carriers of information about the nuclide content in salt or cadmium will be various g-lines (Reilly et al. 1999, Akimov 2014, Bushuev et al. 2006). It is required to conduct an experimental study of the most important emitters that provide information for the process. Until now, a task similar to using the g-spectrometric method for on-line monitoring of SNF reprocessing has not been posed due to the complexity of its implementation. However, if this task is successfully accomplished, it will be possible to speed up the fuel reprocessing processes by reducing the residence time in spent fuel pools. This will certainly increase the profitability of the SNF reprocessing cycle.

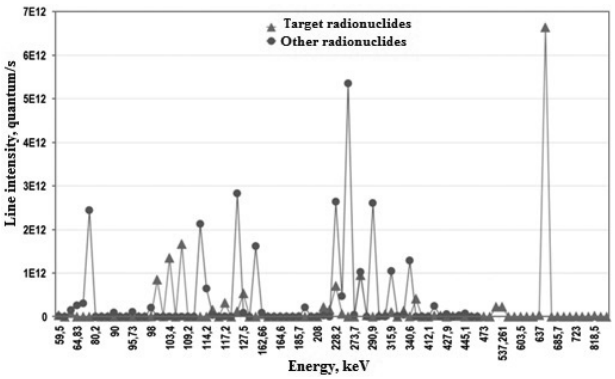

Figure 1. Theoretical -radiation spectrum of the measurement object with divided target and other nuclides.

The task of MNUP SNF g-spectrometry is complicated by a significant total background on which it is necessary to register "useful" lines. Based on the nuclide composition of MNUP SNF, which reached 10% fuel burnout, we calculated the dose rate of γ-radiation in the immediate vicinity (100–200 mm) of the tank with molten salt containing FPs. The calculation was made according to the kerma-constant nuclide values. The approximate dose rate was $P = 5\times10^5$ R/h. Figure 1 shows the theoretical gamma-radiation spectrum obtained using the composition data of the MNUP SNF under study.

Equipment and test methods

The choice of equipment suitable for on-line monitoring of the reprocessing parameters is limited to a large extent by the general technological conditions of the process being carried out. The most critical conditions are the increased temperature (up to 60 °C) in the protective chamber, where the measuring equipment is installed; the difficulty for personnel to access the chamber (therefore, it is impossible to repair or replace the equipment); and the periodic presence of chemically active substances during the chamber clean-up. Data on the conditions applied to the on-line monitoring equipment necessary for solving the set task are presented in (Skiba et al. 2012).

Solid-state detectors (Akimov et al. 1989, Levenets et al. 2018, Rybka et al. 1994, Levenets et al. 2018a), scintillation detectors (Medvedev 1977, Van Loef et al. 2002, Shah et al. 2002) and gas-filled ionization chambers (Ulin et al. 1995, Dmitrenko et al. 1999, Ulin 1999, Bovin et al. 1992, Shustov et al. 2013, Dmitrenko et al. 2000) are used as g-radiation detectors. The work was carried out using a solid-state detector with a CdZnTe crystal, a scintillation detector with a $LaBr_3$ crystal, and an ionization chamber based on compressed xenon. Protection against intense high-energy g-lines and scattered radiation was provided by a collimator consisting of a solid lead "cabin" and a shielding composed of lead bricks.

The solid-state detector based on cadmium telluride is a monoblock containing a detector, a preamplifier, and an analog-to-digital converter (ADC). The quasi-hemispherical detector has a size of 10´10´5 mm. The program for detecting and processing spectra installed on a laptop makes it possible to perform standard operations with spectra (detection for a specified time, energy calibration, "dead" time determination, etc.). Fig. 2 shows photographs of the CdZnTe-based detector combined with the ADC and installed inside the shielding.

The scintillation detection unit is an assembly of a $LaBr_3$ scintillation crystal and a photomultiplier tube (PMT) placed in a light-protective housing (Fig. 3). At the scintillator+photomultiplier assembly output, an ana-

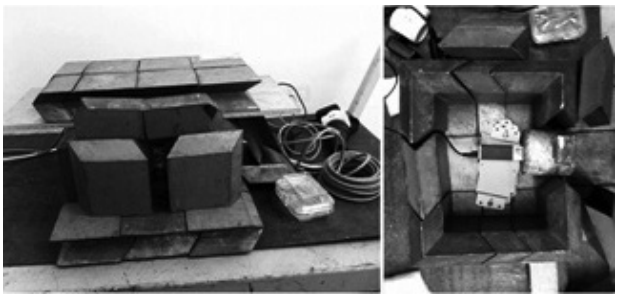

Figure 2. Detector based on CdZnTe inside the lead shielding.

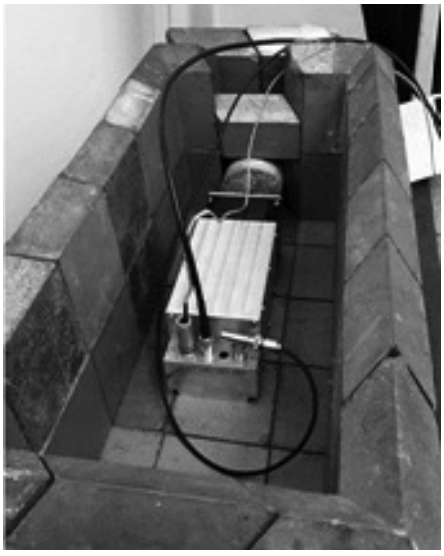

Figure 3. Detector based on a $LaBr_3$ scintillator inside the lead shielding.

log signal is taken to be fed through the connecting wires to the spectrometric board input in a personal computer. The detector dimensions are determined by the size of the assembly, which includes a cylindrical $LaBr_3$ crystal with a diameter of 25 mm and a length of 25 mm and a photomultiplier tube dimensioned 25´100 mm.

Figure 4 shows a photograph of a xenon gamma-detector with a working volume of 0.2 l developed on the basis of a cylindrical ionization chamber with a screening grid. All its internal elements are made of stainless steel and metal-ceramics, the external elements are made of fluoroplastic and aluminum alloys. The detector housing is also the cathode of the ionization chamber and the main supporting element of the structure. A signal from the charge-sensitive amplifier (CSA) output of the g-detector is fed to the digital electronics unit (DEU) via a noise-proof cable and then to a personal computer, where it is processed by special software. The communication line connecting the CSA output to the BCSE is located together with the g-detector inside the lead shielding.

Test results

For a comparative assessment of the detecting properties of the selected detectors, an experiment was conducted to

Figure 4. Detector based on an ionization chamber with compressed xenon inside the lead shielding.

detect the spectra of test γ-radiation sources under conditions of enhanced background. The test sources were of the OSGI type (barium-133, cesium-137). Data on the sources are given in Tab. 1. To create intense background radiation, a GU-200M γ-irradiation facility was used.

A layout of measurement points relative to the radioisotope irradiator is shown in Fig. 5. The photon dose rate values, measured at these points with the help of verified dosimetric devices, are given in Table 2. At each indicated point, a test source γ-radiation spectrum was detected for 300 s with the irradiation facility turned off and on.

The detectors were placed inside the lead shielding, which was composed of standard dovetail bricks with each side 100 mm long and 50 mm thick. A 100 mm long cylindrical lead collimator with an axial hole of 10 mm in diameter was installed in one of the lead shielding walls.

Figures 6–8 show g-radiation spectra measured with the described detectors.

When measuring at Point 1, none of the selected detectors could detect a spectrum due to the extremely high loading of the spectrometric paths. At Point 2, similar difficulties in detecting spectra arose with the gas-filled Xe-based detector and $LaBr_3$ scintillation detector. It was experimentally established that the load limit is 107 cps

Table 1. Characteristics of test gamma radiation sources.

Source	Energy, keV	Quantum yield,%
^{133}Ba	30.973	62.2
	80.998	34.11
	276.398	7.147
	302.853	18.3
	356.017	61.94
^{137}Cs	661.66	85.1

Table 2. Dose loads at various points of the irradiation chamber.

Point number	Photon dose rate, R/h
1	12
2	6.44
3	6
4	1
5	0.48
6	0.22

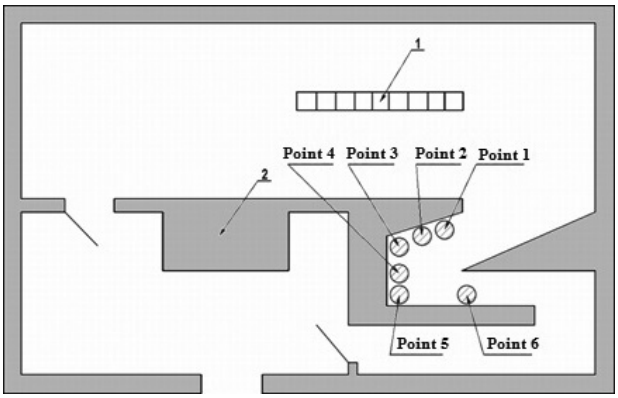

Figure 5. Arrangement of measurement points in the experimental study of different detectors in the field of background gamma radiation: 1. GU-200M irradiator; 2. Operator's position.

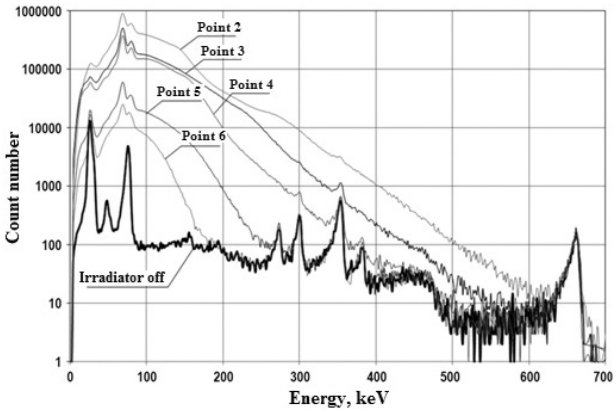

Figure 6. Test source spectra obtained at various points in the maze (CdZnTe detector).

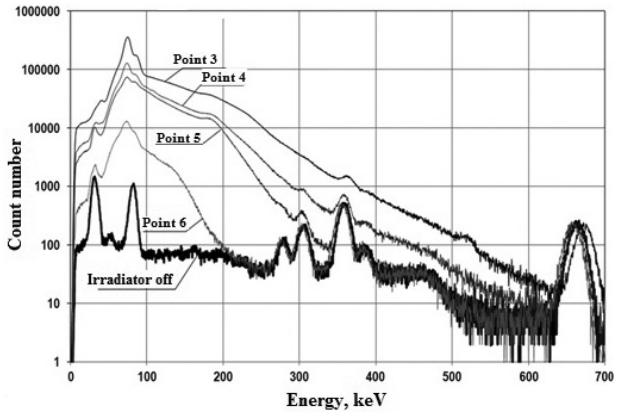

Figure 7. Test source spectra obtained at various points in the maze ($LaBr_3$ detector).

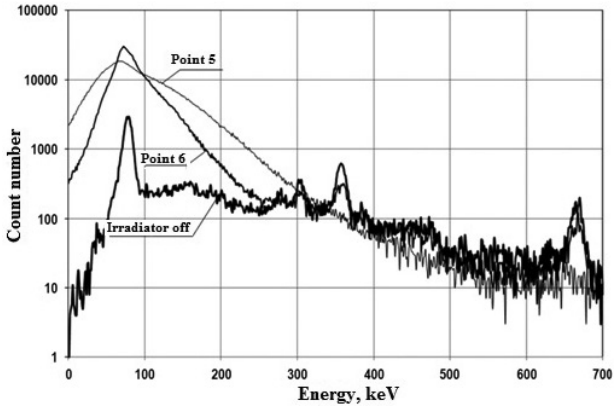

Figure 8. Test source spectra obtained at various points in the maze (compressed xenon ionization chamber detector).

for scintillation detectors and 104 cps for gas-filled detectors, respectively.

Conclusions

Detectors based on CdZnTe or $LaBr_3$ due to their small size and high load limit could be used under the conditions of intense radiation provided that they are protected by a lead shielding with a collimator. The mass of this lead shielding can tentatively reach 300 kg.

Detectors based on an ionization chamber with compressed xenon due to their low g-radiation detection efficiency should have larger dimensions as compared to detectors based on CdZnTe or $LaBr_3$ in order to obtain a comparable energy resolution. This factor, as well as the detector low loading limit, leads to an increase in the lead shielding size, the mass of which (tentatively) can reach 900 kg, which is incompatible with the detector operating conditions.

The main disadvantage of solid-state and scintillation detectors is their sensitivity to neutron radiation. The intensity of interaction between detectors and neutrons of different energies, defined as the integral interaction cross section of detector sensor atoms and neutrons, is approximately 1000:10:1 for detectors based on CdZnTe:$LaBr_3$:Xe, respectively. Therefore, when manufacturing neutron shielding for detectors based on CdZnTe and $LaBr_3$, it is necessary to consider the neutron radiation factor in MNUP SNF processing. Finally, it is possible to surround the collimator with a moderating layer (for example, polyethylene) and create inside it a skin from a thermal neutron absorber (for example, based on cadmium).

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