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Uncertainty of determination of ^{158}Tb in the RBMK nuclear reactor waste

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HIGHLIGHTS

- The activity of ^{158}Tb measurements using gamma-ray spectrometry.
- Uncertainty of determination of ^{158}Tb in the nuclear reactor waste.
- SCALE 6.1 modeling of radionuclide production in the RBMK-1500 reactor.
- Establishment of the origin of ^{158}Tb and other observed gamma-ray emitters in the RBMK-1500 radioactive waste.

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ABSTRACT

The activity of ^{158}Tb was measured in waste samples from the Ignalina NPP Unit I RBMK-1500 reactor using gamma-ray spectrometry. The origin of ^{158}Tb and the other observed gamma-ray emitters has been studied by using SCALE 6.1 modeling and comparing radionuclide ratios in the RBMK-1500 radioactive waste. The results of the calculation of the massic activity of gamma-ray emitters were used for interpretation of the total gamma-ray spectrum and the determination of ^{158}Tb massic activity uncertainty in the waste of RBMK-1500.

1. Introduction

Optimal characterization of radioactive waste from decommissioning of a nuclear power plant (NPP) is based on the use of the method of scaling factors, in which a massic activity of difficult-to-measure nuclides (alpha-, beta-, and long-lived X-ray emitters) in the waste is assessed by measurement of the massic activity of easy-to-measure nuclides (strong gamma-ray emitters). Briefly, the scaling factor is the activity ratio between the massic activity of the difficult to measure radionuclide over the specific activity of the so called key nuclide: $k_i = \frac{A_i}{A_{key}}$, with A_i is the specific activity of the difficult-to-measure radionuclide, A_{key} is the specific activity of the easy-to-measure key radionuclide (Lukauskas et al., 2006). Estimated values for the scaling factors can also be obtained from modeling calculations of the nuclide build-up in the RBMK-1500 reactor. The successful use of simulations saves time and economical resources for radioactive waste characterization (Remeikis et al., 2009). In general, assessment of the massic activity of weak gamma-ray emitters can be done in two ways: (i) by using a scaling factor related to the activity of a strong gamma emitter, if the origin of a weak gamma emitter is related to the one of the strong gamma-ray emitter (e.g. if they are correlated), then the scaling factor can be derived by modeling; (ii) by direct measurements of both gamma-ray emitters with a calibrated gamma-ray spectrometer. In any case, it is important to determine the origin of gamma-ray emitters, which are present in the spectrum.

In this paper we have determined the uncertainty for the determination of ^{158}Tb in the RBMK nuclear reactor waste. The origin of ^{158}Tb and other gamma-ray emitters present in the spectrum have been identified by using modeling. Two possibilities for the presence of ^{158}Tb in the reactor zone were considered: 1) ^{158}Tb originates as a fission/activation product from the nuclear fuel, and 2) ^{158}Tb was generated as an activation product from the absorption rods which contain Gd_2O_3 neutron absorbing material. The build-up and depletion code SCALE 6.1 (Bowman, 2011) was used for the determination of the radionuclides originating in the RBMK-1500 reactor fuel and resulting from the activation of structural material in the reactor during reactor operation and their decay after reactor shutdown. It was assumed, that the isotopes of a same element and elements with the same physical and chemical properties have constant activity ratios in entire reactor zone if there is no specific mixture of the sources. This is also the main assumption for the scaling factors. The calibration of the ^{158}Tb activity and of its associated uncertainty accounting for parameters which may influence the distribution of ^{158}Tb during nuclear plant operation, were

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investigated (e.g. nuclear fuel enrichment, burnable poison enrichment, terbium impurities, fuel burn-up, activation of absorbing material Gd_2O_3).

2. Methodology

2.1. Experimental gamma-ray spectrometry measurements

Terbium-158 has a complex decay scheme: it decays to ^{158}Gd by the EC/β+ branch with the branching ratio of 0.834 and through the β- branch to ^{158}Dy (branching ratio 0.166). The detection of ^{158}Tb is possible with a gamma-ray spectrometer at the following energies when the gamma-ray emission probability is above 5% (data taken from the ENDF/B-VII.1 database): 944.2 keV (emission probability 0.4390), 962.1 keV (0.2027), 79.5 keV (0.1163), 181.9 keV (0.0991), and 780.2 keV (0.0956). The half-life of ^{158}Tb is 180 y; therefore, it is important for decommissioning.

The measurement of low-active ^{158}Tb in the presence of the other activation and fission products such as ^{60}Co, ^{152}Eu, ^{154}Eu, ^{155}Eu, and ^{166m}Ho poses a serious challenge for the non-destructive analysis of the waste from the RBMK-1500 reactors by high resolution gamma-ray spectrometry due to the influence of Compton scattering, mainly from gamma-rays from ^{60}Co and spectral interferences by the full-energy peaks of ^{152}Eu and ^{166m}Ho.

In this work, the activity of ^{158}Tb in the mixture of gamma-ray emitters was measured in the waste samples from the Ignalina NPP Unit I RBMK-1500 reactor using high resolution gamma-ray spectrometry. The coincidence-summing effect was taken into account. Detectors were efficiency calibrated using reference materials traceable to a national standard. The GammaVision software (v. 6.06) was applied for spectra acquisition and analysis. The relative combined standard uncertainty of the ^{158}Tb activity measurements did not exceed 14% (k = 2) in the analyzed samples.

2.2. Modeling of radionuclide activity in the RBMK-1500 radioactive waste

For the calibration of the ^{158}Tb activity and determination of the associated uncertainty, modeling of the activity of the gamma-ray emitters was done. The simplified 4 × 4 core plateau fragment (SCALE 6.1) with 14 fuel assemblies and 2 control rods distributed according to the real RBMK-1500 reactor core geometry was used for the description of the neutron fluence in different parts of the RBMK-1500 reactor (Plukienė et al., 2014). The production of ^{158}Tb was analyzed considering two possible sources: (i) as a fission product from the nuclear fuel and (ii) as an activation product from supplementary absorption rods containing Gd_2O_3 neutron absorbing material. An absorption rod was placed in the middle hole of the fuel assembly and was used with fresh, non-erbium containing, fuel for initial power reduction at the beginning of a reactor cycle. There were about 1300 such absorption rods in total during the nuclear power plant operation. A reactor core fragment with uniformly distributed fuel and a realistic distribution of the control rods of the protection system in the active core was modeled using standard materials composition (Almenas et al., 1998). Terbium-158 activity in the nuclear fuel was calculated using the ORIGEN-ARP libraries for RBMK-1500 fuel (Plukienė et al., 2005) with erbium burnable poison prepared in advance, using the reactor model described above (Barkauskas, 2017). Reactor fuel with different ^{235}U enrichment and Er_2O_3 burnable absorber was considered: 2% ^{235}U enrichment and non-erbium containing, 2.4% enrichment (0.41% Er_2O_3), 2.6% enrichment (0.5% Er_2O_3) and 2.8% enrichment (0.6% Er_2O_3). Natural erbium isotopic composition was used for the calculations (1.7% ^{164}Er, 33.4% ^{166}Er, 22.9% ^{167}Er, 26.9% ^{168}Er, 15.1% ^{170}Er) (Live Chart of Nuclides, 2009–2017). Terbium-158 activity in the other reactor structures (e.g., Gd_2O_3 neutron absorbing material) was evaluated using the transport and depletion module TRITON and the simplified 4 × 4 core plateau fragment. Natural gadolinium isotopic composition was used for calculations (0.2% ^{152}Gd, 2.18% ^{154}Gd, 14.8% ^{155}Gd, 20.47% ^{156}Gd, 15.65% ^{157}Gd, 24.84% ^{158}Gd, 21.86% ^{160}Gd) (Live Chart of Nuclides, 2009–2017).

For the simulation of the different radionuclides and their activity ratios in the nuclear waste, simulation data was compared to experimental data for the following nuclides ($^{166\ m}$Ho, ^{154}Eu, ^{155}Eu). This was done considering different materials: nuclear fuel, absorbing materials (control rods (CR)) checking also the possibility of activation of other construction materials (metal constructions, graphite impurities etc.).

2.3. Samples and gamma-ray measurement equipment

Non-destructive measurements of gamma-ray emitting nuclides in 13 waste samples from the Ignalina NPP Unit I RBMK-1500 reactor were performed using the portable gamma-ray spectroscopic system ISO-CART (ORTEC) with HPGe detectors (INPP Report, 2013) Ten samples were 100 cm^2 wet swipes from the bottom part of control rod tubes surfaces, one sample was a 20 cm^2 wet swipe from the bottom part of fuel channel (FC) tubes surfaces, one sample was a 1.35 kg piece of the bottom part of a control and protection system (CPS) tube and one sample was from the reactor filtration system. The activity of ^{60}Co, ^{137}Cs, ^{154}Eu, ^{155}Eu, $^{166\ m}$Ho, and ^{158}Tb could be determined in almost all analyzed samples (except for the first sample where ^{137}Cs was below the detection limit, and the fifth sample where Eu and Ho isotopes were under detection limits). The counting geometry for the swipe samples was a cylindrical counting container with the swipe placed at the bottom, the sample height was 4 mm, the container was placed on detector end cap. The uncertainty components include an uncertainty component due to geometry corrections (sample height or filling height). The homogeneity of samples has not been tested. The relative uncertainty of the activity measurements was in the range of 10–80% (k = 2) for all radionuclides considered.

3. Results and discussion

As it was indicated above, 13 samples with ^{158}Tb activity above detection limit have been observed. All the samples, except one were wet swipes from the different surfaces of RBMK-1500 reactor constructions. In these samples, ^{60}Co, ^{137}Cs, ^{152}Eu, ^{154}Eu, ^{158}Tb, and ^{166m}Ho have been detected in the gamma-ray spectra. As an example, the gamma-ray spectrum of a volumetric sample (piece of bottom part of CR) is presented in Figs. 1–2. The uncertainty components of this measurement are summarized in Table 1. The uncertainty component associated with the efficiency calibration accounts only for the standard uncertainty of the reference source in case of ^{137}Cs while for the other radionuclides it also accounts for the interpolation as evaluated from

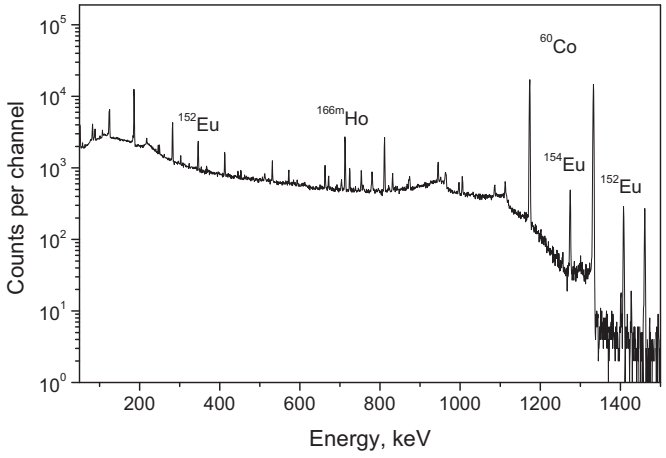

Fig. 1. Gamma-ray spectrum of a radioactive waste sample containing activation and fission products. Acquisition time 1800 s.