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Determination of nuclear material content of items originating from damaged spent fuel assemblies by using collimated gamma-spectrometric scanning

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ABSTRACT

Correlation of the fission product Cs-137, U, and Pu with burn-up has been used to develop a method for determining the U and Pu contents of items originating from 30 spent fuel assemblies (SFAs), which became unusable due to an event in 2003 at Paks Nuclear Power Plant (NPP), Hungary. A mixture of broken fuel rods, pellets, and parts of the cladding were repacked into 72 canisters, whose nuclear material content was to be determined. In this paper, the method has been updated for scanning Cs137 activity and the Cs134/Cs137 ratio along the canisters in 100 s real time intervals by an HPGe detector using a narrow collimator. The effect of mixed burn-up and the irregular fuel geometry was taken into account by using the so-called infinite energy method. The absorption correction factor seems to be constant for all canisters. This is probably due to the overwhelming effect of the thick absorbing water layer in front of canisters lowered down into the service pit, where the measurements were done.

1. Introduction

An event taken place in 2003 at the NPP has made 30 spent fuel assemblies (SFAs) unusable. Fuel material of the damaged assemblies was repacked into 72 canisters. They contained broken fuel rods and pellets from different types of assemblies of different burn-up and irradiation history mixed together, as well as parts of the cladding. The canisters were of three types: type T-28 filled up by one or two relatively intact parts of the SFAs, type T-29 contained mixture of spent fuel pieces of various burn-up distributed in an irregular geometry, and one more type for the non-nuclear construction elements (assembly heads and tails). Their dimensions were similar to those of SFAs, so the loaded canisters could be placed into the lattice in the spent fuel pond. During the recovery process of the damaged fuel, weight measurements had been performed for each loaded canister. Hence, an upper bound for the mass of nuclear material could be estimated for each canister based on total mass of material loaded into them.

Passive non-destructive methods were developed for determining total U, U-235, and total Pu content of each canister for inventory taking of nuclear material.

As described in Ref (Zsigrai et al., 2013), the final method was based on scanning Cs137 activity and the Cs134/Cs137 activity ratio along the canisters in 100 s real time intervals by an HPGe detector using a narrow collimator; and the content of the total U was calculated by a formula with a constant absorption correction factor for all canisters. This ensures good precision of the method.

Because of the high activities of fission products, the U and Pu contents could not be measured directly, they had to be derived from measurable fission and activation products such as Cs and Cm isotopes by gamma and neutron measurements, respectively. As reported in the literature (Tiitta and Hautamäki, 2001; Tiitta et al., 2002; Tiitta et al., 2001; Lebrun et al, 2001), ordinary SFAs of different burn-up were assayed in fixed geometry by measuring Cm neutrons using a fission chamber as well as the Cs-137 content and the Cs-134/Cs-137 ratio by a medium resolution CdZnTe (CZT) detector with a conic collimator.

We considered a principle of measuring the Cs content and burn-up (BU), calibrated by the Cs-134/Cs-137 ratio. Then, U and Pu contents can be derived from the correlation of Cs-137, U and Pu contents with BU. (It is to be noted that the measurements were carried out in 2006–2008, when Cs-134 gamma-ray was still measurable.)

Whereas considering our approach to canister measurements, the irregular geometry of the fuel was concerned for affecting the accuracy of results when measuring the 662 keV gamma ray of Cs-137. Supposing the neutron detection efficiency to be weakly dependent on

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inhomogeneity of the items, the Cs-137 content was estimated upon measuring neutrons emitted from Cm isotopes. Moreover, measuring the Cs-137 content by Cm neutrons may have another advantage, namely decreasing the effect of the gaps between nuclear material containing parts of the canister in a fixed measurement geometry. Thus, neutron counting and measuring the Cs ratio were considered for determining the U and Pu contents. This concept called combined gamma-neutron (CB) method was attempted to develop and test the measurement of 14 reference assemblies in a fixed geometry. Later, use of an HPGe detector with a narrow collimator was considered for improving the accuracy of the Cs134/Cs137 ratio measurement. Our experimental setup consisted of an HPGe, a CdZnTe (CZT) detector, and 2 fission chambers (Zsigrai et al., 2007).

The use of the HPGe detector with a narrow collimator for measuring Cs137 content was not attempted earlier in the fixed geometry because of huge gaps in the geometry of the fuel content in the canister. However, we performed gamma spectrometric scanning of the canisters. Instead of a fixed measurement geometry, canisters were scanned along their lengths in front of a collimator built into the concrete wall of the service pit of the reactor block. Spectra were acquired in every 100 s real time intervals. Due to continuous scanning, the drawback of gaps diminished.

During data analysis, an important observation emerged: the absorption correction factor when measuring the Cs137 content by HPGe seemed to be constant for all items of irregular geometry. Hence, the pure gamma method was estimated and guessed to be better than the CB-method, so it was accepted finally. Tens of thousands spectra were analysed. The results were reported in Ref. (Zsigrai et al., 2013). Due to the constancy of the absorption correction factor for measuring the Cs137 content by HPGe, the accuracy of the pure gamma-method proved to be much better than that of the CB-method.

In this paper, the process to establish the final method by using a simple formula and the prediction of the existence of a constant absorption correction factor for all the 72 canisters and 4 reference assemblies was approved by using an infinite energy method for Cs134 gamma rays. The method was updated by using graphical representation first for a smallest segment, then for the canisters. The effect of mixed burn-up and the irregular geometry is discussed in detail including the infinite energy method. A roughly constant absorption can be explained by the thick water environment of the canisters in the service pit, suppressing the role of other absorbers.

The scheme of the pure gamma spectrometric method is shown in Fig. 1. The distance of a canister or assembly from the wall of the pit was uniformly ~50 cm throughout the experiments.

2. Updating NDA of individual canisters by scanning

2.1. Principle of the method for a smallest segment

A smallest segment was a piece of irradiated fuel material from the damaged SFAs. Canisters contain lots of smallest segments from different damaged SFAs. In principle, all parameters of the smallest segment were the same as those of the original SFA, whereas canister parameters are of a mean average. This means that BU of a smallest segment is a single value, whereas that of canisters is mixed.

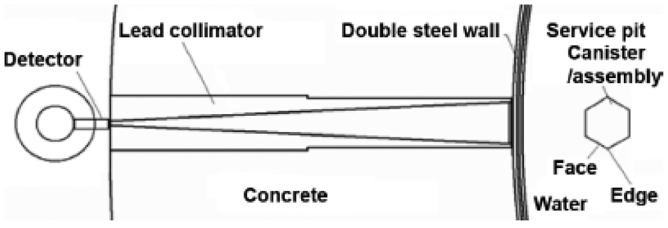

Fig. 1. Scheme of the experimental setup of the pure gamma method.

For establishing a non-destructive gamma spectrometric method to comply with the requests of safeguards authorities, the Cs content and BU calibrated by the Cs134/137 ratio are to be estimated. Then we show how the U and Pu content of a smallest segment can be derived from the Cs content and BU measurements.

Depletion calculation codes for VVER-440 assemblies at the NPP (Basic Technical Character, 2010) give a set of data on isotopic contents in each module of each SFA. An assembly was divided into 20 modules. The module is a standard mass unit, M_m, here it is 6 kg spent fuel, corresponding to its initial total U content.

The method is based on correlations of U, U-235, Pu and some fission products with BU to be determined by gamma measurements. Four correlations were considered, normalizing the three latter to the module mass:

- The Cs137 content vs BU, $M_{137}(BU)$, g/module
- Percentage of total U mass M_U/M_m vs BU, $\rho U(BU),\%$
- Percentage of U-235 M_{235}/M_m vs BU, $\rho 235(BU)$, %
- Percentage of Pu, $M_{Putotal}/M_m$ vs BU, $\rho Pu(BU),\%$.

These correlations obtained by depletion calculation codes for VVER-440 assemblies at the NPP (Basic Technical Character, 2010) are plotted in Fig. 2.

Under spent fuel mass m_{fuel} in a smallest segment as well as in canisters we mean the sum of the masses of the remaining uranium, $m(U_{total})$, produced plutonium, fission products, and minor actinides. Apart from the mass defect, this mass is the same as the initial total mass uranium was in fresh fuel.

We denote the content of Cs137, total U, U235, and total Pu in a smallest segment by $m(Cs137)$, $m(U_{total})$, $m(U_{235})$ and $m(Pu_{total})$, respectively. BU value is a valid parameter for both a smallest segment and a module. For a given BU, the mass ratio of all variables in a smallest segment to that in a module equals uniformly the same value, namely m_{fuel}/M_m. (That is, $m(Cs137)/M_{137} = m(U_{total})/M_U = m(U_{235})/M_{U235} = m(Pu_{total})/M_{Putotal} = m_{fuel}/M_m$.)

For a small irradiated sample the U and Pu contents can be derived as follows:

- From the value of BU, the Cs-137 content $M_{137}(BU)$ in a module is to be derived by using Fig. 2.a, i.e. by formula $M_{137}(BU) = k_1 BU$. Here k_1 is a constant from the linear correlation between Cs-137 content in a module (0.21 g.tU/GWd) and BU.
- From the measured Cs-137 count rate C(662), the Cs-137 content, then the spent fuel mass, m_{fuel} in a smallest segment are derived as

$$m(Cs137) = \frac{C(662)}{\varepsilon . Br}\frac{1}{F}\frac{1}{a(Cs137)},$$
$$m_{fuel} = \frac{m(Cs137)}{k_1 BU}M_m \quad (1)$$

where ε is the detector efficiency, $Br = 0.85$ is the branching ratio of the 662 keV gamma line, F is the absorption factor, and $a(Cs137)$ is the specific activity (3.22E12 Bq/g).

After all,

$$m_{fuel} = K\frac{C(662)}{BU}\frac{1}{F} \quad (2)$$

where K is another constant ($K = M_m/a(Cs137)k_1\varepsilon.Br$).

- Calculating the contents of U-235 and Pu in a smallest segment by using the data in Fig. 2.b and c, respectively.
- Calculating the content of total U, $m(U_{total})$, U-235, $m(U_{235})$, and of Pu, $m(Pu_{total})$, in a smallest segment by multiplying the derived m_{fuel} with ratios plotted in Fig. 2.b, c, and d, i. e. $\rho U(BU)$, $\rho U_{235}(BU)$ and $\rho Pu(BU)$, respectively.

As described in Ref. (Zsigrai et al., 2007), burn-up (BU) was