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Development of neutron spectrum analysis method to assess the content of fissile isotopes in SFA

A.V. Mitskevich

Obninsk Institute for Nuclear Power Engineering, National Nuclear Research University «MEPhI». 1 Studgorodok, Obninsk, Kaluga reg, 249040 Russia

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Abstract

The paper presents the integrated neutron spectrum analysis as a potential method for estimating the contents of fissile isotopes in SFAs. Two method implementation variants are described: (1) measurement of SFA average transmission and (2) measurement of sample average transmission in the spectrum that has passed a SFA. The authors describe the dependences of SFA average transmission on its content of the required isotope obtained by means of two types of detectors: helium counter tube and fission chamber. Also, the authors propose a method to estimate SFA burn-up by means of the integrated NSA. In addition, SFA residence time influence on transmission is estimated.

Keywords: Spent fuel; Neutron spectrum analysis; ^{235}U and ^{239}Pu content assessment; Helium counter tube; Fission chamber; transmission; Fuel assembly; Residence time.

Introduction

According to the IAEA figures of 2012, the total aggregate amount of spent fuel discharged from reactors approximated 360,500 t, of which about 109,800 t have been reprocessed and about 250,700 t placed into reactor spent fuel storage pools or outside reactor storage facilities [1]. The amount of spent fuel discharged for 2012 was approximately 10,000 t while the capacities of fuel reprocessing plants in the world totals only about 5000 t/year. It is obvious that radiochemical reprocessing of irradiated uranium falls far short of its accumulation. Moreover, in many cases for huge amounts of stored spent uranium (and accumulated plutonium) there are only computational estimates of its content and isotopic composition, while the existing NDT techniques do not provide the desired accuracy. In this context there is a need to develop new analysis methods of spent fuel and to determine fission material stores. These methods should be able to conduct a full analysis while maintaining the integrity of commercial reactor fuel assemblies in order to provide data for grouping stored SFAs depending on their nuclear material content as well as spent fuel to be reprocessed now or in the future.

The commonly used gamma-techniques based on intrinsic ^{235}U and ^{239}Pu gamma activity measurements as applied to SFAs are inefficient because of fission-fragment activity dominance. The techniques based on correlation of different burn-up monitors (e.g., ^{137}Cs and ^{134}Cs) provide only a rough idea of fissile isotope content, which in the event of substitution does not ensure the detection of loss. The techniques based on measuring SFAs intrinsic neutron irradiation are weak, because it is practically impossible to separate different sources of their origin. The most efficient method seems to be an active neutron technique, i.e. neutron spectrum analysis (NSA).

Neutron spectrum analysis

The NSA technique is based on the presence of resonances in a reaction of neutron-substance interaction. The intensity of these resonances is unique, i.e. it characterizes each isotope definitely. The NSA is aimed at measuring the number of isotope nuclei rather than its activity; hence it is applicable

E-mail address: korovinyu@mail.ru.

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to any (not only radioactive) isotopes. The proposed active technique showed good results in controlling NM content in thin samples [2–7]. Meanwhile, of the utmost interest is the development of the NSA for controlling the content and isotopic composition of uranium and plutonium in SFAs.

The NSA can be implemented in two approaches: (1) detailed NSA (Neutron Resonance Transmission Analysis) and (2) integrated NSA (Resonance Self-Indication). The detailed NSA became prevailing. The main experimental results here were obtained in the 70–90s. The most important work was done by Shark's team in the National Bureau of Standards (USA) [3]. They examined two cuts from an LWR spent nuclear fuel element with burn-up ~25,000 MW·d/t U about 2.5 cm in length and 1 cm in diameter related to the nuclear fuel element center and its rim. Then the measured transmission spectra were processed with the non-linear least-square method with the involvement of evaluated files from the ENDF-5 library. At the same time, they determined the absolute percentage of 11 actinides (4 uranium isotopes, 5 plutonium isotopes, and 2 americium isotopes) as well as 5 fission products within the accuracy of 0.4–20 percent depending on the line strength.

Currently, the work is underway to adapt the detailed NSA to estimate ^{239}Pu content in SFAs [8–10]. The research is focused on calculations and numerical simulation.

A detailed time-of-flight NSA variant implements the fullest isotopic analysis of a test sample. It is, however, not easy to use and inefficient. At the same time, very often it is not required to know the contents of many isotopes, but operational efficiency should be high. This is the main problem of non-destructive nuclear material testing in SFAs. Here the contents of only fissile isotopes ^{235}U or ^{239}Pu are of interest and hence the informative value of the detailed NSA becomes excessive. The integrated NSA, on the contrary, provides information on the content of one particular isotope [11–14], in addition, its implementation is easier and operational efficiency is higher. Using the integrated NSA, the content a desired isotope is determined by the attenuation of neutron flux that has passed the test object:

$$T(n) = \frac{\int^f (E)\varepsilon(E)e^{-n\sigma^{tot}}dE}{\int^f (E)\varepsilon(E)dE},$$

where $T(n)$ is the average transmission, $f(E)$ is the neutron spectrum, n is the isotope content (n/barn), $\sigma^{tot}(E)$ is the isotope full cross-section (barn), $\varepsilon(E)$ is the detector efficiency.

The simplest integrated NSA variant is measurement of average full transmission of the test object, where a helium counter tube is used as a detector. Its efficiency smoothly varies depending on energy; hence it will properly register neutrons of all energies in the resonance region. This fact greatly reduces the estimation accuracy of the required isotope content in multi-isotope compositions. The estimation accuracy can be improved by creating a selective-sensitive detector based on a helium counter tube. However, for this it will be necessary to involve time-of-flight spectrometry. The idea is to register only those neutrons that fall within the resonance energies. This can be achieved by allocating time slots covering the required isotope resonance energies.

Another variant of a selective-sensitive detector is a fission chamber. A thin layer of fissile isotope (^{235}U or ^{239}Pu) used in it is highly sensitive to neutrons which energy coincides with the energy of fission cross-section resonances of this isotope. Thus, the chamber registers not the whole neutron spectrum but only sections falling within the fission cross-section resonances of the required isotope. Neutrons of other energies available in the neutron flux are hardly detected by the chamber. As a result, the fission chamber energy selectivity makes it possible to jointly use the required isotope lines without measuring the detailed structure of the spectrum that has passed a SFA, which also significantly accelerates statistics gathering. A selective-sensitive detector can be created based on a helium count tube.

The integrated NSA can be implemented in two variants: (1) measuring the average transmission of the object under investigation and (2) measuring the average transmission of a sample in the spectrum that has passed the object under investigation.

Weak points of this technique include strong dependence of the average transmission on the shape of neutron spectrum (hence careful sample calibration is required). Also it is possible to involve overlapping of full cross-section resonances of foreign isotopes available in the sample under investigation with cross-section resonances of the required one. This problem can be solved provided that the content of foreign isotopes is known.

One may choose another way, i.e. to allocate time slots that exclude overlapped resonances. This approach, however, will significantly reduce the operative efficiency.

Simulation and calculations

A mathematical model was created that makes it possible to calculate average transmission of objects with different isotopic content and composition as well as transmission of a sample in the spectrum that has passed the test object.

This mathematical model sets up the isotopic composition of 11 actinides (^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{247}Np) and 5 fission products (^{99}Tc, ^{131}Xe, ^{133}Cs, ^{145}Nd, ^{152}Sm), geometry of the required object, neutron spectrum shape (degradation, cadmium coating) and detector type (fission chamber or helium counter tube). The data on isotope cross-sections were taken from the ENDF5 library.

Range of applicability

In the case of large contents of the desired isotope in the object under investigation the resonances become self-shielded and only their tails are active. The problem was to determine the content at which the average transmission will be described by the exponential relationship. The calculations showed that the maximum value of metallic ^{235}U content at which the method works, is equal to ~0.08 n/barn, for ^{239}Pu