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Determination of nuclear fuel burn-up axial profile by neutron emission measurement

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ABSTRACT

Burning-up of nuclear fuel is usually not a space-isotropic phenomenon. It depends on both the neutron flux density and energy spectrum distribution during fuel operation in a nuclear reactor. This paper presents the method of measurement of burn-up spatial distribution of spent nuclear fuel element. The method is based on recording of the neutron emission from investigated fuel element. Based on performed analyses and calculations, a suitable measuring setup has been designed and constructed. The subjects of investigation were fuel elements used in the MARIA research reactor, operated by National Centre for Nuclear Research in Świerk, Poland. The results of measurements made over a period of several years by means of the described method are presented in the paper.

1. Introduction

Burning-up of nuclear fuel consists in decreasing of ^{235}U content as an effect of neutron-induced reactions in the reactor core. The burn-up quantity is often defined as an amount of heat generated during fission reactions in a mass unit of fuel or in a whole fuel element. Besides nuclear fission reactions, the neutron radiative captures occurs in nuclear fuel during its operation as well. Both fission and radiative capture reactions lead to many nuclides, usually radioactive. Therefore, the burn-up value is directly connected with the activity of nuclides accumulated in the fuel. This has significant impact on the heat generation in spent fuel elements and accordingly their storage security. The spatial distribution of ^{235}U concentration in spent fuel elements need to be determined due to storage criticality safety as well.

Various methods are used to determine burn-up of spent nuclear fuel. In principle, that can be done by using calculation schemes, taking into account fuel inventory and its operation conditions. Among experimental methods, beside destructive chemical treatment, a number of non-destructive assay techniques are used. They are based on measurement of Cherenkov radiation intensity, total gamma-ray activity, single fission product gamma-ray activity, fission products activity ratios [1,2], fission neutron emission (spontaneous and induced) [3–5]. Most of these methods are used in safeguard systems [6] or fuel reprocessing [7].

2. Principles of the method

In the described method the measurement of burn-up distribution consists in recording of neutron emission from separate sections of a fuel element. The neutron emission is proportional to the burn-up value of a particular section of spent fuel element.

The subject of measurements were MR-6/80 fuel elements which had been used in the MARIA research reactor until 1999.

The MARIA reactor is a multipurpose high-flux research reactor with nominal thermal power of 30 MW. It started operation in 1974. From 1985 to 1992 the reactor was out of operation due to major alteration works. Until 1999 the 80% enriched fuel was used. Then it has been replaced with 36% enriched fuel. From 2014 only LEU fuel (enrichment $<20\%$) is used. Since 2005 the MARIA reactor is under continuous operation – over 4500 h per year in 100–200 h duty cycles. The fuel elements are shuffled periodically – during each operation break, according to the needed reactor core configuration, due to criticality conditions and irradiation demands.

The MARIA reactor is equipped with two independent primary cooling systems: the fuel channels cooling system and the reactor pool cooling system. The fuel channels are pressurized up to 1.7 MPa, whereas the reactor pool is open. The inlet and outlet temperatures and flow rates are measured at each fuel cooling loop independently. The power generated separately in each fuel element is, therefore, determined accurately in real time. The total thermal energy output from each fuel element is recorded. This means that the total burn-up of each fuel element is determined by the operator. Based on other analyses the uncertainty of this

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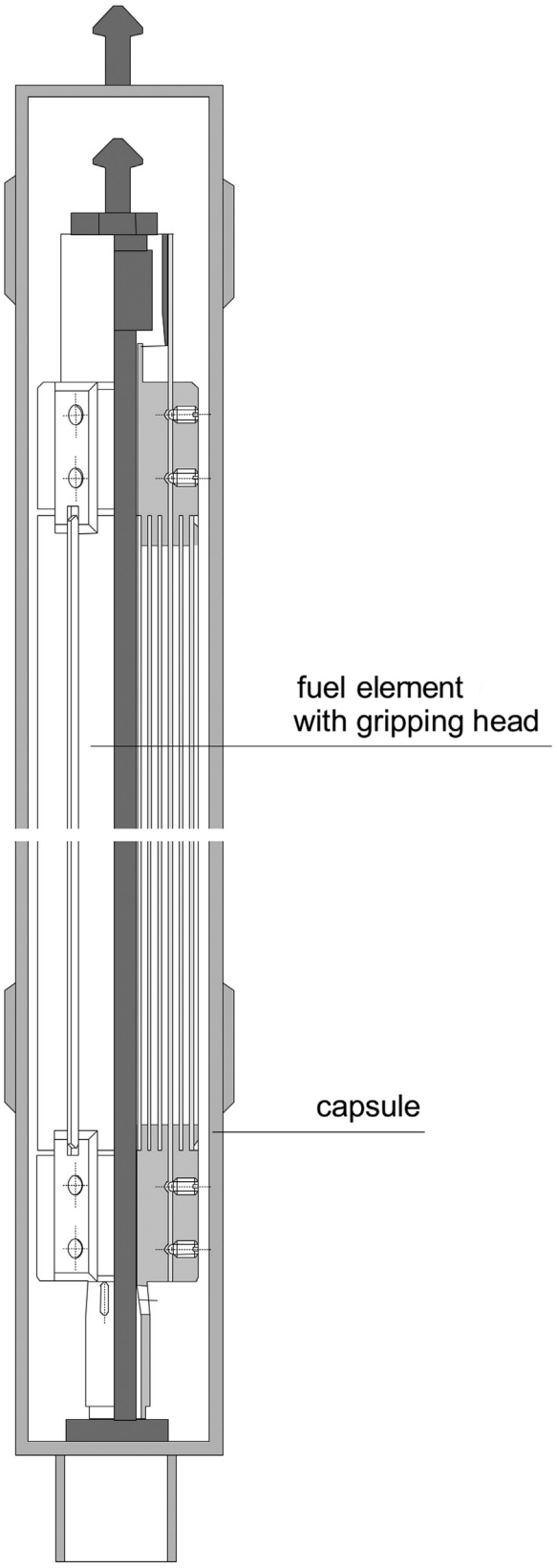

Fig. 1. Steel capsule with MR-6/80 fuel element [9].

determination can be estimate at ca. 5%. That enables the correctness of the results obtained by means of the described measuring method to be verified.

Nuclear fuel in an MR-6/80 element consists of aluminium with a dispersion of uranium dioxide or alloy of uranium and aluminium [8]. The total initial average uranium mass in the MR-6/80 element is 345 ± 22 g and is determined for most of elements with 0.1 g accuracy. Uranium is enriched to 80% of ^{235}U isotope. The active part of a fuel element consists of six concentric tubes, 1000 mm high, 2 mm in thickness and 21, 30, 39, 48, 57, 66 mm in outer diameter respectively. The tubes are built of three layers, each of about 0.7 mm thick. The middle layer is nuclear fuel, the inner and outer ones are aluminium cladding.

Spent MR-6/80 fuel elements after a few years of cooling, had been enclosed under a helium atmosphere in stainless steel capsules of 85 mm in outer diameter and 2 mm thick [9] (cf. Fig. 1). They had been stored in the MARIA reactor storage pool. The measurements of these elements were performed from 2004 to 2006, before shipment of the spent fuel elements in 2008 to their manufacturer, according to the Global Threat Reduction Initiative.

The investigations were performed on 110 MR-6/80 fuel elements with total burn-up values varying from 40 MWd up to 127 MWd and cooling time from 5 to 27 years.

The only neutron source in spent nuclear fuel is spontaneous fission of nuclides accumulated in the fuel during its operation.

Based on the cross-sections of particular nuclear reactions and decay constants of their products [10,11] the main spontaneous fission neutron sources have been identified.

They have been recognized as transuranic elements, which are formed in series of nuclear reactions and transformations (mainly neutron activation and β^-–decay) of nuclides exposed to high neutron flux in the reactor core.

These reactions and transformations are initiated by neutron reactions with ^{235}U and ^{238}U in nuclear fuel (cf. Fig. 2). Moreover, calculations performed by means of ORIGEN code [12] reveal that the overwhelming majority of neutron emitters in the considered spent nuclear fuel come from reactions originating in ^{238}U.

The spatial distribution of the neutron emission rate from spontaneous fission of transuranic nuclides in spent nuclear fuel is strongly correlated with the local neutron fluence during fuel operation and, therefore, with the fuel burn-up value. This relation is not trivial due to several factors' influence on neutron emission, e.g. operating conditions in the reactor, cooling time, initial uranium enrichment and initial uranium mass.

To define the relation of the neutron emission rate with the burn-up value, the isotopic composition (inventory) of spent nuclear fuel has been estimated. This composition depends on initial fuel enrichment and reactor conditions during fuel operation, i.e. on neutron fluence and reactor operation cycles.

The isotopic composition of spent fuel has been calculated by means of the ORIGEN numerical code [12]. The input data are: initial masses of ^{235}U and ^{238}U, average (total) burn-up of the fuel element and cooling time.

Fluctuations of the vertical burn-up distribution in fuel elements are the result of the heterogeneous distribution of neutron flux density in the reactor core, which is influenced by the surrounding moderator, fuel elements, absorbing rods, irradiation targets etc.

To reconstruct the burn-up distribution, calculations assuming different values of neutron flux density were performed by means of the ORIGEN code.

The assumption that fuel elements had been irradiated during standard MARIA reactor, 100-h duty cycles, in a fixed neutron flux density was made. This assumption, however, can lead to incorrect estimations, because it does not take into account variable operation conditions of each fuel element.

A series of calculations were performed in order to determine the inventory of neutron emitters in the spent nuclear elements. The relative concentration of neutron emitters depends on the fuel burn-up and cooling time. According to the performed calculations, the major contributors to neutron emission from well cooled-down (more than 5 years) spent MR-6/80 type fuel elements appear to be