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FISSION PRODUCT RELEASE FROM HIGH AND LOW-ENRICHED URANIUM FUELS OF THE IVG.1M RESEARCH REACTOR

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The research involved conducting resource tests on two experimental water-cooled technological channels (WCTCs) utilizing low-enriched uranium (LEU) fuel within the IVG1.M research reactor. This testing was a crucial step in the reactor's conversion from highly enriched uranium (HEU) fuel to LEU. The research focused on two key parameters, namely the specific activity and the relative release of fission products (FPs) into the coolant, to evaluate the tightness of the fuel element cladding. A gamma-spectrometric sampling method was proposed to determine the relative release of FPs, which involved assessing the specific activity of the coolant, calculating the release rate (Release), the born rate (Born), and the R/B ratio of FPs. Comparative gamma-spectrometric measurements were conducted to analyze the content of FPs and activation products (AP) in the coolant of WCTCs utilizing both LEU and HEU during the tests. From the comprehensive list of detected radionuclides in the IVG.1M reactor coolant, well-identified reference radionuclides recommended for monitoring fuel element cladding tightness were carefully selected. The results of the study provided insights into the specific activity and relative release of FPs, demonstrating that quantitative values for the relative release of FPs from WCTCs using LEU and HEU fuel were comparable.

Keywords: coolant, fuel element, fuel element cladding, fission products, relative release of fission products, gamma-spectrometry

1. Introduction

One of the requirements for fuel elements developed within the project's framework for converting the IVG.1M research reactor to low-enriched uranium fuel is the value of the permissible release of fission products (FP) from fuel elements into the coolant. Measuring the quantity and distribution of radioactive isotopes in reactor fuel provides a wealth of information regarding fuel behavior. This data is invaluable for studying the assessing fuel element performance during irradiation, and various aspects of nuclear fuel safeguarding. Chemical analysis of spent fuel is laborious and time-consuming, often yielding incomplete results. As a result, non-destructive techniques like passive and active neutron counting, calorimetric measurements, and gamma spectroscopy studies are gaining importance. Among these techniques, gamma scanning stands out as it is the sole non-destructive method for the quantitative measurement of gamma-emitting fission or activation products in spent fuel [1].

A suitable method for power distribution determination in the reactor core based on measurement and analysis of the short-living fission products in lightly irradiated fuel pins has been developed on the experimental facility for gamma scanning at the LR-0 experimental reactor [2-4]. The analysis of fuel rod failure character is the key to a real-time detection system for fuel rod failure in a pressurized water reactor (PWR) of great significance for the safe operation of nuclear reactors [5,6].

A mathematical treatment has been developed to predict the release of volatile fission products from operating defective nuclear fuel elements. [7-10] for type CANDU and LWR, WWER reactors.

For any nuclear reactor, the allowable release of fission products (FP) is determined with the objective of ensuring the necessary level of operational safety. This determination hinges upon the effectiveness of protective barriers against the propagation of radionuclides, as well as the reliability and accuracy of methods used to monitor FP concentrations in the reactor coolant [11-16].

One practical approach to address the challenge of monitoring FP levels in a nuclear reactor's coolant involves the implementation of Fuel Element Cladding Tightness Monitoring (CTM) systems. These systems facilitate the timely identification of cladding damage in fuel elements when FP concentrations exceed established limits. They enable continuous monitoring of the situation's evolution and empower decision-making regarding the continued operation of compromised elements [17].

In the context of the IVG.1M reactor, FP concentrations and APs in the coolant are monitored using the CTM system. This system, established in 1990, serves as a critical component of the reactor's safety systems.

Between 2017 and 2019, the CTM system played a crucial role in verifying the cladding integrity of experimental water-cooled technological channels with low-enriched uranium fuel (WCTC-LEU) during their operational lifespan within the IVG.1M reactor [17].

This article focuses on the findings related to FP and AP concentrations in the coolant during the IVG.1M reactor startup. These measurements were comparative in nature, as samples of coolant were extracted from individual cooling paths within two WCTC-LEU systems and standard water-cooled technological channels with highly enriched uranium fuel (WCTC-HEU).

2. Material and methods

Within the framework of the accepted research procedure, the content of FPs and APs in the coolant samples of the IVG.1M reactor was determined. Coolant samples were taken from the WCTCs-LEU cooling paths loaded into cells No. 14 and 24 of the IVG.1M reactor core and from the WCTCs –HEU cooling paths loaded into the remaining 28 cells of the reactor core (Fig. 1).

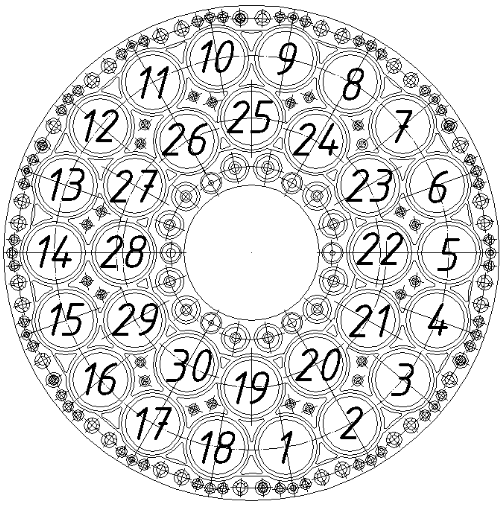


Fig. 1. The cartogram of the WCTC layout in the reactor core

The gamma spectra of the coolant samples were measured using an InSpector-2000 gamma spectrometer with a GC1020 coaxial detector. The spectra were processed in the GENIE-2000 environment and further analyzed using the IPF interactive peak fitting program. The values of the specific activity of AP or FP in the coolant (A0) at the time of sampling were calculated using Formula (1):

$$A_0 = A_i(t_1) \cdot \exp(\lambda t_1) \quad (1)$$

where:

A(t1) – represents the specific activity of AP (FP) at the time of measurement in Bq/l;

λ – is the decay constant in s^{-1};

t_1 – represents the time elapsed from sampling to the start of measuring coolant activity in seconds.

The degree of fuel element tightness is characterized by the relative release of FP, defined as the ratio of the FP release rate into the coolant (R) to the rate of its creation (B). The creation rate of a B nuclide, accounting for the formation of its predecessors, is calculated as shown in Formula (2):

$$B = P_c \cdot 3{,}2 \cdot 10^{13} \eta \quad (2)$$

where:

P_c – represents WCTC power in kW;

η – stands for the relative yield of this nuclide and its predecessors per fission of ^{235}U in relative units;

$3.2 * 10^{13}$ - is the number of ^{235}U fissions required to release 1 kJ (for a reactor operating for a long time at a constant power level).

The release rate of R radionuclides from FAs into the coolant per unit time is determined from their measured specific activity A(t1) using Formula (3):

$$R = \frac{A(t_1) \cdot Q_c \cdot 10^{-3}}{\rho \cdot \lambda \cdot F} \quad (3)$$

where:

$A(t_1)$ – is the specific activity of FP during measurement in Bq/l;

Q_c – is the coolant flow rate through the WCTC in g/s;

ρ – represents the coolant density in g/cm³, with ρ = 1 g/cm³;

λ – is the decay constant of the measured nuclide in s^{-1};

F – is a correction factor for decay from sampling to measurement (F = exp (λ * t_1) in relative units).

The relative yield of the i-th FP into the coolant (R/B) for each WCTC is determined as shown in Formula (4):

$$(R/B)_i = \frac{A(t) \cdot Q_c \cdot 10^{-16}}{P_c \cdot 3{,}2 \cdot \eta \cdot \rho \cdot \lambda \cdot F} \quad (4)$$

The average relative yield for *m* FP analytes of the j-th WCTC is calculated using Formula (5):

$$R/B_{cp\ j} = \sum_{i=1}^{m} (R/B)_i \Big/ m \quad (5)$$

The arithmetic means of the relative yield of activation products for the 28 standard WCTCs is determined by Formula (6):

$$R/B_{cp} = \sum_{j=1}^{n} (R/B)_j \Big/ n \quad (6)$$

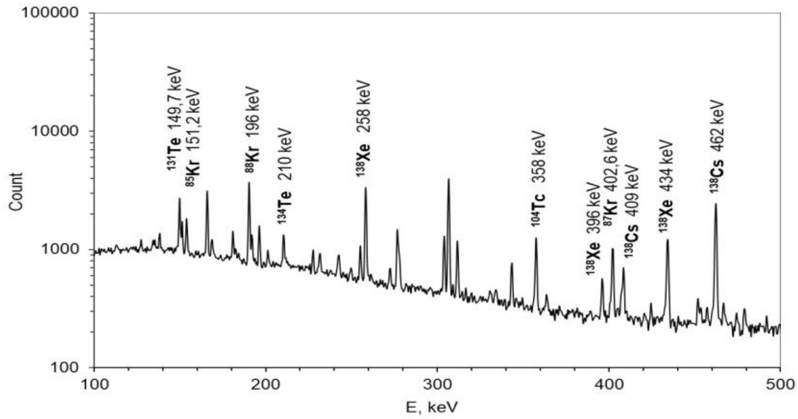
The root-mean-square deviation of the relative yield of FP for the 28 standard WCTCs is calculated using Formula (7):

$$\sigma = \sqrt{\frac{\sum_{j=1}^{n}((R/B)_j - (R/B)_{cp})^2}{n-1}} \quad (7)$$

3. Results

The results of gamma-spectrometric studies of the coolant of experimental and standard WCTC are presented in [17]. Figure 2 shows a typical gamma spectrum for the IVG.1M reactor of the coolant sample taken from the WCTC-LEU cooling path after the reactor was brought to a constant power level of 6 MW.

After sampling and subsequent measurements, Ar-41 was identified in the gamma radiation spectra of the coolant. In contrast, the gamma radiation spectra of the WCTC-HEU coolant revealed the presence of AP elements that are part of the AMg6 alloy. This alloy is used in the construction of the shell, end grids of the fuel assemblies (FAs), and casing of standard WCTCs-HEU. It's worth noting that WCTC-LEU doesn't contain any parts made of aluminum alloys, which explains the significantly lower activity levels of isotopes Mg-27, Na-24, and Mn-56 in the WCTC-LEU coolant when compared to the WCTC-HEU coolant. Weighted average values of specific activity of AP in the coolant at startup of the reactor IVG.1M are given in Table 1.



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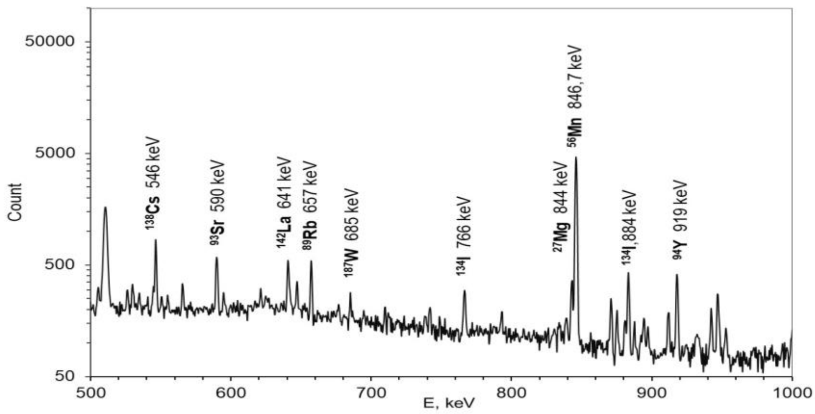

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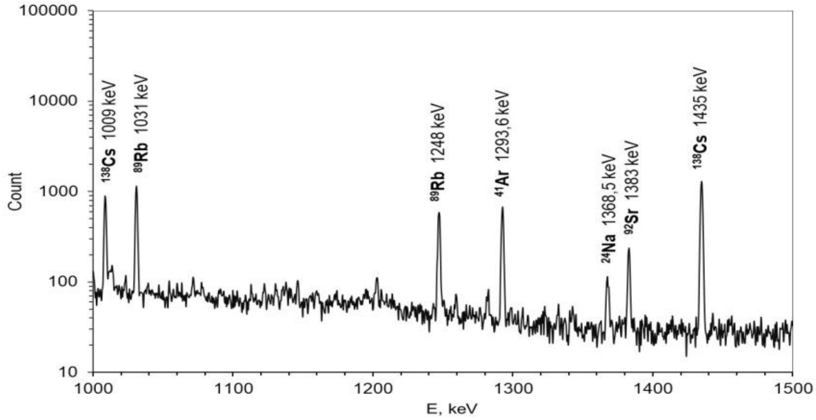

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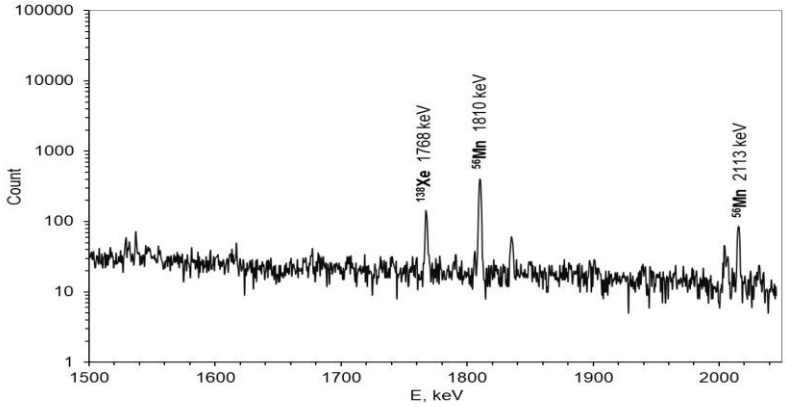

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Fig. 2. Gamma spectrum of the IVG.1M reactor coolant:
a) spectral region from 100 to 500 keV; b) 500 to 1000 keV; c) 1000 to 1500 keV; d) 1500 to 2000 keV.

Table 2 summarizes the results of coolant samples analyzed during a typical reactor startup, providing information on specific activity and the relative release of FPs.

Table 1. Weighted average values of the AP specific activity in the coolant at the start-up of the IVG.1M reactor.

Isotope-AP	A_0, Bq/l			
	WCTC-LEU #14	WCTC-HEU #18	WCTC-LEU #24	WCTC-HEU #22
Na-24	1.8E+03	5.9E+03	1.8E+03	6.9E+03
Ar-41	1.5E+04	1.4E+04	1.9E+04	2.3E+04
Mn-56	5.8E+04	9.2E+04	6.0E+04	1.0E+05
W-187	3.1E+03	5.4E+03	2.2E+03	6.9E+03
Mg-27	5.7E+04	4.1E+05	6.0E+04	5.7E+05

Table 2. Weighted average values of the relative release of FPs into the coolant at the start-up of the IVG.1M reactor

Isotope - FP	WCTC-LEU #14		WCTC-HEU #18		WCTC-LEU #24		WCTC-HEU #22	
	A(t), Bq/l	R/B, rel. units	A(t), Bq/l	R/B, rel. units	A(t), Bq/l	R/B, rel. units	A(t), Bq/l	R/B, rel. units
Kr-85	1.4E+03	7.8E-07	4.0E+02	2.4E-07	2.0E+03	1.1E-06	8.5E+02	4.7E-07
Kr-87	6.1E+03	7.1E-07	1.8E+03	2.1E-07	1.1E+04	1.1E-06	4.3E+03	4.5E-07
Kr-88	5.4E+03	7.7E-07	1.1E+03	1.6E-07	8.2E+03	1.0E-06	3.3E+03	4.3E-07
Rb-89	1.5E+04	9.7E-07	4.9E+03	2.8E-07	3.2E+04	1.1E-06	9.5E+03	4.4E-07
Sr-92	4.0E+03	3.3E-07	1.9E+03	1.7E-07	4.0E+03	3.0E-07	2.2E+03	1.7E-07
Y-94	4.8E+03	2.1E-07	2.3E+03	9.1E-08	7.2E+03	1.9E-07	3.1E+03	1.0E-07
Tc-104	2.3E+03	3.5E-07	1.2E+03	1.7E-07	3.6E+03	3.2E-07	1.7E+03	2.0E-07
Te-131	3.1E+03	3.1E-07	1.2E+03	1.1E-07	3.2E+03	2.1E-07	1.8E+03	1.4E-07
I-133	8.3E+02	3.8E-07	5.1E+02	2.6E-07	1.4E+03	4.1E-07	8.4E+02	5.6E-08
I-134	6.6E+03	2.5E-07	1.6E+03	6.7E-08	4.0E+03	1.3E-07	3.1E+03	1.0E-07
Te-134	4.6E+03	1.6E-07	1.1E+03	3.7E-08	4.4E+03	1.2E-07	4.3E+02	3.8E-08
Cs-138	3.0E+04	1.1E-06	8.2E+03	3.0E-07	4.8E+04	1.3E-06	1.8E+04	5.5E-07
Xe-138	1.2E+04	6.4E-07	3.9E+03	1.7E-07	3.4E+04	9.2E-07	1.0E+04	3.3E-07
La-142	5.2E+03	2.9E-07	1.8E+03	1.1E-07	5.4E+03	2.6E-07	3.4E+03	1.7E-07
$\overline{R/B}$		5.2E-07		1.7E-07		6.0E-07		2.6E-07

The outcomes of assessing the average relative release of FP for all 28 WCTC-HEU are graphically represented in Fig.3.

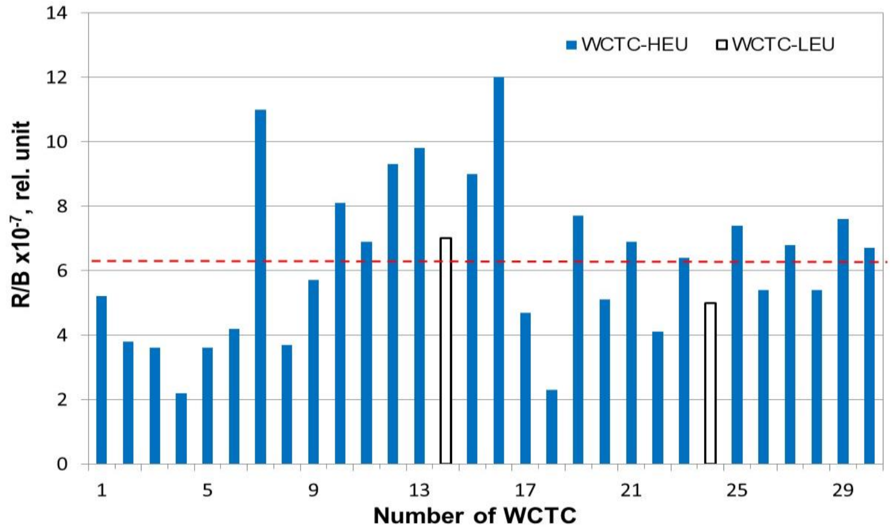

Fig.3. Vales of FP release from WCTC-HEU and WCTC-LEU

The same figure shows the results of R/B determination for two experimental WCTC-LEU averaged for three experiments completing the IVG.1M reactor core operation with 90% enriched fuel. The dashed line in the figure denotes the average relative FP release observed across these 28 WCTCs-HEU. Additionally, the figure depicts the relative release results for two WCTCs-LEU. For the 28 WCTCs-HEU, the average value of the relative release of FP into the coolant, as shown in Figure 3, amounted to 6.3 10^{-7}, with a corresponding standard deviation of 2.5 10^{-7}.

4. Discussion

The obtained results highlight the difference between the relative release of FP for the two experimental WCTCs-LEU and the average relative FP release of standard WCTCs-HEU is within one standard deviation. This observation leads to the conclusion that these parameters closely align, indicating that the quality of fuel element cladding in WCTCs-LEU is comparable to that in WCTCs-HEU. Essentially, WCTCs-LEU exhibit cladding tightness that is on par with the quality observed in fuel elements using high-enriched uranium.

During the analysis of the coolant spectra, the activity of fourteen FPs was determined, encompassing various groups of chemical elements, including halogens, noble gases, alkali metals, metals, and non-metals. Notably, specific radionuclide analytes were carefully chosen for exclusive content measurement in the coolant. This selection enabled the comprehensive monitoring of fuel element cladding tightness concerning the parameters of FP release into the coolant.

The criteria for selecting FP-analytes were based on the presence of prominent FP gamma lines in the spectrum and the reliability of FP identification ensuring the absence of other competing lines near the gamma line of interest. Equally significant was the capability of reliably identifying these FP analytes in the spectra of coolant obtained from WCTCs with varying relative FP releases.

The analysis of measurement results emphasizes that the technique of experimental determination of FP content in samples provides reliable control of fuel element cladding tightness. This technique allows to effectively reduce the workload in the fuel element cladding tightness monitoring system (CTM) without reducing the quality of this control.

5. Conclusion

In conclusion, based on the analysis of the results of measuring the release of fission products and activation products into the coolant of the IVG.1M reactor during life tests of the WCTCs with LEU fuel, the following conclusions can be drawn:

- the release of fission products from the WCTCs loaded with LEU is comparable to the release from the WCTCs loaded with HEU. This indicates that the quality of the fuel element cladding with LEU fuel is acceptable and is not different from the quality of fuel element cladding with HEU fuel in terms of fuel tightness.

- the coolant of WCTCs-LEU contains significantly lower levels of Mg-27, Na-24, and Mn-56 isotopes compared to the coolant of WCTCs-HEU. This reduction is due to the phased-out use of parts and assemblies made of aluminum alloy AMg-6 in the structure of the WCTCs-LEU.

- the tightness of the fuel element cladding in the IVG.1M reactor can be effectively monitored by conducting exceptional measurements of analyte radionuclide content in the coolant. This method is characterized by intense FP gamma lines and the absence of competing lines of other radionuclides nearby, which enhances control efficiency and reduces operational intensity.

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