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Optimization of conditions for topaz irradiation in the WWR-K reactor

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Abstract. Activation of impurities in topazes happens due to irradiation with thermal neutrons (induced radioactivity occurs), which complicates their further handling. This leads to the need for their long cooling for safe use. The Institute of Nuclear Physics (Kazakhstan) is conducting R&D to develop a method for the effective formation of color centers in topaz during their irradiation in the WWR-K reactor. An irradiation capsule design has been developed in which optimized conditions for irradiating stones in the neutron field of the reactor are formed. The capsule uses shielding materials made of boron carbide and tantalum to cut off thermal neutrons, resulting in a reduction in induced radioactivity in topaz. The effectiveness of the irradiation capsule was tested in the core of the critical facility. As a result, thermal neutron flux is reduced by 5.7 times and the induced activity of the tantalum is reduced by 2.2 times.

Keywords: Critical facility • Sandwich screen • Tantalum • Topaz • WWR-K reactor

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Introduction

In recent years, radiation treatment of semiprecious stones, in particular, topaz based on research reactors, has been actively developed as one of the methods for radiation processing of materials [1–7]. This is due to the fact that most of the semiprecious stones mined are colorless, as a result of which they have practically no jewelry value. During radiation treatment of topazes, in particular, by neutron radiation, they are colored in light blue (Sky blue), blue (London blue), and dark blue (Swiss blue) [8, 9]. The color change occurs due to the formation of defects in the lattice of stones, which are formed upon interaction with fast neutrons [10]. Therefore, in the irradiation position of the reactor, it is necessary to have a "hard" neutron spectrum, which will lead to effective staining of stones. To obtain the desired color of topazes, it is necessary to accumulate a fluence of fast neutrons $\geq 10^{18}$ n·cm^{-2} in them [7]. At the same time, it should be borne in mind that topazes contain impurities that are activated during irradiation, while the induced activity is proportional to the flux of thermal and epithermal neutrons (most operating research reactors have a thermal neutron spectrum). The presence of such induced activity leads to an increase in the time of sedimentation of stones after irradiation, which is a negative effect. Based on this, it is necessary to create conditions in the irradiation position for effective coloring of topazes (fast "hard" neutron spectrum) and minimizing the activation of impurities [3, 5, 6]. Another important factor in topaz irradiation is the irradiation temperature, since at high temperatures (>200°C), stones crack and/or defects in the lattice anneal, which leads to a change in the optical and mechanical properties of topaz [1, 7]. It is possible to form the necessary conditions in the irradiation position due to the correct development of the device design (capsule).

Materials and methods

After carrying out complex neutron-physical and thermophysical calculations and considering five options for the design of the irradiation device [11], the optimal option was determined – a device with a sandwich screen made of boron carbide and tantalum. Subsequently, a mock-up of the selected option of the irradiation device design was made.

Experimental work has been carried out on a critical stand to study the neutron-physical characteristics of a model of an irradiation device with a sandwich screen. The critical facility is a zero-power reactor and is intended for modeling the cores of light water reactors (LWRs) and studying the neutronic characteristics of various core configurations and designs of experimental/irradiation devices. The critical facility consists of a critical assembly and a set of equipment for controlling physical processes. The critical assembly makes it possible to simulate the core of various pressurized water research (PWR) reactors, in particular, the core of the WWR-K reactor. The main physical and technical characteristics of the critical assembly are given in Table 1.

The model of the irradiation device (Fig. 1) is a capsule made of aluminum alloy SAV-1 and a radiation screen (Fig. 2). The model of the irradiation device has holes for the flow of coolant and cooling of topazes. The height of the useful volume in the capsule is 50 mm, taking into account the installed screen.

The sandwich screen covers the end and inner surfaces of the capsule case and is a hollow cylinder with double walls, with boron carbide powder poured into the space between them. The thickness of the boron carbide backfill was 1.5 mm. The backfill height was 50 mm and the backfill density was 1.53 g·cm^{-3}. On the outside, the cylindrical screen was covered with a second screen made of tantalum foil 0.2 mm thick.

Table 1. Critical facility characteristics

Reactor type	Tank
Thermal power (W)	100
Reflector	Light water and/or beryllium
Moderator	Light water and/or beryllium
Maximum thermal neutron flux (cm^{-2}·s^{-1})	3×10^9
Maximum fast neutron flux (cm^{-2}·s^{-1})	2×10^8
Fuel composition	UO_2 + Al
U-235 enrichment (%)	19.7
Experimental channel's diameter (mm)	65, 96, 140

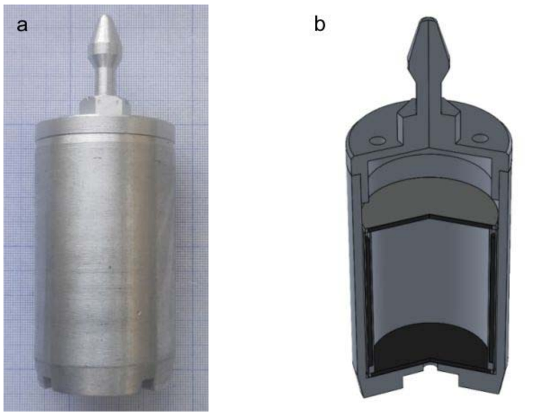

Fig. 1. Layout of the irradiation device: (a) appearance; (b) three-dimensional cross-sectional image.

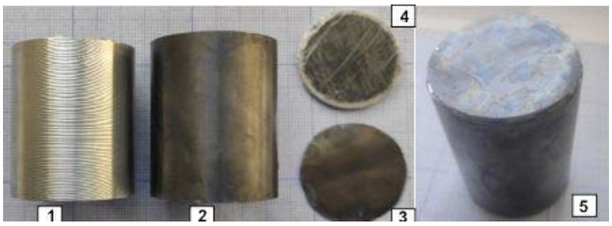

Fig. 2. Appearance of a sandwich screen made of tantalum foil and boron carbide: 1 – screen made of boron carbide, 2 – screen made of tantalum, 3 – end tantalum screen of a round shape, 4 – screen of boron carbide of a round shape, 5 – view of a sandwich screen assembled.

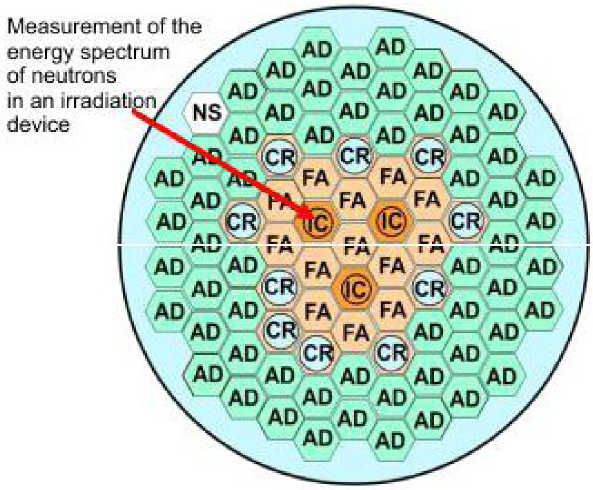

Fig. 3. The core of the critical facility. AD, aluminum displacer; CR, type 2 fuel assemblies with an absorbing rod; FA, type 1 fuel assemblies; IC, irradiation channel; NS, neutron source.

The model of the irradiation device was installed in cell 9-9 (central position) of the critical assembly core (Fig. 3). Moreover, the center of the layout coincided with the central plane of the core. In all experiments, the internal cavity of the model was filled with topazes (Fig. 4).

To measure the energy distribution of neutrons in the layout of the irradiation device, the neutron activation method was used, the essence of which is to irradiate activation detectors in the studied

Fig. 4. Non-irradiated topaz.

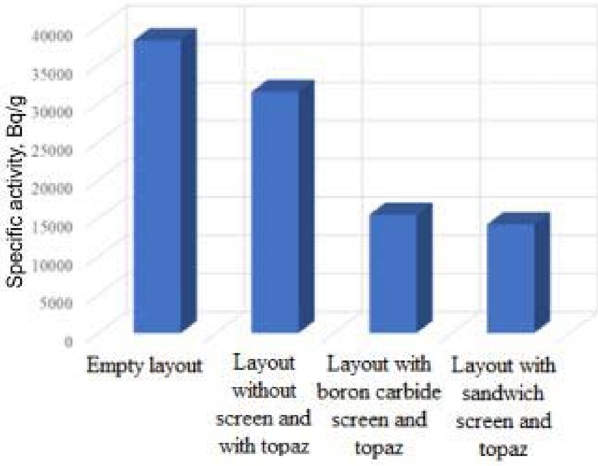

Fig. 5. Dynamics of changes in the induced activity of a tantalum monitor.

neutron field, with further measurement of the induced activity of the detector. Then, the neutron flux density is determined [12].

The study of the neutron energy spectrum in a mock-up irradiation device was carried out for two regions: the thermal region (E_n <0.625 eV) and the fast region (E_n ≤1.15 MeV). The thermal neutron flux density was measured using activation detectors made of gold, the activation of which took place according to the nuclear reaction of radiative capture $^{197}Au(n,\gamma)^{198}Au$. Gold detectors were irradiated with and without cadmium screens. The thermal neutron flux density was determined from the difference in induced activity in gold detectors with and without a cadmium screen. The fast neutron flux density was measured using indium activation detectors, the activation of which took place according to the following nuclear reaction $^{115}In(n,n')^{115m}In$, which is a threshold reaction. Indium detectors were irradiated in cadmium screens. Detailed information on the nuclear physical characteristics of the activation detectors used is given in Table 2.

The induced gamma-ray activity of the detectors was measured using a Canberra GX 2518 wide-range (3 keV – 3 MeV) germanium gamma spectrometer, with a relative efficiency of 25%. Before starting work, the gamma spectrometer was calibrated with a set of standard reference sources of gamma radiation.

The detectors were irradiated at a critical assembly power of about 40 W. The flux density of thermal and fast neutrons was measured at least twice for each layout configuration. To obtain the most reliable values of the influence of the screen on the energy distribution of neutrons, the detectors were irradiated together with topazes. In addition, it was decided to use a tantalum monitor as a reference material to determine the efficiency of cutting off neutrons with energies that activate tantalum in topazes, which is one of the main impurities.

Results and discussion

The results of the energy distribution of neutrons obtained from a series of experiments are given in Table 3. The dynamics of changes in the activity of the tantalum monitor with a change in the configuration of the layout of the irradiation device are shown in Fig. 5. It should be noted that the data given in Table 3 correspond to the average values of the neutron flux density obtained from a series of detector irradiations.

Table 3 shows that when using a sandwich screen, the density of the thermal neutron flux decreases by 5.7 times, which will also lead to a decrease in the activity of impurities by so many times. At the same time, the density of the fast neutron

Table 2. Nuclear physical characteristics of activation detectors

Detector type	Gold	Indium
Neutron energy region	Thermal	Fast
Energy threshold	$0 \le E_n \le 0.625$ eV	E_n >1.15 MeV
Nuclear reactions	$^{197}Au(n,\gamma)^{198}Au$	$^{115}In(n,n')^{115m}In$
Half-life	2.696 days	4.486 h
Reaction cross-section (barn)	98.7	0.286

Table 3. The flux density of thermal and fast neutrons in the layout of the irradiation device

Configuration	Neutron flux density ($cm^{-2}\cdot s^{-1}$)	
	E_n <0.625 eV	E_n >1.15 MeV
Empty layout	$(1.28 \pm 0.11) \times 10^9$	$(2.38 \pm 0.20) \times 10^8$
Layout with topaz	$(8.76 \pm 0.62) \times 10^8$	$(2.45 \pm 0.19) \times 10^8$
Model with boron carbide screen and topaz	$(1.72 \pm 0.14) \times 10^8$	$(2.15 \pm 0.18) \times 10^8$
Layout with sandwich screen and topaz	$(1.54 \pm 0.62) \times 10^8$	$(2.28 \pm 0.17) \times 10^8$

Table 4. Radionuclide composition of topaz after each stage of experiments

Layout type	Specific activity (Bq·g^{-1})		
	Ga-72 ($T_{1/2}$ = 14.1 h)	Na-24 ($T_{1/2}$ = 14.9 h)	Ta-182 ($T_{1/2}$ = 114 days)
Layout without a screen	1290 ± 70	28 658 ± 1146	23 ± 3
Layout with boron carbide screen	316 ± 16	12 600 ± 447	15 ± 4
Layout with sandwich screen	302 ± 43	13 666 ± 548	–

flux decreases slightly, by 7% – another aspect of the effectiveness of the sandwich screen used is the activation of tantalum-182 in the monitor. Figure 5 shows that the use of a sandwich screen leads to a 2.2-fold decrease in the activation of the tantalum monitor, which is a direct proof of the effectiveness of the studied design of the irradiation device.

In addition, a gamma-spectrometric analysis of topaz irradiated in different configurations of the device layout was carried out. The results of this analysis are shown in Table 4. Only three gamma--ray emitters were detected: Ga-72, Na-24, and Ta-182, which is due to the low power and duration of irradiation of the stones. From the data given in Table 4, a decrease in the activation of impurities in topaz is clearly visible (at least twice).

When using a sandwich screen, the induced activity of tantalum (Ta-182) is below the sensitivity limits of the gamma spectrometer. From this, it can be concluded that tantalum was practically not activated. Of course, the full-fledged effectiveness of the developed screen can be determined by experiments at the WWR-K reactor (planned as the next stage of research) since due to the low power of the critical facility, impurities in topaz are activated insignificantly.

Conclusions

The neutron-physical characteristics of the irradiation device layout were studied at the critical facility. In particular, the energy distributions of neutrons in the model of an irradiation device with various configurations were investigated. The density of the thermal neutron flux in the model of the irradiation device with topaz was 8.8 × 10^{9} cm^{-2}·s^{-1}, and the density of the fast neutron flux was 2.5 × 10^{8} cm^{-2}·s^{-1}. When using a boron carbide screen, the density of the thermal neutron flux decreases by 5.1 times, and the density of the fast neutron flux by 14%. And when using a sandwich screen made of boron carbide and tantalum, the density of the thermal neutron flux decreases by 5.7 times, and the density of the fast neutron flux by 7%. In addition, the use of a tantalum monitor further confirms the effectiveness of the developed design of the irradiation device. The induced activity of the tantalum monitor was reduced by 2.2 times when using a sandwich screen. The results of gamma-spectrometric analysis of irradiated topaz showed that there is a decrease in the induced activity of radioisotopes Ga-72, Na-24, and Ta-182 by at least 2 times.

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