JOINT INSTITUTE FOR NUCLEAR RESEARCH

Frank Laboratory of Neutron Physics

FINAL REPORT ON THE
SUMMER STUDENT PROGRAM

Radiation coloring of topazes at the IBR-2 reactor

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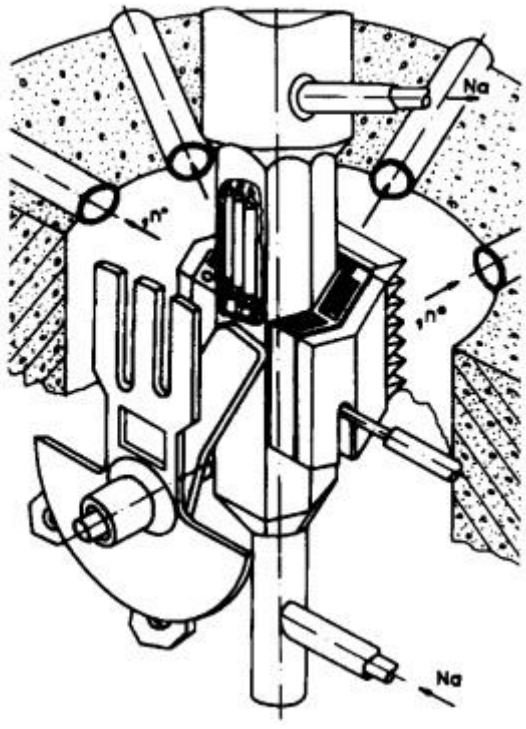
Contents:

1. Literature analysis

1.1 IBR-2

IBR-2 is a pulsed research nuclear reactor with compact active core (Bulavin M. et al, 2015). The core of the reactor is an irregular hexahedron composed of fuel element subassemblies. There are 7 fuel elements (plutonium dioxide) in each subassembly. The cooling system has three circuits and two loops. In the first and second circuits the coolant is liquid sodium and in the third it is air. The core is installed in a double-walled steel vessel and is surrounded by a number of stationary reflectors, control and safety among them. Around the reactor are water moderators which are scanned by 14 horizontal channels for neutron extraction. Two of the moderators are grooved (Fig. 1). The power pulses at 5 Hz are generated by reactivity modulators: the main moveable reflector (MMR) and the auxiliary moveable reflector (AMR). When they both approach the core a power pulse develops. For a pulse repetition frequency of 25 Hz the AMR is slowed down (Belushkin A., 2006).



The IBR-2 reaserch fast neutron reactor (JINR, Dubna) has been put into operation in 1984 and shut down in 2006 for modernization because of the expiration of his service life. In this connection the main equipment (vessel, drivers and movable operating units of the control and safety system) and fuel loadings of the reactor were replaced. The biological shield, reactivity modulator and technological system remained the same (Ananiev V. et al, 2013). The parameters before and after the modernization are given in Table 1. The reactor provides activation with thermal, epithermal and fast neutrons for analytic purposes (Frontasyeva M. et al, 2000).

Fig. 1. A schematic view ofprincipal IBR-2 parts. Shown are the main and auxialiary refzectors,oneof the control reflectors,core,water moderators, sodium cooling system and horizontal neutron beams.

Table 1. The IBR-2 reactor parameters before and after the modernization.

Parameter	IBR-2 before modernization	IBR-2 after modernization
Average power, MW	2	2
Fuel type	PuO_2	PuO_2
Number of fuel assemblies	78	69
Maximum burn-up, %	6.5	9
Pulse frequency, Hz	5; 25	5; 10
Pulse half-width, μs	215	200
Rotation rate, rev/min: main reactivity modulator auxiliary reactivity modulator	 1500 300	 600 300
Material of main and auxiliary reactivity modulators	steel	nickel
Moveable reflector life, hr	20000	50000
Number of satellites at 5 Hz	4	1
Coolant	Sodium	Sodium
Sodium flow rate through the core, m^3/hr	80-110	80-110

The loading of IBR-2 took 14 months (from 17.12.2010 until 14.02.2011) and the rated power of 2MW was achieved at 12.10.2011. The critical loading of the modernized reactor is 64 fuel assemblies. The remaining uncharged cells will ensure the reactor service life for no less than 20 years (Fig. 2) (Ananiev V. et al, 2013).

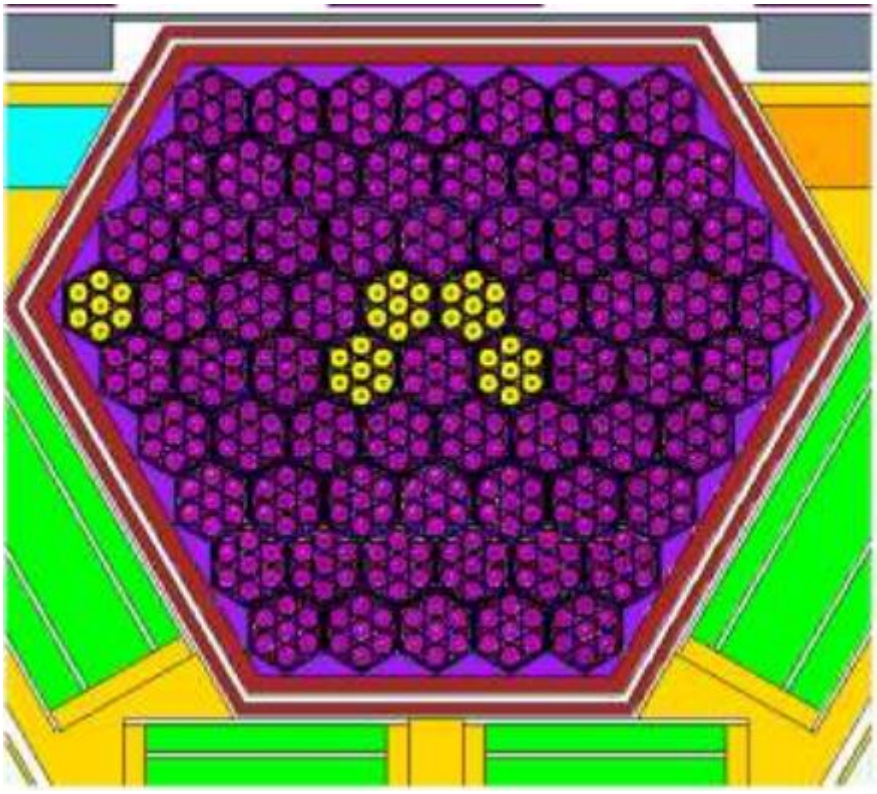


Fig. 2. Final core loading pattern of the IBR-2 modernized reactor with 64 fuel assemblies.

Two safety system units are intended for fast emergency power cutback. In 0.1s each of them reduces reactivity by 0.12%, which reliably brings the reactor below prompt criticality and suppresses neutron generation. The response of the reactor to external reactivity perturbations due to a change in the operation mode or failure of various technological systems that ensure normal functioning of the reactor has been studied. All main reactivity effects are negative. As the power increases, various power feedback effect components start to develop. Numerous experiments in the power range from 500 to 2MW and at a sodium flow rate through the core from 60 to 100m^3/h have demonstrated that power, flow rate and temperature effects are negative. The power pulse shape is the most important characteristic of IBR-2M both for the reactor personnel and the neutron beam users in neutron time-of-flight experiments. The IBR-2 pulse shape is close to a truncated Gaussian distribution with half-width and half-maximum of 200±4μs (Fig. 3) (Ananiev V. et al, 2013).

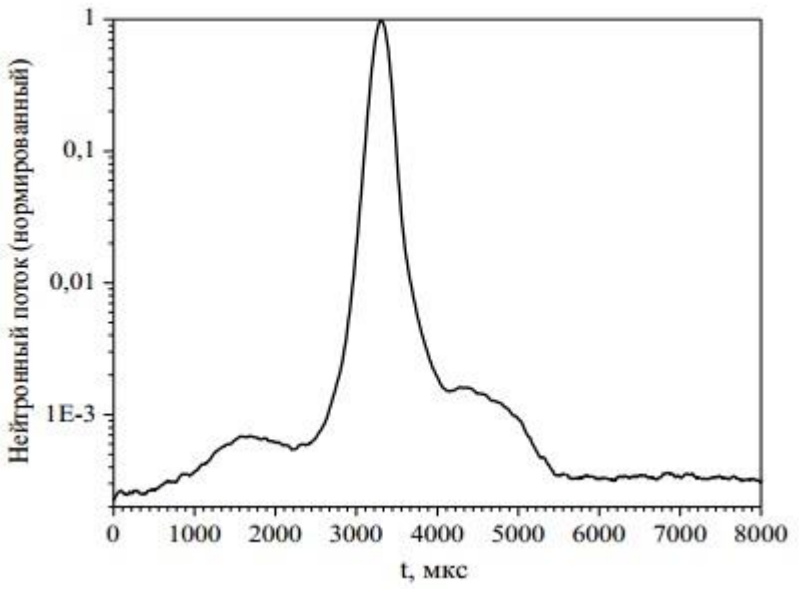


Fig. 3. IBR-2 power pulse shape. The data is normalized to the pulse maximum.

A distinctive feature of IBR-2 as compared with steady-state reactors is a high sensitivity of IBR-2 to external reactivity fluctuations: about 40 times higher than that of steady-state reactors with uranium fuel. A normal operation of IBR-2 requires minimization of all reactivity noise sources (Ananiev V. et al, 2013).

The investigations have shown that the main source of pulse energy fluctuations are axial (towards the core) vibrations of the blades of the movable reflectors. Other effects, for instance, fluctuation of the temperature and flow rate of sodium passing through the core in a turbulent flow manifest themselves to a much lesser degree and their influence on the fluctuations on power may be neglected (Ananiev V. et al, 2013). Table 2 presents some nuclear-physical parameters of IBR-2 characterizing the reactor both as a nuclear physical facility and a pulsed neutron source.

Table 2. The IBR-2 nuclear-physical characteristics.

Parameter	Parameter notation	Value
Half-width at half-maximum	$\Theta_{1/2}$, μs	200±4
Pulse power	W, MW	1830
Power in background (between pulses)	W_b, MW W_b, %W_{aver}	0.2 8.6
Average peak fast neutron flux density in safety system	Φ_f, n/(cm^2/s)	$2.26 \cdot 10^{17}$
Average thermal neutron flux density on grooved water moderator surface	Φ_t, n/(cm^2/s)	$\sim \cdot 10^{13}$

In order to conduct experiments at IBR-2 on neutron activation analysis and radiation studies there are: a pneumatic system 'Regata,' channel 11 (for multipurpose studies), several experimental sites with 3 hot chambers to irradiate samples for material studies (Belushkin A., 2006). Table 3 shows the analytic investigations that have been done at IBR-2 reactor through the years:

Table 3. Analytic investigations at IBR-2 reactor.

Instrumental neutron activation analysis Conventional INAA	Epithermal neutron activation analysis ENAA	Cyclic neutron activation analysis CNAA

Environmental Studies	Material Science
• Air pollution studies (air filters) • Biomonitoring atmospheric depositions (moss, lichens) • Assessment of the environmental situations (waste dumps, oil accidents, aquatic pollution (Moscow Sea, Oka River Basin, Lake Baikal)	• High purity materials (aluminium, semiconductors - silicon, germanium, gallium arsenide) • Biotechnologies: (i) biochemical leaching of elements by aerobic and anaerobic bacteria; (ii) Se-containing pharmaceuticals based on blue-green algae Spirulina Platensis • Construction materials in the problem of decommissioning of nuclear power plants

1.2 Irradiation facility

The irradiation facility is installed in the channel No. 3 zone and it is the modified version of the set-up previously created and used at that place (Bulavin M. et al, 2015).

The inner concrete wall in the direction of the 3d channel has an opening of the rectangular cross-section with 200 mm 400 mm dimensions for extraction of neutron beam. The second (external) wall of the biological shield in the channel № 3 zone has a hole with 800 mm diameter for entering the irradiation facility to the circled corridor. The irradiation facility is intended for studying of a radiation hardness of any materials. The samples for irradiation may be placed not closer than 40 mm of the water moderator surface, behind which the IBR-2 reactor core is. The final distance from the sample under irradiation and the water moderator is selected according to the needed value of neutron integral fluence (Bulavin M. et al, 2015).

The irradiation facility represents horizontally positioned 800 mm diameter steel cylinder filled with water (Fig. 4). The transport beam (2) is fixed to the front end of the cylinder (6). The samples to be irradiated are placed on the metallic container (3) made from Al alloy AMG6 with the transverse dimensions 160 mm 160 mm. This alloy contains minimum quantity of admixtures, and its induced gamma activity quickly drops off after irradiation. The container is fixed to the front end of the transport beam, which is made from AMG6 alloy and H-shaped in crosssection with the following dimensions: width – 100 mm, height – 82 mm and length – 2.7 m. The transport beam is connected to the massive part of the irradiation facility (6) by folding flanged joint. The irradiation facility may be moved along the railway with the help of mechanism with

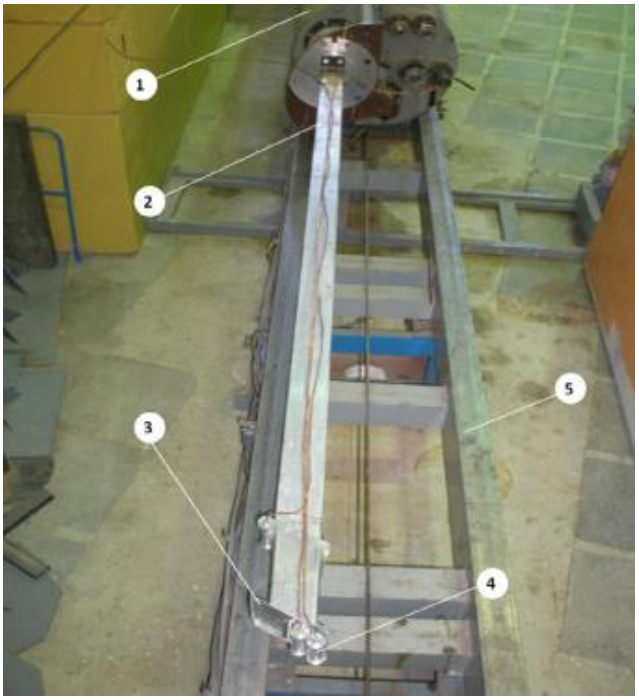


Fig. 4. The irradiation facility at the channel No. 3 of IBR-2 reactor experimental hall, the view from the external biological shield side: 1 – massive part of the irradiation facility, 2 – transport beam, 3 – metallic container for samples fastening, 4 – samples, 5 – rail way.

electric drive (Bulavin M. et al, 2015).

The movement of this facility along the rails are restricted (Fig. 4) both by terminal switches and emergency stops, positioned on both ends of the railway, and by the seen externally mechanical stop, which defines the minimal distance from the samples to the surface of the IBR-2 water moderator. The irradiation facility has a biological shield (Fig. 5) which prevents the experimentalists from the ionizing irradiation during an operation of the reactor (Bulavin M. et al, 2015). An irradiation time depends of the neutron fluence required and is varied from the few hours up to 11 days – the normal duration of an operation cycle of the IBR-2 reactor. The samples temperature during irradiation does not exceed 50ºC, that partially guaranteed by the extracting ventilation system. In case of necessity, irradiation of samples may be provided at cryogenic temperature. The control of neutron fluence is done (with an accuracy of about 5–10%) by placing activation foils near the irradiated materials with subsequent measurement of the foils induced activity. In the steel cylinder of the irradiation facility bended tubes were introduced. That permits to lay the power and signal cables to and from devices, which should be monitored during an assigned doze accumulation. More detailed picture of the forward part of the irradiation facility is shown at the Fig. 3. The samples under investigation would not exceed the size of the metallic container (160 mm 160 mm) (Bulavin M. et al, 2015).

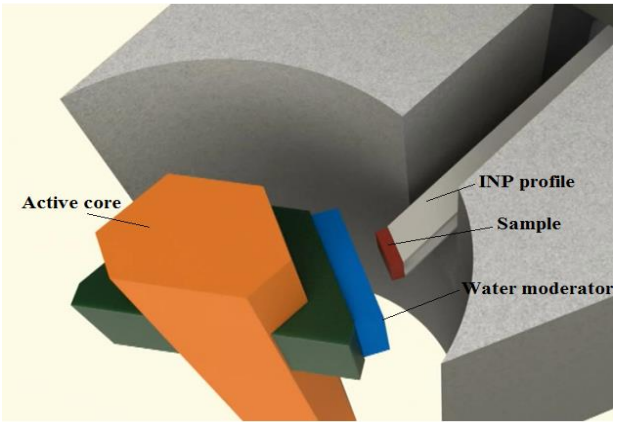
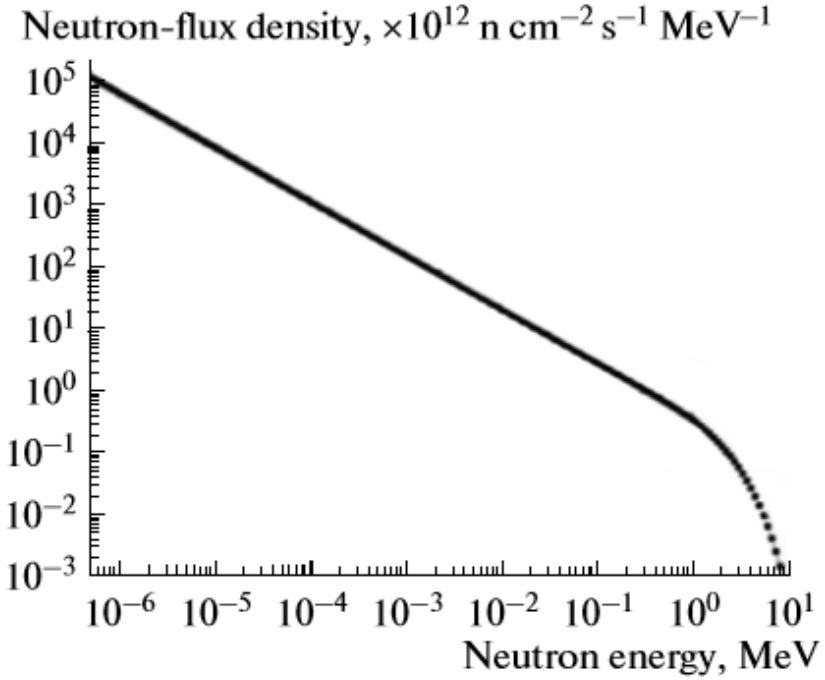


Fig. 5. Irradiation facility at IBR-2.

1.3 Neutron energy spectrum and flux densities in channel № 3

Neutron energy spectrum and flux densities were measured by method of the neutron activation analysis (NAA). Threshold detectors were positioned on the transport beam at 300 mm, 500 mm and 3150 mm distances off the outer surface of the neutron moderator. The measurements of the induced activity of the the

threshold detectors are giving possibility to get differential energy density of the neutron flux (DEDF) for the energy ranges E > 1 MeV and E < 0.1 MeV



(Shabalin E. et al, 2015).

Fig. 6. Neutron spectrum of IBR-2

The next three figures show the depencence of the neutron flux from the distance from the moderator for fast, thermal and resonance neutrons.

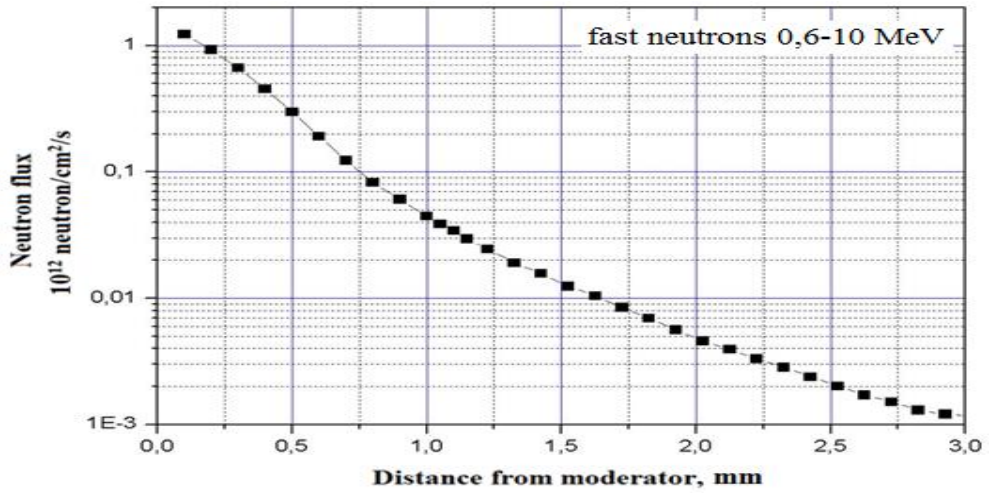


Fig. 7. Dependence of the neutron flux from distance from the moderator for fast neutrons

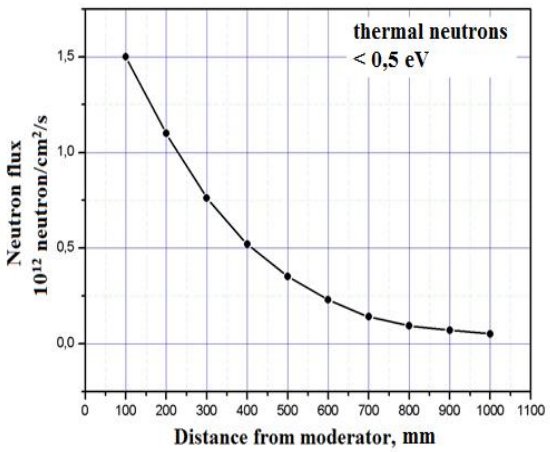


Fig. 8. Dependence of the neutron flux from the distance from the moderator for thermal neutrons.

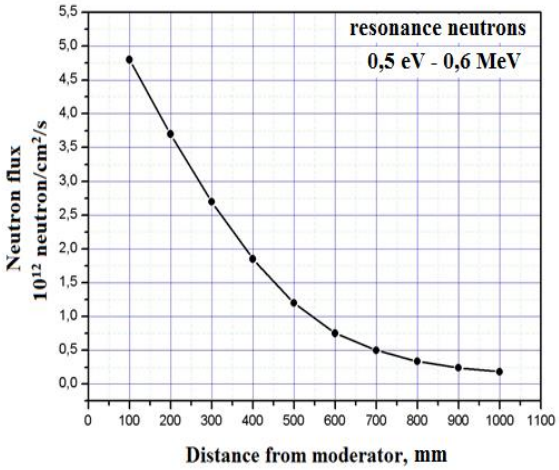


Fig. 9. Dependence of the neutron flux from the distance from the moderator for resonance neutrons.

The neutron spectrum was also measured at 3.125 m distance from the reactor moderator surface – at the point where the transport beam was fixed to the irradiation facility butt end. For such distance the spectrum becomes much harder: for the energy greater than 5 MeV neutron flux near the butt end is 40 times lower than at 0.5 m off moderator, for the middle neutron energy 1–5MeV flux decreases by a factor of 100, and a thermal neutron flux – by a factor of 170. An induced activity of Ni samples, which were positioned at different distances along the transport beam, was measured. The neutron flux intensity distribution at distances between 0.5 m and 1 m of the reactor water moderator surface is described by an exponential law. An intensity decreases by half at a distance of 18 cm. In other words, flux gradient value is around 4% per 1 cm (Bulavin M. et al, 2015).

2. Topazes

2.1 Structure and morphological characteristics

Topaz is a nesosilicate, $Al_2[(F,OH)_2/SiO_4]$ (Fig. 10), and 8^{th} on the hardness scale. It is in the orthorhombic crystal system, which is reflected in the commonly found prismatic crystal form with orthorhombic pyramid terminations. It occurs commonly in high temperature veins, probably formed by igneous intrusions in the presence of fluorine and water vapour. This results in the substitution **of** fluorine atoms by hydroxyl. In most of the cases topazes contain 15 to 20% fluorine (Barton M. et al, 1982). The composition of topaz usually ranges from a nearly OH-free endmember, $Al_2SiO_4F_2$, in acid igneous rocks, to $Al_2SiO_4F_{1.4}(OH)_{0.6}$ in hydrothermal deposits (Alberico A. et al, 2003).

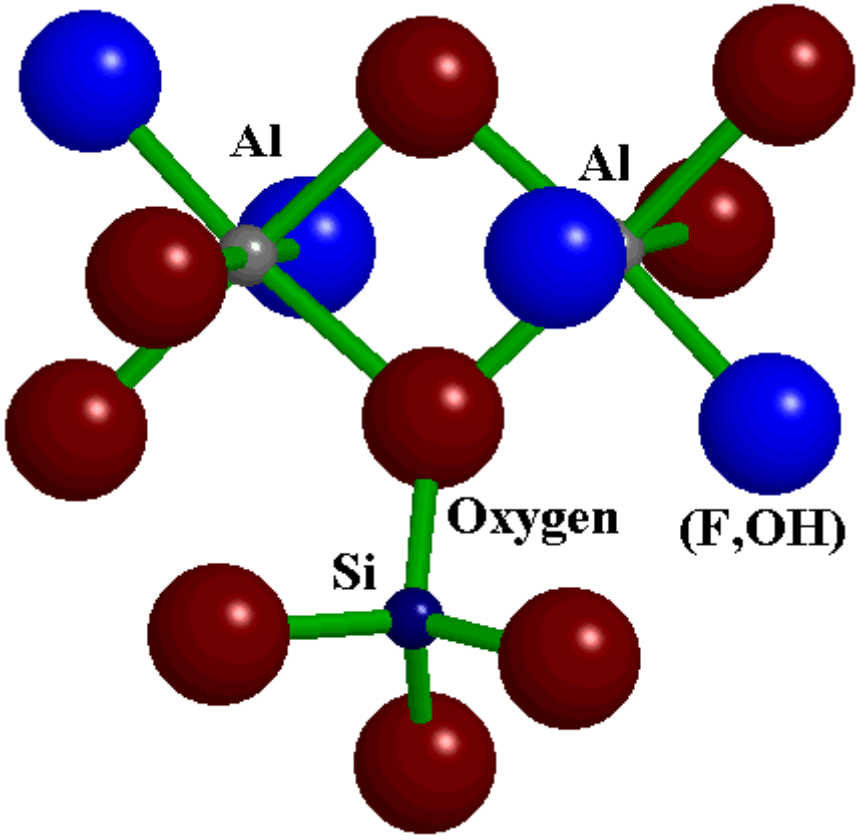


Fig. 10. The molecular structure of topazes.

The specific gravity varies from 3.4 to 3.6 and the optic axial angle varies from about 48”to about 65”with decreasing replacement of F by OH (Albuquerque A. et al, 1988).

If topazes are free of impurities, they should be colorless. In nature, however, topaz generally occurs as yellow or yellow brown, pale blue (Rossman G., 1981).

Fig. 11. A natural-colour blue topaz (left) is notably lighter than its electron-irradiated counterpart (right). Although a wide variety of yellow to brown colours occure naturally or are produced by treatment, the two shades of brown produced by a short dose of gamma rays in the stones on the bottom left provide an interesting comparison with the natural-colour yellow topaz on the bottom right.

When it is heated, yellow topaz often becomes orangy pink. Natural pink topaz, however, is extremely rare; this color is usually obtained by heating certain yellow to reddish brown Brazilian material that contains the chromium required to produce pink. Some natural yellow to brown topaz, such as is found in Utah and in some Mexican locations, fades on exposure to bright light and is therefore not used in jewelry (Nassau K., 1985). Pink and red topazes are the result of chromium replacing the aluminium in the structure, while orange varieties are caused by chromium and colour centers; blue, yellow and brown topazes all contain colour centers.

In Table 4 are shown the most commonly found impurities in topazes from South America.

Table 4. Impurities found by neutron activation in topaz samples from different regions in Brazil – State of Rodonia (TR), State of Tocantins (TT), State of Minas Gerais, Hematita (TH) and Marambaia (TM).

Element	Nuclide	Half-life	Sample			
			TR	TM	TT	TH
Ag	Ag-110	249.76d			X	
Al	Al-28	2.24m			X	
As	As-76	26.32h	X	X		
Au	Au-198	2.70d	X	X	X	
Br	Br-82	35.3h	X	X		
Ce	Ce-141	32.5d		X		
Cl	Cl-38	37.24m	X	X	X	X
Co	Co-60	5.27y	X	X	X	
Cr	Cr-51	27.7d		X		
Cs	Cs-134	2.062y		X		
Eu	Eu-152	13.33y	X	X		
Fe	Fe-59	44.4d	X	X	X	X
Ga	Ga-72	14.1h	X	X	X	
Hf	Hf-181	42.39d	X	X		
K	K-40	1.28e + 09y	X	X	X	
La	La-140	40.27h	X	X	X	
Mg	Mg-27	9.46m	X	X	X	X
Na	Na-24	14.96h	X	X	X	X
Rb	Rb-86	18.66d		X		
Sb	Sb-122	2.7d	X	X	X	
Sc	Sc-46	83.1d	X	X	X	
Ta	Ta-182	114.5d	X		X	X
Th	Th-233	22.3m	X	X		
Tm	Tm-170	128.6d		X		
U	U-239	23.47m		X		
W	W-187	23.9h	X	X	X	
Yb	Yb-175	4.19d	X		X	
Zn	Zn-65	243d	X	X	X	

From the analysis of the samples, it is shown that the blue color, induced by neutrons, is independent from the origin of the topaz and correlated with O^- defect (Leal A. et al, 2007).

2.2 Methods for coloring

In gem minerals there are two different colour mechanisms. One is related to optical transitions involving transition metals, including also charge transfer transitions between two of them or its oxygen neighbors. The other cause of colour is related to intrinsic colour centers. Two types of centers, electron and hole centers, commonly produce colour in minerals and they are produced in general by irradiation (Leal A. et al, 2007).

Upon exposure to natural radiation, most topaz will turn cinnamon brown. This color, which has some value for gemstones, can be readily generated in the

laboratory by artificial irradiation as well. A more interesting case is provided by the blue topaz that results when irradiated brown topaz from some mines is carefully heated. The fact that most properties of irradiated blue topaz are essentially the same as those of natural blue topaz has suggested the possibility that the natural color is itself a product of natural irradiation (Rossman G. et al, 1981).

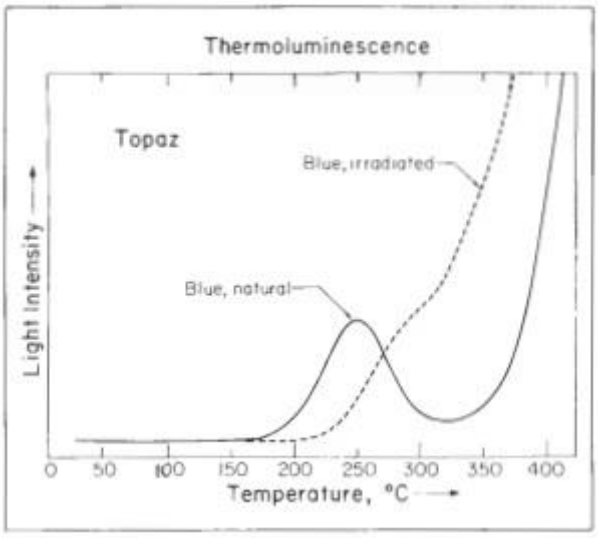


Fig. 12. Comparison of the thermoluminescence curves of natural and irradiated blue topaz. Only the hightemperature behavior is useful for distinction. The broad emission at 250°C is not present in all natural samples.

Because the properties-chemistry, color, indices of refraction, etc.-of natural and irradiated blue topaz are almost identical, the gemologist is faced with the formidable task of distinguishing the natural from the irradiated material. The developed method for making this distinction involves thermoluminescence, that is, light caused by heat. When an irradiated topaz is heated, a small amount of light will be emitted at temperatures below those that cause incandescence. The emitted light represents the energy released by the trapped electrons as they return to their most stable configuration (Rossman G. et al, 1981).

The thermoluminescence behavior of a natural blue topaz is compared to that of its irradiated counterpart in Fig. 12. With natural blue samples, the rise begins around 350°C whereas for irradiated blue topaz, it begins at lower temperatures, typically at or below 300ºC. This difference exists because in nature topaz is exposed to low levels of radiation over long periods of time, whereas the artificially irradiated material is given massive doses of radiation in a short time. Electron traps that have low to moderate stability will decay spontaneously in nature because of the long time over which the natural irradiation occurs. These same traps will be occupied in large numbers in the irradiated material. If, however, their lifetime is tens to hundreds of years they will not fade over the course of a single human lifetime. These moderately stable traps may not necessarily contribute to the color of the topaz, but they will contribute to the stone's thermoluminescence behavior at high temperatures (Rossman G. et al, 1981).

Several types of irradiation can be used to alter the color in topaz: X-rays, gamma rays, neutrons, and high-energy charged particles such as electrons, protons, and the like. Some of these are not in common use: X-rays of the usual low energies have only a very shallow penetration, and highenergy particles other than electrons are more costly to generate and provide no advantage over electrons (Nassau K. et al, 1985).

2.2.1 Irradiation with gamma rays

Gamma rays are produced within a gamma cell, a device containing a quantity of a radioactive material, such as the mass 60 isotope of cobalt (^{60}Co),which emits these rays. The rays are very penetrating and will produce uniform coloration if the material is uniform. Relatively little heat is generated during this process and this heat is produced uniformly throughout the specimen, so that cracking is not a problem; to avoid excessive temperatures, the rate of irradiation is lzept at a reasonable level, usually less than 5 megarads per hour, depending on the size of the specimen (Nassau K. et al, 1985).

When colorless or pale-colored topaz is exposed to gamma rays, a color in the sequence yellow to brown to reddish brown to very dark brown is usually produced, with significant color already appearing at quite low radiation doses (e.g., less than one megarad of ^{60}Co).Because of variations in the nature of the topaz (impurities and other defects), these colors are frequently not uniform and will vary even among zones within a single crystal (Nassau K. et al, 1985).

Fig. 13. This 6-ct topaz illustrates the "steely" blue color that is often producedin stones subjected to gamma or neutron irracliation.

2.2.2 Irradiation with high energy electrons

Electrons are accelerated to high energies in a variety of machines, including linear accelerators (linacs),Van de Graaff generators, and betatrons, among others. After reaching the selected energy, the beam of electrons is electrically deflected in a zig-zag pattern to cover an area, typically a few to many centimeters across, or a sample container is moved in such a way as to expose the whole specimen holder to the electrons. Such high-energy-electronfacilities are large, complex, expensive to build, expensive to operate, and must be well shielded, hence the higher cost of electron irradiation versus gamma irradiation. For the coloration of blue topaz, irradiation energies in the 10 to 20 mega electronvolt range are most commonly used (Nassau K. et al, 1985).

High-energy electrons act quite differently from gamma rays. They produce considerable heat, with much of the heat generated at the surface of the specimen. The samples are usually cooled with cold running water during the radiation procedure; even so, cracking is common if certain inclusions or defects are present, and melting can occur if the water supply is interrupted or the beam of electrons remains fixed in one spot. A large amount of negative electricity is also carried by the beam into the specimen, and an internal electrical discharge or "internal lightning" (also referred to as "treeing effect" or "Lichtenburg figure" in other contexts) can occur as shown in Fig. 14, and may cause severe damage. If the energy used is high enough, most of the beam can be made to pass through the specimen to avoid damage from this effect (Nassau K. et al, 1985).

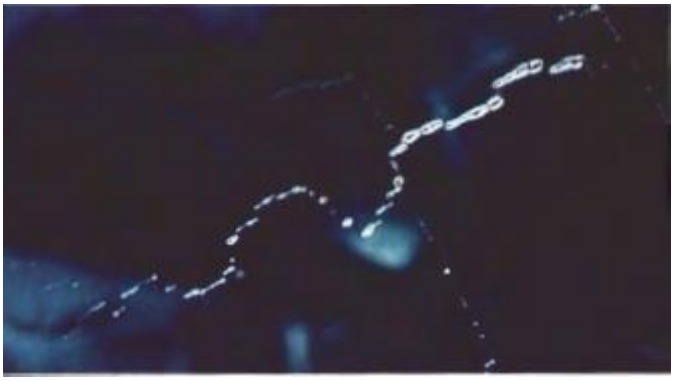


Fig. 14. Internal lightning, caused by internal electrical discharge during irradiation in linear accelerator, is evident when this 52.3-ct blue topaz is viwed through with 3 x magnification.

2.2.3 Irradiation with neutrons

Neutrons, produced in nuclear reactors, can also induce radioactivity in all but the purest of topaz crystals. However, they have excellent penetration, so there are no surface heating or coloration problems, and the colors produced are usually uniform and deep. Because there is no risk of cracking, size is not the problem it is with high-energy electron irradiation. The neutrons in a nuclear

reactor can be of varying energy and are also accompanied by gamma rays and other rays and particles. By placing the material to be irradiated into a cadmium-lined iron container, the thermal neutrons that do essentially all of the activating are absorbed by the metals, which then also generate additional gamma rays. To use neutron irradiation, however, the treater must have access to a reactor facility that is able to handle the very high radioactivities involved with the special cadmium-lined iron container. Doses of up to 1,000 megarads are said to be adequate to produce, after heating, a deep blue. The color may be darker than that produced by electrons, often "inky" or "steely." Zoning of color can be expected to be similar to that observed with other irradiation techniques used for the blue product (Nassau K. et al, 1985).

3. Experiments

3.1 Radiation colouring research of topaz samples at the irradiation facility of the IBR-2 reactor

Three groups of naturally colorless topazes were irradiated in the zone of channel № 3 of the IBR-2 reactor, in a modified version of the installation existing at this site, which makes it possible to study the radiation properties of materials in a wide range of energies and processes of neutrons and gamma quants. The irradiation unit is surrounded by biological protection, which protects the personnel during operation from ionizing radiation. In the zone of protection close to the active zone, there is a rectangular opening with dimensions of 20 cm × 40 cm for the output of a neutron beam. In the second wall of biological protection in the zone of the 3rd channel there is a circular hole 800 mm in diameter for the introduction of the irradiation unit into the annular corridor. Samples for irradiation were placed in a metal container with transverse dimensions not exceeding 160 mm × 160 mm, made of an aluminum alloy AMG6 (Fig.15). The main reason for the choice of the material is explained by the fact that after irradiation its gamma activity decreases rapidly. The container is attached to the end of the transport I-beam, also made of AMG6 alloy. The transport I-beam is connected to the massive part of the irradiation unit by means of a collapsible flange connection. The irradiation unit is moved along the track using an electric drive mechanism. Relocation of the facility is limited by end switches and emergency stops (ES) located on the both sides of the tracking, as well as visible mechanical stop (VMS) which regulates the

distance from the samples to the surface of the IBR-2 water moderator from the side of area of the beam 3 in the experimental chamber.

The period of irradiation depends on the required neutron fluence and varies from a few hours to 11 days (the common duration of the session of the IBR-2 reactor activity). The temperature in samples at irradiation is below 50°C which partly is supplied by the exhaust system of ventilation. The maximum value of neutron fluence with energy of more than 1MeV at arranging the sample at a distance of 40mm from the moderator is $4 \cdot 10^{17}$ n cm^{-2}, while installing the sample at a distance of 40mm from the moderator – 10^{18} n cm^{-2}, the minimum neutron fluence – 10^{15} n cm^{-2}.

The neutron fluence control is carried out by means of replacing activation foils next to the irradiation materials with the succeeding measurement of induced activity. The maximum absorbed dose in water while installing the sample at a distance of 40mm from the moderator composes 100MGr (108 J/kg), and at a distance of 30mm – 40 MGr.

In the steel cylinder of the massive part of the irradiation facility bent pipes are placed which allow to lay power supply cables and to display signals out from the facilities to be monitored during the assembly of the given dose. The maximum value of the absorbed dose of gamma-quants corresponds to about 60 kSv/hour (at fast neutron flux - about 10^{12}n cm^{-2} sec^{-1}), the minimum value of the absorbed dose – less than 400Sv/hour.

The possibility of coloring of nature topazes into blue color at neutron irradiation on the IBR-2 reactor is the key of the current paper.

Studies have been carried out on 15 (unpolished) colorless (except the middle range) samples of nature topazes of Brazilian, Mozambique and Ukrainian fileds (Fig. 15).

Before the irradiation of samples neither analysis nor testing has been carried out.

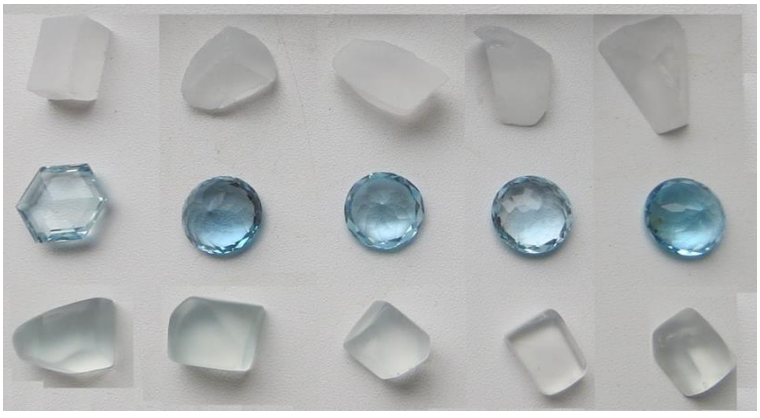


Fig. 15. Samples of the nature topaz. The upper range – of the Mozambique origin, the middle range – of the Ukrainian origin and the lower range – of the Brazilian origin.

The topaz samples with a size of about 1,5cm × 1,5cm × 0,5cm are installed at a distance of 210mm, 350mm, 1030mm, 1530mm and 2040mm from the moderator. The irradiation of samples was carried out in the VII cycle in 2016 in a water moderator mode with duration of 411 hours (17days). The average power of the reactor within the cycle is 1875kVT.

At neutron irradiation both simple and complex defects occur (defect complexes) in the crystalline structure of minerals. Atoms develop sample defects shifted from the site of the lattice. They are produced at energy of neutrons in dozens of electron-volt and provide cloud of crystal, yet they are easily burnt via heat treatment. Defect complexes depend on density of simple defects they are developed from. Accordingly, the higher the density of simple defects is, the more possible the development of complex ones is. For this reason, the possibility of development of complex defects increases with the increase of energy in neutrons and reaches saturation in defect production at fast neutron density with energy of over 1MeV. That is the defect complexes to develop electronic and hole-type colouring centers.

The induced activity of topazes is the key problem, i.e. the occurrence of radioactivity destroys the consumption value of these minerals. The neutron flux in the nuclear reactor can be of various energies, gamma rays and other particles are also associated with it. The induced activity is measured by the availability of impurities in the structure of topaz samples and neutron flux in which the treatment took place. In order to essentially decrease the activity occurred by thermal and resonance neutrons (and to increase the number of productive colouring centers of fast neutrons), a filter consisting of powder Boron was used.

The neutron fluence with energy of over 1MeV at installation of the samples of topaz 5 at a distance of 2,04m from the moderator composed $3{,}9 \cdot 10^{15}$ n cm^{-2} (at flux density - $2{,}6 \cdot 10^{9}$ $n/cm^2/c$). The impact of fast neutron flux resulted in the development of colouring centers in topazes (Fig. 16). The report is shown on Table 5 which presents the comparison of fast neutron flux measurements with energy of over 1 MeV in topaz samples with irradiation duration (17 days). The current values were acquired by the method of neutron activation analysis in FLNP JINR.

Table 5. Report on measurements of fast neutron flux density with energy of n^0 > 1 MeV, measured by the method of neutron-activation analysis with usage of nickel satellite.

Sample	Distance from the moderator, mm	Flux density, **n/cm²/c**	Fluence, **n/cm²**
№1 Topaz	210	$9{,}5\cdot10^{11}$	$1{,}4\cdot10^{18}$
№2 Topaz	530	$9{,}4\cdot10^{10}$	$1{,}4\cdot10^{17}$
№3 Topaz	1030	$2{,}2\cdot10^{10}$	$3{,}3\cdot10^{16}$
№4 Topaz	1530	$8{,}3\cdot10^{9}$	$2{,}2\cdot10^{16}$
№5 Topaz	2040	$2{,}6\cdot10^{9}$	$3{,}9\cdot10^{15}$

On fig. 16 is shownthat the faster neutron flux density is, the better the color specification of topaz becomes. The important colouring centers were developed in the samples of topaz 1 after the irradiation with neutron fluence $1{,}4\cdot10^{18}$ n cm^{-2} at installation of samples at a distance of 0,21m from the moderator (at flux density of $9{,}5\cdot10^{11}$ n/cm²/c).

Fig. 16. The irradiated samples of topazes (from left to right).

3.2 Research of the heat treatment impact

In the used samples the irradiation developed certain colouring centers simultaneously, and especially the required blue and the unrequired brown ones. It's possible to get rid of the unrequired brown centers of colourings via heat treatment. It's well-known, that the heating of the coloured topaz can make it either change its color or return to its former colourless state. Such as for instance, annealing yellow topazes up to brown color is possible at heating them up to 200-300 °C, and at heating up to 500 °C within several hours the natural

blue topaz can be turned into colourless one. In general, colors disappear at heating from 400 ºC up to 750ºC. Various processes of irradiation and heat treatment, applied nowadays, are summed up on fig. 17.

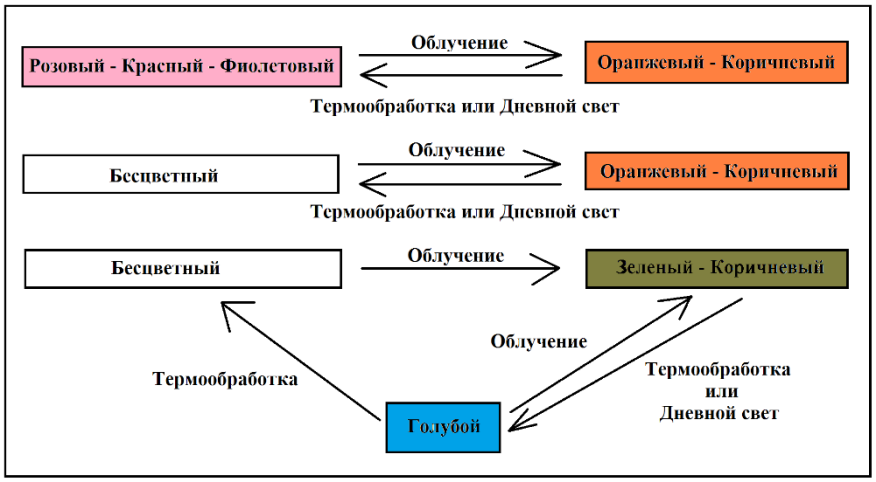


Fig. 17. The color change of topazes at treatment with irradiation and/or annealing (in some cases at exposure of daylight).

In order to get rid of unrequired brown centers, the theoretical data mentioned above were applied. In the result of isothermal annealing (i.e. time-temperature treatment in certain period) of irradiated samples of topazes at 500ºC with duration of 3hours, the great part of samples became colorless (Fig. 16).

As it is shown on Figure 18, the samples of topaz 1 (after the irradiation with fluence of neutrons $1{,}4 \cdot 10^{18}$ n cm^{-2}) acquired light-blue color with respect to the annealing of brown centers of colouring (radiation defects). A majority of topazes are very sensible to high temperatures and they are quite often destroyed, broken; even at temperatures up to 200ºC the thermal annealing is applied in a very slow and careful way. Especially, occurrence of thresholds makes the samples apt to destroying; the so-called samples under research of Mozambique origin were destroyed in the result of heat treatment.

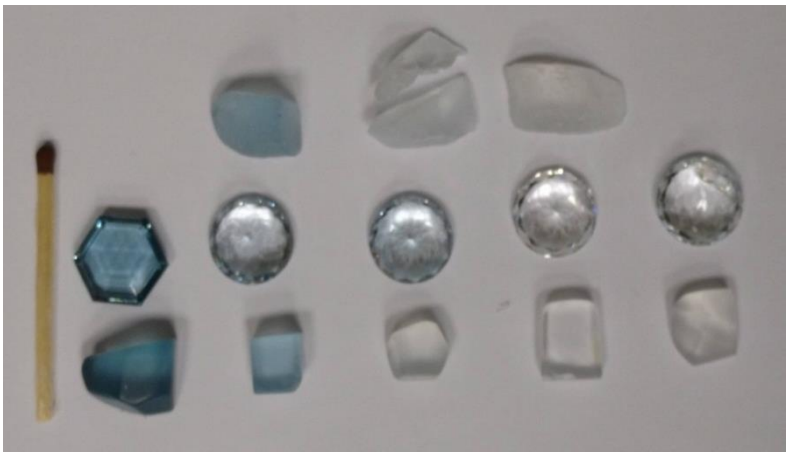


Fig. 18. Topaz samples after the heat treatment (from left to right).

3.3 Induced activity in topaz samples

The period of half-decay of the main elements of topaz O,F, Al and Si at radiative neutron capture composes 29,11.4,134,4 and 9432 second correspondingly. That means the period of half-decay of the main elements of topaz is quite short. Consequently, such kinds of jewels are worth to be used as much time for the decay of their persisting radioactivity isn't needed. The present conclusion is actual in the case when samples don't possess high concentrations of impurity elements which have been in half-decay for a long period of time. Yet, topaz, as a rule, quite frequently consists of various impurities. It's worth pointing out that occurrence of this or that impurity element depends on the localities of topaz; what refers to the content of topazes of Brazilian origin, ^{59}Fe and ^{182}Te are included in it as compounds. After the irradiation, the samples of topazes require long-term scintillation for elimination of induced activity.

Table 6. The half-decay period of impurity elements of irradiated topazes.

Radioactive isotopes	The half-decay period (days)
^{51}Cr	27,7
^{59}Fe	44,51
^{46}Sc	83,81
^{182}Ta	114,4
^{65}Zn	243,8
^{54}Mn	312,2
^{134}Cs	753,7
^{60}Co	1924

In order to essentially decrease the induced activity of samples conditioned with thermal and resonance neutrons which are produced in active volume on account of delay of fast neutrons, substances or mixture of substances are used such as cadmium and boron (in our case, boron).

4. Conclusions

1. With these experiments it was stated that treatment with neutron fluence $1{,}4 \cdot 10^{18}$ led to changes in colouring of topaz samples under study from transparent to blue which increased the jewel value of the mineral in 2-4 times.
2. Irradiation, besides increasing the intensity of colouring of samples, simultaneously increased their activity too, which accordingly increased the period of the required endurance.

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