STUDY OF NEUTRON IRRADIATION-INDUCED COLOR IN TOPAZ AT THE PULSED REACTOR IBR-2

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Abstract

In the present work, a method of a blue topaz production at the reactor IBR-2 is studied. To obtain deep blue color the fast neutron fluence needs to be about 10^{18} cm^{-2}. The optimal sample position in the channel №3 was determined. The special irradiation containers filled with boron carbide were created to reduce activation by thermal and resonance neutrons.

1. Introduction

Most of the natural topazes $Al_2SiO_4(F,OH)_2$ are colorless. To enhance these gemstones various types of radiation are widely used [1]. The irradiation forms the color centers in the crystal structure which change the mineral's color and, thereby, increase its consumer value. Reactor neutron irradiation is commonly applied to create an attractive deep blue color, called «London Blue», in the almost colorless topazes (Fig. 1). The production of blue centers is established to be caused by fast neutrons, while thermal and resonance neutrons lead to induced radioactivity of trace-element in topaz. This induced radioactivity doesn't allow using topazes in jewelry immediately after irradiation, because it can harm the health. The irradiated topazes can be used only after several years, when the radiation level will decrease to appropriate levels. Fast reactor IBR-2 makes it possible to achieve effective color modification with a minimum of induced activity.

This work is devoted to the optimization of the irradiation conditions in the channel №3 of the reactor IBR-2 [2] for production of «London Blue» topazes. In addition, the mechanism of blue centers formation were investigated by the optical absorption and Raman spectroscopy methods.

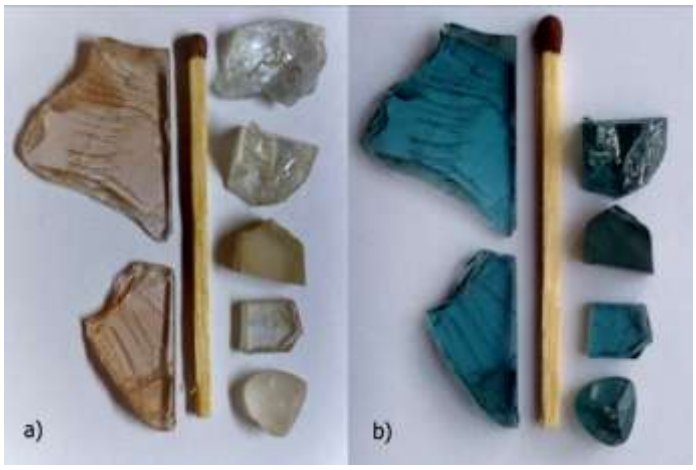

Fig. 1. Color changes of Ural topazes before (a) and after (b) neutron irradiation at the reactor IBR-2. Fast neutron fluence was about 10^{18} cm^{-2}.

2. Experiment

Topazes of different deposits were irradiated by neutrons at the reactor IBR-2. Samples were placed on the I-bar in the research channel №3 in the aluminum capsules (Fig. 2).

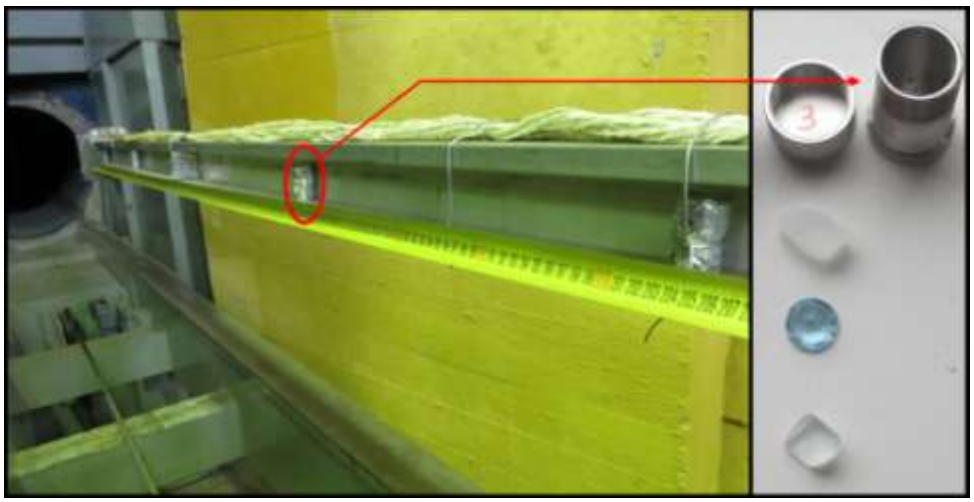

Fig. 2. Capsules for samples in the research channel №3 at IBR-2.

Fluence value was varied by positioning the samples at different distances from the moderator. Fast (>1 MeV) fluence value was measured by the method of the neutron activation analysis. Threshold detectors based on the reaction $^{58}Ni(n,p)^{58}Co$ were positioned near each samples group. The reactor operation cycle was 264 hours. The activity of irradiated samples was analyzed using gamma spectrometer with HPGe detector (Canberra) with relative efficiency of 40 %, an energy resolution of 1.8 keV at 1332 keV for ^{60}Co. The annealing was carried out in a furnace in air.

3. Results and discussion

Sample positions in channel №3 and the color changes depending on the fast neutron (>1 MeV) fluence value are presented in Fig. 3.

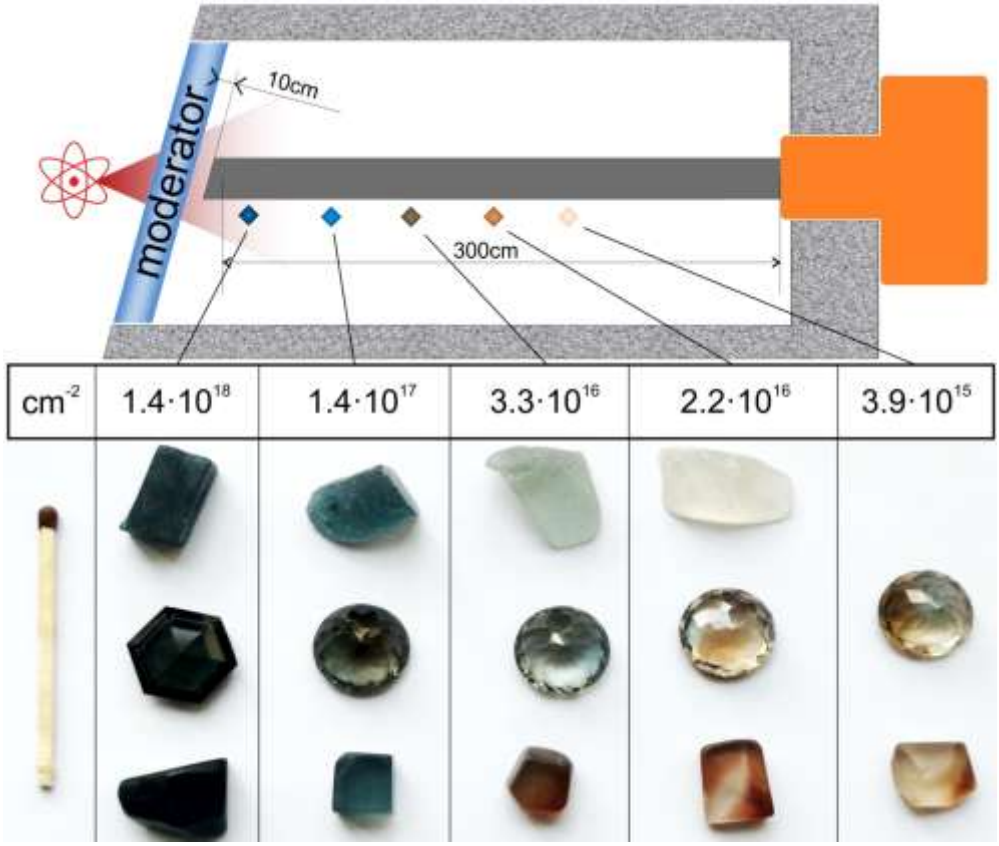

Fig. 3. Sample positions in channel №3 and the color changes depending on the fast neutron (>1 MeV) fluence value.

It can be seen that «London blue» color can be obtained with the fast neutron fluence of about 10^{18} cm^{-2}. This fluence value is reached at the I-bar end.

Some samples have a brown tint which is easy removed by heating in air at the temperature (200÷300)°C (Fig. 4). Various annealing modes at temperatures above 500 °C give any shade of blue color (the optical absorption band is centered at 620 nm). Achieved color remains stable at room temperature for a long time.

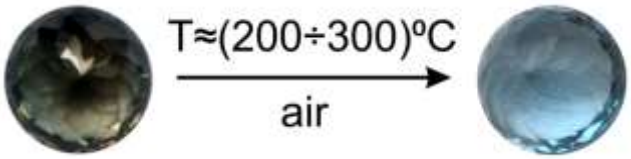

Fig. 4. Removing a brown tint by heat treatment.

Gamma-ray spectroscopy detected the presence of different radionuclides in the irradiated topazes. To reduce activation the special irradiation containers were created. The containers sides were made of boron carbide placed between two aluminum layers (Fig. 5). These containers significantly reduced the residual activity level. The residual activity values of two Ural samples at the end of irradiation are stated in Table 1.

Table 1. Residual activity of two Ural samples at the end of irradiation.

Nuclide	**Half-life**	**Residual activity (Bq/g)**	
		Sample №1	**Sample №2**
^{134}Cs	753.6 d	0.51±0.04	8.17±0.13
^{54}Mn	312.3 d	14.53±0.08	49.58±0.28
^{110m}Ag	249.79 d	0.27±0.04	9.85±0.18
^{65}Zn	244.26 d	8.70±0.09	42.06±0.37
^{182}Ta	114.43 d	22.94±0.25	59.58±0.79
^{46}Sc	83.79 d	28.27±0.11	39.91±0.24
^{58}Co	70.86 d	1.52±0.03	11.53±0.14
^{160}Tb	72.3 d	8.88±0.13	97.09±0.69
^{124}Sb	60.2 d	2.94±0.10	19.60±0.42
^{59}Fe	44.5 d	11.28±0.13	37.88±0.43
^{181}Hf	42.39 d	3.39±0.04	42.22±0.22
^{141}Ce	32.5 d	3.36±0.03	23.07±0.17
^{51}Cr	27.7 d	35.92±0.29	281.79±1.38
^{233}Pa	26.96 d	70.59±0.19	798.57±1.12
^{86}Rb	18.63 d	5.71±0.52	1267.60±5.85
^{122}Sb	2.72 d	37.94±0.87	251.87±2.65
^{198}Au	2.69 d	11.45±0.39	4041.18±6.55
^{239}Np	2.35 d	906.7±3.69	13107.59±18.00
^{140}La	1.67 d	269.08±7.40	2152.36±23.72
^{99}Mo	65.94 h	2.69±0.19	85.36±1.76
^{153}Sm	46.28 h	2516.38±9.27	27414.81±37.09
^{24}Na	14.95 h	(7.07±0.10)E+06	(8.44±0.07)E+06

Fig. 5. The irradiation containers filled with boron carbide between aluminum layers

To optimize this method, we need to investigate the mechanism of blue centers formation. The preliminary results of Raman spectroscopy showed intensity decreasing of peaks, associated with the OH stretching mode (3650 cm^{-1}), after neutron irradiation. Taking into account that natural blue topazes have a low OH-content [3], we can suppose that the blue centers formation is caused by collision mechanism of hydroxyl destruction by fast neutrons. The same effect is observed with an electron beam irradiation, but it is much less pronounced.

4. Conclusion

The optimal irradiation conditions for blue topazes production were determined: To obtain «London blue» color the fast neutron fluence needs to be about 10^{18} cm^{-2}. This fluence value is reached at the I-bar end in the channel №3 of the reactor IBR-2 with operation cycle of 264 hours. The containers with double sides filled with boron carbide significantly reduce the residual activity level. Based on the preliminary results of Raman spectroscopy we can suppose that the formation of blue centers is caused by collision mechanism of hydroxyl destruction by fast neutrons.

References

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