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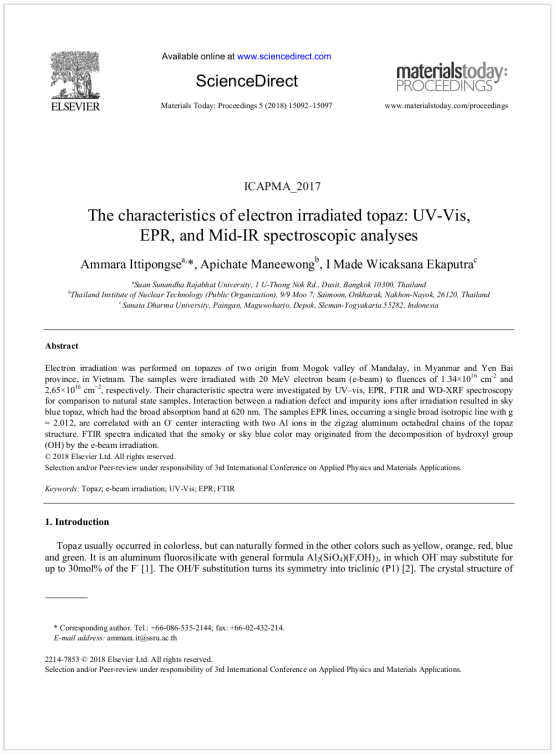

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The characteristics of electron irradiated topaz: UV-Vis, EPR, and Mid-IR spectroscopic analyses

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Abstract

Electron irradiation was performed on topazes of two origin from Mogok valley of Mandalay, in Myanmar and Yen Bai province, in Vietnam. The samples were irradiated with 20 MeV electron beam (e-beam) to fluences of 1.34×10^{16} cm^{-2} and 2.65×10^{16} cm^{-2}, respectively. Their characteristic spectra were investigated by UV–vis, EPR, FTIR and WD-XRF spectroscopy for comparison to natural state samples. Interaction between a radiation defect and impurity ions after irradiation resulted in sky blue topaz, which had the broad absorption band at 620 nm. The samples EPR lines, occurring a single broad isotropic line with g = 2.012, are correlated with an O$^-$ center interacting with two Al ions in the zigzag aluminum octahedral chains of the topaz structure. FTIR spectra indicated that the smoky or sky blue color may originated from the decomposition of hydroxyl group (OH) by the e-beam irradiation.

Selection and/or Peer-review under responsibility of 3rd International Conference on Applied Physics and Materials Applications.

Keywords: Topaz; e-beam irradiation; UV-Vis; EPR; FTIR

1. Introduction

Topaz usually occurred in colorless, but can naturally formed in the other colors such as yellow, orange, red, blue and green. It is an aluminum fluorosilicate with general formula $Al_2(SiO_4)(F,OH)_2$, in which OH may substitute for up to 30mol% of the F [1]. The OH/F substitution turns its symmetry into triclinic (P1) [2]. The crystal structure of

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by Ekaputra I Made Wicaksana

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topaz is belong to the orthorhombic with space group Pbnm (D_{2h}^{16}) [3] and four molecules per unit cell. It's structure consists chains of a pair of edge-shared AlO_4F_2 octahedra and corner-shared SiO_4 tetrahedra that linking octahedral chains in a zigzag fashion parallel to the crystalline c-axis. Topaz is most commonly irradiated with a variety of radiations. Sky blue color, a light pure blue color, can produce by exposure the white topaz to gamma rays and continue by heating it to burn off undesired brown overtones that appear during the exposure process. On the other hand, if exposure the white topaz to neutrons in a research reactor and /or electrons in a linear electron accelerator will produce the darker shades which we know as London blue and Swiss blue.

The coloration of topaz is frequently governed by transition metal impurities or by irradiation. Many authors has been attributed the blue color to an O^- defect, produced by irradiation and located in OH^- positions of topaz structure, interacting with two equivalent structural Al ions [4-5]. The O^- defect is related to the absorption band at 620 nm. Some authors have been ascribed this absorption band to the presence of Cr^{3+}, Fe^{2+} and Mn^{2+} impurities [6-7]. Irradiation treatment caused exchange interaction between these impurities and radiation defect. The chemical composition, different valence states, and occupancy in each site of the topaz structure appear to be associated with one another in the color change caused by irradiation treatment. However, disagreement exists over the detailed origins of the colors. Specifically, the reason for the color changes after e-beam irradiation is still unclear and not well understood.

In the present work, we used Ultraviolet–visible Spectroscopy (UV-Vis), including Wavelength Dispersive X-ray Fluorescence (WDXRF), Electron Paramagnetic Resonance (EPR) measurement and Fourier-transform infrared (FTIR) spectroscopy analyses to study the chemical behaviours of two topaz samples, one originated from Mogok valley of Mandalay, in Myanmar and the other from Yen Bai province, in Vietnam, by e-beam irradiation with two different fluence levels to understand the color change mechanism. We discuss the color change mechanism in relation to e-beam irradiation in terms of the initial point of the jewellery manufacturing process.

2. Materials and methods

Natural colorless topaz of two origin from Mogok valley of Mandalay, in Myanmar and Yen Bai province, in Vietnam were investigated. Each sample was cut in half perpendicular to the crystallographic c-axis. Subsequently, all samples are polished by silicon carbide abrasive paper. After cutting and polishing, the samples thickness was in the range of 2.1 –3.2 mm. The samples were irradiated with 10 kW power and 20 MeV electrons to fluences of 4×10^{16} cm^{-2} and 2.65×10^{16} cm^{-2}, respectively. The linear electron accelerator used in this study is located at Gems Irradiation Center, Thailand Institute of Nuclear Technology (TINT) and can produce high energy 5-20 Million Volts (MeV) and high power 10-20 kilo Watt (kW). The chemical compositions of the samples were recorded by using the S8 TIGER high-end wavelength dispersive X-ray fluorescence (WDXRF) spectrometer. The spectrometer was installed at the TINT's Service Center. The samples were excited by X-rays (Rh tube) with the power of 50 kW and the current of 50 mA. The optical absorption spectra were measured on the platelets oriented perpendicular to the c-axis before and after irradiation by using the PerkinElmer lambda 750 UV/Vis/NIR spectrophotometer located at the Gems Irradiation Center, Ongkharak Branch, TINT. The wavelength for the optical spectra collected was recorded from 300 to 800 nm by setting a spectral resolution of 2.0 nm with a scan speed of 250 nm/min. All EPR measurements were performed at room temperature on a Bruker EPR spectrometer (ESP 300 series) operated at an X-band microwave frequency at Department of Physics, Sanata Dharma University. The spectrometer's operating conditions adopted during the experiment were a 350.0-mT central magnetic field, a 140- to 600-mT scan ranges, a 9.64-GHz microwave frequency, a 1.0-mW microwave power, a 100-kHz field modulation frequency, a 1.0-mT field modulation amplitude and a 0.02-s time constant. 1, 1-diphenyl-2-picrylhydra-2l (DPPH) with a g factor of 2.0036 was used as an internal standard for g-factor calculations. After the spectra had been measured, the positions of the EPR signals were labelled by using their effective g (g_{eff}) values. The g_{eff} was calculated by using the relationship, $g_{eff} = h\nu/\beta H$, where h, ν, β, and H are Planck's constant, the microwave frequency, the electron Bohr magneton and the external field, respectively. The line width (ΔB_{pp}) of each signal was also observed. The mid-infrared spectra of the samples were recorded in the region of 400-5000 cm^{-1} by using a Bruker ALPHA Spectrometer with a resolution of 4 cm^{-1} at the Department of Mineral Resources under the Ministry

of Natural Resources and Environment, Thailand. Band fittings were done with Gauss- Lorentz functions by using the fitting program OriginPro 2015.

3. Results and discussion

The chemical analyses of a given samples obtained by WDXRF were listed in table 1. The WDXRF analysis indicated that both samples were topaz pegmatite. The analyzed topazes reveal a low Fe content and minor amounts of Na, Mg, Br, and Rb, while Cs are present about two time (0.69wt%/0.27wt%) higher in the topaz originated from Mogok valley (M) than the topaz from Yen Bai province (V). In M also present minor quantities of Ni and Ti higher than in V. Other contents, such as Cr and Cu, were detected in subordinate concentrations. There is no significant difference of both samples. In the same way, the correlation of color is not related to chemical composition.

Table 1 Chemical composition of the two topaz samples as measured by using WDXRF analyses.

	Mogok valley (M)	Yen Bai province (V)
Oxide: wt%		
Al_2O_3	59.47	59.49
SiO_2	37.47	37.40
Na_2O	0.97	1.44
MgO	1.16	1.30
Fe_2O_3	0.02	0.04
Br_2O	0.08	0.01
Rb_2O	0.04	0.03
Cs_2O	0.69	0.27
Oxide: wt ppm		
CuO	41	40
Ni_2O_3	413	33
TiO_2	133	52
Total	99.96	99.98

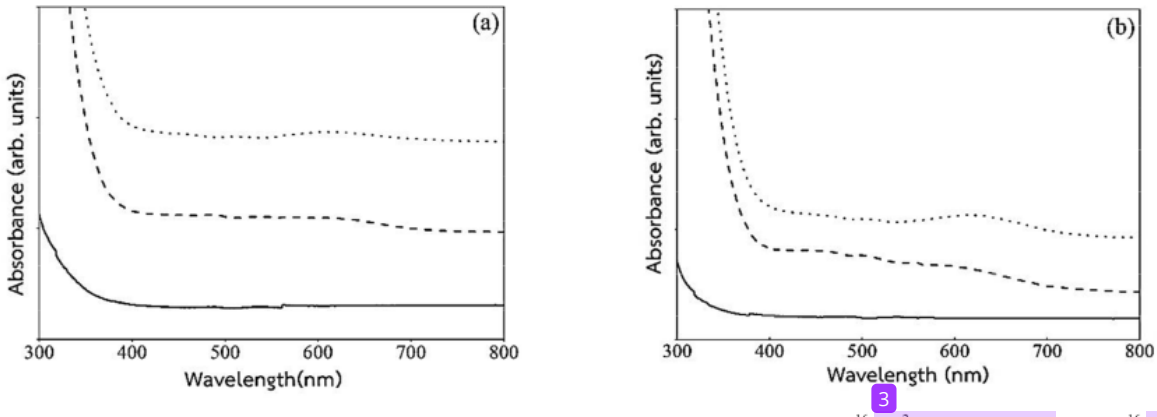

Fig. 1. UV-Vis spectra of (a) M and (b) V: natural state (solid line), after e-beam irradiations at 1.34×10^{16} cm^{-2} (dashed line) and 2.65×10^{16} cm^{-2} (dotted line).

The UV-Vis absorption spectra (E⊥c) of the colorless topaz before irradiation (solid line), after e-beam irradiations at $1.34x10^{16}$ cm^{-2} (dashed line) and $2.65x10^{16}$ cm^{-2} (dotted line) are given in Fig. 1. The natural colorless topazes exhibited no absorption band in both specimens. After e-beam irradiation at $1.34x10^{16}$ cm^{-2}, both samples appear a broad band with maxima at 620 nm and this band increased the intensity, after add the level of e-beam irradiation up to $2.65x10^{16}$ cm^{-2}, resulting in the crystals obtained pale (sky) blue color.

The blue color in topaz can produced by exposure the colorless topaz to gamma ray, neutron and electron. The detail origin of blue color by each irradiation method are difference. The e-beam irradiation lead to appearing of broad band at 620 nm ascribed to the presence of Cr^{3+}, Fe^{2+} and Mn^{2+} impurities by many authors [6, 7]. Because both topazes have no Mn ions (Table 1) and any other absorption peak for Cr^{3+} and Fe^{2+}, one possible reason for the appearance of the absorption band at around 620 nm may be the O^- defect, as suggested in a previous work based on the results obtained by using e-beam irradiation [8].

Fig. 2 shows the EPR spectra of both e-beam irradiated topazes for the magnetic field perpendicular to the c-axis. The paramagnetic defects are observed in the same position after electron irradiation. The spectra exhibited an intense broad resonance signal at g ~ 2.012 with peak-to-peak line width of ~14.20 mT. This resonance signal is the O^- hole center, which has been identified as an O^- ion on an $(OH)^-$ site having resolved hyperfine interactions with two equivalent nearest Al^{3+} ions [9]. After e-beam irradiation, the both topazes shows the peroxyl radical O_2^- [10, 11] responded to the broad isotropic resonance line. It is observe that the EPR lines at g ~ 2.012 increased the intensity when the electrons fluence level rising up.

The OH stretching and bending mode of topaz response in three regions, 600-1200 cm^{-1}, 2500-3800 cm^{-1}, 4700-4900 cm^{-1}, of mid-infrared (Mid-IR) spectra. In the 600-1200 cm^{-1} region are occasionally appears for the OH bending mode in crystal together with OH stretching mode [12]. However, the absorption peaks of OH bending mode in minerals are difficult to assign, due to this wavenumber region is overlapped by Si-O or Al-O fundamental mode of minerals. Next, the OH stretching modes are produced strong absorption in the 2500-3800 cm^{-1} region [13]. The final region, 4700-4900 cm^{-1}, is assigned to a combination of OH stretching and bending modes [14].

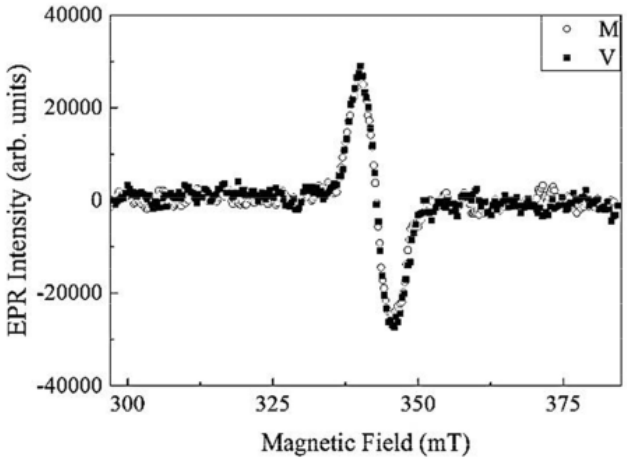

Fig. 2. EPR Spectra of M (circle-symbol line) and V (square-symbol line).

In Fig. 3, the Mid-IR spectra of the topaz samples, M and V, obtained from the Department of Mineral Resources are shown. The spectra of all two samples in their natural states (solid line) were rather similar. M showed a negligible lower spectra intensity than V throughout the Mid-IR region. The weak bands appeared in range from 4780-4830 cm^{-1} with the peak at 4800 cm^{-1}, and strong intensity bands appeared in range from 3610-3690 cm^{-1} with the peak at 3650 cm^{-1}. The 4800 cm^{-1} absorption band is defined as combination of OH stretching and bending modes, while the 3650 cm^{-1} absorption band is assigned to OH stretching modes. The low wavenumber region which referred to OH bending mode cannot observed in both topaz samples. All samples exhibited similar

behaviors, decreasing in the intensity of absorption bands at 3650 cm^{-1} and 4800 cm^{-1} after e-beam irradiation (dashed and dotted line). The decomposing by electron of the OH^- groups into O^- (hole trap) and H^o (electron trap) resulted in a decrease of the intensity.

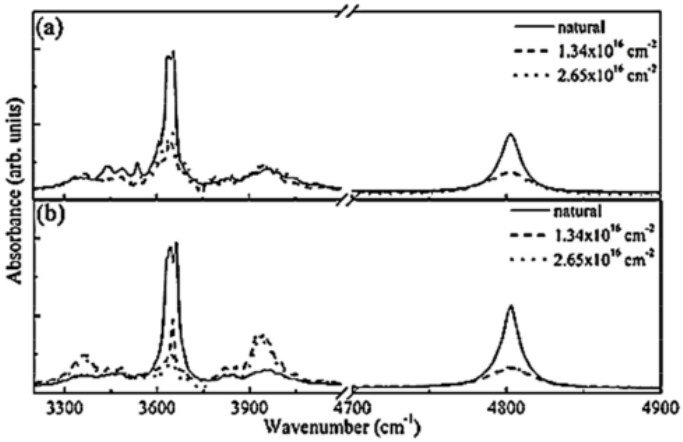

Fig. 3. Mid-infrared spectra in the 3300 – 4900 cm^{-1} region for (a) M and (b) V: natural state (solid line), after e-beam irradiations at $1.34x10^{16}$ cm^{-2} (dashed line) and $2.65x10^{16}$ cm^{-2} (dotted line).

4. Conclusion

Colorless topazes with different chemical composition changed to a pale blue color after e-beam irradiation. Therefore, the absorption bands at 620 nm were observed. This band is correlated with the creation of O^- defect by electron which supported by EPR spectra. The EPR signal at g ~ 2.012 which has been identified as an O^- ion on an $(OH)^-$ site increased the intensity when the electrons fluency level rising up. Decomposing of OH^- groups are occurred after e-beam irradiation from evidence of Mid-IR spectra. Furthermore, decomposing of OH^- groups are occurred after e-beam irradiation from evidence of Mid-IR spectra. The absorption bands at 3650 cm^{-1} and 4800 cm^{-1} after e-beam irradiation indicated OH stretching modes and combination of OH stretching and bending modes, respectively.

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