Gamma ray evaluation of fast neutron irradiated on topaz from Sri Lanka by HPGe gamma ray spectrometry

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Abstract. The purpose of this study was to evaluate the radionuclide concentrations of London blue topaz after fast neutron irradiation. The London blue topaz was obtained from Sri Lanka which classified into dark and light colors in the shape of an oval and rectangle with small, medium and large size. The optical property and radionuclide concentrations of London blue topaz have been examine by UV-Visible spectroscopy and HPGe gamma ray spectrometry, respectively. The UV-absorption spectra of topaz was taken in the range of 300 to 800 nm at room temperature. The results showed that the absorption peak of topaz was observed with only broad peaks in the range of 550 to 700 nm and 630 nm that correlated to the O - center in hydroxyl sites which substitutes for fluorine in topaz structure. The radioactivity of dark and light colors in the shape of an oval and rectangle London blue topaz was in the range of 1.437 ± 0.014 to 21.551 ± 0.037 nCi/g (oval dark), 2.958 ± 0.031 to 6.748 ± 0.054 nCi/g (oval light) and 2.350 ± 0.014 to 43.952 ± 0.088 nCi/g (rectangle dark), 1.442 ± 0.023 to 6.748 ± 0.054 nCi/g (rectangle light), respectively. The decay rates of ⁴⁶Sc, ¹⁸⁶Ta and ⁵⁴Mn isotopes created by irradiation showed that the decay time of the radioactive element depended on the size of the topaz so increased with decreasing the size of topaz. Moreover, the size of topaz also affect the absorption coefficient. This study is applied to predict time of residue dose of topaz for enhancement colorless topaz by neutron radiation treatment.

1. Introduction

Sri Lanka is one of the most popular and important producers of topaz in the world. Most of this topaz has a very low commercial value, because it is colorless. Topaz is the best example for enhancement of color by commercial application of neutron irradiation which leads to a deep blue color, called London blue topaz [1]. The neutron irradiation induces radionuclides with different half-lives, resulting to radioactivity. Therefore, blue topaz colored by neutron irradiation requires several months to allow the residual radioactivity to reach a safe level of less than 74 Bq/g or 2 nCi/g, as defined by the nuclear regulatory commission (NRC) [2]. The radioactive decay of isotopes created by irradiation topaz depends on several factors include the individual isotope, type of irradiation and cutting style of topaz [3]. Especially, color, shape and size of irradiated topaz should be restricted, because of the surface area added radiation exposure, which affects the radioactivity and decay time of topaz [4]. In this paper presents study of UV-Visible absorption spectra and radionuclide concentrations of London blue topaz from Sri Lanka after fast neutron irradiation. Topaz was classified into dark and light colors in the shape of an oval and rectangle with small, medium and large size. The optical property the radionuclide concentrations, decay time and absorption coefficient of topaz was characterized by UV-Visible spectroscopy and HPGe gamma ray spectrometry, respectively. This study is applied for time prediction
of residue dose of topaz before return to customers who have to enhance colorless topaz by neutron radiation treatment.

2. Experimental
In this work, topaz after fast neutron irradiation from Sri Lanka which classified into dark and light colors in the shape of oval and rectangle were analyzed. Topaz was prepared by cleaving and cutting large pieces to size of about 25×29×16 mm (large size), 19×29×15 mm (medium size) and 18×21×10 mm (small size) as shown in figure 1. Then all topaz was investigated by UV-Visible spectroscopy and HPGe gamma ray spectrometry, respectively. The optical absorption measurements were performed in the 300 to 800 nm wavelength range, with a Lambda 650 UV-Visible Spectrophotometer.

![Figure 1. London blue topaz in the shape of (a) an oval and (b) rectangle.](image)

The radioactivity, decay rate and self-absorption of all 12 topaz samples were investigated by HPGe gamma ray spectrometry. The HPGe detector (CanberraTM; 1.96 keV resolution at the 1332 keV photons of 60Co peak; 30% relative efficiency) was connected to a PC-based multi-channel analyzer (MCA) with preloaded filter. The detector efficiency was calibrated with standard point source of 60Co (1173 and 1332 keV), 137Cs (661.66 keV) and 133Ba (81, 276.4, 302.85, 356.02 and 383.85 keV), respectively and calculated using the following equation (1):

\[
\text{% efficiency} = \frac{\text{peak area (count/sec)}}{\text{emission rate (s}^{-1})} \tag{1}
\]

Then, the radioactivity, decay rate measured represent the combined radioactivity of specific nuclides in the process of decay. The measurements were obtained for 1800 seconds of active time and calculated using as in equation (2) and (3):

\[
\text{activity (A)} = \frac{\text{net peak area}}{\text{efficiency} \times \text{intensity}} \tag{2}
\]

\[
\ln \left( \frac{A}{A_0} \right) = -\frac{ln}{\lambda} \tag{3}
\]

3. Results and Discussion
The UV-absorption spectra of oval and rectangle topaz samples after fast neutron irradiation were shown in figure 2. The spectra were taken in the range of 300 to 800 nm at room temperature. The results show that the absorption spectra was observed a broad peak in the range of 550 to 700 nm and appeared band with maximum at 630 nm, which absorbed red color, result in topaz obtained blue color. For absorption peak at 630 nm in London blue topaz was closely correlated with an O¯ center interacting with two Al ions of the topaz structure. This O¯ center is produced by the irradiation in the hydroxyl sites which...
substitutes for fluorine in the topaz structure [5]. The absorbance of topaz varied with the number of molecules absorbed and cutting styles, lead to the dark topaz was more absorbed than the light topaz. The different colors were due to the chemical and physical defects into the topaz. Furthermore, the absorption spectra of oval topaz was greater than rectangle topaz due to charge transfer of differently charged and oxidation states when topaz was irradiated with fast neutron. The gamma ray spectrum of oval dark and rectangle dark topaz after fast neutron irradiation as shown in figure 3. The results show that the gamma ray spectrum of all topaz were quite similar and observed the energy in range of 84.68 to 1373.8 keV. The high radioactivity of topaz result in detection rate and energy peak is high too. From the energy peak, we can determine the type of radioisotopes of the elements. There are three radioisotopes consist of $^{54}$Mn, $^{46}$Sc and $^{182}$Ta. For $^{54}$Mn and $^{46}$Sc were found at the energy 834.81 keV and 889.28 keV, respectively, while $^{182}$Ta was found at 1189.05 and 1221.41 keV that presented in figure 4. Then, the radionuclide concentrations of topaz were calculated using the following equation (2)

![Figure 2](image1.png)

**Figure 2.** UV-absorption spectra of London topaz from Sri Lanka after fast neutron irradiation.

![Figure 3](image2.png)

**Figure 3.** Gamma ray spectrum of (a) oval dark and (b) rectangle dark London topaz with small, medium and large size after fast neutron irradiation.

The samples consists of dark and light colors in the shape of oval and rectangle with small, medium and large size which irradiated and later measured under the same counting condition were used to calculate the radionuclide concentrations of the elements by activity measurement were shown in figure 5.
Comparisons the specific activity of radioisotopes (\textsuperscript{54}Mn, \textsuperscript{46}Sc and \textsuperscript{182}Ta) for (a) dark and (b) light topaz.

When calculating the total of specific activity of oval dark topaz with small, medium and large sizes were 21.551 ± 0.037, 21.182 ± 0.107 and 1.437 ± 0.014, respectively. It was found that the small oval shape had the highest radioactivity of 21.551 ± 0.037 nCi/g and the large oval shape had the lowest was 1.437 ± 0.014 nCi/g. Moreover, the total of specific activity of rectangle dark topaz, with small, medium and large sizes were 43.952 ± 0.088, 6.603 ± 0.036 and 2.350 ± 0.014, respectively. It was found that the total of specific activity for small shape had the highest radioactivity of 43.952 ± 0.088 nCi/g and the large rectangle shape had the lowest was 2.350 ± 0.014 nCi/g. Then, calculating the total of specific activity of oval light topaz, with small, medium and large sizes were 6.748 ± 0.054, 5.010 ± 0.050 and 2.958 ± 0.031, respectively. It was found that the small oval shape had the highest radioactivity of 6.748 ± 0.054 nCi/g and the large oval shape had the lowest radioactivity was 2.958 ± 0.031 nCi/g. Moreover, the total of specific activity of rectangle light topaz, with small, medium and large sizes were 35.221 ± 0.138, 6.946 ± 0.052 and 1.442 ± 0.023, respectively. It was found that the total of specific activity for small shape had the highest radioactivity of 35.221 ± 0.138 nCi/g and the large rectangle shape had the lowest radioactivity 1.442 ± 0.023 nCi/g. From the results indicated that the radioactivity of the both colors and shape of large size had the lowest radioactivity and small size had the highest radioactivity. Since the topaz was cut into small size, they had more surface area than the large topaz as a result, small topaz better reacts to neutrons [6]. Based on the radioactive concentration analysis of London blue topaz, were used to calculate and predict the return period as shown in the following table 1. In the table 1 shows the decay time of topaz that can calculate the duration of radiation reduction to safe level prior before return to customer according to equation (3). The results shows that the decay time of dark and light in the shape of oval with small, medium and large size were 892, 837 and 160 days and 599, 416 and 234 days, respectively. For rectangle topaz, the dark and light color with small, medium and large size were 625, 400 and 312 days and 542, 535 and 532 days, respectively, which indicated that the size of topaz affected the decay time so increased with decreasing the size of topaz. The decay time of topaz depend on the activity concentrations and half-lives of radionuclides of topaz. Some elements produced long-lived radionuclides which in turn will require long storage time to bring the induced radioactivity within the safe limits. Furthermore, the absorption coefficient of standard point source of \textsuperscript{137}Cs with topaz was found that the small, medium and large size of the oval topaz was 1.074, 1.102 and 1.257, respectively while, the small, medium and large size of the rectangle topaz was 1.071, 1.078 and 1.164, respectively as shown in table 2. The results obtained from the absorption of gamma photons that affected the intensity so the larger topaz was more capable of absorbing radiation than medium and small topaz. As a result, the size of topaz also affects the absorption coefficient and
corresponding to the previous results. Finally, this study is applied to predict time of residue dose of topaz for enhancement colorless topaz by neutron radiation treatment.

**Table 1.** Decay time of dark and light topaz from Sri Lanka.

| Shape of topaz | Size of topaz | Decay time (days) |
|----------------|---------------|-------------------|
|                |               | Dark color | Light color |
| Oval topaz     | Small         | 892        | 599         |
|                | Medium        | 837        | 416         |
|                | Large         | 160        | 234         |
| Rectangle topaz| Small         | 625        | 542         |
|                | Medium        | 400        | 535         |
|                | Large         | 312        | 532         |

**Table 2.** Absorption coefficient of standard point source of Cs-137 with topaz.

| Size of topaz | Peak area (count/sec) | Absorption coefficient |
|---------------|-----------------------|------------------------|
| Standard point source of Cs-137 Cs-137 | 147.038 | - |
| Standard point source of Cs-137 with oval topaz Small | 127.334 | 1.074 |
| Medium        | 120.581               | 1.102                  |
| Large         | 91.338                | 1.257                  |
| Standard point source of Cs-137 with rectangle topaz Small | 128.071 | 1.071 |
| Medium        | 126.405               | 1.078                  |
| Large         | 107.708               | 1.164                  |

4. Conclusion

In this paper, the London blue topaz from Sri Lanka after fast neutron irradiation was investigated by UV-Visible spectroscopy and HPGe gamma ray spectrometry. The UV-absorption spectra of topaz was observed with only broad peaks in the range of 550 to 700 nm and appeared band with maximum at 630 nm which absorbed red color, result in topaz obtained blue color. At 630 nm was correlated with an O\(^{-}\) center interacting with two Al ions of the topaz structure. This O\(^{-}\) center is produced by the irradiation in the hydroxyl sites which substitutes for fluorine in the topaz structure. The radioactivity of dark and light topaz was found that the small rectangle topaz had the highest radioactivity of 43.952 ± 0.088 nCi/g and 35.221 ± 0.138 nCi/g, respectively because there is more surface area than the large topaz. The decay time of the radioactive element depended on the size of topaz which the dark color in the shape of small oval topaz had the longest decay time of 892 days. Moreover, the size of topaz also affected the absorption coefficient.

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