Radioactivity of the Treated Topaz
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ABSTRACT

Raw topaz stone samples are subjected to irradiation by neutrons from the Egyptian second research reactor and by gamma (γ) rays from a gamma source. Changes in the color of the stones are observed.

Irradiation induces radionuclides, resulting to radioactivity. Therefore, some time is required for the radioactivity level of stones to fall to exempted value; hence, radioactivity of the stones after irradiation has to be followed and monitored till their release from the reactor. Storage time might be several months to several years.

The decay rates of isotopes created by irradiation depend on the individual isotope. Neutron-irradiated topaz treated in a nuclear reactor facility can present a potential health hazard, if not properly controlled.

A special license is required for transportation and distribution of such treated gemstones, because radiation exposures associated.

Experience gained in management of irradiated topaz shall be reviewed.

Key Words: treated topaz, neutron irradiation, topaz decay and transportation.

1-INTRODUCTION

Natural gemstones have for long human desire beside its economic value. They have achieved this status by possessing the desirable attributes of beauty, rarity, durability, portability.

Gems are frequently subjected to various treatments in order to improve their appearance, and hence increase their commercial value. Different techniques are used to treat the color that is the most important property of gemstones. The treatment has a significant effect on the value of a gemstone. Irradiation treatment is done by gamma or by neutrons. Particles of high energy such as electrons or neutrons and electromagnetic ionizing radiations like gamma rays can produce color centers in the stone which can transfer it to a gem stone. The primary key to the value of gemstones lies in their beauty. The fashioning, sorting, marketing and selling of treated gemstones and jewellery are a large multibillion dollar industry, employing people from all over the world. Topaz is the best example for enhancement of color by commercial application of neutron irradiation.

2-EXPERIMENTAL

The sample

Natural topaz stone has the structure of an aluminum silicate fluoride hydroxide Al₂(SiO₄)(OH, F)₂, and is usually colorless or has unattractive color. Therefore, it is not considered before irradiation treatment to be a gemstone. Measurements are carried on cut topaz samples were prepared from large piece of raw and treated topaz to have the size of (~ 1.0 cm x 1.0 cm x 0.2 cm).
Irradiation Methods and Irradiation Facilities

Irradiation is the process by which materials are exposed to various types of nuclear radiation to change some parts of the crystal structure. This change in crystal structure of the raw gemstone causes to alter its color, either by the addition or subtraction of some part of the crystal structure.

The color that is produced of $^{60}$Co gamma rays is not uniform while the blue color produced by neutron bombardment of topaz is usually uniformly deep, regardless of the size of the topaz, because neutrons have excellent penetrating properties. Neutron irradiation produces a deeper blue color than irradiation by gammas because neutrons are more heavily ionizing, inducing more change to the crystal structure, and penetrate the whole crystal \(^{(5)}\). Unfortunately, radioactive by-products are produced with such treatment.

The samples are irradiated for \((2,4,8\text{ and }12)\) hours at 22 MW- with an average of thermal neutron flux of \(2.37\times10^{14} \text{n/(cm}^2\cdot\text{s)}\). After irradiation of the stones, they have to be transferred to the auxiliary pool until their radioactivity reduces to permissible level, and then to hot cells before any treatments.

\[ \text{a. Gamma-Ray Facility} \]

Gamma -1 unit of the national center of radiation research and technology was used. Cobalt-60 radioactive source was used as gamma source.

\[ \text{b. Neutron Irradiation Facility} \]

The Facility used for neutron irradiation is the Egyptian second research reactor, ETRR-2. It is a multi purpose reactor, (MPR), a 22 MW power, open pool type, with maximum thermal neutron flux of \(2.7\times10^{14} \text{n.cm}^{-2}\cdot\text{s}^{-1}\). ETRR-2 is light water cooled and moderated, with Beryllium reflectors. Fuel elements are MTR type and 19.75 % enrichment. Specific irradiation container has been designed to ensure proper cooling of the stones during irradiations. Several irradiation tests have been conducted to define the optimum parameters of the facility.

3-RESULTS AND DISCUSSION

Topaz as a material under investigation Topaz was selected because of its high desire in the gem trade market and for its experience (June1990) in the nuclear reactors (6). Neutron activation analysis is used to investigate the isotopes kind and its concentrations in the irradiated samples for the residual radioactivity calculation.

Trace elements cause residual radioactivity in topaz after irradiation. It is not allowed to distribute topaz until it decays and reaches a safe level of use for the public. Storage of irradiated samples and isotopes activity for bulk were studied.

Neutron Activation Analysis (NAA) technique and instrumentation

Neutron activation analysis (NAA) has been applied for the analysis of treated gemstones as a very accurate and precise technique \(^{(7)}\). Short and long irradiations were implemented depending on the half-life of the induced radionuclides. NAA is a technique in which gamma ray emissions are detected. It is used for samples analysis after irradiation in the swimming pool nuclear research reactor (ETRR-2) of EAEA, Egypt. The samples are prepared for irradiation with certain procedure. The gamma-ray spectroscopy system includes a computer, associated electronics, a lead shield that contains a detector, and a liquid-nitrogen storage tank. The heart of the system is the detector, which is made from a very-high-purity germanium (HPGe) crystal. The goal was to obtain a profile of the main impurities existing in the stones. The analysis begins with eliminating all removable radioactive surface contamination. NAA measurements were carried out according to the neutron cross section of the parent nuclides of elements to be determined \(^{(8)}\). These measurements are carried out at a 7 cm
away from the top of the high pure germanium detector with relative efficiency of 100%, and a resolution at full width at half maximum (FWHM), namely 2.1 KeV for the 1332.4 KeV photons of Co-60.

**Topaz short irradiation measurement**

The samples were measured individually for their γ -radiation emission. This short irradiation methodology was used for the determination of Al, Cl, Mg, Na, Ti, V and K.

Gamma spectrum of short irradiated topaz shows that it consists of many isotopes with short half-life. Qualitative and quantitative studies by NAA technique for topaz samples were performed.

**Topaz Long Irradiation measurement**

For the analysis, each stone was placed inside the one-ton lead shield in the same position as the stimulants to which the system had been calibrated. Counting times for (10,000 seconds) were established on the basis of the gem's distance above the detector and the gross radiation readings from the Geiger counter. For the bulk quantities in the range of 1 Kg were measured.

Some residual radioactivity was observed still high till after 6 months of irradiation. This activity is due to the isotopes (Sc, Fe, Mn and Ta) with half-lives of (83.79, 44.4, 312.3 and 113.5) days, respectively.

Qualitative and quantitative studies by NAA technique for long irradiated topaz show that it consists of Sc-46(889.25), Mn-54(834.83), Co-60(1332.5 and 1173.2), Cs-134(604.7 and 795.8) and Ta-182(1121.3, 1189.1 and 1221.4) Bq/g as isotopes mainly producing the residual radioactivity.

The isotopes with short half-life times disappeared and the isotopes with long half-life times such as Sc-46 (83.85 d) and Ta-182 (114.74 d) started to appear.

**NAA Calculations**

One of the objectives of this study is to calculate the concentrations of isotopes in neutron irradiated topaz. This is important to identify the specific nuclides responsible for the radiations. It helps to calculate the dose to people attributable to the induced activity. The samples which irradiated and later measured under the same counting conditions were used to calculate the concentration of the elements of interest by comparison of the measured activity between the sample and the standard. There are two main methods used to calculate isotopes concentration namely, comparator and relative methods. Relative method depends mainly on the ratio between concentration of standard material and its gamma peak intensity.

![Energy Calibration (using Co 60-Cs 137 and Ba 133 isotopes)](image)

Figure (1); Energy Calibration (using Co 60-Cs 137 and Ba 133 isotopes)
Table (1); Comparison between concentrations calculations of topaz irradiated by neutrons by using Comparator and Relative methods

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Concentration (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Comparator</td>
</tr>
<tr>
<td>Sc-46</td>
<td>8.62</td>
</tr>
<tr>
<td>Ta-182</td>
<td>0.53</td>
</tr>
<tr>
<td>Fe-59</td>
<td>3782.71</td>
</tr>
<tr>
<td>Mn-54</td>
<td>0.06</td>
</tr>
<tr>
<td>Co-60</td>
<td>0.03</td>
</tr>
<tr>
<td>Cs-134</td>
<td>0.01</td>
</tr>
<tr>
<td>Cr-51</td>
<td>0.48</td>
</tr>
<tr>
<td>Zr-95</td>
<td>5524.53</td>
</tr>
<tr>
<td>Zn-65</td>
<td>21328.59</td>
</tr>
<tr>
<td>Na-24</td>
<td>5.21</td>
</tr>
<tr>
<td>La-140</td>
<td>1.82</td>
</tr>
<tr>
<td>As-76</td>
<td>64.43</td>
</tr>
<tr>
<td>Al-28</td>
<td>265372.42</td>
</tr>
<tr>
<td>Ti-208</td>
<td>2825.78</td>
</tr>
<tr>
<td>Cs-138</td>
<td>48.53</td>
</tr>
<tr>
<td>Mg-27</td>
<td>53.91</td>
</tr>
<tr>
<td>Mn-56</td>
<td>152.62</td>
</tr>
<tr>
<td>Cl-38</td>
<td>1.35</td>
</tr>
</tbody>
</table>

Table (2); Comparison between concentrations calculations of topaz as irradiated by neutrons and irradiated by neutrons after γ irradiation

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Topaz irradiated by neutrons</th>
<th>Topaz irradiated by neutrons after γ irradiation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sc-46</td>
<td>7.24</td>
<td>98.6</td>
</tr>
<tr>
<td>Co-60</td>
<td>0.02</td>
<td>0.00191</td>
</tr>
<tr>
<td>Ta-182</td>
<td>0.44</td>
<td>0.149</td>
</tr>
<tr>
<td>Fe-59</td>
<td>5012.82</td>
<td>15600</td>
</tr>
<tr>
<td>Mn-54</td>
<td>0.08</td>
<td>0.0416</td>
</tr>
</tbody>
</table>
The Concentration of \(^{46}\text{Sc}\) at topaz as irradiated by neutrons after \(\gamma\) irradiation is equal to that at Topaz irradiated by neutrons about "13" times while it is equal to \((^{60}\text{Co})\). \((^{182}\text{Ta})\) at the first sample is "3" times the second sample while the opposite for \((^{59}\text{Fe})\) and finally \((^{54}\text{Mn})\) at the first sample is twice the second sample.

**Radiation protection Instrumentations**

Radiation protection measuring devices are used for:

a) Monitoring of workplace.

b) Follow up irradiated topaz during storage.

c) Transportation of irradiated topaz.

The safety of personnel against ionizing radiation emitted from gemstones previously exposed to neutrons is achieved by using suitable tools such as hand-held or portable survey meters and fixed instruments. These instruments are essential for determination of radiation dose, samples analysis and concentrations estimation for isotopes.

**a. Radiation Detection Equipments**

1. The radiation measuring devices in use in the present search are grouped according to type of exposure:

a) Radiation Worker

b) Member of the public.

a) For radiation worker exposure and occupational control

1. Radiation measuring devices
2. Radioactivity concentration measuring devices
3. Area monitoring devices in the workplace
4. Personnel monitoring devices in the workplace
5. Measurement during storage of the irradiated gemstones
6. Radiation measurements during transport of gemstones containers by air

b) For members of the public

A thorough understanding of radiation detection equipment and information on radiation exposure is necessary to estimate the health risk from irradiated blue topaz. Domestic processors will need to understand the equipment and its limitations to assess quantities such as exempt release concentration levels.

**Radiation Detection Equipment results**

"Thermo FH 40 G" radiometer model "DB-033-961017 E" exposure rate measuring unit with FH 40 TG teleprobe was used for these measurements and Portable contaminations monitor "CONTAMAT" FHT 111 M was used for these measurements (table 3).
The alpha content of raw samples was determined during long time of counting in the hours range, using a contamat. No alpha activity above background was detected. The CONTAMAT FHT 111 M was used for these measurements.

**Cooling time calculations**

Calculations were done using FORTRAN Program depending on Newton-Raphson method which is an iterative procedure that can be used to calculate maximum likelihood estimates (MLEs).

**Table (4); calculated cooling time of some samples**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Time* (yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Topaz irradiated by neutrons</td>
<td>1.386</td>
</tr>
<tr>
<td>Topaz irradiated by neutrons after γ irradiation</td>
<td>1.520</td>
</tr>
<tr>
<td>Topaz irradiated by neutrons twice</td>
<td>2.318</td>
</tr>
</tbody>
</table>

* Time (yr) required for activity to reach the safety level for transportation by air according to IATA regulations (74 Bq/g) \(^8\).

* General decay any sample after (γ, neutron, γ +neutron and neutron twice) irradiation increase gradually.

Topaz as irradiated in a nuclear reactor contains many types and amounts of radionuclides as reported before \(^8\). It takes to be releasable, after radioactive decay 1.38 year according to table (4) while it takes a little more than two years after irradiation \(^6\) at some cases.

**Table (5); Isotopes in the sample of Topaz irradiated by neutrons**

<table>
<thead>
<tr>
<th>Isotope</th>
<th>(A_e) (Bq/g)</th>
<th>Radiation emitted</th>
<th>Half-life (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sc-46</td>
<td>652.24</td>
<td>β + γ</td>
<td>83.79</td>
</tr>
<tr>
<td>Co-60</td>
<td>18.85</td>
<td>4.98</td>
<td>1925.37885</td>
</tr>
<tr>
<td>Ta-182</td>
<td>18.85</td>
<td>39.78</td>
<td>114.43</td>
</tr>
<tr>
<td>Fe-59</td>
<td>24.68</td>
<td>21.8</td>
<td>44.503</td>
</tr>
<tr>
<td>Mn-54</td>
<td>11.73</td>
<td>4.03</td>
<td>312.3</td>
</tr>
</tbody>
</table>

Where element mass (m_e) = (0.46484 and 0.54778) g for neutrons and (neutrons+ γ) at columns 2 and 3 respectively.
γ is the more effective from the radiation protection point of view because of its high ability for penetration while β could be protected easily. Between all isotopes in topaz Co-60 is highly considered because of its very long half life which increases the time of decay even if its concentration is low.

Usually the activity of Sc-46 at most times for most samples is the higher than the sum of all other activities. The ratios in activities differ according to the concentrations of these isotopes in the samples according to its origin.

<table>
<thead>
<tr>
<th>Decay dependent on type of treatment and calculation</th>
<th>Time (years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Topaz irradiated by neutrons (Experimental)</td>
<td>1.12</td>
</tr>
<tr>
<td>Topaz irradiated by neutrons (Experimental considering all isotopes)</td>
<td>1.39</td>
</tr>
<tr>
<td>Topaz irradiated by neutrons (Theoretical)</td>
<td>1.42</td>
</tr>
<tr>
<td>Topaz irradiated by neutrons after gamma irradiation (Theoretical)</td>
<td>1.52</td>
</tr>
<tr>
<td>Topaz irradiated by neutrons twice (Theoretical)</td>
<td>2.31</td>
</tr>
</tbody>
</table>

Transportation

a. Condition of transport of previously irradiated stones abroad

The transport of radioactive material is subject to a radiation protection program, which must consist of systematic arrangements aimed at providing adequate consideration of radiation protection measures. The nature and extent of the measures to be employed in the program must be related to the magnitude and likelihood of radiation exposure.

The activities are measured many times before transportation. IATA does not permit transportation for gemstones with activity more than 74(Bq/g). The transportation regulations are followed.

Transport Index (Ti)

Is defined as the maximum radiation level in microsieverts per hour (µSv/h) at 1 m from the external surface of the package, divided by 10.

The united Nations Number (UNN) or UN IDs are four-digit numbers that identify hazardous substances in the framework of international transport. The (UNN) is displayed on the placards (lower half).

The distribution of irradiated materials, even with low levels of induced radioactivity, to unlicensed person is prohibited unless the distributor of such materials has a specific license, which permits such distribution. Topaz is irradiated to cause it to change color. Irradiation can induce radioactivity, albeit in exempt concentrations. Blue topaz is still the most common gemstone being irradiated at the market. The color of topaz natural or irradiated by γ-rays easy to be removed or deformed if the stones suffered from temperature higher than 450 °C while the stones irradiated by neutrons still stable without any deformation under highest temperatures treated. The topaz is used mainly as gemstones, for scientific and technological applications.

Dependent on experimental observations the optimum way to obtain best irradiated topaz by the lowest cost it is recommended to irradiate it first by γ then by neutrons at the reactor, will needs only short irradiation time at this case, and finally irradiate for very short time in accelerator to give it its attractive shape. Several elements were determined with different concentrations. In particular elements such as Sc, Ta, Mn and Fe are especially significant to the activities at neutrons irradiated
topaz. More gemstone could be produced from Egypt by irradiation technique and new kinds of
gemstone irradiation may become to be technically or economically available, enlarging the gemstone
irradiation service market.

**for radiological concerns:**

a. No contamination is observed.

b. Radionuclide concentrations is only a fraction of exempt concentrations.

c. Radiation dose and dose rates expected to be low, even for large quantities of gemstones.

**for licensing**

a. Need to be able to demonstrate that “hot "gems do not escape detection.

b. Need to be able to demonstrate that gems do not exceed exempt concentrations for any nuclides (the "sum of fractions" rule applies).

c. Need to demonstrate that the licensee can make these determinations to the satisfaction of regulators.

Prospective licensees must be able to convince the regulators that they are competent to determine that
gems are exempt from regulation.

**Commercial Factors include** topaz irradiation is carried out between the activities to reduce the cost.

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