REACTOR-IRRADIATED GREEN TOPAZ

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Examination of samples of faceted "Ocean green" topaz reveals that this material has been irradiated in a nuclear reactor. An earlier reactor-irradiation experiment suggests that temperature conditions during the irradiation of this material may be higher than those used to produce typical "London-blue" topaz. Like such reactor-irradiated blue topaz, this material may be radioactive. In addition, the green color is not stable to direct sunlight.

A relatively new color variety of topaz is being sold under the trade name "Ocean green" topaz (figure 1), with several thousand carats of this material now in the marketplace. Inquiries to dealers revealed that most of the original, untreated topaz comes from Sri Lanka, is currently being irradiated at the research reactor at Texas A&M University, and is being released under their license from the U.S. Nuclear Regulatory Commission (NRC). Material of this same color may also originate from, and be irradiated in, other countries.

GIA's current interest in testing reactor-irradiated blue topaz for radioactivity, and in determining whether it can be released to consumers in compliance with U.S. regulations regarding reactor-irradiated gem materials, now extends to reactor-irradiated green topaz as well. Telephone inquiries to the reactor facility at Texas A&M were unsuccessful in obtaining any information related to the treatment process for this kind of topaz. Therefore, this article reports the results of our gemological examination, chemical analysis, and radioactivity and color-stability testing of several specimens of this material.

MATERIALS AND METHODS

For this study, we selected five faceted samples of irradiated green topaz to examine in detail (table 1). For color-origin and stability testing, we also examined a variety of additional green and other types of topaz (see table 1 and figure 2). Samples 1–4, 9, and 12–13 represent the Ocean-green topaz being sold commercially at this time. Sample 5 was irradiated by one of us (CEA) in a nuclear reactor in 1986. This treatment was performed in a cadmium-shielded "dry rabbit" type of cannister without water cooling (and, therefore, at an elevated temperature) for seven hours. Under these conditions, a bluish green color resulted from a colorless starting material. Green topaz has been reported to occur in nature, but it seems to be rare (Webster, 1983; Deer et al., 1982; Hoover, 1992), and we were unable to obtain natural specimens for examination.

We used standard gemological tests and equipment to characterize green samples 1–5. We then recorded absorption spectra for samples 1–8 over the range of 250–2500 nm using a Hitachi U-4001 spectrophotometer. These spectra were recorded with unpolarized light, with each topaz oriented such that the light beam traveled through the polished girdle. Because it is usually impractical to orient faceted stones optically, we did not attempt to do so; this should be kept in mind when comparing the visible absorption spectra. In addition, we determined the

chemical composition of three samples (nos. 1, 5, and 6) from their polished table facets using a Tracor Spectrace 5000 X-ray fluorescence (EDXRF) analysis system.

We also tested irradiated green topaz samples 1–5, 9, 12, and 13 for radioactivity using GIA's gamma-ray spectroscopy system (Ashbaugh, 1992). Each sample was placed with its table facet positioned above the high-purity germanium (HpGe) detector, and then "counted" for a one-hour period.

TABLE 1. Topaz samples examined.

Sample no.	Color	Weight (ct)	Comments[b]
1[a]	Green	3.85	Emerald cut, irradiated, (R1154)
2[a]	Bluish green	4.34	Emerald cut, irradiated, (R1155)
3[a]	Bluish green	4.71	Emerald cut, irradiated, (R1614)
4[a]	Bluish green	8.70	Emerald cut, irradiated, (R1613)
5	Bluish green	4.10	Emerald cut, reactor irradiated at elevated temperatures (no cooling), (R1156)
6	Blue	3.38	Emerald cut, irradiated and heat treated, from Sri Lanka, (R1160)
7	Brownish yellow	11.26	Emerald cut, from Brazil, (R1161)
8	Brown	14.51	Oval cut, irradiated, (R1186)
9[a]	Green	9.20	Emerald cut, irradiated, cut in half for control (A) and test (B) samples; one piece (B) heat treated (now blue); half of A subsequently exposed to direct sunlight (now blue)
10	Brown	1.84	Modified triangle cut, linac-irradiated; exposed to direct sunlight (now blue)
11	Brown	2.00	Modified triangle cut, linac-irradiated; heat treated (now blue)
12[a]	Green	4.50	Modified pear shape, irradiated; tested for color stability to combined fluorescent and incandescent light
13[a]	Green	4.94	Modified pear shape, irradiated; tested for color stability to direct sunlight (now blue)

[a] Sample represents the material being sold today as "Ocean green" topaz.
[b] Number in parentheses refers to GIA Research sample number. Information after semicolons for samples 9–13 refers to type of color-stability testing, as appropriate.

Figure 1. A new color variety of irradiated topaz has appeared in the trade, and is being sold under the name Ocean-green topaz. These samples (3.85–8.70 ct) show a typical range of color for this material. Photo © GIA and Tino Hammid.

As a preliminary test of color stability to heat, green sample 9 was cut into two pieces, with one half (9A) retained as a control (together with linear accelerator [linac] irradiated brown topaz sample 10). The other half of sample 9 (9B) and brown sample 11 were progressively heated in air over a temperature range of 180°–400°C in 10° increments, with a half-hour period at each step. The stones were visually examined at the end of each period to determine at what temperature a color change occurred.

Last, we tested the color stability of the irradiated green topaz to light. We cut green sample 9A in half again and placed one piece with green sample 13 (and, for comparison, brown sample 10) outdoors in direct sunlight for one day. We exposed green sample 12 to normal indoor lighting—that is, a combination of simultaneous incandescent (30 cm from a 100-watt bulb) and fluorescent lighting—for 48 hours. To determine the extent of the color change, if any, we compared the test stones to Munsell color chips before and after each exposure.

Figure 2. These five samples (from left—nos. 1, 8, 7, 6, and 5) represent the different types of topaz examined in this study and, specifically, those for which absorption spectra are given in figure 3. Photo by Maha DeMaggio.

RESULTS

Gemological Examination. The indices of refraction of green topaz samples 1–5 fell within the range 1.610–1.620, typical values for both untreated and treated topaz (Liddicoat, 1990, p. 208). To long-wave ultraviolet radiation, these samples fluoresced a weak-to-moderate, turbid, yellowish green. The short-wave fluorescence was similar in color but less intense. This U.V. luminescence has been seen in some other colors of topaz (Webster, 1983, p. 141). When the stones were examined with a microscope, no distinctive inclusions or other features were observed.

Absorption Spectra and Chemistry. The visible absorption spectra of two representative green topaz samples (nos. 1 and 5) revealed three main features, all of which have been described previously in colored topaz (see, e.g., Petrov, 1977). These three spectral features are all too weak and too broad to be seen as distinct bands with a handheld spectroscope. Comparison of the visible spectra of these samples with those of other colors of untreated and treated topaz (figure 3) provides an explanation for the green color.

The spectrum of each of the two green samples displays a very weak, broad region of absorption between 500 and 700 nm that is centered at about 600 nm. This same broad band is also present in the spectrum of the blue topaz that has been irradiated and heat treated (no. 6; see also figure 5 in Petrov, 1977, p. 296).

The spectra of the two green samples also display a broad and very intense region of absorption centered in the ultraviolet, a portion of which extends into the visible region up to about 500 nm. This

appears to be the same as the prominent feature seen in the spectrum of irradiated brown sample 8. It is also evident, though much weaker, in blue sample 6. Nassau and Prescott (1975) described this broad band as being produced when topaz is irradiated.

Figure 3. These visible and near-ultraviolet absorption spectra of two samples of irradiated green (nos. 1 and 5) and other natural-color (no. 7, brownish yellow) and laboratory-irradiated (nos. 6—blue and 8—brown) topaz samples illustrate the three main absorption features present in the spectra of irradiated green topaz that, in combination, produce the Ocean-green color: (1) a broad, weak absorption centered at about 600 nm (giving rise to the blue component; e.g., sample 6); (2) the broad region of absorption in the ultraviolet and extending into the visible (causing a brown component; sample 8); and (3) the broad, very weak absorption centered at about 425 nm, and seen on the shoulder of the ultraviolet absorption (contributing to a yellow component; sample 7).
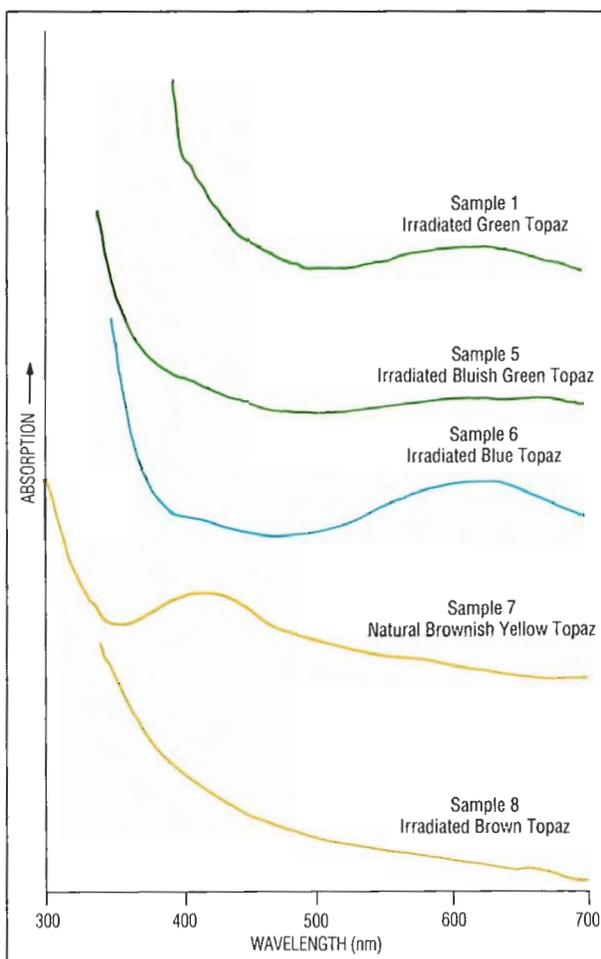
The third feature that we observed in the green samples is a very weak, broad region of absorption centered at about 425 nm that appears to be the same as a band that is especially prominent in the spectrum of the natural-color brownish yellow (no. 7) topaz (also see figure 2 in Petrov, 1977, p. 294). In the spectra of the two green topaz samples (nos. 1 and 5), this broad band is barely visible and only as a very weak shoulder on the side of the large absorption band in the ultraviolet.

Besides the expected aluminum and silicon (the fluorine in most topaz cannot be detected by this method), chemical analysis by EDXRF revealed the presence of germanium (Ge) in all three green topaz samples (nos. 1, 5, and 6) analyzed. This trace element substitutes for silicon in the topaz crystal structure in concentrations up to several hundred parts per million (ppm), and is occasionally reported in chemical analyses of topaz (see, e.g., El-Hinnawi and Hofmann, 1966; Deer et al., 1982, p. 806). When this trace element is present in topaz, irradiation in a linear accelerator generates the radionuclide germanium-69 (Ge-69); as a result, the treated stones must be withheld from the market for a few weeks to await radioactive decay (see Ashbaugh, 1988).

Radioactivity. We determined that each of the six green and bluish green topaz samples that registered radioactivity contained the radionuclides commonly found in reactor-irradiated topaz (Schmetzer, 1987; Ashbaugh, 1991). These included scandium-46 (Sc-46), tantalum-182 (Ta-182), and manganese-54 (Mn-54;
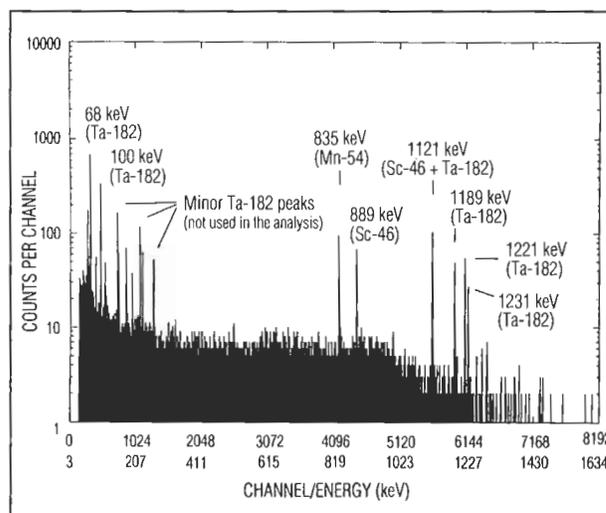

Figure 4. The gamma-ray spectrum of green topaz sample 9 (a one-hour counting period) shows the gamma-ray peaks of Sc-46, Ta-182, and Mn-54. The gamma-ray energy range of the spectrum shown is from 50 to 1500 keV. The radionuclides are represented by peaks in the spectrum. The quantities of each of the radionuclides can be calculated by computer analysis from these counting data.

see the gamma-ray spectrum of sample 9 in figure 4). Sample 1 showed a small amount of cesium-134 (Cs-134). These radionuclides would not likely be found in topaz irradiated by other methods (Ashbaugh, 1991). The concentration levels of these radionuclides in each sample are shown in table 2. The number shown

TABLE 2. Radionuclide data for samples of irradiated green topaz.[a]

Radionuclide	NRC release limit [b]	Sample no. (and carat weight)					
		1 (3.85)	2 (4.34)	3 (4.71)	4 (8.70)	5 (4.10)	9 (9.20)
Sc-46	0.4	Trace[d]	0.02	0.73	0.10	Trace	0.15
Ta-182	0.4	0.20	Trace	0.28	0.20	Trace	0.76
Mn-54	1.0	Trace	Trace	0.17	0.14	Trace	0.17
Cs-134	0.09	0.01	None[e]	None	None	None	None
Sum of ratios[c]	1.0	0.61	0.05	2.70	0.89	0.01	2.45

[a] Radionuclide data in nanocuries per gram (nCi/gm) of radioactivity. Data collected on October 27, 1992; the radionuclide concentration values will decrease over time due to radioactive decay. Because no radioactivity was detected for samples 12 and 13, they are not included in this table.
[b] Current NRC release concentration limits for these radionuclides—see U.S. NRC Rules and Regulations, Title 10, Chapter 1, Part 30.70, Schedule A (August 30, 1991); and Part 20, Appendix B (May 31, 1991).
[c] Sum of the ratios = summation of the concentration of each radionuclide present divided by the NRC release limit for that radionuclide. This number must be less than or equal to 1.0 for the individual irradiated topaz sample to be sold or distributed legally in the United States.
[d] Trace = trace quantities (less than 0.01 nCi/gm) of these radionuclides were detected.
[e] None = the radionuclide was not detected.

in the bottom row (labeled as the "sum of the ratios") must be less than or equal to 1.0 for the topaz sample to be sold or distributed legally in the United States. As can be seen, samples 3 and 9 were above this limit.

Color Stability. With regard to color stability to heat, at about 250°C the brown color of sample 11 became blue; beginning at about 310° up to 375°C, sample 9B changed progressively from green to blue, with no residual green color present above the latter temperature (C. Habib, pers. comm., 1992; figure 5). According to our experience, blue topaz is most color stable to heat, while irradiated brown and (now) green topaz are less stable.

The two green topazes (no. 13 and the portion of 9A) and one brown (no. 10) sample that were tested for stability to direct sunlight all became blue within one day of exposure (see, e.g., figure 6). Subsequent exposure of two additional Ocean-green topaz samples not in the original study gave the same result: Both turned blue in the course of one day in direct sunlight. However, there was no change in the green color of sample 12 after 48 hours of exposure to a combination of simultaneous incandescent and fluorescent (normal indoor) lighting. The visible absorption spectrum of sample 13, after it had turned blue in the sunlight, was identical to that of irradiated blue topaz (see, e.g., the spectrum of sample 6 in figure 3).

DISCUSSION

Because the samples of green topaz that we tested revealed the same radionuclides as are typically found in neutron-irradiated blue topaz, we believe that the

Figure 6. Ocean-green topaz sample 12 (top) showed no color change when exposed to combined fluorescent and incandescent (indoor) lighting. Sample 13 (bottom) changed from green to blue within a day of exposure to direct sunlight. Photo © GIA and Tino Hammid.

green topaz was also neutron irradiated, but probably at a higher temperature. This conclusion is substantiated by the fact that one of us (CEA) produced the bluish green color of sample no. 5 by irradiation in a non-water-cooled area of a nuclear reactor; that is, the topaz was subjected to an irradiation temperature that was higher than is typical in this procedure. We have also seen some green topaz in parcels after electron treatment in a linear accelerator and prior to the heat treatment normally used to produce the "Sky-blue" material (pers. knowledge CEA; D. Duke, pers. comm., 1992). However, this type of irradiation would not have produced the radionuclides found in reactor-irradiated topaz.

The color of this irradiated green topaz can be understood by referring to the visible absorption spectra in figure 3. The green results from the cumulative effect of the causes that individually produce brown, yellow, or blue. In the spectrum of the green topaz, there is the broad absorption in the ultraviolet extending into the visible (causing the brown component), the broad but very weak absorption centered at about 425 nm (providing the yellow component), and a broad, weak absorption centered at about 600 nm

Figure 5. Heat-treatment experiments revealed that exposure to high heat will turn either brown or green topaz blue. On the left are the originally green control (sample 9A) and heat-treated (sample 9B) topaz samples; on the right are the originally brown control (sample 10) and heat-treated (sample 11) topazes. Photo by Maha DeMaggio.

(giving rise to the blue component). This agrees with the interpretation of Petrov (1977).

CONCLUSION

The Ocean-green topaz currently being marketed has been neutron irradiated in a nuclear reactor under conditions that are believed to be different—in terms of a higher irradiation temperature—than the conditions used to produce normal London-blue topaz.
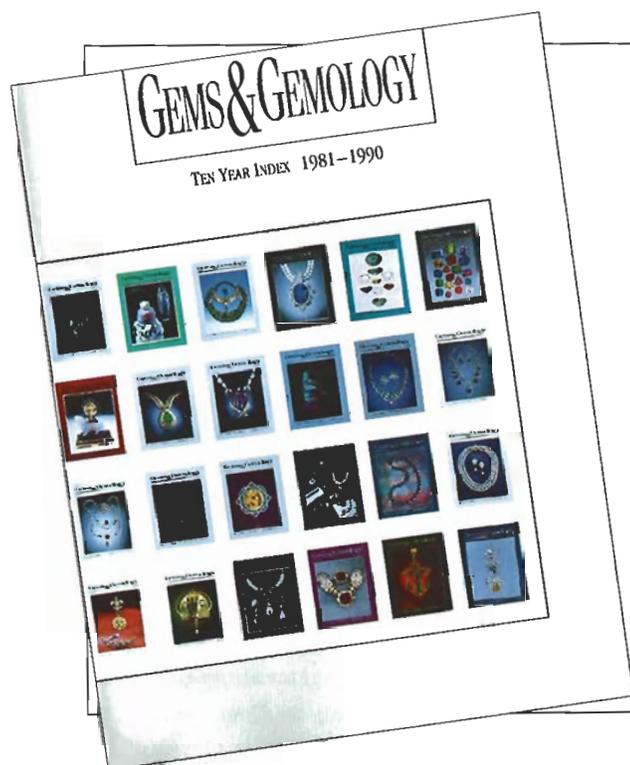
As is the case with reactor-irradiated blue topaz, this green topaz is radioactive when first removed from the reactor. NRC regulations require that it be tested for residual radioactivity before it is made available for sale to the general public. Two samples tested in this study (nos. 3 and 9) were found to be above the current NRC release limits and, as such, are illegal to sell or distribute in the United States. Therefore, all green topaz should be tested for residual radioactivity by an NRC-licensed testing facility before being placed on the market.

We also found that irradiated Ocean-green topaz turned blue when exposed to direct sunlight for only a day. Exposure to fluorescent and/or incandescent light indoors did not appear to affect the color. Disclosure of the instability of the green color to sunlight should be made in conjunction with any sale of this material.

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