GEMSTONE IRRADIATION AND RADIOACTIVITY

By Charles E. Ashbaugh III

With the increasing use of radiation to color enhance gemstones, the issue of radioactivity has become a source of concern for gemologists worldwide. This article examines the basic characteristics of radiation and radioactivity, the sources of radiation in the earth and in the laboratory, the radiation treatment of gemstones in particular, and the detection and measurement of radiation. Also included is a discussion of potential health hazards and current government regulations regarding gemstone radioactivity.

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Acknowledgments: The author wishes to thank the following for their assistance and critical evaluation of this article: Dr. Kurt Nassau of AT&T Bell Labs; Dr. Karl Bernstein of Trintec; Dr. Junaid Razvi and Dr. William Whittemore of General Atomics; Dr. Robert Block of the Rensselaer Polytechnic Institute; Anthony Zane and other colleagues at UCLA; George Drake of CN&L Investment Corp.; Charles Key; and especially Dr. Emmanuel Fritsch and Dr. James Shigley of GIA. Ruth Patchick was invaluable for typing the many drafts.

"Nothing in life needs to be feared. It only needs to be understood."—Marie Curie, discoverer of radium and the recipient of two Nobel prizes.

Exposure to radiation, either from natural or man-made sources, will enhance the color and beauty of many different gemstones. The use of man-made sources of radiation to treat gem materials was experimented with soon after radiation was discovered, almost 100 years ago. In recent years, gemstone irradiation has become a common practice, as evidenced by the literally millions of carats of irradiated blue topaz, and increasing amounts of dark pink tourmaline, yellow beryl, red zircon, and colored diamonds, encountered in the marketplace (figure 1).

Accurate information on gemstone treatments, however, is difficult to obtain. Individuals involved in commercial gemstone irradiation are reluctant to reveal details of their treatment processes, which they consider to be proprietary information. Also, the general subject of radiation itself is rather technical and complicated. At the same time, government policy on the handling and distribution of radioactive irradiated gems to the general public is still being slowly and very cautiously formulated. Finally, only relatively recently have some irradiated gemstones been examined in detail to understand better the changes produced by such treatments. This article reviews the nature of radiation and its various sources, both natural and man-made, as well as the laboratory irradiation of gemstones. Also covered are the issues of radioactivity, its detection and measurement, and current pertinent government guidelines. While there is no question that some gemstones continue to be radioactive after treatment (and, in some cases, after simply being taken from the ground), only in very rare instances would these gems pose any health hazard.

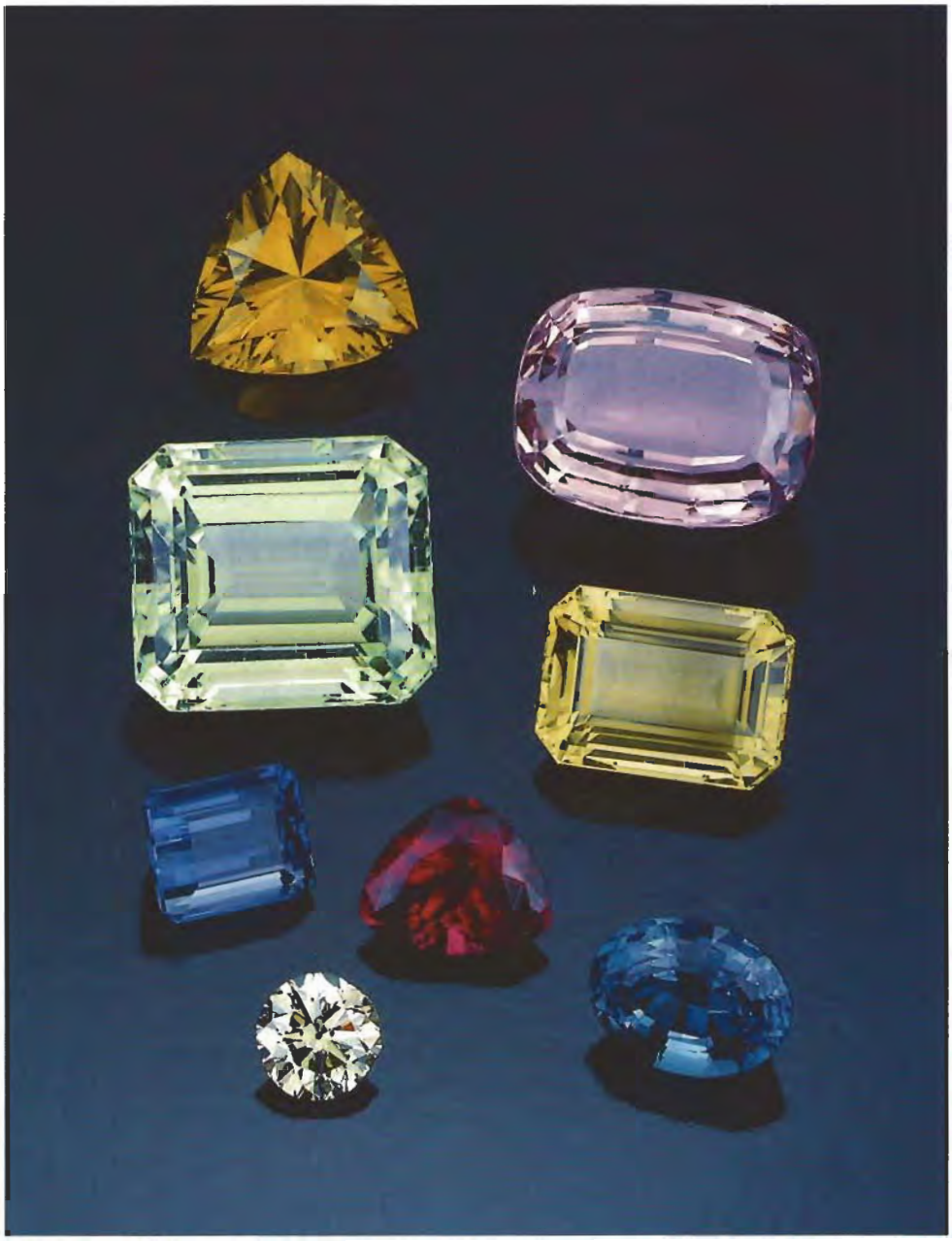


Figure 1. A variety of gems are now being irradiated for color enhancement. Illustrated here are (clockwise from top right) irradiated kunzite, citrine, blue topaz, rubellite, diamond, Maxixe beryl, green spodumene, and yellow beryl. The largest stone, the green spodumene, is 48.68 ct. Stones courtesy of George Drake and the GIA collection. Photo © Tino Hammid.

NATURE OF RADIATION

Radiation can be simply defined as energy emitted in the form of particles or electromagnetic waves. It is given off by a variety of sources. For example, an ordinary light bulb, a fire, and a disintegrating radioactive atom all give off energy as some form of radiation. However, only a particular type or class of radiation, known as ionizing radiation, has enough energy to disrupt and dislodge electrons and sometimes atoms within a gem crystal. As the ionizing radiation passes through, it imparts this energy to the crystal, thereby creating color centers (see Nassau, 1983; Fritsch and Rossman, 1988.)

During the decay of radioactive atoms or nuclides, one or more of four basic types of ionizing radiation are released:

Alpha particles: High-speed helium atoms without any electrons.

Beta particles: High-speed electrons.

Gamma rays: High-energy photons of electromagnetic radiation identical to X-rays. (X-rays originate from electrons and are nor-

UNITS OF MEASUREMENT

Becquerel (Bq): Radioactive content of a material equal to one nuclear disintegration or transformation per second.

Curie (Ci): Radioactive content of a material (originally defined as the amount of radioactivity in one gram of radium-226) equal to 37 billion Bq.

1 nanocurie (nCi) = 1 billionth of a curie = 37 Bq; 1 Bq = 0.027 nCi

Roentgen (R): Unit of exposure that defines the ability of radiation to ionize air (i.e., remove electrons from atoms and molecules). It is the quantity of X-ray or gamma-ray radiation able to produce 2.580×10^{-4} coulombs of charge in one kilogram of dry air (1 coulomb = 6.24×10^{18} [billion-billion] electrons worth of charge). 1 microroentgen (μr) = 1 millionth of a roentgen.

Rad (Radiation Absorbed Dose): Unit of absorbed dose. The quantity of radiation able to deposit 100 ergs of energy into a gram of any material. This unit is independent of the type and energy of the radiation. (An erg is a unit of energy equivalent to a force of one dyne [force able to accelerate a one gram object 1 cm per second per second] moving 1 cm.)

1 Mrad = 1 million rads

SI unit: Gray (Gy); 1 Gy = 100 rad

Rem (Roentgen Equivalent Man): Unit of biological radiation dose equivalent that places on a common scale the biological damage produced by ionizating radiation. A rem equals a rad multiplied by a quality factor that varies between 1 and 20 depending on the type and energy of the radiation.

1 mrem = 1 thousandth of a rem

SI unit: Sievert (Sv); 1 Sv = 100 rem

A simple analogy may help to explain these terms. Imagine going to the beach on a sunny day. Becquerels or curies would represent the total photon or light output of the sun, roentgens would relate to the amount of ultraviolet sunlight at the beach, rads would correspond to the energy absorbed by the sunbather's skin, and rems would be a measure of the amount of tan (or sunburn) produced. For beta and gamma radiation, the roentgen, rad, and rem are relatively equivalent radiation exposure and dosage units.

mally lower in energy, while gamma rays originate from the nucleus and are generally higher in energy.)

Neutrons: Neutral subatomic particles that are rarely found outside the nucleus of an atom and have a weight about the same as that of a simple hydrogen atom.

A *nuclide* is a type of atom that is defined by the number of protons and neutrons in the nucleus. There are roughly 1,400 different nuclides known to date. Radiation energy is described in units of electron volts (eV), which is the energy or energy equivalent of a subatomic particle that has been accelerated through an electric potential of one volt: 1 KeV = 1 thousand eV, 1 MeV = 1 million eV. Several terms are used to quantify radioactivity and radiation effects—*becquerel, curie, roentgen, rad,* and *rem* (see box). Because SI units (International System of Units) are also used in the literature, although not in this article, these equivalents are provided as well.

With respect to the possible health hazards of radioactive gemstones, we have to be concerned only with gamma radiation. Alpha radiation is essentially nonpenetrating and can be stopped by a piece of paper, while neutron radiation is virtually nonexistent outside neutron-producing devices. Beta particles are mostly absorbed within the gemstone itself, and what does exit the gemstone is weak and produces only a shallow, relatively harmless skin dose. Generally, only gamma radiation has the potential to affect those who wear or otherwise come into contact with a radioactive gem.

RADIATION IN NATURE

We exist in a "sea" of natural background radiation. This radiation originates from the decay of radioactive elements such as uranium, thorium, and the nuclide potassium-40 that are naturally present in the earth's crust; from cosmic radiation; from radon gas in our homes; and so on. Natural radioactivity is exhibited by more than 50 naturally occurring radioactive nuclides (Villforth and Shultz, 1970). Consequently, low levels of radiation are always found in the air, water, and ground. However, only terrestrial background radiation is relevant to the coloration of natural gemstones.

Terrestrial Background Radiation. Uranium-238 (U-238), thorium-232 (Th-232), and potassium-40

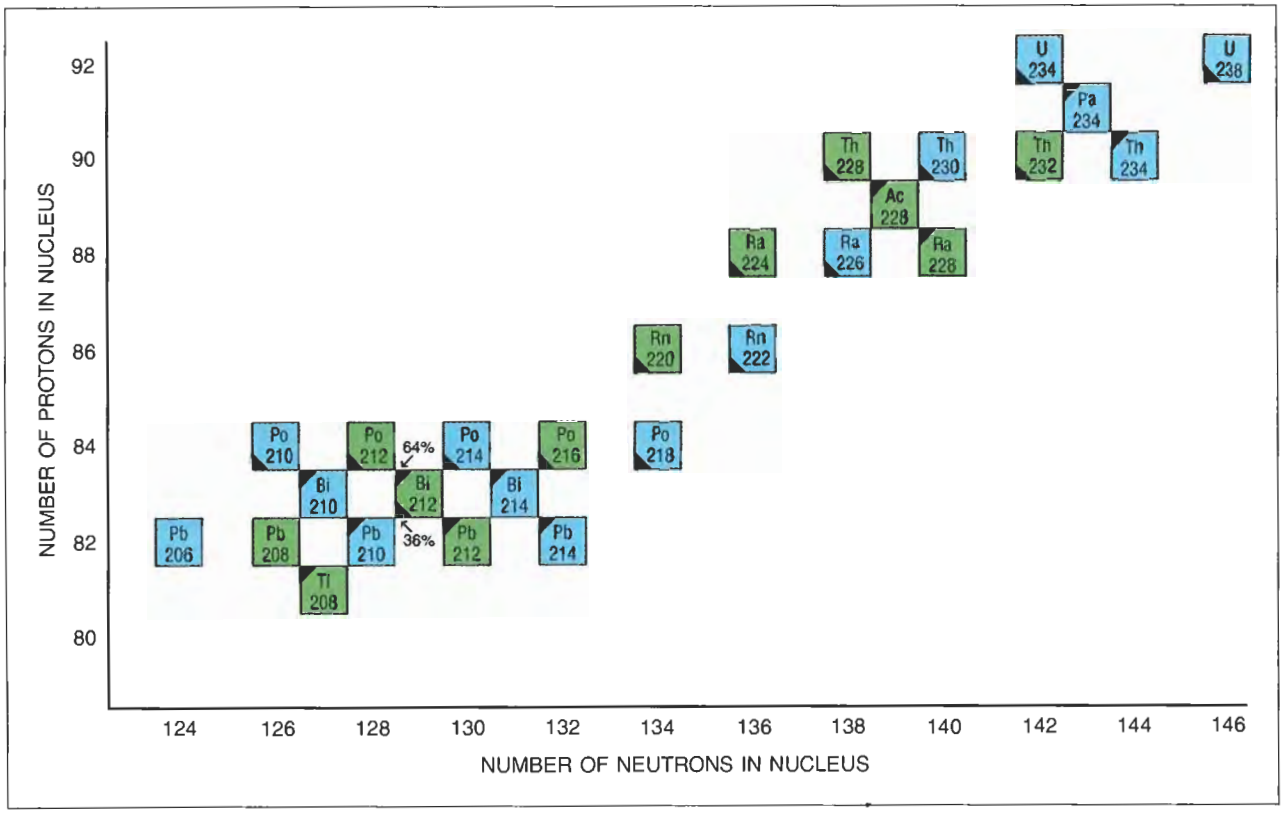


Figure 2. The decay chains of uranium-238 and thorium-232 show the major alpha and beta decays (as indicated by the black corner arrows) from parent to stable lead. Gamma emission is relatively weak for all nuclides except Pb-214, Bi-214, Ac-228, and Tl-208.

(K-40) are the three major radioactive nuclides found in the earth's crust. Ever since their origin inside exploding stars (long before their incorporation into primordial earth), these nuclides have undergone continuous radioactive decay or spontaneous disintegrations. In doing so, they constantly give off alpha, beta, and gamma radiation. Any radioactive atom or nuclide decays by the emission of radioactive particles or rays in a specific sequence of steps. At each step, a particular new type of atom or nuclide is formed until one is created that is not radioactive and does not decay any further. The length of time for one-half of a group of atoms of a particular type to decay into another type is called the half-life. Each nuclide has a specific half-life which can vary from much less than a second to as long as billions of years.

Over 99% of natural uranium is composed of the nuclide U-238, while nearly 100% of natural thorium is Th-232. These nuclides are referred to as "parents"; they decay by the emission of alpha particles which lead to a sequence of a dozen or so radioactive decay products called "daughters." These two radioactive decay sequences release many alpha, beta, and gamma rays before U-238 and Th-232 arrive at their respective nonradioactive daughter isotopes of lead (figure 2). For example, U-238 decays by alpha emission to Th-234, then Th-234 decays by beta emission to protactinium-234 (Pa-234), and so on.

Potassium-40 comprises about one-hundredth of one percent of natural potassium (which is much more abundant in nature than uranium and thorium). This nuclide decays by the emission of a beta particle followed, 10% of the time, by a high-energy gamma ray. The result is the stable, nonradioactive daughter calcium-40.

Naturally Radioactive Gems. Uranium, thorium, and potassium occur in many minerals and gemstones (Fleischer, 1986). When uranium and thorium are present, enough time has usually elapsed since the material crystallized that radioactive equilibrium has been established between parents and daughters. For example, if a gemstone contains 1 nCi/gm of U-238, it also contains 1 nCi/gm of each of its 13 radioactive daughters. For both uranium and thorium, though, only a couple of the daughters produce the bulk of the energetic beta and gamma radiation.

Potassium is found in many gem materials, including feldspars such as orthoclase and labradorite. Since the nuclide K-40 has such a long half-life and makes up such a small percentage of the natural element potassium, gemstones containing potassium rarely contain more than about 0.1 nCi/gm of K-40. (Pure potassium contains only 0.855 nCi/gm of radioactivity.)

Although U-238, Th-232, and K-40 generally occur as minor trace elements, they may be present in rather high concentrations. Table 1 lists various naturally radioactive gem materials in increasing order of radioactivity; most exceed the legal U.S. release limits for manufactured items containing uranium (0.168 nCi/gm) and thorium (0.055 nCi/gm). Although the emerald-cut thorianite is primarily a rare collector's item, it does illustrate just how radioactive natural gemstones can be (in this case, more than 1,500 times the stated legal limit for manufactured goods containing uranium). GIA has examined gemstones of similar radioactive content in the recent past (Editorial Forum, 1987). The author measured all of the zircons in figure 3 and found them to be radioactive.

TABLE 1. Naturally radioactive gem materials.[a]

Gemstone	Nuclide	Concentration[b] (nCi/gm)
Zircon[c]	U-238	
red, yellow, blue		0.03 to 0.3
green		0.5 to 2.1
Ekanite[d]	Th-232/U-238	26/6.5
Euxenite	U-238	29
Fergusonite	U-238/Th-232	40/3
Thorianite	Th-232/U-238	87/19

[a] *Measured by the author.*
[b] *The legal release limit for manufactured items containing uranium is 0.168 nCi/gm and thorium, 0.055 nCi/gm. Because the radioactivity is naturally occurring, such gemstones are not subject to any form of regulation.*
[c] *Sometimes includes trace amounts of Th-232 and daughters. Similar values were reported by Müllenmeister (1986).*
[d] *Similar values were reported by Perrault and Szymanski (1982); see also Fryer et al. (1986).*

Figure 3. Many gemstones are naturally radioactive. In fact, radioactivity was measured in all of the zircons illustrated here (the largest is 4.77 ct). Photo © Tino Hammid.

Naturally Irradiated Gemstones. Terrestrial background radiation in the host rocks of a gem deposit can alter the color of the gem material if the radiation dose is high enough and the ambient temperature low enough. That is, the gem material must be in close proximity to a sufficient amount of radioactive nuclides for a long enough time and at a temperature that will not anneal or bleach out the radiation-induced color.

For example, tourmalines from gem pegmatites become pink or red from exposure to high-energy (1.46 MeV) gamma rays from K-40 over periods of millions of years (Reinitz and Rossman, 1988). The natural blue color of some topaz is thought to be produced by natural irradiation, as is the deep blue color of Maxixe beryl and some fluorites (Rossman, 1981). Surface coloration of yellow and yellow-green diamonds has also been attributed to natural radioactivity (Dugdale, 1953). The presence of radiation from uranium produces the color centers responsible for red in zircons (Fielding, 1970). The color of blue-green amazonite is also radiation induced (Hofmeister and Rossman, 1985). In addition, radioactive solutions in gem deposits can produce color, as in smoky quartz (Koivula, 1986). Although this list is not exhaustive, it does show the magnitude of gemstone coloration by natural radiation. As research

progresses, more gemstones may prove to owe their color and beauty to natural terrestrial background radiation.

GEMSTONE IRRADIATION IN THE LABORATORY

It was not long after natural "radioactivity" was discovered by A. Henri Becquerel in 1896, and the term coined by Marie Curie in 1898, that gemstones began to be treated with radiation. Published reports of diamond coloration by radium treatment appeared as early as 1909 (Crookes, 1909). Today, a wide variety of gems are color enhanced by laboratory irradiation (table 2), including many fancy-color diamonds (figures 4 and 5).

Radiation produced in the laboratory is often more efficient in the coloration of gem materials than natural terrestrial background radiation. The higher radiation energy and doses obtainable can produce more desirable coloration, and the temperature conditions can be controlled so that the induced colors are not annealed during the process. Currently, three major types of laboratory radiation sources are used to irradiate gemstones: cobalt-60 facilities, which produce gamma radiation; linear accelerators, which generate high-energy electrons; and nuclear reactors, which produce high-energy neutrons.

TABLE 2. Effects of irradiation treatment on various gem materials.[a]

Material	Starting color	Ending color
Beryl	Colorless	Yellow
	Blue	Green
Maxixe-type	Pale or colorless	Blue
Corundum	Colorless	Yellow
	Pink	Padparadscha
Diamond	Colorless or pale to yellow and brown	Green or blue (with heating, turns yellow, orange, brown, pink, red)
Fluorite	Colorless	Various colors
Pearl	Light colors	Gray, brown, "blue," "black"
Quartz	Colorless to yellow or pale green	Brown, amethyst, "smoky," rose
Scapolite[b]	Colorless, "straw," pink, or light blue	Blue, lavender, amethyst, red
Spodumene	Colorless to pink	Orange, yellow, green, pink[c]
Topaz	Yellow, orange	Intensify colors
	Colorless, pale blue	Brown, blue (may require heat to turn blue), green
Tourmaline	Colorless to pale colors	Yellow, brown, pink, red, bicolor green-red
	Blue	Purple
Zircon	Colorless	Brown to red

[a] *Adapted from Nassau (1984).*
[b] *Charles Key, pers. comm., 1988.*
[c] *George Drake, pers. comm., 1988.*

Figure 4. Diamonds were the first gemstones to be irradiated. Today, irradiation produces a variety of fancy colors in diamond, of which these are only a few. Stones courtesy of Theodore and Irwin Moed; photo © Tino Hammid.

Gamma-Ray Facilities. A typical gamma-ray facility (figure 6) is essentially a heavily shielded concrete room that contains up to several million curies of cobalt-60 (Co-60). The radioactive material is first encapsulated and then sealed inside hundreds of small stainless steel rods called pencils, which are grouped together to form modules (Wallace Hall, pers. comm., 1988). During irradia-

Figure 5. Irradiated diamonds are increasingly seen in fine jewelry. An example is the fancy-color irradiated pear-shaped diamond in this attractive geometric necklace that was recently sold at auction. Photo by Tino Hammid; courtesy of Christie's New York.

tions, the cobalt sources are raised up out of the shielding water to expose the subject material to the gamma rays.

During radioactive decay, Co-60 releases a beta particle and two gamma rays, with energies of 1.17 and 1.33 MeV respectively, in quick succession. The beta particles from the Co-60 do not contribute to the radiation dose since they are completely absorbed within the pencils. This type of facility can achieve gamma-ray dose rates as high as several Mrads per hour.

Many types of gemstones are currently irradiated with gamma rays to produce or improve their color. Colorless quartz is irradiated to produce smoky quartz. Iron-containing quartz is irradiated to produce amethyst and then heat treated to produce citrine (Rossman, 1981). Light pink tourmalines are irradiated to a couple hundred Mrads (Camargo and Isotani, 1988) to produce dark pink to red stones (figure 7). Gamma irradiation is also used to increase the color saturation of red zircons (Mike Gray, pers. comm., 1987). Near-colorless topaz is irradiated to several hundred Mrads and higher to produce a light blue color, called "Cobalt Blue" in the trade, and to even higher doses for a steely gray-blue color. Gamma irradiation is sometimes also used to screen out unwanted material such as beryl or quartz, or to prescreen topaz for subsequent treatments, since material that turns light blue under these conditions is likely to become much darker blue with additional radiation exposure from high-energy electrons.

Linear Accelerators. A linear accelerator, or "linac" (figure 8), is an electron "gun" that fires a pulsed beam of electrons at energies of 10 to 15 MeV and at a current of several hundred microamperes, producing dose rates to gemstones up to and exceeding several hundred Mrads per hour. At these dose rates, the gem materials must be water cooled to prevent elevated temperatures and ther-

Figure 6. This view down into a gamma-ray facility shows the many modules positioned on two racks held by steel cables that have been lowered into the storage pool. During irradiations, the racks are raised up out of the shielding water. Photo courtesy of Radiation Sterilizers, Tustin, CA, and Pirih Productions, Pasadena, CA.

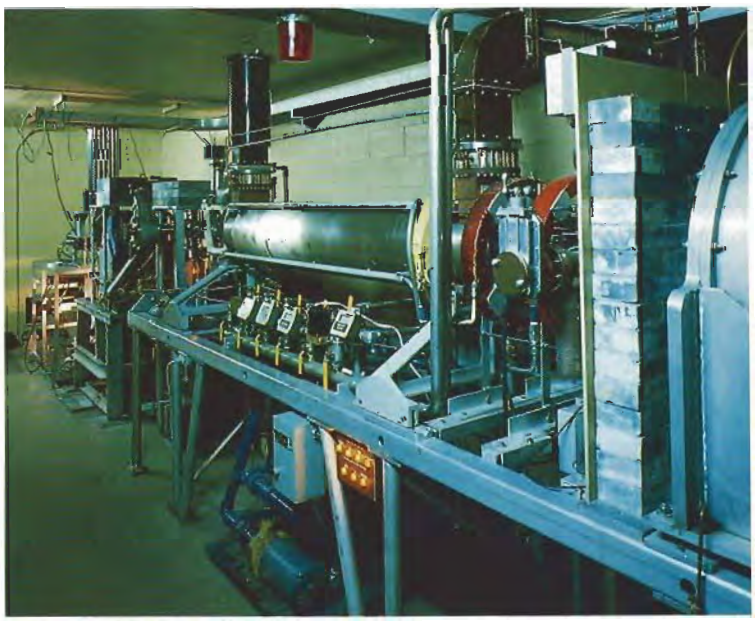
Figure 7. Today, pale colored tourmalines similar to those on the left are commonly irradiated with gamma rays to produce darker stones, like the irradiated tourmalines on the right (the largest is 5.91 ct). Stones courtesy of George Drake and the GIA collection; photo © Tino Hammid.

mal shock. High temperatures will anneal or destroy the color centers in the gem materials, while thermal shock will crack or shatter them. For instance, topaz receiving a typical dose rate of 250 to 500 Mrads per hour will increase in temperature at the rate of 50°C to 100°C per minute if it is not properly cooled.

Some linacs produce electron beams with maximum energies up to about 3 MeV, values on the order of the maximum energies of beta particles from radioactivity. Such facilities are useful only for irradiating small gemstones because the electron beam will not completely penetrate and provide an even radiation dose throughout stones over a few carats in size. If the electron beam stops within the gemstone, a large negative static charge can build up which can cause the stone to fracture or even shatter. By contrast, beam energies above 15 MeV are not generally used for gem materials because such high energies can induce radioactivity within the material.

Normally, gem materials that are irradiated in gamma-ray facilities, such as topaz, tourmaline, zircon, and diamond, can also be irradiated in linacs. Linacs are usually preferred for gemstones that require relatively high doses of radiation. Yellow beryl and blue topaz, for example, generally require irradiation doses from 1,000 to 10,000 Mrads (George Drake, pers. comm., 1988). Also, treatment in a linac will often produce much deeper colors, undoubtedly because higher-energy electrons deposit their energy inside the stones; with gamma rays, lower-energy electrons are produced as these rays travel through or are absorbed

Figure 8. Literally millions of carats of gemstones have been irradiated in linear accelerators (linacs) such as this one. Photo courtesy of IRT, San Diego, CA.



into the material. Currently, topaz is the most frequently irradiated gemstone; the linac-irradiated material becomes an attractive "Sky Blue" after subsequent heat treatment of the greenish or brownish product.

Nuclear Reactors. Nuclear research reactors found at universities and other research centers produce high-energy neutrons along with associated gamma rays. A typical research reactor consists of a large metal or concrete reactor vessel that contains a matrix of uranium fuel rods or plates, control rods for power control, water for cooling and neutron energy tailoring, and ports where materials can be placed for irradiation (figure 9).

Figure 9. This photo was taken looking down into the pool of a TRIGA nuclear research reactor. The bluish glow seen here and in the gamma-ray facility in figure 6 is called Cherenkov radiation. It is produced by gamma rays interacting with the electrons of the water molecules and causing them to very briefly travel faster than light does in water. The electrons must slow down, and consequently lose energy by emitting photons of light. Photo courtesy of General Atomics, San Diego, CA.

Since all gem materials become radioactive when bombarded by neutrons, residual radiation is a serious problem for reactor-irradiated gemstones (but not for gems properly selected and irradiated by the other two methods described above).

Currently, near-colorless topaz is the gemstone most often irradiated in a reactor. Therefore, the following discussion will focus on the reactor irradiation of topaz (called "London Blue" in the trade), although the comments apply equally well to other gems, such as spodumenes (Rossman and Qiu, 1982) and diamonds (Dugdale, 1953).

Theoretically, a few weeks after neutron irradiation in a reactor facility, chemically pure topaz would not be radioactive because the neutron-activated major constituents of topaz (fluorine, aluminum, oxygen, and silicon) have half-lives of only seconds to hours. Topaz from various localities, however, is likely to contain different concentrations of trace-element impurities such as tantalum, scandium, and manganese. Isotopes of these elements (Ta-182, Sc-46, and Mn-54) are the longer-lived radioactive nuclides found in reactor-treated topaz (Crowningshield, 1981).

High-energy (fast) neutrons produce the important color centers in topaz as well as most of the crystalline damage; low-energy (known as thermal, or slow) neutrons produce most of the radioactive nuclides. Therefore, to reduce the amount of neutron-induced radioactivity, gem treaters try to maximize the fast-neutron, and minimize the slow-neutron, components irradiating their material. They can accomplish this by placing the topaz in containers covered with materials such as cadmium or boron compounds that very easily absorb slow neutrons. They can also surround the containers holding the topaz with uranium converter plates; only the fast neutrons will get through the plate to the topaz, while the slower neutrons will be absorbed by the uranium in the plate, causing fissions that produce more high-energy neutrons to irradiate the topaz. In addition, since cooling water for topaz slows neutrons down to lower energies, the treaters try to use either as little water as possible, or a gas such as nitrogen for cooling. Because there are important differences in the designs of various nuclear reactors, and because of the variability in impurities among topaz from different localities, reactor irradiation can yield a variety of results in both induced color and induced radioactivity.

A relatively recent development in topaz en-

hancement is to combine treatment processes to produce a "brighter" blue color. The topaz is irradiated first in a nuclear reactor and then in a linac, after which it is heat treated (Fournier, 1988). The product is referred to in the trade by names such as "American," "Electra," "California," "Swiss," or "Super Blue" topaz. Figure 10 illustrates three types of irradiated blue topaz. Heat treating is applied to gamma-ray and electron-beam treated topaz to remove the undesirable and less stable brown and green colors. Heat treatment is generally not required for neutron-treated topaz except to remove the tell-tale inky or steely component and produce a lighter shade of blue (Schmetzer, 1987). For a further explanation of topaz treatments, see Nassau (1985).

Figure 10. Far more topaz than any other gem material is being irradiated in laboratory facilities. The American Gem Trade Association (Willett, 1987) has estimated that 6,000 kg (30 million carats) of topaz are irradiated annually, 40% of this total in the U.S. Each of the stones shown here was treated by a different method (from lightest to darkest): linac-treated "Sky Blue," combination reactor- and linac-treated "California Blue," and reactor-treated "London Blue." Photo © Tino Hammid.

INDUCED RESIDUAL RADIOACTIVITY IN LABORATORY-IRRADIATED GEMSTONES

Atoms become radioactive whenever there is an excess of energy in their nuclei, brought on by a nuclear reaction or by the disruption of the proper ratio of protons to neutrons. They shed this energy by undergoing some type of radioactive decay or radiation emission. A nuclear reaction between the nucleus of an atom and a subatomic particle or photon can provide that excess energy. Radioactivity is induced by two out of the three treatment processes discussed, linac and nuclear reactor, although the latter poses the potentially most serious problem.

Linac Induced. High-energy electrons produce X-rays, called *bremsstrahlung* or braking radiation, as they lose energy going through a material. Some of these X-ray photons will have enough energy to enter the nucleus of an atom and make it radioactive. This is called photoactivation. A subatomic particle such as a neutron can be released in this nuclear reaction (called a photoneutron reaction) and proceed to enter another gemstone nuclide and make it radioactive. For instance, an X-ray photon can interact with a sodium-23 nuclide and transform it into sodium-22. The free neutron released during this nuclear reaction can go on to make another nuclide radioactive, transforming, for example, cesium-133 into cesium-134.

Photoneutron reactions are produced only above a certain energy and thus are called threshold reactions. They generally occur with photons of energies of from 7 to 18 MeV. As a general rule, the lower in atomic weight the atom is, the higher the photon energy needed to cause the reaction, the fewer the number of neutrons released, and the shorter the half-life of the new radioactive atom (De Voe, 1969). For most gem materials, if the electron-beam energy is kept below 12 MeV, the half-lives of the induced radioactive nuclides are short enough that the radioactivity decays to background levels within a few weeks and only an insignificant number of neutrons are produced.

Gemstones containing the elements beryllium (such as beryls), lithium (such as spodumene and some tourmalines), and uranium and thorium (such as zircons) produce neutrons during linac irradiation; all of these elements have low energy thresholds for photoneutron reactions. The lowest, for beryllium, is only 1.67 MeV (Berman, 1974).

For example, the author has measured sodium-22 and cesium-134 in linac-treated beryls, and manganese-54 in linac-treated tourmalines, which testify to the photoneutron and neutron-absorbing reactions that have taken place. As for topaz irradiated properly in a linac, the most significant radionuclide produced is believed to be germanium-69, the result of a photoneutron reaction with the stable nuclide germanium-70. Since this nuclide has a half-life of 39 hours, the gemstones are not released for many half-lives (a few weeks) to allow the activity to die out (Robert Block, pers. comm., 1988).

Reactor Induced. Nuclear reactors produce copious amounts of neutrons which vary in energy from a hundredth of an eV to over 10 MeV. Unlike most other nuclear reactions with the nuclei of atoms, neutron absorption occurs at all energies but becomes much easier at thermal neutron energies. A neutron is not affected by the positive and negative charges of atoms, as are alpha, beta, and gamma radiation. Therefore, a neutron can enter a nucleus relatively easily, depending on the type of nuclide, and alter the neutron-to-proton ratio. The nuclide then has too many neutrons in its nucleus, and thus undergoes beta decay to correct this imbalance; that is, a neutron turns into a proton and a beta particle is released from the nucleus, normally along with a gamma ray. Consequently, much larger amounts of induced radioactivity (i.e., beta- and gamma-emitting nuclides) are generated per Mrad during irradiation in a reactor.

DETECTION AND MEASUREMENT OF RADIATION

As radiation travels through a material, it loses its energy to the surrounding atoms by ionization, where atoms and molecules lose one or more electrons, and by electron excitation, where electrons are raised to higher energy levels. These processes can lead to other effects such as altering the electrical resistance of a material, producing minute flashes of light, causing slight temperature increases, and so on.

To detect or measure radiation, one takes advantage of one of these effects and either amplifies it or uses it as a gauge or reference. There are many kinds of instruments and methods used to detect and measure radiation. Three types of instruments—the Geiger counter, the sodium iodide scintillation crystal, and the lithium-drifted germanium detector—are particularly useful for measuring radioactive gem materials. For further information on these and other radiation detection instruments, see Knoll (1979).

Geiger Counter. This is perhaps the most commonly recognized and widely used radiation detection instrument (figure 11). Geiger counters are inexpensive, ruggedly built, easy to operate, and work relatively well for measuring low levels of beta radiation and somewhat higher levels of gamma radiation. A G-M (Geiger-Müller) tube, the detector part of a Geiger counter, is a hollow gas-filled metal tube with a charged wire running down its center. As radiation passes through the tube, it causes ionizations in the gas. Because of the high voltage within the tube, each single ionization gives rise to millions of additional ionizations, producing an electrical pulse that is then converted to an audible "click."

The response of a Geiger counter depends on the type and the energy of the radiation. Essentially, a single beta particle has anywhere from less than 1% to as much as a 20% chance of being

Figure 11. The most common, and least expensive, radiation detector is a Geiger counter such as this Victoreen Model 290 Survey Meter with a Model 489 pancake probe. Photo courtesy of Victoreen, Inc., Cleveland, OH.

TABLE 3. Level of radioactivity detectable in various gemstones with a Geiger counter.[a]

Gemstone	Weight (ct)	Radio-nuclides present[b]	Activity[b] (nCi)	Direct readings[c] (cpm)	nCi of detectable radioactivity[d]
Green zircon	2.15	U-238[e]	0.75	≃ 300	0.12
Green zircon	3.71	U-238[e]	0.5	≃ 200	0.13
Ekanite	3.86	U-238[e]	5		
		Th-232[e]	20		
		Total	25	≃ 5500	0.18
Blue topaz (neutron irradiated)	4.62	Sc-46	0.75	≃ 75	0.86
Blue topaz (neutron irradiated)	4.72	Ta-182	2.0	≃ 110	1.1

[a]*Victoreen Model 290 Survey Meter with a pancake probe Model 489-110B (see figure 11). Data generated by Dr. Emmanuel Fritsch of GIA, November 17, 1988. This instrument is able to detect alpha particles above 3.5 MeV, beta particles above 35 KeV, and gamma rays above 6 KeV.*
[b]*As measured with NaI and Ge(Li) systems.*
[c]*Average of visible readings—background is approximately 40 counts per minute (cpm).*
[d]*Nanocuries of radioactivity needed to cause direct readings to be twice background (i.e., 80 cpm).*
[e]*U-238 and Th-232 include daughters.*

counted, because nuclides emit beta particles over a broad energy spectrum and only the highest ones have any likelihood of arriving inside the G-M tube to be registered. A gamma ray has less than a 0.1% to 2% chance of being counted, because a gamma ray must interact within a critical layer in the wall (cathode) of the G-M tube to be registered. In general, then, a Geiger counter is less than 5% efficient in quantifying radioactivity. However, as shown in table 3, the Geiger counter does indicate whether radioactivity is present within a certain range. Note also that the response per nCi depends on the type of nuclide being measured and the size of the stone. For example, stones of a few carats would have to contain at least 0.1 to 0.2 nCi of uranium and thorium, or about 1 nCi of typical mixed by-product nuclides, to double the count rate above that of the background and therefore be measurable with any confidence within a short period of time (e.g., half a minute). Smaller stones with similar amounts of radioactivity should be more easily measured.

NaI Scintillation Crystal. A more refined and sensitive method to measure radiation is through the use of a scintillation crystal such as sodium iodide—thallium doped to 0.1%, NaI(Tl), set within a low-background lead shield. When a gamma-ray enters this crystal, it loses its energy by causing electrons to be moved out of their normal sites in the structure of the NaI(Tl) crystal. These electrons then move very quickly to lower-energy levels at the thallium activator sites, producing tiny pulses of visible fluorescent light (photons) that combine to form one single large light pulse, the intensity of which is proportional to the energy of the absorbed gamma ray. Each type of radioactive atom releases gamma rays at specific energies. When a scintillation crystal is coupled to a photomultiplier tube, which in turn is connected to an analog-to-digital converter and the output manipulated by a computer, it is possible to determine the type and amounts of radioactive nuclides present in a material (again, see table 3). Note, however, that this system is considerably more expensive than a good Geiger counter and requires technical expertise to operate and interpret the data.

Lithium-Drifted Germanium Detector. The best method for measuring a large number of different gamma rays at a single time is with a semiconductor detector. The most popular is the lithium-drifted (doped) germanium detector, Ge(Li), which produces electron-hole pairs when gamma rays are absorbed into it. The motion of these pairs in an applied electrical field generates the basic electrical signal that is fed into a multichannel analyzer (Knoll, 1979). The Ge(Li) most clearly distinguishes between different gamma energy peaks for different radioactive nuclides, but it is less sensitive, requires constant liquid nitrogen cooling, and is much more expensive than the NaI system. Both systems use the same type of multichannel an-

Figure 12. The ND 65B Multi Channel Analyzer (MCA) shown here is a pulse-height analyzer that converts electrical signals from either a NaI or a Ge(Li) detector into useful information such as the type and quantity of radioactive nuclides present in a gemstone. Courtesy of Nuclear Data, Inc., Schaumburg, IL.

alyzer (figure 12). Figure 13 compares the energy spectra for two nuclides generated by the NaI and the Ge(Li) systems. The areas under the peaks or curves on the screen are proportional to the concentrations of the radioactive nuclides.

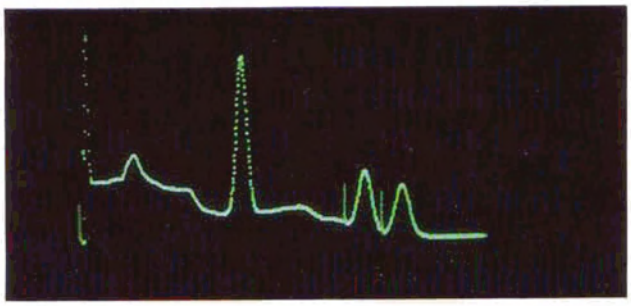
SOME CONSIDERATIONS REGARDING HEALTH HAZARDS

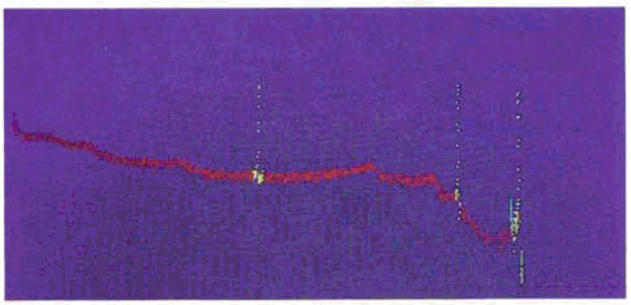
What are the safe limits of radiation and how hazardous are radioactive gems? The answer is highly controversial. Some maintain that any and all radiation is harmful, others feel that low levels are harmless since people live in a sea of background radiation, and still others regard even relatively high levels of radiation as safe.

Figure 14 gives the current estimate of the contributions of background radiation that an average American receives per year (Sinclair, 1987). The total average whole-body dose equivalent is about 360 mrem per year, or 1 mrem per day. In other parts of the world, it may reach as high as 3,000 mrem per year (Eisenbud, 1987). Whole-body dose equivalent can be illustrated as follows: From radon we receive 2,400 mrem per year to the lung tissue; this is the equivalent of about 200 mrem to the entire body. To place these numbers in perspective, those who work in the U.S. radiation industry are allowed by government regulations to receive up to 5,000 mrem to their entire body, 30,000 mrem to the skin of the whole body, and up to a total of 75,000 mrem per year to their hands, forearms, feet, and ankles (U.S. Code of Federal Regulations, Title 10, Part 20, Section 20.101 – 10 CFR 20). These dose levels are considered acceptable (safe) for workers. The 75,000 mrem is comparable to wearing radioactive rings, watches, or bracelets that produce dose rates to these relatively small areas that are more than 8.5 mrem per hour, 24 hours per day, 365 days per year. For comparison, dental X-rays can give doses from 100 mrem to as high as 1,500 mrem.

Table 4 quantifies the variables involved in determining the potential health hazard of a gem material. The half-life and the decay mode of a radioactive nuclide in a gemstone determine its relative radiation hazard. The total gamma-ray energy (γ KeV) emitted per radioactive decay can help quantify the relative gamma-ray dose that a person will receive from a radioactive gemstone. Because radiation doses to humans are very difficult to compute and vary with depth of penetration into tissue, they can be more closely approximated

Figure 13. These two displays illustrate the gamma-ray spectra of cesium-137 (0.662 MeV) and cobalt-60 (1.17 MeV and 1.33 MeV) as shown using a NaI scintillation crystal with an ND 6 multi-channel analyzer (left) and a Ge(Li) detector with an ND 76 multi-channel analyzer (right). Photos courtesy of Nuclear Data, Inc., Schaumburg, IL.





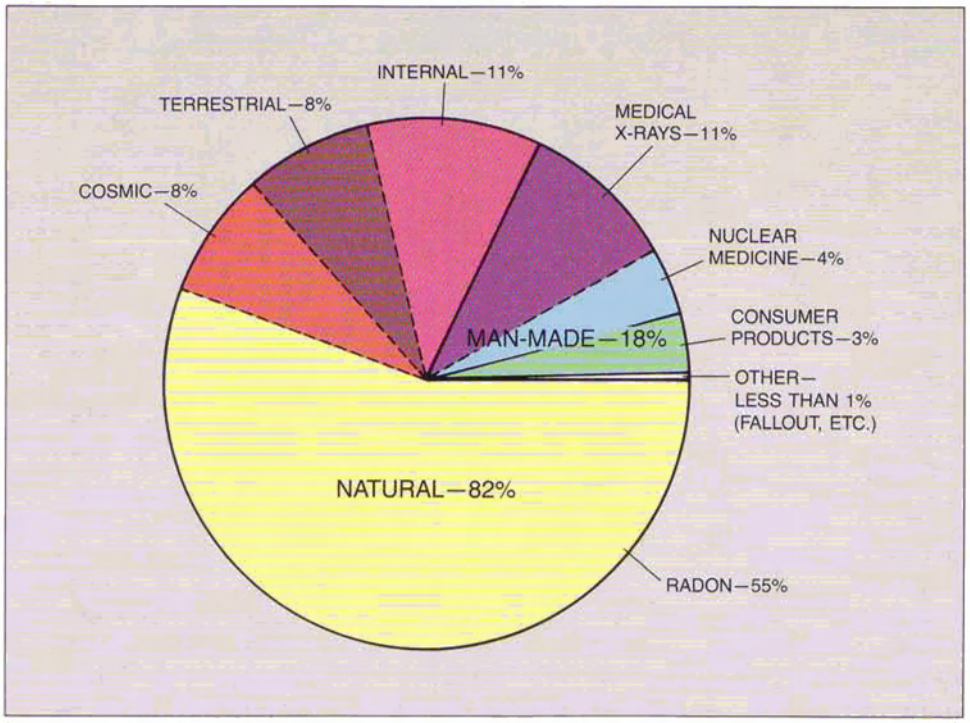


Figure 14. This illustration shows the percentage contribution of various radiation sources to the background radiation whole-body dose equivalent that an average American receives in a year (360 mrem/yr). Adapted from Sinclair, 1987.

by the specific gamma-ray constant (SGRC). This gives the gamma-ray exposure rate in air in microroentgens (μr) per hour, 1 cm from a 1-nCi-point (very small) source.

Consider a series of gemstones, each of which contains a different radioactive nuclide yet produces the same gamma-ray exposure. If we use the data in table 4, and divide each SGRC for uranium-238 and daughters by the SGRC for each nuclide, we get a term for each nuclide called the inverse relative exposure rate index, IRERI. This value is equal to the number of nCi of a nuclide that produces the same exposure rate as one nCi of U-238. Therefore, a gemstone that contains 3 nCi/gm of zinc-65 (Zn-65) or antimony-125 (Sb-125) produces the same exposure rate per hour as a similar gemstone containing 1 nCi/gm of U-238. The relative exposure rate is simply the inverse of the IRERI; that is, for example, Zn-65 and Sb-125 are one-third as radiant as U-238.

When time, half-lives, and the resultant decreases in radioactivity are taken into consideration, the indices will become even more varied. Assuming that the average lifetime or ownership of a typical piece of jewelry is 10 years, the original concentration of each nuclide that would give the same radioactive exposure as 1 nCi of U-238 would be the inverse relative exposure index, IREI. For example, for scandium-46 or tantalum-182, the most prevalent radionuclides in reactor-treated blue topaz, the concentration would be about 25 nCi/gm. In other words, two stones of the same size, a green zircon containing 1.0 nCi/gm of uranium-238 and a blue topaz containing about 25 nCi/gm of either scandium or tantalum, would give roughly the same total radiation exposure over a 10-year period.

Release limits are concentration limits of radioactive nuclides contained in material made radioactive by man that are allowed to be released to the general public. The concentrations are deemed safe and are based on political issues as well as scientific data. The values for various nuclides are found in U.S. Code of Federal Regulations Title 10, Part 30 (10 CFR 30), Section 30.70, Schedule (Table) A. For example, for scandium-46 or tantalum-182, the limit is 0.4 nCi/gm. If a gemstone contains more than one type of nuclide, the sum of the ratios (the concentration divided by the release limit concentration for each isotope) must be less than or equal to 1.

Accordingly, we can see that with respect to long-lived natural radioactive nuclides and shorter-lived man-made radionuclides, the energy and type of decay, as well as the SGRC and half-lives, must be taken into account to arrive at a proper perspective when considering radiation exposures, doses, and release limits for radioactive gemstones.

U.S. Code of Federal Regulations Title 10, Part 40 (10 CFR 40) states that any person is exempt from license requirements for materials manufac-

TABLE 4. Radioactive nuclide data for radioactive gemstones.[a]

Nuclide	Half-life[b]	γ KeV[c]	Γ SGRC[d]	IRERI[e] (nCi nuclide/nCi U-238)	IREI[f] (nCi nuclide/nCi U-238)	Release limit[g] (nCi/gm)
Chromium-51	28 d	33	0.16	51.6	4710	20.0
Cerium-141	32.5 d	77	0.35	23.6	1840	0.09
Niobium-95	35 d	764	4.2	1.96	142	1.00
Iron-59	44.5 d	1188	6.4	1.29	73.3	0.60
Antimony-124	60 d	1852	9.8	0.842	35.4	0.20
Zirconium-95	64 d	733	4.1	2.01	79.6	0.60
Strontium-85	65 d	518	3.0	2.75	107	1.00
Cobalt-58	71 d	977	5.5	1.50	53.6	1.00
Iridium-192	74 d	813	4.8	1.72	58.9	0.40
Scandium-46	84 d	2009	10.9	0.757	22.9	0.40
Tantalum-182	115 d	1301	6.8	1.21	26.7	0.40
Tin-113	115 d	280	1.7	4.85	107	0.90
Zinc-65	244 d	585	2.7	3.06	31.7	1.00
Manganese-54	312 d	836	4.7	1.76	14.2	1.00
Cesium-134	2.06 a	1555	8.7	0.948	3.30	0.09
Sodium-22	2.60 a	2187	12.0	0.688	1.97	0.40[k]
Antimony-125	2.73 a	443	2.7	3.06	8.42	1.00
Cobalt-60	5.27 a	2504	13.2	0.625	1.12	0.50
Barium-133	10.54 a	404	2.4	3.44	4.69	—[l]
Europium-152	13.33 a	1162	5.8	1.42	1.82	0.60
Potassium-40	1.28 Ga	157	0.78[i]	10.6	10.6	n/a
Uranium-238 plus daughters	4.47 Ga	1796[h]	~8.25[j]	1.00	1.00	n/a
Thorium-232 plus daughters	14.06 Ga	2470[h]	~5.1[j]	1.62	1.62	n/a

[a]*This list is not all-inclusive; it covers the major man-made nuclides encountered in irradiated gemstones with half-lives between a month to a dozen years. Shorter-lived nuclides die out quickly and longer-lived nuclides are too difficult to produce in sufficient quantity to be a problem. The nuclides are those identified in gemstones by the author, by J. Razvi and W. Whittemore of General Atomics (pers. comm., 1988), and by D. Alger of the University of Missouri (pers. comm., 1988). The data were taken, and calculations made, from Brown et al. (1986), Kocher (1981), Johns (1983), and Jaeger (1968). The three major natural radioactive nuclides (K-40, U-238, and Th-232) are also included.*

[b]*Half-life: The time it takes for the activity (nCi) of a nuclide type to decay to one-half its original value. d = days, a = years, Ga = billion years.*

[c]*γ KeV: The total energy (in KeV) of the gamma rays emitted per radioactive decay (Bq). X-rays are not included.*

[d]*SGRC: Specific Gamma-Ray Constant. This is the exposure rate in air (roentgens per hour) from the gamma rays emitted by a millicurie point source of radioactive nuclides at a distance of one centimeter. Author converted R to μr and mCi to nCi.*

$$\Gamma = \frac{\mu r - cm^2}{hr - nCi}$$

Example: Exposure rate in air from a 2 nCi point source of Ta-182 at a distance of 0.5 cm

$$\mu r/hr = 2\ nCi \times 6.8/(0.5\ cm)^2 = \underline{54.4}\ \mu r/hr \text{ or } \sim 0.05\ mr/hr$$

[e]*IRERI: Inverse Relative Exposure Rate Index. This is the number of nCi of a particular nuclide needed to give the same gamma-ray exposure rate in air as one nCi of U-238 plus daughters.*

[f]*IREI: Inverse Relative Exposure Index. This is the original number of nCi of a particular nuclide required to give the same gamma-ray exposure in air over a 10-year period as one nCi of U-238 plus daughters.*

[g]*Current release limits for reactor-irradiated faceted blue topaz as listed in Schedule A of 10 CFR 30. For nuclides that are not listed and that have a half-life of less than three years, the limit is 0.001 nCi/gm.*

[h]*Energy computed by assuming all daughters in equilibrium with parent nuclide.*

[i]*Computed by the author using Johns (1983).*

[j]*Includes radium-226 and radium-228 and their respective daughters only—not the parents U-238 and Th-232 (Jaeger et al., 1968).*

[k]*Limit changed in 1988 from 0.001 to 0.4 for faceted blue topaz only, by license request from General Atomics.*

[l]*Not listed in 10 CFR 30 and has a half-life greater than three years.*

tured that contain less than 1/20 of 1% of uranium and thorium source material. For instance, eyeglass lenses cannot be sold unless they contain less than these amounts (Moghissi, 1978). This translates into 0.168 nCi/gm of U-238 and 0.055 nCi/gm of Th-232. From the IREI in table 4, one can calculate that the exposure from a gemstone containing about 4.5 nCi/gm of Sc-46 or Ta-182 would produce about the same gamma-ray exposure or dose over a 10-year period as a gemstone contain-

ing source uranium at exempt (0.168 nCi/gm) concentrations. As another comparison, the average human contains about 200 nCi of K-40 and eats and drinks about 140 nCi of uranium per year (Eisenbud, 1987). Therefore, with respect to the small quantities of radioactivity found in gemstones and the resultant radiation doses people may receive, 2 nCi of any radioactive nuclide per gram of material can be judged to be harmless and safe, and a very small fraction of background radiation.

U.S. REGULATION OF IRRADIATED MATERIAL

All irradiation facilities in the United States are controlled by one or more of several regulatory agencies: the U.S. Nuclear Regulatory Commission, the U.S. Department of Energy, and the health and safety agencies of the various states. The Atomic Energy Act of 1946 established control over nuclear energy by the federal government. Further legislation in 1954 provided for the civilian licensing of nuclear power plants, allowed civilian access to nonmilitary uses of atomic energy, and gave the Atomic Energy Commission (AEC) the responsibility for protecting public health, safety, and property in matters concerning radiation.

A 1959 amendment to the Atomic Energy Act of 1954 transferred some of the federal licensing authority and responsibility to certain qualified states called agreement states. Currently, there are 29 agreement states, each with its own radiation control programs. New York and California were among the first states in this group, while Illinois was the most recent to join.

In 1974, the AEC was separated by legislation into two groups, with the regulatory side being renamed the Nuclear Regulatory Commission (NRC). The other side eventually became the Department of Energy.

If a gem is made radioactive during irradiation treatment in a nuclear reactor facility anywhere in the United States, the NRC has total jurisdiction over its handling and release to the general public. The NRC also has control over reactor-irradiated radioactive gem materials entering or leaving this country. If the material is made radioactive in a linac or some other device not regulated by the NRC, then the state has jurisdiction.

On March 16, 1965, the AEC issued its first and only policy statement concerning the use of either radioactive source materials (uranium and thorium) or by-product materials (those rendered radioactive by exposure to radiation in a reactor) in products intended for use by the general public without the imposition of regulatory controls. This policy statement set forth criteria for exempting, on a case-by-case basis, the possession and use of approved items from licensing requirements. Approved possession and use by the general public would depend both on the associated radiation doses people could receive and on the apparent usefulness of the products. In particular, the AEC at that time considered that "the use of radioactive material in toys, novelties, and adornments [gemstones] was of marginal benefit" ("Products intended . . .," 1965).

On June 25, 1986, the NRC issued a letter to all non-power reactors (NRC, 1986) that states in part that "the distribution of irradiated materials, even with low levels of induced radioactivity, to unlicensed persons is prohibited unless the distributor of such materials has a specific license . . . , which permits such distribution. The staff [NRC] considers gems to be adornments and has not granted licenses for the distribution of irradiated gems. If you directly distribute the irradiated products to unlicensed persons, you must obtain a license to reflect this activity." Thus, the distribution of reactor-treated gemstones to the general public was prohibited. For the next two years, however, enforcement was uneven and no distribution licenses were issued.

On February 23, 1988, after much debate (Stello, 1987a and b), the NRC announced that "applications will now be considered for interim licenses authorizing the distribution of neutron-irradiated gems, particularly topaz, to unlicensed persons" (Miraglia and Cunningham, 1988). In March 1988, the NRC stated that this applies only to cut, finished gems and that the "NRC staff plans to control distribution of irradiated gemstones at the source, and thus envisions two principal groups of applicants for distribution licenses: domestic reactor facilities and initial importers" (Michael Lamastra, letter to author, March 3, 1988).

These interim guidelines appear to place an undue burden on domestic irradiation operations, since few reactor facilities are willing to assume the many responsibilities that certification entails. A licensed importer can certify stones irradiated outside the U.S. for distribution in this country, but cannot certify domestically irradiated material. The importer still must contract with a U.S.–

licensed reactor facility to determine radiation concentrations for release to the general public. The NRC goal here is to "limit the impact of licensing requirements on the jewelry industry. If domestic reactors and importers obtain licenses, then no one else in the jewelry distribution chain need obtain a license" (M. Lamastra, letter to the author, October 31, 1988). The net result, however, is that more constraints are placed on gems irradiated domestically than those irradiated outside the U.S.

An additional problem or inequity is that the exempt concentrations for the various nuclides in 10 CFR 30 (again, see table 4) are based on *"ingestion"* or *"inhalation"* (Stello, 1987b), and have not been adjusted for solid, nonsoluble materials (such as gemstones) that remain outside the body. The author has tried to find legal definitions of "radioactive" material and the only state or federal regulation found so far was 49 CFR 173.403 (X), issued by the Department of Transportation: "'Radioactive material' means any material having a specific activity greater than 0.002 microcuries per gram" (2 nCi/gm). At this time, the U.S. federal release limits for the great majority of the nuclides are 1 nCi/gm or less (again, see table 4), but the NRC prefers 0.4 nCi/gm (J. Razvi and W. Whittemore, pers. comm., 1988).

Most other countries—e.g., West Germany, Italy, Japan, Taiwan, and Hong Kong—regard 2 nCi/gm as releasable (Scott Kohn, pers. comm., 1987). Canada temporarily set the releasable value at 1 nCi/gm while waiting for the U.S. NRC to make a final determination; Great Britain uses 2.7 nCi/gm (see Ilari, 1985, for further information on international recommendations in this area).

It is the author's opinion that two nanocuries of induced radioactivity per gram of irradiated gemstones would be a fair and safe limit for distribution to the general public, considering natural background radiation doses we receive during our lives, the amount of radioactivity contained in natural gemstones and other consumer products, and other data presented in this article.

CONCLUSION

Gemstone irradiation and radioactivity are very complex issues. This article has summarized the various components of these issues in an effort to clarify the nature of gemstone radioactivity and the potential health hazards involved. Current U.S. regulations appear to be unrealistic.

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