"EMERALDOLITE": A NEW SYNTHETIC EMERALD OVERGROWTH ON NATURAL BERYL

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This article describes a new manufactured gem material marketed under the trade name "Emeraldolite," which is an epitaxial growth of flux synthetic emerald on opaque white beryl. The material has a distinctive visual appearance, and is commonly used for jewelry in its "rough" form. The gemological properties of "Emeraldolite" are very similar to those normally shown by flux-grown synthetic emeralds.

"Emeraldolite" is a new manufactured gem material that consists of a substrate of common opaque white natural beryl with an epitaxial deposit of flux-grown synthetic emerald (figure 1). Although it is not presently being produced in commercial quantities, the capacity exists for a large-scale operation. It has been used in jewelry in its "natural" form, but is also fashioned *en cabochon*. Unlike most synthetic emeralds, it is usually not faceted because of its opacity.

This article provides a general description of the procedure used to produce this new manmade gem material, and reports on its gemological properties and chemistry.

METHOD OF SYNTHESIS

"Emeraldolite" is manufactured by one of the authors (D.R.) in France. A flux process (a chemical reaction in anhydrous solvents) is used to produce the synthetic emerald overgrowth on a natural beryl seed; fluoroberyllate compounds play an important role (see Robert, 1987).

The resulting product is reminiscent of the Lechleitner overgrowth of hydrothermal emerald on prefaceted colorless beryl (see, e.g., Schmetzer et al., 1981). Both are based on the same principle: the synthesis of an epitaxial overgrowth on less expensive beryl, rather than the (more costly and more time-consuming) synthesis of an entire stone. The two products vary greatly in appearance, however, with the Lechleitner material usually translucent to transparent, while virtually all of the "Emeraldolite" produced to date has been opaque.

Figure 1. This 76.42-ct "Emeraldolite" specimen consists of a flux-grown synthetic emerald overgrowth on natural white beryl. Photo by Robert Weldon.

The flux process that produces the synthetic emerald overgrowth for "Emeraldolite" is also significantly different from that used to grow synthetic emeralds such as the Lennix product (see Graziani et al., 1987). In the Lennix process, Al_2O_3 and SiO_2 are dissolved from the crucible itself, and the flux is a typical mixture of lithium molybdate and a boron salt. By contrast, the synthetic emerald layer in "Emeraldolite" is grown using a chemical – not a physical – reaction involving fluorine as a transport agent. This allows for faster growth than the Lennix process. Further information on the procedure use to grow "Emeraldolite" must remain proprietary.

CURRENT PRODUCTION

Because "Emeraldolite" only requires the growth of a thin (0.3–1 mm) layer of synthetic emerald, it is relatively inexpensive to manufacture (see "Synthetic emeralds . . . ," 1988). With only a small furnace, 15,000–20,000 ct of material can be produced annually. The process could be adapted to accommodate larger production in a variety of shapes and styles. Presently, production is variable, with ongoing experimentation into the most attractive product for jewelry use. It has been found, for example, that the overgrowth is unpleasantly asymmetrical on all isometric shapes, such as spheres, because of preferred growth along the optic axis.

DESCRIPTION OF THE MATERIAL

A view from the base of the "Emeraldolite" sample in figure 1 shows a thin layer of transparent dark green, flux-grown synthetic emerald covering a milky-white natural beryl crystal (figure 2). The substrate, which plays the role of seed crystal, can be of any size or shape. The linear dimension of the beryl core used is not a critical parameter (pieces over 10 cm have been successfully processed); rather, the total surface area is key because it determines the amount of feed material necessary to obtain a smooth, homogeneous coverage.

The dark color of the overgrowth is very even, and is reproducible from run to run. The synthetic emerald layer has a step-like appearance caused by the juxtaposition of numerous tiny (1–5 mm in longest dimension) crystal faces that are all oriented in the same crystallographic direction. This crystallographic orientation also contributes to the homogeneity of color.

ABOUT THE AUTHORS

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Figure 2. A different view of the specimen in figure 1 shows the thin flux synthetic emerald overgrowth. The color contrast between the white seed and the overgrowth is obvious. The numerous parallel crystal faces demonstrate the epitaxial character of the overgrowth. Photo by Robert Weldon.

Before deposition of the overgrowth, the substrate can be preformed in a particular shape (cabochon, for example) to fit in standard mountings. The "Emeraldolite" can then be polished (with greater ease than if the overgrowth were random, because there is only one crystallographic orientation) or incorporated as grown into pieces of jewelry (figure 3). Even polished, though, the surface is irregular, so visual appearance will easily separate "Emeraldolite" from natural emerald and even other synthetic emeralds.

GEMOLOGICAL PROPERTIES

To characterize "Emeraldolite" gemologically, we studied a total of 10 samples: one large (37.94 × 24.31 × 18.13 mm; 76.42 ct), very dark, irregular crystalline mass (figure 1); five smaller (20 mm in maximum dimension) rough pieces; one 20-mm piece polished as a flat tablet; and three "rough" pieces that had been mounted in jewelry (again, see figure 3). The results of our examination are summarized below. In general, the gemological properties of the "Emeraldolite" overgrowth are consistent with those of other flux-grown synthetic emeralds (Koivula and Keller, 1985).

Figure 3. These three pieces of "Emeraldolite" jewelry represent a typical style in which this new gem material is employed. The central piece measures 5.5 × 3.5 cm. Photo by Robert Weldon.

Refractive Index. Because the essentially rough surface is covered with tiny crystal faces, we used the spot method (on a Duplex II refractometer with a white light source) on four different areas of the large crystal and one spot on each of the other pieces. All yielded an R.I. of 1.56, which is typical of flux synthetic emerald (Liddicoat, 1989; p. 101). Because of the limitations imposed by the irregular surface condition of the "Emeraldolite," no birefringence could be determined.

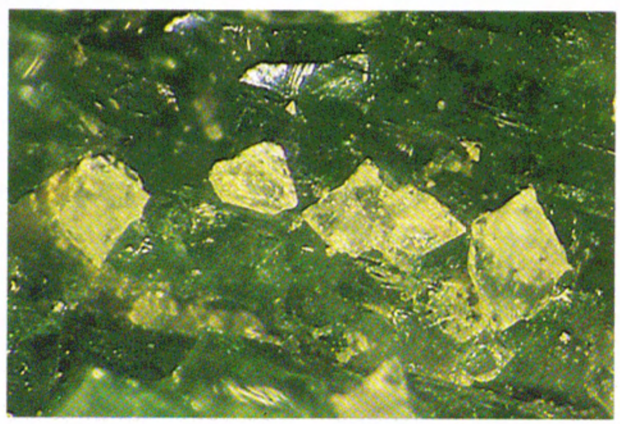
Specific Gravity. We could not determine an accurate specific gravity on any of the study specimens because the irregular surfaces trapped excessive amounts of air that could not be completely dislodged. Moreover, the specific gravity of an entire piece of "Emeraldolite" would vary depending on the specific gravity of the core. We did, however, test a 0.05-ct fragment of the synthetic overgrowth, using a mixture of methylene iodide and benzyl benzoate calibrated at 2.67. The sample floated very slowly to the surface of the liquid, on the basis of which we estimated the S.G. to be 2.66, which is typical of flux-grown synthetic emerald (Liddicoat, 1989; p. 101).

Ultraviolet Fluorescence. When examined in darkroom conditions with both long- and short-wave ultraviolet radiation, the synthetic emerald overgrowth and the white core material were inert, with no visible phosphorescence. This contrasts with the weak red fluorescence typical of most flux-grown synthetic emeralds. Although the iron content of the material is relatively low (0.28–0.34 wt.% FeO; see "Chemical Analysis" below), other synthetic emeralds containing similar amounts of iron do fluoresce red (Koivula and Keller, 1985; Graziani et al., 1987). Quenching by vanadium is possible, but unlikely. Instead, the absence of fluorescence may be due to "self-quenching," which occurs when nearby ions of the same type (here, Cr^{3+} ions) interact because of their high concentration, and the energy absorbed is given off in a form other than visible luminescence (Waychunas, 1988).

The surfaces of several specimens are speckled with numerous tiny, white, translucent, octahedral crystals (figure 4) and crusts of what appears to be "lithium feldspar"—the type of crystals obtained by Hautefeuille and Perrey when they first synthesized emerald by a flux method in 1888 (Nassau, 1976). These spots fluoresce a weak, chalky, brownish yellow to long-wave U.V. radiation, and a strong, chalky, slightly brownish yellow to short-wave U.V.

Visible-Light Spectroscopy. Using a Pye Unicam 8800 spectrophotometer, we took the visible-light absorption spectrum of a small fragment of the synthetic emerald overgrowth in a random orientation. The spectrum produced is typical of synthetic emerald: two broad bands with apparent maxima at about 430 and 610 nm, and a number of

Figure 4. Translucent, white, octahedral "lithium feldspar" crystals form as a by-product on and in the "Emeraldolite" overgrowth. Photomicrograph by John I. Koivula; magnified 30×.



sharp bands at 470, 476, 636, 660, 680, and 683 nm. "Cr lines" were easily seen with a Beck hand spectroscope.

Pleochroism. Although "Emeraldolite" is essentially opaque and very dark, we did see moderate pleochroism (very slightly yellowish green and bluish green) when all specimens were viewed through a dichroscope with reflected light.

Chelsea Color Filter Reaction. When bathed in white light from a fiber-optic illuminator and examined with a Chelsea color filter, the surfaces of all the "Emeraldolite" specimens appeared intense brownish red.

Hardness. Using a set of Mohs hardness points, we found that "Emeraldolite" was easily scratched with the corundum point (hardness 9), but the topaz point (hardness 8) did not produce a scratch even when a reasonable amount of pressure was applied. We therefore estimated the Mohs hardness to be approximately 8, which is within the expected range for synthetic emerald.

Toughness. The epitaxial growth process seems to begin with a short etching period that guarantees complete continuity between the crystalline lattice of the substrate and that of the overgrowth. This results in a very good toughness and mechanical solidity that would not be achieved if the emerald overgrowth had a random orientation. The overgrowth cannot be broken off the substrate, so the toughness of "Emeraldolite" is basically that of the substrate. If this latter contains big fractures, it will shatter when put in the flux; if it contains small fractures, the epitaxy will seal them, reinforcing the piece.

Microscopic Features. Using oblique illumination, we carefully examined the transparent overgrowth of the "Emeraldolite" samples with a gemological microscope. The features observed can be used to identify this material as a synthetic.

Specifically, magnification revealed several families of tiny parallel crystal faces that cover the flux synthetic overgrowth layer like a mosaic. In some few areas, the overgrowth had been chipped away, exposing the white beryl substrate (figures 2 and 5).

The morphology of the emerald crystals in the overgrowth is typical of emerald, with essentially

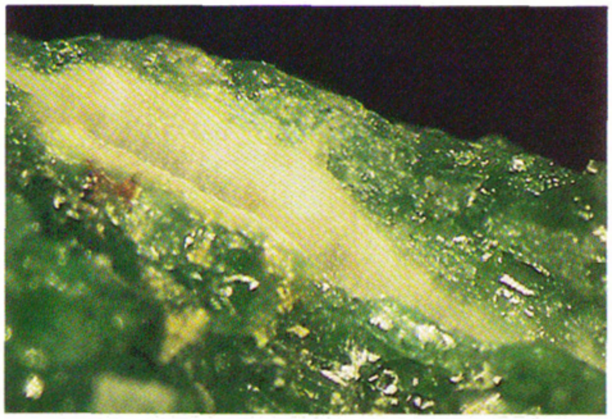


Figure 5. The 0.5-mm-thick flux synthetic emerald overgrowth layer contrasts with the underlying white beryl substrate. Photomicrograph by John I. Koivula; magnified 6×.

prism faces and basal planes. The occurrence of bipyramidal faces is strongly related to the somewhat complicated end of the thermal synthesis cycle. These faces are less common in the "evaporation facies" obtained on the surface of the melt and on the walls of the crucible (Robert, 1987). A detailed description of the crystal shapes and associations is given in Robert (1988a and b). Note that the stress cracks typical of the hydrothermally grown Lechleitner synthetic emerald overgrowth were not seen in the flux overgrowth of "Emeraldolite."

The small, colorless octahedral crystals of "lithium feldspar" that were seen to fluoresce on the surface of the samples were also seen inside the synthetic emerald overgrowth. This "lithium feldspar" crystallizes in the tetragonal system, and its crystal structure is currently under study.

In the synthetic emerald layer, we also observed comparatively large primary flux inclusions of an off-white to brown color (figure 6), as well as high-relief spherical to subspherical voids that greatly resemble the ordinary gas bubbles common to most glasses (figure 7). The latter may be partially open at the surface, forming crater-like pits. Although the flux inclusions were typical of those observed in flux-grown synthetic emeralds (Gübelin and Koivula, 1986), the bubble-like features have not been seen before in synthetic emeralds and are more characteristic of inclusions found in the Knischka synthetic ruby (Gübelin, 1982), also a flux product.

Figure 6. These primary flux inclusions were commonly observed in the "Emeraldolite" overgrowth. Photomicrograph by John I. Koivula; magnified 20×.

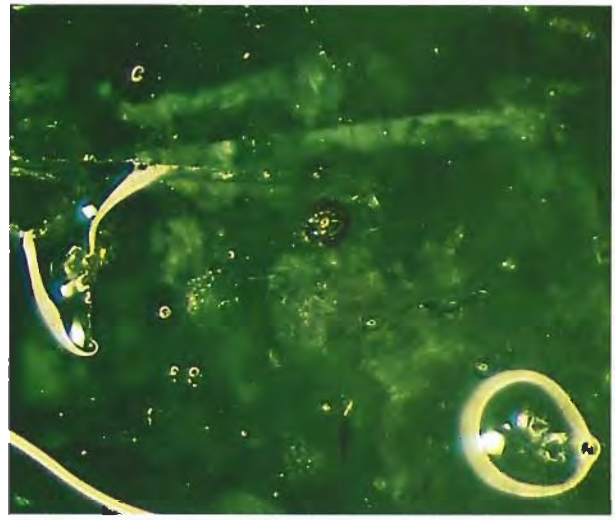


Figure 7. High-relief spherical to subspherical voids are commonly seen in the "Emeraldolite" overgrowth layer. Photomicrograph by John I. Koivula; magnified 50×.

CHEMICAL ANALYSIS

Using an energy-dispersive electron microprobe, Dr. Henry Hänni and George Bosshart, both at the time with the Swiss Foundation for Research on Gemstones (SSEF), analyzed the chemistry of the "Emeraldolite" epitaxial overgrowth. The range of compositions obtained is shown in table 1.

Note in particular that the concentration levels of Cr^{3+} are significantly higher than those found in most natural emeralds (approximately 1 wt.%; Hänni, 1982), but are lower than those found in the Lechleitner synthetic emerald overgrowth (7.64 to 13.20 wt.%; Schmetzer et al., 1981). These high concentrations of Cr^{3+} are responsible for the dark green color. The low concentrations of magnesium also contrast with the high values (more than 0.8 wt.% MgO) found in natural emeralds. Concentrations of Na_2O in "Emeraldolite" compare favorably with those of synthetic emeralds (less than 0.2 wt.%) and contrast with the 0.4 wt.% usually found in natural emeralds (Hänni, 1982).

The higher concentrations of Mg and Na in nature are due to Mg^{2+} substituting for Al^{3+} in the beryl crystal structure, the charge deficit being compensated by the presence of a Na^+ ion nearby. This mechanism does not occur during the growth of the "Emeraldolite" layer or any flux-grown synthetic emerald because Mg and Na are not voluntarily introduced in the growth environment.

TABLE 1. Chemical composition of the "Emeraldolite" synthetic emerald overgrowth.[a]

Oxide	Weight %
SiO_2	66.88 – 67.28
Al_2O_3	16.58 – 18.21
TiO_2	nd – 0.06
FeO	0.28 – 0.34
MgO	0.17 – 0.25
MnO	nd – 0.13
Cr_2O_3	1.76 – 4.15
V_2O_5	nd – 0.12
Na_2O	bdl

[a]*Chemical analyses performed by Henry Hänni and George Bosshart, SSEF, on an electron microprobe equipped with an energy-dispersive spectrometer. Details of the operating conditions can be obtained from the analysts. nd = not determined; bdl = below detection limits.*

CONCLUSION

Manufactured in France, "Emeraldolite" is an overgrowth of deep green flux synthetic emerald on opaque white natural beryl. It is based on a principle similar to that of the Lechleitner synthetic emerald overgrowth on transparent to translucent near-colorless natural beryl. Its unique appearance makes it easy to separate from any other natural gem or synthetic material. In addition, one can find in the synthetic emerald overgrowth characteristics typical of a flux-grown synthetic emerald.

"Emeraldolite" has been used by jewelry manufacturers in its original state, without polishing, because its numerous crystal faces reflect light well. The material can, however, be polished, and lends itself to intarsia and sculpture, as well as to cabochons.

REFERENCES

Graziani G., Gübelin E., Martini M. (1987) The Lennix synthetic emerald. *Gems & Gemology*, Vol. 23, No. 3, pp. 140–147.

Gübelin E.J. (1982) New synthetic rubies made by Professor P. O. Knischka. *Gems & Gemology*, Vol. 18, No. 3, pp. 165–168.

Gübelin E.J., Koivula J.I. (1986) *Photoatlas of Inclusions in Gemstones*, ABC Edition, Zurich, Switzerland.

Hänni H.A. (1982) A contribution to the separability of natural and synthetic emeralds. *Journal of Gemmology*, Vol. 18, No. 2, pp. 138–144.

Koivula J.I., Keller P.C. (1985) Russian flux-grown synthetic emeralds. *Gems & Gemology*, Vol. 21, No. 2, pp. 79–85.

Liddicoat R.T. (1989) *Handbook of Gem Identification*, 12th ed., 2nd rev. printing. Gemological Institute of America, Santa Monica, CA.

Nassau K. (1976) Synthetic emerald: The confusing history and the current technologies. *Journal of Crystal Growth*, Vol. 35, pp. 211–222.

Robert D. (1987) Genèse du beryl, synthèse de l'émeraude. *Monde & Minéraux*, No. 80, pp 39–42.

Robert D. (1988a) L'Emeraldolite, une nouvelle matière. *Revue de Gemmologie a.f.g.*, No. 94, pp. 9–10.

Robert D. (1988b) Qu'est-ce que l'Emeraldonite *[sic]*. *Revue de Gemmologie a.f.g.*, No. 95, pp. 19–20.

Schmetzer K., Bank H., Stähle V. (1981) The chromium content of Lechleitner synthetic emerald overgrowth. *Gems & Gemology*, Vol. 17, No. 2, pp. 98–100.

Synthetic emeralds: New growth in an ancient gem (1988). *Solitaire*, November, pp. 27–30.

Waychunas G.A. (1988) Luminescence, X-ray emission and new spectroscopies. In F. C. Hawthorne, Ed., *Spectroscopic Methods in Mineralogy and Geology, Reviews in Mineralogy*, Vol. 18, Mineralogical Society of America, Washington, DC, pp. 635–698.