SERENDIBITE FROM SRI LANKA

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Two samples of faceted serendibite, a boron-bearing mineral that is only rarely found in gem quality, are described. These samples reportedly were cut from rough discovered in secondary deposits of the Ratnapura area in Sri Lanka. The bluish green to green-blue gems are similar in color, physical and optical properties, and chemical composition to low-iron-bearing non-gem-quality serendibite from various occurrences. These gems may be confused with sapphirine and zoisite, but can be identified as serendibite on the basis of refractive indices, twinning, and spectroscopic features.

Sri Lanka, which for centuries has supplied the gem market with a large number and wide variety of gemstones, has in recent years been overshadowed by the discovery of comparably propitious gem deposits in East Africa and Madagascar. Nevertheless, during the second half of the 20th century, Sri Lanka yielded rare gem varieties of minerals such as dumortierite, ekanite, sinhalite, and taaffeite. In the 1990s, yet another new gem variety was recovered in Sri Lanka, of the mineral serendibite. Although the mineral serendibite was first described in 1903, also from Sri Lanka, not until 1997 was the first report of a gem-quality sample published (Reinitz and Johnson, 1997).

Serendibite derives from the old Arab name for Sri Lanka—Serendib—in recognition of this country as the type locality for this mineral. The first description of the mineral serendibite was published in 1903 by Prior and Coomaraswamy for the boron-bearing Ca-Mg-Al-silicate that was discovered in a contact zone between granulite and limestone at Gangapitiya, near Ambakotte, about 20 km (12 miles) east of Kandy. This first description, however, was the only publication about this mineral from a primary (as opposed to secondary) deposit in Sri Lanka. The discovery of gem-quality serendibite

from secondary deposits in the Ratnapura area in the second half of the 1990s increases the number of gems containing essential boron from this island (tourmaline, dumortierite, boron-rich korerupine [or prismaticine, according to recent mineralogical studies], and sinhalite).

To date, serendibite has been reported from 11 localities: Sri Lanka (type locality near Kandy), United States (three occurrences), Russia (two occurrences), Ukraine, Tanzania, Canada, and Madagascar (two localities). At all of these localities, serendibite occurs in metasomatic high-temperature calc-silicate rocks (skarns), mostly of granulite facies (Grew et al., 1990, 1991a,b; Grew, 1996). The occurrence of gem-quality serendibite crystals in secondary deposits near Ratnapura, Sri Lanka, is consistent with the derivation of these gem gravels from granulite-facies metamorphic rocks. According to modern mineralogical examination (see, e.g., Kunzman, 1999), serendibite is a triclinic Ca-Mg-Al-B-silicate which can also contain distinct amounts of Fe^{2+} and Fe^{3+} .

Since 1997, gemologist and gem dealer D. Palitha Gunasekera of Ratnapura has reported encountering three gem-quality samples of serendibite, weighing 0.35, 0.55, and 0.56 ct as faceted stones (pers. comm., 1997–2001). A merchant in Kolonne showed him the first (smallest) sample as a 1.25 ct pebble. This rough sample was said to originate from Ginigalgoda, near Kolonne in the Ratnapura district. Mr. Gunasekera submitted the 0.35 ct stone faceted from this piece of rough (figure 1) to the GIA Gem Trade Laboratory for identification in autumn of 1996 (Reinitz and Johnson, 1997). It was subsequently purchased by one of the present authors (EG) and identified independently as serendibite by KS in 1997, also using X-ray powder diffraction. A review

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Figure 1. These two stones, both from Sri Lanka, are the first faceted serendibites recorded in the gemological literature. The sample on the left weighs 0.35 ct and the one on the right weighs 0.55 ct.

of the literature did not turn up any other references to gem-quality serendibite.

Subsequent to the identification of the 0.35 ct sample, Mr. Gunasekera found what he believed to be a second—0.55 ct—sample in a mixed parcel of unidentified faceted stones in his possession. This stone (again, see figure 1) was purchased by the same author (EG) and identified as serendibite on the basis of the properties gathered for the 0.35 ct sample. Mr. Gunasekera reported that a third serendibite recently was found at Mudunkotuwa in the Ratnapura district. The 1.24 ct pebble was sold to another gem merchant, who cut a 0.56 ct. faceted stone from it (D. P. Gunasekera, pers. comm., 2001). We were not able to obtain this stone for identification or examination as part of this study.

In view of this gemstone's rarity, the authors took the opportunity to perform a detailed gemological, chemical, and spectroscopic study of the first two available faceted samples.

MATERIALS AND METHODS

In addition to the 0.35 ct and 0.55 ct faceted gemstones from Sri Lanka (see again figure 1), we obtained four polycrystalline, non-gem-quality serendibite samples in rock matrix from Johnsbury, Warren County, New York, for comparison (we were not able to obtain any nongem samples from Sri Lanka). This second reported serendibite locality was described in 1932 by Larsen and Schaller and studied later by various authors (e.g., Schaller and Hildebrand, 1955; Grew et al., 1991a, 1992). Subsequent to visual inspection, we obtained the X-ray powder diffraction patterns of two small powdered

Johnsbury samples using a Gandolfi camera. These X-ray data were compared with the diffraction pattern of the 0.35 ct sample obtained by the same technique.

The two faceted serendibites were tested by standard gemological methods for refractive indices, fluorescence to long- and short-wave ultraviolet radiation, and specific gravity. The optic character was determined by observation of the optic axis figure with and without the addition of a gypsum plate. We used standard microscopic techniques to examine the internal features under different lighting conditions, both with and without immersion liquids. Both stones also were examined with a desk-model spectroscope.

Qualitative chemical data were obtained by energy-dispersive X-ray fluorescence (EDXRF) spectroscopy using a Tracor Northern Spectrace 5000 system. To determine the quantitative chemical composition, we used a Cameca Camebax SX 50 electron microprobe with traverses of 9 point analyses each, measured across the tables of the faceted stones.

Polarized ultraviolet-visible-near infrared (UV-Vis-NIR, 300–2500 nm) absorption spectra were recorded using a Perkin Elmer Lambda 19 dual-beam spectrophotometer. Because of the small size and twinning of the faceted serendibites, as well as the unknown orientation of their optic axes, we were unable to obtain exactly oriented, polarized spectra in all three axial directions. Thus, two polarized spectra for each sample were taken with the incident beam traversing the stones through a crown facet and opposite pavilion facet, selected so that rotating the polarizer yielded maximum pleochroism. A third spectrum was obtained perpendicular to the direction of maximum pleochroism. In addition, colorimetric data were collected on the basis of non-polarized spectra taken in the 380–760 nm range with a Zeiss MCS 311 multi-channel color spectrometer.

Infrared spectroscopy in the 7000–2000 cm^{-1} range was performed in transmission mode using a Pye Unicam 9624 Fourier Transform infrared (FTIR) spectrometer and a DRIFTS (beam collector) unit, with different orientations of the faceted samples. Furthermore, minute amounts of powder were scraped off the girdle of both gem-quality serendibite specimens. These microsamples were used to prepare 3-mm-diameter KBr pellets (approximately 0.3 wt.% serendibite powder) for infrared spectroscopy in the mid-infrared range (2000–400 cm^{-1}).

Raman and photoluminescence spectra were

obtained with a Renishaw 2000 Raman microspectrometer equipped with a He/Cd laser (excitation in the ultraviolet region at 324.98 nm) and an Ar-ion laser (excitation in the green region at 514.5 nm). Since we were not able to obtain oriented Raman and photoluminescence spectra, we did take measurements on several facets to examine the orientation dependency of the Raman and photoluminescence signals. To obtain any Raman bands, we had to use high integration times per scan sequence.

RESULTS

The X-ray diffraction pattern of the 0.35 ct faceted Sri Lankan stone was identical to those of the Johnsbury nongem serendibites. The gemological properties of the two cut stones are listed in table 1 and discussed below.

Gemological Properties. The 0.35 ct serendibite was bluish green and the 0.55 ct specimen was green-blue (figure 1). Both samples were transparent and only slightly included. The dominant wavelengths derived with the Zeiss color spectrometer were 494.8 nm and 489.6 nm, respectively.

Although we were unable to determine the exact orientation of the optic axes relative to the table facets, it was evident from the color and the strong pleochroism of both specimens that their cuts were

TABLE 1. Gemological properties of two gem-quality serendibites from Sri Lanka.

Property	0.35 ct	0.55 ct
Size	4.42 × 3.80 × 2.80 mm	4.98 × 4.95 × 2.72 mm
Color	Bluish green	Green-blue
Pleochroism	Strong: Light yellowish green Bluish green Violetish blue	Strong: Light yellowish green Bluish green Violetish blue
Clarity	Transparent	Transparent
Refractive indices		
n_x	1.696	1.697
n_y	1.699	1.700
n_z	1.701	1.702
Birefringence	0.005	0.005
Optic character	Biaxial negative	Biaxial negative
Specific gravity	3.43	3.44
UV fluorescence		
Long-wave	Inert	Inert
Short-wave	Inert	Inert
Microscopic features	Isolated twin lamellae, partially healed fissure, fracture, tiny mineral inclusions	Polysynthetic twinning

Figure 2. Both faceted serendibites showed evidence of twinning. At 20× magnification, the 0.55 ct sample reveals two polysynthetically twinned areas within untwinned regions. Immersion, crossed polarizers.

oriented differently. In the 0.35 ct sample, the strongest pleochroism—light yellowish green to violetish blue—was evident in the direction parallel to the table facet. In the 0.55 ct sample, the strongest pleochroism—also light yellowish green to violetish blue—was evident in a direction perpendicular to the table facet. The third pleochroic color, observed perpendicular to the directions of the strongest pleochroism, was bluish green for both samples. Refractive indices and specific gravities are summarized in table 1. The samples were inert to both long- and short-wave UV radiation.

Features Observed with the Microscope. Both samples showed various forms of twinning. The smaller stone contained several isolated small parallel twin lamellae; the larger had two areas with polysynthetic twinning as well as areas free of twinning (figure 2). In the 0.35 ct sample, both a partially healed fissure and a nonhealed fracture were encountered. Small colorless mineral inclusions also were detected in this sample. These inclusions, however, were too small for identification with Raman analysis.

Chemical Composition. EDXRF analysis revealed the main components of serendibite (Mg, Ca, Al, and Si), minor amounts of Na and Fe, and traces of P, K, Ti, V, Cr, Mn, and Ga. The electron microprobe results are given in table 2; no obvious chemical zoning was observed in either sample. The chemical composition of these gem-quality serendibites is similar to the composition of sodium-bearing, low-iron serendibite from Johnsbury, New York (see table 2).

Spectroscopic Properties. *UV-Vis-NIR Spectroscopy.* Both samples revealed a broad, distinctly polarized absorption band in the 600–800 nm range (figure 3). This band was centered at about 720 or 700 nm for the directions of the violetish blue and bluish green pleochroic colors. A weaker band in the 800–900 nm range with a maximum at about 850 nm (not shown on figure 3) was measured for the direction of the yellowish-green coloration. The

TABLE 2. Chemical composition of gem-quality serendibite from Sri Lanka, compared to non-gem-quality serendibite from Johnsbury, New York.

Composition	Sample		
	0.35 ct ^a	0.55 ct ^a	Johnsbury ^b
Oxides (wt.%)			
SiO ₂	27.18	26.56	25.95
TiO ₂	0.02	0.03	0.08
Al ₂ O ₃	34.82	34.24	35.31
B ₂ O ₃	7.63 ^c	7.55 ^c	7.64
Fe ₂ O ₃ ^d	0.84	1.07	0.93
Cr ₂ O ₃	0.01	0.02	nd
V ₂ O ₃	0.02	0.02	nd
MnO	0.02	0.03	0.03
MgO	15.43	15.57	15.78
CaO	13.68	13.79	12.95
Na ₂ O	1.20	1.23	1.25
K ₂ O	0.10	0.01	nd
P ₂ O ₅	0.03	0.02	nd
Total	100.98	100.14	99.98
Cations per 20 oxygens			
Si	3.096	3.058	2.989
B	1.500 ^e	1.500 ^e	1.520
Al	1.401	1.440	1.491
P	0.003	0.002	nd
Sum Z	6.000	6.000	6.000
Al	3.272	3.205	3.302
Fe	0.072	0.093	0.081
Cr	0.001	0.002	nd
V	0.002	0.002	nd
Ti	0.002	0.003	0.007
Mg	2.620	2.672	2.709
Mn	0.002	0.003	0.003
Sum Y	5.971	5.980	6.117 ^f
Ca	1.669	1.701	1.598
Na	0.265	0.275	0.279
K	0.015	0.002	nd
Sum X	1.949	1.978	1.877

^aElectron microprobe analyses, average of 9 analyses each.

^bElectron microprobe analyses (and ion microprobe analyses for B₂O₃) from Grew et al. (1991a). The sample also contained traces of Li, Be, and F; nd = not determined.

^cCalculated from B = 1.500.

^dTotal iron calculated to Fe₂O₃.

^eAccording to the theoretical formula, Ca₂(Mg₃Al₃)(Al_{1.5}Si₃B_{1.5})O₂₀.

^fAlso contains traces of Li and Be.

Figure 3. These polarized UV-Vis-NIR absorption spectra for the 0.35 ct serendibite show strongly polarized absorption bands in the 600–800 nm range that are responsible for the pleochroism. Weak absorption bands also are present between 400 and 600 nm. The absorption coefficient is approximate.

strong polarization of this band is almost exclusively responsible for the distinct pleochroism of the serendibites. In one specific orientation of the polarizer, the strong band in the 600–800 nm range split into two maxima at about 660 and 740 nm, possibly due to interference with twin plane reflections. A number of weak to very weak bands also were found in the 400–600 nm range in both samples, with maxima at approximately 410, 435, 460, and 500 nm. The 460 and 500 nm lines were visible with the desk-model spectroscope.

Infrared Spectroscopy. Both faceted samples showed several absorption bands in the 3700–3200 cm⁻¹ and 2800–2500 cm⁻¹ areas and an absorption edge at about 2200 cm⁻¹, as indicated on figure 4A. The strengths and positions of these absorption bands in our nonpolarized spectra varied only slightly for different IR beam traverses through the samples. The weak bands at 3695 and 3620 cm⁻¹ in the spectrum of the 0.35 ct sample were not observed in the 0.55 ct sample. Thus, they might be caused by the minute inclusions observed only in that sample.

The mid-infrared spectra for the powder microsamples of both specimens (figure 4B) revealed virtually identical absorption spectra. The 0.35 ct sample exhibited major bands at 975, 811, 608, 537, and 489 cm⁻¹. The 0.55 ct sample yielded a set of slightly shifted major bands at 965, 807, 608, 537, and 484 cm⁻¹. Other, minor bands would require reconfirmation by spectra of additional microsamples.

INFRARED SPECTRA

Figure 4. In part A, the infrared spectra of the two gem serendibite samples are shown in the 5000–2000 cm^{-1} range. The major bands are found in the 3700–3200 as well as in the 2800–2500 cm^{-1} range. The weak bands of the 0.35 ct sample at 3695 and 3620 cm^{-1} are probably due to minute mineral inclusions. The absorption coefficient is approximate. Part B shows the mid-infrared spectra of KBr pellets containing minute amounts of serendibite powder, with the KBr absorption subtracted. The main serendibite absorption bands are in the 1200–700 cm^{-1} and 700–400 cm^{-1} ranges.

Raman and Photoluminescence Spectroscopy. We noted no significant differences in the intensities of the weak Raman bands, and only small differences in the intensities of photoluminescence bands (which actually is not typical for anisotropic minerals), when we tested the samples in different orientations. A series of faint, broad Raman bands recorded at ambient temperature with the Ar-ion laser are illustrated in figure 5. As a result of the low intensity of these bands, it is not appropriate to discuss them in further detail.

Although the Raman signals were of low intensity, a series of dominant photoluminescence (PL) bands were present in the spectra taken with both lasers. With the He/Cd laser, we recorded a series of PL bands positioned at approximately 1500, 2770, and 3870 cm^{-1} Raman shift (calculated to about 340, 357, and 370 nm; figure 6A). The Ar-ion laser exhibited an even more intense series of PL bands positioned at 4970, 5005, and 5435 cm^{-1} Raman shift (calculated to 690, 693, and 715 nm accordingly; figure 6B).

DISCUSSION

Gem-quality serendibite from Sri Lanka is similar in composition to low-iron-bearing serendibite from Johnsbury, New York, which also contains small but distinct amounts of sodium replacing part of the calcium in the idealized formula (table 2). The color

of our Johnsbury serendibite samples resembles that of the gem material as well.

The physical and optical properties of these two gem serendibites from the Ratnapura area (table 1) are in the range of those of low-iron-bearing serendibites, such as from Johnsbury and Russell, New York (Larsen and Schaller, 1932; Grew et al., 1990, 1991a) or from Melville Peninsula, District of Franklin, Canada (Hutcheon et al., 1977). In detail, the refractive indices of the gemstones are somewhat lower

Figure 5. The Raman spectrum of the 0.55 ct serendibite reveals several weak bands in the 1000 to 300 cm^{-1} range.

Figure 6. (A) This 325 nm photoluminescence spectrum of the 0.55 ct serendibite reveals a series of PL bands at 340, 357, and 370 nm. (B) The 514.5 nm photoluminescence spectrum of this sample shows PL features at 690, 693, and 715 nm.

than the optical properties of low iron-bearing serendibites, such as from Johnsbury, New York (2.76 wt.% FeO, $n_x = 1.701$, $n_y = 1.703$, $n_z = 1.706$; Larsen and Schaller, 1932) or from Melville Peninsula, Canada (3.48 wt.% FeO, $n_x = 1.700$, $n_y = 1.703$, $n_z = 1.706$, Hutcheon et al., 1977). This undoubtedly is due to the somewhat lower iron contents of the Sri Lankan material. We believe these are the lowest refractive indices reported thus far for any serendibite. Small discrepancies in the properties reported by Reinitz and Johnson (1997) on the 0.35 ct sample are most probably due to differences in the instrumentation used.

Serendibite is triclinic and belongs to the aenigmatite-rhönite group, which has a general formula of $X_2Y_6Z_6O_{20}$. Kunzmann (1999) provides the structural formula of serendibite as $Ca_2(Mg_3Al_3)(Al_{1.5}Si_3B_{1.5})O_{20}$, with (Mg_3Al_3) in octahedral sites and $(Al_{1.5}Si_3B_{1.5})$ in tetrahedral sites. In general, some Fe^{2+} substitutes for part of the Mg and some Fe^{3+} replaces part of the Al. An isomorphic replacement between sodium and calcium, coupled with replacement of magnesium by aluminum and aluminum by silicon, is also observed (see also Machin and Süsse, 1974; Grew et al., 1991a,b; Van Derveer et al., 1993; Burt, 1994). Calculating the cations of our analyses according to the chemical formula of serendibite, that is, with 20 oxygen and 1.5 boron atoms per formula unit, shows that our data are consistent with this formula (table 2).

The absorption bands measured in the visible and near-infrared are probably caused by Fe^{2+} and/or Fe^{3+} replacing Mg or Al in octahedral sites of the serendibite structure. A detailed assignment of all

absorption bands, however, is not possible without further research, such as with Mössbauer spectroscopy. The strong photoluminescence in the red region observed with the Ar-ion laser is found in the range typical for spin-forbidden Cr^{3+} absorption and emission lines. Thus, the small chromium contents found by X-ray fluorescence and microprobe analysis might be responsible for these lines.

Two gem minerals that are found occasionally with similar color and almost identical properties—such as refractive indices, birefringence, and specific gravity—might be mistaken as serendibite, namely, sapphirine and zoisite. The distinction from sapphirine—at least for the low-iron-bearing serendibites known to date in gem quality—can be made by careful measurement of refractive indices. Although the birefringence may be identical for sapphirine and serendibite, the refractive indices for sapphirine are always slightly above 1.700 (Fryer, 1985; Kane, 1987). Note, though, that higher iron contents in serendibite may cause misleading R.I. values. In such a case, identification would require additional examination with microscopy or spectroscopy.

The optical properties and specific gravity of serendibite and zoisite, on the other hand, show a complete overlap. The color of the rare chromium- and even rarer vanadium- and chromium-bearing varieties of zoisite from Tanzania (Schmetzer and Bank, 1979; Barot and Boehm, 1992) may also be quite similar to that of serendibite. Again, a distinction may be made on the basis of the lamellar or polysynthetic twinning of serendibite, which was

also reported as a typical property of non-gem-quality serendibite samples from various localities. However, spectroscopic features as described for the UV-Vis-NIR and infrared ranges, as well as Raman and photoluminescence data, are considerably more reliable for the identification of suspected serendibite gems than the standard gemological properties cited above.

CONCLUSION

The rare gem material serendibite is characterized with regard to gemological, chemical, and spectroscopic properties. Gemological properties, such as refractive indices, are related to the very low iron contents of the samples, as shown by microprobe analysis. Serendibite might be confused with sapphirine or zoisite, but refractive indices, twinning, and spectra can be used to separate these gem materials

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