Fluid and mineral inclusions in corundum from gem gravels in Sri Lanka

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Abstract

Mineral and fluid inclusions were studied in seven gem corundums from gravels of three areas in Sri Lanka. All fluid inclusions are pure CO_2. Microthermometry results on primary fluid inclusions suggest formation of corundum under granulite facies metamorphism (>630 °C, 5.5 kbar). Secondary fluid inclusions indicate different retrograde events of post-metamorphic cooling and uplift for different source areas.

KEYWORDS: corundum, fluid inclusions, gem gravels, Sri Lanka.

Introduction

SRI LANKA is famous for its gem corundum, which occurs with other gemstones in alluvial, eluvial and residual gravels on granulite facies rocks of the Highland Series (Cooray, 1978; Dahanayake and Ranasinghe, 1985). In the Highland Series corundum occurs as blue prisms in feldspathic granulite at Tannahena near Kandy; as violet tablets in corundum–sillimanite rock near the Haputale escarpment (Coomáráswamy, 1903, 1904); as transparent crystals in a pegmatite near Matara, as pale blue crystals in a pegmatitic feldspar vein at Horton Plains (Coates, 1935); as blue crystals in a marble at a syenite contact near Ohiya (Wells, 1956); in a biotite–sillimanite gneiss at Polgahawela (Cooray and Kumarapeli, 1960); in a pegmatite near Kolonné, Ratnapura (Gunaratne, 1976); as an accessory mineral in a skarn at a marble-granite contact in Elahera area (Silva, 1976); and in garnet gneisses and granulites in Weddagala and Kutuganga areas (Dahanayake and Ranasinghe, 1981, 1985). These occurrences suggest various types of source rock for corundum in the gem gravels. However, gemmological and mineralogical data have provided little information on the provenance and source rocks of the gemstones. Mineral and fluid inclusions in the gemstones have been studied in order to gain information about primary formation conditions and possible source rocks.

Description of samples

Four corundum specimens were studied from the Kataragama area (1b from Amarawewa, 10 km S. of Kataragama; 2a, 2b, 2c from Kochipadana, 5 km E. of Kataragama), two from the Embilimpitya area (4a, 4cd), and one from the Ratnapura area (10c) (Fig. 1).

The Kataragama samples 1b, 2a, 2b and 2c are transparent, euhedral corundum crystals; 1b is 8 × 4 mm and colourless, showing parting and dark-stained healed cracks and the mineral and fluid inclusions are regularly disposed along trails, approximately planar surfaces of various orientations; 2a was 6 × 4 mm and colourless, showing dark-stained healed cracks and has mineral and fluid inclusions along two trails and a number of isolated inclusions; 2b is 4 × 3 mm, has a colourless inclusion-free core and a dark-blue/colourless zoned rim with various mineral inclusions, and fluid inclusions along two trails; 2c is 10 × 5 mm, greyish with an indistinct yellowish/colourless zoning, showing healed cracks, containing various solid inclusions, and fluid inclusions on widely separated trails.

The Embilimpitiya samples 4a and 4cd are transparent to translucent, anhedral to subhedral crystals, showing an inclusion-free core and a rim rich in various mineral inclusions; fluid inclusions occur isolated and along trails; 4a is 7 × 5 mm and grey; 4cd is 6 × 4 mm and light-blue.

The Ratnapura sample 10c is a translucent, anhedral, light-blue crystal, 14 × 7 mm, showing parting along (0001) and $(01\bar{1}2)$; mineral inclusions occur in zones, and fluid inclusions isolated and along trails.

The seven corundum crystals are of poor gem quality due to inclusions and cracks, but some could be cut as star stones. For the present study

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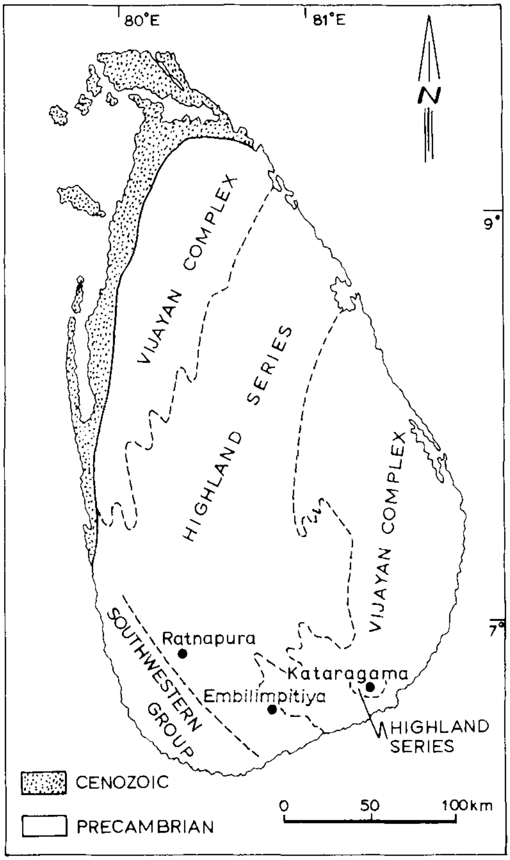


FIG. 1. Geological sketch map of Sri Lanka with sample locations.

0.3 mm thick sections, polished on both sides, were prepared of the corundum crystals.

Mineral inclusions

Very fine (<250 μm) inclusions of rutile, apatite, calcite and zircon were identified in corundum by optical methods and partial qualitative analysis with a Microdil-28 laser Raman microscopectrometer (Mao *et al.*, 1987).

The samples 2b, 4a, 4cd and 10c have a euhedral growth zoning of alternating rutile-free rutile-rich zones around a rutile-free core. The rutile-rich zones had a greyish-milky appearance and are characterized by a very fine to almost submicroscopic crystallographic network intergrowth of corundum and exsolved rutile needles. Isolated rutile inclusions are rare in sample 2b. Subhedral to euhedral apatite occurs in 2b, 2c and 4a. In 2b and 2c the apatite contains fluid inclusions (Fig. 2.1); 2b also encloses round carbonate (calcite with fluid inclusions (Fig. 2.2). Round zircon grains in the rutile-rich zones of corundum 4a and 4cd are surrounded by pleochroic halos with cracks. Opaque inclusions are of various compositions; 2c contains hexagonal tablets up to 0.5 mm across, probably of hematite (Gübelin, 1986); 4cd contains amorphous opaque material, and 1b and 2a finely dispersed opaque matter. Raman microspectrometry indicated the absence of graphite. Inclusions, which have the appearance of glass fragments with gas bubbles (Fig. 2.3) were observed in 4cd. The distribution of solid inclusions in the corundum samples is given in Table 1.

Bluish and greyish corundum of the three areas show rutile exsolution, whereas the colourless transparent cordundum does not show such exsolution. Rutile is common in Sri Lankan gem corundum (Zwaan, 1982; Gübelin, 1986; Gunawardene and Rupasinghe, 1986; Heilmann and Henn, 1986). The blue colour of sapphire is due to titanium/iron solid solution (Schmetzer, 1987); exsolution of rutile causes a milky greyish or whitish colour as in star stones (star effect due to exsolved rutile needles) and in Geuda sapphire [a Ti-rich milky-white corundum which might turn blue on heating (Gunaratne, 1981; Harder and Schneider, 1986)].

Round zircons in the Embilimpitiya samples suggest the presence of zircon-bearing feldspathic gneisses in the source area. Zircon is absent in the Kataragama and Ratnapura corundums studied in this investigation.

Fluid inclusion data

Microthermometric data on fluid inclusions were obtained between −100 and +35 °C, using a Chaixmeca stage (Poty *et al.*, 1976). Temperatures could be read with a precision of 0.2 °C between −25 and +25 °C, and of 0.5 °C outside the latter range. The measured temperatures are denoted as: *Te* = temperature of eutectic melting = temperature of first melting; *Tm* = temperature of final melting; *Th* = homogenization temperature. Fluid densities (or molar volumes) were estimated from *Th* and fluid composition, using the method described by Kerkhof (1987). Fluid inclusions and daughter minerals were analysed by laser Raman microspectrometry (Burke and Lustenhouwer, 1987).

The *fluid inclusions in corundum* range in size from <1 μm to several hundred micrometers. They constitute three groups: (1) isolated, fluid inclusions, often with euhedral negative crystal

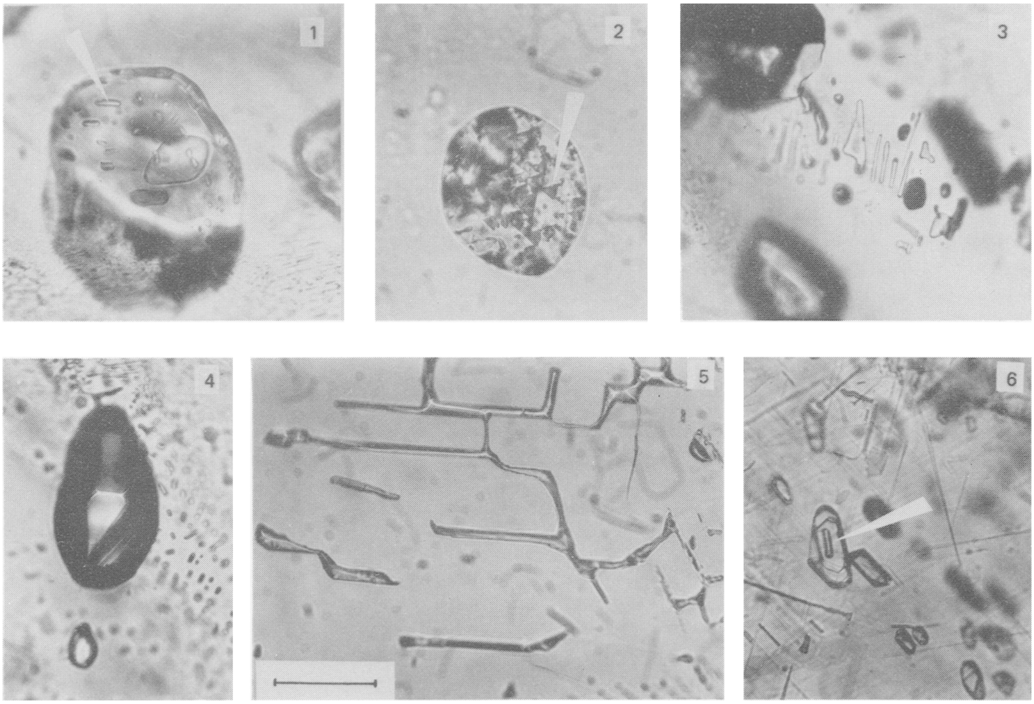


FIG. 2. Photomicrographs of solid and fluid inclusions in Sri Lanka corundum. 1: CO_2 inclusions in subhedral apatite in Kochipadana corundum (arrow). 2: CO_2 inclusions in round calcite in Kochipadana corundum (arrow). 3: Inclusions resembling glass fragments in Embilimpitiya corundum. 4: Euhedral negative crystal fluid inclusion in Embilimpitiya corundum. 5: Rectangular and channel shaped fluid inclusions in corundum from Amarawewa. 6: Daughter minerals in fluid inclusions (arrow) and rutile needles in a Ratnapura corundum. Length of bar (photo 5) 50 μm for all photographs.

Table 1. Distribution of solid inclusions in corundum from Kataragama, Embilimpitiya and Ratnapura area.

colour	area	no.	rutile	apatite	carbonate	zircon	opaque	glass?
colourless		1b					x	
colourless		2a					x	
zoned colour-less/dark blue	KATARAGAMA	2b	x	x	x			
grey-vaguely zoned		2c		x			x	
grey-zoned	EMBILIMPITIYA	4a	x	x		x		
light blue-zoned		4cd	x			x	x	x
light blue	RATNAPURA	10c	x					

forms of the host corundum (Fig. 2.4), found in 2a, 4a, 4cd and 10c; (2) channel and rectangularly shaped (Fig. 2.5), but also subhedral to euhedral negative crystal fluid inclusions on secondary or pseudosecondary trails, found in all samples; (3) fluid inclusions in mineral inclusion apatite (Fig. 2.1) and carbonate (Fig. 2.2).

All fluid inclusions homogenized to the liquid phase, implying a density increase with decreasing *Th*, and have $Te = Tm = -56.3 \pm 0.3\,°C$, characteristic for pure CO_2. Raman microspectrometry has confirmed that the fluid inclusions in the corundums are pure CO_2, without admixture of significant N_2, H_2S, CH_4 or other hydrocarbons.

Daughter minerals occur in fluid inclusions on trails in the Ratnapura corundum 10c (Fig. 2.6); Raman microspectrometry indicated the presence of diaspore (Peaks at $\bar{v} = 795$, 711, 665, 495, 446 cm^{-1}), rutile (peaks at $\bar{v} = 637$, 598, 440 cm^{-1}), corundum (peaks at $\bar{v} = 748$, 574, 413 cm^{-1}) and an unidentified mineral, probably a carbonate, sulphate or phosphate (peaks at $\bar{v} = 1190$, 667 cm^{-1}). Though diaspore daughter minerals suggest the presence of water in these daughter-bearing carbonic fluid inclusions, liquid water was never seen.

Fig. 3 shows histograms of *Th* of carbonic fluid inclusions, isolated, on trails, and in mineral inclusions for the seven corundum crystals. When all data are considered, the isolated fluid inclusions and fluid inclusions on trails show a wide variation of *Th* between −25 and +23 °C, corresponding to densities between 1.05 and 0.79 g/cm^3; fluid inclusions in the mineral inclusions apatite and carbonate have *Th* between −5 and +2 °C, corresponding to densities between 0.95 and 0.92 g/cm^3. Fluid inclusions from individual samples however, lead to some significant observations.

The isolated fluid inclusions in samples 2a and 4cd show well-preserved negative crystal forms and a Th of -25 ± 5 °C (Fig. 3), corresponding to a density of 1.05 ± 0.03 g/cm^3; such high-density inclusions are here considered to be unmodified primary fluid inclusions, representing fluids entrapped during the metamorphic crystallization of corundum in the source area. Isolated fluid inclusions in samples 4a and 10c show higher *Th* (Fig. 3) and thus lower density; they also contain some large (up to 300 μm) isolated inclusions with very low densities of 0.75 to 0.88 g/cm^3, and it is believed that in such cases leakage of primary fluid inclusions has occurred.

The fluid inclusions on trails in samples 1b, 2b, 2c and 4a show a narrow spread of *Th* around peak values of about −9 °C (Fig. 3), corresponding to a density of about 0.975 g/cm^3. Fig. 4 shows Th histograms of fluid inclusions on separate trails in 1b, 2b and 2c; it appears that fluid inclusions on individual trails are characterized by a uniform *Th*, and that between trails there is a narrow spread of *Th*.

The fluid inclusions on trails in 2a show a rather broad spread of *Th* overlapping the spread of *Th* in 1b, 2b, 2c and 4a, but centred around lower *Th* (Fig. 3). The fluid inclusions on trails in 4cd and 10c show a broad spread of *Th* with peaks near −9 °C (density 0.975 g/cm^3) and +9.5 °C (density 0.88 g/cm^3); 10c shows still higher *Th* values with a peak around +22 °C (density 0.795 g/cm^3). Fig. 5 shows histograms of *Th* of fluid inclusions on separate trails in 2a, 4cd and 10c; 4cd and 10c show higher *Th*, lower-density fluid-inclusion trails with uniform *Th* on the same trail, and lower *Th*, higher-density fluid-inclusion trails showing a spread of *Th* on the same trail. This spread of *Th* is due to the occurrence on the same trail of relatively large inclusions with negative crystal form and a relatively low *Th*, besides small rectangular and channel-shaped inclusions with a relatively high *Th*; these features suggest an evolution from high- to low-density fluid during fluid entrapment on trails along healing fractures in the host corundum.

Origin and *PT* history of corundum crystals

Origin. The investigated corundum crystals contain two types of solid inclusions.

(*a*) Regularly oriented abundant rutile exsolution needles in growth zones. This suggests the formation of zoned corundum porphyroblasts in titaniferous aluminous rocks with diffusion-controlled alternating Ti-free and Ti-rich growth zones. The association of Ti-rich blue and Ti-free colourless corundum in the gem gravels suggests source areas with varied lithology of silica-undersaturated aluminous rocks with different TiO_2 contents.

(*b*) Isolated crystals trapped by the host corundum during its growth (primary solid inclusions). Kataragama corundum contains apatite and calcite; Embilimpitiya corundum contains zircon. Some of these solid inclusions contain CO_2 inclusions. Possible glass inclusions have been observed under the microscope in Embilimpitiya corundum. The diversity of solid inclusions in the corundum suggests source areas with a varied lithology of magmatic silica-undersaturated aluminous rocks of different TiO_2-contents. It may also be remarked that the occurrence of calcite, apatite and zircon, minerals which are characteristic of carbonatite rocks, might suggest a genetic relation with carbonatitic rocks, as was proposed by Touret (1984) for granulite rocks from southern Norway.

History. The Highland Series of Sri Lanka is a polymetamorphic Archean–Proterozoic complex. The imprinted granulite facies metamorphism took place most probably about 1100 Ma ago (Kröner *et al.*, 1987). Schumacher and Schenk (1987) have distinguished a first (820 °C, 8.5 kbar) and second stage (630 °C, 5.5 kbar) granulite facies metamorphism. Prame (1987) has provided

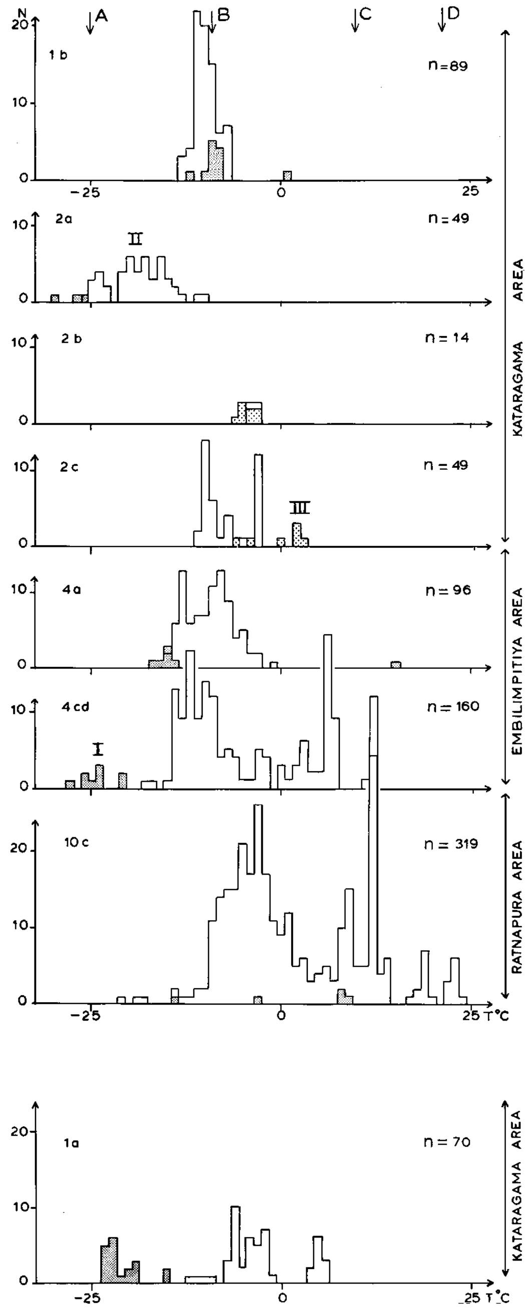
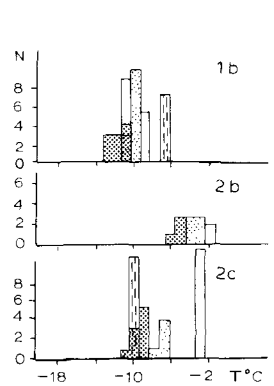
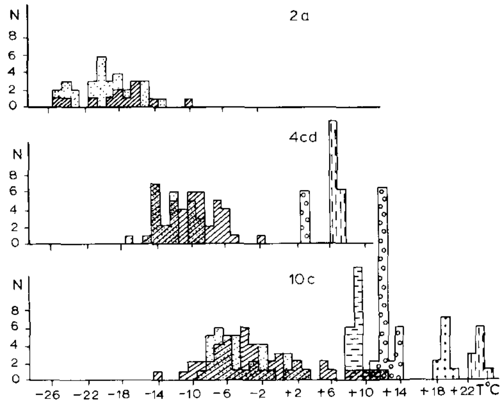


FIG. 3. Frequency histograms of homogenization temperatures of carbonic fluid inclusions. All homogenization in the liquid phase. 1b corundum from Amarawewa; 2a, 2b, 2c corundum from Kochipadana; 4a, 4cd corundum from Embilimpitiya; 10c corundum from Ratnapura; 1a quartz from Amarawewa. I (heavy stippled): isolated (primary) inclusions; II (white): trail bound secondary inclusions; III (light stippled): isolated inclusions, occurring in solid inclusions in corundum (e.g. apatite or carbonate, see Fig. 2–1 and 2). n is total number of measurements. N is number of measurements taken. A, B, C and D represent the position of isochores of Fig. 6.





FIGS. 4 (*left*) and 5 (*right*). Frequency histograms of homogenization temperatures of fluid inclusions on separate trails. Each separate symbol indicates a different trail.

estimates of 700–800 °C, 5.5–6.5 kbar for granulite facies rocks of the Highland Series. Current research by De Maesschalck on granulite facies garnet gneisses of Weddagala (Ratnapura area) Sri Lanka gave 720 ± 20 °C (garnet–biotite thermometer) and 6.7 ± 0.3 kbar (garnet–sillimanite–plagioclase–quartz barometer). These data and also the *P–T* path of Schenk *et al.* (1988) are shown in Fig. 6. Fig. 6 also shows CO_2-isochores for d = 1.05 (A), d = 0.975 (B), d = 0.88 (C) and d = 0.795 (D) g/cm^3, constructed according to Holloway (1977). The A isochore, representing the density of primary carbonic fluid inclusions in corundum, passes through the *PT* boxes of Prame (1987) and De Maesschalck *et al.* (in preparation) and is also close to the second stage *PT* point of Schumacher and Schenk (1987).

The corundum samples contain two groups of carbonic fluid inclusions, which could serve to mark events in the metamorphic history of the source areas:

(1) Primary, high-density fluid inclusions with an average density d = 1.05 g/cm^3, entrapped in corundum during the peak of metamorphic recrystallization in the source area.

(2) Secondary, lower-density carbonic fluid inclusions entrapped in secondary trails and related to retrograde late- or post-metamorphic events; these inclusions have densities clustering around d = 0.975, d = 0.88 and d = 0.795 g/cm^3.

A quartz crystal (1a) from a gem pit in the Kataragama area has yielded comparable results with primary carbonic fluid inclusions clustering around $Th = -23$ °C, d = 1.04 g/cm^3 and secondary carbonic fluid inclusions clustering around $Th = -7$ °C, d = 0.96 g/cm^3 and $Th = +6$ °C, d = 0.89 g/cm^3 (Fig. 3).

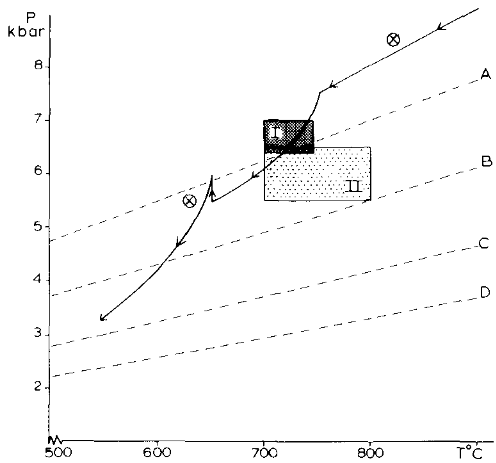


FIG. 6. *P–T* interpretation of fluid inclusion data. Lines A, B, C, and D represent isochores; A for d = 1.05, B for d = 0.975, C for d = 0.88 and D for d = 0.795 g/cm^3. I (De Maesschalck, current research) and II (Prame, 1987) show *P–T* estimates from mineral assemblages for peak metamorphic conditions. ⊗ Represents two stages of granulite-facies metamorphism (Schumacher and Schenk, 1987). The arrow shows the *P–T* path of Schenk *et al.* (1988).

It is concluded that primary unmodified carbonic fluid inclusions, with a high density of about 1.05 g/cm^3 is compatible with the formation of the host corundum under granulite-facies metamorphism (>630 °C, 5.5 kbar) in the Highland Series source rocks. The lower-density carbonic fluid inclusions entrapped along secondary trails are presumably related to retrograde events of cooling and uplift after the granulite-facies metamorphism. The secondary fluid inclusions in Kataragama corundum show a narrow spread of relatively high densities around isochore B (Fig. 6), suggesting a source area with a relatively simple retrogression history with *PT* conditions changing along the B isochore. The Ratnapura corundum shows a very wide spread of carbonic fluid inclusions from high to very low densities (to d = 0.795 g/cm^3), while diaspore in most of the carbonic inclusions indicates admixture with aqueous fluids. It is suggested that the source area of Ratnapura corundum has a more complex history of post-metamorphic uplift and cooling, resulting in entrapment of secondary fluid inclusions under the lower temperature and pressure conditions depicted by the C and D isochores in Fig. 6. The fluid inclusion characteristics of Embilimpitiya corundum is generally intermediate between those of Kataragama and Ratnapura corundum.

The differences in populations of secondary fluid inclusions along trails in corundum from Kataragama, Embilimpitiya and Ratnapura might reflect differences in the post-metamorphic history of cooling and uplift of the different source areas of the gem gravels.

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