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Study of Macroscopic and Microscopic Reaction Textures Associated with Corundum of Balangoda Region, Sri Lanka

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ABSTRACT

Corundum is the most sought after gem variety among Sri Lankan gem minerals. Locations of most of the gem quality stones were identified as alluvial. The in-situ corundums rarely show a gem quality and are mostly associated with macroscopic and microscopic reaction textures. Studies of these textures, associated reactants and the intrusion of corundum by reaction products using electron probe microanalysis and energy dispersive x-ray analysis reveal that hydrous minerals such as phlogopite mica play an important role in promoting such reactions. Because of these reactions corundum is likely to be fractured and the fractures are often filled with reaction products. Hence the gem quality is not retained in most corundum after their formation. Also the natural shielding effect, called plagioclase shielding that protects corundum against the retrograde reactions, is found to be responsible for the production of valuable gem quality stones.

Keywords: Reaction textures, Corundum, Calc-silicate, Granulite, Mineralization

INTRODUCTION

Sri Lanka is famous for many varieties of gem minerals from time immemorial and among them corundum is conspicuous since ancient times [1]. It is the only gem mineral that accounts for more than half of Sri Lanka's gem exports. Furthermore, Sri Lanka is a major supplier of top quality blue and white sapphire. Most of the corundums are found in alluvial gem gravels, while a few corundum bearing source rocks are scattered at several locations. The latter rarely produce gem quality crystals, and one objective of this study is to find out the reason for this phenomenon.

For this study, samples covering the whole gem-bearing regions of Sri Lanka were collected. However, only the samples from Kaltota of Balangoda Region, were investigated in detail because all types of gem-bearing source rocks so far recorded such as corundum bearing granulitic gneiss and calc-silicate gneiss are found there (Fig. 1).

Balangoda is situated approximately half way on the Colombo-Badulla road and Kaltota is 26 kilometers from Balangoda. This area is famous for top quality stones of many varieties, including rubies and sapphires.

Reaction textures of both macroscopic and microscopic nature were studied to identify original characteristics of gems, as well as the transformations that had taken place to deteriorate their quality, subsequent to the formation. The macroscopic reaction textures associated with corundum are important in the sense that they reveal how corundum is affected after the formation.

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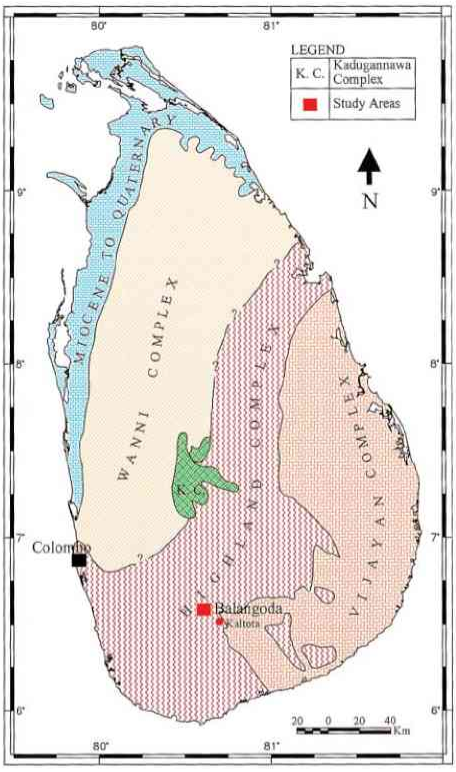

Fig. 1 Geological map illustrating the distribution of crustal units of Sri Lanka (modified after Ref. 2).

GEOLOGICAL SETTING

Geologically, the basement rocks of Sri Lanka are subdivided into three distinct crustal units on the basis of isotopic, geochronological, petrological and geochemical data. They are the Highland Complex (HC), the oldest and the largest unit (2.0–3.4 Ga), the Wanni Complex (WC) (1.1–1.8 Ga), and the Vijayan Complex (VC) (1.1–1.8 Ga) (Fig. 1) [2–4]. Besides these main crustal units, another unit called as Kadugannawa Complex consisting of mainly biotite-hornblende gneisses, resembling the gneisses of the Vijayan Complex, is recognized in an area characterized by upright synformal structural basins around Kandy [5]. The study area belongs to the Highland Complex that mainly consists of garnet-sillimanite-biotite gneiss, charnockitic gneiss, granodioritic gneiss, undifferentiated calc-gneisses, marble, quartzite, hornblende-biotite gneiss, garnetiferous quartzo-feldspathic gneiss, undifferentiated charnockitic biotite gneiss and pegmatite (Fig. 2). The pegmatite is a common occurrence in the area. According to the geological setting (Fig. 2), the presence of calc-gneiss and associated 'skarn' mineralization in proximity to Al-rich pelitic gneiss at relatively high paleo-pressures and temperatures (> 8 kb, > 750°C) coupled with uplift and cooling might have a bearing on the formation of gems. The skarn mineralization is associated with the garnet-sillimanite-biotite gneiss called khondalite mechanically incorporated in the tectonic breccia. The conditions such as these, to which the area had been subjected to, might have influenced the formation, and as such the discovery of in-situ or near in-situ gem occurrences. The magmatic rocks, which include pegmatite and granite, are supposed to be the source rocks for some of the gems found in the study area [6].

Metamorphosed supracrustal rocks, meta-igneous rocks (nearly 30–40%) and a variety of pegmatites occur in the study area. Of metasediments, quartzite is the most abundant. Calc-silicate rock occurs close to the major source rock of corundum and spinel. Metamorphosed supracrustal rocks included quartzite, metapelites, quartzo-feldspathic gneisses and abundant calc-silicate gneisses. The major metamorphosed igneous rock was metadiorite. Deformed basic dykes and orthopyroxene-bearing orthogneiss are also present. In addition some charnockitic gneisses also belonging to the meta-igneous category were present.

Field observations and aerial photographs revealed that the Kaltota area is highly deformed and folded, forming a series of nearly upright and linear antiforms and synforms. Many gem deposits were located closer to the major shear zones, more or less parallel to the axial traces of large-scale folds [7]. A unique feature of this area is the occurrence of platy corundum crystals (short, tabular and prismatic) which are usually rare elsewhere.

REACTION TEXTURES AND RESULTS OF THEIR OBSERVATION

Macroscopic and Microscopic Textures

A variety of reaction textures associated with the minerals of source rock is observed in corundum-bearing rocks. Macroscopic and microscopic reaction textures provide clues to the origin of corundum/spinel and their metamorphic alterations.

The reaction textures, described in this paper, were observed in corundum and spinel-bearing calc-silicate rocks, collected from Neluyaya in the Kaltota area. The rock contained minerals such as scapolite, plagioclase, short- prismatic as well as bipyramidal corundum, spinel, phlogopite and K-feldspar along with traces of graphite, zircon, ilmenite and sapphirine. The upper surface of the rock is highly weathered; hence corundum crystals could be

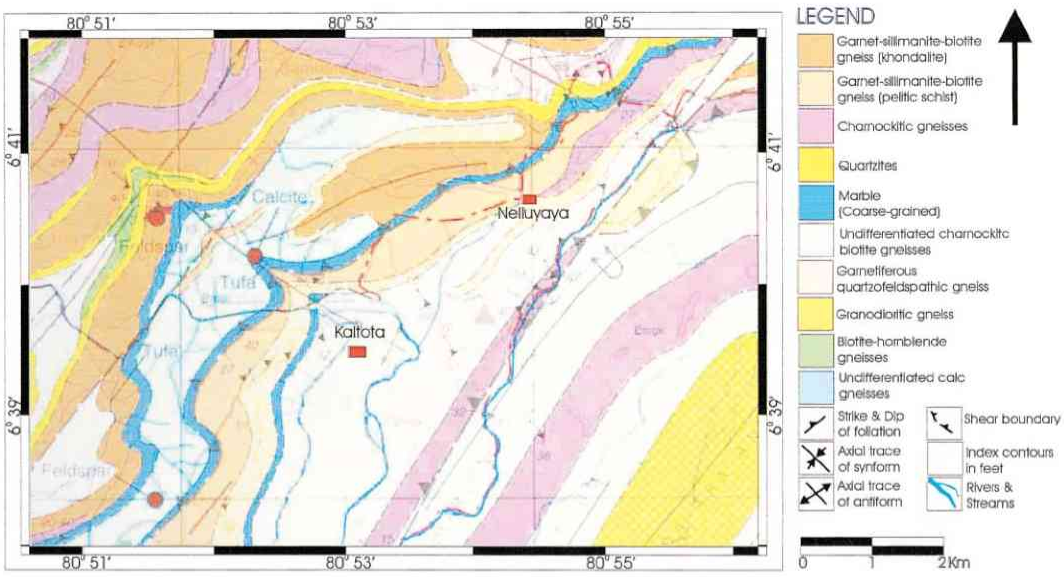

Fig. 2 Geological map of the Kaltota area (Source - GSMB 1 : 100,000 map of Nuwara Eliya- Haputale, after Ref. 6).

hand-picked. The rock is similar to the one found in the Elahera Region [8].

Methodology of Studying Reaction Rims/Textures

Normally most of the reaction rims/textures can be scrutinized with the naked eye or through a 10x lens. Therefore, they are called macroscopic textures. The rest falls into the microscopic category. Gemmological and petrological microscopes facilitate detailed investigation. Scanning electron microscope and transmission electron microprobe enable study at very high magnification, hence finer details could be studied and compared with corundum found elsewhere.

During this study, the samples were first observed under an optical microscope. Thereafter major and minor elements (except trace elements) of included mineral phases were analyzed by electron microprobe (CAMECA SX50). Polished sections of corundum and corundum-bearing rocks were prepared for analysis. The electron microprobe was equipped with four wave dispersive (WDS) spectrometers and an energy dispersive (EDS) attachment. Analyses were made with an accelerating voltage of 15 kV and a sample current of 20 nA.

Finally, x-ray micro-florescence (XRMF) study of rims, surrounding the corundum crystal, was carried out. Data was collected in continuous raster mode using the four wavelength dispersive spectrometers of the microprobe. Resulting element images depicted more than 200,000 pixels per element map. Data collection was performed within a period of 0.2 seconds along each measuring distance of about 6 μm. Special data handling and image-forming software, provided by the microprobe manufacturer, facilitated the image formation.

The image processing software of the analytical system produced images of sample with a color code to display varying concentrations at each location analyzed. Variable element concentrations were displayed pixel by pixel on the X-ray images in a sequence of black→ blue→ green→ yellow→ red→ pink→ white.

Macroscopic Reaction Textures

In most fresh corundum-bearing rocks, corundum crystals were enclosed by thin spinel crusts (Fig. 3). In the same calc-silicate rock, there were some portions with abundant phlogopite mica and some with-

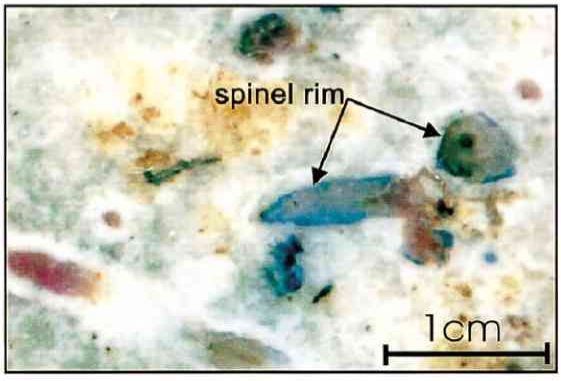

Fig. 3 Corundum crystals with thin spinel crusts.

out (mica exhausted).

Generally, corundum-bearing rock was subjected to regional granulite grade (about 800°C and 8 kb) metamorphism. In spite of these conditions corundum could have been stable during metamorphism. Yet during the isobaric cooling in the hydrous environment, corundum-bearing assemblages probably had retrograded to form mica varieties such as phlogopite. Hence, phlogopite mica could be seen associated with corundum in the source rock.

Within areas devoid of mica, the spinel rim around the corundum crystal was not perfectly hexagonal (Fig. 4 and Table 1). Observations revealed that the outer edge of the corundum (Fig. 4) was eroded to some extent, as a result of a reaction (Fig. 5). Therefore, it is assumed that there was a reaction between corundum and phlogopite mica to form spinel as the major product along with a trace of K-feldspar. This reaction could have been continuous until all the mica was exhausted. The reaction suggested on the basis of investigations of a hand specimen and a thin section is illustrated in Fig. 5 [9].

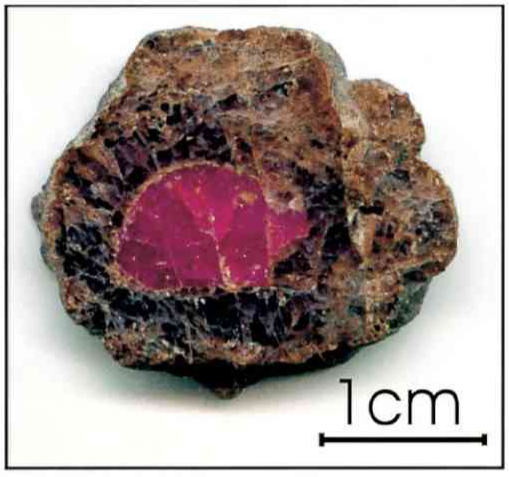

Fig. 4 Corundum crystals with thick spinel rim.

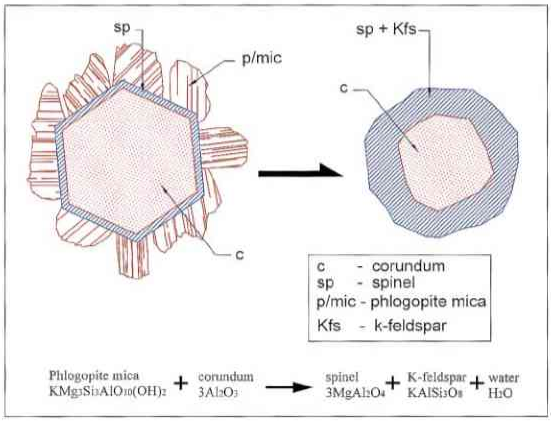

Fig. 5 Schematic illustration of the non-uniform enlargement of spinel rim due to the reaction of corundum and phlogopite (after Ref. 9).

In the later stage of the study, the minerals and the products resulted from such reactions were confirmed by the spot analysis using electron microprobe. This was a semi-quantitative and non-destructive analysis. The spot analysis was carried out for some selected minerals. In order to reduce the errors by multiple spot analyses, each mineral was targeted with a beam spot of around 5 μm. The average value was calculated and it was used for further calculations (Table 1).

Analysis of the spinel by electron microprobe reveals that spinel embedded in the rim surrounding corundum (Fig. 3) before the reaction contains more magnesium and less aluminum than the subsequent spinel (Fig. 4) resulting after reaction. The difference in these spinels of two stages clearly indicates the role of some sort of reaction, which led to the increase in the amount of spinel and enlargement in the thickness of the rim but a reduction in the amount of corundum. Hence, there is decrease in the magnesium content from the initial one (see Fig. 6, for comparison).

In order to confirm the mechanism of this process, backscattered images were taken and XRMF study was carried out for a minute portion of the selected part of the rim that showed the signs of

Table 1 EPMA analyses of corundum and the minerals constituting its rim.

Oxide	Corundum	Spinel	Sapphirine	K-Feldspar
(wt%)	(N = 9)	(N = 7)	(N = 5)	(N = 2)
Na_2O	0.01	0.01	0.02	4.91
MgO	0.02	20.37	17.76	0.01
Al_2O_3	98.99	69.13	64.47	19.31
Cr_2O_3	0.13	0.1	0.09	0
MnO	0.02	0.08	0.03	0.01
FeO_{tot}	0.25	5.68	1.47	0.01
CaO	0.01	0.01	0.03	0.56
TiO_2	0.03	0.01	0.06	0
K_2O	0	0	0.01	9.41
SiO_2	0.05	0.03	12.37	62.17
Total	99.51	95.42	96.31	96.39

Note: FeO_{tot} = Total iron oxide expressed as FeO, Accuracy of EPMA = ± 0.0 1%; Values given are averages for N specimens; Accuracy of EPMA = ± 0.01%

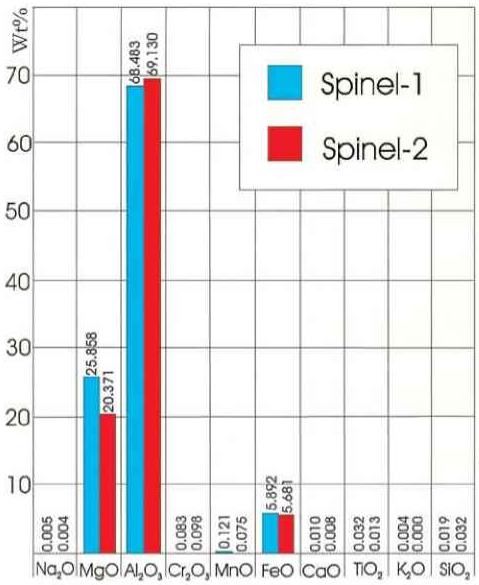

Fig. 6 Comparison of the spinel compositions of the specimens, representing the stages before (Spinel-1) and after (Spinel-2) the reaction.

partial transformation to spinel (Fig. 7). Then the same sample was subjected to XRMF and element mappings of elements such as K, Na, Ca, Al, Si and Mg (Fig. 8).

Occurrence of phlogopite mica is suggested from the red patches in the K map. The red colour patches, however, are no longer visible around the spinel. This indicates that the corundum → spinel reaction had continued until all the adjoining phlogopite was exhausted. For this type of reaction, fluids were necessary to transport material under the chemical potential gradient. As a result of this type of reaction, the quality of corundum was significantly reduced and this was true for almost all the in-situ corundum in Sri Lanka.

Microscopic Reaction Textures

The initial observation clearly indicated what happened to corundum crystals after their formation. But further observation indicated some processes that provide natural shielding to corundum crystals

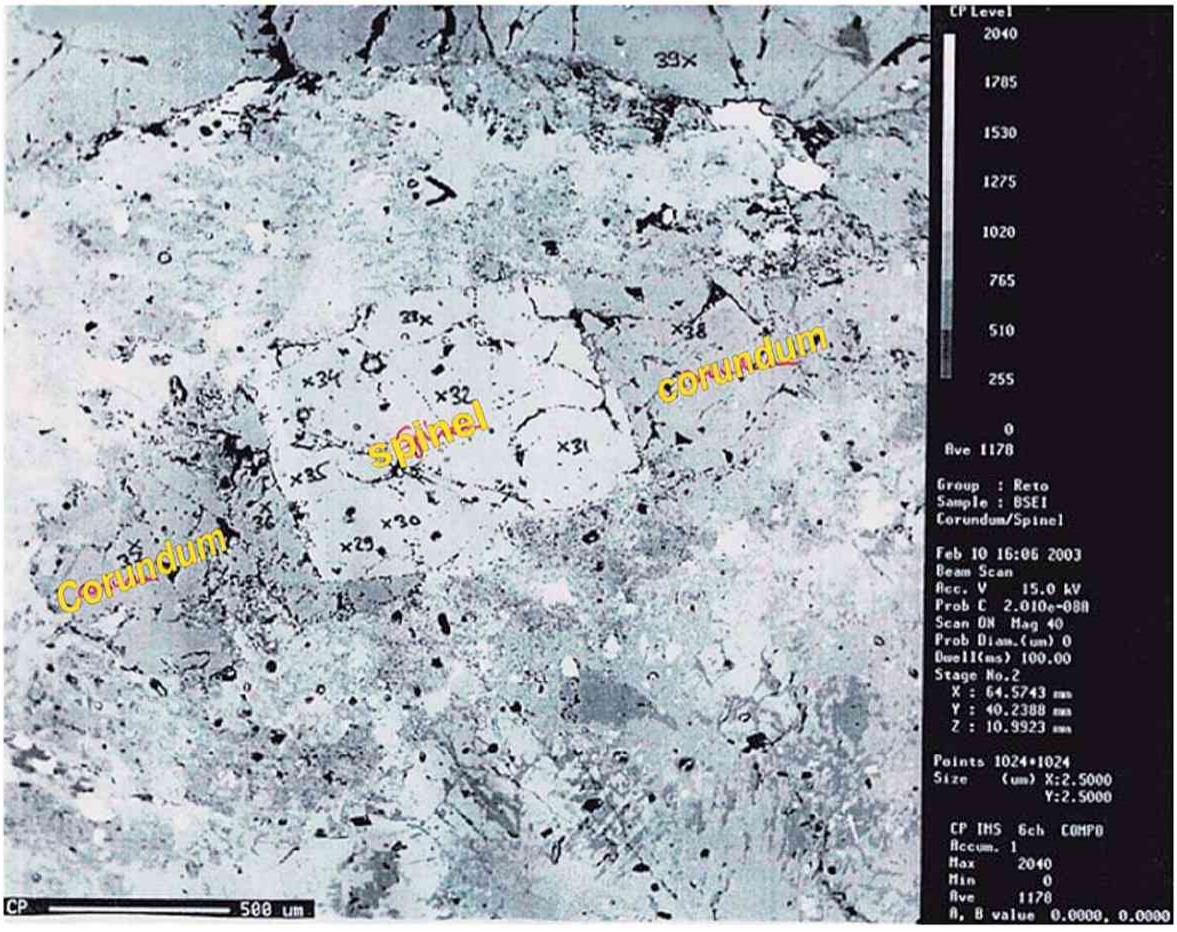

Fig. 7 A small protruded part of spinel rim with microscopic remains of corundum, subjected to backscattered imaging.

to guard them against the quality deterioration resulting from various processes such as retrograde reactions that produce mica minerals, etc. This was also recorded as microscopic reaction textures in corundum bearing calc-silicate rocks. These calc-silicate rocks contain corundum, spinel, plagioclase, scapolite, sillimanite, phlogopite and magnesite along with accessory minerals such as graphite, zircon and

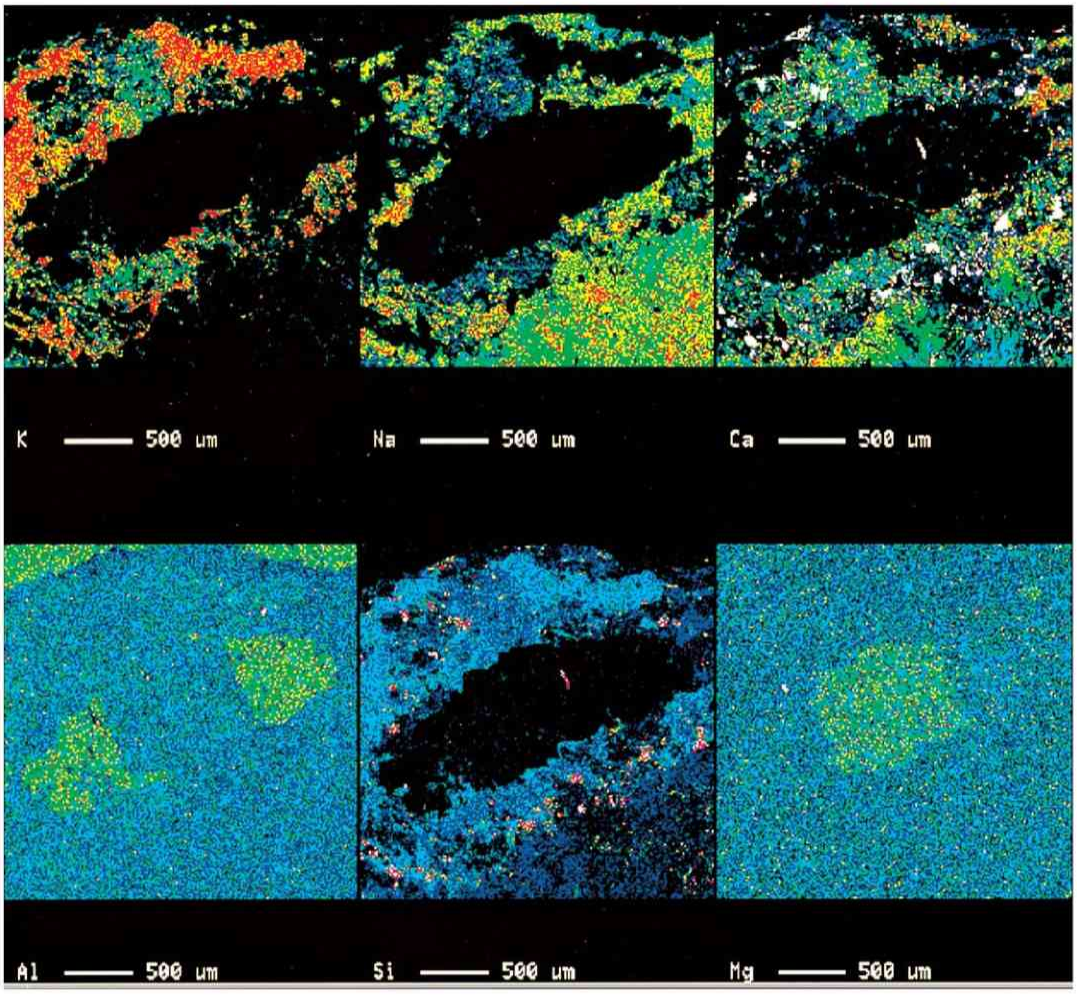

Fig. 8 Element distribution map illustrating the formation of spinel as a byproduct of the reaction between phlogopite mica and corundum. Red and yellow areas within the K map, indicate K rich areas, probably containing phlogopite.

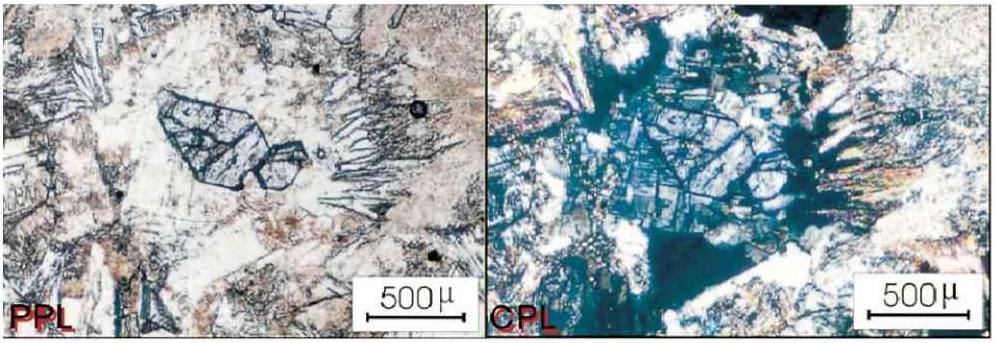

Fig. 9 Photomicrographs illustrating a corundum crystal surrounded by re-crystallized plagioclase grains.

apatite.

As a result of the point-counter analyses, considerable amount of plagioclase (46% - model percentage) was observed in these calc-silicate rocks. This plagioclase is rich in Na and it is composed mostly of re-crystallized grains. Sometimes plagioclase occurs around the corundum crystals as either complete or partial rims. Very rarely, corundum crystals occur without the plagioclase rim.

A corundum crystal completely enclosed by the re-crystallized plagioclase grains was a significant finding. It was seen to be intact with perfect edges, undamaged in spite of the mineral reactions (Fig. 9). But in the case of a corundum crystal which was partially enclosed by re-crystallized plagioclase grains, the uncoated areas were fractured and some of the fractures were filled with the reaction product, namely the phlogopite mica (Fig. 10). Some corun-

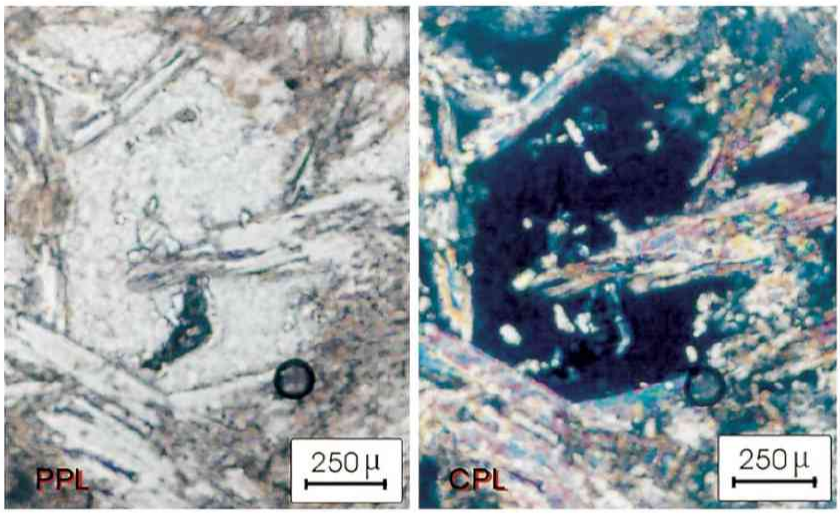

Fig. 10 Photomicrographs illustrating corundum partially surrounded by re-crystallized plagioclase grains. The portion of corundum devoid of plagioclase rim is subjected to the reaction process of phlogopite mica formation.

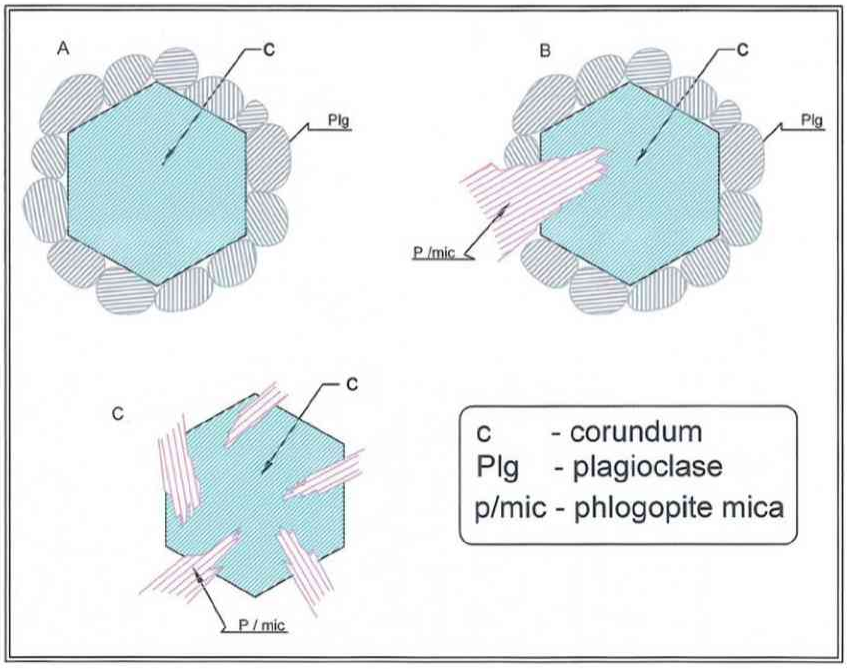

Fig. 11 Schematic illustration of the plagioclase shielding effect over corundum (after Ref. 10).

dum crystals were found totally damaged by the reaction and the spaces created thus were filled with phlogopite mica.

These observations suggest that plagioclase coating acted as a shield for corundum by safeguarding it from the progress of reactions, such as phlogopite mica formation (Fig. 11). The shielding material was analyzed with the use of electron microprobe and was identified as Na-rich plagioclase. In this specific case plagioclase was not present as an inclusion in corundum [10]. For the time being, the reason for the occurrence of plagioclase shielding in certain corundum-bearing rocks is unclear, and the future study would reveal the actual reasons for its occurrence.

DISCUSSION

Most gem quality corundums in Sri Lanka were found as alluvial material but they are believed to have originated as a result of metasomatic processes. In Sri Lanka, corundums found in a few in-situ deposits, which are rather scattered, are rarely of gem quality. This is attributed to the alterations due to reactions described above and the deformation that followed afterwards. However, even then some natural shielding effects probably worked to protect corundum against the retrograde reactions that form mica minerals, up to a certain extent. The plagioclase shielding effect, which is just one such example, provides some insight into the reason why some corundum crystals are far superior to industrial quality corundum. This is the first time to record the plagioclase-rim around corundum in Sri Lanka at microscopic scale. Further study is needed to elaborate at length on this effect.

ACKNOWLEDGMENTS

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