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An update study in micro-inclusions of emerald from worldwide significant deposits

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Abstract. A conclusive study of inclusion in schist-type emeralds from some famous deposits in the world is introduced. The deposists includes Santa Terezinha, Socoto, Capoeirana, Itabira (Brazil), Mananrary (Madagascar), Transwaal (South Africa), Ural (Russia). The typical inclusions for emerald of these localities are quartz, mica, amphibole, fluid inclusions. Particular and unique features of inclusion could provide information of geographic orgirin. Itabira emerald is featurelized with the abundance of multi-phase inclusions containing two liquids, developed in well-formed negative crystals; more or less square, rectangular cavities. Santa Terezinha deposit is charactierized with pyrite inclusion which is not found in other deposits.
Keywords: Inclusion, Brazil, Ural, Madagascar, South Africa, emerald.

1. Introduction

A comparative study in inclusion between schist-type and non-schist type beryl has been pulished by the author (Huong, 2012) [1]. Based on the inclusion suits a gemmologist could identify from which mine/deposit a gemstone originated, i.e. schist type or non-schist type. This paper introduces an update and conclusive study in inclusion of the most favourist gems of this group, i.e. emerald, from some worlwide famous deposits. Following the former study (Huong, 2012) [1], observations from this study leads to the conclusion that inclusion features can provide information to identfy not only between schist and non-schist types but also among geographic orgins of gemstones in many cases.

2. Material and methods

For this research, 179 natural facet-cut emerald samples have been investigated in this study. The samples were collected from Brazil (Carnaiba, Itabira, Santa Terezinha, Socoto), Madagascar (Mananjary), Russia (Ural), and South Africa (Transvaal). All emerald samples studied belong to schist type.

The inclusions were firstly observed, described, and classified using a gemmological microscope with Zeiss optics. Then all of the different types of inclusions were photographed

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and determined using confocal-Raman spectroscopy. All host emerald samples were polished at two parallel sides with the thickness varying from 1 mm to 4 mm. The experiments determining inclusions of emerald were carried out on a LabRam confocal micro-Raman-system HR-800 equipped with an Olympus-BX41 by JOBIN YVON HORIBA. For searching inclusions as well as measuring a certain point, an objective with a 50 times magnifying power and green laser light (514.532 nm) were used. Raman spectroscopy is a non-destructive technique to identify not only solid but also fluid inclusions in gemstones.

3. Results and discussion

3.1. Santa Terezinha (Brazil)

Mineral inclusions are found to be abundant in emeralds from the Santa Terezinha deposit. They are listed as chromite, pyrite, calcite and two-phase inclusions. Other minerals that we considered as typical inclusions are hematite, goethite, amphibole, feldspar, quartz, and magnetite. Pyrite inclusions normally occur as sharp or slightly rounded cubes. They can occur solitaire or in groups. Numerous minute crystals may form tiny clouds.

Figure 1a. Chromite inclusion ($FeCr_2O_4$) in Santa Terezinha emerald as a well shaped octahedron. x50.

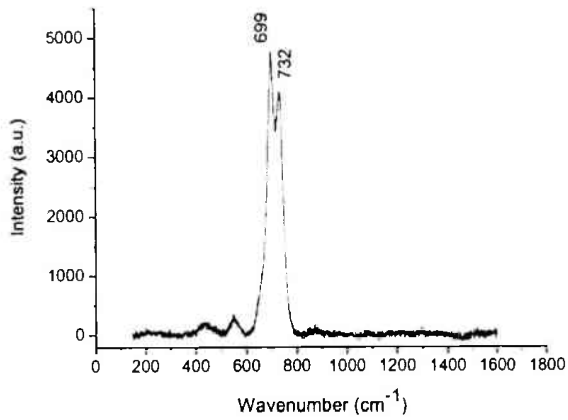

Figure 1b. Raman-spectrum obtained from chromite inclusion in Santa Terezinha emerald.

Chromite is present as black rounded crystals or in octahedrons (figure 1a). The big individual crystals are isolated, and the small ones form irregular clouds or trails parallel to the basal faces. Carbonate minerals are colourless to brown, irregularly bordered grains, found either singly or in groups. They were determined by Raman microscopy to be calcite, dolomite, hydrozincite and magnesite (figure 2a).

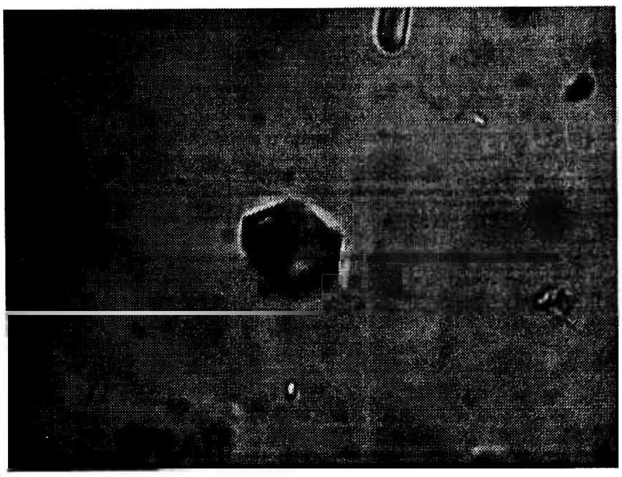

Figure 2a. Magnesite ($MgCO_3$) in Santa Terezinha emerald. x50.

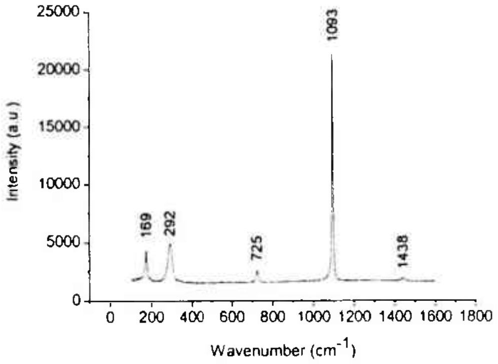

Figure 2b. Raman-spectrum obtained from magnesite in Santa Terezinha emerald.

Talc minerals are colourless, transparent to white or silky, in flake-like shapes (figure 3a). They are sometimes gathered in agglomerations that make the crystals appear cloudy. Mica inclusions were determined to be biotite as brown flakes.

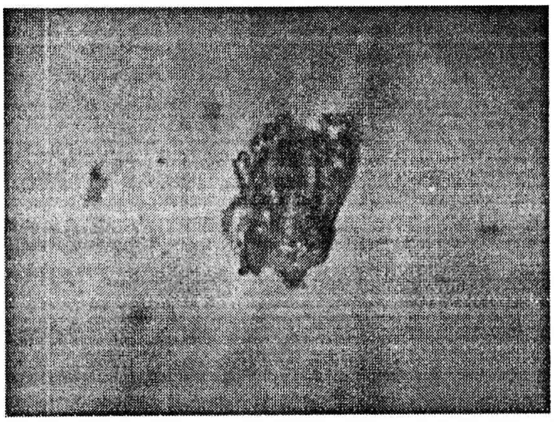

Figure 3a. Talc flakes usually are very small aggregates in Santa Terezinha emerald, the big one as shown in photo is very infrequent. x50.

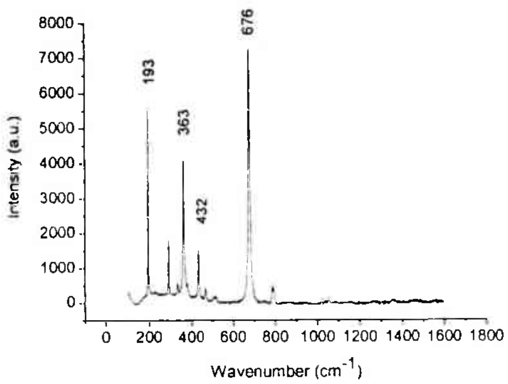

Figure 3b. Raman-spectrum obtained from talc inclusion in Santa Terezinha emerald.

3.2. Socoto (Brazil)

The great variety of different mineral inclusions is found to be the most characteristic feature of which mica is the most frequently observed inclusion. Micas are usually biotite and phlogopite, and less frequent they can be margarite or muscovite. This is due to the fact that the largest portion of the Socoto emeralds is found in a biotite/phlogopite schist host rock. Normally, the mica crystals occur in the form of rounded or irregular-shaped platelets. Their colour is generally light to dark brown. Margarite and muscovite are practically colourless. Mica crystals rarely occur isolated, mostly they form agglomerations. It is the agglomerations of mica that sometimes make the host emerald crystal have dark brown colour (figure 4) and partially appear almost opaque. Sometimes, tubes are observed orientated in the direction of the c-axes and are accompanied with mica (figure 5). A part from mica, chlorite inclusions are observed, which are sometimes not distinguishable from mica without the help of confocal-Raman microscopy.

Figure 4. Only rarely isolated occurring mica crystals, usually forming agglomerations, darkening the crystal. x50.

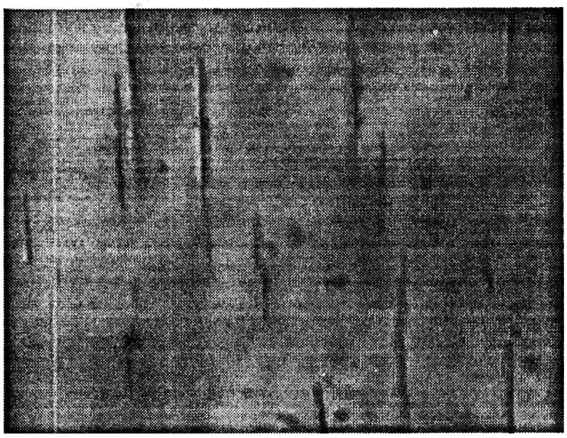

Figure 5. Fissure system parallel to the c axis in emeralds from Socoto. x10.

Other inclusions found in Socoto emeralds are actinolite, tremolite, apatite, talc, quartz, albite, molybdenite and hematite/goethite/ lepidocrocite, in which molybdenite is least frequently observed and considered as a rare mineral inclusion. Actinolite/tremolite sometimes forms thick needles or rods that are practically colourless and transparent. They normally occur isolated but sometimes are found to be bundles of numerous crystals showing no preferred orientation. Besides the colourless, transparent crystals, others of greenish to light brownish crystals occur that sometimes show the characteristics of a bamboo-like appearance.

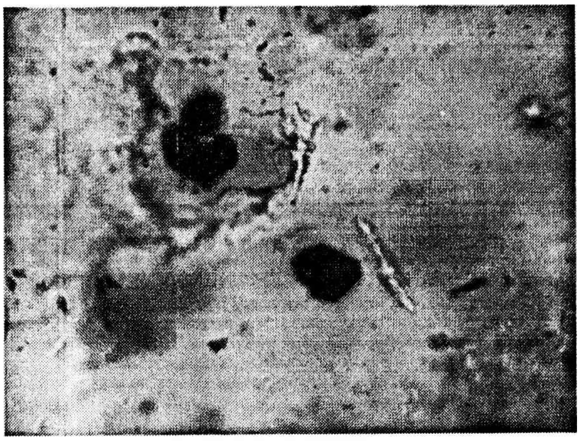

Figure 6. Lepidocrocite (FeOOH) with very intensive red colour, with brownish hematite (Fe_2O_3). x50.

Figure 7. The dark brown core zone is caused by the conglomeration of mica and some carbonate minerals. x50.

Observation in all samples indicates that the importance of actinolite/tremolite as mineral inclusions falls clearly behind that of micas. The appearance of isolated albite crystals and of fractures filled with feldspar in the emeralds can be explained also by the fact that the Socoto emeralds are partly found in feldspar masses. Hematite occurs as small, irregular crystals that can be found mostly within fissures or dispersed over the surface of the emerald crystals. Lepidocrocite can be found sometimes to be associated with hematite and shows a very strong red colour (figure 6). Apatite occurs as prismatic crystals that are sometimes slightly corroded and rounded. They are colourless and they almost always show cleavage planes parallel to the basal phase. Based on their appearance within the emerald host crystal we can define them as protogenetic inclusions. Besides the mineral inclusion mentioned above, there are talc crystals occurring in the form of transparent, colourless platelets. According to Schwarz et al. (1990) [2], Eidt and Schwarz (1986) [3], tourmaline and orthite can be observed as rare inclusions in emeralds from Socoto also. The core with darker colour is caused by the high inclusion density, in general, they are dark brown mica, and may be carbonate minerals, or others. The presence of zonation (figure 7) shows that the growth process of Socoto emeralds is characterized by the repeated abrupt alterations of the forming environment.

3.3. Capoeirana (Brazil)

This study brings out quartz, mica (biotite) inclusions in emeralds from Capoeirana. Quartz inclusions are colourless, transparent, and can be found either as rounded grains or in long prismatic forms. Rounded quartz grains were found usually in groups, distributed irregularly within host crystals (figure 8). Long prismatic quartz crystals were found as singly transparent ones, orientated parallel to the c-axis. In some cases, quartz was found to be associated with fracture systems or liquid inclusions. Mica inclusions were found less frequently than quartz. Not only in one sample but also in the whole sample set, quartz inclusions are more abundant than mica. Micas were observed as transparent to translucent slightly greyish brown flakes and booklets or somewhat rounded grains and they are not distributed in any certain place inside the host crystal. In addition to mica and quartz, siderite crystals are found as atypical mineral inclusions from this region. In the study of Epstein (1989) [4], the slightly rounded, translucent greyish white crystals which were determined as calcite and dolomite were also found in a few cases. Well-formed negative crystals together with two-phase inclusions are another particular feature of emeralds from Capoeirana (figure 9).

Figure 8. Group of quartz grains in Capoeirana emerald. x10.

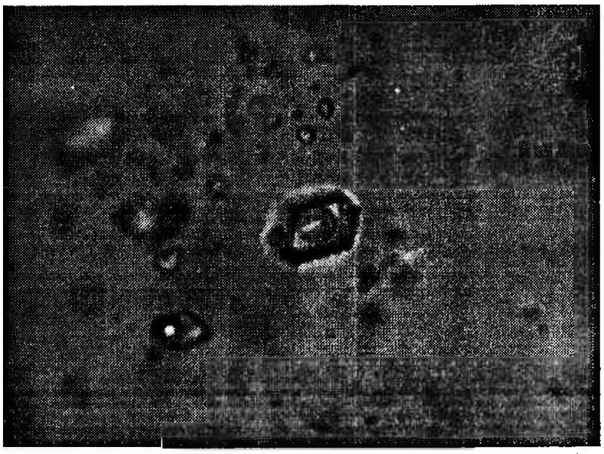

Figure 9. Liquid inclusions in negative crystals in emeralds from Capoeirana. x50.

3.4. Itabira (Brazil)

These emeralds are like those from Capoeirana and in general, are easily distinguished from those from Santa Terezinha and Socoto based on inclusion features. Under the microscope Itabira emeralds were found to contain relatively few types of inclusions. The most frequent observed mineral inclusions are quartz and micas. Other mineral inclusions, which were found much less frequent, are apatite, tremolite and hematite. According to Schwarz (1987) [5] andesine may also be found.

Liquid inclusions built the most abundant group in Itabira emeralds which exhibit a large variety of forms. This indicates a complex and multiphase formation history of emeralds in this region. Among liquid inclusions, two-phase type (liquid-gas) seems to occur more often. They are widely and irregularly distributed within host crystals, and can be observed in various-sized tubes or in almost rectangular bordered cavities as well as in more or less perfectly formed negative crystals. Three-phase inclusions were sometimes found, and differently, they are usually composed of two fluids and a bubble (figures 10, 11), in case of four-phase inclusions, liquid phases are still dominant: 2 liquids, a solid, and a bubble. The type of liquid inclusion containing solid substance is occasionally observed in Itabira emeralds. In general, liquid inclusions are so small that all attempts to determine their phases by confocal-Raman spectroscopy failed.

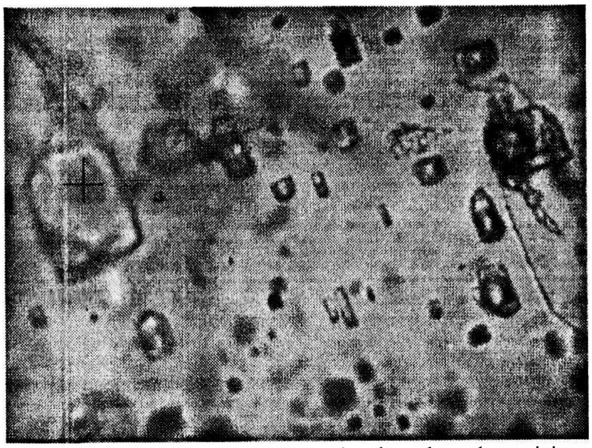

Figure 10. Almost rectangular bordered cavities filled with two or three phase inclusions in Itabira emeralds. x10.

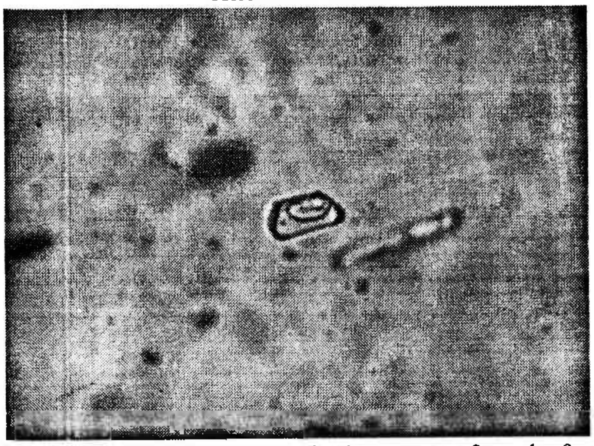

Figure 11. Multi-phase inclusions were found often containing two liquids and gas. x50.

Among mineral inclusions of emeralds, micas were dominant. They vary in a large diversity of forms and colours. Not only does the colour of mica inclusions vary from sample to sample but also within the same sample itself. The colour may be composed of various shades of brown, from yellowish, grey to dark brown. The mica flakes are usually strongly rounded or irregular. In some samples, micas are elongated or have a disc-shaped form and are (001)-oriented parallel to the basic face (001) of the emerald. The strongly rounded micas which show no preferred orientation are considered as protogenetic inclusion and often possess a deep brown colour. The thickness of mica slabs are also very different, some mica slabs are so thick that they appear almost opaque and show very clearly cleavage surfaces (figure 12). Another appearance of protogenetic mica is that the mica crystals are in a state of dissolution (figure 13). The syngenetic micas are mostly thin and transparent flakes which are either elongated or partly exhibit a distorted pseudo-hexagonal shape. These flakes show the following orientation within host crystal: the elongated crystals lie parallel to the c axis, and the pseudo-hexagonal crystal lie parallel to the basal plane. Micas have been determined by Raman spectroscopy as biotite and phlogopite.

Quartz is another type of frequently observable inclusion, but much less frequently than mica. Quartz crystals occur normally colourless, elongated or rounded and sometimes are found to be associated with liquid inclusions. Another type of inclusion in Itabira emeralds are various fissures which remained unhealed. This indicates that these fissures were formed when the crystal growth process had been stopped, thus the crystal had no longer contact with any liquid environment. In a few samples, tremolite, apatite and hematite are rarely found.

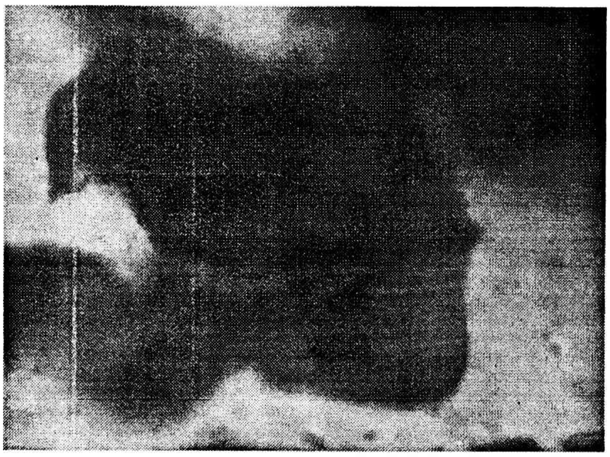

Figure 12. Thick brown mica flake with cleavage or growth surface. x50.

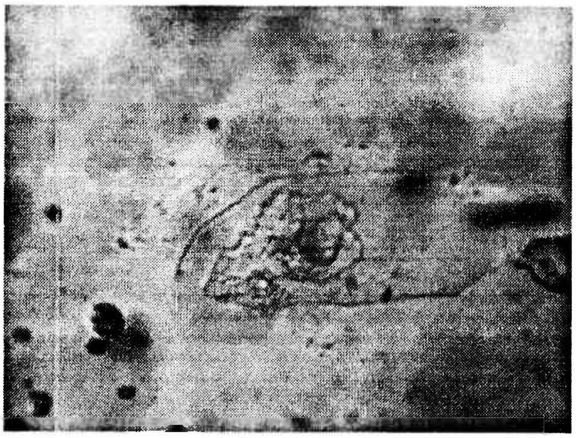

Figure 13. Mica flake with corrosion. x50.

3.5. Mananjary (Madagascar)

Abundant types of mineral inclusions are the most striking internal characteristic observation in Mananjary emeralds. Most of the inclusion features are similar to those found in emeralds from other schist-type deposits. Nevertheless, not all of Mananjary emerald inclusions can be found in any other locality. The association of numerous mineral inclusions such as quartz, mica (biotite, phlogopite), amphibole (actinolite, tremolite) and other minerals such as feldspar (albite, oligoclase), carbonate minerals (magnesite, calcite, dolomite), talc, molybdenite, tourmaline and fluid inclusions is a special feature of emerald from this occurrence.

Quartz inclusions are mentioned in almost all emeralds from different localities [6]. In Mananjary emeralds, quartz appears in a diversity of morphologies. Quartz inclusions can appear as transparent, colourless, elongated (or prismatic) parallel to the c-axis of the host emerald crystals, often associated with primary fluid inclusions (figure 14). They occur as isolated crystals, irregularly distributed throughout the host crystal or dispersed over the planes of healing fissures. The others are postgenetic inclusions that show irregularly rounded crystals, some of which have a badly corroded rough surface.

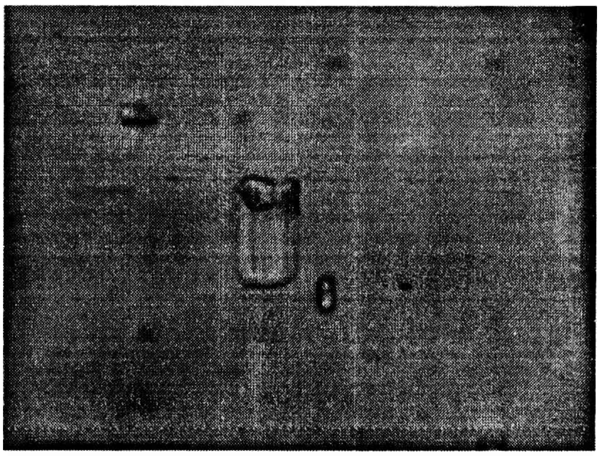

Figure 14. Quartz appears with fluid inclusions in prismatic form. x50.

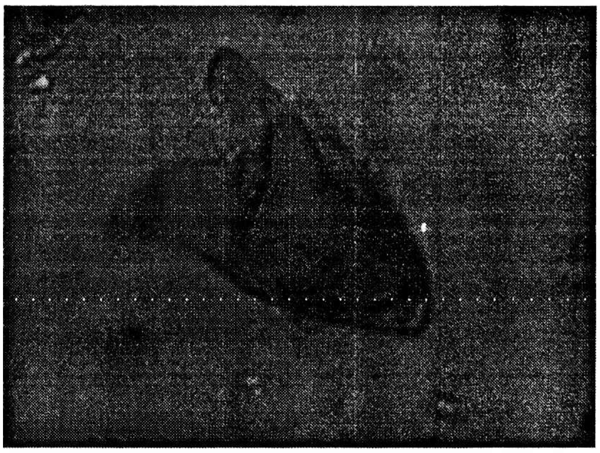

Figure 15. Thin plate of mica inclusion in Mananjary emerald. x50.

Quartz inclusions can also be observed in groups of small grains. They are well rounded or almost spherical. Nevertheless, the appearance of mica inclusions in Mananjary emeralds is even more frequent than that of quartz. They belong to the most common group of mineral inclusions. Micas were determined by Raman spectroscopy to be usually biotite and phlogopite. They often appear as thin plats with usually somewhat rounded edges, or may also have a almost perfect sharp outline (figure 15).

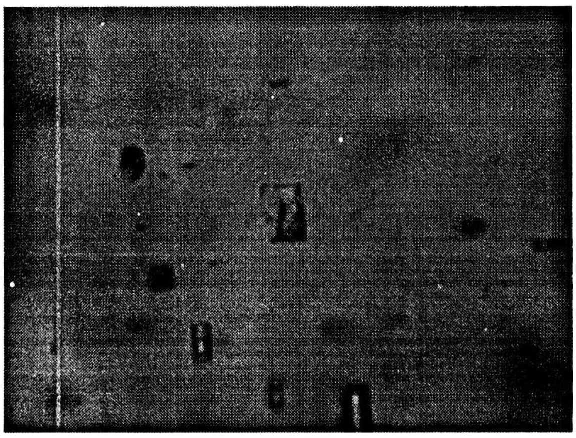

Figure 16a. Oligoclase ($KAlSi_3O_8$) found to be with two-phase inclusions. x50.

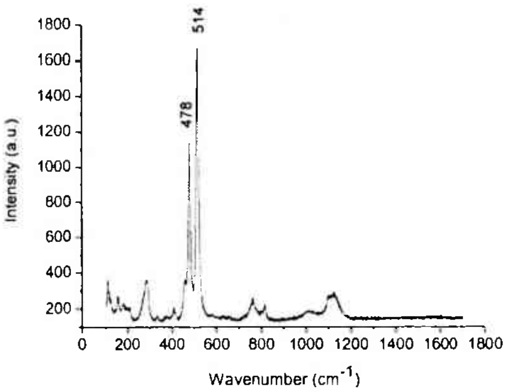

Figure 16b. Raman-spectrum obtained from oligoclase inclusion in Madagascar emerald.

Not only mica schists are host rocks of emeralds in Mananjary but also amphibolite schists (although less frequent). Consequently, amphiboles also belong to the main inclusions in emeralds; the observed amphiboles are actinolite and tremolite. They arrange as chaotic tubular crystals which always knit with others. But that distribution of amphibole is only in certain areas of the host crystals, not in the whole sample as those in emeralds from Habachtal.

In comparison with the above mentioned inclusions, carbonate minerals, feldspar and molybdenite are less frequent. Carbonate minerals are most irregular or rounded grains, although they show some relatively well-developed rhombohedral crystals. In general, they appear transparent and colourless. The surface corrosion made some carbonate crystals appear slightly brown. Feldspar is determined normally to be either albite or oligoclase. They are usually irregular in shape and often very rounded or corroded (figure 16a). Molybdenite occurs as grey or silver platelets with typical metallic luster. They may be slightly rounded or show a well developed hexagonal outline. Tourmaline has been reported in Mananjary emeralds by other studies ([7], [8]). However, according this study, tourmaline belong to rare inclusion caterogy of Mananjary emerald (figure 17a).

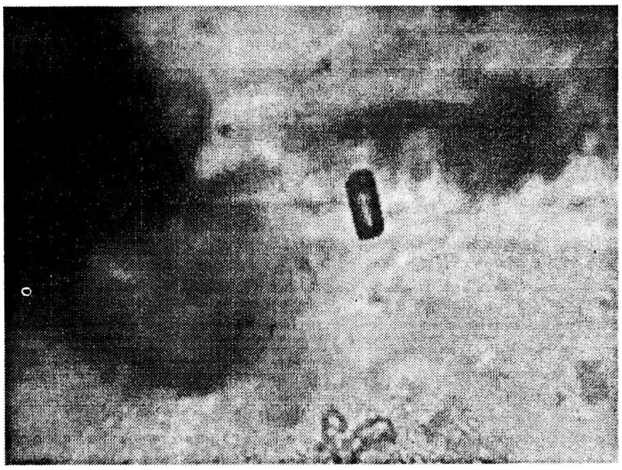

Figure 17a. Occasional case of tourmaline crystal found in emeralds from Mananjary. x50.

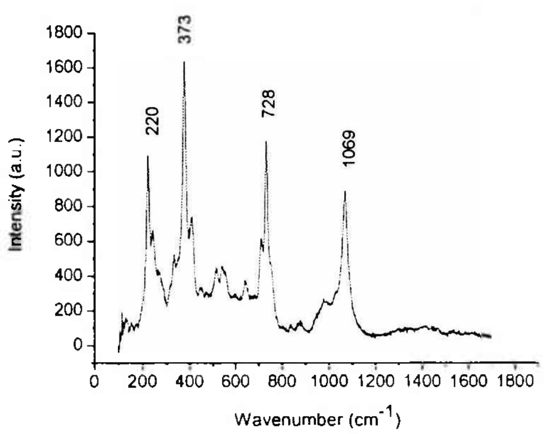

Figure 17b. Raman-spectrum obtained from tourmaline inclusion in Mananjary emeralds.

3.6. Transvaal (South Africa)

As emeralds hosted in biotite schist, Transvaal emeralds present the typical inclusions for this origin: fluids and mica (biotite, muscovite) as well as atypical ones such as calcite, quartz, talc. Notably, fluid inclusions in Transvaal emeralds were found fewer in comparison with mineral inclusions in one sample itself or in comparison with those in other localities of the same host rock (for instances, Itabira). These appear usually to be small, containing two phases, a liquid (water or liquid CO_2) and a CO_2 bubble. Sometimes, they can be seen in elongated cavities. Liquid inclusions in veil-like type or in tubular fissures were also seen. These make groups of irregularly shaped tiny inclusions which look like curved veils. This type of inclusions has also been observed in "emerald" crystals grown by the flux methods, which will be mentioned later in the part on synthetics.

Micas are considered as the most frequently observed inclusion, as been described in many localities; they appear with typical brown colour. Calcite and quartz show more or less well-formed crystals. Quartz crystals present the preferred orientation in host crystals parallel to the c axis.

Growth zoning is another feature of emeralds from Transvaal. They almost appear as fine-scaled parallel lines (figure 18). The colour between the lines is slightly varying. This indicates that the forming environment of Transvaal emeralds was not harshly changed. Hematite and lepidocrocite with the typical red colour were found commonly (figure 19).

Figure 18. Growth zoning in emeralds from Transvaal. x10.

Figure 19. Lepidocrocite, with light brown flakes of mica. x50.

3.7. Ural (Russia)

These emeralds were found to contain several types of liquid and two-phase inclusions. The most noteworthy were found in the form of flat cavities orientated on planes parallel to the pinacoid. In general, extremely thin channel-like structures running parallel to the c-axis, that is, perpendicular to the flat cavities, are confined to these planes. These cavities consist of liquid or two-phase (one liquid and one bubble) inclusions and they have less frequently three-phase inclusions. Channel-like growth tubes parallel to the c-axis and elongated fluid inclusions trapped on growth planes were observed. In addition, numerous, partly healed fractures were observed irregularly traversing through most of the crystals.

In a few cases, fluid inclusions in Uralian emeralds were found to be alike with those in emeralds from Itabira: three-phase inclusion containing 2 liquids and a gas bubble. The gaseous and liquid phases in these inclusions are CO_2. The solid phase in three-phase inclusions is halite, but the halite crystals found here are not in the cubic form as those in Chivor or Gwantu emeralds. The feature of clouds or trails caused by tiny particles found in Carnaiba emeralds were also found in Uralian emeralds (figure 20). As described, these particles are the tiny one or two-phase liquid inclusions that make the host crystals look partially translucent only.

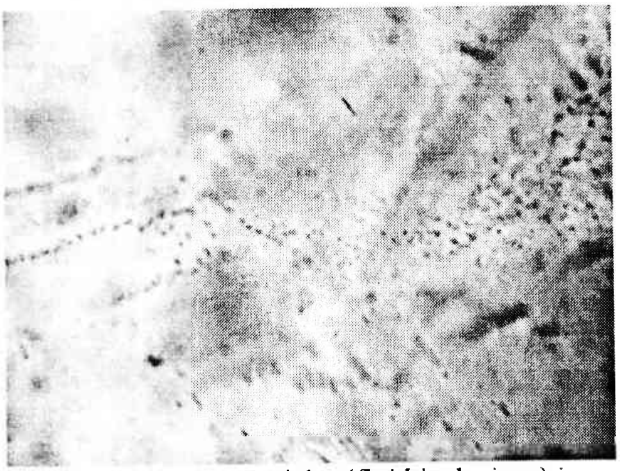

Figure 20. Tiny particles (fluid inclusions) in Uralian emeralds resembling those in emeralds from Capoeirana. x50.

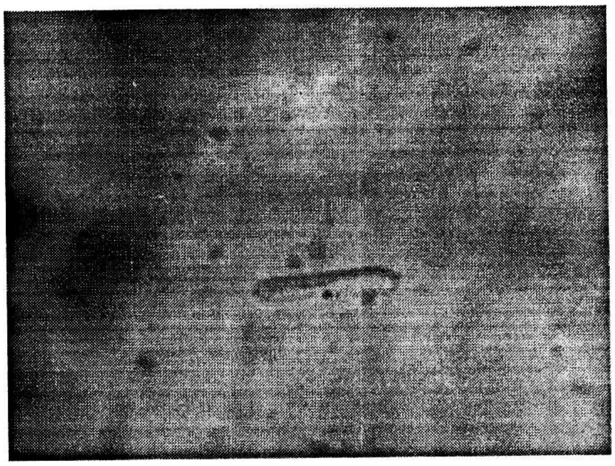

Figure 21. Elongated mica inclusion in Uralian emerald. x50.

Mineral inclusions were found only rarely in Uralian emeralds. The most frequently observed minerals are several forms of mica which were determined by confocal-Raman spectroscopy as phlogopite (and muscovite with the smaller portion) which are known to originate from mica schist host rocks. In general, phlogopites are virtually colourless, only a few of those show the brown hue. Normally, they appear as the more or less corroded flakes which are considered to be the typical form of mica. But sometimes they can appear as small rounded grains which at the first glance look like quartz grains. In other cases, the elongated form makes them to be confused with amphibole (if remembering that amphibole schist is one of the host rocks of these emeralds) (figure 21). Nevertheless, true grains of quartz and actinolite also occur. Actinolite crystals are found usually to occur individually. Due to the much less frequent appearance of mineral inclusions in comparison with liquid ones and the features of liquid inclusions, Uralian emeralds present themselves to be most alike with emeralds from Carnaiba.

4. Conclusion

Based on the study, it leads to conclude that the inclusion suit of quartz, mica, amphibole, fluid inclusions are normally abundant and considered as the typical inclusions for emerald of all of study localities. In many cases, with the keen eyes of experiences, inclusion observation could help not only distinguish mineralization conditions between schist type and non-schist type emeralds but also identify geographical origins of particular sample. The feature of abundant multi-phase inclusions containing two liquids, developed in well-formed negative crystals; more or less square, rectangular cavities is characteristic for Itabira beryls. Beside the typical mineral suit, there are the individual minerals that exist only in certain localities and could be a great value to limit the range of beryl location, for instance, pyrite is found only in Santa Terezinha (Brazil).

Acknowledgments

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