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On corundums from Umba Valley, Tanzania

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Abstract

The many colours exhibited by corundums from Umba Valley in Tanzania are related to their varying contents of Fe, Ti, Cr and V. Ni and Co could not be detected by the EDS-XRF technique used. The presence of abundant inclusions distinquishes these gemstones, and the following were found: zircon, rutile(?), apatite, hematite, mica, monazite, plagioclase, calcite, pyrrhotine and graphite. Healed fractures, negative crystals, colour zoning, twinning with traces of intersecting lamellae and boehmite(?) laths were noted. Techniques for the identification of inclusions are discussed. The densities (3.98-4.01 g/cm^3) and refractive indices (n_e: 1.761-1.768, n_o: 1.769-1.778) were also determined. Spectral features are given.

Introduction

Corundum is economically the most significant of the many unusal gemstones of East Africa. Numerous occurrences of ruby in Kenya and Tanzania (Longido, Lossogonoi, Mangari and Morogoro) have supplied the gem-cutting industry with raw material for a long time, but corundum from the Umba Valley is unique for the large range of colours exhibited by the mineral.

The aim of this study is to describe the properties and characteristics of the corundum from Umba Valley, and to supplement and confirm data and information from earlier publications.

Rubies from alluvial deposits were discovered in the vicinity of the River Umba in north-eastern Tanzania in 1960 (Solesbury, 1967). The host rock of the corundum was found not far from this location in a pegmatite which intrudes a greyish-green serpentinite. The pegmatite varies mineralogically, but where it hosts corundum it contains vermiculite and a calcic plagioclase. Gneisses (in part graphitic), amphibolites and marbles are geologically associated with these rocks. The geology of the area was described by Solesbury (1967). The erosive action of the River Umba resulted in the exposure of these rocks and the formation of alluvial deposits. Many corundum crystals were thus extracted from the host rock and accumulated in pockets and depressions.

Habit and colour of the crystals

The crystals or crystal fragments usually occur in tabular, hexagonal forms without pyramid faces. Generally, the basal faces seem to be corroded and, as with the prism faces, perforated with 'packets' of vermiculite. Intersecting furrows occur on the basal faces, often forming regular triangles. These originate where the traces of the rhombohedral cut the basal faces. The furrows are also exit positions of the fine twin lamellae which also lie parallel to the rhombohedral faces. In many ways, the corundum from Umba resembles that from the Missouri River in Montana (USA), especially as far as colour, habit and inclusion patterns are concerned. However the Umba corundum exhibits colours which are much more varied; all conceivable colour combinations of blue, red, yellow and colourless occur (Pough, 1972; Naftule, 1982). However, pure (spectral) colours are virtually never encountered and variations of bluish-green, bluish-grey, violet, pink, yellowish-green, brownish-yellow, orange or brown predominate. The best colour is exhibited in the direction of the *c*-axis, often paler in the centre of the crystal and more strongly coloured towards the crystal rims. Colour zoning in two different colours within the same crystal sometimes occurs.

Two colour varieties of Umba corundum have attracted particular attention. Firstly, the bright orange to yellowish-brown stones and secondly, the colour-changing alexandrite-like types (Naftule, 1982). At this stage, it should be pointed out that with respect to colour, the superb orange corundum from Umba Valley differs chemically from the padparadschas of Sri Lanka as far as the causes of the colour are concerned. The trace elements chromium (Cr) and iron (Fe) are mainly responsible for the colour of the orange Umba corundum, whereby Cr^{3+} is the cause of the red component, and Fe^{3+} of the yellow. The padparadschas of Sri Lanka owe their colour to a combination of chromium and lattice defects (the latter acting as colour centres), as well as to trace quantities of iron. In the latter case the chromium is the cause of the

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red colour component, but the yellow component of the orange stems mainly from the colour centres (Schmetzer *et al.*, 1982). As the two varieties of corundum are dissimilar both in appearance and as to the origin of their colour, there is no cause to designate the orange corundum from Umba as padparadscha (Gunawardene, 1984).

Many authors have studied the causes of colour in corundum (Harder, 1969, Lehmann *et al.*, 1970, Schmetzer *et al.*, 1981). The following data from corundums from Umba originates from the work of Harder (1969), who investigated a large number of differently coloured corundums from all over the world, using wavelength-dispersive X-ray fluorescence techniques (Table 1).

Table 1. Colouring effect of trace elements in corundum from Umba Valley. (after Harder, 1969)

	Cr	V	Fe	Ti
Red	0.03	–	0.05	0.006
Violet	0.04	0.002	0.1	0.009
Bluish	0.014	–	0.6	0.006
Yellowish-green	0.007	–	0.5	0.006
Greenish	0.002	–	0.4	0.009

(Values in weight %)

As can be seen in Table 1, the four main trace elements in Umba corundum occur in concentrations covering a wide range. Lattice position and oxidation state of Fe as well as its association with other trace elements play an important role in colour considerations. Basically, the following rules apply (after Schmetzer and Bank, 1981): *Yellow* corundum is either coloured by the presence of trivalent iron (Fe^{3+}) or by defects in the crystal lattice (colour centres). The iron in *blue* sapphire is mainly divalent and together with quadrivalent titanium it forms the blue colour of the mineral as (Fe^{2+}/Ti^{4+}) pairs. *Green* corundum probably possesses its colour through mixing of yellow and blue, the latter two colours resulting from the reasons given above. *Red* corundum (ruby) contains trivalent chromium (Cr^{3+}) and often also vanadium (V^{3+}) and iron. *Violet* stones can be expected to contain a mixture of Cr with Fe and Ti, as violet may be considered to be a mixture of red and blue. Vanadium plays a major role in the colour-changing alexandrite-type corundum (Hänni, 1983). On the other hand, the colour effect of such natural stones is often masked by further trace elements resulting in the predominance of other colours. Particularly in the case of Umba Valley corundum, the combined effect of different colour-giving trace elements can be observed. Depending on which combination of elements are present and their relative proportions, a rich palette of colour results.

Twelve corundum samples from Umba exhibiting a wide range of colours, were analysed for the four chromophore elements Cr, V, Ti, Fe by W. B. Stern (Basel). Using an energy-dispersive X-ray fluorescence unit (Stern and Hänni, 1982), the results of Harder (1969) were confirmed qualitatively. Measurable quantities of nickel or cobalt were not detected. Signals from Zr, Pb, Cu, Zn were registered in specimens containing numerous inclusions. There, Zr and Pb is explained by zircon crystals which in many cases reach the surface of the faceted stones. The Cu and Zn signals are most probably due to metal remainders stemming from the cutting wheels. The metal particles are fixed in fractures and open pores in the sample.

Physical Properties

The specific gravity of corundum varies only within a small range; any values outside this range are probably due to the presence of lighter or heavier inclusions. Table 2 shows a compilation of values of specific gravity and refractive indices also exhibit only small variations. The maximum values attained are extremes for corundum, and were measured on an orange-coloured specimen (see also Bank, 1972).

The absorption spectra reflect the many colour varieties and are combinations of the three basic spectra of ruby, sapphire and yellow corundum. Even very small amounts of Cr result in the fluorescence doublet at 693nm. On the other hand, the presence of vanadium or titanium when combined with other chromophores is hard to detect from the spectrum (Bossart, 1981). The Fe^{3+} in blue and bluish-green stones results in a strong band at 450nm. This can be so broad and strong in orange stones, that it merges with the absorption shoulder in the violet. This means that under the spectroscope, darkness occurs with such specimens after violet, below 470nm.

Four typical colour varieties of corundum from Umba exhibit the following absorption spectra (recorded using a Pye-Unicam SP8-100 spectrophotometer):

Orange: 700, 693, 688, 674, 657, 554 (wide, **470** general absorption.

Bluish-green: (693), 560 (wide;, 466, **450,** 388, 376, 360 general absorption.

Olive: (693), 650, 468, 456, **450,** 388, 376, 360 general absorption.

Light violet: **693,** 657, (560 wide), (466), 450, 388, 373, 320 general absorption.

Weak lines are in brackets, strong lines are in bold-faced type, and the last values represent the beginning of general absorption. Values are in nanometres (nm).

Table 2. Physical data of the Umba Valley corundums.

	This study	Zwaan (1974)	Gunawardene (1984)
Density (g/cm^3)	3.98_0-4.01_0	3.97_5-3.99_3	3.99-4.06
Refractive indices			
n_e	1.761-1.769	1.760-1.765	1.763-1.765
n_o	1.768-1.778	1.768-1.774	1.771-1.773

Mineral inclusions and their determination

Of the gemstones which occur in the Umba Valley, garnets have undergone the most in-depth study of their inclusions. Investigations by Schubnel (1972), Zwaan (1974) and Gübelin (1981) have shown that the following inclusions exist in the pyrope-almandine-spessartine-grossular mixed crystals of the garnet group:– apatite, monazite, zircon, rutile, pyrite, pyrrhotine, quartz.

With the exception of quartz, all the above minerals as well as some others can be found in corundum. The following is a list of minerals determined by Zwaan (1974), identified mainly by using X-ray powder techniques: apatite, graphite, pyrrhotine, rutile, spinel, vermiculite, zircon.

Observations of the occurence of a number of these minerals were made previously by Eppler (1973).

Unfortunately, results in publications seldom mention how mineral inclusions were identified. Nowadays, numerous techniques are available, and the commonest are briefly described below.

X-ray powder technique

A small amount of powdered sample material is usually used for this relatively simple investigation. After removal of included grains exposed on a surface or scraping off of powder using a diamond file, the characteristic diffraction pattern of the sample is compared with those of known phases.

Electron beam techniques

The study of tiny areas such as inclusions lying on the surface of a gemstone can be undertaken by means of two similar instruments: the scanning electron microscope (SEM) and the electron microprobe (EMP). Utilizing a fine beam of electrons, the atoms of the area studied are excited and generate characteristic X-rays. They are typical in terms of energy or wavelength for each element present in the sample. The emitted (fluorescence) radiation may be sorted using an energy-dispersive system (EDS) attached to the SEM or EMP. The resulting energy spectra enable one to read the chemical composition at least qualitatively (Figure 10). The SEM has the advantage that it can magnify strongly the analysed area and produce pictures of the surface. The EMP on the other hand is used primarily for fully quantitative chemical analysis rather than for imaging. Both techniques work non-destructively but possess the inherent disadvantage that the lightest elements cannot be analysed.

Raman-laser probe

Using this equipment, solid, liquid ot gaseous inclusions within gemstones can be analysed. A monochromatic (wavelength, e.g., 488nm) laser beam is focused on an inclusion. The laser beam undergoes a frequency change characteristic of the material excited, through interaction with oscillating molecules. The spectra recorded in the infrared region are compared with reference spectra for known solid, liquid and gaseous phases.

Of the three methods discussed above, Raman spectroscopy is still a novel technique in the field of gemmology (Delé *et al.*, 1985).

Optical techniques

Inclusions are often solely identified by their appearance under the microscope (form, colour, relief, etc.) and compared with known phases, especially when other techniques cannot be applied through lack of equipment. On the other hand, identifications purely on the basis of optical comparison are relatively unreliable. Information on inclusions would be more valuable if the technique used for the determination was described.

There are, however, certain types of inclusions, although widely distributed and well-known, whose determination poses great problems: for example, the identification of fine rutile needles in corundum. It can be difficult to decide in certain cases whether, we are dealing with rutile needles, or with hollow cavities (the titanium detected can either occur in the corundum lattice, or as segregated rutile).

In corundum from Umba Valley, ruby from Thailand and Kenya, sapphire from Missouri River deposit (Montana, USA) and corundums from other sources, characteristic inclusions resembling scaffolding are found. These have not been definitely identified due to differing results obtained by various investigators. These linear elements which cross through the corundum in sets parallel to the rhombohedral edges have been described as:

Fig. 1. The most common inclusions in corundum from Umba Valley are zircons. 25x.

Fig. 2. Short and long needles of rutile, partly lanciform as twinned forms. Reflected light showing interference colours. 25x.

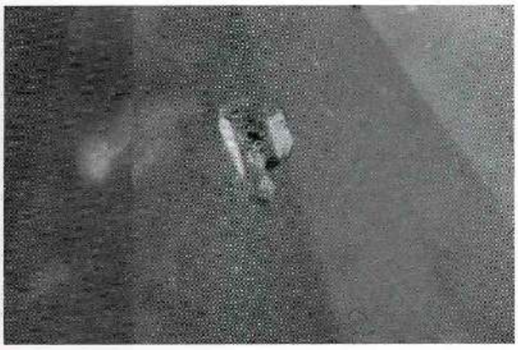

Fig. 3. Idiomorphic crystals of apatite with unidentified dark inclusions. 50x.

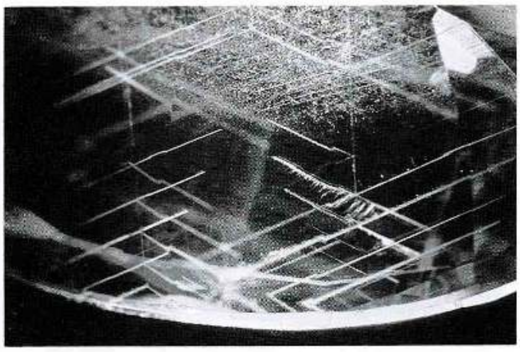

Fig. 4. Twin lamellae intersections and boehmite laths respectively, with fine fissures. Below: fine rutile segregations. 10x.

(a) needles of corundum (Eppler, 1973).
(b) traces of intersecting twin- or dislocation lamellae (Gübelin, 1974; Schubnel, 1972).
(c) boehmite (Keller *et al.*, 1985).

Regardless of the importance of these inclusions (sometimes wrongly taken for rutile) general discussions on their nature have as yet not taken place. The extensive treatment in Schmetzer (1985) will lead hopefully to a generally accepted and used term for this type of inclusion.

The different inclusions found in Umba corundums, as well as structural phenomena exhibited, are described in order of abundance below. The abbreviations following the mineral names denote the method of identification used for this study:
EMP Electron microprobe technique.
OM Optical microscopy.
SEM Scanning electron microscopy.
XRD X-ray diffraction (powder techniques).

Identified inclusions

Zircon (SEM) Figure 1. Small to very small crystals of slightly rounded tetragonal prisms, partly intergrown in groups. They exhibit no preferred orientation in corundum, but are often surrounded by stress fissures. A zoned structure is often observed. The ratio of length: width varies from 2:1 to 30:1. The colourless zircon crystals have a positive relief relative to the surrounding corundum.

Rutile (OM) Figure 2. Very fine, oriented needles, often occurring in three intersecting systems. Mainly very loose and short. Due to the small amounts present and the minute exit positions to the corundum surface, rutile could not be confirmed either chemically or by X-ray techniques.

Apatite (SEM) Figure 3. Individual large to small crystals which occur frequently in reddish corundum and themselves often contain minute dark brown unidentified inclusions. Their hexagonal and prismatic forms with multi-faced to rounded terminations are usually readily recognizable. The mineral grains are colourless and possess a negative relief relative to corundum. Apatite is much rarer than zircon in the blue and green corundum varieties.

Boehmite-laths (OM) Figure 4. Fine to substantial lathlike to track-like structures run through the

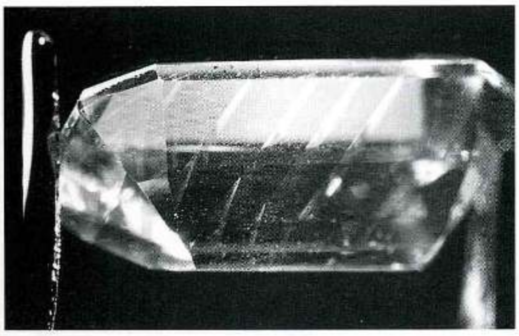

Fig. 5. Polysynthetic twinning, wide and narrow bands alternating. 10x.

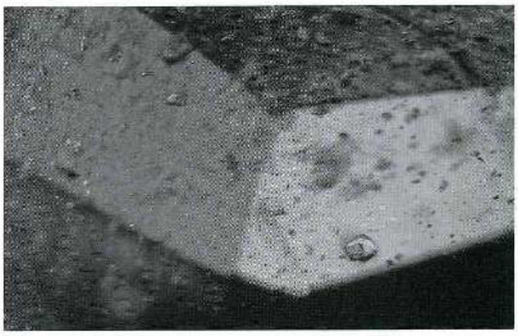

Fig. 6. Tabular greenish-yellow crystal of monazite, together with numerous small grains of zircon. 20x.

Fig. 7. Needles and thin platelets of hematite coloured brown in transmitted light. The regular pattern corresponds to the arrangement of the rutile-silk. 35x.

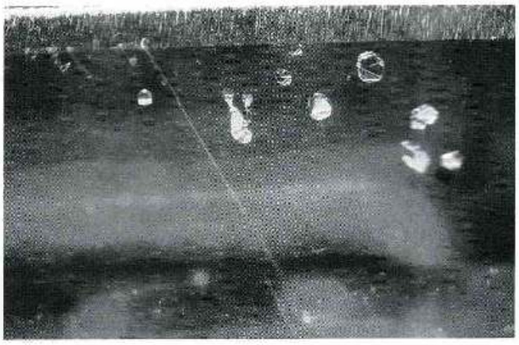

Fig. 8. Flat negative crystals, parallel to the basal face of corundum, in part exhibiting trigonal symmetry. 30x.

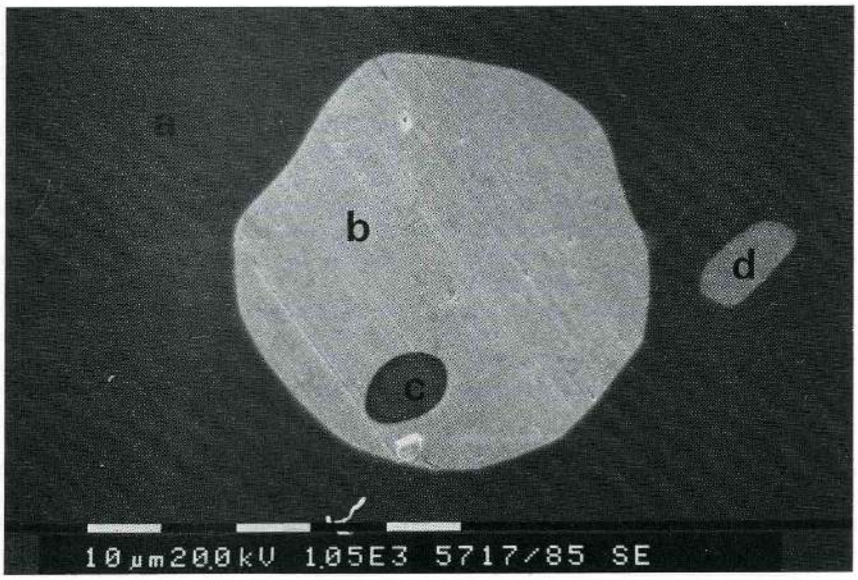

Fig. 9. SEM micrograph of a surface of corundum, showing intersected inclusions. The latter were identified on the basis of their EDS spectra: *a:* corundum, *b:* monazite, *c:* plagioclase, *d:* zircon.

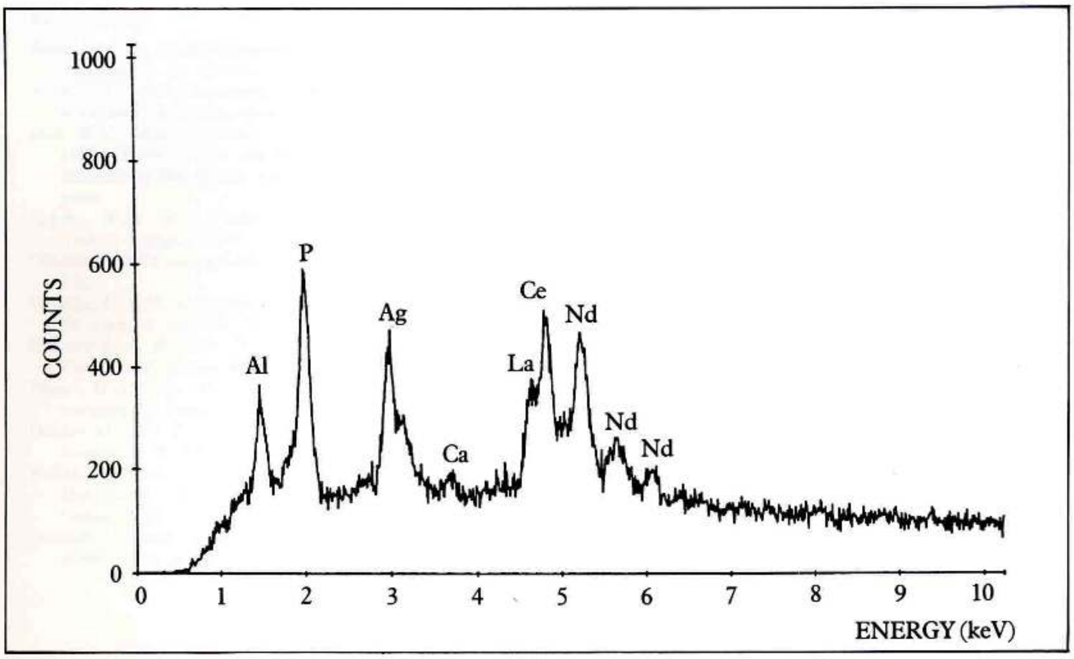

Fig. 10. EDS spectrum of the monazite grain in Figure 9. The Ag line is due to the necessary coating of the sample with a conductive material. The fact that monazite often contains rare earth elements is supported by this EDS spectrum.

corundums parallel to the rhombohedral edges. They occur mostly in three spatial directions and intersect each other at nearly right angles. Often very fine fissures emanate from these lines. The boehmite laths run within the traces of systems of intersecting twin lamellae. The impression arises that the laths are either hollow or filled with polycrystalline material (Delé, pers. com., 1984). As mentioned above, different names have been applied to this type of inclusion.

Twinning (OM) Figure 5. Polysynthetic twinning occurs very commonly in all the colour varieties, mainly in the form of alternating, very fine and broader lamellae. Two other common growth features often encountered are colour zoning, and zoning whereby differing amounts of mineral inclusions occur in definite zones.

Monazite (SEM) Figures 6 and 9. Yellowish-green tabular crystals, containing plagioclase inclusions (SEM). Monazite ($CePO_4$) usually contains different elements of the rare earth group, in this case lanthanum and neodymium (Figure 10).

Hematite (EMP) Figure 7. Very thin, parallel oriented transparent brown platelets to lanciform habits. These are very similar to the hematite inclusions in the brown star corundum of Ban Kha Cha, Thailand (Weibel and Wessicken, 1981). These hematite platelets are the main cause of colour in most of the brown corundums from Umba.

Negative crystals (OM) Figure 8. Flat, crystallographically shaped cavities (?), arranged parallel to each other, have been found, which exhibit interference colours when viewed with oblique illumination. They could also be regarded as liquid films.

Mica (XRD) Colourless and also greenish-brown scales or flakes, presumably vermiculite from the host rock, occur mainly in the reddish varieties.

Calcite (XRD) Whitish rounded crystals, identified in blue corundum.

Pyrrhotine (XRD) Dark brown to golden lustrous round or stepped crystals, identified in reddish corundum.

Graphite (XRD) Very shiny, opaque, black platelets or flakes, flexible, identified in reddish-violet corundum.

Mineral dust (OM) Minute unidentified dots in zones or as 'comets', often clouding the whole stone.

Healed fractures (OM) More or less well-healed fissures containing liquid droplets.

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