THE ROLE OF CARBONATITE DEPOSITS IN THE INDUSTRIAL MINERALS INDUSTRY: MINERALOGICAL ASPECTS OF SELECTED ECONOMICALLY IMPORTANT CARBONATITE-RELATED DEPOSITS

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ABSTRACT

Carbonatites are unique rocks that consist largely of magmatic calcite but also may contain a wide variety of other minerals, many of which are industrial minerals of economic value. They serve, for example, as important host rocks for fluorspar, rare earth elements (REEs), apatite, and vermiculite, as well as a source for calcite. Selected deposits of these industrial minerals hosted by or connected with carbonatites are reviewed here. Emphasis is placed upon the microscopic examination of the deposits.

Fluorspar deposits are especially characteristic of alkaline igneous-carbonatite complexes. Okorusu in north-central Namibia is the world's largest carbonatite-related fluorspar producing mine. Historical mine sites include Amba Dongar, India; Mato Preto, Brazil and several mines in Russia. Sites not previously considered to be hosted by carbonatites but with the mineralogical characteristics of carbonatite-related fluorspar ores include the Kimwarer fluorspar mine in the East African Rift Zone in Kenya and the Hick's Dome deep fluorspar ores in Southern Illinois, USA. Those deposits are characterised by abundant apatite, the local presence of REE's and nearby carbonatite intrusions. The connection of fluorspar with carbonatites is also shown by the 5% fluorspar content in the groundmass of carbonatite lavas of the active Oldoinyo Lengai carbonatite volcano in northern Tanzania. REE deposits are also characteristic of alkaline igneous-carbonatite complexes. Although the Bayan Obo mine in Inner Mongolia, China is primarily a magnetite mine, by-product REE's make it the world's largest REE producer. The Mountain Pass, California, USA carbonatite-hosted REE deposit, formerly the world's largest REE producer, has recently resumed production and the Eldor carbonatite-related REE deposit is currently being explored in the Labrador Trough of Quebec, Canada. Apatite is mined from the Jacupiranga carbonatite bedrock at the Cajati mine in southern Brazil, with calcite recovered as a by-product. Although the Palabora carbonatite of northeast South Africa is best known for its copper production, vermiculite is recovered from weathered phlogopite crystals in pyroxenite intrusions in that igneous complex.

INTRODUCTION

Carbonatites are unusual rocks that consist mostly of calcite (or dolomite) and therefore are mineralogically like limestones. However, they have been conclusively shown to have formed by igneous processes. At Oldoinyo Langai in Tanzania, for example, carbonatite lava is currently extruding from an active volcano. Carbonatites also contain commercially valuable amounts of other minerals many of which are industrial minerals.

This paper reviews some of the mineralogical aspects of ten world-class carbonatite industrial mineral deposits. Some of these deposits have been intensively studied by the author and his former students. Other deposits have been studied using microscopy, by the author.

Brief descriptions of the geology are presented for each of the deposits. An emphasis has been placed upon selected aspects of the mineralogy and on aspects of the economic minerals observed under the microscope. Transmitted and reflected light microscopy, Scanning Electron Microscope (SEM), Electron Probe Micro-Analysis (EPMA) and cathodoluminescence microscopy techniques have been employed in the study of these deposits.

FLUORSPAR DESPOSITS

Okorusu carbonatite-related fluorspar, Namibia

The Okorusu carbonatite-related fluorspar is located in north-central Namibia about two-thirds of the way between Windhoek and Tsumeb and about 20km west of the highway that links those two cities. It is the world's largest carbonatite-related fluorspar mine and it supplies about 50% of Solvay's (Belgian chemical company) fluorspar needs. Fluorspar production at Okorusu peaked at 132,000t (tonnes) of concentrate in 2006; with production ranging between 80,000t and 100,000t concentrate in the past three years (2011-2014), depending upon the vitality of the general economy.

The Okorusu alkaline igneous carbonatite complex is a vertical intrusion from the upper mantle. It is circular in plan view, and about 8 km in diameter (Van Zijl, 1962). It was intruded during early Cretaceous time (125 Ma) into late Precambrian Damara Series regional metasediments. In the vicinity of the complex and especially along its southern margin the Precambrian rocks have been highly metasomatised to green, dense, fine-grained, aegirine-rich sodic fenites (Figure 1). The fenitisation was the result of early sodic and potassic fluids introduced into the complex. The resultant fenites were brecciated prior to the introduction of the carbonatites. Silicate igneous rocks include nepheline syenite, foyaite, tinguaite, and monzonite.

Fluorspar-depositing fluids replaced easily dissolved calcite in the carbonatite intrusions, and left relatively insoluble titaniferous magnetite at the margins of those intrusions (Figure 2), and converted iron-rich diopside pyroxene (Shivdasan-Gebhardt and Hagni, 2008) and pyrrhotite crystals within the carbonatite intrusions into goethite pseudomorphs (Hagni, in press). Apatite crystals in the carbonatites are only partially replaced by the fluorspars-depositing fluids (Figure 3). Cathodoluminescence microscopy provides an especially useful tool to study the mineralogy of the carbonatites, fluorspar ores, and fluorspar mill products (Hagni and Shivdavsan, 2001; Hagni, 2006). Partially replaced apatite crystals form binary locked particles in the fluorspar concentrates that cause an important metallurgical problem for carbonate-related fluorspar concentrates. Phosphorus is a detrimental constituent in steelmaking and carbonatite-related fluorspar concentrates therefore usually cannot be used as a steelmaking flux. At Okorusu the carbonatite-replacement fluorspar ores average 2.5% P_2O_5, and flotation reduces that content in the final concentrates to 0.3-0.5% P_2O_5.

In addition to the characterisation of carbonatite-replacement fluorspar ores by their high phosphorus contents, they typically also are characterised by elevated REE contents. At Okorusu REE-bearing minerals are present as synchysite [Ca (Ce,La,Y,Nd,Gd)CO$_3$F] crystals (Figure 4) that were locally concentrated in the carbonatites by an early hydrothermal event prior to the introduction of the fluorspar-depositing fluids (Hagni, in press).

Where the fluorspar-depositing fluids locally encountered Precambrian marbles they produced fine-grained marble-replacement fluorspar ores (Hagni in press). Some of the fluorspar ores at Okorusu have formed by the replacement of marble. Those ores are

Figure 1. *Field photograph showing partial fenitisation of quartzite. Ameboid remnants of unaltered quartzite (white) are enclosed in metasomatic fenite (green). Area outside the Okorusu fluorspar mine, Namibia. Geological hammer for scale.*

Figure 2. *Open pit mine photograph showing fluorspar replacement of carbonatite (white; lower right) that previously had been intruded into fenite (green; upper left). Titaniferous magnetite (black) crystallized at the rim of the carbonatite was unaffected. Okorusu fluorspar mine, Namibia. Geological hammer for scale.*
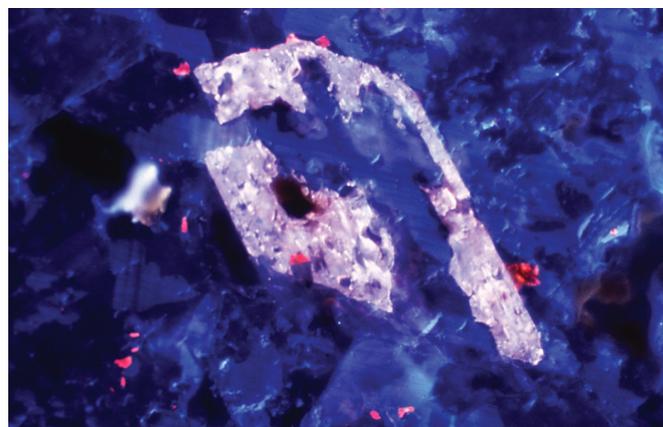

Figure 3. *Cathodoluminescence micrograph showing a partially replaced apatite crystal in fluorspar ore. Carbonatite apatite (violet to white) remains as the replacement remnant along with small patches of carbonatite calcite (orange), most of which have been replaced by fluorspar (blue). Okorusu fluorspar mine, Namibia. Length of the partially replaced apatite crystal is 100 μm.*

not only characterised by a finer grain size than the carbonatite-replacement fluorspar ores, they also are characterised by their negligible phosphorus contents. It has been calculated that 84% of the fluorspar ores at Okorusu have formed by the replacement of carbonatite, 15% by the replacement of marble, and 1% by the filling and replacement of fenite breccias (Hagni, in press).
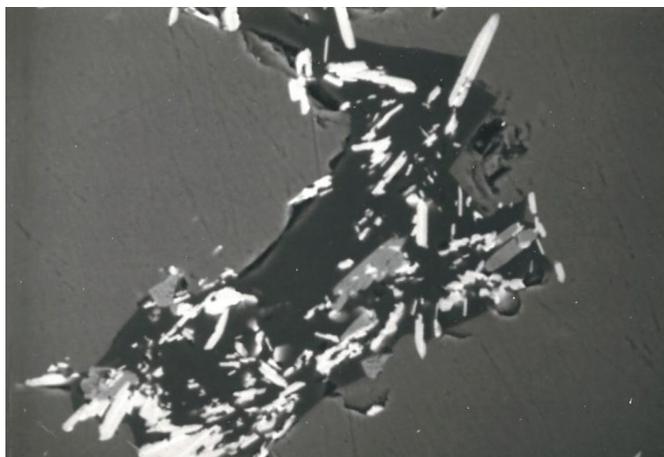

Figure 4. Scanning electron microscope back-scatter micrographic image (SEM-BSI) showing synchysite [Ca(Ce,La,Y,Nd,Gd)CO₃F] crystals (white) in fluorspar ore (grey) and quartz (black). Smaller amounts of apatite (light grey) also are contained in the quartz. Okorusu fluorspar mine, Namibia. The larger synchysite crystals are about 20 μm long

Amba Dongar carbonatite-hosted fluorspar mine, India

The Amba Dongar carbonatite-hosted fluorspar mine (Figure 5) in west-central India (Gujarat State) is very similar to Okorusu (Hagni and Viladkar, 2004), and the two deposits could be considered to be sister deposits geologically. At Amba Dongar all of the fluorspar ores replace carbonatite. The carbonatites occur in two ring dikes, and the inner ring dike is carbonate breccia (Viladkar, 1996). The colour of the fluorspar at Amba Dongar is largely yellow, whereas at Okorusu fluorspar is mostly purple, some is green, and only late fluorspar is yellow. The fluorspar ores are characterised by elevated amounts of phosphorus and relatively low amounts of REEs (Palmer and Williams-Jones, 1996). Production has always been small, usually about 20,000t of concentrate annually, but the mine has recently been closed (Viladkar, S. G., personal Communication, February 2014).
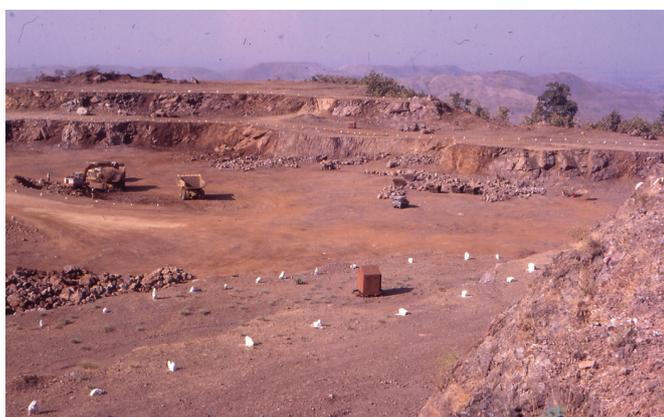
Figure 5. Photograph showing the open pit at the Amba Dongar fluorspar mine, India. Fluorspar ores (purple) have replaced portions of the carbonatite (orange). Mine equipment for scale.

Mato Preto carbonatite-related fluorspar mine, Brazil

The Mato Preto carbonatite-related fluorspar mine (Figure 6) in Southern Brazil was a larger producer of fluorspar concentrate than Okorusu through to 1998 but was closed in 1999 and the property was converted into a park. The fluorspar ores were very fine-grained and challenging to detect in hand specimen. The ores were believed by the mine geologists to have replaced tinguaite dikes but carbonatite was also very closely associated with the ore deposits. Like other carbonatite-related fluorspar ores, the Mato Preto fluorspar ores contained elevated amounts of phosphorus and REE's (Hagni, 1999).

Figure 6. Photograph showing the open pit at the Mato Preto fluorspar mine, Brazil. The lighter areas are carbonatite and the darker areas are tinguaite dikes. The fluorspar mineralisation has been interpreted to have replaced tinguaite and cannot be distinguished at this scale. Mine benches are about 13m high.

Kimwarer fluorspar mine, East African Rift, Kenya

The Kimwarer fluorspar mine (Figure 7) operated by the Kenya Fluorspar Company in the East African Rift Zone in southern Kenya has not previously been thought to be carbonatite related. However, the fluorspar concentrates are characterised by elevated amounts of both phosphorus and REE's and its position in the East African Rift suggests that it may well be carbonatite-related. The fluorspar ores were discovered in 1967. Production in 2013 was at 48,000t of fluorspar concentrate (USGS, 2014), but concentrate production has been as high as 130,000t per year. Fluorspar ores are mined from seven separate open pits in the area and blended.

Figure 7. Photograph showing one of the open pits at the Kimwarer fluorspar mine in Kenya. Ore is extracted from seven separate open pits and is blended. Surficial oxidation has produced fine-grained goethite that colors the ore yellow.

Deep fluorspar ores, Hicks Dome, Southern Illinois, USA

The Hicks Dome deep fluorspar ores in Southern Illinois also have not previously been considered carbonatite-related fluorspar ores, but their high apatite contents, elevated REE contents, and nearby presence of carbonatite all suggest strongly that the ores are carbonatite-related (Hagni, 2015). Carbonatites are exposed at the surface and are known from drilling programmes to be present at depth. Information about the deep fluorspar ores is largely confidential and little has been published. It is uncertain whether the deep fluorspar ores will be mined because the depth at which they occur is 600m (2,000 feet) or more.

The shallower fluorspar ores that have been mined in the Southern Illinois Fluorspar District, (that was the largest fluorspar-producing district in the United States for many years) contain sphalerite and galena making it similar to a Mississippi Valley Type (MVT) deposit but the district differs from most MVT districts in its content of very abundant fluorspar. By contrast, other MVT deposits such as the Tri-State Zinc District of southwest Missouri, southeast Kansas and northeast Oklahoma and the Viburnum Trend Lead District of southeast Missouri do not contain a single crystal of fluorspar.

The unusually high content of fluorspar in the shallow mined ores of the Southern Illinois district and the presence of carbonatite-related fluorspar deep ores at Hicks Dome invite speculation on the relationship between the two ores. One interpretation could be that basin-derived MVT ore fluids were introduced at the same time that carbonatite-related fluids were introduced at Hicks Dome and that the two fluids mixed to produce the fluorspar-sphalerite-galena ores of the Southern Illinois District (Reynolds et al, 1997). Another interpretation could be that the carbonatite-related fluorspar ores were introduced earlier than the MVT fluids, and that the subsequent MVT fluids dissolved fluorspar from the carbonatite-related fluorspar deposits (Hagni, 2015). It appears that the abundant apatite crystals in the carbonatite-related fluorspar deposits at Hicks Dome were largely insoluble in the MVT ore fluids and were left behind in those deposits. The fact that not one crystal of apatite has been found in the shallow mined fluorspar deposits favors the latter interpretation.

Oldoinyo Lengai active carbonatite volcano, Tanzania

Although the Oldoinyo Lengai carbonatite volcano in Northern Tanzania is not a deposit of industrial minerals, it is included here because of its importance to the origin of carbonitites and to the importance of the understanding of carbonatite-related fluorspar deposits. Oldoinyo Lengai is a clear example of the extrusion of present-day carbonatite that leaves no doubt as to its magmatic origin (Hagni, 2015). It is also important because it shows that volcanic carbonatite lava, which contains much of its original volatiles, can have very significant fluorine contents. The carbonatite lava contains 5% fluorine or 2.5% fluorspar in the groundmass (Keller and Krafft, 1990; Hagni, 2015).

APATITE DEPOSITS

Cajati Apatite mine, Jacupiranga cabonatite Brazil

The Cajati carbonatite-hosted apatite mine (Figure 8) in southern Brazil recovers apatite from the world-famous Jacupiranga carbonatite. Calcite is recovered as a by-product from the tailings resulting from beneficiation of apatite, and is used in the cement industry. Mining at Cajati began in 1943 with the extraction of supergene ores with about 20% P_2O_5. 30 years later mining progressed into bedrock ores with 5% P_2O_5.

Carbonatite is younger than the abundant jacupirangite (nepheline-magnetite-titanaugite) in the open pit mine and where the two rocks are in contact the edges of jacupirangite breccia blocks are metasomatised to distinctly banded sodic fenites (Figure 9). Disseminated pyrrhotite and magnetite are common in the carbonatite (Figure 10).

Figure 8. *Photograph showing the open pit at the Cajati apatite mine, Brazil. The darker areas of rock are earlier intruded jacupirangite and the lighter areas are subsequently intruded carbonatite. Mine equipment for scale.*
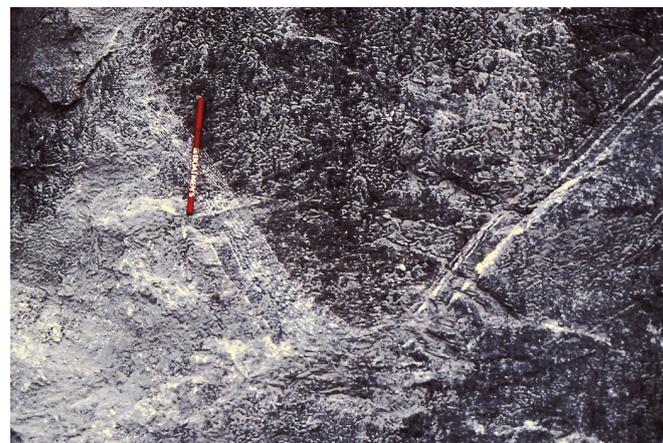

Figure 9. *Mine photograph showing a close view of the contact between jacupirangite (black, upper right) and carbonatite (white; lower left). Soda-rich fluids from the intruding carbonatite have metasomatized the margins of the jacupirangite to form a sodic fenite (banded). Cajati apatite mine, Brazil. Red pen for scale.*
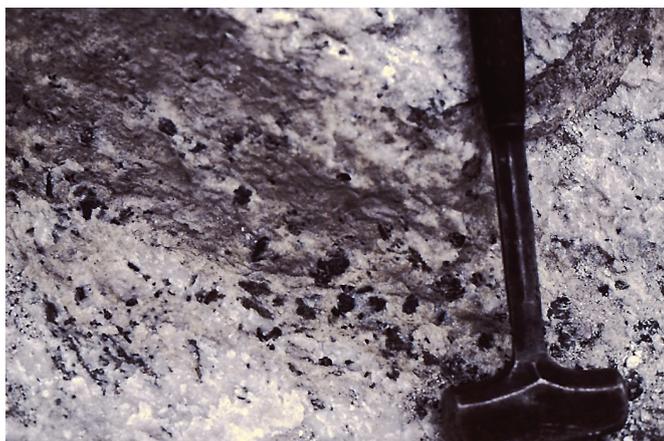
Figure 10. *Mine photograph showing abundant magnetite (black, equant) and pyrrhotite (black; elongate) disseminated in carbonatite (white). Cajati apatite mine, Brazil. Geological hammer for scale.*

Figure 11. *Photograph showing the main open pit at the Bayan Obo magnetite-hematite-REE mine, Inner Mongolia, China. The ores are mined primarily for the iron minerals; magnetite and hematite. The REE minerals are recovered as by-products. Mine equipment for scale.*

RARE EARTH ELEMENT (REE) DEPOSITS

Mountain Pass, California carbonatite-hosted REE mine

The Mountain Pass, California carbonatite-hosted REE mine was the world's largest REE mine from 1966 to 1985. The carbonatite complex is intruded into Precambrian metamorphic rocks. The REEs are present mainly as bastnaesite-(Ce) [(Ce,La)(CO$_3$)F], disseminated within the carbonatite. The average grade of the REE ore is 8.24% but as much as 25% of the REEs were lost to the tailings when the mine was operating. The separation plant was closed in 1998 and the mine was closed in 2002.

The Mountain Pass mine has recently been reopened as a result of restrictions on exports of REEs from China and the resulting increases in the prices of REEs. A new MolyCorp company, with the help of U.S. federal government grants has led to the mine's reopening. The mine has had a challenging startup due to the recent moderation in REE prices, the fact that 49% of the REE production at Mountain Pass is the low-value light REE cerium and only 15% is the higher-value neodymium and praseodymium. Further, MolyCorp is short of ready funds. On the positive side, their new chlor-alkali separation plant was completed in March 2014 (MolyCorp, 2014), and MolyCorp is developing a market for a water-purifying product that uses cerium.

Bayan Obo magnetite-hematite-REE mine, Inner Mongolia, China

The Bayan Obo magnetite-hematite-REE mine in Inner Mongolia, China is the world's largest REE producing mine. The mine consists mainly of two large open pits, the main pit (Figure 11) and the east pit (Figure 12), that mine the iron ore minerals, magnetite and hematite. Two rare earth minerals, bastnaesite and monazite, were first identified by a Chinese geologist, Prof. He Zuolin, in 1934. They were subsequently separated as a byproduct to the iron ore minerals and they now constitute the world's largest source of REEs. As recent as 2010 China was producing 97% of the world's REE minerals (People's Daily Online, 2011), although that figure has now declined slightly to 85% (Gresser, 2013).

Figure 12. *Photograph showing the east open pit at the Bayan Obo magnetite-hematite-REE mine, Inner Mongolia, China, which has been mined to a greater depth than the main open pit. Mine equipment for scale.*

The REEs are contained mostly in bastnaesite and monazite, but several other REE minerals have been identified as well. Monazite-rich ores are characterised by a brown cast (Figure 13), and bastnaesite-rich ores may have a striped texture (Figure 14). Reflected light microscopy provides a useful means for distinguishing
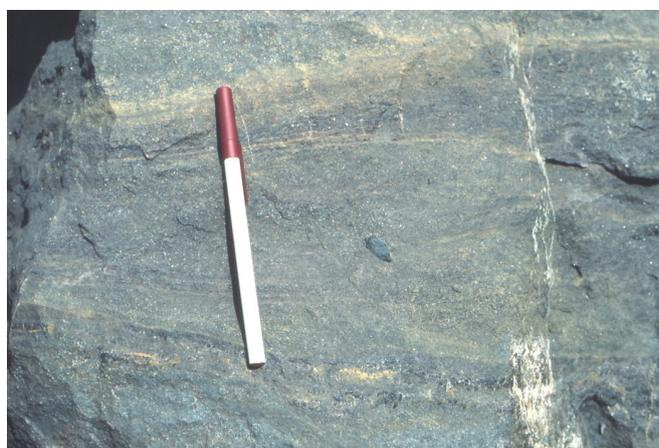
Figure 13. *Mine photograph of a hand specimen of monazite-rich, iron-REE ore. The presence of abundant monazite gives a brownish cast to the ore. Bayan Obo magnetite-hematite-REE mine, Inner Mongolia, China. Red and white pen for scale.*
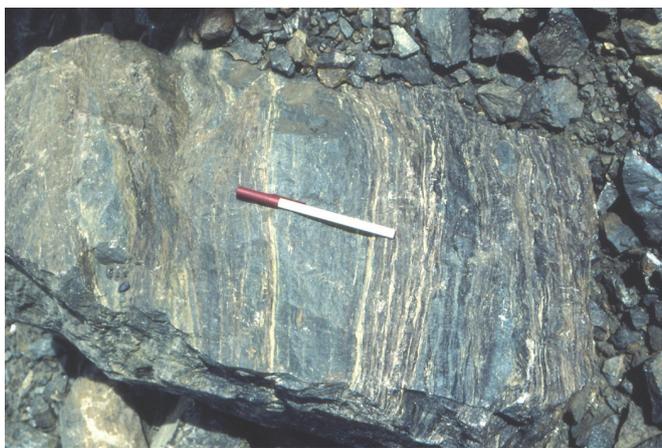
Figure 14. *Mine photograph of a hand specimen of bastnaesite-rich iron-REE ore. This specimen is typical of REE-rich ores called striped ore by mine geologists. Bayan Obo magnetite-hematite-REE mine, Inner Mongolia, China. Red and white pen for scale.*
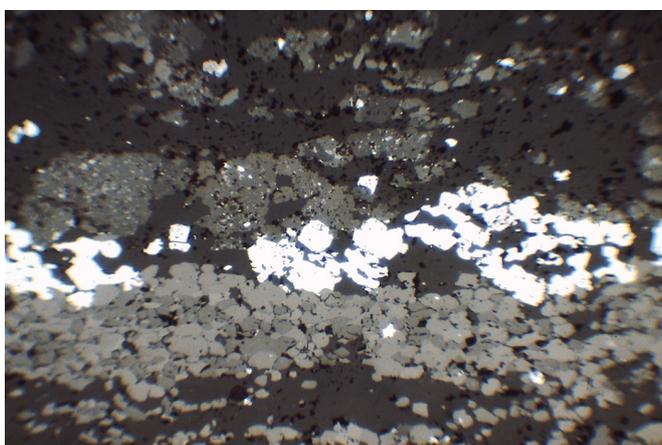

Figure 15. *Reflected light photomicrograph showing monazite (medium brown; R = 8) and fluorspar (darker gray; R = 4) in the upper half of the image, bright magnetite crystals (white, R=21) in the middle and bastnaesite (light brown; R = 9; partly elongate crystals) and quartz (dark gray; R = 4.5) in the bottom half of the image. Bayan Obo magnetite-hematite-REE mine, Inner Mongolia, China. Magnetite grains are about 25 μm across.*
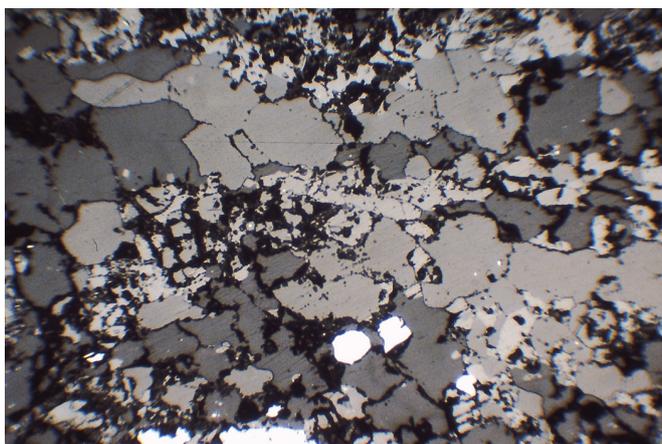

Figure 16. *Reflected light photomicrograph showing relatively abundant monazite (medium brown; R = 8) and closely associated bastnaesite (light brown; R = 9; reflective pleochroic, locally elongate) in the middle of the image. Quartz (dark gray; R = 4.5), fluorspar (even darker gray; R = 4), and small amounts of magnetite (white, R = 21) are also present in the image. Bayan Obo magnetite-hematite-REE mine, Inner Mongolia, China. The elongate basnaesite crystal in the center of the image is about 50 μm long.*

monazite from bastnaesite (Hagni, 2011) in the Bayan Obo ores. Under reflected light, monazite is brighter (reflectance = 8) than quartz and fluorspar (Figure 15), and bastnaesite is brighter yet (reflectance = 9) and locally may show elongate texture (Figure 16).

Although the origin of the ores at Bayan Obo have been debated, the discovery of carbonatite dikes in the vicinity, and the typical mineralogy of a carbonatite deposit strongly supports the interpretation that the ores are related to carbonatites (Le Bas et al., 1992).

Ashram REE deposit, Eldor carbonatite, Labrador Trough, Quebec, Canada

The Ashram REE deposit in the Eldor Carbonatite in the Labrador Trough of Quebec, Canada was discovered by Commerce Resources Corporation in 2009. The discovery has been ranked as one of the largest REE deposits outside China (Commerce Resources Corp News Release, August 25, 2011).

The carbonatite and niobium (Nb)-tantalum (Ta) (pyrochlore) deposits had previously been mapped by Tony Mariano. He graciously provided a map and field samples that he had collected for a microscopic study of lithologies and potential ores, later conducted by Wright (1997). The map (Wright, 1997, Figure 2, p. 4) shows the extent of the carbonatite and the locations of two areas of pyrochlore prospects, a northwest area and a southeast area. The pyrochlore crystals observed under reflected light are large euhdral octahedrons. Most of the monazite crystals that Wright observed under SEM-BSI were very small grains that averaged 15 μm, but some were as large as 50 μm.

The Ashram REE Zone discovered by Commerce Resources is located just to the southeast of the northwest Nb-Ta prospect area. It was discovered by following glacial boulder trains to their source. Based upon 57 drill holes (16,868m drilled) the Ashram deposit has been reported to contain: 29.3 million tonnes indicated ore at 1.9% Total Rare Earth Oxide (TREO), and 219.8 million tonnes inferred ore at 1.88% TREO with a 1.25% cut-off (Commerce Resources Corp News Release, March 20, 2014). In addition to monazite, bastnaesite and xenotime also have been identified in the Ashram deposit.

VERMICULITE DEPOSIT

Palabora carbonatite copper and vermiculite mine, South Africa

The Palabora carbonatite copper and vermiculite mine in northeast South Africa is best known for its copper production formerly from a large open pit (Figure 17) and more recently from underground. However, vermiculite also has been mined from two smaller open pits. Palabora, together with Goias, Brazil, produced 49% of the world's vermiculite in 2012.

Palabora also recovers several by-products as a result of the copper mining. Those by-products include apatite (Figure 18), titaniferous magnetite, uranothorianite [(Th,U)O$_2$] (Figure 19) and baddeleyite (ZrO$_2$) (Figure 20). Although the amounts of by-product minerals in the ores are small, the large volume of ores produced at Palabora results in a very sizeable production of those minerals.

Figure 17. *Photograph showing the open pit at the Palabora mine, South Africa. Subsequently the mine has gone underground. Mine equipment for scale.*
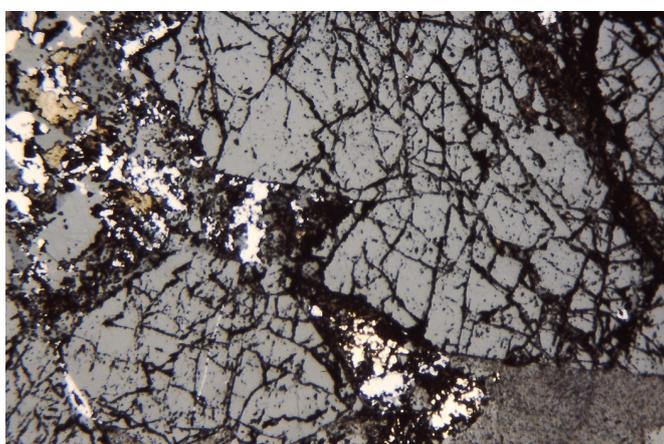

Figure 18. *Reflected light photomicrograph showing several large (about 350 μm long) prismatic apatite crystals (grey and fractured, with a reflectance of about 9) in Palabora copper ores. Also present are small amounts of pyrrhotite (white), chalcopyrite (yellow), valleriite (brown), and serpentine (dark gray, soft, lower right corner).*
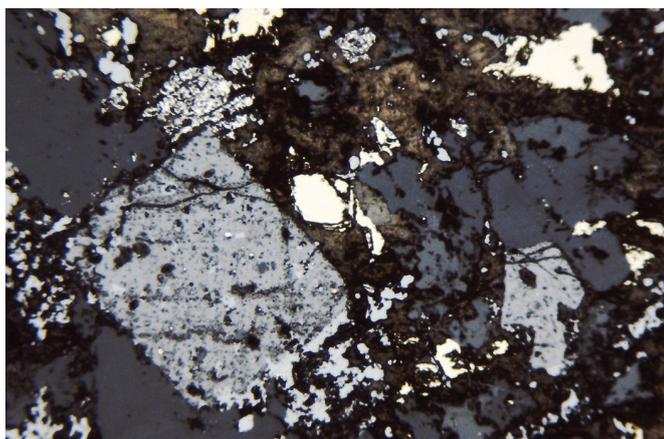

Figure 19. *Reflected light photomicrograph showing a large crystal of uranothorianite [(Th,U)O₂] (light blue grey, euhedral cube, 50 μm) in carbonatite calcite (dark gray) with closely associated radiogenic galena formed by the breakdown of the uranothorianite. Brown to grey grains of reflective pleochroic valleriite are abundant in the upper portion of the image, and small grains of pyrrhotite (yellow brown) are present mainly in the center of the image. Magnetite is locally present (white) and it is slightly brighter than the uranothorianite crystal. Palabora mine, South Africa.*
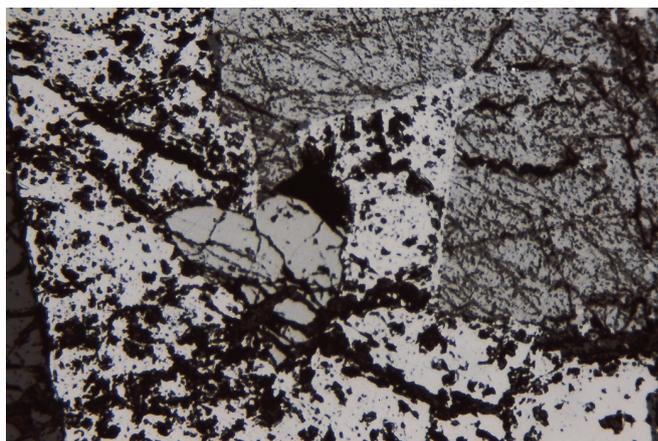

Figure 20. *Reflected light photomicrograph showing two baddeleyite (ZrO₂) crystals (medium grey; elongate prisms, about 40 μm long) left of the center where they are enclosed in magnetite (bright grey to white). Two large uranothorianite [(Th,U)O₂] crystals make up the upper right portion of the image (grey with abundant fine fractures) and calcite (dark gray) is present along the left side of the image. Palabora mine, South Africa.*

Palabora produces several thousand tonnes of thorium and uranium from uranothorianite recovered. The mine is the world's largest reserve of zirconium and produces about 11% of the world's zirconium from the baddeleyite recovered.

Vermiculite is an iron magnesium aluminum silicate that forms by surficial weathering of phlogopite contained in the pyroxinite of the Palabora complex. The vermiculite occurs in a small pit separate from the large copper pit. Palabora is the world's largest vermiculite mine, producing about 32% of the world's supply. The average grade is 22% vermiculite. The vermiculite occurs in large books often about 15cm across and 2.5cm thick.

OTHER INDUSTRIAL MINERAL DEPOSITS RELATED TO OR POSSIBLY RELATED TO CARBONATITES

The Araxa mine in Brazil is the world's largest niobium mine. The Catalao carbonatite complex at Goias, Brazil is the world's second largest vermiculite mine. The Mount Weld REE deposit in Australia has recently been put into production by Lynas Corporation. The Bear Lodge REE deposit has been extensively explored in Montana, USA. The Lemhi Pass REE deposit in western Montana and eastern Idaho, USA has been explored by drilling. The Elk Creek carbonatite deposit in southeast Nebraska has been extensively drilled and was the subject of six papers at the Geological Society of America symposium in 2014 (Carbonatites and Other Alkalic Rocks, 2014). The Elk Creek carbonatite is 7km in diameter, one of the largest known carbonatites, and it contains considerable Nb and REE minerals.

SUMMARY AND CONCLUSIONS

Carbonatites provide an important source for a variety of industrial minerals. Those minerals include fluorspar, rare earth elements, pyrochlore (Nb-Ta), apatite, vermiculite, calcite, magnetite, uranothorianite [(Th,U)O₂], baddeleyite (ZrO₂), and titaniferous

magnetite. For many of these industrial minerals, carbonatites constitute essentially the only economically viable source. This paper illustrates some of the industrial mineral deposits found in carbonatites and has presented some of the salient geological and mineralogical features of those deposits.

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