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Evidence for Fe-Sulfide and Fe-Ti Oxide Liquid Immiscibility in the Itsindro Gabbro Complex, Madagascar

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Abstract. The petrology and mineralogy of the Itsindro complex, south-central Madagascar has been investigated using samples obtained from a borehole drilled in the early 1970s on a geophysical and Ni-Cu geochemical anomaly. The host rocks consist of a pyroxenite unit with interbedded gabbro layers and a gabbro unit. A decrease in the Mg-number of cpx, independent of the lithology, is observed from the deepest samples (316 m) up to 163 m, suggesting a regular fractional crystallization process. The evolution of this parameter is more erratic in the upper part of the borehole. The mineralization consists of Fe-Ti-P oxides and Fe-(Cu-Ni) sulfides. The ilmenite-magnetite±apatite and pyrrhotite±chalcopyrite±pentlandite assemblages are always observed in interstitial positions. The contemporaneous character of the oxide and sulfide assemblages strongly suggests a two-stage evolution of the tholeiitic magma: first a separation between immiscible Si-rich and Fe-Ti-P liquids, followed by sulfide contamination of the latter leading to massive Fe-rich over Cu-Ni sulfides.

Keywords. Fe-Ti-P deposit; Fe-sulfide; liquid immiscibility; mafic-ultramafic complex; Madagascar

1 Introduction

The Itsindro Massif (south-central Madagascar, Fig. 1) was known to contain a base metal sulfide mineralization strongly dominated by pyrrhotite, without economic interest. Previous work described a total thickness of 16 m of sub-massive pyrrhotite between 72 and 90 m in the only drill core obtained from mafic-ultramafic rocks. Further work planned in the area was abandoned because of the lack of Ni-Cu mineralization.

As part of a research project aimed at understanding the parameters controlling the compositions of magmatic base-metal sulfide (BMS) assemblages (and specifically their Ni-Cu-PGE contents), this unconventional mineralization was revisited. The aim of this paper is to describe the mineralized drill core, in which we identified a double immiscibility process, leading to the coexistence of an Fe-Ti apatite mineralization and an Fe-sulfide mineralization (with virtually no Ni-Cu) hosted in pyroxenite and gabbro, throughout the section.

2 Geological environment

The Itsindro gabbro massif, located in the Itremo area, is believed to belong to a nearly continuous 450-km long belt of mafic and granitic plutons of Neoproterozoic age (Tucker et al. 1999) interpreted as the root of a continental magmatic arc emplaced between 804 and 776 Ma, during or slightly preceding the breakup of the Rodinian supercontinent.

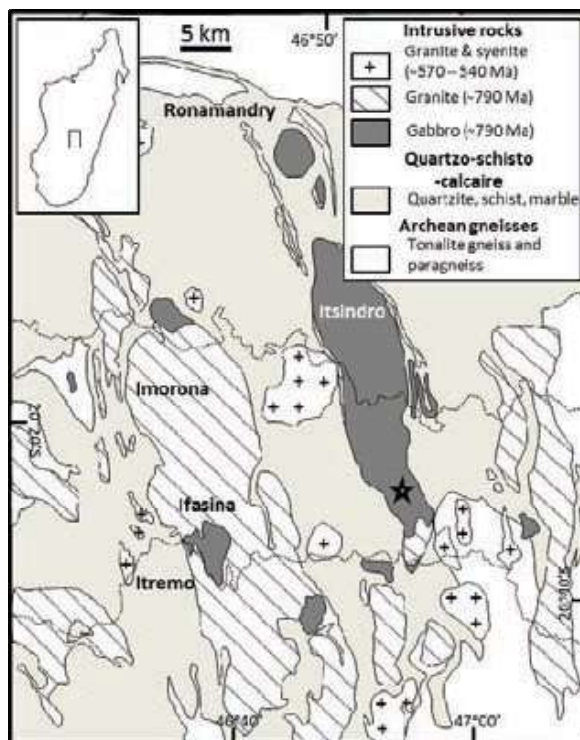


Figure 1. Geologic map of the Itremo region, showing the Itsindro gabbro and the location of the drilling (star). After Handke et al. (1999).

The Itsindro massif was studied in the nineteen-seventies mainly for its potential in Cu, after the discovery of malachite and azurite in a gossan developed on magnetite-rich gabbro. The work undertaken consisted of mapping, sampling (for petrographic studies), and the collection of stream sediments, soils and alluvial minerals over an area of about 500 km² (Coulombeau 1969).

The geological map shows an elongated massif, about 40 x 8 km with a N-S orientation, mainly composed of gabbro emplaced in the Itremo "Quartz-Schisto-Calcaire Series". Detailed mapping was carried out in 1968 under difficult conditions, partly due to the lack of access and outcrops. The massif was interpreted as a series of gabbro-forming sills or laccolites in the metamorphosed sedimentary series.

The geochemical exploration survey, conducted during the exploration campaign (soil analyses) in 1969, targeted two anomalous zones within the complex, with a superposition of the highest Ni and Cu values in both zones. The southern one includes a mineralized occurrence (known as "Lanjanina") discovered in 1958 and described as a gossan with copper carbonate and sulfide, in an environment dominated by magnetite

gabbro, pyroxenite and lherzolite.

A ground geophysical exploration survey (magnetometer, spontaneous and induced polarization, and resistivity) carried out in the gossan area gave positive results, and the superposition of the geochemical and geophysical anomalies was interpreted as indicating the existence of occurrences of massive sulfides at depth.

Following these encouraging results, it was decided to drill a deep vertical borehole where the geochemical and geophysical anomalies were optimally superposed. The drilling was stopped at a depth of 320.75 m. The results were considered to be negative, in spite of the abundance of sub-massive sulfides, which were largely dominated by pyrrhotite, with correspondingly low Cu and Ni values. All exploration work in this area was halted after this negative result.

The drill core recovered in 1969 was very well preserved in the former BRGM office in Antananarivo. The log is relatively simple at the macroscopic scale and comprises five main units:

- 0 – 8.2 m: oxidized iron cap (“gossan”),
- 8.2 – 15.35 m: gabbro (saprolite) unit,
- 15.35 – 34.02 m: granite unit (heterogeneous, weathered, and strongly fractured),
- 34.02 – 287.6 m: pyroxenite unit with dm to m thick dykes of fine grained granite above 120 m, and meter-thick gabbro layers throughout the unit,
- 287.6 – 320.75 m: main gabbro unit, with 3 to 7 m thick pyroxenite layers.

3 Petrography

All the rock types encountered are composed of various proportions of clinopyroxene, plagioclase, olivine, apatite, mica, zircon and secondary amphibole (orthopyroxene is totally lacking). The sulfide ore minerals include pyrrhotite (strongly dominant over the other BMS), chalcopyrite, pentlandite, rare cobaltogersdorffite, and traces of sphalerite. Oxides include ilmenite, magnetite, and spinel-group minerals. Graphite flakes are always found in close association with BMS. Titanite occurs sporadically within the Fe-oxides. Although the ore minerals (oxides and sulfides) are present throughout the drill core, their proportions and the ratio BMS/Fe-Ti oxides are highly variable, as reflected in the observed whole-rock TiO₂ and S contents, which range from 0.75 to 14.2 % and from 0.20 to 18 % respectively.

Pyroxenite with various proportions of ore minerals is the dominant facies in the pyroxenite unit. It is generally coarse-grained, exhibiting an adcumulate texture. Olivine has been found in four samples (out of 17), and does not exceed 5 % in volume. In some samples olivine occurs as rims around the ore minerals, in contact with clinopyroxene, resembling a reaction texture. In some cases the olivine rim is specifically altered to smectite. Some pyroxenites contain up to 10 % interstitial plagioclase; plagioclase may also locally form a very thin (20 µm) rim between the ore minerals and the silicates.

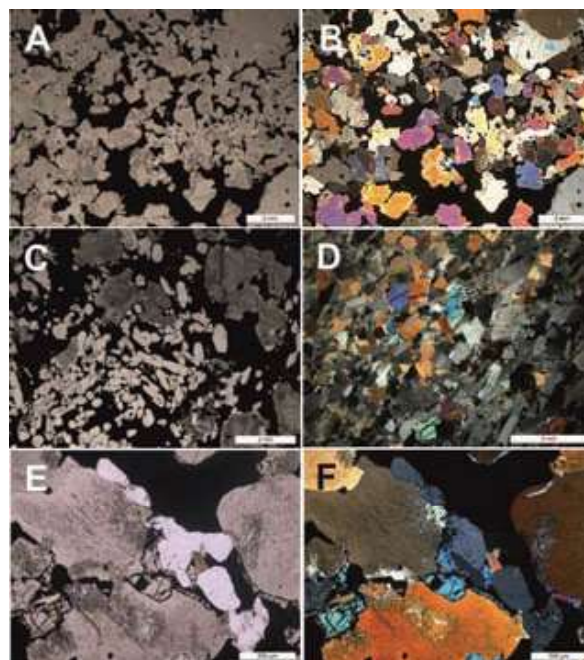


Figure 2. Photomicrographs showing typical facies of the Itsindro Gabbro Complex. **A, B.** mineralized pyroxenite (plane and cross-polarized light respectively), **C.** Apatite-rich mineralized pyroxenite, **D.** Gabbro showing a preferred orientation (cross-polarized light). **E, F** Apatite-bearing, mineralized olivine gabbro (plane and cross-polarized light respectively).

Disseminated ore minerals tend to be euhedral but are generally interstitial with respect to the silicates. Gabbro layers in the pyroxenite unit exhibit a fine-grained equant texture, with a relatively low (20 %) proportion of plagioclase, but a pegmatitic facies in which plagioclase dominates over pyroxene is also found in the same context. Contacts between gabbro and pyroxenite were not observed.

Gabbro from the gabbro unit seems to show a more pronounced magmatic mineral layering (Fig. 2D), marked by a preferred orientation of the plagioclase laths. Olivine seems to be systematically present in the basal gabbros and may be more abundant than in the pyroxenite unit. Similarly, pyroxenite layers in the basal gabbro are olivine-bearing. Their contacts with the host gabbro are sharp.

A hydrothermal alteration affects most of the gabbro and pyroxenite, and is characterized by the amphibolitization of pyroxene and the local development of carbonates.

At hand-specimen scale we saw no rhythmic layering anywhere in the borehole, and no evidence of a “layered complex”, but rather two main units, suggesting that the complex is composed of mega-cyclic units.

The proportions and the ratios between the ore minerals have been carefully studied. There are no occurrences of oxide without sulfide or sulfide without oxides, and in spite of local variations the average proportion may be estimated at 50/50. SEM images (Fig. 3) show the relationships between silicates, sulfides and oxides. Most oxide grains are two-phase associations, composed of ilmenite, free of hematite exsolution lamellae, and magnetite everywhere containing oriented

exsolutions of ilmenite and spinels. The contact between ilmenite and magnetite is highly irregular and marked by tiny spinel-group minerals (Fig. 3D). We find euhedral sulfide grains in oxides and euhedral oxides in BMS, and it was not possible to establish an order of crystallization between the oxide and the sulfide minerals.

When the ore mineral content is less than 60 % the ore displays a characteristic net-texture, and the oxide-sulfide association generally appears interstitial with respect to the euhedral magmatic silicates and apatite (Fig. 2).

Apatite is ubiquitous, and euhedral and is found included in the magmatic silicates, in the oxide phase, in the BMS (Figs. 2, 3), and in the oxide-BMS association. Its volume may locally reach 30 % of the sample.

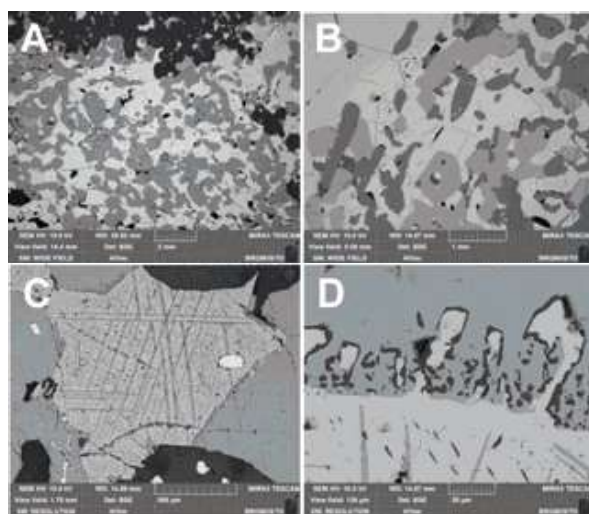


Figure 3. SEM images (BSE mode). **A.** Euhedral Fe-Ti oxide (gray) enclosed in Fe sulfide. Silicates in black. **B.** euhedral (elongated) apatite (dark gray) and euhedral Fe-Ti oxide (gray) enclosed in Fe sulfide. **C.** magnetite (with ilmenite exsolutions) adjacent to ilmenite. Silicate in black, sulfide droplets in white. **D.** Detail of the complex contact between magnetite (with ilmenite exsolution) and ilmenite (dark gray). In black, Al spinel surrounding the interfingering of magnetite in ilmenite.

4 Mineral chemistry

Clinopyroxene was analyzed in 21 samples and shows a wide range of compositions, mainly in the Mg number. The most Mg-rich clinopyroxenes are found in the gabbro, but there is a large compositional overlap between clinopyroxenes in pyroxenite and in gabbro. In the triangular diagram, they plot in the augite field, close to the salite domain. In terms of minor elements, Al_2O_3 content varies between 3.37 and 1.64 wt.%, and TiO_2 1.37 to 0.15, whereas Cr_2O_3 is generally below the detection limit, with a maximum of 0.06 wt.%.

There is a significant decrease in the Al_2O_3 and TiO_2 contents in the pyroxenites and gabbros correlated to an Mg decrease but the presence of ore minerals does not seem to influence the mineral composition.

As a function of depth, two domains appear: the first, from the bottom (316 m) to 162.8 m is marked by a regular decrease in the Mg number (and an accompanying drop in TiO_2 and Al_2O_3). This evolution

(Fig. 4) cuts across the petrographic and macroscopic units described in the log, and is compatible with an undisturbed fractional crystallization process. There is then a reverse trend observed at the 135.3 m level, within the pyroxenite unit, followed by a complex and irregular trend. In the upper part of the hole, Al_2O_3 in pyroxene seems to remain fairly constant whereas TiO_2 seems to vary in an erratic manner.

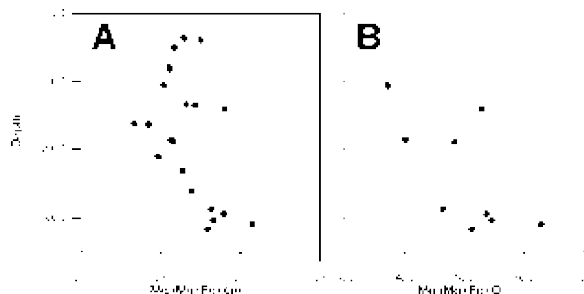


Figure 4. Plot of (A) molar $\text{Mg}/(\text{Mg}+\text{Fe}^{2+})$ in clinopyroxene and (B) Fo content (%) in olivine versus depth, from the Lanjanina sample. Dashed line corresponds to the marker level at around 163 m.

Olivine was analyzed in four pyroxenite and five gabbro samples. The Fo content varies from Fo 62.85 (lowermost gabbro) to 37.2 (uppermost pyroxenite). There is no relationship between the nature of the host rock and the olivine composition. Plotted against depth, there is a general tendency towards a decrease from the bottom to the marker level near 163 m as defined by the clinopyroxene composition (Fig. 4). Interestingly, as with clinopyroxene, above this limit the Mg number increases again, suggesting that the level corresponds to a significant horizon in the evolution of the complex. In terms of minor elements, the NiO content in the olivine appears typically low, <0.05 wt.%, whereas MnO, traditionally correlated with FeO, decreases from 1.13 (Fo 40.18 %) to 0.36 wt.% (Fo 62.85 %).

Plagioclase was analyzed in three gabbros from the basal gabbro unit and in three plagioclase pyroxenites and two gabbros from the pyroxenite unit. Plagioclase tends to be interstitial in pyroxenite, and to form laths in the gabbro. In some mineralized facies, it also occurs as a thin rim between clinopyroxene and ore minerals. Plagioclase is relatively homogeneous in composition within a single sample, but at the scale of the drill core variations from An 50.22 to An 98.14 have been recorded. In more detail, plagioclase from the basal gabbro unit appears very homogeneous (An57-An58) and rather similar to plagioclase from the gabbro in the pyroxenite formation (An around 53) whereas interstitial plagioclases in pyroxenite are more An rich with more variable compositions (An74.3 and An96-97).

Plotted against depth, the An composition seems to reflect changes in the mineral composition already observed around 163 m but there is no obvious correlation between the plagioclase and pyroxene compositions.

Magnetite shows little variation in composition. It is characterized by a low TiO_2 content (av. 0.45 %), a quite elevated V_2O_3 content (from 0.36 up to 2.45 %) and a consistently significant Cr_2O_3 content (av. 0.23 %).

Ilmenite is rich in Ti and Fe and exhibits consistent contents in MnO (av. 0.81 %) and MgO (av. 0.84 %).

The spinel-group minerals range in composition between spinel ss. and hercynite, their $\text{Fe}^{2+}/(\text{Fe}^{2+}+\text{Mg}^{2+})$ ratio ranging between 0.48 and 0.80. Their ZnO contents range between 1.51 and 6.92 % (av. 2.49 %). No correlation was observed between the composition of the oxides and the depth of sampling.

5 Discussion

Although occurrences of apatite-rich oxide rocks (“nelsonites”) are common in gabbroic and ultramafic rocks, the complex association described here between apatite, Fe-Ti oxides, Fe sulfides and mafic silicates is rather uncommon. As far as we know, only one similar occurrence of apatite- and oxide-rich rocks (all containing sulfide minerals in amounts up to several volume percent) has been described, from the Boulder Lake area in the Southern portion of the Duluth Complex, Minnesota, which presents some similarities with the mineralization described here. In the same complex, apatite and Fe-Ti oxide minerals are also found in the massive sulfide mineralization of the Babbith deposit. These occurrences led Ripley et al. (1998) to compare both types of mineralization and to offer speculations on their origin. They suggested that an immiscible sulfide liquid was present together with an apatite-oxide liquid at the origin of the Fe-Ti-P mineralization.

The mechanism leading to the formation of two liquids, an Si-rich liquid (evidenced by the abundance of granitic bodies in the Itsindro Complex) and an Fe-rich liquid, during the evolution of a tholeiitic liquid has been studied and demonstrated (Veksler et al. 2007; Namur et al. 2012). Similarly the mechanisms leading to the formation of an immiscible sulfide liquid are well

documented (Naldrett 1981).

Preliminary results obtained here strongly suggest that the Fe-Ti-P-bearing gabbro and pyroxenite were derived from an Fe-rich liquid formed by immiscibility. This liquid then underwent a certain degree of fractionation and we have evidence of at least two pulses of such a liquid. The abundance of base-metal sulfide must correspond to BMS crystallisation from an immiscible sulfide liquid formed in response to S contamination of the magma. The strong Fe enrichment in the magma explains the abundance of Fe-sulfide over Cu and Ni sulphides.

The potential for Fe-Ti mineralization in this intrusion, never evaluated, should be investigated.

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