

**MINERALIZATION AND WALLROCK ALTERATION IN GOLD-BEARING QUARTZ VEINS IN N.E.
BRAZIL**

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INTRODUCTION

Gold-bearing quartz veins are widespread in Borborema Province, N.E.: Brazil (Figure 1), the development of which seems to be closely related to shear zones reactivated during the Pan-African event (0.9-0.5 Ga; see Coutinho, 1994).

Although the Borborema Province shows a complex crustal evolution, the general structure consists of a mosaic of Archaean-Early Proterozoic massif terranes surrounded by Proterozoic fold belts.

Mineralized veins occur in a variety of host rocks: Archaean basement, Early Proterozoic metavolcanic-sedimentary fold belts, and Early to Late Proterozoic granitoids that intruded both the supracrustal and basement rocks.

In Borborema Province gold was first discovered in the early 1940's, and over the past 50 years extensive small-scale exploration and exploitation have been carried out by 'garimpeiros' (local prospectors). Since the end of the 1970's mining companies are showing more interest in the region. Two gold mines have been developed. The Sao Francisco mine, in Rio Grande do Norte State and Cachoeiras de Minas mine, in Paraíba State. Both are small deposits (metal reserva c.a.10 tonnes Au) with a grade ranging from 2.3 to 3.7 g/tonne of gold. The ore consists of gold-bearing quartz veins which are mined by open cast methods, the gold being recovered by cyanide heap-leaching. No major problem exists in gaining access in the region, and all facilities are available.

METHODS OF STUDY

Eighty six polished blocks of ore minerals and host rock samples from eight gold deposits (Table 1) have been studied by reflected light

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microscopy. This study was combined with scanning electron microscopy (SEM) and X-ray elemental mapping investigations. Many of the ore minerals have also been analyzed quantitatively by electron microprobe. Selected polished blocks of pyrite were etched with warm 20% HNO₃ to study textures and growth features. Samples of the wallrock alteration adjacent to the veins have been analyzed by X-ray fluorescence (XRF) for major and trace elements. Some gangue minerals were also analysed quantitatively by microprobe.

ORE-MINERALS

The mineralized veins are invariably dominated by quartz and locally tourmaline, and these make up at least 95 vol.% of the vein. Usually all the oxides occur disseminated in the wallrocks except the hematite, which is commonly also associated with the sulphides and gold. The other mineral groups are mainly present in the quartz veins. The overall 'ore'-mineral assemblage and paragenesis (see Table 2) identified in all deposits consist of the following: (i) Oxides: Magnetite ($\text{Fe}^{\text{II}}\text{Fe}_2^{\text{III}}\text{O}_4$); hematite (Fe_2O_3); maghemite ($\gamma\text{-Fe}_2\text{O}_3$), goethite ($\text{FeO}\cdot\text{OH}$), rutile (TiO_2); and ilmenite (FeTiO_3). (ii) Sulphides: pyrite (FeS_2), pyrrhotite (FeS); chalcopyrite (CuFeS_2); and, as minor constituents, marcasite (FeS_2), galena (PbS), covellite (CuS), chalcocite (Cu_2S), molybdenite (MoS_2), sphalerite (ZnS), bismuthinite (Bi_2S_3), and greenockite (CdS). (iii) Intergrown native bismuth, selenium/tellurium minerals and alloys of gold-silver and gold-bismuth. The X-ray investigations indicate a very characteristic but complex mineralogical association with native bismuth in symplectic intergrowth with bismuthinite, selenium/tellurium minerals, and alloys of gold-silver and gold-bismuth. The extremely fine grain size of these minerals did not allow the full identity of these phases to be established. The gold-bismuth (Bi = 10.7 wt%) alloy (maldonite) was observed among the intergrowths of uncharacterized selenium/tellurium minerals. (iv) Tellurides: Hessite (Ag_2Te) occurs as inclusions of very small grains within galena. A telluride of silver is also associated with chalcopyrite, and X-ray mapping reveals a clear concentration of Ag and Te at the edges of the chalcopyrite grain. The BSE image shows distinctive spongy aggregates along these areas. Microprobe analyses of these areas indicate very low contents of gold. These features, which are not consistent with the hessite associated with galena, could be indicative of a separate Ag-Au-telluride phase. (v) Native Gold. It occurs in several forms: (a) Very small particles associated with sulphides such as pyrite or chalcopyrite where the grain-size is extremely fine (around a few μm or less-grading into 'invisible gold'); (b) Disseminated

filling microcracks in the quartz veins, where it occurs as visible native gold; the grain size is predominantly $\approx 100 \mu\text{m}$; (c) Disseminated within pyrite as free grains of fine (size $\approx 100 \mu\text{m}$) or coarse (size $\approx 10 \text{mm}$) visible gold; and (d) As coarse grains of gold intergrown with native bismuth, bismuthinite and selenium/tellurium minerals, and associated with pyrite (size 10mm).

Chemical analysis by electron microscopy indicates that the native gold disseminated in quartz veins contains ≈ 95.9 to $73.4 \text{ wt } \% \text{ Au}$; 20.1 to $4.1 \text{ wt } \% \text{ Ag}$; 0.4 to $0.2 \text{ wt } \% \text{ Cu}$, and 0.1 to $0.3 \text{ wt } \% \text{ Bi}$. The gold content in sulphides ('invisible gold') ranges up to $0.72 \text{ wt } \%$.

Quantitative chemical analysis in pyrite shows that the gold distribution is not homogeneous. In the same crystal of pyrite ($\approx 100 \mu\text{m}$ in size) the gold content varies from below detection limit ($\approx 0.10\%$) to $0.72 \text{ wt } \% \text{ Au}$. Thus the gold content varies widely both within small grains and in adjacent grains. It is concluded that the gold occurs as submicron particles of native metal (see Cabri, 1992; Cabri et al., 1989; 1985; Starling et al. 1989). In contrast, the gold distribution pattern revealed by X-ray elemental mapping on the surface of chalcopyrite is more uniform and it does not show the same pattern as for Te and Ag, the distribution of which suggests random concentration areas in these elements.

On the other hand, this silver-gold-tellurium-bearing enrichments are concentrated in fractured grain boundaries and display a spongiform texture. It could indicate the destabilization of aqueous Te-Ag-bearing complexes on the surface of the sulphides as a result of the hydrothermal processes. These observations confirm the importance of sulphide surfaces and the conducting properties of sulphides to metal precipitation as previously mentioned by Starling et al. (1989).

Notwithstanding the very low contents of gold (close to the detection limit of the analytical methods) the random distribution pattern and scattered values of gold contents detected during this study appear to be more consistent with gold occurring as submicron particles within the sulphides rather than as solid solution, which is usually suggested in references.

Electron microscopy and microprobe analyses carried out on native gold grains shows that the values of the Au/Ag weight ratios were within the range from 3.7 to 23.1. The purity ('fineness') of the native gold in quartz ranges from 797 to 959. This result is close to values found in mesothermal lode gold deposits, typically ranging from 750 to 900 (Boyle, 1979). According to that author, gold of very high fineness (990 or more), is

generally the result of oxidation under conditions favourable for the complete removal of silver. In some auriferous deposits there is an indication that the early deposited gold is more silver-rich (lower in fineness) than that deposited later in the paragenetic sequence.

MICROSTRUCTURE OF PYRITE AND MARCASITE

The mechanism of pyrite deformation during metamorphism has been investigated (see McClay and Ellis, 1984). Etching of the pyrite grains in polished blocks during this study has revealed primary and growth structures. This study carried out on the ore samples has allowed recognition of three types of pyrite: (i) Pyrite type I is characterised by relict primary depositional textures such as spheroidal, rhythmic texture formed by fine aggregates of pyrite and marcasite and overgrowth zoning features. Brittle deformation textures (e.g. brecciation) are also found associated with this type of pyrite, revealing that a phase of deformation followed the first stage of pyrite formation. (ii) Pyrite type II is marked by ductile deformation textures such as stylolitic indentation of porphyroblastic euhedral pyrite due to pressure solution; fractures and cleavage filling with remobilised marcasite ("Filament-structure" of marcasite-pyrite; Bonnemaïson and Marcoux, 1990); and slip lines suggesting folding. (iii) Pyrite type III is defined by post deformation textures, e.g. annealing deformation, which has produced grain growth with boundaries at 120° triple junctions and a marked tendency towards large idioblastic pyrite porphyroblasts. Annealing pyrite crystals enclose coarse grains of gold. Locally, formation of a fine-grained pyrite-marcasite aggregate, is associated with pyrrhotite suggests recrystallization.

Pyrite type Y is closely associated with pyrrhotite and marcasite, but the textural evidence suggests that it is actually replaced pyrrhotite.

It is marked by a clear zonal growth pattern with cores of the earliest-formed pyrrhotite.

Pyrite type II is marked by clear deformation textures. It occurs intergrown with marcasite, and sometimes occurs in the shape of fractured prisms, due to brittle deformation. Although marcasite has no field of true stability (Gronvold and Westrum, 1976) it appears that annealing, even at relatively low temperature, results in transformation of the marcasite to pyrite (Craig and Vokes, 1993). The largest porphyroblasts of pyrite exhibit an arrangement of folding suggesting brittle-plastic deformation.

Pyrite type III is marked by recrystallisation to yield typical annealed textures with 120° dihedral angles. Craig and Vokes (1993) suggest

that the recrystallisation of pyrite with typical annealed textures may be related to an isochemical system relative to iron and sulphur, even if some fluids were expelled from or passed through the rocks.

MINERALIZATION EVOLUTION

A 'three-stage model of metallogenesis' based on the progressive increase of gold concentration related to the evolution of the shear zone has been proposed based on Bonnemaïson and Marcoux (1990): (i) The First Stage of Mineralization.

A first stage of gold mineralization in the second-order shear zones is common in Borborema Province. Magnesium and iron, mainly in the mafic rocks, were leached and partially fixed by the formation of opaque minerals (e.g. oxides and sulphides). Chloritization was common and caused release of it from primary Ti-bearing minerals. Ilmenite breakdown resulted in the formation of rutile and/or leucosene along the foliation of the mylonitic rocks. The titanium is thus mobile within the shear zone and indicative of intense hydrothermal activity in this structure during the first stage of evolution. Subsequent hydrothermal processes are accompanied by crystallization of pyrrhotite and pyrite disseminated in the mylonitic rocks.

Deposition of pyrite I is associated with pyrrhotite and marcasite (marcasite I). Spheroidal textures and overgrowth structures are preserved in porphyroblasts of pyrite, thus suggesting the first stage of mineralization.

The increase in the hydrothermal processes produced highly silicified zones where the quartz veins were developed. During this process the fluid flow provided conditions for mass transfer or fluid/rock interaction which caused change in the pyrite-pyrrhotite ratios. These ratios vary with the metamorphism since programmed heating results in sulphur release from pyrrhotite and retrograde cooling permits re-growth of pyrite as pyrrhotite releases sulphur (Craig and Volkes, 1993).

During this process the 'invisible' gold in the pyrrhotite (see Bonnemaïson and MARCOUX, 1990) is liberated and located at the surface of pyrite (in edges and surface defects), and as submicron particles in pyrite (I) and chalcopyrite as demonstrated in this study. Invisible gold associated with sulphides (pyrite and chalcopyrite) is therefore interpreted as gold formed during the first stage of the mineralization (gold I).

(ii) The Second Stage of Mineralization

During the second stage of mineralization rock/fluid mass transfer is more effective as a result of opening and shearing processes. The mineralized fluids rich in elements leached from the host rocks (e.g. Fe, Cu, Zn) cause destabilization of the gold-bearing pyrrhotite-pyrite.

The first mineralization is strongly obliterated. The presence of 'filament-structures' in marcasite-pyrite is characteristic of the transformation of pyrrhotite-pyrite, representing a relict of the first stage (marcasite II).

The second stage is marked by an Au-Pb correlation, with minerals such as galena I (which is not associated with tellurides and shows evidence of deformation) and sphalerite, chalcopyrite, and the first deposition of visible gold (gold II). Gold occurs filling microcracks in the quartz vein where the grain size is predominantly $\approx 100 \mu\text{m}$. Chalcopyrite commonly occurs replacing pyrite and/or pyrrhotite. This generation of pyrite is defined by a typical deformation texture and displays evidence of brittle-plastic deformation (pyrite II).

The mineral assemblage consists of intergrowths of native bismuth, bismuthinite, uncharacterized selenium/tellurium minerals and alloys of gold-silver and gold-bismuth, and marks a second stage of precipitation.

Molybdenite also occurs in this stage. This paragenesis is characterised by enrichments in Cu, Zn, Pb, Te, Bi, and Mo elements which possibly reflect a plutonic influence.

(iii) The Third Stage of the Mineralization

The third stage of the mineralization is marked by the introduction of lead and tellurides, as can be seen in the Igarape Garapa occurrence, host rock of which is the syn- and late-tectonic granites.

The gold (gold III) occurs as coarse grains which are disseminated in sulphides (pyrite) and associated with galena without evidence of deformations and hessite (galena III).

The pyrite is usually characterised by annealing texture and coarse grains (pyrite III). It is often associated with idiomorphic marcasite (marcasite III) which displays coarse twin lamellae.

GEOOTHERMOMETRY

Based on the mineral texture and parageneses, the ore-gold mineralogical study has therefore suggested two main phases of mineralization:

- (i) An oxide phase containing magnetite, ilmenite, and rutile. This assemblage is primarily the result of modifications to the original host rocks. It took place under high temperatures (= 500°C), and a brittle regime.
- (ii) A sulphide/solenoid/telluride stage containing base and precious/rare metals (Au, Bi, Ag). This assemblage was formed at a temperature <275°C under plastic-brittle conditions. The mineral assemblage bismuthinite and native bismuth is also consistent with a temperature of 270°C.

GANGUE MINERAL ASSEMBLAGES

The gangue consists of two types of mineral assemblages: supergene and hypogene. The hypogene minerals are dominated by quartz and tourmaline, and more rarely by: K-feldspar, albite, biotite, muscovite/sericite, fluorite, and chlorite. The supergene assemblage is related to weathering, and consists of jarosite, goethite, maghemite and pyromorphite (see Table 2).

Tourmaline is predominantly associated with the quartz veins and typically has a polyphase occurrence, suggesting several superimposed hydrothermal generations. Epitaxial crystals of tourmaline were analysed by microprobe giving a formula of $\text{Na } 0.8 \text{ (Mg } 1.2 \text{ Fe } 1.9 \text{ Ti } 0.1 \text{ Al } 0.6) \text{ Al } 6[\text{Si } 6.6 \text{ O } 18] [(\text{BO}_3)_3 (\text{OH}, \text{F})_4]$ indicative of a $\approx 50 : 50$ schorl-dravite mixture. The atomic ratio $\text{Fe}/(\text{Mg}+\text{Ti}) = 1.48$ is closest to the values for hydrothermal tourmaline, the ratios of which range from 2 to 6; tourmaline from other environments has values higher than 6 (Charoy, 1979). The iron content is compatible with the analyses of tourmaline from granites, aplites, pegmatites, greisens and lodes (Power, 1968).

The blue-green pleochroism is also consistent with that of hydrothermal tourmaline (Charoy, 1979).

Fluorite (CaF_2) is very rarely seen in the mineral assemblages. It occurs as disseminated grains associated with quartz hosted by the less-deformed syn- and late-tectonic granites (e.g. Igarape Garapa).

Jarosite [$\text{KFe}_3^{+3}(\text{SO}_4)_2(\text{OH})_6$] has been identified by its optical features and confirmed by microprobe analysis, as a very persistent but not predominant mineral in all of the gold mineralized quartz veins.

WALLROCK ALTERATION

In the Borborema Province gold occurs predominantly in quartz veins without extensive development of alteration zones. However four alteration zones were selected with varying host rock lithologies: mafic igneous,

schist, and granitoid. Although the host lithologies show differences they were all subjected to the same metamorphic processes.

The areas are characterized by amphibolite facies domain which have been subjected to metamorphic retrogression. Two of these zones are developed in supracrustal rock types and the other two are related to granitoid rocks. The relationships between the host rocks and the types of alteration, mineralogy and the ratio enrichment/depletion of major and trace elements are shown in the Table 3.

(i) Wallrock Alteration in the Granitoids

Two types of wallrock alteration zones have been identified: (a) potassic-silicic alteration; and (b) tourmaline-potassic-silicic.

The first type of the wallrock alteration exhibits a zone of alteration penetrating the host rock from a few centimeters to 1 m, which is developed along a fault (S 60° E). It is characterised by a pervasive style of alteration where brown biotite is the dominant mineral, although K-feldspar forms thin pink layers. Intercalated in the alteration zone are several very thin quartz veins which are mineralized with gold and sulphide. Their thickness is commonly 20 to 40 cm.

The second type extends to =4 m width, and is dominated by a rhythmic alternation of zones ranging from 20 cm to 60 cm in width. It consists of a wide transition zone of: (i) dark tourmaline; (ii) pink K-feldspar (orthoclase) whose grain size ranges from fine to coarse (5 cm length); and (iii) silicified areas. It forms a mixture of tourmaline-quartz or tourmaline-potassium feldspar zones as a result of early potassium silicate alteration and a later overprinting tourmaline alteration. The alteration style is vein controlled.

(ii) Wallrock Alteration in the Supracrustal Rocks

In the supracrustals, although the alteration products do not extend as far as in the granitoid environment, two types of wallrock alteration were recognized: (a) wallrock alteration in schist; and (b) wallrock alteration in the contact meta-mafic/gneiss rocks.

In the metasediments (Sao Francisco mine) a mixed zone occurs from the unaltered rock inward towards the vein, involving alteration consisting of silicified-hematitic and silicified-potassic-tourmaline zones, with a predominance of the first type. Both types are marked by a high degree of silicification. The hematitic-silicified type consists of quartz, hematite, goethite and jarosite. It usually has a rusty-red colour and pervasive, finely disseminated alteration, while the second type can be recognized by its black colour, which is formed by tourmaline and quartz. The alteration

products extend from a few centimeters to 1 m away from the vein and are vein-controlled.

The silicified-potassium-tourmaline zones within the shear zone in the Itapetim District (Pimenteiras/Desterro areas) provide a second example of wallrock alteration in the supracrustal domain. In this case the alteration is pervasive and vein-controlled and consists of quartz veins with an intense silicification-feldspathisation zone where tourmaline may be locally abundant. The wallrock extends from a few centimetres to 1.5 m into the host rock.

RELATIONSHIPS BETWEEN WALLROCK ALTERATION, DEFORMATION AND GOLD MINERALIZATION

Although different lithologies have been subjected to wallrock alteration, they exhibit a similar pattern of wallrock alteration, and the resulting mineral assemblages reflect the fact that these rocks were all subjected to the same metasomatic processes.

The geochemical study demonstrates that there has been a major introduction of K, Ti, and Mg and locally Fe, and other elements (Pb, Th, Rb, Zr, B, Nb, Y, Sc, Ga) into all rock types. Depletions in Ca and Na are common. So, the petrography and geochemical data reveal that the nature of the local host rock has not controlled the hydrothermal fluid composition.

Major and trace element analyses also suggest a genetic link between the gold mineralization and the granitic magmatism. The high values of K, Ba and Rb, and the high contents in B (assumed by the presence of tourmaline), suggest that the hydrothermal fluid was associated with a K-silicate phase of a typically hydrothermal-metasomatic paragenesis (Kerrick, 1989). Gold occurs in the tourmaline-rich veins. However, there is evidence of deformation taking place in these veins prior to gold mineralization. For example, kinked tourmaline crystals and very fine-grained quartz surrounding porphyroblasts of K-feldspar, or fine-grained quartz surrounding porphyroblasts of K-feldspar, or recrystallised ribbons of quartz parallel to the regional foliation, all indicate that the tourmalinization is pre-syn shear event (D_4) and pre gold mineralization. Thus the wallrock alteration seems to have taken place before the gold mineralization, possibly at the same time as some of the early sulphide-rich stages of mineralization. A similar syn-deformation phase of alteration and later gold mineralization has also been proposed for other Precambrian Au deposits (Colvine et al., 1988).

CONCLUSIONS

The study of ore-mineral and gangue assemblages, wallrock alteration and the sulphide textures has placed several constraints on the evolution of the gold mineralization in Borborema Province:

(i) The ore development and its concentration is characterised by three stages of mineralization. The first stage results predominantly from brittle deformation and hydrothermal alteration. The mineral assemblage is characterised by early, high temperature formation of Ti minerals and Fe oxides from the destabilization of mafic minerals. The Fe and Mg leached and removed from the host rocks provided conditions for the precipitation of the sulphides and some metals (e.g. Au and Ag) on the surface of the sulphides as submicron particles. An increase in the hydrothermal processes produced the quartz veins. During this stage the fluid/rock interaction placed constraints on the mass transfer, causing a change in the pyrite-pyrrhotite ratio (programmed heating released sulphur to form pyrrhotite and retrograde cooling allowed re-growth of pyrite).

(ii) During the second stage, the mineral assemblage is enriched in elements that indicate a plutonic influence, in particular K, F, and B, but also Pb, Bi, Te, Mo and B. This stage was marked by a Au-Pb correlation and deposition of sulphides, oxides and tellurides.

The visible gold liberated from the sulphides is associated with recrystallised sulphides (pyrite and chalcopyrite) or intergrown with bismuth and selenium/tellurium minerals, suggesting deposition at a temperature of $\approx 270^{\circ}\text{C}$. The paragenetic sequence indicates that the mineralization postdated the amphibolite facies metamorphism (major regional metamorphism) and took place mainly between $270\text{-}200^{\circ}\text{C}$. The association of mineralization with early potassic alteration and tourmaline suggests primarily acidic magmatic-hydrothermal fluid.

(iii) The third stage is marked by the enrichment in lead, tellurides, gold and fluorite. Gold occurs disseminated in the annealed sulphides with a fine to medium grain size.

(iv) Weathering provided the conditions for the formation of: maghemite, cerussite, pyromorphite, jarosite and goethite.

(v) Tourmaline, one of the dominant gangue mineral after quartz, is indicative of hydrothermal activity.

(vi) Although alteration haloes are not always well developed, the major and trace element distribution appears to be consistent for all deposits. The geochemical data for the major elements indicate that the wallrock

alteration involves massive additions of K and less Ti, Fe and Mn. The trace element data defines the mineralized areas as a region of a distinctive suite of elements: Ba, Pb, Th, Rb, Cr, Ni, V, Zn, Sc, Ga, Y, Zr, Nb and Nd. Removal of Ca and Na is typical. The high values of K, Ba, and Rb and the high contents in B and F (assumed by the presence of tourmaline and fluorite) suggest that the addition of these elements was effected by a K-silicate phase of a typically hydrothermal-metasomatic paragenetic association (Kerrick, 1989).

(vii) Petrographic and geochemical data reveal that the nature of the local host rock has not been an important influence on the resulting alteration assemblage, and therefore has not controlled the hydrothermal fluid composition.

(viii) The wallrock alteration seems to have taken place before the gold mineralization, possibly at the same time as the early sulphide-rich stages of mineralization.

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