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Cu-ores in quartz-carbonate veins at Estremoz-Alandroal and Barrancos-S^to^ Aleixo regions (Ossa Morena Zone): a result of Late-Variscan hydrothermal activity?

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Abstract

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In SW Iberia, a large number of ore showings and deposits of epigenetic Cu-dominant mineralisation exists. Quartz-carbonate lodes and/or mineralised fault breccias are preferentially hosted in Palaeozoic metasedimentary sequences. Their development is intimately related to faulting of presumable Late-Variscan age. Theoretical considerations concerning Cu solubility in hydrothermal systems suggest that metal deposition might have involved low to moderate saline fluids previously responsible for metal scavenging in metasediments at *ca.* 325-375ºC and $fO_2 < 10^{-40}$ (at pH = 4, $aCl^- \approx 3$ and $a(\Sigma S) = 0.001$). Copper deposition as *cpy* is expected to occur at *ca.* 260-300ºC under $10^{-33} < fO_2 < 10^{-28}$ as a result of different mechanisms.

Introduction

In SW Iberia there are a large number of ore-showings and deposits that show a relatively narrow variety of metal associations and have many features in common. They include epigenetic, structurally controlled sulphide ores, mostly in quartz-carbonate veins along fault zones (usually N-S to NE-SW or ENE-WSW to E-W-trending strike-slip faults with normal component of movement), which are preferentially hosted in metasedimentary series (often, turbiditic in nature) of Ordovician-Silurian to Carboniferous age. According to data presented in many non-published technical reports and in several papers (*e.g.* Gaspar, 1967, 1968; Oliveira, 1984, 1986; Goinhas & Martins, 1986), these ore-showings and deposits can be grouped in three major types characterised by the following metal associations: 1) Sb(-Cu, Ag, Au); 2) Pb(-Ba-Zn, Sb, Ag); and 3) Cu(-As, Sb, Ag, Au). Many of these ore-showings were actively explored in the past and some deposits were extensively mined, particularly during the last decades of the 19^th^ and beginning of the 20^th^ century. Total metal production is difficult to calculate accurately, but the simple addition of the officially declared amounts (only available for some deposits) leads to minimum estimates of several thousands of tons of Cu-ore concentrates @ 25-32%Cu and several hundreds of tons of Sb-ore concentrates @ 35-40%Sb (the grades indicated for both metals represent the most common range of declared values); for Pb- and Zn-ores the available information is very scanty and appears not to reflect suitably the actual production as qualitatively inferred on the basis of the mine tailings volume. Presently, none of these (ore-showings or) deposits is exploitable. However, their widespread distribution and remarkable metal content (especially if the mineralogical nature of the ores, their distribution in lodes and the their average size is considered) pose important questions concerning the sources of metals and fluids, the reasons for a sustained high heat flow, the processes involved in metal transport and the mechanisms implicated in metal deposition. Answering these questions will improve significantly the current knowledge on the geodynamic and metallogenic evolution of Ossa-Morena (OMZ) and South Portuguese Zones.

The present work will focus only on a small part of the third group of ore-showings and deposits (containing Cu-dominated mineralisation), namely those recognised long time ago in the Estremoz-Alandroal and Barrancos-S^to^ Aleixo regions (OMZ). This extended abstract reports the first results obtained in an ongoing research project that intends to examine the origin and evolution of the above mentioned epigenetic ore-forming systems. Work in progress and further research will enable to determine the compositional variability of the major mineral phases that form the mineralisation and their host rocks, as well as to characterize in detail their bulk geochemical signature. These data, along with isotopic (stable and radiogenic) determinations and results of microthermometric and spectroscopic studies of fluid inclusions, will allow testing the hypotheses here articulated in a near future.

General geological features of some epigenetic, Cu-dominated ore-forming systems

In the Estremoz-Alandroal region, the major deposits are Bugalho, Miguel Vacas, Moviços, Mostardeira and Zambujeira, all belonging to the Sousel-Barrancos belt as defined by Oliveira (1986); for additional information see Matos & Rosa (2001) and references therein. In the <u>Bugalho</u> deposit, the main lode (N25ºE,80ºNW) shows a maximum thickness of 1.2 m and was mined in an extent of 800 m up to *ca.* 200 m deep; two other lodes were identified (N-S,50ºE and N-S, sub-vertical), although poorer in sulphides. All these lodes are preferentially hosted in metasediments of the *Charneca Fm.* of Silurian age (mostly grey schists with intercalations of lidites and carbonaceous schists) and are structurally controlled by a major NNE-SSW fault zone; secondary, sub-vertical E-W fault zones can also be observed (Oliveira, 1984). In the Miguel Vacas deposit, the main lode system has an average NNW-SSE trend, dips 50º-70ºE and was recognised in an extent of 2000 m, being preferentially hosted in Silurian carbonaceous schists with intercalations; a second lode of minor importance was also identified 500 m eastwards of the major mineralised system, following a N-S trend and being enclosed in mafic metavolcanics (Oliveira, 1984). Prospecting works carried out during the sixties and seventies of the 20th century enabled to recognise the mineralisation until 330 m in depth, the first 80-90 m corresponding to an important horizon of supergene metal-enrichment. Reserves indicated in 1975 by EMIL point out to 1.17 Mt @ 1.5% Cu of secondary, oxidised ores and 1.73 Mt @ 1.15-1.49%Cu of primary, sulphide ores. In the Moviços deposit, the mineralised domain of the fault zone (N15ºW,80ºE) is hosted in detrital metasediments of the *Serra Colorada Fm.* (Upper Ordovician) and in carbonaceous schists (with intercalations of lidites) of Silurian age that occupy the Moviços synclinal hinge. The lode can be followed for more than 1000 m and the old underground mine works attained 80-90 m in depth. In the Mostardeira deposit, the main lode (<4.5 m thick) follows a sub-vertical, N80ºE, left lateral fault zone, and is hosted in schists and greywackes of the *Terena Fm.* (Upper Devonian). The underground mining works performed in the final decades of the 19th century reached the depth of 120 m. Detailed mapping show that the lode comprises distinct zones of variable enrichment in different sulphides and/or sulphosalts, higher when greywackes are the host rocks.

In the Barrancos-S^{to} Aleixo region, the most paradigmatic deposits are Aparis and Botefa located in the Sousel-Barrancos belt; the S^{to} Aleixo-Achada de João Alves prospect represents a similar epigenetic mineralisation developed in a relatively different geological setting positioned in the Arraiolos-S^{to} Aleixo belt (Oliveira, 1986, 1991) – for additional information see Matos & Rosa (2001) and references therein. In the Aparis deposit, the lodes are related to a conjugate set of fault zones, the main system being sub-vertical and striking N10-20ºE. The sulphides are irregularly distributed and form different ore-shoots (9 were recognised by the SFM up to 150 m in depth), which are preferentially hosted in the turbitic sequence of the *Terena Fm.* (Upper Devonian), although some less mineralised segments of the fault zone can also be found enclosed in the *Xistos Raiados Fm.* (Lower Devonian) – *e.g.* Rhoden (1956), Mendes (1967), Gaspar (1968). Reserves calculated in 1965 by SFM are of 175925 t @ 2.76%Cu. In the Botefa deposit, the NNE-SSW,70ºNW mineralised fault zone can be followed for several hundreds of meters. The mineralisation, preferentially hosted in schists and greywackes of the *Terena Fm.*, was recognised and mined till 176 m depth. In the S^{to} Aleixo-Achada de João Alves prospect, drilling performed in 1991 by IGM intersected the hidden lodes, which are apparently controlled by a late, sub-vertical NNE-SSW fault zone that also cut the S^{to} Aleixo da Restauração Variscan Thrust Zone. The lodes are hosted in strongly hydrothermally altered mafic metavolcanic rocks belonging to a volcano-sedimentary sequence of Middle to Upper Silurian age.

Mineralogical and geochemical features of epigenetic Cu-dominated ores

All the afore-mentioned lodes (massive and usually fractured) are mainly composed of different generations of both milky quartz and carbonates. The latter minerals make up distinct aggregates that form the vein-matrix along with quartz, and fill up diverse fracture systems whose relative chronology is difficult to establish. The main carbonate associations are: *sd + ank ± dol* in Bugalho; *cal ± ank ± sd* in Miguel Vacas and Moviços; *sd ± dol ± cal* in Mostardeira; *dol + ank ± sd ± cal* in Aparis and Botefa; and *ank + cal* in S^{to} Aleixo-Achada de João Alves. Minor amounts of chlorite are also present in lodes at Aparis and Bugalho (filling up trans- and intergranular microfractures only intersected by very late carbonate veinlets), Mostardeira (finely disseminated in late quartz aggregates) and at S^{to} Aleixo-Achada de João Alves (forming relatively coarse-grained aggregates that seal interstitial spaces of the siliceous matrix or fill up distinct sets of transgranular microfractures). Mineralised fault breccias, enclosing heterometric, angular and hydrothermally altered fragments of diverse host rocks, can be found in Botefa, Bugalho, Miguel Vacas, Moviços, and Mostardeira.

An accurate characterisation of primary ore mineralogy is presently a very difficult task because of the problematical access to the old mine works. However, a comprehensive inspection of samples collected in mine tailings has led to results that are consistent with data presented in many technical reports contemporary of mining activity. In general, ore mineralogy consists of copper-iron sulphides, sulpho-arsenides and sulphosalts, often with accessory amounts of lead and zinc sulphides. In Bugalho and Miguel Vacas, *cpy + py ± apy ± tn-td* is the main ore assemblage; *cpy* occurs as fractured masses developed mostly after *py* but before *apy* and *tn-td*, the latter often forming tiny grains deposited in late microfractures. At Moviços and S^{to} Aleixo-Achada de João Alves the mineral assemblage is apparently simple and contains just *cpy + py* that develop coarse-grained aggregates randomly distributed in quartz-carbonate veins. In Mostardeira, the mineralisation is essentially composed of *cpy + py + apy + td ± gn*; *cpy* postdates an earlier (strongly fractured) *py* generation and occurs mainly in carbonate-rich domains of the quartz lodes; a late, euhedral and non-fractured, *py* generation is associated with very fine-grained *apy* and/or *td*; *gn* is rare and seems to be deposited lately. In Botefa, a more extensive mineral assemblage is found, mostly comprising *cpy + py ± po ± gn ± apy ± td*; coarse-grained *cpy* aggregates occur along with late quartz-carbonate generations cementing the angular rock-fragments in fault breccias; early quartz masses include fine disseminations of *cpy* (that incorporate rare *po* inclusions) postdating the fractured grains of *py*; euhedral *apy* grains, locally associated with *td* and rare *gn*, occur in restricted matrix domains of the fault breccias, particularly in those subjected to intense late fracturing. The primary mineralisation in Aparis, the best-studied of this group of deposits (Gaspar, 1968), includes *cpy* + *py* ± *marc* ± *po* ± *td* ± *apy* ± *gn* ± *sph*; different generations of *cpy* can be identified, the early ones forming fractured masses (that may contain *po* relics) and the later ones consisting of coarse-grained aggregates that involve *py* + *marc*, which replace *po* and are intimately related to *sd* ± *ank* deposition; a late *py* generation develops together with *sph* (often showing *cpy* exsolutions) and *td*, and these minerals can be observed in lately fractured domains of the lodes; as in other cases, *gn* is uncommon but can be identified in some intensely fractured domains of the lodes, although always in those where *sph* is absent (Gaspar, 1968).

In all these deposits, an upper horizon of strong oxidation and hydration exists, often leading to a supergene metal enrichment zone of variable thickness that comprises mixtures in diverse proportions of secondary oxides, hydroxides, sulphates, phosphates, arsenates, native copper, hydrated Cu-bearing silicates and carbonates, and occasionally also secondary sulphides. Extensive lists of these mineral assemblages can be found in Gaspar (1968), Oliveira (1984) and Matos & Rosa (2001). According to the main purpose of this extended abstract we will not go into details on this issue, but it is worth noting that, in Miguel Vacas, the supergene horizon go as deep as 80 m and was subjected to extensive exploitation during the seventies and the first-half of the eighties of the 20th century.

As referred to above, the access to the old mine works is currently very difficult, thus limiting any sampling programme ample enough to characterize properly both the primary ore mineralogy and its bulk chemical composition. The geochemical data existing at present are, therefore, insufficient and further sampling efforts should be made in order to analyse drill-core samples when available. Nevertheless, a brief review of data available shows that: 1) in Bugalho, strongly oxidised and leached domains of the supergene horizon have 1795-9167 ppm Cu, 6610-7060 ppm As, and < 256 ppb to 5.2 ppm Au; the host schist with fracture infillings composed of secondary Cu-minerals hold 7.9% Cu, 2510 ppm As, and 2.8 ppm Au; 2) in Miguel Vacas, oxide ores may reach 22% Cu, 2500 ppm As and 356 ppb Au, whereas mineralised fault breccias display 3.6% Cu; 3) in Moliços, the Cu-content of fault breccias with secondary Cu-minerals reaches 6338 ppm, going up to 1.5% in strongly weathered samples; 4) in Mostardeira, fault breccias with fine *py* and *apy* disseminations show 7.4% As, 3.0 ppm Au and 350 ppm Cd, and according to old technical reports primary ores contain, on average, 2.2 % Cu; and 5) in Botefa, the metal contents of fault breccias go up to 7.9% Cu, 199 ppm Pb, 2906 ppm Zn, 1290 ppm As and 63 ppb Au.

Discussion and conclusions: some thoughts concerning hydrothermal circulation and ore genesis

Field evidence indicates that the Cu-dominant mineralisation here concisely described is intimately associated with faulting. This faulting dates presumably from Late-Variscan times (*ca.* 300-270/260 Ma), but precise dating of fault rocks and ore minerals is needed to establish this with certainty; note that the Alpine re-activation of some fault zones is rather evident in some places, which means that a (significant?) late hydrothermal contribution during Early-Alpine times cannot be precluded. Field relationships and the mineralogical-textural characteristics shown by quartz-carbonate lodes, mineralised breccias and host-rocks, also point to an epigenetic origin of this mineralisation. The zoned structural and/or mineralogical character (sometimes following a rhythmic pattern) displayed by many lodes, as well as the polyphase fragmentation revealed by many fault breccias, are taken as evidence for a repeated seismic activity of the major faults and a multi-stage circulation of mineralising, hydrothermal fluids. From this, one may conclude that, as expected, fluid flow focusing within faults is particularly important in areas of highest fracture density and opening, such as damage zones associated with fault jogs, bends and splays. Therefore, fluid-driven growth of hydraulically linked networks of faults and fractures is favoured through constructive interplays between deformation, fluid flow, and fluid pressure.

Macroscopic fluid pathways in fractured-controlled hydrothermal systems are influenced by the evolving connectivity among structural elements during progressive deformation, the most extreme flow focusing occurring at the onset of a system-wide fluid flow for which a percolation threshold is attained. This threshold value requires bulk strains as low as a few percent, but in the presence of rocks with contrasting mechanical behaviour, such as alternating sequences of greywackes and schists, the more competent rock domains will yield much earlier for a fixed temperature and strain rate, becoming gradually the preferential host of mineralisation, as observed at Aparis, Botefa and Mostardeira. In relatively monotonous schist series, like those in Bugalho and Miguel Vacas, for instance, the prior development of a (micro-)fracture network is needed to achieve the critical percolation threshold for lodes formation, preventing the dispersion of the mineralising fluid flow. This contrasting behaviour helps to explain the widespread distribution of a relatively high number of ore showings among which only a few important lodes can indeed develop.

From observation, experimental results and numerical modelling, it is known that fluid pressure gradients, buoyancy effects and permeability distribution are the main parameters that control the fluid pathways between metal sources and the sites of hydrothermal-ore deposition. Active deformation is therefore needed to generate and maintain permeability, and to sustain large-scale fluid flow, provided that anomalous heat flow exists. Accordingly, rocks with strain-dependent permeability, like those forming the Palaeozoic sequences of SW Iberia, will act as good sources for fluid flow, particularly if the initial increments of the Late-Variscan crustal uplift proceeded under, at least, moderate rates (*ca.* 3-5 $mm.year^{-1}$). In these conditions, broad gradients produced by buoyancy or fluid pressure will enhance long distance fluid flow provided that the local perturbations have less magnitude than the broad anomalies thus generated. This would control both the extent and the lifetime of large-scale hydrothermal systems, favouring also the processes of metal scavenging from substantial rock volumes. Local, small fluid flows around structural discontinuities, such as faults and/or fracture arrays of variable complexity, will favour, conversely, the tendency for flow focusing, controlling the deposition of metals. In conclusion, hydrothermal fluids are, expectably, mixtures in variable proportions of chemically modified meteoric waters and residual metamorphic solutions, having extracted their metal contents mainly from Palaeozoic metasediments. Accepting this work-hypothesis, it is important to discuss what are the most suitable physical and chemical conditions for copper leaching and transport, thus constraining some of the most important parameters that may rule the development of Cu-dominant mineralisation.

The most likely conditions of Cu transport in hydrothermal conditions can be roughly evaluated in function of T and fO_2 values, by changing iteratively the pH, aCl^- and $a(\Sigma S)$ of hydrothermal solutions and considering the most stable ionic Cu-complexes (namely $CuCl^0$, $CuCl_3^-$ and $Cu(HS)_2^-$). Since the Palaeozoic metasedimentary sequences in SW Iberia include abundant carbonaceous schists, a reduced ($SO_4/H_2S < 1$) environment for Cu leaching and transport is largely favoured. Additionally, the widespread evidence of feldspar hydrolysis points out to an acidic environment, thus suggesting that Cl-complexes would control copper solubility, being the relative predominance of $CuCl_3^-$ over $CuCl^0$ a direct consequence of fluid salinity increase. From this it may be concluded that leaching and transport of Cu will be very significant at *ca.* 325-375ºC for $fO_2<10^{-40}$, if pH = 4, $aCl^- \approx 3$ and $a(\Sigma S) = 0.001$. Under these chemical conditions, other metals (namely Zn, Pb, Ag and Au) can also be efficiently carried in hydrothermal solutions, provided that they exist also in metasediments (as suggested by some scarce analytical data concerning the Silurian schists at Miguel Vacas). What will be then the preferred conditions to promote the instability of Cu-chloride complexes, leading to copper deposition as *cpy*?

In these locations, the deposition of *cpy* usually postdates *py* development, although post-*cpy* pyrite associated to minor amounts of *sph* and/or *td-tn* is also known; the deposition of *po*, when recorded, is typically an early process; the growth of *gn*, on the contrary, is very restricted and seems to take place lately. On the basis of these observations and the afore-mentioned hypothetical characteristics of hydrothermal solutions, one may conclude that *cpy* deposition will be strongly favoured at temperature conditions of *ca.* 260-300ºC under $10^{-33} < fO_2 < 10^{-28}$, for a wide range of fluid salinities ($2 \leq$ wt% NaCl ≤ 25). In other words, *cpy* precipitation can be envisaged as a result of local temperature decrease and/or H_2S concentration increase and/or salinity depletion and/or reduction and/or pH rise. Many of these processes can easily be attained if local system depressurisation, fluid mixing and/or common fluid/rock interactions are considered, all of them geologically plausible and compatible with faulting during crustal uplift. For that narrow temperature range (*ca.* 260-300ºC), Zn and Pb will remain mostly in solution, thus explaining the scarcity of *sph* and *gn* in the lodes, as well as their late deposition under lower T conditions. In this scenario, very small amounts of Au and Ag are expected to precipitate also.

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