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The structure and mineralogy of the mine

by

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Introduction

Archaeological excavations have revealed prehistoric opencast copper mining at the so-called "La Turquesa Mine" with small, irregular shafts dug by hand with the help of mining artefacts made of porphyritic igneous rocks. However, in recent periods (possibly in the 19th or 20th centuries), the same area was mined using different methods and technologies. These most recent workings include a gallery dug nearby the path that leads to Mas de les Moreres and another gallery by the roadside, just where the Porrera road crosses the Cornudella del Montsant road, and finally a rectangular shaft dug in the area of the prehistoric shafts, which damaged part of the ancient workings.

The objective of this paper is to study the mineralogy of the mined ores and their geological setting.

Methodology

Based on pre-existing cartography and by taking geological sections of the structure, a representative sampling of the mineralised zones and the host rock was performed. The samples were sawn and polished sections for study with reflected light optical microscopy and thin sections for study with transmitted light microscopy were prepared. In addition, the mineral composition of the most complex samples was determined with X-ray powder diffraction technique (XRD) at CTT of the University of Barcelona.

Geographical context

As indicated in the previous chapter, the outcrops of the La Turquesa Mine are in the municipality of Cornudella del Montsant, some 3 km to the SSW of the town, in the central part of Priorat county. Various trenches and shafts were dug on the top of a hill near Mas de les Moreres. A short exploratory gallery to seek out the same mineralisation at a deeper level was cut to the south of the hill and there is another gallery on a hill to the west, on the other side of the Arbolí River (Fig.18).

Geological context

The study zone is in the south-western sector of the Pre-Litoral Range, which contains the most southerly outcrops of Palaeozoic rocks in the Catalan Coastal Ranges. This zone presents a highly complex structure and contains outcrops of some of the oldest rocks in the Catalan Coastal Ranges (Fig. 19).

In our study zone there are outcrops of sedimentary materials from the Upper Devonian and the Carboniferous (the upper part of the Palaeozoic). These series were deformed by the Hercynian orogeny and affected by contact metamorphism produced by granite intrusions. The Mesozoic series unconformably overlies the above materials and is in turn covered, also in a discordant manner, by detrital series from the Tertiary Period. The ensemble had been compartmented by faults from the Alpine Period. We will

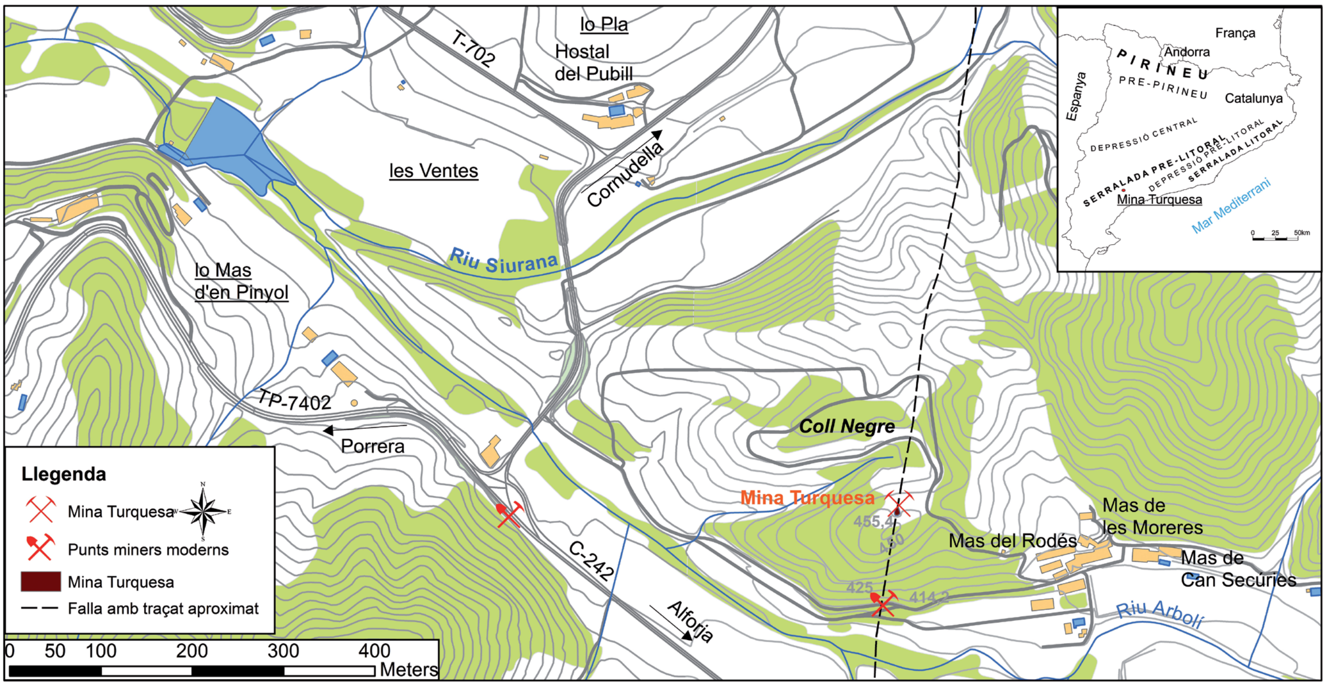

Figure 18. Map of the geographic situation of La Turquesa mine indicating the mineralised fault zone and the relation to other mines in the surrounding area.

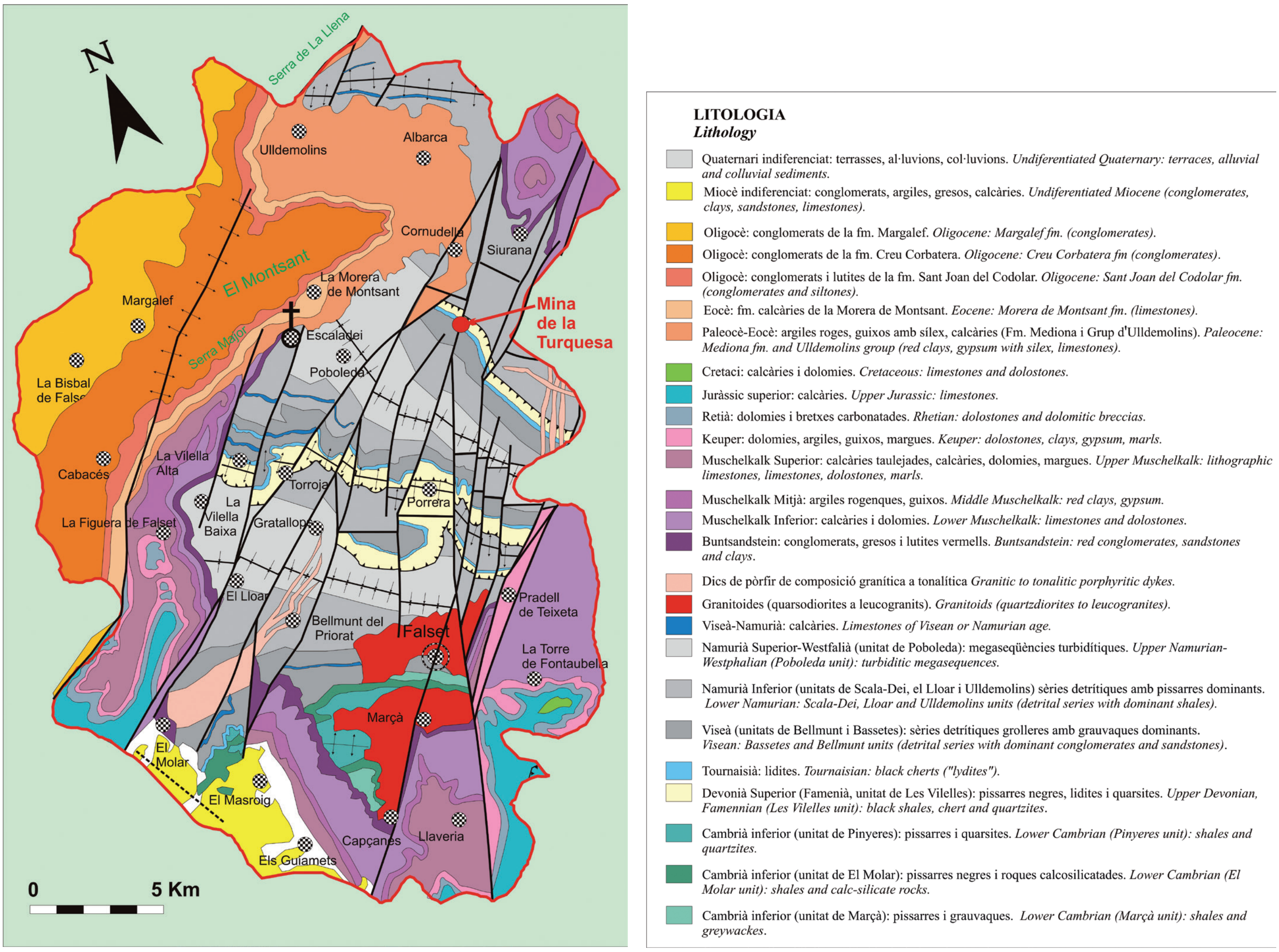

Figure 19. Geology of the Priorat and the location of La Turquesa mine (based on Melgarejo 1987).

describe each of these elements in broad strokes, but we will focus more on those that are directly related to the mineralisation, such as the hosting rocks or faults linked to them.

The Mid-Upper Devonian Series

The Upper Devonian materials in the Priorat are series of black slates (Raymond and Caridroit 1993). Their dark colouring is because they are rich in organic matter as the result of a global reducing phenomenon (Moreno *et al*. 2018), and, in fact, they can contain beds with petroleum and abundant disseminations of sulphurs, mainly of very fine-grain pyrite. Thin beds (with millimetric to centimetric thickness) of dark chert and quartzites are interbedded with the black shales. Small, white-coloured phosphatised levels of millimetric thickness may also occur interbedded with the former; these phosphates are composed of cryptocrystalline aggregates of minerals from the apatite group. All these materials are generally highly tectonized and can be dozens of metres in thickness. They crop out in the study zone in a NW-SE trending belt from Mas de les Moreres to Coll Negre; it can also be followed on the other side of the Siurana River. They have been dated from the Eifelian to the Famennian by their microfossil content (pollen) (González *et al.* 2015).

The Carboniferous Series

In broad strokes, they consist of a basal unit of chert, unconformably covered by thick coarse detrital series (Melgarejo 1992).

The Tournaisian cherts ("lidites")

The base of the Carboniferous series of this zone is made up by a thick packet of multicoloured cherts from the Tournaisian Period (Lower Carboniferous) up to 10 m thick. Where it is possible to examine the contact of these cherts on the majority of the Pre-Hercynian materials, they are generally found to be in angular unconformity with the older series. However, when they are situated above the Upper Devonian series, as in Cornudella, it can be seen that the contact is progressive. These cherts are a very common sedimentary facies throughout Western Europe in this period, and they can be considered a guide level. Due to their cryptocrystalline texture and siliceous constitution, they have a conchoidal fracture, a waxy shine and are very hard. They are generally dark in colour due to their high content of organic material, except where they have been subjected to a high degree of contact metamorphism and recrystallisation, in which case they take on a lighter colour because the organic matter has been destroyed. The chert units are made up by centimeter-thick beds and crop out precisely on the hill where the mineralisation is located. The thin section study of the cherts showed that they are made up of cryptocrystalline quartz; moreover, it is possible to observe in them abundant radiolarian and conodont remains. The strong internal folding of the cherts that can be observed in some packets near the mine can be attributed to underwater slumping processes, as has been described in other parts of the Priorat (Scherer 1969).

Elsewhere, towards the base of the packet of cherts, in various places in the Priorat, there may be a bed of manganese carbonates that, due to the effect of low- to mid-degree metamorphism induced by contact metamorphism, can produce very varied manganese silicate mineralisations accompanied by a complex paragenesis of abundant, very fine-grain sized metallic minerals (with pyrite, chalcopyrite, Ni-Co arsenides, selenides and tellurides, molybdenite, W minerals, etc). These mineralisations appear in the south of the Priorat at the Serrana mine nearby to El Molar (Melgarejo 1987, Melgarejo and Ayora 1994, Escusa and Melgarejo 1998), as well as in La Selva del Camp in Baix Camp county (Melgarejo 1987). This manganese packet and the complex associated paragenesis have not been located in this zone. However, a disseminated mineralisation of sulphides, mainly consisting of framboidal pyrite, occurs in the cherts of the Cornudella area. Pyrite occurs as spherical bodies just a few dozen microns in diameter, composed in turn of small microspheres a few microns in diameter, that tend to recrystallise into cubic shapes.

In addition, the beds of chert in the Priorat, as in most of Western Europe, tend to present millimetric levels of cryptocrystalline apatite, often accompanied by centimetric-sized phosphate nodules, as is the case of some levels in the cherts of the mine itself. However, none of these indices has ever presented worldwide a sufficient quantity of phosphate to make exploration or exploitation.

The detrital Carboniferous series

A deep packet of coarse detrital material was deposited over the cherts. It is mainly made up of coarse greywackes more than 400 m thick. The age of this packet is Visean (Low Carboniferous; Anadón *et al.* 1983 and 1985a, Sáez and Anadón 1989, Melgarejo 1987 and 1992).

In turn, above this unit, there is another some 400 m thick of finer materials, essentially slates. They are mainly the typical greenish slates of the Priorat of Namurian Age (Mid-Carboniferous; Anadón *et al.* 1983 and 1985a, Sáez and Anadón 1989, Melgarejo 1987 and 1992). The series continues with a thick packet of alternating sandstones and slates with a turbiditic affinity of Westphalian age, (Upper Carboniferous) organised into mega-sequences of more than 1500 m thick (Anadón *et al.* 1983 and 1985a, Sáez and Anadón 1989, Melgarejo 1987 and 1992, Maestro-Maideu *et al.* 1998).

Hercynian tectonomagmatic episodes

The ensemble of Palaeozoic materials is folded by the Hercynian orogeny producing in this sector large-radius NW-SE trending folds, with the SW flank vertical and even inverted. Detachment in the flanks of these folds may produce thrusting. There are episodes of folding with very similar axes but with inverse vergence; the first has a SW vergence and the second is NW (Melgarejo 1987 and 1992).

The regional metamorphism associated with the Hercynian deformation is of a very low degree and the schistosity is almost always very little developed, it can only be appreciated in silty materials on the flanks of the large folds (Melgarejo 1987, Valenzuela 2005 and 2016). In contrast, the materials are affected by the contact metamorphism caused by the intrusion of the granitoids of the late-Hercynian pluton of L'Alforja (Enrique 1990), although in this sector, distanced from the intrusion, the degree of contact metamorphism is also low and the recrystallisation is hard to see with the naked eye.

Some granitic porphyry dykes associated with granitic pluton cross the Palaeozoic series, especially towards the east of the study area.

The pre-orogenic state of the Alpine cycle: the Mesozoic megasequence

The alpine cycle began with an erosion that formed an immense peneplain with rubefaction, over which the Mesozoic series was deposited in angular unconformity. These materials do not crop out in the area studied, but can be subdivided into a set of depositional sequences separated from each other by lower-order unconformities or discontinuities. The Mesozoic series of the Prades mountains roughly consists of a Trias in complete Germanic facies (Calvet 1986), with a Buntsandstein of reddish conglomerates, sandstones and clays (up to 50 m deep), followed by two sections of Muschelkalk limestones and dolostones, each with a similar thickness; at the top of the Triassic sequence separated by a clayey and evaporitic section of a similar thickness as above at the top of the Triasic sequence the Keuper facies are made up by multicolour clays, evaporates and finally dolomites, and have also a decametric tickness. In addition, although outside the studied areas of the Priorat, there are much thick series, essentially carbonated, from the Jurassic and the Cretaceous.

The synorogenic state of the Alpine cycle (Palaeogene)

The materials of the Paleogene period in the Priorat consist of continental detrital series from the Palaeocene to the Oligocene (Barnolas *et al.* 1987). The Palaeocene consists of series with gypsums and red clays with flint nodules; they only crop out in the north of the Priorat and above them there are Eocene materials of the Cornudella Group, with 300 m of red lutites, carbonates and gypsums at the base, evolving towards the top to limestones and lutites (Colombo *et al.* 1995). These series can also be deposited discordantly over the Palaeozoic.

Above the aforementioned materials, there are Oligocene materials from the Escaladei Group, which essentially comprise calcareous conglomerates interbedded with lutites (Pérez Lacunza and Colombo 2001).

The compressive alpine structures

A compressive episode occurred during the Eocene-Oligocene in the western Mediterranean. In the Catalan Coastal Mountain Ranges this episode reactivated the old late-Hercynian or Mesozoic faults as strike-slip faults and new ones also formed. Some of the larger faults cross the study area and continue for dozens of kilometres and produce hectometric displacements. One system runs approximately NNE-SSW and NE-SW (such as the Porrera-Cornudella fault) and other smaller ones run in a NW-SE and WNW-ESE direction (such as the Coll d'Alforja fault). These faults are responsible for the elevation of the Catalan Coastal Ranges with respect to the Ebro Depression (Anadón *et al.* 1979, Teixell 1986 and 1988). On the edges of the Ebro Depression the synsedimentary activity of the faults favoured the formation of progressive unconformities (Anadón *et al.* 1979 and 1985b, Colombo and Vergés 1992).

The most important fault in the study area is that of Cornudella, accompanied by the those following the Siurana River and the Argentera ravine, which places the Tertiary sedimentary materials into contact with the metasedimentary series of the Carboniferous; finally, the Coll d'Alforja fault produdes the tectonic contact between the Late Hercynian granitoids with the sedimentary materials of the Mesozoic.

The alpine faults, as in the previous examples, allowed the circulation of hydrothermal fluids, thus enabling the formation of hydrothermal deposits.

The late-Orogenic state of the Alpine cycle (Neogene)

During this stage there was a generalised extension that in some sectors of the Catalan Mountains —the northernmost part— would be accompanied by vulcanism. In response to this extension, many of the pre-existing faults were reactivated, in this case as normal faults. These faults caused large tectonic grabens across the Catalan Coastal Ranges (Guimerà 1988), which gradually filled with detrital materials produced after the consequent erosion of the adjoining horsts. The nearby Mora Depression, for example, is interpreted as a Miocene era graben, with a fill of detrital materials. Likewise, these materials do not crop out in the central Priorat.

The section excavated by the Quaternary fluvial network developed sediments in the form of terraces and other alluvial materials, at the same time as sloping colluvial deposits were also produced in slopes.

The structure of the mineralisation

La Turquesa mine exploited a subvertical vein system that formed over the satellite faults of the Cornudella fault that run approximately NNE-SSW. They are in reality veinlets with a complex structure. The veinlets host into the Tournaisian decametric-depth chert packet. These rigid and very compact silicic materials demonstrated a very fragile behaviour during the Alpine fracturing processes. Therefore, rather than a vein in the strict sense, it is a fault zone with a principal vein of very variable width, from few centimeters to some decimeters, and a series of randomly oriented small veinlets that cement the brecciated zones. The fragile deformation was very

Figure 20. Aerial view of the mined area in the Carboniferous cherts. Of particular note is the presence of an extensive, highly fractured mineralised zone many veinlets cutting the banded cherts. The mineralisation was explored with trenches (in the right of the picture) and mined through shafts.

Figure 21. Part of the previous picture in which we can see the greenish patina of minerals formed by the supergenic alteration of the sulphurs.

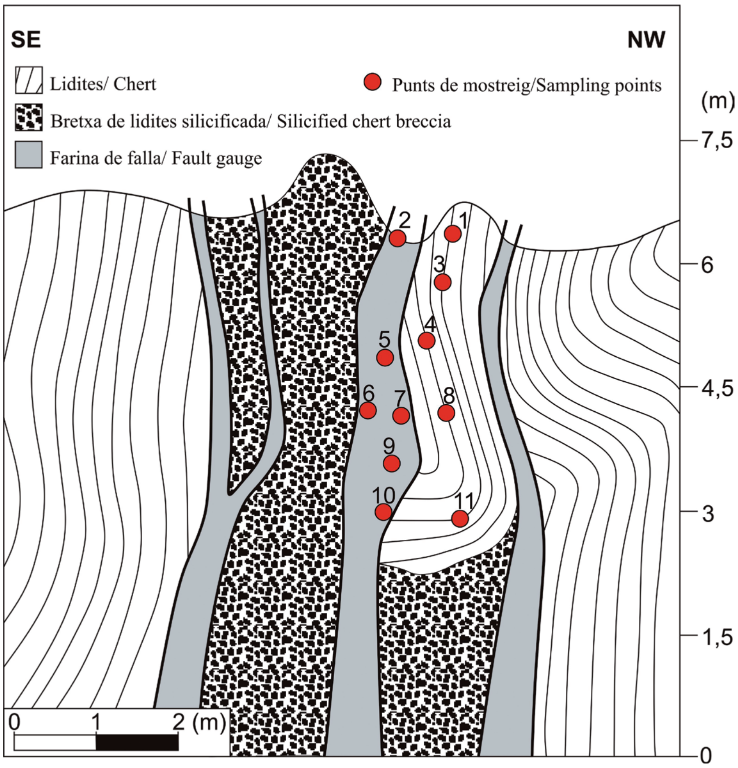

Figure 22. A section of the vein mineralisation from La Turquesa mine, with the locations of the sampling points.

intense (Figs. 20 and 21) and therefore developed into wide fault gauge bands (Fig. 22). The whole mineralised zone reaches a width of 5 metres. Altogether it determines a very large mineralised zone that crops out extensively. Therefore, and due to its proximity to the topographic surface, a notable zone of gossan and supergenic enrichment developed.

Given the size of the outcrop and the oxidised zone, it is not strange that the mineralisation was easy to find on the surface, even in prehistoric times. Initially work was carried out on the surface in the form of shafts and trenches to recognise and exploit the ores. Subsequently, galleries were dug into the base of the hill to reach the primary mineralisation at depth (these, in much more recent times, in the 19th century). These last operations did not meet expectations and were soon abandoned.

At the same time, the hydrothermal fluids that circulated there brought associated silicifications of the encasing rock, which caused the recrystallisation of the cherts.

Mineralogy and textures

As previously indicated, the next three parts are distinguished in the mineralisation, from the deepest part to the surface: the primary mineralisation, supergene enrichment zones and gossan zones (Fig. 22).

Primary mineralisation

It is possible to recognise two types of primary mineralisation. A first type consists of fine disseminations of pyrite (FeS_2) with a framboidal texture dispersed in the cherts. These framboids are spheroidal bodies of less than 25 microns in diameter, in turn composed of small pyrite spherules of a few microns in diameter (Pyrite 1, Fig. 23A). It has often been suggested that they are of bacterial origin, which would fit in with the fact that they are found in cherts that would originally have contained abundant organic material. In any case, this generation of pyrite would have been formed through processes contemporary with the sedimentation or with early diagenesis.

Where it can be recognised, the primary ore mineralisation in the veins consists of pyrite and chalcopyrite, with a style very similar to that observed in the mines of the Barranc Fondo ravine in Cornudella, to the east of these. At La Turquesa mine the primary minerals are of fine grain (less than 1 mm in diameter) and consist of idiomorphic pyrite (FeS_2, in form of cubes) and allotriomorphic chalcopyrite ($FeCuS_2$) dispersed between microcrystalline milky quartz (Fig. 23A). We have not observed geodic cavities in the mineralisation. Pyrite 2 often replaces pyrite 1 (Fig. 23A). This generation would have formed in association with the alpine fracturing processes.

Supergene enrichment mineralisation

Supergene enrichment mineralisation is formed by the replacement of the earlier one below the phreatic level. It consists of pseudomorphosis of the primary copper sulphides by grey-coloured chalcocite (CuS_2) with a metallic luster on hand sample (Fig. 23B and 23C). The degree of primary sulphide replacement is very variable and diminishes at depth until it finally disappears.

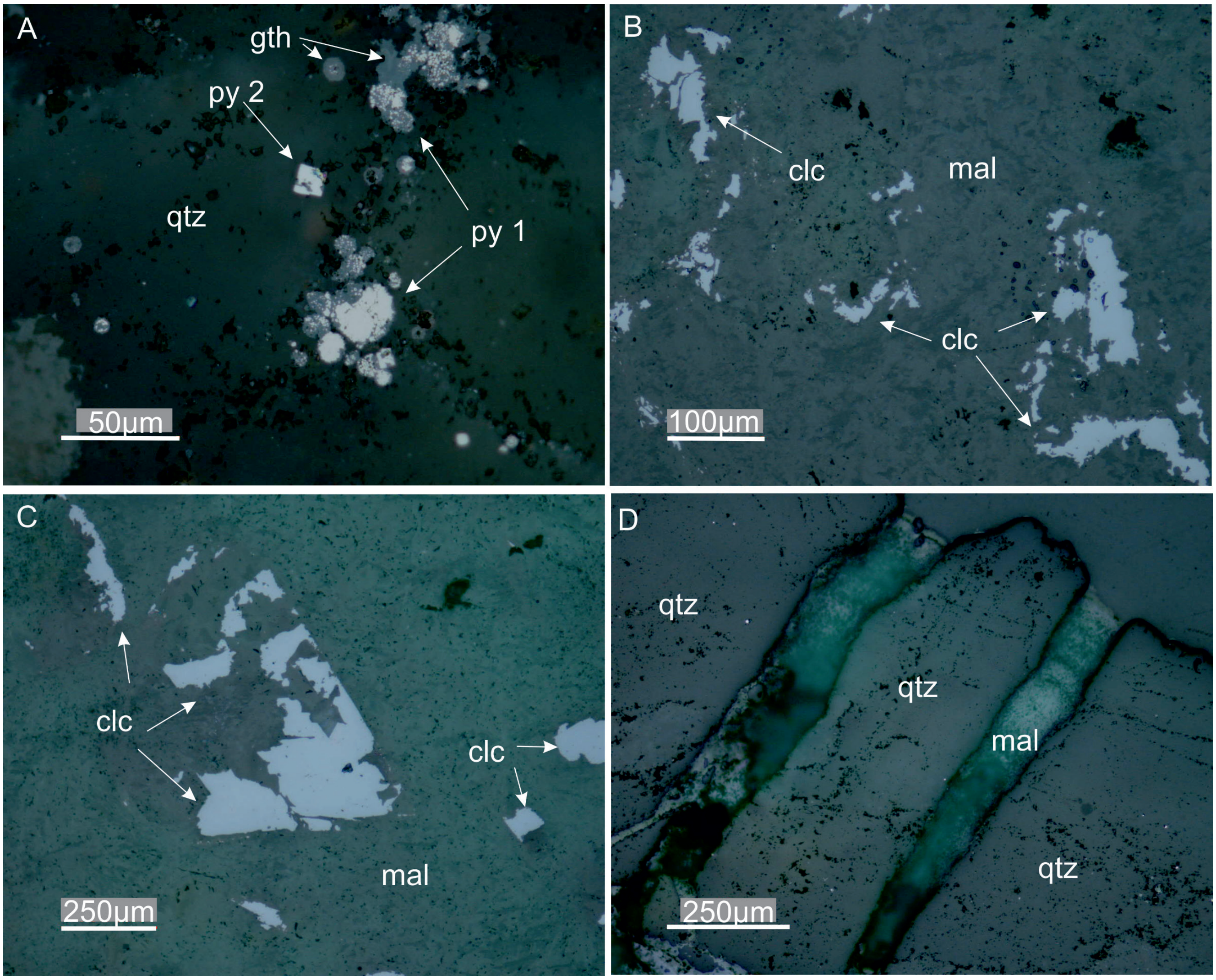

Figure 23. Reflected-light optical microscopy images without analyser. A, primary pyrite framboids from the first generation (py1) dispersed among the quartz (qtz) of the chert, replaced by second generation pyrite (py2); both generations of pyrite have been replaced by goethite (gth). B and C, details of the supergenic enrichment mineralisation, with chalcocite (clc) partially replaced by malachite (mal). D, Malachite (mal) and goethite (gth) veins among the quartz of the chert.

Gossan

The mineralisation in the form of gossan is the most spectacular in the deposit. Even on the surface it is possible to see outcrops of the vein with well-developed alteration, which implies the replacement of the sulphides by oxidised paragenesis. These minerals normally form growths in the form of crusts of millimetric thickness that fill various types of porosity, but above all that produced by the dissolution of the sulphides, which can also fill small fractures and geodes. Seldom they also form textures of the boxwork type. It is not possible to identify their crystalline forms with the naked eye because most of the minerals are cryptocrystalline and have a waxy or dull sheen in the hand sample. The minerals that have been determined there are cupriferous crandallite (in the highest part of the deposit), goethite, malachite (Fig. 23D) and azurite (rare and in contact with the primary sulphides).

Crandallite ($CaAl_3(PO_4)(PO_3OH)(OH)_6$) is the most common mineral in the superficial zone of the deposit. It forms fine crusts with very little compactness, of a pale blue to greenish-blue colour, with a dull shine, that easily crumbles and is hosted between the bleached cherts. Under the transmitted light microscope it is very little crystalline, with first order interference colours. The anomalous blue colour of the mineral is due to the presence of small quantities of copper, possibly in the position of the Ca.

Malachite ($Cu_2CO_3(OH)_2$) is the most abundant secondary mineral at a depth of 2-4 metres. It forms fine crusts of a characteristic green colour. Its identification has been confirmed by X-ray diffraction analysis.

The goethite (FeO(OH)) forms brownish crusts or yellowish patinas on the cherts.

In very small quantities and on the limit of the X-ray diffraction detection method, small quantities of other minerals have been detected in the crandallite crusts. These include minerals of the alunite group, including alunite and jarosite. These minerals are found associated with altered fragments of slates from the Carboniferous or the Devonian. They are cryptocrystalline and white (alunite) or ochre (jarosite). This latter mineral is easily confused with goethite, given that it is also formed by the supergenic alteration of pyrite and is associated with this other mineral. In much smaller proportions, there are other phosphates in the higher zones. Members of the goyazite-gorceixite series ($(SrAl_3(PO_4)(PO_3OH)(OH)_6$-$BaAl_3(PO_4)(PO_3OH)(OH)_6$) and pseudomalachite ($(Cu_5(PO_4)_2(OH)_4$), a mineral with a green-colour very similar to that of the malachite), have been identified by X-ray diffraction, as well as corkite ($PbFe_3^{3+}(SO_4)(PO_4)(OH)_6$), that may have formed due to the alteration of galena. In addition, there are small quantities of fluorapatite ($Ca_5PO_4)_3F$).

Discussion

The mineralisation of La Turquesa mine could seem to be anomalous due to the presence of crandallite in high quantities and the fact that this mineral is blue, if we take into account that it should normally be colourless given its original composition. Likewise, the presence of blue crandallite, which looks like turquoise, is not an isolated case in various copper mineralisation alteration zones in Catalonia. In fact, it has been found in great abundance in some outcrops adjacent to the Neolithic variscite mines of Gavà, forming veinlets of centimetric thickness with a pale blue colour. In this case, they were formed by simultaneous supergenic alteration of pyrite, chalcopyrite and apatite dispersed among the slates of the Silurian (Camprubí *et al.* 1994, Costa *et al.* 1994, Camprubí *et al.* 2003).

A similar situation can be seen at the Cornudella mine. The slates and cherts of the Devonian and the cherts of the Carboniferous have small levels of apatite, along with disseminations of framboidal pyrite of a very fine grain and, therefore, chemically highly reactive. There is also abundant pyrite among the primary veins into the vein mineralisation. Therefore, during the weathering processes, acidic fluids are formed by the oxidation of the pyrite. This oxidation, in addition to forming goethite, can also determine the formation of highly oxidising and acidic descending fluids. These fluids have a high capacity to react with the minerals in their path, such as the phyllosilicates of the slates, the apatite of the slates and the cherts, and the sulphides. Therefore, anion phosphate could be added to the solutions, as well as anion sulphate. This meant that phosphates could form in the meteorisation zones, thanks to the availability of phosphorus. On the other hand, aluminium is a highly immobile element in most supergene environments. Given that the fluids interact with slates, which are rich in micas and therefore in aluminium, this aluminium removal can explain the formation of sulphates and phosphates with aluminium in the highest parts of the deposit. This would explain the formation of the alunite group minerals and crandallite in the zones nearer the surface; the presence of corkite or goyazite-gorceixite series minerals can be explained by the same phenomenon. Copper can become fixed in the crandallite structure during this process, because crandallite has various positions suitable for retaining elements of very diverse ionic radiuses and oxidation. The presence of corkite can be explained by the same mechanism.

Special mention should be made of the existence of malachite, especially if we take into account that the copper mineralisation is hosted in quartz and not in carbonates. Therefore, the source of the carbon must be atmospheric. The fact that there is no formation of sulphates such as brochantite or antlerite can be explained by the fact that anion sulphate is not so abundant, because the quantity of pyrite in the original ore is not very high, or because most of the sulphate remains fixed in the form of alunite or jarosite in the highest levels.

In reality, the proportion of copper in the crandallite is low and its exploitation, especially in prehistoric times, could have been complicated, although there is no data on its mining. It is possible that the exploitation would have focused more on the malachite, which increased in proportion at depths of just a few metres. This would explain the development of relatively deep shafts and not superficial trenches following the outcrop of the vein on the crest.

The mineralisation is very fractured due to the effect of the continuous activity of the faults and their weathering on the upper levels has favoured the development of a rock that is even more disposed to crumbling. Therefore, it would not have been difficult to mine using the tools available in prehistory. In contrast, the exploitation appears to come to a halt in the deeper levels where the primary sulphides begin to appear, suggesting that those minerals offered technical complications to the early metallurgists.

Conclusions

La Turquesa mine was opened to exploit the secondary copper ores. These ores are in the upper part of a vein zone with pyrite and primary chalcopyrite that, due to supergene alteration, results in a gossan with crandallite and malachite, accompanied by minerals of the alunite group, and a zone of supergenic enrichment with chalcocite. The indicated paragenesis has its remote origin in the simultaneous meteorisation of pyrite and apatite in the hosting rocks, on the one hand, and of the chalcopyrite veinlets, on the other.

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