Comment on "Berthierine and chamosite hydrothermal: genetic guides in the Peña Colorada magnetite-bearing ore deposit, Mexico"

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1. Introduction

Rivas-Sanchez *et al.* (2006) studied the composition and structure of berthierine and chamosite in the Peña Colorada magnetite deposit (Colima, Southwestern Mexico), which they interpreted genetically to represent (1) an early SEDEX stage of mineralization and (2) a late "hydrothermal" stage of mineralization. Their mineralogical study has some deficiencies that will be discussed. However, it is the deficiencies in their genetic interpretation that are our major concern. Their paper lacks any description of the regional geologic context and any real discussion on the origin of the deposits, which is surprising because there are several recent publications on both topics (Klemic, 1970; Zürcher *et al.*, 2001; Tritlla *et al.*, 2003; Corona-Esquivel and Henríquez, 2004). Rivas-Sanchez *et al.* (2006) do not even consider models for similar deposits in Mexico that are better undestood than Peña Colorada, like Cerro de Mercado (Swanson *et al.*, 1978; Lyons, 1988).

In this discussion we describe first the basic published information that the paper by Rivas-Sanchez *et al.* (2006) lacks. Then, we discuss what, to our knowledge, are inconsistencies and misconceptions in the paper. Finally, we suggest that a "classical" geologic approach to the Peña Colorada deposit provides a much stronger rationale than the mineralogical characterization of only two minerals to establish the general conditions for mineral deposition that lead to plausible genetic models.

2. Geological Setting

The basement of the sedimentary sequence at Peña Colorada contains highly deformed Triassic rocks (Centeno-García *et al.*, 1993). The Cretaceous sedimentary suite starts with the Alberca Fm. (Valanginian-Hauterivian), which comprises biopelites, marls, limestones, fine-grained sandstones, and andesitic tuffs (Centeno-García, 1994). It is conformably overlain by the Tecalitlán Fm. (Barremian-Aptian), which is comprised of andesitic flows, rhyolitic pyroclastic flows, sandstones and conglomerates. It is overlain in turn by the Tepalcatepec Fm. (Albian-Cenomanian),

which is ubiquitous in the study area and correlates stratigraphically with a regional series that contains peperites (Elena Centeno, personal communication). The Tepalcatepec Fm. hosts most of the magnetite ores in the Peña Colorada deposit. The base of this formation is comprised of marls, carbonaceous pelites, limolites, sandstones, evaporites, and occasional carbonate reefs. The middle part of the Tepalcatepec Fm. contains micritic marine carbonate rocks interbedded with limolites, and the upper part contains interbedded calc-alkaline andesitic tuffs, which are characteristic of a back-arc extensional environment (Centeno-García *et al.*, 1993; Zürcher *et al.*, 2001). The recrystallized carbonate rocks and interstratified andesitic tuff beds have been dated as Aptian-Albian by Corona-Esquivel and Alencaster (1995). These rocks are capped by reddish conglomerates of the Cerro de la Vieja Fm. (Cenomanian).

These Cretaceous strata were intruded by the calc-alkaline Manzanillo batholith (Schaaf *et al.*, 1995; Calmus *et al.*, 1999), which contains mostly granodiorites with minor plagiogranite and gabbro intrusions. At Peña Colorada, the sedimentary sequence is intruded by andesitic porphyry dikes that are probably Tertiary.

3. Structure of the Peña Colorada Iron Deposit

Peña Colorada is a complex polyphase deposit. Several magmatic and hydrothermal events produced iron mineralization, garnet-rich rocks (granatites) as skarns or skarnoids, and late dikes and faults that crosscut the mineralized bodies. The main mineralization events are: (a) a massive orebody, (b) a disseminated orebody, (c) a layered barren exoskarn/skarnoid, (d) a polymictic breccia, (e) mineralized conglomerates, and (f) late andesitic dikes (Zürcher *et al.*, 2001; Tritlla *et al.*, 2003).

The massive orebody is tabular and generally massive, up to 40 m thick, over 1000 m long, and nearly parallel to the local layering of host andesite flows and carbonate rocks. The SW part of this magnetite orebody is in contact with lutites and recrystallized limestones of the Tepalcatepec Fm. The contact is usually discordant and sharp, although it may occasionally contain granatites up to 10 cm thick within. The mineralogy of this orebody is dominated by subhedral to euhedral magnetite (>85% in volume), partially martitized, with subordinate pyrite, chalcopyrite,

pyrrhotite, K-feldspars, pyroxenes (basically hedenbergite), chlorite, apatite, and carbonates. Fragments of feldspathic rocks are found sporadically in this orebody that were apparently dragged from a pre-existing mineralization (Tritlla *et al.*, 2003). Ferroan chlorite is intimately associated with K-feldspar and the distribution of crystals in this particular association suggests that these minerals are pseudomorphs of pyroxenes and, notably, garnet. Potassic metasomatism has been reported in other allegedly Iron Oxide-Copper-Gold (IOCG) deposits such Candelaria in Chile (Marschik and Fontboté, 2001). The K-feldspar gives a K-Ar date of 57.3±2.1 Ma (Tritlla *et al.*, 2003).

The disseminated orebody, which is found ∼40 m below the massive ore body, is over 10 m thick, and its hanging-wall is subconcordant to the local layering of the host andesitic rocks. This orebody has reserves of >90 Mt grading 26% magnetite. It shows a laminated internal arrangement due to alternating K-feldspar, chlorite, and pyroxene layering. The groups of layers are a few mm to over 30 cm thick with coarse-grained magnetite-rich layers and euhedral pyrite crystals at their base. The pyrite content and size of magnetite crystals decrease upwards, as the content of fine-grained pyroxene increases. Laterally to these layered deposits are accumulations of poikilitic euhedral to anhedral non-oriented K-feldspar crystals (similar to a syenite) with subordinate euhedral magnetite crystals that form irregular bodies that extend for several meters. Such accumulations may represent portions of deeper bodies that were upthrown by faulting. Such a "syenite" was dated at 65.3±1.5 Ma (K-Ar in K-feldspar, Tritlla *et al.*, 2003).

The polymictic breccia is a subvertical zone of brecciation that cuts the massive and disseminated bodies (Tritlla *et al.*, 2003). The structure, morphology and mineralogy of the breccia are highly variable through its vertical extent of >300 m. This breccia is ∼5 m wide in its lower part and contains sharp-edged andesite blocks in a fine-grained magnetite matrix that resemble a breccia formed by hydraulic fracturing. Other fragments up to 1 m in diameter are made up of aggregates of cm-sized crystals of magnetite, apatite and hedenbergite. Tritlla *et al.* (2003) interpreted these fragments to be xenoliths that were dragged from a deeper magnetite body. There are similar mineral associations in other similar deposits in the region and in the Cerro de Mercado deposit (Lyons, 1988). In the central section of the Peña Colorada deposit the breccia is >10 m thick, >100 m long, and contains (1) massive magnetite blocks from the massive body, (2) fragments of underlying host aplites and andesites that are overgrown by coarse magnetite crystals, and (3) fragments from the disseminated body. An array of calcite and magnetite-bearing veinlets of hydrothermal origin are found in the polymictic breccia and extend into the upper part of the deposit. The partial replacement of conglomerate clasts on top of the volcanosedimentary rocks is thought to be coeval with the hydrothermal veinlets.

4. Relative Chronology of Stages of Mineralization

The events that led to the formation of the deposit can be grouped as follows (based on Zürcher *et al.*, 2001; Tritlla *et al.*, 2003): (1) intrusion of a microgranite that is found only in a few drill core samples in the deepest parts of the deposit; it is unclear whether it was the first mineralization event; (2) barren stratabound skarn or skarnoid, not in contact with the microgranite; (3) disseminated orebody; the rock rich in K-feldspar and magnetite in fault contact with this body may be older than it; (4) massive orebody (may be coeval with the disseminated body); it contains granatite fragments replaced by K-feldspar that were probably dragged from the barren stratabound skarnoid; (5) polymictic breccia and associated veins and veinlets; (6) intrusion of andesitic dikes with associated small barren skarn-like associations of garnets and pyroxenes.

The time span between the ages obtained by Tritlla *et al.* (2003) is, accounting for the standard deviations, at least 4.4 M.y. This duration is too long for the whole deposit to be a typical skarn, as proposed by Zürcher *et al.* (2001). Consequently, Tritlla *et al.* (2003) proposed that this deposit is a Phanerozoic equivalent of IOCG-type deposits.

5. Inconsistencies, Misconceptions and Misleads in Rivas-Sanchez *et al.* (2006)

5.1 Why SEDEX?

Rivas-Sanchez *et al.* (2006) propose a sedimentary-exhalative (SEDEX) model for the formation of the Peña Colorada magnetite deposits, but offer no evidence to support such a model. The high-grade *diagenetic origin* (sic) of some mineral associations is equally dogmatic and unsustained. These authors assume the SEDEX option but they support it with neither solid consistent geological data nor citations of other authors. To date this deposit has been explained as a skarn (Zürcher *et al.*, 2001) or as an orthomagmatic or IOCG-like deposit (Klemic, 1970; Tritlla *et al.*, 2003; Corona-Esquivel and Henríquez, 2004). Conversely, SEDEX models have been disregarded as inconsistent with the vast majority of the geologic evidence.

Rivas-Sanchez *et al.* (2006) do not describe the local and regional geology, although it is a key factor for understanding the genesis of any deposit. Accordingly, they fail to describe sedimentary sequences that are characteristic of SEDEX environments such as turbidites and marine clastic sequences deposited in intracontinental rift basins or in passive continental margins (e.g., Russell *et al.*, 1981; Large, 1983), which, by the way, are virtually absent in the region. The sedimentary control is extremely important in the formation of SEDEX deposits, unlike IOCG or volcanogenic massive sulfide (VMS) deposits. Further, these authors did not provide any stratigraphic evidence from the local basins that would have helped to establish a favorable setting for a SEDEX model. Although carbonate rocks are not uncommon in stratigraphic sequences that contain SEDEX deposits (e.g., Sangster, 1990), Rivas-Sanchez *et al.* (2006) fail to discuss the abundance of such rocks at Peña Colorada.

Most SEDEX deposits are unrelated to magmatism and thus, unlike VMS deposits, do not form in subduction settings. SEDEX deposits form in intracontinental rift basins or in passive continental margins, with associated synsedimentary faulting (e.g., Russell *et al.*, 1981; Large, 1983). Rivas-Sanchez *et al.* (2006) did not describe such a regional geologic context for the formation of SEDEX deposits in

Southwestern Mexico. Conversely, Schaaf *et al.* (1995) and Calmus *et al.* (1999) have described clearly a continental arc setting in the region when the iron deposits were formed. Further, Rivas-Sanchez *et al.* (2006) do not describe a geometry, a structure, textures, or rock or mineralization assemblages that are typical for SEDEX deposits such as tabular lenses, growth faults, massive sulfides, exhalites, *inhalites*, bedded ores, or vent complexes (Large, 1983; McClay, 1991; Sangster and Hillary, 2000). They also fail to explain why this allegedly SEDEX deposit consists mostly of magnetite rather than massive lead and zinc sulfides; why it is mainly found as a thick ore body with a low lateral extent vs stratigraphic thickness ratio; and why it does not consist of several cycles of exhalation and sedimentation that are typical of SEDEX deposits. Furthermore, the main orebody is not clearly stratiform but is only partially stratabound. There are indeed Fe- and Mn-rich SEDEX deposits, but the mineralogical, textural and structural characteristics of the Peña Colorada deposit do not compare in any way with them (see e.g. Lydon, 1996). Also, regretfully, the authors do not offer a physicochemical model that supports the deposition of magnetite in a relatively shallow marine environment (see Corona and Alencaster, 1995) that they favor. They do not explain why most magnetite crystals are subhedral to euhedral, no matter what the crystal size is, or why the most common crystal size in all mineralized bodies is in the order of millimeters and centimeters, not nanometers, given that this deposit was not affected by regional or contact metamorphism or by hydrothermal activity to a degree that could have induced the recrystallization of magnetite or other minerals.

In fact, the main support for a SEDEX model in Rivas-Sanchez *et al.* (2006) is that the chemical compositions of berthierine and chamosite (figures 14, 15 and 16 in their paper) fall into the "marine" field in the discriminatory diagrams of Damyanov and Vassileva (2001, in Rivas-Sanchez *et al.*, 2001) who used the compositional diagrams for several phyllosilicates to estimate geological conditions of formation (e.g., marine, non-marine, hydrothermal) for different mineral associations of a well-established SEDEX deposit that, contrary to Peña Colorada, has an ore mineralogy of siderite, barite and base-metal sulfides. However, what does "marine" mean? Marine depositional environments are not homogeneous and, moreover, they may involve very different sources and processes such as detrital, authigenic, early diagenetic or even hydrothermal. Thus, a "marine" berthierine does not necessarily imply a SEDEX deposit. On page 1399, Rivas-Sanchez *et al.* (2006) state that *"berthierine is in the marine zone, corresponding to a sedimentary exhalative hydrothermal process (SEDEX). Marine chimneys produced the hydrothermal Fe rich brines, in a marine clay floor of relatively shallow depth, provoking the simultaneous precipitation of intergranular berthierine and magnetite"*. They state further that (1) a "marine" composition supports a SEDEX model, (2) the hydrothermal discharge in the seafloor occurred through chimneys, that no one has ever described, and (3) the hydrothermal discharge occurred at a shallow depth, that is not estimated in any way. All three are highly speculative and unsubstantiated statements.

On page 1399, Rivas-Sanchez *et al.* (2006) state that *"the botryoidal texture of magnetite in berthierine shows the typical texture of deposition in a sedimentary exhalative environment (figure 17)"*. However, no botryoidal aggregates can be seen in figure 17. Also, botryoidal textures cannot be ascribed to any specific type of ore deposit: they occur in a very wide variety of environments under a wide range of physicochemical conditions. In addition, the authors fail to report the most usual textural type of magnetite crystals, which is certainly not botryoidal elsewhere in the deposit.

On page 1399, Rivas-Sanchez *et al.* (2006) apparently use the composition of chamosite and berthierine (their "octahedric mean") as geothermometers, yielding temperatures of $360°$ and $150°C$. However, they do not cite the source for such geothermometers. They also fail to discuss that $360°C$ is probably far too high a temperature for mineral precipitation at the sediment-seawater interface in SEDEX depositional environments.

As the final statement in their conclusions, Rivas-Sanchez *et al.* (2006) declare that the homogeneous distribution of *microparticles and nanoparticles* of magnetite within *amorphous* (sic) berthierine indicates that they formed in a sedimentary-exhalative environment. Why does the grain size of the magnetite particles indicate that they formed in such environment? The paper lacks any evidence or reference to previous studies that support such an interpretation, and thus we must conclude that the SEDEX model for the Peña Colorada deposit is exclusively based on a working hypothesis from an undisclosed source and that the mineralogic data collected by them is interpreted to fit only such an hypothesis.

5.2 Berthierine and chamosite, indicators for specific geologic environments?

Some minerals in nature are good indicators of very specific geologic settings, but berthierine and chamosite are not among them. It is true that these minerals have not been reported in many papers, but this does not necessarily mean that they are scarce in nature. Berthierine and chamosite cannot be easily differentiated by optical methods or even by more sophisticated analytical methods and so are often confused with each other, or even with other members of the Kaolinite-Serpentinite and the Chlorite groups to which they respectively belong. However, berthierine and chamosite have been effectively identified in many more places and geological environments than the "about 15 localities around the world" that Rivas-Sanchez *et al.* (2006) acknowledge. These authors identify SEDEX deposits, VMS deposits, metamorphic rocks, bauxites and laterites, ironstones, paleosoils, thrust zones, pegmatites, and coal deposits as for berthierine. According to Gaines *et al.* (1997), berthierine is generally *"found in unconsolidated estuarine and marine sediments in iron reduction zones and in arctic soils. Can be found in phosphatic or sideritic sedimentary rocks, also laterites or coal beds, rarely found in granitic pegmatite. In banded iron formations (...) also in fault breccias and in fluorite-pyrrhotite veins"*. Berthierine has been even found in the Mont Saint-Hilaire (Gaines *et al.*, 1997) and the Khibiny (www.mindat.org) alkaline complexes (!), as well as >50 other localities worldwide. Thus it is unlikely that berthierine is found only in very specific

environments, and it is risky to use berthierine for deducing environments of formation when basic geological criteria (e.g., stratigraphy, tectonics) are disregarded. This observation is no less true for chamosite. According to Gaines *et al*. (1997), chamosite *"formed in low-grade regional and contact-metamorphosed rocks, granite pegmatites, (...) as a hydrothermal alteration of wall rock around sulfide deposits; amygdules in volcanic rocks. Abundant in soils and marine clays. (...) in coal deposits. Found variously associated with* (several minerals). *(...) as a replacement of a wide variety of silicates, including garnet..."*. It may also be found in sedimentary iron deposits, in hydrothermally altered basic igneous rocks, in many metallic mineral associations, and in metamorphosed banded iron formations (Melgarejo, 1997). Recall that (1) some of the host rocks in the Peña Colorada deposit are submarine intermediate-basic volcanic rocks, (2) there is a conspicuous late hydrothermal breccia in the deposit, and (3) chlorite is part of a mineral assemblage that pseudomorphosed granatite (Tritlla *et al*., 2003). The bottom-line is that both berthierine and chamosite may be found in a wide variety of geologic environments, and their origin may not be discriminated with chemical diagrams when such environments lack detailed geological studies. On pages 1389 and 1390 Rivas-Sanchez *et al*. (2006) state that *"berthierine and chamosite are considered good indicators of the geologic processes and conditions under which this deposit was formed"*. Are they considered so by themselves or by the other authors cited by them? Further, why are berthierine and chamosite the only relevant indicators of the genesis in the deposit? Rivas-Sanchez *et al*. (2006) state that these minerals are important components in the Peña Colorada deposit. That is not correct because the most conspicuous minerals in the deposit are magnetite, hematite, apatite, K-feldspar, chlorite and sulfides.

We agree with the contention of Rivas-Sanchez *et al*. (2006) that the mineralogy of sheet silicates, no matter how difficult their determination is, should not be treated lightly in any mineralogical study, but the interpretation that they make from the berthierine and chamosite could hardly be farther from a systematic acquisition of relevant data.

5.3 Other mineralogical issues

Rivas-Sanchez *et al*. (2006) succeed in presenting apparently sound mineralogical data from berthierine and chamosite that are difficult to work with. Unfortunately, this virtue of their work has been masked by the clear intention of the authors to make a point about the genesis of the deposit through the use of these minerals. Nonetheless, there are some comments to be made about their mineralogical work.

The authors did not justify the use of "non-canonical" forms to describe the chemical formula of berthierine, $(Fe^{2+}, Fe^{3+}, Mg)_{2-3}(Si, Al)_2O_5(OH)_4$ (Blackburn and Dennen, 1997) or $Fe_2^{2+}Al(SiAl)O_5(OH)_4$ (Gaines *et al*., 1997), and chamosite, $(Fe^{2+}, Mg, Fe^{3+}, Al)_5Al(Si_3Al)O_{10}(OH, O)_8$ (Blackburn and Dennen, 1997) or $Fe_3^{2+}(Fe_2^{2+}Al)(Si_3AlO_{10})(OH)_8$ (Gaines *et al*., 1997). On page 1392, they state that berthierine belongs to the Chlorite Group. Gaines *et al*. (1997) and www.mindat.org think otherwise as they include this mineral in the Kaolinite-Serpentinite Group. Moreover, on page 1393, they talk about "kaolin-type chlorite", a non-existent category. After that, the authors continue to treat berthierine as a member of the Chlorite Group.

On page 1393, Rivas-Sanchez *et al*. (2006) describe sulfur as an opaque mineral, although native sulfur, which was not previously described, is obviously not opaque. On the same page, in the mineral chemistry section, they report a "$Ca2O_3$" (sic) content; the Ca oxide is always reported as CaO because the oxidation state of Ca is known to be 2 alone, not 3.

On pages 1394 and 1396, Rivas-Sanchez *et al*. (2006) note the occurrence of iron as Fe^{3+} and Fe^{2+} from Mössbauer spectroscopy in both berthierine and chamosite. However, in the calculations from analyses from Electron Probe Micro-Analysis (EPMA) of these minerals in tables 2 and 3, they considered FeO to be the only iron oxide, although values for Fe^{3+} and Fe^{2+} are indeed reported. H_2O is not considered in the oxides, and thus one can only guess how the hydroxide groups were calculated. Further, water weight was not considered in the total oxide sum, although it can attain $\sim$10 wt.% in such minerals. Thus, if we sum water to total oxide weights, we will obtain unacceptable total oxide percentages that are generally much greater than 101%. In addition, such total oxide percentages have been even more underestimated because the 50% additional O of Fe_2O_3 with respect to FeO was not considered in the sum. The calculation of the atoms per formula unit (*apfu*) also shows major mistakes. For instance, on page 1393, it is stated that the calculations of berthierine were performed on the basis of 28 oxygens. This is not possible for such calculations because it is necessary to consider a whole multiple of 9. Considering the values presented in table 2 of Rivas-Sanchez *et al*. (2006), the *apfu* calculations were probably based on 36 oxygens. Moreover, these authors state that they assumed *full site occupancy* (page 1393) to calculate the structural formula of berthierine, although it is usually non-stoichiometric. Surprisingly, also on page 1393, they discuss *vacant octahedral positions*. In addition, there are basic errors of presentation in the EPMA tables of Rivas-Sanchez *et al*. (2006). In their table 2 the values corresponding to elements below detection limit are expressed randomly as 0 or 0.000 or *n.d.* (not detected). For instance, some elements are initially considered as 0.00 (oxide, wt.%), and then *n.d.* (element, *apfu*). All these mistakes and inaccuracies undermine the credibility of the formula calculations that are the basis for the geological deductions stated by the authors.

At the beginning of their discussion (page 1396), Rivas-Sanchez *et al*. (2006) describe the mineral associations that accompany berthierine and chamosite. Some of the accompanying minerals are (1) "sulphide", in a context that suggests that it is a mineral, not a mineralogical class or group of minerals, and (2) "sulfurs". Such use of mineralogical terms is clearly inappropriate. The authors also report the occurrence of "clay minerals", which is surprising in a paper that apparently stands for an accurate characterization of sheet silicates.

5.4 Miscellaneous issues

Although figure 1 of Rivas-Sanchez *et al.* (2006) is suspiciously similar to figure 1 of Tritlla *et al.* (2003), the authors do not indicate that their map is "modified after Tritlla *et al.* (2003)", the customary practice in most papers. We can only wonder why the evidence presented by Zürcher *et al.* (2001), Tritlla *et al.* (2003) and other authors was not cited by Rivas-Sanchez *et al.* (2006), given that they used their data and diagrams.

Rivas-Sanchez *et al.* (2006) conclude that *"the first hydrothermal process (...) took place at the end of the Cretaceous epoch"* and that *"the berthierine precipitation (...) indicate the end of the hydrothermal phase* (sic), *possibly at the beginning of Tertiary"*. Such precise ages can be obtained only by absolute dating. However, the ages "suggested" by the authors sound correct to us because they coincide with the K-Ar ages of 65.3 ± 1.5 Ma and 57.3 ± 2.1 Ma that Tritlla *et al.* (2003) obtained, which correspond to *"the end of the Cretaceous epoch"* and *"the beginning of Tertiary"*, respectively. Sadly, Rivas-Sanchez *et al.* (2006) fail to explicitly refer to the paper by Tritlla *et al.* (2003) or to any other paper, and fail to provide any evidence, reason or analysis of their own to support their interpretation.

6. A Note of Caution

It is tempting to try to use a single mineral or a simple mineral association as an indicator of a geologic setting, environment, type of ore deposit, etc., but only a few minerals can do the trick, and usually then only in a given geological context (e.g. a metamorphic facies). It is irresponsible to ignore the regional geology, previously published work, and virtually all the other aspects that help to constrain the genesis of an ore deposit. It is no less irresponsible to propose a genetic model, SEDEX in this case, which is exclusively based on unsubstantiated speculation. We dare state this note of caution because Rivas-Sanchez *et al.* (2006) provide no geologic evidence to sustain their only explanation. Their contribution may be valuable for providing new knowledge of berthierine and chamosite, but it is worthless when trying to shed some light on the genesis of the Peña Colorada deposit.

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