Fluid Rock Interactions based on the Water Saturation Condition in Hypogene Zone within the Sar-Cheshmeh Porphyry Cu-Mo Deposit, Iran

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ABSTRACT

The Sar-Cheshmeh porphyry Cu-Mo deposit is located in Southeastern Iran (90 km. Southwestern Kerman). Four main vein Groups have been identified: I) quartz + molybdenite + anhydrite ± K-feldspar with minor pyrite, chalcopyrite and bornite; II) quartz + chalcopyrite + pyrite ± molybdenite ± Calcite; III) quartz + pyrite + calcite ± chalcopyrite ± anhydrite (gypsum) ± molybdenite; IV) quartz ± calcite ± gypsum ± pyrite ± Dolomite. Based on the previous works (e.g., fluid inclusions and mass transfer investigations), three different alteration types of potassic, phyllic and propylitic have been occurred within the deposit by varying the magmatic and meteoric fluids. Thermodynamic data are used to illustrate the stability fields of alteration and ore assemblages as a function of fO_2 and pH for the two alteration/mineralization stages of potassic and phyllic under the water saturation conditions. These calculations show that in the water saturation conditions (assumed condition for Sar-Cheshmeh deposit), at 450 °C, the fO_2 was between -26 and -20, and the pH >7, and that at 350 °C the corresponding values were -32.5 to -25, and >4, respectively. These calculations are consistent with observations that the bulk of the chalcopyrite deposited at Sar-Cheshmeh is hosted by veins formed at the approximate temperature of <350 °C which is situated in phyllic alteration zone. It is also suggested that other thermodynamic factors such as pH and fO_2 may reduce chalcopyrite solubility.

KEYWORDS

Porphyry, Copper, Sar-Cheshmeh, Thermodynamic, Rock interaction,

1. INTRODUCTION

Within the Sar-Cheshmeh porphyry deposit, the early hydrothermal alteration produced a central potassic assemblage (orthoclase-biotite), the propylitic alteration occurred contemporaneously with potassic alteration, but in the peripheral parts of the stock, and phyllic alteration occurred later, overprinting these earlier alteration events. The early hydrothermal fluids are represented by high temperature (350 °C to 520 °C), high salinity (up to 61 wt % NaCl equiv.) liquid-rich fluid inclusions, and high temperature (340 °C to 570 °C), low-salinity, vapor-rich inclusions [10]. These fluids are interpreted to represent an orthomagmatic fluid; the brines are interpreted to have caused potassic alteration, and deposition of Group I and II quartz veins containing molybdenite and chalcopyrite. Propylitic alteration is attributed to a liquid-rich, lower temperature (220 °C to 310 °C), Ca-rich, evolved meteoric fluid [10]. Influx of meteoric water into the central part of the system, and mixing with magmatic fluid produced deep albitization and shallow phyllic alteration. This influx also caused dissolution of early formed copper sulphides and remobilization of Cu into the sericitic zone where it was redeposited in response to a decrease in temperature (as the main zone of the copper deposition in Sar-Cheshmeh).

The purpose of this paper is to attempt to investigate the physical and chemical environment at the time of the formation of this deposit. The quantitative evaluation of the processes potentially responsible for the transport and deposition of copper is also provided. To aim this goal, the works of Brimhall [4] by incorporating experimental data on the solubility of copper in hydrothermal brines [9,10,20] have been applied.

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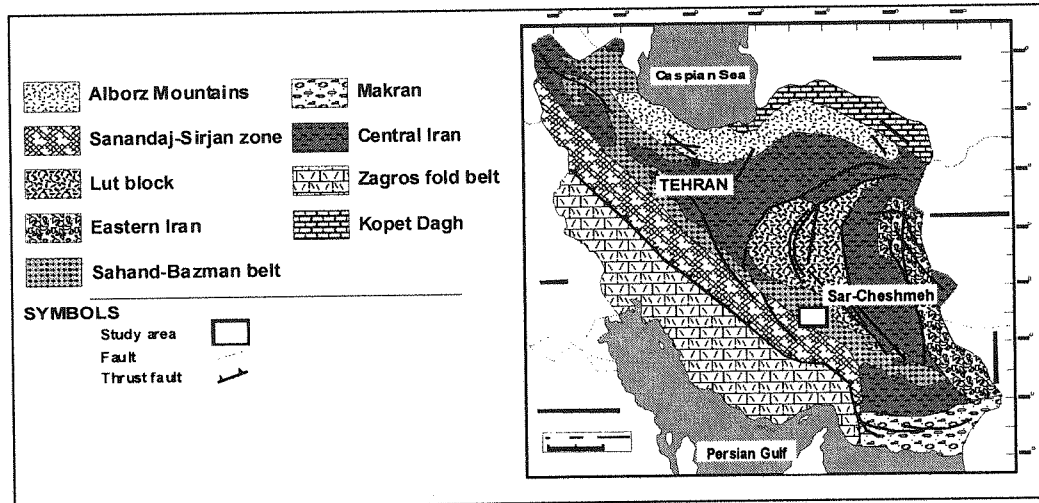
Figure 1: Simplified geological map of Iran showing the Sar-Cheshmeh area [12].

2. GEOLOGICAL SETTING

The Sar-Cheshmeh porphyry copper deposit is hosted by a diorite to granodiorite stock [18], located 67 km southwest of Kerman city in the Kerman province of southwestern Iran (Fig. 1). The stock is part of the Sahand-Bazman igneous and metallogenic belt [17] (Figs. 2, 3 and 4). The volcanics were laid down unconformably over folded and eroded Upper Cretaceous andesitic volcanic and sedimentary rocks. Subduction and subsequent continental collision during the Paleocene to Oligocene caused extensive alkaline and calc-alkaline volcanic and plutonic igneous activity [3,8,17], including intrusion of a porphyritic calc-alkaline stock at Sar-Cheshmeh during the Miocene.
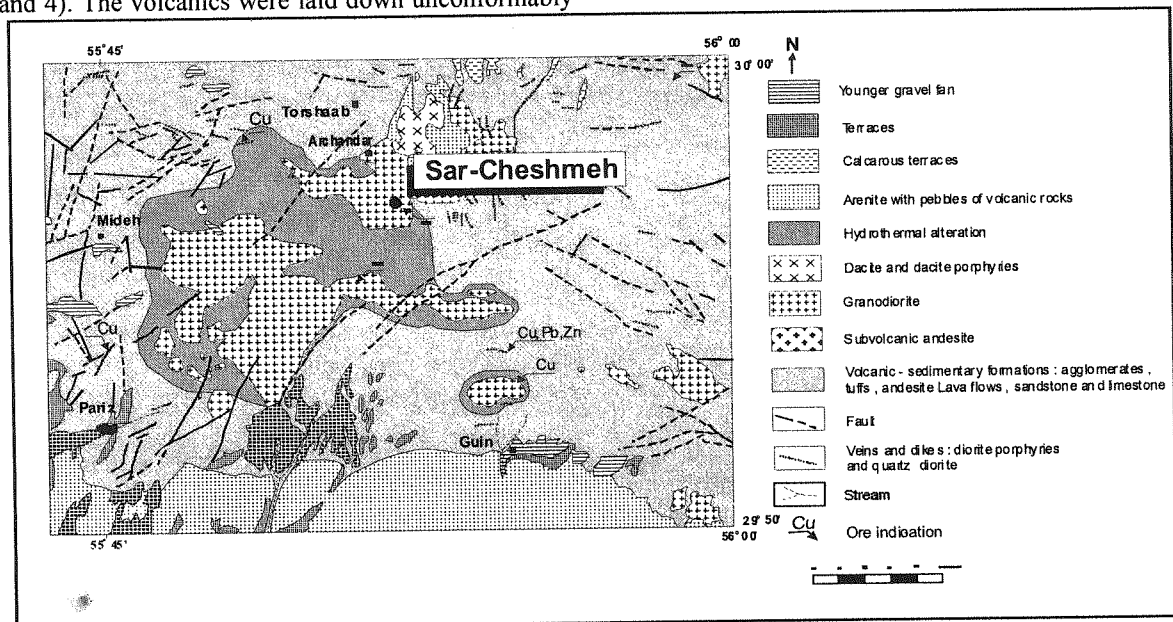

Figure 2: Detailed geological map of the Sar-Cheshmeh area showing the distribution of different igneous suites [15].

3. MINERALOGY OF THE VEIN SYSTEMS

On the basis of mineralogy and cross-cutting relationships, it is possible to distinguish at least four main Groups of veins representing four episodes of vein formation (Fig. 5): I) Group I veins consist of quartz + molybdenite + anhydrite ± K-feldspar with sporadic pyrite, chalcopyrite and bornite. These group veins are discontinuous, vary in thickness between 0.1 and 12 mm, and formed during the early fracturing of the porphyry stock [11,15]. II) Group II veins consist of quartz + chalcopyrite + pyrite ± molybdenite, generally cross-cut and in places off-set the Group I veins [11, 15]. III) Group III veins consist of quartz + pyrite + calcite ± anhydrite ± chalcopyrite ± gypsum ± molybdenite. Group III veins are most abundant in the phyllic alteration zone, cross-cut both Group I and II veins, and in some cases offset the earlier-formed veins [11]. Chalcopyrite is the only copper mineral, which is observed mainly as small blebs and inclusions in pyrite [15]. As it is discussed in [10], in

terms of fluid inclusions, the primary saline fluid inclusions in this Group veins are similar to those in Group II veins. The occurrence of chalcopyrite and other copper minerals in Group III veins suggests that they

could have been leached by later fluid circulating in these veins. Group IV veins consist of quartz, and/or calcite, and/or gypsum ± pyrite.
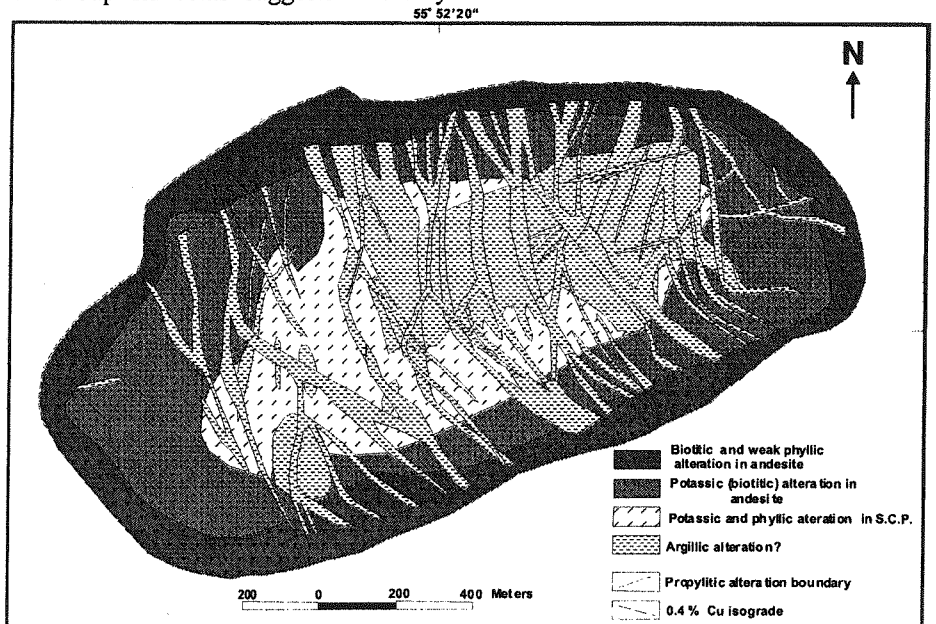

Figure 3: Detailed alteration map of the Sar-Cheshmeh deposit [18]. (S.C.P) stands for Sar-Cheshmeh Porphyry.
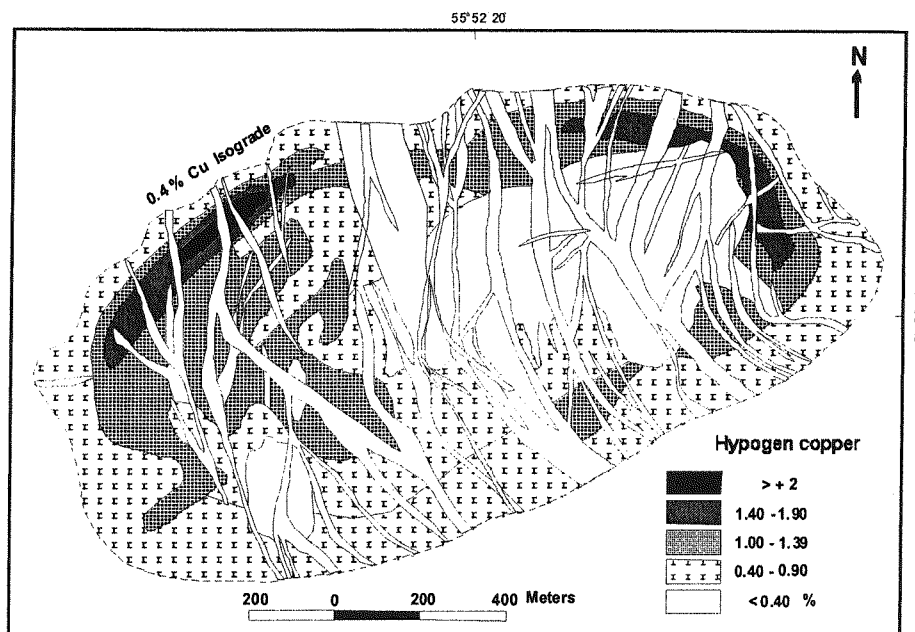

Figure 4: Copper mineralization map within the Sar-Cheshmeh stock [18].

4. ALTERATION AND MINERALIZATION

Hydrothermal alteration and mineralization at Sar-Cheshmeh are centered on the stock and were broadly synchronous with its emplacement (Figs. 2, 3 and 4). Early hydrothermal alteration was dominantly potassic and propylitic, and was followed by later phyllic, silicic and argillic alteration. Potassic alteration is represented by mineral assemblages developed pervasively and as halos around veins in the deep and central parts of the Sar-

Cheshmeh stock (Figs. 3 and 4). Potassic alteration is characterized by K-feldspar, irregularly shaped crystals of Mg-enriched biotite, and anhydrite [16, 19]. The hydrothermal biotite is generally interstitial to feldspar and quartz, and locally replaced hornblende and primary biotite phenocrysts [15]. Electron microprobe data indicate that some grains of potassium feldspar are rimmed by albite (An_{4-12}) suggesting a later sodic alteration event [15]. Propylitic alteration is pervasive and represented mainly by chloritization of primary and

secondary biotite and groundmass material in rocks peripheral to the central potassic zone. Epidote replaced plagioclase, but this alteration is less pervasive and intense than chloritization [10,15, 17].
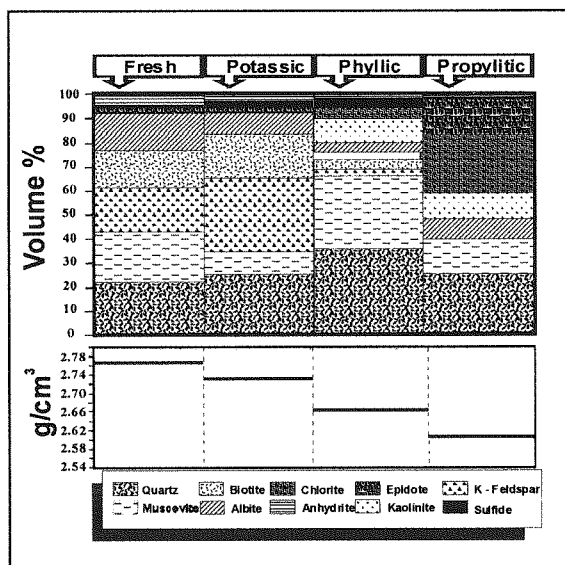

Figure 5: Relative mineral abundances in various alteration zones in the Sar-Cheshmeh deposit. Widths of bars denote qualitative abundances.

Hypogene copper mineralization was introduced during phyllic alteration and to a lesser extent during potassic alteration, and occures as disseminations and veinlets form. During potassic alteration, the copper was deposited as chalcopyrite and minor bornite; later hypogene copper was deposited mainly as chalcopyrite [13]. Hypogene molybdenite was concentrated mainly in the deep part of the stock, and is associated exclusively with potassic alteration, where it is found in quartz veins accompanied by K-feldspar, anhydrite, sericite, and minor chalcopyrite [10].

5. PHYSICOCHEMICAL CHARACTERIZATION

Thermodynamic data for ore and gangue minerals are used to delineate the stability fields of alteration and ore assemblages as a function of fS_2, fO_2, pH, water and aqueous species for the alteration/mineralization stages [14]. a software package for calculating the standard molal thermodynamic properties of minerals, gases, aqueous species, and reactions from 1 to 5000 bar and 0 to 1000 ° C) were used to delineate the stability fields of the various alteration and mineralization assemblages as a function of the intensive parameters such as fO_2 and pH in liquid saturation conditions (since the Sar-Cheshmeh magmatic fluid was rich in liquid phase instead of gas). Temperatures for early and late stages of alteration and mineralization have been estimated from fluid inclusion data [10]. These data indicate that Group II Cu-rich veins were deposited by a high salinity aqueous fluid (about 61 wt % NaCl equivalent) in the temperature range ~300 °C

to ~400 °C, with a mode at 350 °C. In contrast, the Group I Mo-rich veins formed at higher temperature (>400 °C). For the present calculations, the author has adopted a K/Na ratio of 0.37, which falls within the range of values (0.06 to 0.59) determined by microthermometric analyses of fluid inclusions containing halite, chalcopyrite and sylvite daughter minerals from Group I and II quartz veins [10]. Total aqueous S and C concentrations were set to 0.1 molal. These estimates are consistent with analyses of the S content of fluid inclusion decrepitates (1 to 2 wt %), and agree with previous estimates of their concentrations in porphyry fluids [2]. Total concentrations of other components (e.g., aqueous Cl, K, Ca) were chosen to be consistent with the fluid inclusion data [10], see below). The speciation and chemical activities of the important aqueous species were obtained using the program EQBRM [2]. According to Anderson and Crerar, 1993, the EQBRM is a program written in FORTRAN 77 which calculates the concentrations of all aqueous species in a system at equilibrium. The program calculates activity coefficient of ionic species by calling a subroutine called ACTCF, which calculates activity coefficients using the Davies revision of the Debye-Huckel equation:

$$\log \gamma_i = \frac{-Z_i^2 A\sqrt{I}}{1+\sqrt{I}} + 0.2AZ_i^2 \bar{I} \tag{1}$$

where I is the true ionic strength, representing concentrations of all species as corrected for ion-pair and complex formation:

$$\bar{I} = \frac{1}{2}\sum_{i=1}^{N} m_i Z_i^2 \tag{2}$$

This is a fairly crude approximation of the activity coefficients, especially if it is a try to model a system at high T and P [1].

Isothermal fO_2-pH (Fig. 6) diagrams have been constructed at 350 °C, the inferred temperature of deposition for most of the hypogene copper mineralization under water saturation conditions, at the Sar-Cheshmeh deposit [10]. Figure 6 shows the stability fields of pyrrhotite, pyrite, chalcopyrite, bornite, magnetite, titanite, rutile, and hematite, and alteration minerals such as K-feldspar, muscovite and biotite. The activity of the annite endmember in hydrothermal biotite was calculated based on microprobe analyses, following the methods outlined in Wood and Fraser [19]. The log $a_{activity}$ value obtained is -3.3 (in water saturation conditions). The stability field of anhydrite has been contoured over a range of Ca^{+2} activities. The latter value was difficult to estimate.
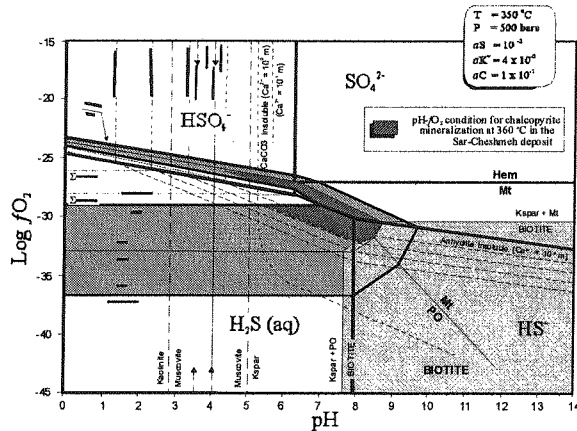
Figure 6: Log fO_2-pH diagram showing approximate conditions (shaded areas) for Group II and III mineralized quartz veins, relative to mineral stabilities field of ore, silicate and oxide minerals at 360 °C and 500 bars in the systems Cu-FeS- O, K-Al-Si-O-H, and calcite/anhydrite, with ionic strength = 1, m Ca^{+2} = 0.1 to 0.001 , aK^+ = 0.04 and aS = 10^{-1}. The inferred conditions of copper deposition at Sar-Cheshmeh are dense and light shaded, refer to the potassic and phyllic alteration zones, respectively. See text for further details and discussion. Abbreviations: CP = chalcopyrite, PO = pyrrhotite, PY = pyrite, BN = bornite, Hem = hematite, Kspar = K-feldspar, CV = covellite, CC = chalcocite.as in Figure 8, plus Qz = quartz, Mt = magnetite, Mus = muscovite.

In Figure 6, the boundaries have been calculated for the dominant silicate and Ca-bearing minerals that co-exist with economic copper mineralization, i.e., orthoclase, muscovite, calcite, and anhydrite at a Ca^{+2} molality of 0.1 to 0.01 in water saturation conditions. The pH value at 350 °C for the muscovite field boundary is between 2.8 to 5.01. At or near the shaded area all of the major ore and gangue minerals in the phyllic and potassic alteration zones at Sar-Cheshmeh are stable. This includes the phases chalcopyrite, pyrite, hematite, muscovite, K-feldspar, anhydrite, and hydrothermal biotite. Although not shown, molybdenite would also be stable at this area. However, as discussed earlier, most of the molybdenite at Sar-Cheshmeh was deposited at higher temperatures.

Figure 7 is also a fO_2-pH diagram drawn for the same conditions (in water saturation conditions) as those in Figure 6. The inferred conditions of copper deposition at Sar-Cheshmeh are dense and light shaded areas, which refer to potassic, and phyllic alteration zones, respectively [10].

The stability fields of anhydrite and calcite are also shown in Figure 7, contoured for different Ca^{2+} activities. Both minerals are present in hypogene ore at Sar-Cheshmeh, although anhydrite is by far the more abundant phase, especially in the higher temperature stages of mineralization. This is consistent with the fact that the anhydrite insoluble boundary occurs at a much lower pH than that for calcite at any given aCa^{2+}. In fact, according to the calculations, calcite could only form at pH values above 5, assuming a maximum aCa^{2+} value of 0.1, and that the estimate of total C = 0.1 molal is correct.
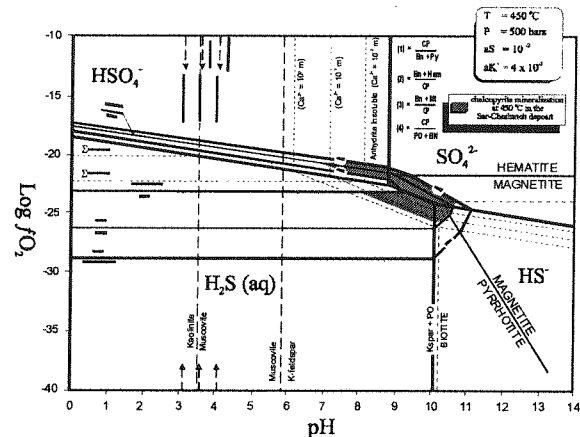
Figure 7: Log fO_2-pH diagram showing the approximate condition (dark shaded area) of the Group I Mo-quartz veins, relative to mineral stabilities in the systems Cu-FeS- O and K-Al-Si-O-H, and anhydrite at 450 °C, 500 bars, ionic strength = 1, m Ca^{+2} = 0.1 to 0.01, aK^+ = 0.004 and aS = 10^{-1}. Activities were estimated from speciation calculations using compositional data observed from fluid inclusions. Phase boundaries, predominance fields and stabilities of mineral aqueous species were calculated as described in Figure 7.
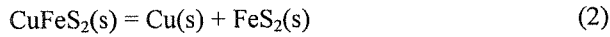
Figure 8 shows the thermodynamic conditions at high temperature, 450 °C and 500 bars, which is estimated to be the suitable for molybdenite mineralization and associated potassic alteration at Sar-Cheshmeh. The muscovite-K-feldspar and muscovite-kaolinite boundaries occur at pH values of 2.8 and 5.0, respectively. It should also be noted that magnetite may coexist with hydrothermal biotite at high temperatures (450 °C and higher), whereas at 350 °C and lower temperature hydrothermal biotite lies in the stability field of hematite. Figure 9 illustrates the mineral stability fields for early potassic and later phyllic alterations on log (aK^+/aH^+) versus log (aNa^+/aH^+) diagrams. As fluids migrate away from an intrusive heat source and down a thermal gradient, the stability field of K-feldspar expands at the expense of albite, which is reflected by potassic alteration of wall-rock.

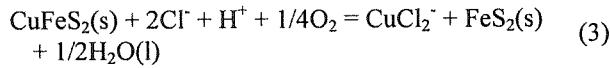## 6. COPPER SOLUBILITY AND CHALCOPYRITE DEPOSITION IN WATER SATURATION CONDITIONS

It has been re-evaluated the solubility of copper in porphyry ore fluids (in water saturation conditions), using as guides the physico-chemical conditions for ore formation at Sar-Cheshmeh outlined above; aCl$^-$ (~1.00 m) was estimated from speciation calculations using the computer program EQBRM and data from fluid inclusions [10]. The mineral solubility contours in Fig. 7 are based on the thermodynamic data of Zotov et al. for the following reaction:

$$Cu(s) + 2Cl^- + H^+ + 1/4O_2 = CuCl_2^- + 1/2H_2O(l) \qquad (1)$$
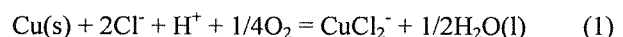
Since the native copper is absent in hypogene porphyry copper deposits, it is possible to recast reaction (1) in terms of chalcopyrite. This is accomplished by adding the following reaction:

$$CuFeS_2(s) = Cu(s) + FeS_2(s) \qquad (2)$$

to obtain:

$$CuFeS_2(s) + 2Cl^- + H^+ + 1/4O_2 = CuCl_2^- + FeS_2(s) + 1/2H_2O(l) \qquad (3)$$

The calculated value of log K_3 at 350 °C, and 500 bars is +9.01. As is evident from reaction (3), copper solubility for the assemblage pyrite + chalcopyrite is highest at conditions of high fO_2, low pH and high chloride concentration. The range in these physico-chemical conditions for the Sar-Cheshmeh deposit has been determined previously (dense and light shaded regions Figure 6).
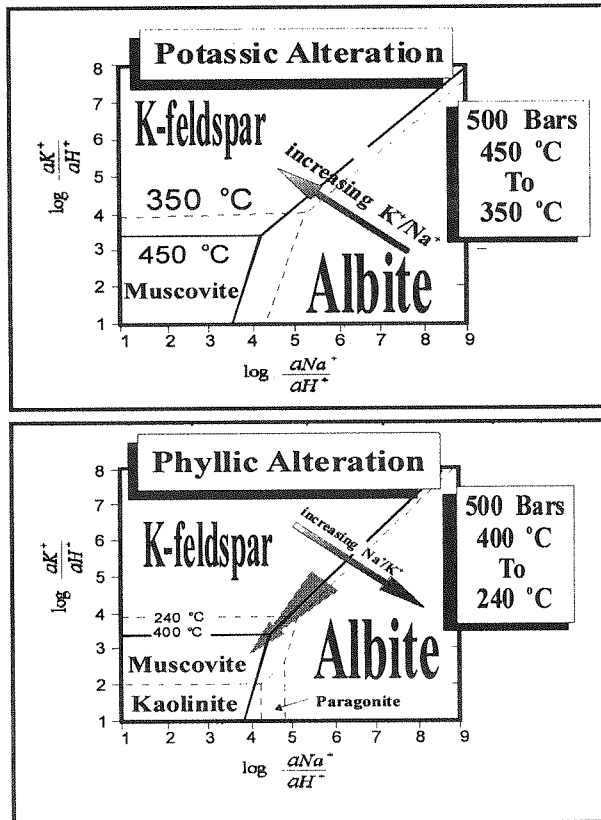

Figure 8: Log aK^+/aH^+ versus log aNa^+/aH^+ diagram showing the thermal paths and stability relationships of minerals in the system K-Al-Si-OH, at different stages of Sar-Cheshmeh hydrothermal system. Diagram (up) is drawn for 360-450 °C and 500 bars. As fluids migrate away from the intrusive heat source and down a thermal gradient (solution retrograde), the log aK^+/aH^+ increases whereas the log aNa^+/aH^+ decreases. This expands the stability field of K-feldspar at the expense of albite, which is reflected by potassic alteration of wall rock. The reverse reaction is operative when the fluid migrates toward the heat source and up a thermal gradient (solution prograde). The albite stability field expands at the expense of K-feldspar. Diagram (down), illustrates the sericitic (phyllic) alteration which overprints the early-stage potassic alteration. Conditions are 240-400 °C, and 500 bars. Under solution retrograde conditions, the muscovite stability field expands at the expense of both K-feldspar and albite.
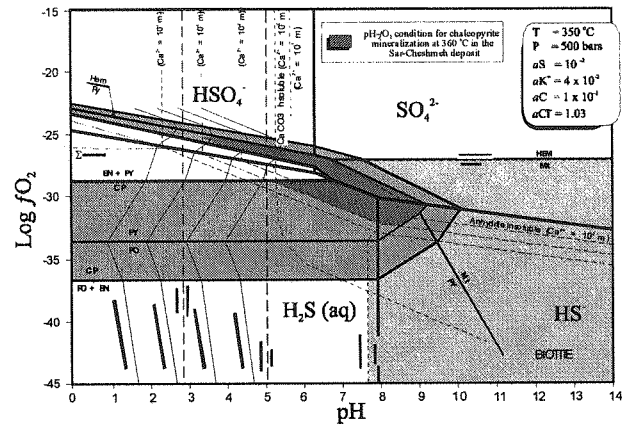

Figure 9: Log fO_2-pH diagram showing Cu solubility contours for Group II mineralized quartz veins at 350 °C (see text for details). Phase boundaries, predominance fields and stabilities of mineral aqueous species were calculated as described in Figure 7. The value of aCl^- (1.04 m) was estimated from speciation calculations using the computer program EQBRM and data from fluid inclusions.

Within this range, copper solubility varies over several orders of magnitude, from over 1,000 to less than 1 ppm. The former value is close to the estimated copper content of the initial fluid at Sar-Cheshmeh (>1000 ppm, Type I fluid inclusions), based on SEM/EDS analyses of fluid inclusion decrepitates and the volume of chalcopyrite daughter minerals [10]. As shown in Figure 7, copper solubility decreases rapidly with increase in pH. Such an increase could occur, for example, during mixing of acidic magmatic waters with heated meteoric waters of lower salinity and near-neutral pH. Mixing would also serve to dilute the aqueous phase with respect to Cl⁻ ion, further augmenting the precipitation of chalcopyrite. A sudden increase in pH could also result from boiling, due to the partitioning of acidic components (e.g., HCl, CO_2, SO_2, H_2S) into vapour during phase separation [7]. Both these processes, boiling and fluid mixing, are inferred to have been important in the genesis of the Sar-Cheshmeh deposit, based on fluid inclusion and ore petrology constraints.

As discussed earlier, there was a transition from early molybdenite precipitation to later copper deposition, which could imply that decreasing temperature was an important control of mineralization. The temperature dependence of copper solubility for a solution with $aCl^- = 1$ m (at 350 °C aCl^- is estimated to have been ~1.00 m, see earlier discussion) and P = 500 bars is shown in Figure 6. The calculations are based on equation 3, and therefore assume equilibrium between chalcopyrite and pyrite. Oxygen fugacity was fixed by coexistence of magnetite and hematite, both of which are present in hypogene alteration/mineralization at Sar-Cheshmeh. The pH was calculated assuming coexistence of muscovite, K-feldspar and quartz, with aK^+ set to 0.04 (see preceding discussion). Thermodynamic data for copper solubility were taken from [20], and all other reactions from

SUPCRT92 [14].

7. DISCUSSION

One of the main objectives of this study was to try to define in some detail the physico-chemical conditions attending mineralization at Sar-Cheshmeh with its water saturation conditions. A second objective was to try to combine these thermodynamic constraints with the results of the fluid inclusion study of Hezarkhani (2006) to build a chemical model describing the process of ore deposition. This proved a much more difficult task, largely because the inherent complexity of porphyry Cu systems can lead to non-unique solutions. For example, the analysis of chalcopyrite solubility (see reaction (3)) indicates that this mineral could have deposited in response to: (1) a decrease in fO_2; (2) an increase in pH; (3) a decrease in temperature; and (4) a decrease in chloride concentration. To determine which of these processes was most responsible, it is necessary to re-evaluate all of the fluid inclusion, thermodynamic and paragenetic evidence. The first process, reduction, can probably be ruled out. There is little evidence for significant redox reactions in the mineral assemblages at Sar-Cheshmeh. If anything, the transition in the dominant Fe-oxide from magnetite at high temperature to hematite at low temperature suggests an increase in relative oxidation state with reaction progress. The second process, pH increase, could have been important during fluid boiling and resultant potassic alteration. However, it is demonstrated that at the inferred temperature of potassic alteration at Sar-Cheshmeh, the ore fluids were probably highly under-saturated with respect to chalcopyrite, or any other copper mineral for that matter. Moreover, it has shown that most of the main-stage mineralization at Sar-Cheshmeh appears to be associated with phyllic, rather than potassic alteration. Thus, although boiling may have played an important role as a means of generating a highly saline, copper-rich brine, little copper deposition appears to have taken place at this time. The third process, temperature decrease, almost certainly played an important role. A significant drop in temperature occurred between early potassic alteration (about 450 °C) and later phyllic alteration and main-stage Cu mineralization (about 350 °C). This fact, combined with the thermodynamic analysis of chalcopyrite solubility, suggests that deposition of copper was delayed until the metal-rich brine had cooled several hundred degrees below the temperature at which it exited the parent magma. Cooling may also have indirectly served to lower the acidity of the ore fluid. Although the boiled brine was in equilibrium with K-feldspar and hydrothermal biotite at high temperature, a decrease in temperature would eventually have placed the fluid composition in the stability field of muscovite [5]. Because the conversion of feldspar to muscovite consumes protons, the acidity of the brine would have tended to decrease with cooling, provided that the system remained rock-buffered. Any increase in pH would have served to reinforce the drop in solubility due to cooling alone.

Cooling may also have played an important role as a means of increasing the H_2S content of the ore fluid via hydrolysis (disproportionation) of magmatic SO_2.

Theoretical studies [5,6] have shown that this reaction proceeds from left to right with decrease in temperature, especially below 400 °C. The aqueous sulfide produced could then combine with dissolved copper and iron to precipitate chalcopyrite:

$$CuCl_2^- + FeCl_2(aq) + 2H_2S + 1/4O_2 = CuFeS_2 + 4Cl^- + 3H^+ + 1/2H_2O \quad (4)$$
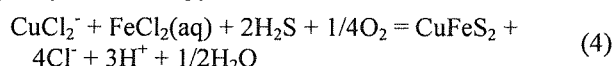
However, it should be noted that the production of H^+ by dissociation of sulfuric acid in reaction (4) could largely outweigh the effect of increasing H_2S concentration on chalcopyrite solubility according to reaction (4), especially if the immediate wall rocks have no pH-buffering capacity (e.g., all feldspar already converted to muscovite). For this reason, hydrolysis of SO_2 could actually lead to the dissolution of chalcopyrite, rather than its precipitation.

The final mechanism, decrease in Cl concentration, is also of potential importance. The fluid inclusion study of Hezarkhani (2006) presented evidence that significant mixing did indeed occur between hypersaline brines of magmatic origin and lower salinity fluids of presumed meteoric origin. Any such mixing would decrease the Cl concentration of the metal-carrying fluid, thus destabilizing the $CuCl_2^-$ complex and depositing chalcopyrite. If the meteoric water was cooler than the magmatic water, mixing would also have resulted in a temperature decrease, further promoting metal deposition, as discussed above. Previous studies have indicated a major involvement of circulating meteoric fluids in the origin of other porphyry Cu deposits [4].

8. CONCLUSIONS

1) The main Cu mineralization at Sar-Cheshmeh occurred in late Groups of II and III veins, as a result of the precipitation of chalcopyrite from high salinity aqueous fluids in water saturation conditions.

2) Copper solubilities established for the chalcopyrite-pyrite assemblages are highest at conditions of high fO_2, low pH and high chloride concentration. At Sar-Cheshmeh, fluid conditions varied from >400 °C, initially to about 300 °C. Solubility decreases rapidly with increasing pH, and mixing of acidic magmatic waters with heated meteoric waters of lower salinity and near-neutral pH could have caused chalcopyrite deposition.

3) Copper deposition occurred at saturation temperature of (<400 °C), and the thermodynamic modeling of the stability fields for alteration and mineralization phases support this interpretation. These conditions are very close to those in Butt Montana copper deposit.

9. ACKNOWLEDGMENTS

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