TRANSPORT AND DEPOSITION OF COPPER AND GOLD IN PORPHYRY DEPOSIT: A CONSTRAINT FROM MICROTHERMOMETRY AND HYDROTHERMAL BIOTITE CHEMISTRY

Arifudin Idrus*

ABSTRACT

The nature of ore-related hydrothermal fluids as well as partition, transport and deposition of metals particularly copper and gold in the fluids are essential issues in the study of mineral deposits. In this current work, microthermometry of fluid inclusions in quartz veins/veinlets and mineral chemistry of hydrothermal biotite associated with copper-gold-bearing potassic alteration have been employed to constrain the transport and deposition of copper and gold in the Batu Hijau porphyry deposit, Sumbawa Island, Indonesia. The ore-bearing quartz veins/veinlets contain abundant hypersaline liquid (~45 NaCl wt.% eq.) and low saline vapour-rich inclusions, which were trapped at high temperature of ~510°C and inferred to contain chalcopyrite daughter crystals. This may suggest a strong partition of copper-(gold) from melt into the coexisting hypersaline liquid and vapour phases. Mineral chemistry analysis indicates that the hydrothermal biotites associated with copper-gold-bearing potassic alteration contain high halogens (particularly chlorine). This implies the significant roles of chlorine during the transport and deposition of the Cu and Au metals through the formation metal-chloride complexes. Copper may transport in form of CuCl₂, whereas gold mainly dissolves as AuCl₃ at a highly temperature and saline hydrothermal fluids.

Keywords: Copper-gold transport and deposition, fluid inclusion microthermometry, biotite halogen chemistry.

INTRODUCTION

A large part of world copper and gold production comes from porphyry deposit. Copper is mostly associated with copper-sulphides, whereas gold is rarely seen in the deposit. We have limited information on the form in which copper and gold were precipitated from solution, and this limits our ability to understand the geochemistry of copper and gold during formation of the deposit. Particularly, we assume that gold in porphyry copper deposit was precipitated as native gold or electrum, just as it was in many greenstone and epithermal gold deposits where visible gold is much more common. Growing information on the actual mineralogical form of gold suggests that this assumption is not always correct and that gold in many deposits is largely in solid solution in other sulphide minerals.

This paper is dealing with the partition, transport and deposition of copper and gold in the porphyry deposit, which is constrained from fluid inclusion microthermometry of quartz veins/veinlets and mineral chemistry of hydrothermal (secondary) biotite associated with copper-gold-bearing potassic alteration zone. As a case study, this contribution will be focused on the partition, transport and deposition of copper and gold in hydrothermal fluids responsible for the formation of the Batu Hijau porphyry deposit, Indonesia.

THE BATU HIJAU PORPHYRY DEPOSIT

The Batu Hijau porphyry deposit is situated in southwest Sumbawa Island, Indonesia (Figure 1). It is a world-class deposit of copper and gold, containing mineable reserves of 914 million tonnes grading 0.525 % Cu and 0.403 g/t Au (Clode et al., 1999). The major wall rock units within the Batu Hijau deposit consist of interbedded andesitic volcanioclastic rocks and quartz diorite intrusions. These rock units are intruded by at least two texturally distinct stages of tonalite porphyries, which are referred to as "intermediate tonalite" and "young tonalite", respectively.

*Dr. Arifudin Idrus is a lecturer/researcher at Department of Geological Engineering, Faculty of Engineering, Gadjah Mada University.

Figure 1. Location map of the Batu Hijau porphyry deposit in the Contract of Work area of Newmont Nusa Tenggara Company in Sumbawa Island, Indonesia.

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The copper and gold-bearing tonalite porphyries were emplaced along the contact between the andesitic volcanic rocks and the quartz diorite. The U-Pb SHRIMP zircon dating indicates that the emplacement of the tonalite porphyry intrusions was rapid within 90 ± 160 thousand years from 3.76 ± 0.12 to 3.67 ± 0.10 Ma (Garwin, 2000). During the emplacement and cooling of the causative tonalite porphyry intrusions, the magmatic hydrothermal fluids are exsolved and interact with the wall rocks, mixing with meteoric waters and form multiple stages of alteration zones. The ore-related hydrothermal alteration and mineralisation developed in four temporally and spatially overlapping stages (Idrus, 2006). The early stage is represented by the central Cu-Au-bearing biotite (potassic), proximal actinolite (inner propylitic) and the distal chlorite-epidote (outer propylitic) zones. The transitional stage is characterised by the chlorite-sericite (intermediate argillic) zone. The late stages are distinguished into the sericite-paragonite (argillic) and pyrophyllite-andalusite (advanced argillic) zones. The very late stage is typified by the illite-sericite zone, developed along structural-controlled quartz-base metal veins.

LITERATURE REVIEWS

The nature of ore-related fluids, transport and deposition of metals are essential issues in the study of mineral deposits. Three approaches are commonly used to constrain the characteristics of the ore fluids, transport and deposition of metals including stable isotopes, fluid inclusions and mineral chemistry. Stable isotope study is mostly employed to determine the source of ore fluids. Fluid inclusion microthermometry and mineral chemistry are utilized to evaluate the physicochemistry and evolution of the ore-related hydrothermal fluids as well as transport and deposition of copper and gold in the deposit (e.g. Yang and Lentz, 2005).

Study on the fluid inclusion microthermometry related to the early central biotite (potassic) zone has been done by Turner (1995). Author pursued the work, which is focused on the fluid inclusions hosted by the quartz veins/veinlets associated with the early distal chlorite-epidote (outer propylitic) and the late sericite-paragonite (argillic) zones. The classification and terminology for the fluid inclusion types follow those of Nash (1976). These fluid inclusion types include: (1) Type I liquid-rich, liquid-vapour (10-40 vol. % vapour) inclusions. (2) Type II vapour-rich, liquid-vapour without or with one or more daughter minerals (60-90 vol. % vapours). (3) Type III liquid-rich, liquid-vapour with more than one daughter minerals. The Type II and III inclusions are closely related to the potassic alteration zone, and are typical of inclusions within the early quartz veins/veinlets from the porphyry systems.

The mineral chemistry particularly halogen fluorine (F) and chlorine (Cl) of hydrothermal biotite has been used to interpret the transport and deposition of copper and gold in the porphyry deposits. The greater exchangeability of elements residing in the hydroxyl site relative to those in the tetrahedral and octahedral sites of biotite opens the possibility that the biotite will undergo F-Cl-OH equilibrium with the hydrothermal fluids, which is responsible for the formation of the alteration zones (Selby and Nesbitt, 2000). Mg and Fe contents of the Batu Hijau biotites are directly related to F-Cl-OH exchange between the biotites and hydrothermal fluids, as indicated by a good agreement of their compositions to the “Mg-Cl and F-Fe avoidance rules”. The theoretical estimates of F-Cl-OH exchange between biotite and hydrothermal fluids are based on the model equilibrium reaction (Munoz, 1984):

$$\text{OH}^{(\text{biotite})} + \text{HX}^{(\text{fluid})} = X^{(\text{biotite})} + \text{H}_2\text{O}^{(\text{fluid})}$$

The high content of halogens particularly Cl in the hydrothermal biotite may be related to the transport and deposition of metals Cu and Au in the deposits (Selby and Nesbitt, 2000).

ANALYTICAL PROCEDURES

The microthermometry of fluid inclusions was analysed using an adapted US Geological Survey gas-flow heating-freezing stage (Fluid Inc., Denver, Colorado, USA). The doubly polished samples of quartz (~100 μm thick) were used in this analysis. The stage was calibrated using synthetic inclusions of pure water at the freezing point (0°C) and critical point (374.1°C) and the triple point of carbon dioxide at -56.6°C. The inclusions were first frozen at about -150°C before heating to the higher temperatures, with a measurement rate of 1 °C/second. The temperature of eutectic (T_e), temperature of melting (T_m), temperature of homogenisation (T_h) of the phases and melting temperatures of halite and sylvite (T_m NaCl and T_m KCl, respectively) were recorded during the heating.

The chemical composition of hydrothermal (secondary) biotite identified optically was determined quantitatively by using a JEOL JXA-8900R Electron Probe Micro Analyser (EPMA). Sample specimens (polished and polished thin sections) were cleaned and coated with graphite prior to the analysis. The analysed points were located with the assistance of a point logger or locally with backscatter imaging. The machine was operated at
voltage of 15 kV with a beam diameter of ~10 μm. A beam of electrons excites the sample, x-rays are analysed and their intensities are converted to concentrations. A microprobe analysis of biotite is defined as the arithmetic mean of 2-10 spot measurements, depending on the mineral grain size. This is aimed to attain sufficient X-ray count rates (i.e. a number of x-rays emitted from the sample surface), which is expressed in count times (it should be ~1000 counts during minimal 10 seconds) for the analysis of halogens (fluorine and chlorine) in the mica. A computer software program MINPET 2.02 was primarily used to calculate the structural formula of the analysed mineral. Both microthermometry and mineral chemistry analyses were carried out at RWTH Aachen University, Germany.

RESULTS

Fluid inclusion microthermometry

The fluid inclusions within the quartz veins/veinlets record the microthermometry (physicochemistry) conditions of fluids at the Batu Hijau porphyry copper deposit. The early-transitional quartz veins/veinlets contain abundant hypersaline liquid (~45 NaCl wt.% eq.) and low saline vapour-rich inclusions. The inclusions were trapped together at a temperature of about 500°C and a lithostatic pressure of 400 bars at a paleodepth of ~1.5 km (Turner, 1995; Garwin, 2000). The estimate of trapping conditions of the fluid inclusions represents a minimum pressure, temperature and salinity, for conditions that prevailed during precipitation of early copper sulphide-bearing quartz veins/veinlets associated with the early potassic alteration in the core of the deposit. The microthermometry data shows that the Type II vapour-rich and Type III liquid-rich fluid inclusions are inferred to contain chalcopyrite daughter crystals.

The quartz veins/veinlets related to the outer propylitic and argillic alterations contain the Type I, II and III fluid inclusions: the Type I inclusions appear to be more abundant than the other types. In contrast, within the early-transitional copper sulphide-bearing quartz veins/veinlets, the abundance of the Type I inclusions is less prevalent. The abundance of the Type I inclusion increases from the central to the peripheral parts of the deposit, which may depict the increase of the influx capacity from meteoric waters outward. The fluids forming the propylitic alteration have a salinity of 17-20 NaCl wt.% eq., and trapped at temperatures of 240-260°C. The evidences indicate that the distal propylitic alteration zone formed under a hydrostatic regime. The hydrostatic pressures of about 100-150 bars correspond to the paleodepth of about 1-1.5 km, representing upper part of the propylitic alteration.

The more dilute fluids associated with the argillic alteration are estimated to have been trapped within the late quartz veins/veinlets at temperatures of 220-230°C and a salinity of 14-19 NaCl wt.% eq. The pressures of trapping were under a hydrostatic regime at about 100-125 bars, which corresponds to a paleodepth of ~1-1.25 km.

Halogen (F, Cl) chemistry of hydrothermal biotite

Hydrothermal biotites from early Cu-Au bearing potassic alteration zone have been analysed in term of chemical composition including their halogen (F, Cl) contents. Mole fraction of magnesium (X_Mg) and halogen (F, Cl) contents are well correlated showing a good agreement with the experimental results of Munoz (1984). The biotite with high Mg tends to incorporate more F and less Cl compared to the biotite with lower Mg; a crystal-chemical effect referred to as “Fe-F and Mg-Cl avoidance rules” (cf. Munoz, 1984). This suggests that the octahedral Mg and Fe in the hydrothermal mica control the halogen-hydroxy exchange of the hydrothermal fluids. Halogen chemistry particularly Cl of hydrothermal biotite exhibits a systematic variation through the alteration zones from inner part to outer part of the deposit. Chlorine in hydrothermal biotites associated with early Cu-Au bearing potassic alteration zone show a highest concentration.

DISCUSSION

Partition of copper and gold

As outlined in the previous section, the microthermometric data indicates that the Type II vapour-rich and Type III liquid-rich fluid inclusions in the Batu Hijau porphyry deposit are inferred to contain chalcopyrite daughter crystals. Study on the inclusion types using Proton-Induced X-ray Emission (PIXE) microanalysis supports the optical observation, which is indicated by a significant amount of copper and other metals within both fluid inclusions (Ryan et al., 2001). The Cu concentrations are about 2.5 and 1.5 wt.% in the Type II and Type III inclusions, respectively, which may suggest a strong partition of copper-(gold) from melt into vapour phase. The appreciable Cu and Au concentrations in the vapour-rich inclusions have also been recorded in several porphyry deposits (e.g. Bingham-Utah, Bodnar, 1995; Alumbrera-Argentina, Ulrich et al., 1999). However, most researchers believe that the transport of Cu, Au and other metals is by brine (e.g. Candela and Holland, 1986). The Grasberg porphyry copper deposit, Indonesia, for example, shows that the Type II vapour-rich inclusions contain ~1.5 wt.% Cu.
which is considerably lower than that in the hypersaline Type III liquid-rich inclusions containing ~5 wt.% Cu (Williams et al., 2003). In the case of the Alumbra, Ulrich et al. (2002) suggested that the selective vapour transport was probably not significant as a mineralising process at the scale of the entire deposit. Considering an average concentration of 0.3 wt.% Cu for both phases, the extent of boiling required to increase the salinity of the brine from 50 to 70 wt.% NaCl eq. will cause only a small fraction of the total Cu to be transferred to the vapour, and some of this Cu may be returned to the brine by recondensation as the vapour phase cools (Eastoe, 1982). Thus, the assumption that the Cu-Au metals partition whether strongly into the coexisting hypersaline liquid or vapour phases needs re-examination.

Transport and deposition of copper

The chloride is commonly an important component of hydrothermal fluids and has been shown to be a potential complexing agent for copper, gold and other metals in porphyry deposit (Shinohara, 1994). The relative high concentrations of halogens fluorine and chlorine in hydrothermal biotites associated with the early potassic alteration may support the evidence of significant roles of the halogens, particularly chlorine during the transport and deposition of the metals through the formation metal-chloride complexes (Zhu and Sverjensky, 1991).

There are lacking of data and literatures, which explain the transport and deposition conditions of copper by the chloride complex. Crerar and Barnes (1976) measured the solubility of various copper-bearing mineral assemblages in chloride solutions to 350°C. They concluded that chloride complexes of copper were of major importance in the formation of the ore deposits, and that CuCl\textsubscript{(aq)} was the dominant form of aqueous copper above temperature of 300°C. However, latest experiments (e.g. Hezarkhani and Williams-Jones, 1999) indicate that CuCl\textsubscript{2}⁻ is the dominant copper complex at the conditions of interest, which is represented by the following reaction:

\[
\text{Cu}_{(s)} + 2\text{Cl}^- + \text{H}^+ + 0.25\text{O}_2 = \text{CuCl}_2^- + 0.5\text{H}_2\text{O} + n \text{H}_3\text{O}^+_n
\]

As noted in the preceding section, the occurrence of copper and gold mineralisation is closely related to the early central potassic alteration, which is characterized by major minerals such as biotite, sodic plagioclase, quartz, magnetite, and hornite. This mineral assemblage is relatively similar that has been used by Hemley and Hunt (1992) in their exploratory experiments for the solubility of Fe-, Cu-, Pb- and Zn-sulphides in chloride solutions at temperature of 500°C and pressure of 1 kbar. Thus, it may be more applicable for the Batu Hijau deposit. The general decrease of copper from central to distal deposit is largely a function of the decreasing solubility of Cu in chloride complex, with decreasing temperature and increasing distance from the causative intrusion that provide the heat source.

Transport and deposition of gold

As described before, it is still debatable, whether the hypersaline liquid or vapour phases playing a significant role in the transport and deposition of copper and gold in the porphyry deposit. However, the experimental result of Gammons and Williams-Jones (1997) indicates that solubility quotients for the metals transported as chloride complexes in coexisting brine and vapour are similar. Gammons and Williams-Jones (1997) introduce two models for gold solubility, i.e. isobaric model and boiling model. The boiling model consisting of boiled brine and vapour models (Figure 2A and B, respectively), are more appropriate to be used in characterising gold solubility within the high crustal-level porphyry copper deposit, and it may represent the role of the liquid and vapour phases in the transport and deposition of the metals.

Four main gold species are considered in their experiments, including AuCl\textsubscript{2}⁻, AuHS\textsuperscript{−}, Au(HS)\textsubscript{2}⁻ and AuOH\textsuperscript{−} (Gammons and Williams-Jones, 1997). The aqueous speciation of gold is a function of ligand concentration ratios. At high Cl⁻/HS\textsuperscript{−} ratio, AuCl\textsubscript{2}⁻ is the dominant species, whereas at high H\textsubscript{2}S/Cl⁻ ratio, Au(HS)\textsubscript{2}⁻ is the dominant species. At low concentrations of both ligands, AuOH\textsuperscript{−} controls the gold solubility. Gold mainly dissolves as AuCl\textsubscript{2}⁻ at high temperature, whereas Au(HS)\textsubscript{2}⁻ predominates below 450°C. The neutral complexes AuOH\textsuperscript{−} and AuHS\textsuperscript{−} are less important at all temperatures. The gold solubility transported as AuCl\textsubscript{2}⁻ increases with an increase in K/Na ratio and oxygen fugacity of the fluid. Therefore, highly saline ore fluids, with potassium-rich and highly oxidised are especially good candidates for dissolving high concentrations of gold. Microthermometric study shows that the quartz veins/veinlets associated copper-gold-bearing alterations form at K-rich as well as highly salinity and temperature suggesting that gold was most probably transported as chloride complexes (AuCl\textsubscript{2}⁻) in coexisting brine and vapour.
Figure 2. Boiling models for gold solubility in the porphyry copper deposit: (A) The solubility of gold in saline brine with $\Sigma$Cl = 2.0 m that separates from a vapour phase at 500°C, and 0.5 kbar, and then cools with no further pressure decrease to 300°C, and (B) The solubility of gold in low-salinity vapour phase with $\Sigma$Cl = 2.0 m that cools from 500°C to 300°C at constant pressure of 0.5 kbar (simplified from Gammons and Williams-Jones, 1997).

Deposition of copper-(gold) sulphides

The bornite and chalcopyrite are the most abundant copper-bearing sulphides in the Batu Hijau deposit. Simon et al. (2000) have been undertaken an experiment to determine how much gold can be accommodated by bornite and chalcopyrite. The results show that the range of gold concentrations in bornite and chalcopyrite increases with increasing temperature from 400 to 700°C. For all temperatures, the bornite contains an order of magnitude more gold than chalcopyrite. The result is consistent with the gold deportment study of the Batu Hijau deposit, which indicates that the average gold concentrations are 3.83 and 0.16 ppm in the bornite and chalcopyrite, respectively (Arif, 2002).

Figure 3. Sulphide stability diagrams: (A) T-fO$_2$ diagram for the system Fe-Cu-S-O at 1.5 kbar under feldspar stable conditions (modified from Simon et al., 2000). The contours of AuCl$^-$ are indicated as log molality, and (B) $[\text{Fe}^{3+}]/[\text{H}^+]^2$ against $[\text{Cu}^+]_2$ at $[\text{H}_2\text{S}]=10^{-2}$ for the system FeO-CuO-H$_2$O at 300°C and 500 bars (modified from Beane and Tittley, 1981). The schematic location of the major alteration zones in Batu Hijau deposit is symbolised by solid-grey circles. For both figures, the grey arrows represent changes in mineral assemblages from early, transitional, to late alteration zones.
The deposition of sulphide and oxide minerals is largely controlled by the physicochemical conditions of the mineralising fluid, e.g. temperature, oxygen fugacity and activity of metals. Figure 3A represents the stability conditions for deposition of sulphide and oxide minerals within the Batu Hijau deposit, with respect to the temperature and oxygen fugacity relationships. Digenite (chalcolite)-bornite and magnetite in the early central biotite (potassic) alteration are deposited at relatively high oxygen fugacity ($f_{O_2}$) ranging from $10^{-15}$ to $10^{-25}$ that approach the hematite-magnetite buffer, at temperatures of about 500-600°C. This estimated temperature range is consistent with the homogenisation temperature (Th) of 510°C, which is determined from Type II and III fluid inclusions in early copper sulphide-bearing quartz veins/veinlets (Turner, 1995).

Figure 3A also illustrates that the highest value contour of AuCl$_4^-$ (10$^{-6}$ molal $\approx$ 100 ppb AuCl$_4^-$) cuts across the darker shaded area of the early central potassic alteration, which suggests that high grade of gold and copper attached in the bornite-digenite (chalcolite) are closely related to the central potassic alteration zone. The copper sulphides in the Batu Hijau deposit are associated with magnetite, which is consistent with the studies of Cox and Singer (1982) that Cu and Au contents of porphyry copper deposits directly correlate with magnetite rather than pyrite contents. The occurrence of copper and gold mineralisation in the early central biotite (potassic) zone is associated with hydrothermal minerals, such as biotite, sodic plagioclase, quartz, magnetite, and bornite-(digenite-chalcolite). This mineral assemblage is relatively similar to that has been used by Hemley and Hunt (1992) in their exploratory experiments for the solubility of Fe-, Cu-, Pb- and Zn-sulphides in chloride solutions at temperature of 500°C and pressure of 1 kbar. Thus, this may be applicable for the Batu Hijau deposit. They found that the general decrease of copper from the central to the distal deposit is largely a function of the decreasing solubility of Cu in the chloride complexes, with decreasing temperature and increasing distance from the causative intrusions that provide the heat source.

The transitional chloride-sericite (intermediate argillic) alteration zone is typically characterised by a chalcopyrite-magnetite assemblage (Figure 3A). Petrographical observation indicates that the chalcopyrite is, partially, a product of sulphidation of early copper-bearing sulphides, e.g. bornite, digenite and chalcolite during the temperature decline. This sulphide-oxide mineral assemblage is probably deposited at temperatures of ~450-500°C and oxygen fugacities of about $10^{20}$-$10^{25}$, on the basis of the stability relationships shown in Figure 3A. The late ore mineral assemblage is characteristically dominated by pyrite and hematite, which formed at a relatively low temperature and oxygen fugacity. The stability conditions for the ore mineral deposition with respect to ion activity ratios of [Fe$^{3+}$]/[H$^+$] and [Cu$^{+}$]/[H$^+$] are shown in Figure 3B. The progression for the formation of the ore mineral assemblage from the early to late alteration events is displayed by the white arrow (after Bodnar and Titly, 1981). The activity of Cu$^+$ and Fe$^{2+}$ upon H$^+$ decreases systematically from the early central biotite (potassic), through transitional chloride-sericite (intermediate argillic), to late argillic alteration zones within the Batu Hijau deposit. Figure 3B also indicates that the Cu$^+$ activity decreases with decreasing Fe$^{2+}$ activity in hydrothermal fluids related to the early, transitional and late alteration zones, respectively.

CONCLUSION

Microthermometry of fluid inclusions in quartz veins/veinlets and mineral chemistry of hydrothermal biotite associated with copper-gold-bearing alteration zone record the nature of ore-related hydrothermal fluids as well as partition, transport and deposition of copper and gold in the deposit. The ore-bearing quartz veins/veinlets contain abundant hypersaline liquid (~45 NaCl wt.% eq.) and low saline vapour-rich inclusions, which were trapped at high temperature of about 510°C and inferred to contain chalcopyrite daughter crystals. The presence of chalcopyrite daughter crystals in the fluid inclusion is supported by PIXE study (Ryan et al., 2001) indicating a significant amount of copper and other metals within both Type II and Type III inclusions at Cu concentrations of 2.5 and 1.5 wt.%, respectively. This may suggest a strong partition of copper and gold from melt into the coexisting hypersaline liquid or vapour phases.

Hydrothermal biotites from Cu-Au bearing alteration (potassic) zone have been analysed in term of chemical composition including their halogen (F, Cl) contents, showing chlorine in hydrothermal biotites associated with the early alteration zone has a highest concentration. The relative high concentration of halogens particularly chlorine in hydrothermal biotites associated with the early Cu-Au bearing potassic alteration may support the evidence of significant roles of chlorine during the transport and deposition of the Cu and Au metals through the formation metal-chloride complexes. Copper transports in form of CuCl$_2^-$, whereas gold mainly dissolves as AuCl$_4^-$ at a high temperature (above 450°C) in highly saline fluids as also shown by fluid inclusion microthermometric data. Copper and gold
concentration systematically decreases from central to distal part of the deposit, which is largely a function of the decreasing solubility of Cu-Au in chloride complex, with decreasing temperature and increasing distance from the causative tonalite porphyry intrusion that provides the heat source.

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