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Partitioning of copper in a two phase fluid system at 600 °C and 700 bar - a study on synthetic fluid inclusions

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Studies on natural assemblages of coexisting vapor and brine inclusions revealed that certain ore-forming elements (Cu, Au, As, ±Ag, ±Mo) occur in higher concentrations in the vapor phase than in the brine. It is believed that these elements are bound in the vapor phase by some sort of hydrosulfide complex, but so far it was not possible to reproduce this phenomenon experimentally at geologically realistic conditions.

We have tried out many different experimental setups to trap Cu and S-bearing fluid in form of synthetic fluid inclusions. In all cases, quartz cores were filled together with some fluid, ± mineral buffers, ± trace elements (Ag, Co, Mo, Pb, As, Zn), and ± quartz glass into gold or platinum capsules, and subjected to 600 °C and 700 bar for 4-21 days in rapid quench cold-seal pressure vessels. The resulting fluid inclusions were analyzed by microthermometry and LA-ICP-MS.

With differing fluid compositions (e.g., amount of added HCl; presence/absence of reducing agents like Na2S), mineral buffers (biotite + magnetite + orthoclase + chalcopyrite + bornite ± andalusite ± gypsum; magnetite + pyrrhotite + pyrite), and capsule material (gold or platinum; single or double capsule), different results were obtained.

In most runs, measured Cu concentrations were not well reproducible, precluding the calculation of precise vapor–brine partition coefficients. The following processes may have contributed to this variation in Cu concentrations: (1) loss of copper by alloying with the gold capsule and/or precipitation of Cu-bearing mineral phases, (2) changes in fO2 and fS2 due to alloying, (3) incomplete separation of brine and vapor during fluid entrapment (co-entrapment of small amounts of brine is very common in vapor inclusions), (4) loss of Cu-bearing daughter crystals during LA-ICP-MS analysis of the fluid inclusions. We consider the latter process subordinate because the inclusions were opened in a controlled way.

Processes (1) through (3) probably all occurred, hence it is important to differentiate between the individual contributions. The amount of co-entrapped brine in vapor inclusions can be quantified based on measured Na concentrations and assuming that the lowest value is representative of the pure vapor endmember. After subtracting this effect, the re-calculated vapor compositions still show a large scatter in Cu concentrations. However, in many experiments these corrected Cu concentrations correlate positively with the concentrations of Co and Pb (which also tend to alloy with gold), suggesting that the vapor inclusions with the highest Cu contents were trapped earliest and thus most closely approach the initial conditions before Cu started to be lost to the gold capsule. Similar trends can be observed also in brine inclusions, although less pronounced due to apparently faster closure times compared to vapor inclusions. By mass balance calculations it can be further demonstrated that the brine inclusions with the highest Cu content did not lose any Co until they were closed off, hence also their Cu content should be representative of the initial conditions. The latter is of great importance, as it allows the initial Cu content of the vapor phase to be calculated by mass balance.

To prevent any change in fO2 or fS2 during the runs we also performed internally buffered experiments (magnetite + pyrite + pyrrhotite with chalcopyrite as the Cu-source). This reduced the scatter of data, but on the other hand excludes the possibility of calculating the Cu content of the vapor phase by mass balance.

The ideal setup would be a system in which no Cu could be lost to the capsule material, but geologic realistic conditions nevertheless can be achieved.