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Preferential partitioning of copper into the vapor phase: fact or a natural artifact?

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Studies on natural assemblages of coexisting vapor and brine inclusions revealed that copper (± Au, As, B, Mo) commonly occurs in higher concentrations in the vapor phase than in the brine. It is believed that this is due to the formation of stable hydrosulfide-bearing complexes in the vapor phase, a theory that is supported by experiments proving the existence of copper-hydrosulfide complexes, experiments noting an increase in the vapor/brine partition coefficients of Cu ($D_{Cu}^{vap/brine}$) with increasing sulfur content of the fluid, and observations showing that in natural boiling assemblages $D_{Cu}^{vap/brine}$ correlates positively with $D_{S}^{vap/brine}$. On the other hand, none of the experimental studies conducted so far has succeeded in reproducing conditions under which copper clearly fractionates into the vapor phase (i.e., $D_{Cu}^{vap/brine} > 1$). An exception seemed to be the study of Nagaseki and Hayashi (2008) who claimed to have obtained $D_{Cu}^{vap/brine}$ values up to 30. However, mass balance constraints and results of experiments performed at identical conditions in our laboratory suggest that these values are wrong and in fact are below unity.

In view of recent experimental studies demonstrating that quartz-hosted fluid inclusions can diffusively loose or gain copper after their formation we wanted to check whether such a process could also lead to artificially high vapor/brine partition coefficients. For this purpose we synthesized vapor and brine inclusions from a Cu_2S-saturated H_2O-NaCl-S fluid at 800 °C / 1.3 kbar and re-equilibrated them in a second experiment with similar fluid at 800 °C / 700 bar. After each step some of the inclusions were analyzed by LA-ICP-MS. Although the Au capsule featured a leak after the second experiment, the result is quite spectacular: while vapor and brine inclusions analyzed after the first step contained only 250 ppm Cu (n=1) and 530 ± 150 ppm Cu (n=8), respectively, vapor and brine inclusions analyzed after re-equilibration contained 5.1 ± 3.2 wt% Cu (n=7) and 0.4 ± 0.2 wt% Cu (n=7), respectively. Results of subsequent experiments suggest that copper in these inclusions was gained in response to a change in pH, which changed from <1 in the first experiment to 9-10 in the second (leaking) experiment. We also found that the fluid inclusions need to contain significant amounts of sulfur in order to be able to take up significant amounts of copper. Both the constraint of pH change and the correlation with the sulfur content in the inclusion are compatible with evidence from natural fluid inclusion assemblages: cooling magmatic-hydrothermal fluids (and inclusion-containing quartz crystals immersed therein) almost inevitably experience a change from acidic to neutral/basic pH due to buffering along the feldspar-mica join, promoting a loss of H^+ from existing fluid inclusions and the corresponding uptake of Cu^+. $D_{Cu}^{vap/brine}$ values greater than unity, observed on natural boiling assemblages could thus be a result of post-entrapment copper gain rather than representing a primary feature.

References:

Nagaseki H. and Hayashi K. I. (2008) Experimental study of the behavior of copper and zinc in a boiling hydrothermal system. *Geology* **36**, 27-30.