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Understanding a million dollar reaction - the mineral replacement of chalcopyrite by chalcocite - insights from in-situ XRD and in-situ XAS techniques.

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Australia is host to world's 2nd largest copper deposits - yet it's only the 5th largest copper exporter in the world. The copper industry in Australia faces significant challenges in mineral process owing to the complicated paragenesis of these copper deposits. Chalcopyrite (CuFeS_2) is among the most common Cu minerals. The replacement of chalcopyrite to chalcocite (Cu_2S) in a copper-rich aqueous medium at mild hydrothermal conditions is an emerging method in mineral processing to upgrade the chalcopyrite-rich concentrates while rejecting deleterious elements. Despite its potential as a cost-effective method to upgrade copper ores, the underlying reaction mechanism and kinetics of the mineral replacement of chalcopyrite by chalcocite remains poorly understood.

In fluid-mediated mineral replacement reactions, a comprehensive understanding of the reactions involves the careful observation of changes in the mineral phase (phase transitions, mineralogical changes) and the fluid phase (concentrations and redox state of dissolved components) during the reaction. To probe the mineral replacement of chalcopyrite by chalcocite we have performed two sets of in-situ experiments at two different beamlines at the Australian Synchrotron.

At the PD beamline we have carried out a series of in-situ XRD experiments to follow the reaction at high temperatures ($180^\circ\text{C} - 240^\circ\text{C}$). The results revealed the reaction pathway as well the metastable reaction species generated in the reaction. The major findings from the in-situ XRD experiments include, i) observation of in-situ replacement of chalcopyrite by digenite-high/covellite in all experiments, ii) observation of szomolnokite ($\text{FeSO}_4 \cdot \text{H}_2\text{O}$) and djurleite ($\text{Cu}_{1.96}\text{S}$) as the metastable species during the mineral replacement reaction.

Pivoted on the observation from the in-situ XRD experiments we have been successfully awarded beamtime to study the redox evolution of the fluid during the replacement of chalcopyrite to chalcocite using our in-house high pressure - high temperature 'mAESTRO' cell for in-situ XAS spectroscopy at the Australian synchrotron. Combining the results from the in-situ XRD experiments (nature and relative proportions of solids) and the in-situ XAS experiments (Fe and Cu concentrations and oxidation state in the fluid during the reaction) - we'll be able to construct a coherent understanding of the reaction mechanism governing the replacement of chalcopyrite by chalcocite.

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