

Cu isotope fractionation during the replacement of pyrite by copper-bearing sulfides

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Mineral replacement is an important way for the re-equilibrium of a mineral in nature. This topic, recently, has received increasing attention with respect to the reaction mechanisms, the evolution of textures and compositions, the generation of the porosity, and the geochemical cycle of elements. However, little is known about the isotope fractionation behavior during this process. In this study, Cu isotope fractionation between solid products and aqueous solutions has been investigated during the replacement of pyrite by copper-bearing sulfides. Results show that small mounds of products formed at the pyrite surface at the primary stage of reactions, and then connected together and covered pyrite grains with the proceeding of reactions. The Cu isotope results show that the $\delta^{65}\text{Cu}_{\text{aqueous-solid}}$ is close to the equilibrium line of Cu(I)-sulfides in the primary stage of reactions, whereas quickly far away with prolonged time of reactions. It is important to note that pyrite grain size and Cu concentration have important effects on the Cu isotope fractionation behavior.

Textural observations demonstrate that little pores can be found in the inner layer of chalcopyrite. The mass transfer between pyrite dissolution interface and bulk solutions was hindered by compacted product layers. We propose that the redox of Cu and diffusion of aqueous Cu(I) species in product layers result in the anomalous Cu isotope fractionation between solid and bulk aqueous Cu(I) species. This study implies that the anomalous depletion of ^{65}Cu in low-temperature deposits was probably controlled by local fluid chemistry in product layers, not bulk solutions. This study highlights local conditions, such as the redox reaction, the diffusion of species in product layers control the isotope fractionation pathways, and may open a new window to study the local fluid chemistry that controls isotope fractionation during mineral replacements or isotope exchange between minerals and fluids.