Thermodynamic prediction of aqueous As concentration in scorodite-rich tailings

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Hydrometallurgical processing of As-rich ores for the extraction of gold or other metals produce voluminous fine-grained, As-rich waste which is disposed of in the tailing ponds and stored in these facilities over a long time. Under predominantly acidic conditions, the principal As carrier in these tailings is the mineral scorodite, $FeAsO_4 \cdot 2H_2O$. Field and laboratory studies revealed a wide variation of the aqueous As concentrations in such tailings or in aqueous solutions with scorodite. This study explains these concentrations and their large scatter by thermodynamic modelling of scorodite-rich waste.

For the calculations, we have used recently measured or estimated thermodynamic properties from our work of As-rich hydrous ferric oxide (As-HFO) [1], scorodite [2], crystalline iron oxides goethite and lepidocrocite [3], and ferrihydrite [4].

At pH < 2.5, scorodite dissolves congruently and controls and aqueous As concentrations in the tailings. In this region, most of the experimental studies agree with respect to the measured and predicted As concentrations.

Above pH of ~2.5, Fe^{3+} phases (As-HFO, goethite, lepidocrocite, ferrihydrite) will start precipitating. These reactions drive the aqueous Fe^{3+} concentration down, force further dissolution of scorodite, and hence elevate the aqueous As^{5+} concentration. As each of these phases has a different stability in the environment of the tailings, the influence of each phase on the Fe^{3+} and As concentration is different. Yet, we show that the majority of the scattered experimental measurements fall into the field defined by the models where scorodite dissolves and Fe oxides precipitate. This field is bound for the curves which represent the scorodite-goethite (scorodite dissolves, goethite precipitates) and scorodite-ferrihydrite model. These two phases are expected to constitute the boundaries of the field because goethite is the most stable and ferrihydrite the least stable phase in the system.

Hence, using our data and models, we can reconcile all published experimental and field data with the thermodynamic values for the participating phases. We can also show that the As concentrations is likely to change in time as the Fe oxides age and transform to more stable phases or phase assemblages.

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Fracture Mineral Investigation in Crystalline Rock, SW Greenland

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Abstract

This current study is associated with the Greenland Analogue Project (GAP), an international collaboration between the Nuclear Waste Management Organization (Canada), Posiva (Finland), and SKB (Sweden). The objective of this project is to further understand groundwater evolution in bedrock as influenced by the presence of a continental ice-sheet. Physical and chemical characteristics of secondary minerals within fracture networks in the bedrock were examined to provide insights into paleo-hydrological processes (e.g. hydrothermal, metamorphic, or glacial events).

Two boreholes were drilled into granitic gneiss near the margin of the Greenland ice-sheet. The first was drilled beneath a talik lake and instrumented to sample groundwater (GAP01). The second borehole was drilled to ~350 m to determine the depth of the permafrost (GAP03). Preliminary results, specifically, carbon ($\delta^{13}C$) and oxygen ($\delta^{18}O$) isotope geochemistry of carbonate fracture fillings, are now available (Fig. 1). Calcite analyses reveal a vertical $\delta^{18}O$ trend in the GAP01 borehole, indicative of hydrothermal carbonates. This is further corroborated by the ^{18}O depleted dolomite in that borehole. Dolomite in the GAP03 borehole corresponds to a fracture filling that is coated with a thin layer of calcite (arrow identifies isotopic signature in Fig.1), indicative of two different crystallization events based on the large difference in $\delta^{18}O$ values.

Ongoing fluid inclusion studies are anticipated to elucidate minimum carbonate mineral crystallization (trapping) temperatures and fluid salinities. Formation temperatures are used to calculate the isotopic composition of the parent fluids from which these carbonates precipitated. Estimates for $\delta^{13}C$ of DIC and $\delta^{18}O$ of paleo-fluids are powerful tools to distinguish meteoric or glacial water from fluids that have been subjected to water-rock interaction.

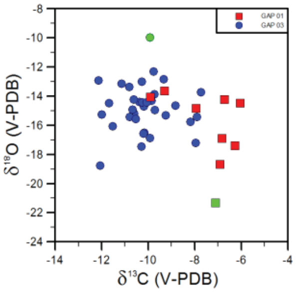

Figure 1: $\delta^{13}C$ and $\delta^{18}O$ isotope signatures of carbonates sampled from Greenland core - calcite (red and blue) and dolomite (green).