A comparative study of rare earth element concentration in copper-sulfides from different hydrothermal sites on the MAR

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Rare earth elements (REE) are good tracers of the evolution of geochemical systems. Their geochemistry is classically studied in hydrothermal fluids [1, 2], mid-ocean ridge basalts and seawater [3]. Because of their low concentration, few studies exist on the REE geochemistry in hydrothermal sulfides. The aim of this study is to determine the REE concentrations and their evolutions in sulfides close to equilibrium with the endmember fluid. At the core of the chimney, Cu-sulfides (chalcopyrite, isocubanite…) are generally dominant. Minerals were sampled from three site associated with ultramafic rocks: Rainbow (36°14'N), Logatchev (14°45'N), Ashadze (12°58'N), and one site associated to basalt: Snake Pit (23°22'N). All sulfides were collected on active black smokers (fluids at 250-350 °C) or in massive sulfide from the internal part of large spires. The differences in the REE concentrations will be discussed in term of basement rock settings and in term of zonation across the sulfides precipitation. We will also discuss how the fluid evolves during the growth of the chimney. The normalisation by REE concentration of hydrothermal fluids from the same area [4] is a way to trace the relationship between the precipitated chalcopyrite and the endmember black smoker fluid.

[1] Douville *et al*. (2002) *Chemical Geology* **184**(1-2), 37–48. [2] Schmidt *et al*. (2007) *Chemical Geology* **242**(1-2), 1–21. [3] Elderfield (1988) **325**(1583), 105–126. [4] Mills & Elderfield (1995) *GCA* **59**(17), 3511–3524.

Critical processes in the release and transport of radionuclides in the near-field

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A total system analysis of the performance of a geologic repository generally involves the simulation of a connected series of processes, beginning with radionuclide release from the waste form and ending with a calculation of dose to an exposed individual. The simulation consists of a series of cascading models, and the uncertainty in the analysis increases with increasing spatial and temporal scales. When the point of compliance is tens of kilometers from the source and the time of compliance extends for hundreds of thousands of years, a positive judgement of the veracity of the analysis requires a remarkable level of credulity. Although the limitations of such analyses have been discussed in detail [1, 2], this begs of question of whether there is not a better approach. An alternative may be to break the analysis into 'parts' that are focused on specific barriers.

I will argue that the barrier most amenable to analysis is the waste form and its interactions with the near-field. To the extent that the mechanisms of release and transport of specific radionuclides can be understood and the uncertainties decreased, the subsequent analysis of the far-field barriers becomes less important. Radionuclide release will be critically sensitive to variations in the temperature, the radiation field, redox conditions, pH, pCO_2, surface area to solution volume and the presence and type of near-field materials. Among the important processes are: 1.) the kinetics of waste form corrosion; 2.) the mechanisms of waste form corrosion; 3.) the formation of secondary, alteration phases; 4.) the sorption/reduction reactions at the surfaces of near-field materials; 5.) the formation and mobility of colloids; 6.) microbial interactions with radionuclides and materials in the near-field. Each of these processes are complicated and are expected to vary in importance with changing conditions over time. One approach may be to consider the radionuclide inventory as it changes with time and to identify the critical processes within each time frame that hold the promise or potential for reducing the mobility of specific radionuclides. Such an approach may reduce the complexity of safety assessments.

[1] Ewing *et al*. (1999) *Risk Analysis* **19**, 933–958. [2] Ewing (2006) In *Uncertainty Underground–Yucca Mountain & the Nations's High-Level Nuclear Waste*. MIT Press, pp.71–83.