Linking Impurity Incorporation to their Effect on Mineral Growth

J.WEBER[1], J. N. BRACCO[3], M. LORENZ[1], A. IEVLEV[1], K. MORE[1], S. HIGGINS[2], J. D. POPLAWSKY[1], V. STARCHENKO[1], A. BERTAGNI[2], S. JINDRA[2], A. G. STACK[1]

[1]Oak Ridge National Laboratory, USA (weberj@ornl.gov)
[2]Wright State University, USA
[3]Argonne National Laboratory, USA

Many minerals show useful capabilities to immobilize toxic heavy metals, e.g. in nuclear waste management or oil/gas extraction. The link between heavy metal incorporation and their effects on growth rates and morphology has not been fully established. Here, we chose the model system of barite ($BaSO_4$) and Sr to elucidate the relationship between growth and incorporation rate. We combined in-situ hydrothermal growth observations by atomic force microscopy (AFM) with high-resolution ex-situ chemical imaging. To profile the Sr-distribution within the grown solid, we employed atom probe tomography (APT), time of flight secondary ion mass spectrometry equipped with AFM (AFM-ToF-SIMS) and transmission electron microscopy (TEM). Barite growth experiments under incrementally increasing Sr-concentration showed a decrease in growth rate for aqueous [Sr]/[Ba] < 1, shifting to an increase at aqueous [Sr]/[Ba] > 1 by a transition from <120> step growth to [010] step growth. By chemical imaging, we observed the preferential incorporation at high aqueous [Sr]/[Ba]. For aqueous [Sr]/[Ba] < 5, we found a correlation between aqueous [Sr]/[Ba] and the newly grown solid composition, whereas this was not the case for higher aqueous [Sr]/[Ba]. We will discuss the results based on current thermodynamic models and demonstrate the need for kinetic process models to interpret our observations.