Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.20012

Uptake of iodine by crystallization of phosphate minerals

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To study immobilization mechanisms for iodine-129 (^{129}I, a mobile fission product), we investigate the iodine entrapment by phosphate minerals from aqueous solution at a range of temperatures similar to those expected at waste repository sites, where thermal peak of the waste package (heated by radioactive decay) is ~300°C (Greenburg and Wen 2013). Experiments were conducted in the plastic containers or Teflon lined autoclaves at 40 and 200°C and pressures of saturated water vapor, where metastable phosphate (brushite) was transformed to apatite or apatite/monetite mixtures in iodate and iodide bearing aqueous solutions.

X-ray diffraction (XRD), scanning electron microscopy (SEM) equipped with energy dispersive spectroscopy (EDS) and backscattered electron diffraction (EBSD) detectors allowed mineral identification and selection of sample areas for atom probe tomography (APT) analysis (Figure 1). Iodine content in crystals and its concentration in co-existed solution were evaluated with electron microprobe analyzer (EMPA) and ultraviolet visible (UV-VIS) spectrophotometer respectively, which allowed calculation of apparent Nernst partition coefficient of iodine ($D^I = I_{crystal} / I_{fluid}$) between crystal and fluid phases.

Our results showed: 1) The highest concentration of iodine (5.6 wt%) was observed in apatite precipitated in chloride (0.5M NaCl) and iodate bearing (0.1M $NaIO_3$) solution at 40°C; 2) D^I decreases by a factor of two (from 9.6 to 4.7) when iodate content in solution increases by a factor of ~ 3 (from 0.03 to 0.1M of KIO_3); 3) A few nm size clusters of iodine in apatite were identified with APT (Figure 1); 3) Monetite or apatite crystallized from NaF-bearing solution incorporated less iodate compared to apatite precipitated from fluoride free solution; 4) Positive correlation between iodate and chlorine was observed in apatite; 5) Iodine uptake by apatite is more effective from iodate bearing solutions compared to iodide bearing solutions by an order of magnitude.

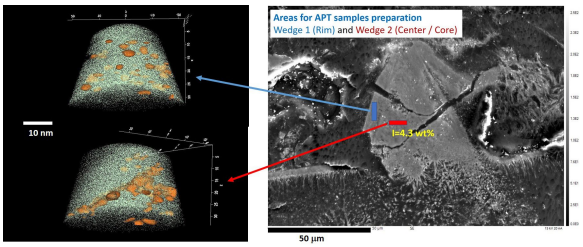

Figure 1. SEM image on the areas selection for APT analysis (right image): iodine content was determined with EMPA. Atomic distribution evaluated with APT (two left images) within apatite tips (prepared with focused ion beam): iodine clusters are shown in orange and homogeneous distribution of calcium is shown in cyan color.

References

Greenburg H.R. and Wen J. (2013) LLNL-TR639869-DRAFT, 38.