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Leaching of metamict and recrystallized zircon in H_2O+HCl: an in situ SR-XRF study

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The radioactive decay of U and Th isotopes and their unstable daughter isotopes to form Pb can transform crystalline zircon to an amorphous, “metamict”, state. Metamict zircon is more susceptible to alteration by aqueous fluids, which may result in an increased loss of radiogenic Pb and He and, thus, too low U-Th-Pb and (U+Th)/He ages [1,2]. The effect of metamictization on the durability of zircon is also of interest for the performance assessment of this mineral for the safe long-term storage of actinides from radioactive waste [3].

We studied the leaching behavior of an initially nearly amorphous zircon in H_2O+HCl at temperatures between 260 and 600˚C using modified hydrothermal diamond-anvil cells and in situ SR-XRF analyses [4] at HASYLAB beamline L. The concentrations of Zr and U in the fluid were monitored as a function of time at several temperatures and pressures.

The results show that metamictization has a large effect on the solubility of zircon. At 260˚C, a constant Zr molality in the fluid of 0.096 mol/kg was reached in 4200 seconds during leaching of a single grain of zircon with an amorphous fraction of about 90% [2] in 7.2 m HCl. At 300˚C, the zircon recrystallized and the Zr concentration in the fluid decreased for about 10000 seconds until it became constant at 0.0052 mol/kg. At 450˚C, the Zr molality in the fluid increased to 0.0083 mol/kg. In contrast, the U concentration in the fluid increased continuously from 0.0013 mol/kg at 260˚C to 0.0015 mol/kg at 450˚C. This indicates that U released into the fluid was not reincorporated in precipitating and recrystallizing zircon. The recrystallized zircon was very fine-grained (grain size 30 to 50 nm) and had an amorphous fraction of about 0.01%

(estimated from the linewidth of the Raman band at about 1000 cm^{-1}). Similar results were obtained in leaching experiments to 600˚C at other HCl concentrations.

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