

LETTER

Experimental observation of an interface-controlled pseudomorphic replacement reaction in a natural crystalline pyrochlore

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ABSTRACT

Pyrochlore-type ($A_2B_2O_6O'$) ceramics are considered for the immobilization of highly radioactive waste. Understanding the alteration process of such potential nuclear waste form materials in aqueous media is critical for the prediction of their long-term stability in a nuclear repository. Current models on pyrochlore alteration are based on a diffusion-controlled hydration and ion exchange process. However, we present results of a hydrothermal experiment at 200 °C with a natural, polycrystalline pyrochlore and ^{18}O -enriched aqueous solution, which are not compatible with a process based on solid-state diffusion. TOF-SIMS and confocal μ -Raman mapping of the run product revealed the occurrence of ^{18}O -enriched alteration zones with sharp chemical gradients to relict unreacted areas when compared to the extent of the alteration zones. The data are consistent with a pseudomorphic reaction that involves the dissolution of the pyrochlore parent accompanied by the simultaneous reprecipitation of a defect pyrochlore at a moving dissolution-reprecipitation front.