## <sup>18</sup>O-tracing of the hydrothermal alteration of pyrochlore

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The disposition and long-term storage of high-level nuclear waste and "surplus" plutonium is a topic of particular concern for the modern society. Minerals of the pyrochlore group have been suggested as a host matrix for plutonium. In this study we compare the hydrothermal alteration of crystalline and metamict, i.e. self-irradiation-damaged pyrochlore, using <sup>18</sup>O-enriched solutions. The <sup>18</sup>O-tracing experiments were conducted on (1) a natural crystalline, Tabased pyrochlore from Congo, (2) a natural heavily radiationdamaged Ti-based pyrochlore-(Y) from Norway, and (3) a synthetic crystalline pyrochlore, (Ca<sub>0.76</sub>Ce<sub>0.75</sub>Gd<sub>0.23</sub>Hf<sub>0.21</sub>)Ti<sub>2</sub>O<sub>7</sub>. From each sample a polished cuboid with edge sizes around 3 mm was prepared for the experiments. The letters "WWU" were engraved into the surfaces of the Ti-based pyrochlore samples using an Excimer laser. Each cuboid was treated in a cold-sealed silver reactor in 1 ml of a 1 M HCl solution containing 43.5 % <sup>18</sup>O at 250°C for 72 hours (Ti-based pyrochlores) or at 200°C for 14 days (Ta-based pyrochlore).

After the experiments the colours of the samples turned from brown or black to almost white. All three cuboids were either intact or parts of it (e.g., an edge) could be recovered. This observation indicates that the reaction is pseudomorphic. Backscattered electron imaging on cut sections revealed both altered and unaltered areas. Whereas the Ta-based pyrochlore was partly transformed into a defect-pyrochlore characterized by a deficit in the A-site cations (i.e., Ca and Na), the Tibased pyrochlore samples were transformed into a mixture of rutile/anatase (TiO<sub>2</sub>). In the case of the metamict Ti-based pyrochlore aeschynite-(Y) was found at the interface between the rutile/anatase assemblage and the unreacted pyrochlore. Imaging the <sup>18</sup>O distribution in the reacted material by timeof-flight mass spectrometry reveals in all cases a strong enrichment in <sup>18</sup>O with a sharp gradient (on a micrometer scale). Our observations suggest that the alteration mechanism is a dissolution-reprecipitation mechanism that operates at a moving front rather than a diffusion-controlled solid-state process. We emphasize here that our results produced under relatively extreme conditions agree well with those derived from experiments conducted under moderate conditions rather expected in a nuclear repository [1].

## References

[1] Zhang Z., Li H., Vance E.R., McLeod T., and Scales N. (2004) *Mat. Res. Soc. Symp. Proc.* **824**.