Goldschmidt 2021 Abstract
https://doi.org/10.7185/gold2021.7426

Uranyl superoxide linked to formation of carbonate corrosion products observed on spent nuclear fuel.

TORI Z. FORBES

University of Iowa, USA

Presenting Author: tori-forbes@uiowa.edu

The high radiation field associated with spent nuclear fuel ($U(IV)O_2$) pellets produces an array of reactive radical species that impact the corrosion and formation of secondary alteration phases [1]. Dioxygen radicals are particularly important as radiolysis products, but the interaction between these reactive oxygen species and $U(VI)O_2^{2+}$ and its effects on the resultant alteration phases is unclear. Herein, we report the first example of a U(VI) superoxide compound and explore its reactivity in the environments relevant to the storage of spent nuclear fuel. We utilized X-ray diffraction and Raman scattering techniques to demonstrate that the uranyl superoxide material reacts with CO_2 in air to afford a mixed uranyl peroxide/carbonate phase within three days, both in solution and under atmospheric conditions. An additional transformation occurs over the course of three months to form the uranyl carbonate mineral, grimselite, which is isomorphic to the čejkaite phase observed as an alteration product on Chernobyl corium [2]. Our results demonstrate the presence and significance of radiolysis in the alteration of spent nuclear fuel and indicate the impact of uranyl superoxide chemistry on high prevalence of carbonate minerals in the corrosion of spent nuclear fuel.

References:

[1] Burns, Ewing, Navrotsky (2012) *Science* 335, 1184-1186.

[2] Barlow, Bailey, Fisher, Stennett, Gausse, Ding, Krasnov, Sayenko, Hyatt, Corkhill (2020)*npj Materials Degradation* 3.