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Tetravalent Uranium Migration and Unrecoverable Uranium Resource Due to Fluorite

YANRU LIANG[1], ZIRU PEI[1], CUI LI[1], DAVID M. PATERSON[2], PETER J. KOPITTKE[3], WENMEI WU[1] AND **PROF. YUHENG WANG, PHD**[1]

[1]Northwestern Polytechnical University

[2]Australian Synchrotron

[3]The University of Queensland

Presenting Author: yuheng.wang.univ@gmail.com

The nuclear power industry generated 2657 TWh of electricity in 2019, representing ~10% of the world's total electricity production and ~29% of low carbon power. The industry is mainly fueled by uranium, a radioactive and toxic element. Its enrichment in the environment can occur due to both geogenic processes, *e.g.*, granite weathering, and anthropogenic activities, *e.g.*, mining and milling. Uranium migration in the environment is controlled by its oxidation state, with hexavalent uranium [U(VI)] being highly soluble in aqueous phases whilst tetravalent uranium [U(IV)] is considered insoluble and immobile. Recent studies[1-6] show that noncrystalline U(IV) species are common in U contaminated reducing environments, with these U(IV) species more susceptible to oxidation and mobilization than crystalline uraninite (UO_2). Therefore, the transformation of noncrystalline U(IV) into UO_2 is considered favorable for U immobilization. Nevertheless, knowledge regarding the migration and fate of residual U from mine and mill tailings, especially the role of common accessory minerals such as fluorite (CaF_2), is still scarce.

Here, we investigated the mechanisms by which U is transported from a decommisioned granite-related mine to a downstream area. Uranium speciation throughout the successive transport bodies was characterized using electron microscopy, X-ray fluorescence microscopy and microscale X-ray absorption spectroscopy. Surprisingly, we discovered evidence of mobile $U^{IV}O_2$-CaF_2 colloids in mine seepage and $U^{IV/VI}O_x$-CaF_2 aggregates in seepage sediment, with this demonstarting a role of CaF_2 in facilitating U migration. These findings are important for two reasons. Firstly, the identification of these colloids demonstrates a remarkable but not-yet-reported mechanism of crystalline U(IV) transport in the environment. Secondly, the U in these colloids is encapsulated by CaF_2 and is therefore not recoverable. As fluorite is widespread in various U deposits[7], our discovery is of broad relevance to global recoverable U resources and the nuclear power industry.

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