Sustainable Approaches to Uranium Remediation: Insights from Neutron Powder Diffraction

C. Wu [1], F.M MacIver-Jones[1], C.A. Morrison[1] C.A. Kirk[1]

1 School of Chemistry, University of Edinburgh, Edinburgh, UK

Caroline.Kirk@ed.ac.uk

Environmental concerns related to uranium (U) and arsenic (As) present significant challenges, particularly regarding their presence in mine tailings and solid waste streams. Metatorbernite ($Cu(UO_2)_2(PO_4)_2{\cdot}8H_2O$) offers a potential solution as a remediation material, capable of immobilising soluble U(VI) ions and preventing their environmental spread. However, current remediation methods often rely on energy-intensive thermal treatments or solvent-based extraction, both of which generate secondary waste streams.

We propose a novel, scalable approach using mechanochemistry to convert unstable As and U species into stable/non soluble mineral phases, suitable for long-term storage. Our preliminary studies have successfully synthesised metatorbernite and metazeunerite ($Cu(UO_2)_2(AsO_4)_2{\cdot}8H_2O$), secondary minerals that naturally occur near uranium mining sites due to the weathering process.[1] The structure of these minerals are comprised of uranyl phosphate/arsenate sheets with hydrated interlayers that contain water coordinated Cu(II) ions. Water plays a key role in their structural stability. When heated from room temperature to 315°C, they undergo stepwise dehydration which coincides with a major reconstruction of the uranyl phosphate/arsenate sheets.[2] However due to the dominant X-ray scattering of uranium atoms, previous X-ray diffraction studies have been unable to identify how water is structurally incorporated into the partly dehydrated, high temperature phases.

This study uses neutron powder diffraction alongside Ab initio Random Structure Searching (AIRSS) to explore hydrogen bonding between the uranyl phosphate/arsenate sheets and provide an accurate model for the interlayer of these mineral structures. [3] We have carried out variable neutron powder diffraction studies over the temperature range RT to >315°C to investigate the structure of the partly dehydrated high temperature phases.

Here we present new structural models for the high temperature phases and show that metatorbernite and metazeunerite have different dehydration and thermal reconstruction pathways. By understanding how hydrogen bonding influences phase stability, we can refine strategies for the effective immobilization of U(VI) and As, ultimately advancing remediation efforts for hazardous waste.

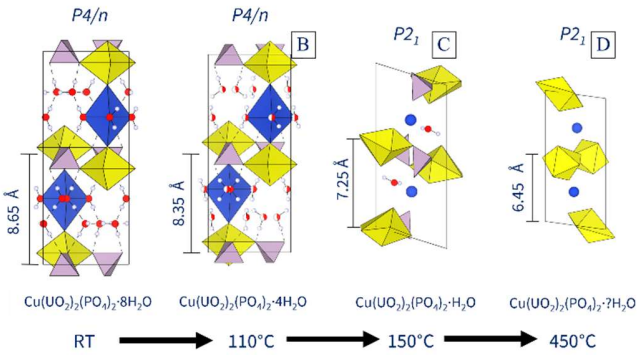


Figure 1. *Dehydration pathway of metatorbernite U polyhedra (yellow), P tetrahedra (lilac), Cu polyhedra/atoms (blue); O atoms (red),H (white)*

[1] C.A. Kirk, F.M. MacIver-Jones, C. Wu, B. Johnson, *Mine Closure 2023,* doi.org/10.36487/ACG_repo/2315_074
[2] J.E. Stubbs, J.E. Post, D.C. Elbert, P.J. Heaney, D.R. Veblen. (2010), *Am. Mineral.*, **95**, 1132–1140
[3] Fiona M. MacIver-Jones, Polly Sutcliffe, Margaret Graham, Carole A. Morrison and Caroline Kirk, *IUCr J.*, 2021, Volume 8(6) 963-972