

Nanocrystalline and non-crystalline U(IV) in the subsurface

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Both nanocrystalline uraninite (UO₂) and non-crystalline U(IV) form in anoxic sediments, controlling the fate and transport of U in contaminated aquifers. It is important to distinguish between these forms of U because non-crystalline species are more reactive towards oxidants and aqueous complexing ligands, increasing the likelihood of U re-release into groundwater in the presence of such solutes.

Much work has been done to elucidate microbiological and geochemical conditions favouring non-crystalline U(IV) or UO₂ formation. Iron reducing and sulphate reducing bacteria can produce non-crystalline species. Non-crystalline U(IV) formation is also favoured in the presence of a strong complexing ligand (e.g. phosphorus). We have developed a model system, analogous to a high-organic matter sediment, to study U(VI) reduction and U(IV) speciation. The model comprises homogenized plant roots incubated in artificial groundwater (containing ~7 mM sulphate, bicarbonate, and Ca²⁺). Sulphate reduction is carried out by the native microbial community associated with the roots, which utilizes the products of root decomposition (no electron donor is added). We use this model to address the following questions: (1) does the ratio of U to sorbent influence the ratio of non-crystalline U(IV) to uraninite? (2) Does reduction of U occur predominantly via enzymatic reduction (direct biological) or by the reaction of U with the reduced products of anaerobic metabolism (indirect biological)? And, finally, (3) Does the mode of U reduction (direct biological vs. indirect biological) lead to differences in the ratio of non-crystalline U(IV) to UO₂?