Nanoscience of Actinide Migration: Development of Reactive Nanomaterials for Cleanup and Monitoring of Actinide Contaminated Sites

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Actinides pose a significant danger to the environment because of a high migrating ability of the oxidized high-valent species. New approaches to monitoring actinide distributions and to preventing their migration are of immediate concern. Particular efforts are needed to develop rapidly deployable immobilizing agents that could be applied to contaminated soils, surface waters, aquifers, buildings, etc. These reactive agents could be comprised of a permeable support embedded with a range of reactive nanomaterials tailored to induce the reduction of high-valent actinides to less soluble lower oxidation state. Such reactive nanomaterials could ideally suit multiple purposes of monitoring, cleanup, and removal of actinides.

The goal of the proposed research is to develop reactive humic nanomaterials that could be easily embedded into the permeable matrix and would possess the ability to induce the reduction and binding of high-valent actinides. These sorbents could be used both for monitoring and removal of actinides in surface and ground waters. The proposed novel approach will use Humic Substances (HS), - natural refractory macromolecular compounds stable to radiolysis, - which have been specifically modified to enhance their reducing and complexing properties with respect to actinides. Two general types of chemical modification will be undertaken: functionalization and cross-linking of humic molecular backbone. Specific reactive moieties (e.g., quinoic units) will be introduced to facilitate actinide reduction and binding, whereas cross-linking will ensure formation of nanosize humic aggregates. The latter are insoluble and bring about irreversible immobilization of actinides. The obtained reactive nanomaterials will be embedded into different permeable matrices (urethane foam or polymeric tissue) using the previously developed fibrous-adsorbent approach (Myasoedova, G.V., et al. 2000. *J. Anal. Chem.* 55, 549-553).

The immobilizing performance of the obtained nanomaterials will be demonstrated with six- and pentavalent Pu and Np. Corresponding sets of redox and binding experiments will be undertaken to determine thermodynamic and kinetic parameters of interfacial reduction and complexation reactions of actinides induced by the developed humic nanomaterials. Mechanistic understanding of these molecular reactions will allow to scale them up to the interfacial phenomena observed under environmental conditions. The corresponding predictions will be made using geochemical modeling of high-valent actinide transport through the porous media embedded with the humic nanomaterials. The obtained model estimates will define the humic nanomaterials possessing desired properties and advance the current modeling of actinide migration in the environment facilitated by HS.