Functionalized clay buffer materials for the long-term sequestration of iodine and technetium

PI: Nathalie A. Wall
University of Florida

Collaborators: Carolyn Pearce (PNNL); James Szecsody (PNNL); Simon R. Phillpot (UF); Juan Nino (UF)

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ABSTRACT:

Total System Performance Assessment of nuclear waste disposal options requires the implementation of effective buffer materials. Buffer materials must meet longevity requirements and be able to scavenge particularly challenging key radioisotopes, such as iodine-129 and technetium-99, both highly mobile species in oxic environments. The main challenge for evaluating the role of I and Tc in performance assessment arises from their complex chemistry and high mobility under oxidizing conditions. Iodine can exist in the -1, 0 +1, +3, +5 and +7 oxidation states in aqueous solution. Iodide (I^-) and iodate (IO_3^-) are the dominant inorganic species of I found in the environment, whereas elemental iodine (I_2^0) only exists as a major species at low pH and over a narrow Eh range. The oxidation state of Tc in aqueous solutions can vary from +3 to +7, but Tc(VII) (as the pertechnetate oxyanion, TcO_4^-) is considered the most stable oxidation state under natural oxic conditions. TcO_4^- is mobile in the subsurface environment due to high aqueous solubility and essentially nonadsorptive properties towards minerals and sediments. However, TcO_4^- can be reduced to produce relatively immobile and sparingly soluble Tc(IV) species. In many proposed nuclear waste repositories, the heat-generating radioactive waste-form will be surrounded by a clay buffer material, which is intended to swell and fill the gap between the waste package and the host geology, restricting radionuclide transport to diffusive processes for several hundred years. The clay buffer is ultimately breached by groundwater, but the clay can be functionalized to have the necessary chemical properties to limit the release of TcO_4^-, IO_3^- and I^-. This functionalized buffer material must also retain its properties of plasticity under conditions relevant to used fuel storage, and remain cost-effective.

The goal of this research is to provide DOE with key data on a novel functionalized clay material for the sequestration of TcO_4^-, IO_3^- and I^-, and to understand the mechanisms associated with long-term contaminant retention. The proposed specific objectives are **A.** Develop new functionalized clays to sequester the various anionic species of I and Tc; **B.** Test the influence of key parameters on the sequestration of I and Tc by the functionalized clay; **C.** Model the sequestration of I and Tc by the functionalized clay. The rationale for this program-supporting work is that a well-designed buffer material will enable increased reliability on waste form performance in repository environments. This effort supports the U.S. DOE R&D program vision to develop strategies and technologies by mid-century for the safe, long-term management and eventual disposal of used nuclear fuel and HLW. Additionally, this work will contribute to the public good by advancing the fundamental science associated with nuclear waste disposal and the training of new students in nuclear sciences to allow for the building of a knowledgeable workforce in the nuclear energy field.