

Understanding Technetium Transport in Porous Media with Batch Geochemical Tests, 4D Emission Imaging Experiments and Modeling Studies

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In this work key parameters controlling technetium (Tc) transport in porous media will be quantified and modeled. Particular focus will be on how transitions in redox state are related to the transport and fate of Tc in the environment. Fundamental geochemical controls have been investigated in equilibrium and kinetic batch sorption studies and have shown substantial differences under oxic versus anoxic conditions as well as in transitions between these conditions. It has also been shown that the amendment of the porous media with strong reducing agents, such as titano-magnetite nanoparticles, can substantially enhance the sorption behavior. This fundamental understanding was then transitioned to column-scale studies intended to investigate the interplay between geochemical conditions and the flow regime. Initial experiments focused on developing and evaluating a novel, one dimensional gamma-ray scanning system to monitor the distribution of Tc-99m within the column over time. While ^{99m}Tc was found to be a useful tracer for one-dimensional transport, we have further investigated the use of SPECT (single-photon emission computed tomography) for 4D (3D with time-lapse) emission imaging of ^{99m}Tc distributions within a column. The value of SPECT imaging, when combined with computed tomography scans of the material, is that it is possible to both image transport behavior in 3D and related this to the structural characteristics of the porous medium, such as preferential flow paths. We are implementing reactive transport models in COMSOL to both improve the design of SPECT imaging experiments as well as to understand how redox transitions affect the local mobility of Tc. One case study, for example, integrates our findings throughout the project by predicting the Tc concentration distributions we might expect in the vicinity of a redox active inclusion (i.e., a zone impregnated with titano-magnetite particles). The model clearly shows a dynamic interplay between the availability of oxygen and Tc reactants, which are controlled by the flow system, and the reactive zone in the column. SPECT imaging experiments are now being designed to validate these modeling results, which are parameterized based on the results of the batch and column experiments.

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