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Sorption and diffusion changes of ¹³⁴Cs, ⁹⁹Tc and ¹²⁹I radionuclides on bentonites at various conditions

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Radionuclides of 99Tc and 129I belong among the fission products of spent nuclear fuel and are the largest contributions with long-term nuclear waste. The high proportion of risk from these radionuclides is due to their large inventories in many types of waste, long half-lives, and the perception that they are highly mobile in the environment and therefore play potentially large role in long-term dose assessment. Computer-modeling methods were used to calculate equilibrium thermodynamic principles, the distributions of predominant aqueous species, and potential solubility controls for the environmentally important oxidation states of each of the contaminants of concern. The Eh-pH diagrams of individual chemical species of the tested radionuclides were calculated by the geochemical software tool Geochemist's Workbench. The data obtained from the model calculations corresponded with experimental results. The chemical stability of the bentonite was studied on a series of long-term pressure and temperature loaded bentonite samples that were selected with the aim of constructing a behavior profile of the bentonite buffer material in the experimental container. The chemical stability of the loaded bentonite samples was evaluated on the basis of their ion exchange capacity, the migration behavior of selected radionuclides and a determination of the leachable components of bentonite into redistilled water. The results obtained in this study were compared with the values for the unloaded bentonite material. It was found that long-term pressure and temperature gradients do not have a significant influence on the changes in ion exchange capacity of the bentonite material. These results make a major contribution towards determining the migration of radionuclides on bentonite, especially cationic forms of radionuclides. Electromigration methods and thin layer chromatography with radiometric detection were used for the identification of technetium and iodine chemical forms in the studied solid-liquid systems. Migration studies of the radionuclides 134Cs in the form of the Cs+ cation and 99Tc in the form of the pertechnetate anion were described on the basis of two dominant processes that both include sorption and diffusion. Dominant chemical forms of technetium under these redox conditions is insoluble TcO2.nH2O, which is formed by the reduction of pertechnetate anion to the oxidation state Tc(IV). The determined values of distribution and diffusion coefficients of the loaded bentonite samples corresponded with the values of the unloaded bentonite material. Ion exchange at the surface sites of bentonite corresponded with the control mechanism of Cs+ cation sorption on bentonite in that both were simultaneously influenced by competitive cations present in the aqueous phase(Na+, K+, Ca2+, Mg2+). To retard the migration of selected radionuclides (99TcO4-, 125I-) released from spent nuclear fuel after the failure of a container, the reducing effects on the concentration of these radionuclides by container corrosion products and of some additives on the bases of Fe compounds in various oxidation states were examined in aqueous media in contact with bentonite.

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