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Chemical Behavior of Strontium at the Solid-Liquid Interface of Amorphous Manganese Oxides: A Molecular-level Study Using EXAFS

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Strontium-90 (half-life = 28 years), a product of uranium fission, is released to the environment from the normal operation of nuclear reactors, nuclear plant accidents, nuclear weapons testing, waste streams associated with fuel processing and leakage from radioactive waste storage facilities. Based on total inventory, half-life and biological activity strontium-90 is one of the most important radionuclides released from nuclear reactors in the first one hundred years of disposal (Martin and Simon, 1972). As a result, the selective retention of strontium by specific components of soil and water is extremely important because these retardation mechanisms (e.g. adsorption, precipitation, etc.) may prevent the migration of the radionuclide for times that assures its decay to insignificance.

Several cases of ^{90}Sr contamination of groundwater and surface water resources have been reported in the literature (e.g., Jackson and Inch, 1983; Kastenberg and Gratton, 1997). As a prominent example, concentrations of ^{90}Sr exceed the drinking water standard by almost 10-fold in ground water in some areas around Hanford in Washington state (US DOE, 1998). In addition, plumes containing ^{90}Sr intersect the Columbia River. Interactions of aqueous strontium ions with natural solid phases have been recognized as playing a key role in controlling the transport, fate, and bio-availability of ^{90}Sr.

Manganese (hydr)oxides are particularly important solid phases in the environment, where they occur as fine-grained aggregates, fresh-water nodules or concretions, crusts and coatings on mineral particles and rock surfaces. Numerous laboratory and field studies have revealed an exceptional reactivity of natural manganese phases [Duff et al., 1999]. Given their reactivity and ubiquity in the environment, it is important to understand their impact on Sr mobility.

Macroscopic uptake and molecular-level spectroscopic investigations were conducted to elucidate the chemical behavior of aqueous Sr at the δ - Mn oxide solid-solution interface. Sorption isotherm measurements revealed a pronounced reactivity of the Mn oxide - Sr system documented by fast reaction kinetics and a high affinity of Sr for the Mn oxide surface. The observed uptake behavior is characterized by a pronounced nonlinearity of sorption isotherms indicative of chemical heterogeneity of the Mn oxide (multiple reactive sorption sites). Furthermore, variation in pH and ionic strength had only a minor effect on the uptake behavior.

Synchrotron-based Extended X-ray Absorption Fine Structure Spectroscopy (EXAFS) was used to identify the local co-ordination environment of the predominant interfacial Sr species. The obtained molecular-level information is fundamental in determining the chemical reactions responsible for the observed macroscopic uptake behavior. The EXAFS results reveal that manganese occurs in the second co-ordination shell around Sr at a characteristic Sr-Mn distance of approximately 3.6Å over a wide range of solution conditions. This finding is consistent with the formation of inner-sphere Sr sorption complexes on the Mn-oxide surface.

At elevated Sr and/or carbonate concentration, $SrCO_3$ precipitates were identified by EXAFS. Under the conditions at which precipitation was observed, the formation of $SrCO_3$ is predicted using existing thermodynamic data. However, the identified $SrCO_3$ precipitates revealed a higher degree of disorder compared to $SrCO_3$ formed by homogeneous precipitation, which is interpreted as indicative of a surface precipitation process.

The present findings are of importance with respect to reliable risk assessment of nuclear (waste) facilities and technologies as well as for the development of sustainable environmental management and protection strategies. Furthermore, the knowledge gained of the chemical behavior of Sr at the manganese oxide - aqueous solution interface has implications for the development of future (off-site) ^{90}Sr waste treatment technologies as well as the design of adequate *in-situ* remediation schemes.

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