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Interactions of uranium and organics with calcium aluminium silicate hydrate

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Organic compounds are routinely present in radioactive wastes of low and medium activity. Sources include waste from effluent treatment (e.g., EDTA, NTA); ion exchange resins; PVC materials and their degradation products (e.g., adipate, phthalate); and cellulose, its derivatives and associated degradation products (e.g., isosaccharinate). These compounds have the potential to form complexes with the radionuclides present in the waste. This complexation may increase radionuclide mobility by (i) increasing their solubility and (ii) decreasing their sorption onto cement-based materials. Current knowledge of cement-organic interactions in these types of systems is limited despite their implications for ensuring the safe storage of nuclear waste. Here, we study the interactions of three organic compounds (α-isosaccharinic acid, phthalate, adipate) as well as uranium with calcium-aluminium-silicate-hydrate (CASH), a hydration product of Al-rich blended cements. Isosaccharinate is a principal degradation product of cellulose, an important organic component in radioactive residues of low and intermediate activity. Phthalate and adipate are produced from the hydrolysis and radiolytic degradation of PVC.

All materials were prepared and handled under a N_2 atmosphere. Sorption of organic molecules was performed with a solid:liquid ratio of 10 - 20 g/L and initial concentrations of 10^{-2}, 10^{-3}, and 10^{-4} M. CASH was prepared with initial Ca:Si and Al:Si ratios of 1.2 and 0.05, respectively. Initial solution pH in the experiments was 11.2 and represented the value obtained after equilibrating the solid phases with deoxygenated deionized water for a minimum of 28 days. Uptake of the organic phases was limited after 14 days' equilibration. Modeling of the experimental systems was consistent with this limited sorption of organics. Our data suggests that due to their low sorption on cement phases at elevated pH, organic molecules such as adipate, phthalate, and ISA may have the potential to increase the mobility of radionuclides such as uranium.