

# Strontium immobilization in Portland cement matrices – influence of low-pH systems, minor phases and carbonation

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Strontium-90 ( $^{90}\text{Sr}^{2+}$ ) is common in low- and intermediate-level radioactive waste streams, often solidified and stabilized by Ordinary Portland Cement (OPC) matrices. Low-pH cementitious systems, where part of the OPC is replaced by amorphous silica-rich additives, are often used for radioactive waste disposal applications. Such formulations can improve the performance of radioactive waste-forms, due to lower heat of hydration, refined porosity and lower pH, decreasing radionuclide migration to the host rock. Although Sr behavior in cementitious systems has been widely studied, full understanding of the phases and mechanisms controlling Sr immobilization in these systems has not been achieved yet. Furthermore, waste-forms are expected to experience carbonation, which may further affect the immobilization process. This study integrates experimental work with geochemical modelling to address these open questions and can provide tools for cementitious waste-forms formulation design, optimizing Sr retention.

In this study, cementitious specimens were spiked with non-radioactive Sr during casting to simulate the immobilized Sr ions. Leaching experiments were held to study Sr retention, in equilibrium and in diffusion-controlled conditions. The experimental approach is based on the use of synthetic alite- (tricalcium silicate) based pastes, as a model material, compared to traditional OPC pastes, to study the effect of minor OPC hydrates on Sr immobilization separately from the major hydrates – C-S-H and portlandite. Low-pH systems were prepared by amorphous silica addition to alite/OPC-based mixtures and compared to their high-pH corresponding systems. Batch sorption tests, where Sr was dissolved in various concentrations and equilibrated with crushed, non-Sr-spiked matrices, were conducted as well. To study the carbonation effect, specimens were subjected to accelerated carbonation followed by leaching tests. For non-carbonated samples, Sr retention was significantly improved in low-pH compared to high-pH systems, and in OPC over their alite model systems. To conclude, Sr retention was improved in the low-pH systems due to sorption to low Ca/Si C-S-H (Figure 1). The Kd values calculated from leaching and sorption experiments suggest that an additional retention mechanism other than sorption takes place in OPC-based systems and absent in their alite-based systems (Figure 2). Carbonation influence on Sr

retention will be discussed as well (experiments underway).

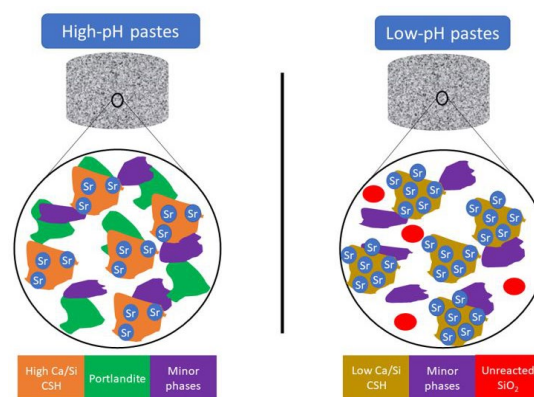


Figure 1

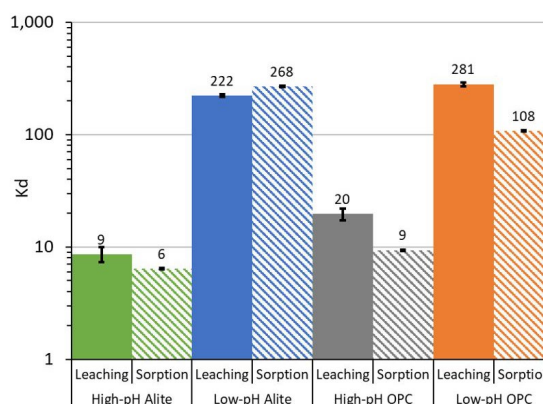


Figure 2