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Effects of Ceria on the Mineralogical and Mechanical Properties of Cement Matrices

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Cementation for the treatment, storage and disposal of nuclear waste is used at a global scale. As a waste form, the hydrating reactions that create the cementitious matrix can be initiated by water in liquid nuclear wastes, thus immobilizing and solidifying the waste once cured. Engineered cement barriers at disposal sites protect the environment by providing sorption and reaction sites that slow radionuclide transport. However, research and development of cements that meet specific application needs often begin with a scoping effort to identify formulations with desirable fresh and cured properties. These efforts often rely on substituting non-radiological surrogates for radionuclides to reduce cost and safety risks. For example, cerium (Ce) is often used as a surrogate for actinides, like uranium (U) and plutonium (Pu) in spent nuclear fuel. Yet, few studies have explored how Ce impacts the properties used to qualify a cement for use.

This research evaluated the fresh, cured, and mineralogical properties of cement materials to evaluate the impact of CeO_2 on the cementing matrix. Three classes of cements were considered, including ordinary Portland cements (OPC), magnesium phosphate cements (MPC), and ultra-high-performance cement (UHPC) composites. An early finding of this work was that CeO_2 is capable of sorbing free water (up to 54 vol%) that effectively decreases the water available to hydrate and form binding cement phases. This led to the formation and/or persistence of anhydrous mineral phases and a decrease in the amorphous content, such as calcium(-aluminum)-silicate-hydrates, that facilitate mineral dissolution and precipitation reactions attributed to strengthening the matrix as it ages. The impact of free water sorbing to CeO_2 resulted in observed shifts in the cements' set time, flowability, residual free liquids, heat of hydration, saturated hydraulic conductivity, heat capacity, coefficient of thermal expansion, and dimensional and mass change. These findings suggest that CeO_2 may not be representative of the matrix behaviors when using actual U and Pu sources. This work also underscores the importance of these studies when using surrogates to extrapolate formulations for immobilizing radionuclides and meeting the needs of specific nuclear waste immobilization applications.