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Chemo-mechanical study of cement paste degradation subjected to external weak sulfate attacks in geological nuclear waste disposals

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This study assesses the containment durability of cementitious material in the context of deep geological nuclear waste repositories. Concrete would be used as a buffer material located around steel waste packages or as a mechanical support of sealing systems. The geological groundwater in contact with waste packages or sealings contains sulfate ions in low quantities which may diffuse in concrete porous media, leading to external weak sulfate attacks. Low concentration sulfate attacks are characterized by the precipitation of secondary ettringite and gypsum that affects chemical and microstructural properties. A significant precipitation of these minerals in the porous media eventually leads to both swelling and cracking by differential strain mechanisms. The novelty of this work is to study the coupled chemo-mechanical evolution of CEM I cement pastes under such conditions, by combining mineralogical (DRX, SEM-EDS, microtomography, autoradiography) and mechanical analyses (microindentation) with reactive transport and mechanical modeling.

The experimental setup consisted in immersing CEM I hydrated cement pastes (w/c = 0.5) in a low-concentration sodium sulfate solution (30 mmol/L), representative of realistic service conditions, which were removed at specific timeframes for analysis (from 15 days to 6 months). Different C_3A ratios were used to boost either ettringite or gypsum formation during the chemical attack. An initial crack was also generated in one sample to estimate whether mineral precipitation in cracks may accelerate its propagation. Results showed sample decalcification contributing to the weakening of the material. Furthermore, apparition of cracks located in the calcium-depleted area was revealed, induced by swelling of the paste due to secondary ettringite and gypsum formation.

Chemical degradation was modeled with the reactive transport code HYTEC to estimate mineralogy and porosity evolution of samples over time. These modeling results were then used to estimate the local evolution of mechanical properties by analytical homogenization techniques. The calculated chemical fronts and Young Modulus values fitted well with experimental results, highlighting a decrease of the material mechanical properties related to the paste decalcification. HYTEC modeling was then applied to the pre-cracked sample, which corroborated the enhanced propagation of the sulfate attack along the crack.