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Leaching behavior of magnesium phosphate cement pastes

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Introduction

The reprocessing of spent nuclear fuel designed for the first generation of nuclear reactors (of the natural uranium – graphite – gas type) has generated some waste with aluminum metal, which need to be stabilized and solidified before their final disposal. Portland cement (PC) is extensively used for the conditioning of low- or intermediate-level radioactive waste. However, its high alkalinity is a serious problem to aluminum stabilization since it leads to strong corrosion of the metal and the release of dihydrogen. Recent work has shown that magnesium phosphate cements may present higher chemical compatibility with aluminum metal as compared to usual calcium silicate cements (*e.g.*, PC). Their main constituents are magnesium oxide (MgO) and potassium dihydrogen phosphate (KH₂PO₄). Their hydration mainly yields K-struvite (MgKPO₄·6H₂O), a low-solubility mineral that sets the pore solution pH to a value close to 8 (*i.e.*, within the passivation domain of aluminum). Nonetheless, the understanding of these binders under leaching conditions is limited and thus, there is the need to gain insights into their long-term durability. Therefore, this work focuses on the leaching behavior of magnesium phosphate cement pastes by using experimental and modeling approaches.

Materials and methods

Magnesium phosphate cement pastes were prepared with a MgO/KH₂PO₄ molar ratio and a water to binder (MgO + KH₂PO₄) weight ratio equal to 1 and 0.51, respectively. To improve rheological properties, a low-calcium fly ash (class F) was added to the mixture, and boric acid was used as a set retarder. After 90 days of curing, paste samples were investigated to obtain information on their porosity and mineral composition. Moreover, semi-dynamic leaching tests were performed by using two different leaching solutions: pure water (used as the reference solution) and an alkaline solution, simulating the pore solution of conventional concrete, which could be placed in the near field of the cemented waste packages in a repository. In order to investigate the degradation mechanisms of magnesium phosphate cement under leaching, microstructural changes were studied by SEM/EDS. EDS mapping was carried out on polished samples, from the exposed surface to the core, to estimate the degradation depths and to identify either the dissolution or precipitation of phases in the solid fraction. Moreover, XRD, TGA and ³¹P MAS-NMR analyses were performed on crushed solid samples to investigate eventual mineralogical changes. Furthermore, ICP-AES and ionic chromatography techniques were used to analyze the leachates obtained over time. These data helped to gain insights into the leaching kinetics of magnesium phosphate cements and to better understand their mechanisms of degradation.

Conclusion

Results obtained in this investigation helped to gain insights into the leaching kinetics of magnesium phosphate cements and to better understand their mechanisms of degradation. These results will be used as input data for reactive transport modeling of the degradation of magnesium phosphate cement pastes under leaching.

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