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### UNDERSTANDING THE BEHAVIOUR OF MAGNESIUM PHOSPHATE CEMENT PASTE UNDER LEACHING: EXPERIMENTAL AND MODELLING APPROACHES

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Some legacy radioactive wastes containing aluminum metal are still waiting to be stabilized and solidified before being sent to disposal. Currently, Portland cement (PC) is extensively used for the conditioning of low- or intermediate-level radioactive waste. Nevertheless, the high alkalinity of PC leads to strong corrosion of Al metal, which is associated with important dihydrogen release [1]. Therefore, there is a need to investigate alternative binders showing better chemical compatibility with Al metal. Magnesium phosphate cements (MPCs) are possible candidates since their pore solution pH may fall within the passivation domain of Al. MPCs are formed through an acid-base reaction between MgO and a soluble acid phosphate salt (e.g.,  $\text{KH}_2\text{PO}_4$ ,  $\text{NaH}_2\text{PO}_4$ ,  $\text{NH}_4\text{H}_2\text{PO}_4$ ). When the reactants are in equimolar amounts, K-struvite ( $\text{MgKPO}_4 \cdot 6\text{H}_2\text{O}$ ) is the main mineral formed during hydration [2]. The understanding of the long-term durability of these binders is still incomplete. Hence, this study aims at investigating the degradation mechanisms of MPC paste under leaching by using experimental and modelling approaches.

Semi-dynamic leaching tests were performed on cylindrical MPC paste samples using well-controlled conditions (pH maintained at 7, decarbonated and thermo-regulated  $25 \text{ }^\circ\text{C} \pm 1 \text{ }^\circ\text{C}$  solutions under nitrogen atmosphere). Leaching solutions were periodically renewed over time to avoid accumulation of dissolved species. Leachates were analysed by ICP-AES, and leached solids were characterized by XRD, SEM/EDS,  $^{11}\text{B}$  and  $^{31}\text{P}$  MAS-NMR analyses. Experimental data were used as an input to numerical simulations. One-dimension (1D) model was developed by using the reactive transport code HYTEC, which takes into the account the geochemical reactions and the migration of ions (diffusive transport) through the cementitious material.

Leaching was mainly controlled by diffusion. Characterization of the leached solids revealed a dissolution front and a zonation process. Three zones were identified:

- (i) a residual and very fragile layer at the interface with the leaching solution, where K-struvite was fully depleted,
- (ii) an intermediate zone, where K-struvite coexisted with cattite ( $\text{Mg}_3(\text{PO}_4)_2 \cdot 22\text{H}_2\text{O}$ ),
- (iii) the sound core (where no cattite was detected).

Simulations reproduced fairly well the mineralogical changes observed experimentally and helped to gain insights into the degradation process of MPC materials under leaching.

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#### References

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