



Development of low pH cement systems forming magnesium silicate hydrate (M-S-H)

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ABSTRACT

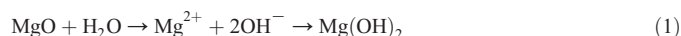
This work aimed to develop novel cement systems for waste encapsulation that would form with a pH of around 10. The approach taken was to investigate the formation of brucite by hydration of a light burned periclase (MgO). Commercially available MgO powders often contain some CaO, and therefore silica fume was added to form C-S-H gel. Identification of the hydrated phases in MgO/silica fume samples showed that brucite formed in substantial quantities as expected. However, brucite reacted with the silica fume to produce a magnesium silicate hydrate (M-S-H) gel phase. After 28 days, the pH of systems rich in MgO tended towards the pH controlled by residual brucite (~10.5), whereas when all brucite reacts with silica fume a cement with an equilibrium pH just below 10 was achieved.

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1. Introduction

Cements are widely used for waste encapsulation to limit contaminant mobility [1–8]. The cement forms a low permeability matrix and results in immobilisation of contaminants by incorporation into hydrated phases and by precipitation due to the prevailing pH in the pore solution [4]. The solubility of many heavy metals is low at around pH 10, and therefore cements with a pH in this range would be highly desirable for such applications. Such moderate pH cements are likely to be particularly desirable for nuclear waste management [9]. For example, in the UK there are significant quantities of mixed wastes containing both Mg and Al alloys. Whilst the high pH in Portland cement based binders passivates the corrosion of Mg alloys, Al alloys corrode under high pH conditions with evolution of H₂ gas. The pH range in which Al alloys show passive corrosion behaviour extends up to about pH 10 and therefore cements with equilibrium pore solutions in this pH range may balance the need for Mg alloy passivation whilst avoiding Al alloy corrosion.

The pH imposed by cement internally and on its environment is a function of the phases present. It follows that control of the pH is easier if the number of phases in the cement paste is limited. For example, the hydration of MgO is given by:



Magnesium hydroxide (Mg(OH)₂, brucite) is a useful starting point for the development of low pH cements because the pH of excess brucite in equilibrium with water is calculated readily from the solubility product of brucite to be around pH 10.5. Hence, in principle a cement based on the hydration of MgO powder, calcined at low temperature to ensure fast hydration, should yield a desirable pH.

However, most commercially available magnesium oxide powders contain some calcium oxide (lime, CaO) impurities. The presence of CaO has the potential to significantly increase the pH through dissolution of the oxide:



Only when a pH of 12.5 is reached, will precipitation of calcium hydroxide (Ca(OH)₂) mitigate further increases in pH.



A simple strategy to mitigate the pH increase due to the presence of CaO consists in adding a pozzolan capable of reaction with the Ca²⁺ ions or the calcium hydroxide (Ca(OH)₂, portlandite) formed during hydration of CaO, resulting in the formation of C-S-H gel. The aim of this paper was therefore to investigate whether mixtures of silica fume and commercial MgO could yield a pH close to 10.

2. Experimental methods

Blends of a commercially available MgO (MgO, MagChem 30, M.A.F. Magnesite B.V., The Netherlands) and silica fume (SF, Elkem Materials Ltd) were prepared. The chemical composition of the raw materials is reported in Table 1. Note that the MgO contains about 0.8 wt.% of CaO.

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