

The effect of silica fume on biodegradation of cement paste and its capacity to immobilize strontium during exposure to microbial sulfur oxidation

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Abstract The disposal of low-level radioactive waste containing isotopes such as strontium by immobilization in cement paste has become common practice. However, the stability of cement paste in the environment may be impaired by sulfuric acid produced by sulfur-oxidizing bacteria. Since biodegradation rates in the environment of most radioactive waste burial sites are too low to be measured, determination of the degradation kinetics of cement paste is a difficult task. This study reports on the development of an accelerated biodegradation system for cement pastes in which the cement paste is exposed to a continuous culture of the sulfur-oxidizing bacterium *Halothiobacillus neapolitanus*. This system facilitated detection of the biodegradation processes in cement paste after as short a time as 15 days. A comparison of the durability of a cement paste blended with silica fume with that of unblended cement paste showed that the silica fume induced an increase in the leaching of Ca^{+2} and Si and enhanced weight loss, indicating rapid deterioration in the structural integrity of the cement paste. The leaching of Sr^{+2} from the silica fume amended cement paste was slightly

reduced as compared with the non amended cement paste, indicating an increase in immobilization of strontium. Nevertheless, our findings do not support the use of silica fume as a suitable additive for immobilization of low-level radioactive waste.

Keywords Cement paste · Biodegradation · Sulfur-oxidizing bacteria · Silica fume · Radioactive waste

Introduction

Immobilization of low-level radioactive waste in cementitious mixtures that are buried in soil or in engineered repositories is becoming a common practice for the disposal of short-lived isotopes such as strontium and cesium (Gougar et al. 1996), raising concerns regarding the fate of the radioactive waste in the environment. Being highly alkaline ($\text{pH} > 10$; Lea 1970), cementitious mixtures dramatically reduce microbial activity in their close vicinity. However, as the cement paste ages it undergoes processes such as carbonation that reduce its alkalinity (Ismail et al. 1993). It is widely accepted that nuclear waste disposal sites are not free of active microbial populations (Francis et al. 1980). Microorganisms can have a significant impact on the stability of such burial sites by consecutive reactions with the external protective barriers (such as metal barrels, clay layer and concrete) and/or consequently react with the waste itself, causing leakage that pollutes the surrounding

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area and groundwater with radioactive elements. The sensitivity of cement products to acids, especially sulfuric acid from biogenic or chemical sources, is well known (Gani 1997; Monteny et al. 2000). Sulfuric acid, the main oxidation product of sulfur-oxidizing bacteria, induces corrosion and degradation of cementitious mixtures. Sulfuric acid reacts with free lime [$\text{Ca}(\text{OH})_2$] in the cement paste to form gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). That compound in turn produces a corroding layer on the surface of the paste, which may cause cracking and degradation due to the large difference in density between the reaction products and the cement paste (Atkins and Glasser 1992; Morti et al. 1992). In addition, a far more destructive reaction occurs between the newly formed gypsum and the Calcium-aluminate-hydrate present in the cement paste. This reaction leads to the production of ettringite ($3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{CaSO}_4 \cdot 32\text{H}_2\text{O}$), which further contributes to the degradation of the cement paste by increasing the internal pressure, leading to the formation of cracks. Thus, the surface area for corrosion processes increases, providing additional sites for acid penetration (Davis et al. 1998). In recent years, it has been realized that available data on the chemical degradation of cementitious mixtures do not reflect the actual biological degradation picture (Monteny et al. 2000). The formation of biofilms containing sulfur-oxidizing bacteria on the cement paste surface and inside the corrosive layer can make the acidic attack more localized and aggressive than chemical degradation (Monteny et al. 2000).

Various additives have been tested to enhance the durability of cement paste against sulfuric acid attack, among them silica fume (Torii and Kawamura 1994; Roy et al. 2001). Silica fume is a pozzolanic material that reacts with the free lime in cement to produce calcium silicate hydrate (CSH). The resulting cement paste is denser and less permeable than non-supplemented cement paste (Gani 1997; Glasser 1992).

The biodegradation of cement paste in natural environments due to exposure to microbially generated sulfuric acid is a very slow process which may take many years. There is therefore a need to devise bioassays based on accelerated biodegradation that provide rapid evaluation of the durability of cement-paste. Most documented systems have been developed for evaluating the biodegradation of concrete sewer pipes (Sand et al. 1987; Gu et al. 1998; Vincke et al. 2002) and are therefore not directly applicable

to testing the leaching of radioactive elements since in burial sites the cement paste is in a relatively dry environment usually in a neutral to alkaline pH levels and not immerse in sewer where the pH levels can be very acidic. We have recently developed an accelerated biodegradation system in which cement paste samples are exposed to semi-continuous cultures of sulfur-oxidizing bacteria. This system enables assessment of the biodegradation of cement paste in as short a time as 30 days (Aviam et al. 2004).

In this paper we report the results of a study focusing on the effects of silica fume augmentation on the biodegradation kinetics of cement paste in terms of leaching of elements and gravimetric weight loss. The study was carried out in an optimized accelerated biodegradation system developed by the authors, in which cement paste samples were continuously exposed to an exponential culture of the neutral sulfur-oxidizing bacterium (NSOB) *Halothio-bacillus neapolitanus*. NSOB are more likely to inhibit radioactive waste burial sites as they can grow in a relatively high pH environment.

Materials and methods

Preparation of cement paste samples

To simulate radioactive waste immobilized in cement paste, Sr^{+2} containing cement paste specimens were prepared by mixing the following materials in an N-50 mixer (Hobart, Troy, Ohio): the binder, i.e., either 2,500 g of Portland cement (PC 250; Nesher Israel Cement Enterprises, Ramla, Israel) or a mixture of 2,000 of Portland cement with 500 g of silica fume (Scancem Chemicals, Norway), plus 760 ml of distilled water containing 16 g of $\text{Sr}(\text{NO}_3)_2$. The mixture was cast into a cylindrical mold (5 cm diameter) and allowed to stand for 24 h. The demolded paste was then sealed in a polypropylene bag and cured at room temperature for 28 days. After curing, the specimen was cut into cubes (approximately 1 cm^3) with an electric saw equipped with a 35-cm diamond blade (Buehler, Lake Bluff, IL, USA) from the internal part of the cylindrical mold.

pH adjustment of cement paste samples

To reduce the alkalinity of the fresh cement paste, which could hinder the growth of sulfur-oxidizing

bacteria in the medium containing the cement paste specimens, the following steps were taken: The samples were first washed for 4 days in a stirred tank containing 5 l of distilled water that was replaced daily. The cubes were dried for 24 h at room temperature and then exposed to 100% CO₂ in a sealed jar for 14 days.

Disinfection of the cement paste samples

The cement paste cubes were immersed in 70% ethanol for 24 h and then dried at 80°C for 24 h to facilitate evaporation.

Bacterium and growth conditions

The sulfur-oxidizing bacterium *H. neapolitanus* strain ATCC 23638 was purchased from the American Type Culture Collection. The cultures were maintained at 30°C in 250-ml flasks containing 50 ml of a mineral salt medium consisting of: 10 g/l of Na₂S₂O₃·5H₂O; 1 g/l of NH₄Cl; 0.5 g/l of MgCl₂; 0.6 g/l of K₂HPO₄; 0.4 g/l of KH₂PO₄; 0.02 g/l of FeCl₃; and 0.02 g/l of MnCl₂. The medium was supplemented with thiosulfate as the sole energy source. In the solidified medium (1.5% agar), 0.08 g/l chlorophenol red was added as a pH indicator.

Continuous accelerated biodegradation system for cement paste

To facilitate rapid biodegradation of the cement paste samples, a continuous culture system for *H. neapolitanus* was developed (Fig. 1). The exposure chamber consisted of a horizontal glass tube equipped with a shelf to hold the cement paste samples (five for each experiment). A polypropylene tube fitted in the top section of the exposure chamber was provided with nozzles through which the exponential culture of *H. neapolitanus* was dripped onto the cement paste samples at a rate that kept the samples constantly wet and maximally aerated. The culture circulated between the growth tank and the exposure chamber with the aid of a peristaltic pump. Fresh medium was pumped into the system at a constant flow of 2 ml/min and the effluent was discarded from the growth tank into a collecting tank, maintaining the culture at a constant volume of 300 ml. All parts of the system were aerated (Fig. 1); the growth chamber was constantly stirred. The pH of the thiosulfate-mineral

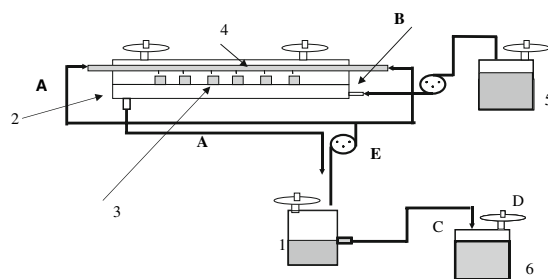


Fig. 1 Schematic diagram of the chemostat-like continuous culture system in which the sulfur oxidizing bacterium *Halothiobacillus neapolitanus* was circulated between the culture growth tank (1) and the cement paste-sample exposure chamber (2). The culture was continuously dripped onto the cement paste samples (3) while flowing through a polypropylene pipe (1 cm in diameter) equipped with 0.5 mm nozzles (one nozzle per cement paste cube) (4). Stable exponential growth of *H. neapolitanus* was assured by supplying fresh medium (5) and discarding excess culture to the collecting tank (6) at the same flow rate. (A) culture circulation; (B) fresh medium inlet to the exposure chamber; (C) outflow to collecting tank; (D) vent filters; (E) peristaltic pumps

salt medium was adjusted to 7 (using 0.2 g/l of KH₂PO₄ and 0.8 g/l of K₂HPO₄) so as to minimize degradation of the cement paste samples by the medium itself. All parts of the system were autoclaved prior to use. In each experiment the combined leachate of 5 cubes, placed in the exposure chamber, were sampled in triplicates from the collecting tank. Samples were withdrawn periodically from the collecting tank to determine ion concentrations in the leachate of the cement paste and from the growth tank to monitor the pH and the bacterial cell density. In addition, at four-day intervals one cement paste cube was removed aseptically from the exposure chamber for dry weight determination.

To compare the degradation effect of the biogenic sulfuric acid with that of synthetic acid, experiments were also performed in sterile medium amended with mineral sulfuric acid to yield a pH of 6.0 (the same pH level as that of the bacterial culture). Sulfate concentration was maintained by the addition of sodium sulfate. Leachate of cement paste cubes exposed to a sterile medium served as control.

Analyses of ions in the *H. neapolitanus* culture

Aliquots withdrawn from the collecting tank were centrifuged at 21,000g for 15 min at 4.0°C to pellet the bacterial biomass. The concentration of thiosulfate

was determined by titration with a KI solution (0.005 N), with starch as the indicator. Sulfate was determined with a Dionex (Sunnyvale, CA, USA) ion chromatograph. The concentrations of Ca^{2+} , Si^{2+} , and Si that had been leached or dissolved from cement paste samples were analyzed, after removal of bacterial cells from the medium, with an optical inductively coupled plasma (ICP) spectrometer (Perkin-Elmer, Wellesley, MA, USA). An earlier study showed that ion uptake by the bacteria is negligible (unpublished data).

Gravimetric determinations

Weight change of cement-paste samples was determined according to Aviam et al. (2004). Briefly, the cubes were first immersed in distilled water for 24 h, dried in an oven at 80°C for 3 days and kept at 100% humidity for 24 h. Samples were washed in 2% sodium dodecyl sulfate (SDS) over night before weighing.

Scanning electron microscopy (SEM) of cement paste samples

Cement paste samples were removed from the medium and washed with 0.1 M phosphate buffer (pH 7.2) to remove medium residue. The samples were fixed in a solution of 2% (vol/vol) glutaraldehyde in 0.1 M phosphate buffer (pH 7.2) for 2 h and then washed three times (20 min each) in 0.1 M phosphate buffer (pH 7.2). Dehydration of the samples was done stepwise by immersing the samples in serial concentrations of ethanol (50, 70, 95 and 100%). The samples were then dried under vacuum in a desiccator at room temperature until scanning (24 h minimum). The samples were gold coated in a deep vacuum and visualized with a JSM-35CF SEM (JOEL, Tokyo, Japan).

Results

Operation of the accelerated biodegradation system

A dedicated bioreactor was developed to accelerate biodegradation of cement paste (Fig. 1). Biodegradation was accelerated by using a continuous culture

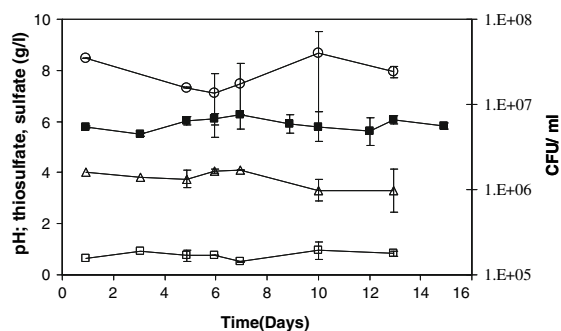


Fig. 2 Parameters used for monitoring the performance of the continuous culture system during incubation of silica fume amended cement paste cubes. Incubated in an exponential culture of *Halothiobacillus neapolitanus*. (○) colony forming units (CFU); (■) pH; (△) thiosulfate (g/l); (□) sulfate (g/l)

of *H. neapolitanus* and aeration of the cement paste samples. To verify the stability of the bacterial culture, we monitored population density, dilution rate and pH in the culture growth tank during operation of the bioreactor with the cement paste samples (Fig. 2). The data show that the bacterial concentration remains stable at about 10^7 CFU/ml and the pH of the culture at 6.0. This moderately acidic environment suits the optimal growth pH of *H. neapolitanus* and the common range of soil pH at radioactive waste burial sites.

Biodegradation of cement paste blended with silica fume

In order to evaluate the effect of silica fume on biodegradation of cement paste, we monitored leaching of Ca^{+2} and Si, the major constituents of the cementitious matrix, from amended and plain cement paste samples during incubation with *H. neapolitanus* (Fig. 3). Leaching of Ca^{+2} proceeded twice as rapidly in microbial culture than in sterile medium in the case of the amended cement paste (Fig. 3A). On the other hand, no difference in Ca^{+2} leaching was detected between the bacterial culture and the control when plain cement paste was used (Fig. 3C). Microbially induced leaching of Si was similar for both types of cement paste (Fig. 3B, D).

The silica fume amended samples lost more weight than the plain cement-paste. After incubation of 15 days with *H. neapolitanus* silica fume amended cement-paste lost 7.2% of its initial weight while that of a plain cement paste samples lost only 2%. A

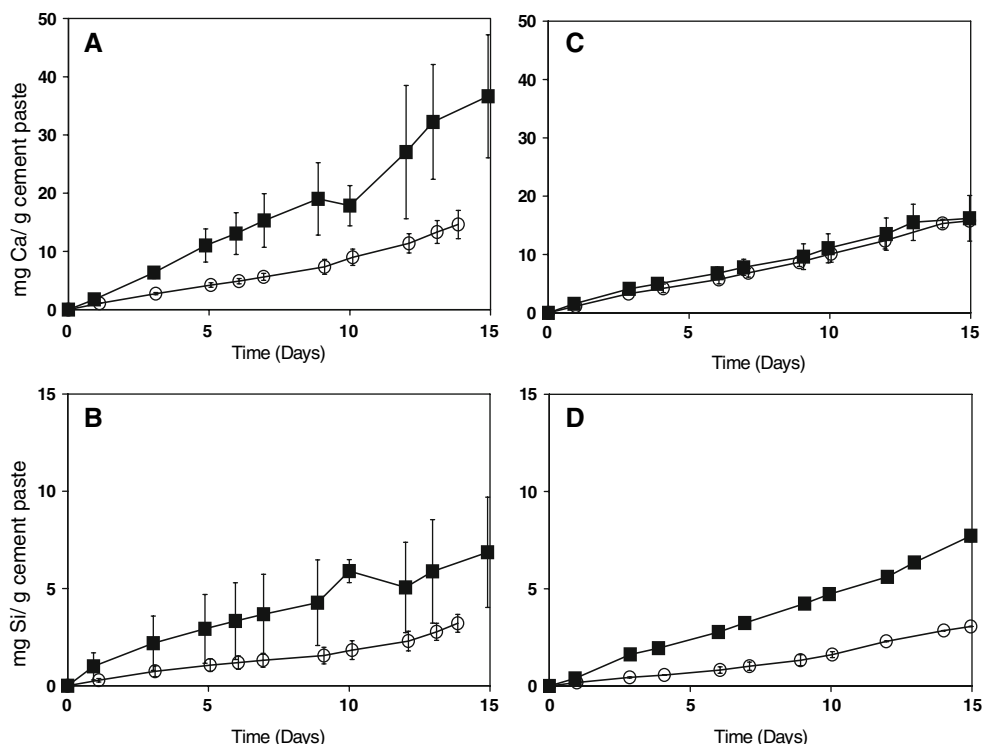


Fig. 3 Leaching of Ca^{2+} (A, C) and silicon (B, D) from cement paste cubes incubated in the continuous system in an exponential culture of *Halothiobacillus neapolitanus*. (A, B) samples amended with silica fume; (C, D) non amended samples; (■) cement paste cubes incubated in bacterial culture; (○) cubes incubated in a sterile mineral medium, pH 7.0, serving as control. Ion concentrations in cell free aliquots

withdrawn from the continuous system every 1 to 2 days were analyzed with an ICP spectrometer. The combined leachate of 5 cubes, placed in the exposure chamber, were sampled in triplicates from the collecting tank. Data represent the mean values of two sets of triplicates, from two experiments \pm standard deviation

similar pattern of weight loss was obtained after incubation in sterile medium (control). SEM analysis combined with energy dispersive spectrometry (EDS) revealed that the main biodegradation products were gypsum and ettringite on the cement paste surface and inside the pores, respectively. A dense biofilm of *H. neapolitanus* was observed on the surface of the gypsum layer (Fig. 4). Furthermore, SEM analysis revealed a higher incidence of cracks in the silica fume amended samples as compared with the non amended samples (data not shown).

Immobilization of strontium

In order to simulate the immobilization of radioisotopes in a cement paste matrix, we added non radioactive Sr^{+2} ions to amended and non amended cement paste samples. On exposure to an exponential culture of *H. neapolitanus*, the samples amended with

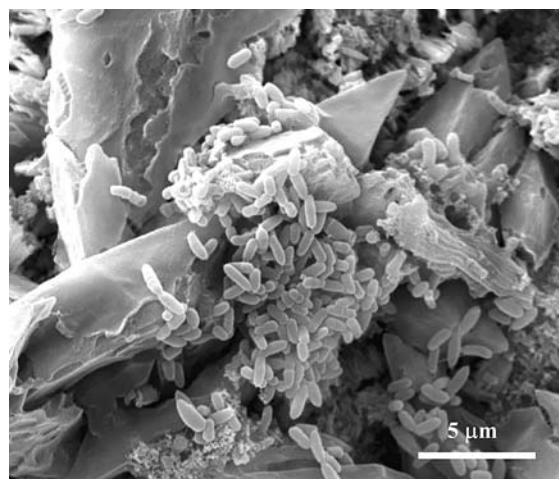


Fig. 4 Biofilm formation on gypsum crystals. SEM photomicrograph ($\times 10,000$) of cement paste surface exposed to an exponential culture of *H. neapolitanus* after 14 days of incubation in the continuous culture system

silica fume exhibited slightly improved immobilization of Sr^{2+} as compared with the silica-free samples (Fig. 5). On the other hand, no difference was found between the bacterial culture and the sterile medium in regard to the amount of Sr^{2+} leached from non-amended samples (Fig. 5). A comparison of the two control curves shows that the presence of silica fume leads to a reduction in the leaching rate (i.e. improved immobilization of the ion; Fig. 5).

Degradation of cementitious mixtures by biogenic and synthetic sulfuric acid

In order to simulate biodegradation conditions in bacterial culture, we exposed non amended cement paste samples to a sterile growth medium supplemented with sulfuric acid at a concentration designed to provide the same pH level as that of the bacterial culture. The synthetic sulfuric acid induced a 2.5-fold increase in the leaching-rate of Ca^{2+} from 2.8 mg Ca^{2+} /g cement paste/day of the non-amended cement paste to 1.1 mg Ca^{2+} /g cement paste/day of similar samples exposed to biogenic acid. However, in the case of Si the leaching-rate was five times higher on exposure to the biogenic acid (0.49 mg Si/g cement

paste/day) compared to exposure to synthetic acid (0.12 mg Si/g cement paste/day). No significant differences in leaching-rate of Sr^{2+} were found between the two media.

Discussion

The concept of accelerated biodegradation systems as a tool for rapid monitoring of microbial induced corrosion (MIC) of concrete was developed in the pioneering studies of Sand (1987) and Sand et al. (1984, 1987). These were followed by various types of accelerated systems (Gu et al. 1998; Vincke et al. 2002 and Aviam et al. 2004). Here we present a new accelerated chemostat-like system designed to screen cement paste compositions for their effectiveness in immobilizing metals, including radioactive elements such as strontium. In this system the cement paste samples are exposed to an exponential bacterial culture that drips onto the samples, providing the maximal aeration favorable for sulfur-oxidizing bacteria. The high degree of stability of the system may be attributed to the combination of an exponential culture, on one hand, and small-sized samples that do not affect culture conditions over time, on the other (Fig. 2).

Surprisingly, the addition of silica fume to the cement mix reduced the structural stability of the cement paste, as demonstrated by the accelerated leaching of Ca^{2+} (Fig. 3) and parallel weight loss. The effect of silica fume on the structure of cement paste has been the object of some debate. While several reports have shown that silica fume increases the chemical resistance of mortars to sulfuric acid (Torii and Kawamura 1994; Roy et al. 2001), another study demonstrated decreased durability of silica fume supplemented mortars exposed to a biogenic sulfuric acid environment (Vincke 2002). In the presence of Silica Fume the content of free lime in the paste is reduced due to the pozzolanic reaction; Torii and Kawamura (1994) report that the addition of 20% silica fume to cement paste resulted in a 70% decrease in the content of $\text{Ca}(\text{OH})_2$. Although free lime itself is vulnerable to acidic attack it still serves as a buffer whose buffering activity protects cement paste against acidic attacks. Therefore, reducing the free lime content can impair the durability of the paste and facilitate microbial attack. Indeed, our

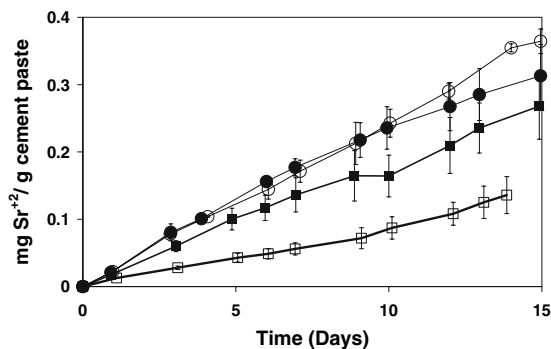


Fig. 5 Leaching of Sr^{2+} from cement paste cubes exposed to an exponential culture of *H. neapolitanus* in the continuous system: (●) cement paste without additives, (■) cement paste amended with silica fume. Leaching of Sr^{2+} was also determined from cement paste cubes in a sterile mineral medium, pH 7.0, to serve as control: (○) cement paste without additives, (□) cement paste amended with silica fume. Ion concentrations in cell free aliquots withdrawn from the continuous system every 1 to 2 days were analyzed with an ICP spectrometer. The combined leachate of 5 cubes, placed in the exposure chamber, were sampled in triplicates from the collecting tank. Data represent the mean values of two sets of triplicates, from two experiments \pm standard deviation

results show that exposure to *H. neapolitanus* led to accelerated leaching of Ca^{2+} and Si ions from silica fume amended cement paste. Alternatively, an increased content of silica fume in the cement paste could also lead to alkali silica reaction (ASR), in which alkaline ions react with siliceous aggregates at high pH to yield a hydrous alkali silicate gel. This gel could increase the internal pressure, leading to the formation of cracks (Diamond 1997). In our study SEM analysis revealed increased cracking in the silica fume amended samples and the presence of spherical aggregates that were not present in the plain cement paste samples and therefore are presumed to be siliceous aggregates. The presence of such agglomerated particles of silica fume has been already reported (Bonen and Diamond 1992; Diamond 1997) that also demonstrated (by SEM and EDS) that these particles are composed mainly of silica which has not been properly dispersed in the paste and therefore did not undergo the pozzolanic reaction (Bar-Nes 2006). Several reports have shown that the presence of such particles can reduce the positive effect of the silica fume on the microstructure and mechanical properties of the pastes (Yajun and Cahyadi 2003). Furthermore, in the presence of high alkali content silica-fume agglomerates can result in matrix expansion due to the alkali-silica reaction (Diamond 1997; Diamons et al. 2004).

The reduced structural integrity of the cement paste was due to the combined effect of elevated pH in the cement paste, alkali cations (i.e., sodium and potassium from the growth medium), and the silica fume additive (including the siliceous aggregates; Bonen and Diamond 1992).

The opposite effects of biogenic and synthetic acid on leaching-rate of Ca^{2+} and Si suggest that the two acids act through different degradation mechanisms, notwithstanding the finding that both acids induced similar leaching of strontium. Previous attempts to compare the degradation efficacies of biogenic sulfuric acid and synthetic acid have also proved inconclusive (Knight et al. 2002). Numerous processes are involved in the deterioration of cement paste, as reflected in our finding that biofilms of *H. neapolitanus* lead to faster leaching of Ca^{2+} , Sr^{2+} , Ba^{2+} and Cd from cement paste than bacterial cells trapped in a nutrient-permeable membrane to prevent contact with the cement paste (Sivan et al. 2006). Hence any attempt to compare biogenic and synthetic

chemical degradation is likely to prove inadequate and probably inaccurate. It would therefore seem that synthetic acid cannot be used in place of biogenic acid to mimic accelerated biodegradation.

The role of silica fume in improving strontium immobilization remains unclear. There are indications that strontium ions can become immobilized through interaction with the CSH phase and to a limited extent may even replace some of the Ca^{+2} ions (Atkins and Glasser 1992). Consequently, one would expect the lower permeability of cement paste amended with silica fume to delay strontium leaching. Furthermore, the reported reduction in the calcium/silicon ratio due to silica fume supplement. This should facilitate interactions between strontium and CSH, thereby improving immobilization (Heath et al. 1996). Alternatively, the high concentration of phosphate in the culture medium may have induced precipitation of strontium (Ksp value of 1.0×10^{-31}) in both cement types. Indeed, this type of precipitation had probably been obtained when a similar medium and bacterium were used in a semi continuous accelerated biodegradation system (Aviam et al. 2004). However, the very limited reduction in strontium leaching recorded for the silica fume amended samples relative to plain cement paste suggests that deterioration in structural integrity played a major role.

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