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Behaviour of different cementitious material formulations in sewer networks

J. Herisson, M. Guéguen-Minerbe, E. D. van Hullebusch and T. Chaussadent

ABSTRACT

Sewer networks are subjected to degradation including biodeterioration of materials in the presence of biogenic sulfuric acid, leading to costly rehabilitation. To ensure durable structures, it is necessary to select the most adapted materials. Two cementitious materials based on Ordinary Portland Cement (OPC) or Calcium Aluminate Cement (CAC), were submitted to biodeterioration in the headspace of an operating sewer network. After a few months OPC materials start to deteriorate whereas CAC materials are still intact. The better durability of CAC materials is due to the presence of alumina providing a combination of protective mechanisms. On-site environmental parameters have been monitored and analysed in the context of the biological and chemical mechanisms involved in material degradation. These data will eventually feed the development of a representative, reproducible and accelerated laboratory test.

Key words | aluminium, biodeterioration, calcium aluminate cement, cementitious materials, hydrogen sulfide (H_2S), portland cement

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INTRODUCTION

Sewer networks are structures built to transport wastewater from production to treatment plants. Severe constraints are applied to these structures which result in premature deterioration. These include root intrusion, joint displacement, cracks, and hole formation leading to significant volume of leakage with an overall risk for the environment and public health. For example it is estimated that 500 million cubic meters of contaminated water per year can leak into soil and ground water in Germany (Kaempfer & Berndt 2009).

Among these degradations, hydrogen sulfide (H_2S) is indirectly responsible for the biodeterioration phenomenon (US EPA 1974, 1985; Vollertsen *et al.* 2008). This poisonous gas is produced by a reduction of sulfates, sulfites or other oxidized sulfur species in sludge and sediments accumulating at the bottom of the sewer creating anoxic conditions favourable for the establishment of a biofilm that may contain sulfate reducing microorganisms (i.e. *Desulfovibrio* spp., *Desulfobulbus* spp.) (Vincke *et al.* 1999). H_2S is then emitted into the aerial part of the structure. In the presence of this gas, a second biofilm colonizes the surface of the structure. It is composed of a succession of sulfur-oxidizing microorganisms that oxidize H_2S into sulfuric acid. This acid streams on the wall of the structure, dissolves metals and cementitious matrix, and leads to the formation of non-cohesive and/or expansive products such as gypsum and/or ettringite. If biodeterioration of cementitious materials is not properly managed, it may lead to the severe structural deterioration with significant cost for its rehabilitation.

However, this model is based on the study of Ordinary Portland Cement (OPC) and is not adapted to all cementitious materials. As an example, Calcium Aluminate Cement (CAC) materials performed much better in sewer network conditions submitted to biogenic deterioration (Alexander & Fourie 2011) despite the fact that they can dissolve in sulfuric acid at pH below 3 (Herisson *et al.* 2013). A report from the Concrete Manufacture Associate in South

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Table 1 | M_F values of various materials (PIPES 2009)

Cement	Aggregates	Deterioration rates (in mm y^{-1})	M_F
OPC	Siliceous	7.5	1
CAC	Siliceous	1.9	0.25
CAC	CAC		0.025

Africa (PIPES 2009) proposes a material factor (M_F) to consider the deterioration of different types of cementitious materials used in sewer networks, based on fourteen years observation of a real sewer. M_F is the ratio of corrosion rate for the alternative material being considered and a standard concrete (OPC and siliceous aggregate). As shown by Table 1, composition of cementitious materials is a crucial factor in biodeterioration phenomenon.

Nevertheless, there is no universal laboratory test that can mimic biogenic acid formation in sewer networks in order to assess any material durability. As a result, currently there is no standard to compare most adapted materials in those deleterious environments.

This paper aims at biodeterioration monitoring of CAC and OPC specimens exposed in severely corroded sewer networks. Such monitoring is meant to gain better knowledge regarding the intrinsic and extrinsic bio-physicochemical parameters governing cement materials biodeterioration.

MATERIALS AND METHODS

Specimens studied

Mortars are generally used as repairing materials or as coating for precast concrete or iron-cast pipes. The behaviour of this type of material can be extrapolated to that of concrete. For the various test series presented in this paper, mortars specimens were cast with OPC or CAC for which chemical oxide compositions are given in Table 2. For all specimens, the water to cement and siliceous sand to cement ratios are respectively 0.37 and 1.44. After demoulding, specimens were stored at room temperature and kept in these conditions to allow natural carbonation. Porosities are similar for both materials (9.8% for OPC and 8.9% for CAC).

Cylindrical mortar specimens (6 cm diameter, 14 cm high) were prepared with a PVC tube anchor for their suspension in the sewer network. Specimens were exposed in the headspace of a sewer network after 6 months conditioning in a controlled lab atmosphere (20 ± 2 °C and 40% RH).

On site exposition

Site characterization

The exposed sewer has experienced severe deterioration of the concrete walls. The cementitious matrix was non cohesive and exposed aggregates were clearly visible. In some areas, a significant gypsum layer had formed. In the atmospheric part of the sewer network, a temperature and H_2S sensor (myDatasensH_2S® from Microtronics) was used to measure the previously mentioned parameters every minute. Hence it is possible to observe the annual temperature cycle with a maximum in summer (around 30 °C) and a minimum in late winter (around 8 °C). H_2S gaseous concentration values measured during one year are between 0 and 250 ppmv with an average value of 100 ppmv.

Sampling and analyses

At each sampling time (i.e. every four months), samples were photographed, weighed and the surface pH was measured. Three sets of samples were exposed. While one of them was left undisturbed, a second one was gently brushed at each sampling time with a nail brush and then washed under tap water before being re-weighed to estimate the proportion of degradation by-products generated. The later one was used to study microbial diversity.

Microbial diversity. Biofilm microbial diversity was studied using molecular techniques. One set of specimens was scraped with a brush impregnated with a guanidine thiocyanate solution (pH 7.5) to extract and preserve total DNA from biofilms. DNA specimens were held in the guanidine

Table 2 | Composition of the two cements (in %)

	Al_2O_3	CaO	SiO_2	Fe_2O_3	MgO	TiO_2	SO_3	K_2O	Na_2O	P_2O_3	LOI	IR
OPC	5.07	63.93	20.87	3.31	0.83	0.24	3.39	1.01	0.20	0.94	0.94	0.26
CAC	51.87	37.06	5.31	2.25	0.54	2.19	0.15	0.31	0.05	0.19	–	–

LOI: Loss On Ignition; IR: Insoluble Residue.

thiocyanate solution at −80 °C for transport and storage until analysis. DNA was extracted using an MP Biomedical kit, the Fast DNA® SPIN Kit for Soil as described by Ettenauer *et al.* (2012). The extracted DNA was amplified by PCR and used for CE-SSCP analysis (Capillary Electrophoresis Single-Strand Conformation Polymorphism). Two specific sets of primers had been used to target prokaryotes and eukaryotes species. Primers w104 (6-FAM labeled) and w49 (Godon *et al.* 1997) were used for prokaryotes while primers w131 (TET labelled) and w16 were used for eukaryotes. The amplification mix contained 1.25 units of Pfu Turbo (Stratagene, La Jolla, CA), 5 mL of 10× buffer, 200 mM of dNTPs, and 200 ng of each primer, with water added to a final volume of 50 mL. The thermal profile used for amplification is as follows: incubation at 94 °C for 2 min, and then 25 cycles of denaturation at 94 °C for 30 s, 61 °C for 30 s, and 72 °C for 30 s. This robust molecular fingerprint tool allows the study of microbial diversity present in environmental ecosystems. One microliter of total extracted DNA was used per PCR-SSCP amplification. The CE-SSCP analysis was carried out with ABI 3130 (Applied Biosystems) as described by Delbes *et al.* (2000, 2001).

ATP analyses. On site, condensed water in equilibrium with mortar was collected and analysed to determine Adenosine TriPhosphate (ATP). It is the molecular unit of currency of intracellular energy transfer of every living entity. The higher the ATP content, the higher the activity of cell or microorganism. Water condensation in the PVC tube anchor and in contact with mortar was recovered in order to measure ATP content by an ATPmeter type Novalum® from Charm Science using WaterGiene® swabs. These swabs, initially made for use in the food industry, are quantitative, easy to use and give repeatable results.

Aluminium ion. *In situ* condensed water was also analysed to determine of dissolved aluminium ion. Aluminium ion concentrations were measured with an inductively coupled plasma atomic emission spectroscopy (ULTIMA 2000 from Jobin-Yvon) according to NF EN ISO 11885 standard.

RESULTS AND DISCUSSION

Table 3 gives an overview of the visual evolution of cementitious materials of specimens exposed on site that are not disturbed. After two years exposition, the OPC mortars are highly deteriorated. They have lost their physical integrity and deterioration products are detached inducing a mass loss of around 14% with a surface pH around 1.5. In contrast, CAC mortars have good behaviour; they keep their original geometry and present a slight weight gain (≈ +1%) with a surface pH of 3.5. As specimens have been exposed in the same environment, the difference of durability is due to matrix composition and more particularly to the cement type.

During the exposition some specimens are brushed (series 2) to mimic tidal zone effect. The monitoring of the mass before and after brushing enables a diagram to be plotted as a function of time (Figure 1). The masses brushed away are calculated by the mass of the degradation products removed during brushing divided by the mass before brushing. The mass losses are not uniform and one can notice that mass loss rates are high during the autumn/winter season. These cooler seasons favour the condensation of water vapour on the specimen surface which in general enhances biofilm development. Moreover, wetting and drying cycles accelerate the process of acidic corrosion. During wetting events, acidic solution penetrates into the material. If

Table 3 | Visual evolutions of OPC and CAC mortars exposed on site over two years

	T 0	4 months	1 year	2 years
OPC				
CAC				

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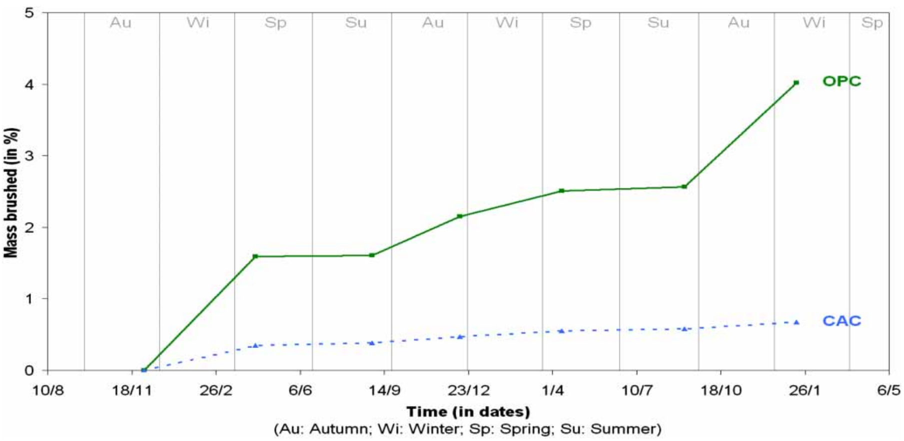

Figure 1 | Evolution of mass loss following OPC and CAC specimens brushing exposed in a sewer network.

dryness causes desiccation, it will result in salt crystal formation. Generally, crystals require an increase volume causing development of a porous network within the material promoting cracks (Sand 1997; Allahverdi & Škvára 2000). Colonization of cracks may be particularly harmful to cementitious materials because these cracks are a way for the spread of aggressive substances. They also allow penetration of microorganism and metabolised substances in the interstices that may lead to an increase in the material internal pressure thus worsening biodeterioration phenomenon (Roux 2008).

Biodeterioration phenomena are caused by the metabolic activity of microorganisms that are well known in the case of OPC (Islander *et al.* 1991; Okabe *et al.* 2007). The specimens were studied for microbial diversity by SSCP analyses in order to have an overview of biodiversity without identification of microorganisms involved in the process. Figure 2 shows the prokaryotic SSCP spectra of two mortars exposed during four months. On such a graph, each peak represents a species and the peak height reflects the relative abundance of the individual among the population. The prokaryotic (Figure 2) and eukaryotic (Figure 3) biodiversity appears to differ depending on the cement type and it evolves completely differently over time (data not shown). However, as only single specimens were analysed on this instance, definite conclusions regarding differences in the microbial community compositions cannot be drawn from this study.

Hence, the matrices generated by the two cement types have a role in microbial selection but they also have a role in the amount of biomass developing on the surface of materials as shown in the DNA concentration extracted from biofilms at several sampling time (Table 4).

Aluminium is known to have a potential bacteriostatic effect (Illmer & Erlebach 2003). The aluminium ion dissolved in the condensed water in the PVC tubes of the specimens was monitored, regardless of the formulation, in order to ascertain its impact on bacterial activity. The evolution of ATP as function of total dissolved aluminium (Figure 4) shows that there is an inhibition of microorganisms at 350 mg L^{-1} of aluminium ion, also reported by Ehrich (1998) for a similar environment.

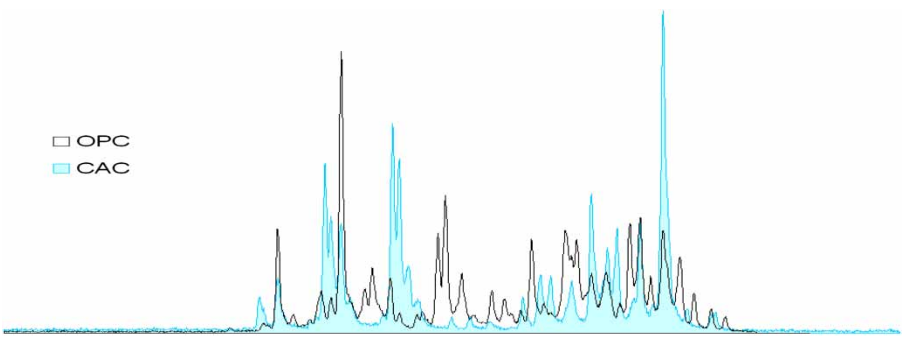

Figure 2 | SSCP diagram of bacterial DNA extracted on OPC and CAC mortars after an exposition of 4 months.

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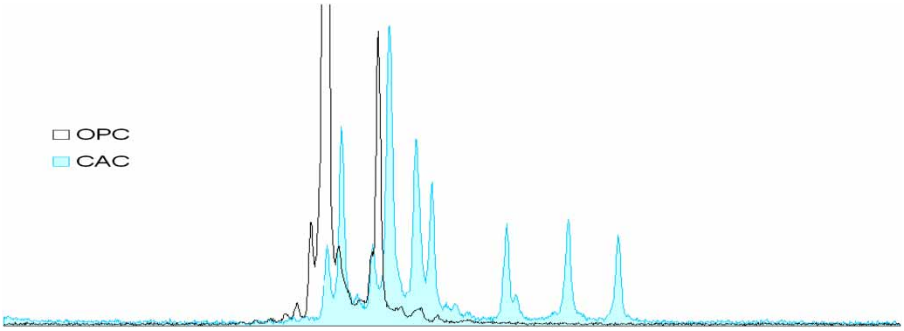

Figure 3 | SSCP diagram of eukaryote DNA extracted on OPC and CAC mortars after an exposition of 8 months.

Table 4 | Total DNA content (in ng cm^{-2} of material) extracted on the material surface exposed

	T 0	4 months	9 months	1 year	1.3 years
OPC	0.8	10.3	1.3	70.6	16.9
CAC	0.5	3.5	3.1	8.1	3.8

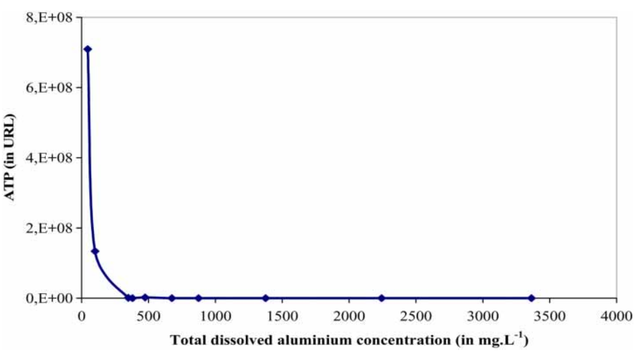

Figure 4 | ATP measurement on *in situ* specimens condensed water of OPC and CAC as a function of total aluminium ions concentration at low pH (1–2) after an exposition year.

The difference of durability between OPC and CAC materials is due to a difference in colonisation resulting from the presence of aluminium ions at the surface of CAC materials. During biogenic acid attack, CAC hydrates react to form an alumina gel (AH_3, in cement chemist notation) which is stable between pH 9 and 3–4 (Figure 5). Below this acidic threshold, the alumina gel dissolves releasing aluminium ions that likely have a bacteriostatic effect on some microorganisms (Figure 4; Herisson 2012). The stability of this layer over a wide pH range ensures a better acid neutralization capacity and prevents the establishment of acidophilic microorganisms like *Acidithiobacillus thiooxidans*, known to be one of the greatest acid producer responsible for deterioration (Parker 1947; Roberts *et al.* 2002). Another advantage of CAC materials is that alumina gel will precipitate in the porosity and on the surface limiting acid penetration hence protecting the material from the aggressive environment (Fryda *et al.* 2010). The formation of this AH_3 layer makes the surface smoother and could limit the biofilm adhesion.

CAC materials may be wrongly excluded from a use in sewer networks because they are not resistant in highly acidic condition (pH 1–2). However, due to their particular properties they can perform better than OPC materials. According to Scrivener & De Belie (2013) chemical acid attack and biogenic acid attack involved two different processes because, as one can see with the case of CAC materials, the material matrix could have an influence on the growth of microorganisms, hence on the acid production. Moreover, the deteriorated layer plays a different role in the two phenomena (Alexander & Fourie 2011). For chemical acid attack, this layer becomes a beneficial barrier against acid penetration, limiting exchanges. However for biogenic acid attack, a deteriorated layer composed mainly of gypsum, creates excellent conditions for the growth of microorganisms. They penetrate into this soft layer and produce sulfuric acid close to the intact material. Under these conditions, the deteriorated layer contributes to the aggression of the parent cement material.

Identification of durable materials in sewer networks needs a laboratory accelerated test that mimics the conditions of these aggressive structures involving a range of selected microorganisms. A few groups have worked to design an accelerated test (Hormann *et al.* 1997; Vincke *et al.* 1999) but they are very different from real conditions except that of Hamburg University (Ehrich 1998) which is out of operation. According to Scrivener & De Belie (2013), the Hamburg accelerated test is the most representative test of bacteriogenic deterioration developed to date but it used as the inoculum sludge from the Hamburg sewage system. Aiming at standardizing this method, DNA extracted on site from biofilms at each sampling will be used to

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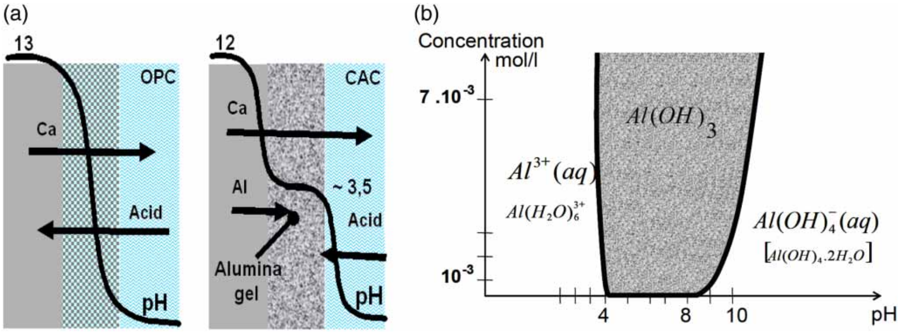

Figure 5 | a – Schematic representation of acid attack on OPC and CAC (Fryda *et al.* 2010); b – Stability diagram of alumina ($Al(OH)_3$) (Lamberet *et al.* 2008).

perform pyrosequencing analyses in order to identify microorganisms probably involved in the biodeterioration process. Using those interesting microorganisms in such a test, instead of an unknown microbial consortium sampled from a local sewer system, and growing them properly, will accelerate biodeterioration phenomenon of any materials sensitive to biogenic acid. This reproducible test will allow ranking materials durability within almost one year.

CONCLUSION

Sewer networks are prone to many deterioration causes including, in the presence of H_2S, biodeterioration of cementitious materials by acid produced by microorganisms. This complex phenomenon has been studied in detail for OPC materials and mechanisms are suggested in the literature but these mechanisms are not accurate for CAC materials.

Experiments confirm that, CAC materials are more durable on account of their composition. They are based on calcium aluminate hydrates which react with acid resulting in the precipitation of an alumina gel. This alumina layer creates a physical barrier to acid penetration, stabilizes pH around alumina solubility threshold (3–4) and has a bacteriostatic effect. This study highlights that active biofilms will not readily colonize CAC materials surfaces and will not evolve as biofilm developed on OPC materials.

Considering the necessity to build and repair sewer networks subjected to biogenic deterioration, there is a necessity to design a representative laboratory test that mimics realistic deleterious conditions. This test must involve selected acid producing microorganisms able to grow on cementitious materials in order to observe the interaction between cementitious matrix and biofilm development with optimized conditions acquired based on *in situ* experiment. Eventually, such a reproducible test will be a way to try any new material before using them in sewer networks.

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