



## Degradation modeling of concrete submitted to biogenic acid attack



Haifeng Yuan<sup>a,b,\*</sup>, Patrick Dangla<sup>a</sup>, Patrice Chatellier<sup>c</sup>, Thierry Chaussadent<sup>c</sup>

<sup>a</sup> Université Paris-Est, UMR Navier, UMR8205 CNRS/École des Ponts ParisTech 2 Allée Kepler, 77420 Champs-sur-Marne, France

<sup>b</sup> Vanke Architecture Research Center, 8 Gongye East Road, 523808 Dongguan, China

<sup>c</sup> Université Paris-Est, IFSTTAR, 14-20 Boulevard Newton, Cité Descartes, 77447 Marne la Vallée Cedex 2, France

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### ABSTRACT

Biodeterioration of concrete, which is very common in sewer system, results in significant structure degradation. The process can be described by the 3 following steps: Concrete surface neutralization providing appropriate environment for sulfur oxidizing bacteria (SOB) to grow, sulfuric acid ( $H_2SO_4$ ) production by SOB on concrete surface and chemical reaction between  $H_2SO_4$  and cement hydration products. A reactive transport model is proposed to simulate the whole biodeterioration processes of concrete in contact with  $H_2S$  gas and SOB. This model aims at solving simultaneously transport and biochemistry/chemistry in biofilm and concrete by a global coupled approach. To simulate the neutralization process, the absorption of  $H_2S$  gas, the dissolution of portlandite (CH), the decalcification of calcium silicate hydrates (C–S–H) and the precipitation of calcium sulfide (CaS) are considered. To obtain the amount of biogenic acid, the production rate of  $H_2SO_4$  by SOB is calculated via a set of simplified models governed by pH. Coupling with the modeling of  $H_2SO_4$  degradation process, the biodeterioration depth and the solid composition evolution could be predicted. A laboratory experiment reported in literature is simulated and the simulation results are compared with experimental results.

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### 1. Introduction

Huge amount of concrete and cementitious materials is used in the wastewater systems. Some of the microorganisms in sewer pipes can generate aggressive aqueous solutions which may damage cementitious materials and reduce the material service time. It was estimated that the United States will need 390 billion dollars during the next 20 years to repair the existing wastewater infrastructure [1]. Thus, there is great interest in predicting the corrosion rate and the service life of sewer pipes.

The most prominent biodegradation is biogenic sulfuric acid (BSA) corrosion which can be found in sewer pipes containing  $H_2S$  gas. The mechanism of such biodeterioration of concrete can be briefly described by the following three steps:

Step (1) Neutralization of concrete surface: The pH of fresh concrete (11–13) is too high for the sulfur-oxidizing bacteria (SOB) to survive. However,  $H_2S$  gases can be absorbed into concrete surface to react with portlandite (CH)<sup>1</sup> and calcium silicate hydrate (C–S–H) which are the main Portland cement hydrates. Consequently, the pH of concrete surface can be reduced to less than 9 [2,3].

Step (2) Production of  $H_2SO_4$  by SOB: Once the pH of concrete surface is reduced to 9, some strains of SOB start to grow and form biofilms on the concrete surface [4,5]. Although the bio-activity in biofilms is not fully understood, it is generally believed that firstly  $H_2S$  in biofilms is slowly oxidized to  $H_2SO_4$  by neutrophilic sulfur-oxidizing microorganisms (NSOM) [6]. After pH has decreased to 4–5, acidophilic sulfur-oxidizing microorganisms (ASOM) produce large amount of  $H_2SO_4$ , which makes the main contribution to the biodeterioration of sewer pipe [7,8].  $H_2SO_4$  solution and sulfate is destructive to concrete. With sufficient sulfur source, the pH of concrete surface can be even reduced to 1 [5].

Step (3)  $H_2SO_4$  attack of concrete: Biogenic sulfuric acid penetrates into concrete and reacts with CH and C–S–H. The ample supply of sulfate and the intrusion of the acid could result in the formation of ettringite ( $3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O$ ) which is expansive [5,9]. However, some experimental observations [10,11] found that the boundary between corrosion products and uncorroded concrete is quite clear and ettringite was not present. According to the investigations conducted by Gabrisova et al. [12,13], ettringite starts to form at pH ranging from 12.5 to 12. When pH decreases below 10.7, ettringite starts to decompose into gypsum. Since normally pH in biofilms is less than 3 [15], ettringite exists temporarily and occasionally appears in the corrosion products during the biodeterioration of sewer pipes. Therefore, in this study only the precipitation of gypsum is considered during the sulfuric

\* Corresponding author at: Université Paris-Est, UMR Navier, UMR8205 CNRS/École des Ponts ParisTech 2 Allée Kepler, 77420 Champs-sur-Marne, France. Tel.: + 86 18319040310. E-mail address: [marsgarnett@gmail.com](mailto:marsgarnett@gmail.com) (H. Yuan).

<sup>1</sup> Cement chemistry notation is used throughout the paper: C = CaO, S =  $SiO_2$ , H =  $H_2O$ , and  $\bar{S}$  =  $SO_3$ .

acid attack process which takes place between the concrete surface and biofilm–cement matrix interface. This step is characterized by the production of a corroded layer consisting of gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ , noted as  $\text{C}\bar{\text{S}}\text{H}_2$ ), silica gel ( $\text{SiO}_2$ , noted as S) and moisture [14,16].

This step could decrease the performance of concrete since gypsum has barely no strength.

For the chemical corrosion process, which directly results to the degradation of materials, our previous research [17] has proposed a reactive transport model to predict the behavior of Portland cement concrete in contact with a given  $\text{H}_2\text{SO}_4$  solution. However, in such a biogenic degradation process, the concentration of sulfuric acid is not constant or given, but dominated by the biochemical reactions in the SOB community. Thus the biochemistry reactions taking place in the biofilm system (step (1) and step (2)) should be simulated as well. Several models were reported to simulate the biochemical process, such as “Wastewater Aerobic/anaerobic Transformations in Sewer (WATS)” model [18] and sulfide oxidation model [19].

Yet, to our knowledge, no attempt has been made to model the neutralization of concrete surface and few models focus on the coupling of biochemical process and chemical corrosion process. With HYTEC, De Windt et al. [20] modeled a bioleaching test applied to ordinary Portland cement pastes during 15 months. But the production rate of  $\text{H}_2\text{SO}_4$  with different pH, diffusion of different species, equilibrium between the different solid phases, and the coupling between transport and reactions were not investigated specifically.

In this paper, a set of reactive transport modeling is expected to simulate the neutralization process of concrete surface and  $\text{H}_2\text{SO}_4$  production in SOB community. Thus, the reduction of pH of concrete surface and the pH evolution in SOB community can be predicted. Coupling with the  $\text{H}_2\text{SO}_4$  degradation modeling of concrete, the change of pH in SOB community and the composition of pipe concrete are expected to be calculated during the biodeterioration process. Furthermore, the degradation depth of concrete submitted to biogenic acid attack will be predicted. Simulations results of a set of accelerated microbiological tests conducted by De Muynck et al. [21] are presented and compared with experimental results.

## 2. Modeling of the neutralization process of concrete

The initial pH of ordinary Portland cement concrete is too high for sulfur-oxidizing bacteria (SOB) to grow. Rigdon et al. [4,5] proposed that the microbial activity on the concrete surface is initiated at pH around 9. Thereafter, a succession of microbial communities, which can utilize sulfide and/or its oxidized forms (such as element sulfur [3]) develops. The abiotic pH reduction of concrete surface results to an initial lag period before the start of active corrosion phase [22].

Therefore, the primary stage is the reduction of pH at the concrete surface, where pH decreases from about 12.5 to 9 by chemical acid reactions of  $\text{CO}_2$  and  $\text{H}_2\text{S}$  with concrete. However, the experimental research conducted by Joseph et al. [3] suggested that  $\text{H}_2\text{S}$  gas is the major factor for the surface pH reduction in sewers during early stages of exposure rather than carbonation. Thus this study concentrates on the effect of  $\text{H}_2\text{S}$ . Similar consideration was taken in the study of Lin [23].

### 2.1. Absorption of $\text{H}_2\text{S}$

In sewer pipes,  $\text{H}_2\text{S}$  can be absorbed from gas phase into pore solution of concrete and biofilms from gas phase. Since only aqueous  $\text{H}_2\text{S}$  can react with concrete or be oxidized by SOB, the absorption of  $\text{H}_2\text{S}$  governs the time of surface neutralization and the production of  $\text{H}_2\text{SO}_4$ . Therefore, the transferring of  $\text{H}_2\text{S}$  from sewer gas to concrete surface or biofilms is a crucial step.

The overall removal of hydrogen sulfide from a sewer atmosphere depends on numerous conditions, such as  $\text{H}_2\text{S}$  concentration levels, temperature, and relative humidity of the sewer atmosphere [3,25].

Richard et al. [24] suggested that all released  $\text{H}_2\text{S}$  from wastewater is immediately absorbed by concrete surface, resulting in zero  $\text{H}_2\text{S}$  in the gas phase. However, experimental results of Vollertsen et al. and Nielsen et al. [25,26] revealed that transferring of  $\text{H}_2\text{S}$  from the gas phase to the concrete surface is not an instantaneous process and absorption kinetics should be taken into account. By experiments with pilot-scale sewer reactors, Vollertsen et al. [25] found that the absorption rate ( $F_{\text{H}_2\text{S}}$ ) could be described as a power function in the gas phase  $\text{H}_2\text{S}$  concentration ( $p_{\text{H}_2\text{S}}$ ).

Furthermore, it is found that the corrosion is more severe at downstream of some particular areas e.g. force main discharge points or drop structures, where release rates are higher than in other parts of the sewer [27]. Nielsen et al. [28] studied the influence of the air-flow in sewer on the absorption of  $\text{H}_2\text{S}$ . Increased air-flow will provide better mixing of the sewer atmosphere, as well as reduce the thickness of the diffusive boundary layer near surfaces.

Nielsen et al. [28] presented an empirical relationship between the Reynolds (Re) number of the gas flow and the absorption rate. For the sake of simplicity, the effect of gas flow regime and temperature are ignored in this paper. The  $n$ th order kinetics of  $\text{H}_2\text{S}$  absorption of concrete surface and biofilms is described as follows:

$$F_{\text{H}_2\text{S}} = k_{\text{abs}} p_{\text{H}_2\text{S}}^n \quad (1)$$

where,  $p_{\text{H}_2\text{S}}$  is the content of  $\text{H}_2\text{S}$  gas (ppm).  $k_{\text{abs}}$  is the surface specific  $\text{H}_2\text{S}$  absorption rate constant, which is reported to vary between  $6.25 \times 10^{-8}$  and  $3.12 \times 10^{-7} \text{ mol S m}^{-2} \text{ s}^{-1} (\text{ppm H}_2\text{S})^{-n}$  depending on temperature [25]. The reaction order  $n$  is correlated with  $k_{\text{abs}}$ , with  $n$  approximately 0.5 for low  $k_{\text{abs}}$  and 0.8 for high  $k_{\text{abs}}$ . However, the quantitative relationship among  $k_{\text{abs}}$ ,  $n$  and temperature is still unclear. According to the study of Joseph et al. [3], the rate of neutralization process significantly enhanced by the increase in temperature from 16 °C to 30 °C. Since temperature is constant and equal to 25 °C in this study,  $k_{\text{abs}}$  is determined as  $2 \times 10^{-7} \text{ mol S m}^{-2} \text{ s}^{-1} (\text{ppm H}_2\text{S})^{-n}$  and  $n = 0.75$ . Then the absorption rate of  $\text{H}_2\text{S}$  versus content of  $\text{H}_2\text{S}$  in gas phase is plotted in Fig. 1. With a higher concentration,  $\text{H}_2\text{S}$  gas will be absorbed into concrete surface faster.

### 2.2. Chemical reactions of aqueous $\text{H}_2\text{S}$ and concrete

Aqueous hydrogen sulfide dissociates in pore solution and releases  $\text{H}^+$ . As Portland cementitious materials, portlandite (CH) and calcium silicate hydrates (C–S–H) are the solid phases able to react with  $\text{H}_2\text{S}$ , calcium hydroxide can be dissolved and calcium sulfide (CaS) forms

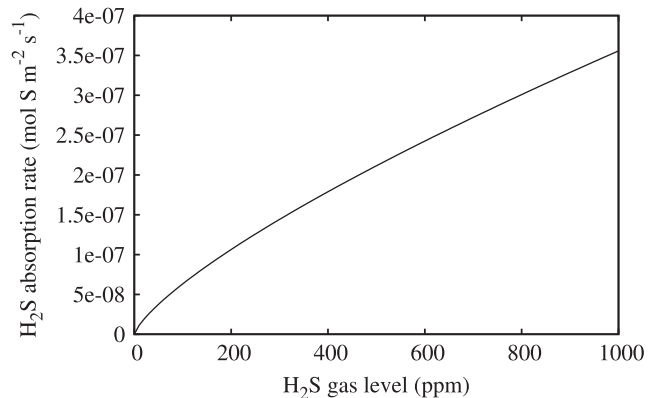


Fig. 1.  $\text{H}_2\text{S}$  absorption rate at different  $\text{H}_2\text{S}$  gas contents.

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