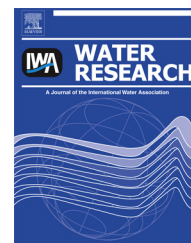


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# The role of iron in sulfide induced corrosion of sewer concrete



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## ABSTRACT

The sulfide-induced corrosion of concrete sewer is a widespread and expensive problem for water utilities worldwide. Fundamental knowledge of the initiation and propagation of sewer corrosion, especially the interactions between chemical reactions and physical structure changes, is still largely unknown. Advanced mineral analytical techniques were applied to identify the distribution of corrosion products and the micro-cracking that developed along the corrosion boundary. It was found that sewer concrete corrosion caused by reactions with sulfuric acid progressed uniformly in the cement of concrete. In contrast to conventional knowledge, iron rust rather than gypsum and ettringite was likely the factor responsible for cracking ahead of the corrosion front. The analysis also allowed quantitative determination of the major corrosion products, i.e., gypsum and ettringite, with the latter found closer to the corrosion front. The conceptual model based on these findings clearly demonstrated the complex interactions among different chemical reactions, diffusion, and micro-structure changes.

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## 1. Introduction

Concrete corrosion is a major deteriorating process affecting sewer systems worldwide. Corrosion causes loss of concrete mass, cracking of the sewer pipes and ultimately, structural collapse. The rehabilitation and replacement of corrosion damaged sewers involves very high costs. It is estimated that the annual cost of concrete corrosion within the water and wastewater infrastructure is about US\$ 36 billion in USA (Koch et al., 2002). This cost is expected to increase as the aging infrastructure continues to fail (Sydney et al., 1996; US EPA, 1991).

Concrete corrosion in sewers is a result of a range of abiotic and biotic processes. The corrosion proceeds from carbonation (CO<sub>2</sub>) and H<sub>2</sub>S acidification during early stages, to biological production of sulfuric acid from oxidation of hydrogen sulfide present in the sewer gas phase (Parker, 1945a,b; 1947; Pomeroy and Bowlus, 1946) during later stages, which causes mass loss of concrete (Islander et al., 1991; Ismail et al., 1993). Recent studies identified various microorganisms, including both bacteria and fungi, which are involved in acid production (Hernandez et al., 2002; Kelly and Wood, 2000; Okabe et al., 2007). For example, *Acidithiobacillus thiooxidans*

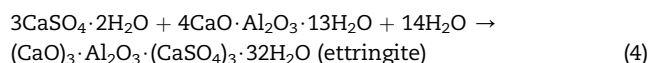
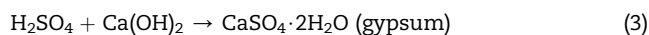
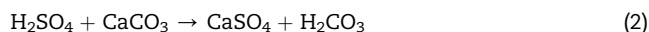
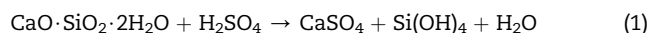
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**Nomenclature**

BSE	Backscattered electron
EDS	Energy dispersive spectroscopy
MLA	Mineral liberation analyzer
RTD	Resistance temperature detector
SEM	Scanning electron microscope

has been shown to be a key organism involved in the oxidation of hydrogen sulfide is corrosion biofilms (Jensen et al., 2008; Kelly and Wood, 2000). A range of corrosion products forms during various stages of the corrosion process. The components of uncorroded cement are mainly hydrated calcium silicate ( $\text{CaO}\cdot\text{SiO}_2\cdot 2\text{H}_2\text{O}$ ) and portlandite ( $\text{Ca}(\text{OH})_2$ ). Abiotic processes that include carbonation and  $\text{H}_2\text{S}$  acidification, result in  $\text{CaCO}_3$ , and  $\text{Ca}(\text{HS})_2$  and  $\text{S}^0$  as the main products (Parker, 1947). The initial acidification processes can reduce the pH of concrete sewer from around 12 to 9, this can provide favorable conditions for the propagation of sulfur and sulfide oxidizing microorganisms on the concrete surface (Joseph et al., 2012). These oxidizing microorganisms then develop and cause more active production of sulfuric acid, which leads to the formation of two important corrosion products: gypsum and ettringite, according to the reactions below.



Both gypsum and ettringite have significantly higher volumes than intact cement, estimated to range from 124% to 700% (Monteny et al., 2000; Parande et al., 2006). The expansion is believed to cause internal cracking and pitting, which in turn, exposes more surface area for acid attack. It is thus essential to identify how the formation of expansive corrosion products correlates with the physical cracking processes. Traditionally, research has focused on concrete cracking that is caused by the rust forms by rebar corrosion in the reinforced concrete (Wei et al., 2010; Zhao et al., 2012). This is usually caused by chloride other than  $\text{H}_2\text{S}$  induced corrosion. Micro-cracking at the corrosion front is another important feature of concrete corrosion. This occurs on a smaller scale, compared to the big cracks caused by rebar rust, which leads to concrete surface spalling.

This study aims to enhance understanding of the correlation between corrosion products and the microstructure of concrete. In particular, how the nature, amount and distribution of the corrosion products correlate to the corrosion propagation. Using concrete samples exposed to simulated sewer conditions with controlled  $\text{H}_2\text{S}$  concentration, relative humidity (RH) and temperature, the formation of major

corrosion products on the transition zone from a corrosion layer to the intact concrete core was measured. Especially, profiles of different corrosion products formed at different depths inside concrete coupons were determined using advanced mineral analytical techniques including mineral liberation analyzer (MLA) and energy dispersive spectroscopy (EDS). The results provide new insights into the temporal development of corrosion, leading to a new conceptual model for concrete corrosion.

## 2. Material and methods

### 2.1. Concrete coupons

The concrete coupons were prepared from corroded concrete slabs obtained from a main trunk sewer of Sydney Water Corporation, Australia. The dimensions of coupons were approximately 100 mm × 70 mm × 70 mm (thickness). The concrete has been under active corrosion, with about 2–4 mm of corrosion products on the surface. After cutting with a water-cooled saw, the coupons were washed in fresh water to remove corrosion layer and any surface contamination. Washed coupons were then dried in an oven (Thermotec 2000, Contherm) at 60 °C for 3 days to achieve similar and stable initial water content (Joseph et al., 2010).

One of the original surfaces of the coupons, i.e. the internal surface of the pipe, was designated as the experimentally exposed surface. After cutting, the coupons were embedded in stainless steel frames using epoxy (FGI R180 epoxy & H180 hardener) with the steel frame providing a reference point for determining the change in thickness due to corrosion (Fig. 1).

### 2.2. Corrosion chamber and exposure condition

The corrosion chamber was constructed to achieve a controlled environment simulating that of real sewers. The chamber was constructed of glass panels of 4 mm thickness. The dimensions of the chamber are 550 mm (L) × 450 mm (D) × 250 mm (H) (Fig. 2). It contained 2.5 L of domestic sewage that was collected from a local sewer pumping station and replaced every two weeks. The coupons were exposed to the gas phase within the chambers with the exposed surface facing downwards approximately 110 mm above the sewage surface. This coupon arrangement simulated the sewer pipe crown, a location which is reported to be highly susceptible to sulfide induced corrosion (Mori et al., 1992; Vollertsen et al., 2008).

To achieve the specified  $\text{H}_2\text{S}$  gaseous concentration, i.e. 50 ppm, in the corrosion chamber,  $\text{Na}_2\text{S}$  solution was injected into a container partially filled with acid (13% HCl), using a corrosion-resistant solenoid pump (Bio-chem Fluidics, model: 120SP2440-4TV) with a dispense volume of 40  $\mu\text{L}$ . The  $\text{H}_2\text{S}$  concentration was monitored using a  $\text{H}_2\text{S}$  gas detector (Oda-Log Type 2) with a range between 0 and 200 ppm (App-Tek International Pty Ltd, Brendale, Australia). A programmable logic controller was employed to monitor the  $\text{H}_2\text{S}$  concentration and to trigger the dosing pump for  $\text{Na}_2\text{S}$  addition to maintain the specified  $\text{H}_2\text{S}$  concentration at approximately 50 ppm.

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