



Study on the degradation mechanism of sulphoaluminate cement sea sand concrete eroded by biological sulfuric acid



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HIGHLIGHTS

- Sea sand and sulphoaluminate cement (SAC) were used to make the concrete.
- Biological sulfuric acid (BSA) was applied to simulate the corrosion.
- The degradation mechanism of SAC sea sand concrete eroded by BSA was analyzed.
- The BSA had a more negative effect on performance of SAC sea sand concrete.

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ABSTRACT

Two different types sulfuric acids, i.e. the biological sulfuric acid (BSA) produced by T.t bacteria (*Thiobacillus thiooxidans*) and chemical sulfuric acid (CSA), were applied to simulate the corrosion. The sulphoaluminate cement (SAC) and the sea sand were mixed to make the concrete. The mass loss, compressive strength, chloride binding rate, pH value and pore structure were measured in the process of corrosion. Furthermore, X-ray diffraction (XRD) and scanning electron microscopy-energy dispersive spectroscopy (SEM-EDS) were used to analyze the products of the reaction. It could be found that under the corrosion of sulfuric acid, the initial hydration products of SAC, including ettringite (AFT) and Friedel's salt, were decomposed continuously and the main corrosion product was dihydrate gypsum. Under corrosion of BSA, the corrosion became more intense due to the influence of bacterial effect. Moreover, the corrosion of BSA was more severe than that of CSA, which was obvious in terms of mass loss, strength decline and pore structure. The BSA has a more negative effect on the performance of SAC concrete.

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1. Introduction

At present, extensive attention has been attracted on the corrosion degradation of concrete structures induced by microbe. In the United States, approximately 10% of the sewer pipes are subjected to corrosion due to biological sulfuric acid attack. In Germany, microbial corrosion has caused 10–20% of the damage of building materials [1–3]. Microbes that produce sulfuric acid could accelerate the deterioration of concrete. In order to avoid the huge cost of building maintenance, researchers began to study the microbial corrosion on concrete. Parker [4] has found that in the marine engineering, one of main reasons responsible for the concrete corrosion is the biological sulfuric acid (BSA) corrosion. BSA is a kind of sulfuric acid, which is produced by the microbial decomposition of sulfur-containing organic matter and sulfur-containing inorganic

compounds. The Acidophilic sulfur-oxidizing microorganisms (ASOM), which is the major acid producing bacteria, has been confirmed to be attached to the surface of concrete [4,5].

Compared to ordinary portland cement (OPC), the sulphoaluminate cement (SAC) shows lower energy consumption, environmental benefits and many engineering advantages. In the terms of durability, SAC could effectively reduce the chloride penetration and resist the erosion of sulfate. Research [6] has indicated that chloride migration coefficient of SAC concrete is distinctly smaller than that of OPC, especially at early stage. With better compactness and durability [7–10], SAC concrete has been widely used in marine environment, especially in coastal cities.

Because of the lack of river sands, sea sand has been considered to use in the industrial and residential buildings. Some studies indicated that use sea sand instead of river sand to make concrete would not affect the mechanical property of concrete [11], and could improve the impermeability of concrete [12]. Fu [13] has found that using sea sand in marine concrete structures is practicable. The artificial

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island of Chek Lap Kok, where Hong Kong airport is located, was constructed with marine aggregates [14]. Chen et al. [15] reported that SAC concrete prepared with sea sand gave a good workability and mechanical properties. It is expected that compared to OPC sea sand concrete, the SAC sea sand concrete could have high corrosion resistance and better durability, particularly in marine environment.

For concrete under the attack of sulfuric acid, it has been proved that the corrosion of sulfuric acid can decrease the ultimate bending tensile strength and the elastic modulus of steel fiber concrete [16]. There are some studies about sewer pipe made by concrete under BSA corrosion [17,18], but little research on the corrosion of SAC sea sand concrete by BSA was performed. Since BSA corrosion is a complex and long-term process, it is incorrect to investigate the corrosion of concrete only by chemical sulfuric acid (CSA). It showed significant differences between BSA and CSA [19–22]. This paper used T.t bacteria (*Thiobacillus thiooxidans*, T.t) to simulate the BSA corrosion. CSA was used as a contrast to study the corrosion effect of BSA on SAC sea sand concrete in order to reveal the mechanism of SAC sea sand concrete by BSA corrosion.

2. Experimental

2.1. Raw materials

This study used sulphoaluminate cement (SAC 42.5), and its mineral composition is shown in Table 1. The coarse aggregate was ordinary stone, which was selected in accordance with GB/T 14685–2001 [23]; Fine aggregate was sea sand, each performance index measured according to GB/T 14684–2001 [24] is listed in Table 2. A superplasticizer (SP) KDSP-1 (Polycarboxylate retarding type) with a water-reducing ratio of 25% was used. A retarder for SAC, produced by Tianjin BASF Chemicals Co. Ltd, was used.

2.2. Mix proportion

The mix proportion of C50 sea sand concrete was designed according to the method of ordinary concrete [25] and practical experience. The dosage of the water reducing agent and the retarder were 1.1% and 0.3% of the cement mass respectively. Mix proportion is shown in Table 3. The size of the sample was $100 \times 100 \times 100 \text{ mm}^3$. The samples were demolded after 24 h and standard cured for 28 days. Then the samples were taken out in the room to air dry for 2 days. After that, the samples were placed in the prepared CSA solution and BSA solution for corrosion test.

2.3. Simulated sulfuric acid corrosion method

According to the Berger Bacteria Identification Manual [26], *Thiobacillus thiooxidans* (T.t) was selected as the representative bacterium of ASOM. The bacterial culture medium components were: $(\text{NH}_4)_2\text{SO}_4$, CaCl_2 , K_2HPO_4 , $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ and sublimation sulfur. The concentration of each component is shown in Table 4.

For the BSA, different amount of sublimation sulfur was added to the culture medium to simulate different pH value of the BSA corrosion. The samples were soaked in T.t bacterial medium, and the medium was inoculated with 10% T.t bacteria. In the initial stage, the culture medium could acidify the concrete. When concrete was acidified to the appropriate degree, the T.t bacteria could adhere to the concrete surface and produce sulfuric acid to corrode the concrete. For the CSA, the samples were placed in a simulated acid rain corrosion test chamber and soaked into CSA solution. The pH value of the solution was monitored daily with a pH meter.

In the following test results, the first letter H in all figures represents the CSA corrosion, S represents the BSA corrosion; the second letter S represents SAC sea sand concrete; 1.0, 1.5, 2.0 represents different pH value.

2.4. Test methods

The mass loss and compressive strength of the samples under the corrosion of CSA and BSA were tested for every 7 days. Each group was measured as a set of three samples. The thickness of corrosion layer studied in this study was based on some previous research which claimed that the surface peeling depth of concrete is less than 5 mm [27,28]. The samples were cut with a thickness of 5 mm from the concrete surface. The mortar was knocked out from the cut corroded layer and grounded into a powder, then sampled for pH value and chloride binding rate test. The pH value of the samples was measured by pH meter produced by DENVER INSTRUMENT, USA. Besides, the free chlorine content (C_f) in each sample was measured according to the chemical titration method [29]. The total chloride ion content (C_t) of the corrosion layer was calculated according to the chloride content in sea sand. The chloride binding rate (W_b) was calculated per Eq. (1):

$$W_b = \frac{C_t - C_f}{C_t} \times 100\% \quad (1)$$

After that, the cement picked from the fragments of the corrosion layer was knocked into particles with a diameter of about 3 mm by a hammer. After removing the impurities, the cement particles were put into a sample tube, then placed in the ASAP 2020M automatic surface and pore size analyzer made by Micrometric Company to measure the pore structure.

In this study, the hydration products of concrete cured for 28 days and the corrosion products at the end of the corrosion were tested by the Philips X/Pert Pro MPD X-ray diffraction (XRD), Netherlands. The pore structure of the SAC sea sand concrete under two kinds of corrosion was observed and analyzed by S-3400N scanning electron microscopy (SEM), Japan.

3. Results and discussion

3.1. Variation of mass loss

Fig. 1 shows the mass loss of SAC sea sand concrete corroded by BSA and CSA. As can be seen from the Fig. 1, when the pH = 1.0, the mass loss was the most serious in CSA and BSA. The mass decreased by 5.82% after corroded by CSA for 84 days. While under BSA corrosion, the concrete mass decreased by 8.21%. Some studies indicated that the hydration products such as C-S-H gel, Friedels' salt and CaCO_3 in corrosion layer could be decomposed by sulfuric acid and to produce CaSO_4 [30]. The concrete surface gradually peeled off and eventually led to the mass loss.

The corrosion of sulfuric acid is a chemical reaction in which both the erosion of H^+ and the precipitation of SO_4^{2-} are combined [28,31]. Under the corrosion of CSA, the mass loss was not obvious at the beginning. After 42 days, the mass loss suddenly increased and the loss amplitude was greater. This showed that with the increase of time, the erosion of H^+ had been greater than the precipitation of SO_4^{2-} . With lower pH value, more H^+ occurred and the erosion by H^+ became more serious, so the mass loss increased. In the case of BSA corrosion, the mass of concrete decreased linearly. This indicated that concrete had always been corroded by sulfuric acid in the test period due to the bacterial effects. The bacterial activity was higher due to the lower pH value which means more bacterial can be produced resulting in more production of sulfuric acid. So the BSA has a more detrimental effect on the mass loss of concrete.

3.2. Variation of compressive strength

The compressive strength of SAC sea sand concrete corroded by BSA and CSA is shown in Fig. 2. Under the CSA corrosion, the compressive strength of the concrete was slightly improved in the first 21 days. Since the CaSO_4 precipitated into the internal pores at the beginning of the corrosion, the concrete compactness was

Table 1
Mineral composition of SAC (%).

Cement	C ₃ S	C ₂ S	C ₃ A	SO ₃	C ₄ AF	C ₄ A ₃ S
SAC	–	31.46	–	2.96	11.28	52.85

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