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Study on the applicability of bactericides to prevent concrete microbial corrosion

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HIGHLIGHTS

- $C_{32}H_{16}CuN_8$ is the best bactericide to prevent concrete microbial corrosion.
- NaBr has good effect on the resistance of concrete microbial corrosion in short term.
- Both ZnO and $C_{21}H_{38}NCl$ have adverse effect on the performance of concrete.
- The bactericidal ability of Na_2WO_4 is the worst.

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ABSTRACT

For a comprehensive evaluation of the effect of bactericide on the concrete microbial corrosion, five different types of bactericide were selected, the methods of high-throughput sequencing, inductively coupled plasma mass spectrometry, confocal laser scanning microscope and so on were employed, and their bactericidal ability, dissolving property, effective content as well as the influence on the fluidity and strength of concrete were studied. The results show that the addition of copper phthalocyanine can increase the fluidity and strength of concrete greatly, and its killing rate in sewage is 90.82%, and its retention rate in concrete is still as high as 99.69% after being immersed in sewage for 120 days, meanwhile, both the lower calcium dissolution content of specimens in sewage and a large number of dead microbes observed on concrete surface have proved that copper phthalocyanine is a good bactericide for preparing the anti-microbial corrosion concrete. Although the bactericidal rate of sodium bromide is only inferior to copper phthalocyanine, its retention rate in concrete is only 87.14% after 120 days immersion. However, the calcium leaching concentration results demonstrate a high inhibitory effect of sodium bromide on the deterioration of concrete structure in sewage in the short term. Moreover, it has no significant influence on the performance of concrete. Despite the high retention rates of both zinc oxide and dodecyl dimethyl benzyl ammonium chloride in concrete, their contents should be no more than 0.05%, otherwise, they will have a significant adverse effect on the fluidity and strength of concrete. The influence of sodium tungstate on concrete performance is least while its content is 0.1%, but its bactericidal ability is poor, it even promote the growth and reproduction of Proteobacteria, and has a higher cost, so it is not suitable for preparing the anti-microbial corrosion concrete.

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1. Introduction

In urban environmental engineering, most of the facilities for sewage transportation and treatment are reinforced concrete structure. They are generally underground and long term subjected to the acid [1,2], erosion [3], microbial [4,5] and other corrosive effect. The growing problem of concrete deterioration results in a huge economic losses, which has been paid more and more attention around the world. In Hamburg (Germany), back in 1970s, only the maintenance costs of concrete microbial corrosion (CMC) of sewer pipelines reached up to 25 million Euros. In Los Angeles (USA), the sewer pipe of 208km in a total length of 1900 km had been damaged by CMC, and the rehabilitation costs were as high as $400 million [6]. Not only the construction and replacement of sewer pipes is very expensive, but the failure of sewer pipes causes extensive damage to roads and pavements. In Zhengzhou (China),

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24 cases of road collapse frequently occurred within a year of 2015, which are mainly caused by the aging of underground sewage pipe network [7]. Therefore, it is necessary to pay more attention on the durability of concrete in sewage environment. Especially in China, with the accelerating process of urbanization in our country, a large number of municipal sewage projects are required to build, so the research of prevention of CMC has important practical significance.

As early as 1945, Parker discovered that bacteria were involved in the deterioration of internal sewer concrete, and proposed the corrosion mechanism is a result of the sulfur cycle, which has also been extensively accepted [8,9]. First, hydrogen sulfide (H_2S) is produced by sulfate-reducing bacteria (SRB) present in wastewater and sediments under anaerobic conditions. Then, part of H_2S reacts with atmospheric oxygen to produce elemental sulfur and thiosulfate in the headspace above the wastewater. Finally, sulfur-oxidizing bacteria (SOB) present in the condensate on the concrete surface oxidize the dissolved H_2S and other sulfur compounds to sulfuric acid, which corrodes the concrete.

Several methods have been developed to control CMC in wastewater structures. At present, there are mainly three kinds of measures, including concrete modification [10–13], coating protection [14–16] and biological killing [17–19]. The object of concrete modification is to improve the acid resistance, impermeability and crack resistance of concrete. However, the simple modification cannot reduce the corrosion of concrete significantly. Although the inert coating can be used to isolate the contact between concrete and aggressive medium, it has some defects like cracking, spalling and wearing, etc. Therefore, these two types of measures can only delay the concrete corrosion to a certain extent. The application of bactericides, which based on the microbial corrosion mechanism, is an initiative measure [20]. It is very effective to control the microbial induced concrete corrosion by preventing microbial reproduction and metabolism in sewage, and thus inhibiting or reducing the formation of biological acid. So far, the bactericides suitable for concrete, which have been reported by foreign patents, are halogenated compound, quaternary ammonium compound, heterocyclic amine, iodopropargyl compound, (copper, zinc, lead, nickel) metal oxide, (copper, zinc, lead, manganese) phthalocyanine, tungsten power or tungsten compound, silver, organic tin, and so on [21]. However, as a functional component in concrete, the selection of bactericide types and contents has not been systematically studied. Moreover, the long-term retention rate of bactericide and its influence on the other properties of concrete are also poorly understood.

In this study, five different types of bactericide with four different contents were determined based on the reported patents. To investigate the influence of bactericide on the performance of concrete itself, the workability of fresh concrete and compressive strength of hardened concrete were tested, respectively, as well as the compositions of cement paste. In addition, the sterilizing rate of bactericides in sewage was studied through the change of microbe species and quantity, which obtained from DNA sequencing. To study the retention rate of bactericides in concrete, the elements dissolution content of concrete specimens was also tested by inductive coupled plasma (ICP) and ion chromatograph. Furthermore, the dead and live microorganisms within biofilm developed on concrete surface were observed by using confocal scanning laser microscopy (CSLM). The results obtained are expected to provide some theoretical basis for the development of bactericides of efficiency, safe and not affecting concrete performance.

2. Experimental program

2.1. Materials

Grade 42.5 Ordinary Portland Cement was obtained from Dingxin Cement Limited, China. Class I fly ash was obtained from was obtained from Shijingshan Thermal Power Plant, Beijing, China. River sand with a fineness modulus of 2.8 and an apparent density of 2.61 g/cm^3 was obtained from Zhengding, and limestone aggregate with a specific gravity of 2.64 g/cm^3 and particle size between 5 and 20 mm was obtained from Luquan. Polycarboxylic superplasticizer was obtained from Ruidisi Building Materials Limited, China. The properties of all raw materials meet the requirement of standard. In addition, based on the considerations of bactericidal capability, less toxic, low pollution, low cost and other issues of bactericides, five different types of bactericide were selected for the study including sodium bromide, sodium tungstate, zinc oxide, copper phthalocyanine and dodecyl dimethyl benzyl ammonium chloride. Their specific indexes are listed in Table 1.

2.2. Mixture proportions

In this experiment, concrete strength grade was designed for C30 with water-cement ratio (w/c) of 0.47 according to common concrete strength in municipal engineering. The concrete mixture was composed of 278 kg cement, 64 kg fly ash, 160 kg water, 687 kg sand, 1154 kg coarse aggregates and 1.5 kg superplasticizer per cubic meter. The reference concrete without bactericide was designated as BC. The other five kinds of concrete mixed with sodium bromide, sodium tungstate, zinc oxide, copper phthalocyanine and dodecyl dimethyl benzyl ammonium chloride were designated as SBC, STC, ZOC, CPC and DDC, respectively. The contents of each bactericide are 0.05%, 0.1%, 0.2% and 0.4% of cementitious materials by weight, so that the effect of bactericides types and contents on concrete performance can be studied. Cement paste specimens with the same w/c ratio were prepared for the microscopic test.

2.3. Test methods

2.3.1. Performance of concrete

The workability of fresh concrete and strength of hardened concrete were tested according to the related contents of GB/T 50080=2002 "Standard for test method of performance on ordinary fresh concrete" (ASTM C143/C143M-15a) and GB/T 50081-2002 "Standard for test method of mechanical properties on ordinary

Table 1
Physical indexes of the bactericide.

Bactericide	Molecular formula	Appearance	Density (g/cm^3)	Solubility	Cost (Yuan/500 g)
Sodium bromide	$NaBr$	White crystal or powder	3.203	Soluble	20
Sodium tungstate	Na_2WO_4	White crystalline powder	3.23–3.25	Soluble	120
Zinc oxide	ZnO	White powder	5.606	Insoluble	15
Copper phthalocyanine	$C_{32}H_{16}CuN_8$	Blue powder	1.34–1.46	Insoluble	25
Dodecyl dimethyl benzyl ammonium chloride	$C_{21}H_{38}NCl$	Light yellow liquid	0.980	Soluble	13

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