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Cement and Concrete Composites

journal homepage: www.elsevier.com/locate/cemconcomp

Effects of Deoxyribonucleic acid on cement paste properties and chloride-induced corrosion of reinforcing steel in cement mortars



Shaobo Jiang^a, Song Gao^a, Linhua Jiang^{a,b,c,*}, Ming-Zhi Guo^a, Yu Jiang^d, Chen Chen^a, Ming Jin^a, Shuya Bai^a

^a College of Mechanics and Materials, Hohai University, Nanjing, 210098, PR China

^b National Engineering Research Center of Water Resources Efficient Utilization and Engineering Safety, PR China

^c Engineering Research Center on New Materials and Protection in Hydraulic, Jiangsu Province, 1 Xikang Rd, Nanjing 210098, PR China

^d Jiangsu Testing Center for Quality of Construction Engineering Co., Ltd., 210028, PR China

ARTICLE INFO

Keywords:

DNA
Corrosion inhibitor
Steel reinforced mortar
Electrochemical methods
Cement mechanical properties

ABSTRACT

The corrosion inhibition effect of Deoxyribonucleic acid (DNA) on steel reinforcement in cement mortar was investigated by electrochemical techniques, including linear polarization resistance (LPR) and electrochemical impedance spectroscopy (EIS). In addition, the effect of DNA on the mechanical properties of cement paste was also examined. The hydration products and pore-size distribution of DNA incorporated cement pastes were detected by X-ray diffraction (XRD) and Mercury Intrusion Porosimetry (MIP), respectively. The results revealed that the addition of DNA in cement mortar greatly improved the corrosion resistance of embedded steel reinforcement. Moreover, the incorporation of DNA in cement paste led to a decrease in the 3d compressive strength, but an increase in the 28d flexural strength. DNA, however, exerted a negligible effect on the cement hydration products and the 28d compressive strength. Instead, DNA induced positive effects on the pore-size distribution of cement paste, such as lowering the proportion of harmful pores and increasing harmless pores.

1. Introduction

The chloride-induced corrosion of reinforcing steel has been one of the most widespread damage phenomena of reinforced concrete structures (RCS), particularly in the coastal marine environment. Several prevention measures have been employed to reduce the corrosion risk, such as using improved materials (concrete surface coatings [1] and steel galvanisation [2]), using stainless steel [3], cathodic protection [4,5], steel coating [6], and adding corrosion inhibitors [7,8]. If corrosion has already occurred, repair techniques, such as realkalization, can be employed to repair the eroded reinforced concrete [9]. Among all the available methods, the use of corrosion inhibitors represents a promising and viable method due to their low cost, easy handling, and good corrosion inhibition effect [10–14]. However, the existing uncertainties regarding their long-term influence on the reinforcement corrosion process and the properties of concrete limit their wide applications.

Traditional corrosion inhibitors are normally divided into three groups: inorganic inhibitors, organic inhibitors, and mixed inhibitors. They can either be added into the fresh concrete during the mixing process or be migrated into the hardened concrete at a later stage. In

the past, nitrite-based inhibitors, typical inorganic inhibitors, have long been applied effectively in RCS. Nowadays, however, they are banned in many countries because of its toxicity [15–17]. Therefore, there is growing interest to develop environmentally friendly alternatives to replace the nitrite-based inhibitors. Several studies have concentrated on the development of organic inhibitors and mixed inhibitors, such as amines, aldehydes, and zincates [10,18,19]. Meanwhile, attempts have also been made to develop biological corrosion inhibitors, such as extracts from *Lannea coromandelica* and biomolecules derived from bacteria cells [20–24]. But the synthetic processes of most organic and biological inhibitors are too complicated.

It has been well known that the efficiency of the inhibiting action for steel is the most concern. But the influence of inhibitors on the properties of cement and concrete is equally important [25–27]. Fayala et al. [25] reported that inhibitors can make the concrete matrix more denser and promote a decrease in the chloride ion diffusion. Ann et al. [26] found that the addition of calcium nitrite based corrosion inhibitors increased the compressive strength at an early age. In contrast, Sun et al. [27] reported that the addition of inhibitors caused changes in the hydration products and thereby reduced the strength, which limits their applications. Therefore, there is an urgent need to develop

* Corresponding author. College of Mechanics and Materials, Hohai University, Nanjing, 210098, PR China.
E-mail address: lhjiang@hhu.edu.cn (L. Jiang).

non-toxic and long-term efficient inhibitors without adverse effects on the properties of cement and concrete. To achieve this goal, we have employed DNA as a new corrosion inhibitor and demonstrated that DNA greatly improved the corrosion resistance of steel reinforcement in simulated concrete pore solutions [28]. But it remains unclear whether DNA will still be effective in corrosion resistance when added in cement mortar, and how the properties of cement paste will be affected.

To the best of our knowledge, DNA was for the first time used as a corrosion inhibitor in cement mortar. This paper examined the effect of DNA on the corrosion resistance of reinforcing steels in cement mortars. Meanwhile, its effects on mechanical properties of cement paste, cement hydration products, and pore size structures of the cement composites were also investigated. A wet-dry cyclic test, which simulates marine environments, was used to accelerate the corrosion behavior of reinforcing steel. The linear polarization resistance and electrochemical impedance spectroscopy (EIS) were performed to evaluate the corrosion behaviors of reinforcing steel in mortar samples. In addition, the effects of DNA on flexural strength and compressive strength of cement pastes were investigated. Moreover, cement hydration products of samples with and without the addition of DNA were analyzed by X-ray diffraction (XRD), and the pore size structures of the corresponding composites were measured by Mercury Intrusion Porosimetry (MIP).

2. Materials and methods

2.1. Materials and specimens preparation

In this study, the same DNA corrosion inhibitor used in our previous study [28] was adopted. A widely used commercial corrosion inhibitor, phosphate (purchased from Sobute New Materials Co., Ltd), was also employed as the reference. Ordinary Portland Cement (OPC) (made in China) was used to prepare cement pastes and mortars. The chemical compositions of OPC are provided in Table 1.

The fine aggregate used in the mortar mix was a river sand. For all the mortar mixes, the cement to sand ratio (c/s) was kept at 0.4, and the water to cement ratio (w/c) was kept at 0.45. The DNA corrosion inhibitor was first added to the water with the concentration of 0.0025% and 0.0050% by weight of cement during the mixing phase. Smooth ordinary carbon steel rebars of 10 mm diameter and 14 cm length were used and embedded centrally in mortar specimens with the cover depth of 15 mm. A copper wire was soldered to one end of the reinforcing steel, and both ends were covered with epoxy resin coating, leaving a free exposure surface of approximately 31.4 cm². The diagram of steel reinforced mortar is presented in Fig. 1.

Steel reinforced mortar specimens were used to investigate the corrosion inhibition effect of DNA. For each mortar mixture, 40 mm × 40 mm × 160 mm prisms were cast in steel moulds. All the specimens were demoulded at 1d after casting and cured in a standard curing box at a temperature of 20 ± 2 °C and RH of > 95%. After 28d of curing, the whole specimens, except one side (40 mm × 160 mm), were covered with epoxy resin coating to promote unidirectional diffusion of chloride ions. Then, the samples were subjected to alternate wetting and drying in sodium chloride solutions to accelerate chloride penetration. One cycle consisted of 3d immersion in 3.5% NaCl solutions and 3d drying in open atmosphere. The electrochemical experimentation went on up to the 20th wet-dry cycle for all the samples. In order to measure the corrosion inhibition effect of DNA, referred to as the optimal dosage of DNA in simulated concrete pore solutions [28], four groups of mixtures were added into the steel reinforced mortar: 0%

DNA (as the control mixture), 0.0025% DNA, 0.0050% DNA, and 2% commercial corrosion inhibitor (as the reference mixture; the optimal dosage indicated by the instruction manual). A total of twelve steel reinforced mortars were prepared (three replicates for each mixture group).

Cement paste samples were tested for mechanical properties, and used for XRD and MIP analysis. The DNA corrosion inhibitor was added into the mixing water with the DNA to cement ratios (by mass) of 0.0025% and 0.0050%. 40 mm × 40 mm × 160 mm prisms with w/c ratios of 0.35, 0.45, and 0.55 were cast in steel moulds. All the specimens were demoulded at 1d after casting and cured in a standard curing box at a temperature of 20 ± 2 °C and RH of > 95% for 3d and 28d. Three replicates were prepared for each cement paste mixture.

2.2. Electrochemical and analytical methods

A common three-electrode system was used for the electrochemical experiments with the steel reinforcement being the working electrode, a saturated calomel electrode (SCE) serving as the reference electrode, and a platinum electrode serving as the counter electrode. The electrochemical measurements were conducted on a PARSTAT 2273 electrochemical workstation. All the electrochemical tests were conducted after the open circuit potential (OCP) of the working electrode was stable at room temperature. The corrosion potential (E_{corr}) and corrosion current density (i_{corr}) were monitored by means of linear polarization resistance (LPR) and electrochemical impedance spectroscopy (EIS). The LPR tests were performed in the potential range from −15 mV to +15 mV vs. OCP with a scan rate of 0.2 mV/s. EIS measurements were carried out within a frequency range from 100 KHz to 10 mHz, the ac signal used in EIS was 10 mV, and the data analysis was carried out using ZSimpWin software with proper electrical equivalent circuit. The experimental set-up of LPR and EIS is shown in Fig. 2.

The flexural strength and compressive strength of the cement paste samples were tested at both 3d and 28 d. After the strength measurement, small pieces of the cement paste samples were first immersed in acetone to stop the hydration. After that, samples were dried in an oven at 65 °C and ground to fine powders. The mineralogy of the crystalline phases in the samples was analyzed using a D8 Advance X-ray diffractometer (XRD, Bruker AXS, Germany). The scanning range was from 10° to 70° 2θ with 5° min^{−1} speed duration at 45 kV and 200 mA. The pore size distribution of the samples was measured using a mercury intrusion porosimeter (MIP, Poremaster GT-60, Quantachrome, USA). The samples were crushed to 5–10 mm in size, immersed in acetone for 30d, and dried at 60 °C for 7d before test.

3. Results and discussion

3.1. Corrosion potential and corrosion current density

Fig. 3 shows the corrosion potential and corrosion current density of reinforcing steel in cement mortars after exposure to 3.5% NaCl solutions for varying alternate wetting and drying cycles. It can be seen from Fig. 3(a) that the E_{corr} of all the specimens was between −150 mV and −200 mV before 8 cycles, which is an indication of less than 10% probability of corrosion as recommended in ASTM C876. This may be due to a passivating layer formed on the steel surface. Especially for the control specimen, the chloride ion either did not diffuse to the steel surface or did not accumulate to a certain concentration. It should be noted that the E_{corr} of specimens incorporated with the DNA inhibitor

Table 1
Chemical composition of OPC (wt.%).

Composition	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	SO ₃	Na ₂ O	K ₂ O	TiO ₂	Mn ₂ O ₃	LOI
Content	63.26	21.62	5.10	4.79	1.46	1.68	0.21	0.69	0.07	0.05	0.46

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