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Novel surface treatment of concrete bricks using acid-resistance mineral precipitation

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HIGHLIGHTS

- New method to treat concrete bricks through precipitating HAP is proposed.
- HAP produced by the proposed treatment method is acid resistant.
- Compressive strength of bricks can be improved by 100% by the treatment.
- Higher pH value of the treatment solution can accelerate the formation of HAP.

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ABSTRACT

Mineral precipitation is a promising surface treatment method to enhance the mechanical properties and durability of concrete and similar porous materials. Both chemically and microbiologically induced mineral precipitations have been proposed, in which calcium carbonate is used as the precipitated mineral. Since calcium carbonate can be dissolved in low pH value solution, existing mineral precipitation method is vulnerable to acid attack. To overcome this drawback, this study proposes to use a new mineral, Hydroxyapatite (HAP) as the precipitate to treat concrete bricks. Compared with calcium carbonate, HAP has much lower solubility and dissolution rate and therefore much higher resistance to acid attack. To demonstrate the effectiveness of this method, low grade concrete bricks made from recycled aggregates were treated by the proposed method. Experimental study shows that up to 100% enhancement of the compressive strength of the concrete bricks can be achieved by this treatment. Meanwhile, the water absorption of the bricks is also significantly reduced. Scanning electron microscope (SEM) imaging and X-ray diffraction (XRD) analysis were carried out to examine the mineralogy and morphology of the precipitates produced by the proposed treatment. Results suggest that all precipitates produced in this treating method eventually transform into HAP, and the transforming process can be accelerated by using treating solution with higher pH value.

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1. Introduction

Most deterioration mechanisms of concrete and similar porous materials are associated with water transport. The durability of these materials can be significantly enhanced by reducing their permeability. To this end, mineral precipitation method has been proposed [1], in which mineral precipitates were produced on the surface of the material to seal/fill the surface cracks/voids of the porous material. Both the durability and strength of the material can be significantly improved by this treatment.

Two major methods have been proposed to produce mineral precipitation: microbiological method and chemical methods. The former method is commonly known as microbiologically induced calcite precipitation (MICP) [1–6]. To treat the surface using MICP, a water-based medium consisting of bacteria, nutrients for bacteria, urea and calcium ions is applied to the surface to be treated. The applied medium can penetrate into the cracks and pores. The bacteria can produce urease as part of metabolism to catalyze the hydrolysis of the urea to produce carbonate and ammonium [2]. The produced carbonate then combines with calcium ions to produce calcium carbonate precipitate, which fills vacant spaces in the treated material and blocks the pathway of water to enter the material. As a result, the water absorption of the treated material is reduced, as well as the potential of harmful

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chemicals entering the material. The durability of the material is thus significantly improved [2–6].

However, the practical application of the MICP treatment is prohibited by its high cost. A recent study by De Muynck et al. [4] shows that the cost of the MICP treatment based on the price of the microorganisms and the price of the nutrients could be as high as $11–19/m^2. The inconvenient application procedure is another major barrier preventing the MICP from practical application in construction industry. To overcome these shortcomings of the MICP, chemical method to introduce mineral precipitation was recently proposed by Amidi and Wang [7]. In the MICP, bacteria are mainly used to produce urease which catalyzes the hydrolysis of urea in ambient environment to produce CO_2. In the chemical method, an organic carbonate, dimethyl carbonate (DMC) is used to produce the required carbonate for the generation of calcium carbonate precipitates. This is because DMC can be hydrolyzed slowly into methanol and carbonate in the basic environment of the concrete at ambient temperature and pressure [8]. In this way, carbonate can be produced in controlled manner in ambient temperature without using any urease, which can then react with calcium ions to form calcium carbonate deposition.

However, calcium carbonate is the precipitate used in these studies, which is a relatively soluble mineral (solubility product, $K_{sp} = 5 \times 10^{-9}$) and thus susceptible to dissolution in acidic environment. To overcome this shortcoming of the existing mineral precipitation methods, this study exploits an acid-resistant mineral, hydroxyapatite (HAP) as the precipitate to treat the concrete-type porous materials. HAP is the main inorganic component of vertebrate bones and teeth and has excellent biocompatibility and bioactivity. Compared with calcium carbonate, HAP has much lower solubility product (K_{sp} ($\sim 10^{-59}$)) [9] and dissolution rate ($R_{diss} = \sim 10^{-14}$ mols cm^{-2} s^{-1} at pH 5.6) [10], and therefore, is much more resistant to acid attack [6].

To demonstrate the feasibility and potential of using HAP as precipitated mineral in surface treatment, low grade concrete bricks made from recycled aggregates were treated by the proposed method. Compared with ordinary concrete bricks, these bricks make good use of waste or demolished concrete and are lighter. However, they suffer from low strength and poor durability because of their high porosity. To enhance the properties of these bricks using the proposed treating method, we can first soak the bricks into diammonium hydrogen phosphate ($(NH_4)_2HPO_4$ (DAP)) solution to absorb phosphate source, and then submerge them into a calcium ion-rich solution. DAP absorbed into the pores of the bricks will react with the calcium ions to produce HAP. Since HAP is much more difficult to dissolve in water than calcium carbonate, it will precipitate and fill or seal the pores of the bricks. In this way, the pathway of water permeation will be blocked and permeability of the brick will be reduced, leading to significantly improvement of the mechanical properties and durability of the bricks.

2. Materials and methods

2.1. Materials

Commercially available concrete bricks with size of 240 mm × 115 mm × 53 mm were used in this study. They were made from recycled concrete aggregates with water absorption in the range of 19–21% and bulk density in the range of 1590–1630 kg/m^3. The average compressive strength of three samples was measured as 11.89 MPa. DAP solution (0.5 mol/L) was chosen as phosphate radical source. Two treating solutions were used to provide calcium ions needed to produce HAP precipitant: a low pH value solution produced by using pure $CaCl_2$ (0.5 mol/L) solution and a high pH value solution produced by adding 4 g/L NaOH into 0.5 mol/L $CaCl_2$ solution.

2.2. Treating method

Two methods, single-treatment and multiple-treatment, were used to treat the bricks. Before the treatment, all specimens were cleaned using a banister brush to remove dust and loose particles. In the single-treatment, the specimens were submerged in the DAP solution for one day to absorb phosphate ions, and then submerged in the treating solution to allow for calcium ions to reach the pores of the specimens. These calcium ions reacted with DAP to produce precipitate in the pores. The duration of submerging in the treating solution, which determines the availability of the calcium ions and reaction time, plays an important role on the mineral precipitation. Therefore, four durations (1, 3, 7, and 14 days) were used in submerging the specimens in the treating solutions to evaluate their effect on the properties of the treated bricks.

The multiple-treatment method follows the same procedure used by the single-treatment, but with a number of identical treating cycles. In each treating cycle, the brick specimens were first submerged into the DAP solution for one day, and then soaked into the treating solution for two submersion durations (1 day and 3 days). All treatments were carried out at ambient temperature of the laboratory. After the treatment, all treated specimens were submerged in fresh water to remove residual salts, loose particles and unbounded precipitates. Finally, the specimens were dried in an oven at temperature of 105 °C until their weights reached constants. These dried specimens were used in the tests described thereafter.

2.3. Characterizing the treated bricks

The effects of the proposed treatment methods were evaluated by measuring the water absorption, bulk density, and compressive strength of the specimens according to China standard (GB/T 2542-2012) [11] (equivalent to ASTM C67 [12]).

2.3.1. Water absorption and bulk density

Water absorption and bulk density were determined as the average of six treated samples. The treated bricks were first completely submerged in water at ambient temperature for 24 h. Then they were taken out and dried with a damp towel to remove the visible water on their surface. Their weights were then immediately measured as the saturated weight (m_s). After that, the bricks were dried in an oven at 105 °C ± 5 °C until a constant weight was reached, which was recorded as the oven-dried weight (m_d) of the specimen. The geometry of the bricks (length (L), width (W) and height (H)) were also measured. The water absorption (W_a) and bulk density (ρ_d) of the specimen can be calculated by

$$W_a = [(m_s - m_d)/m_d] \times 100\%, \quad (1)$$

$$\rho_d = m_d/(L \times W \times H). \quad (2)$$

2.3.2. Compressive strength

To measure its compressive strength, each brick specimen was cut into halves along the width direction. These two halves were stacked along the surface perpendicular to the cutting surface, as shown in Fig. 1, with a stacking length no less than 100 mm. Then, the stacked two halves were compressed by a loading machine until a maximum load P was reached. The compressive strength of the recycled bricks (σ_p) can be calculated by

$$\sigma_p = P/(a \times b), \quad (3)$$

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