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Surface treatment of recycled concrete aggregates through microbial carbonate precipitation

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HIGHLIGHTS

- Recycled concrete aggregates (RCA) were treated by microbial carbonate precipitation (MCP).
- MCP-treated RCA show increased weight and reduced water absorption.
- Rate, amount, and type of MCP can be modified by control of precipitation conditions.

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ABSTRACT

Reuse of recycled concrete aggregates (RCA) in new concrete structures is limited due to their high water absorption and weak bonding to new matrix. These unfavorable properties are associated with the thin layer of old mortar attached to the surface of original aggregates. In this paper, a novel approach of RCA surface treatment by means of microbial carbonate precipitation (MCP) was studied. Factors influencing MCP on RCA were investigated. It was found that the amount of MCP peaks at pH = 9.5 and increases with higher temperature, bacteria concentration, or calcium concentration. Enhanced MCP on RCA can be achieved through proper control of culturing and precipitating conditions. Results show that surface modification of RCA through MCP is feasible evidenced by increasing weight and reducing water absorption of treated RCA.

1. Introduction

Interest in using recycled concrete aggregates (RCA) from demolished old concrete structures in new construction is growing fast recently [1]. Such interest derives from two problems associated with sustainability, i.e. lack of land for disposing demolished concrete debris and depletion of quality primary natural aggregates for concrete production. This is of particular importance to small country like Singapore. As an island city-state with limited land space and nature resource, it is crucial for Singapore to develop strategies to prolong the lifespan of landfills and to recover waste into resources. Recent aggregate crises in Singapore highlight the need of identifying alternative aggregate resources [2]. The heavy dependency on import of aggregates from neighboring countries and soaring prices make the use of RCA an attractive solution.

RCA are mainly used as fillers for road bases and backfills [3,4]. When RCA are used for concrete production, they are usually mixed in small proportion (20% or less) with the natural aggregates [5]. The concern on using RCA in structural concrete comes from laboratory work showing inclusion of RCA deteriorates properties of both the fresh and the hardened concrete. It has been reported that concrete incorporating RCA has lower workability and greater slump loss due to its high water absorption. Pre-wetting RCA before mixing is normally necessary [6,7]. While the water absorption of natural aggregate is usually below 1%, water absorptions between 3.6% and 8.0% were found for recycled coarse aggregates and absorptions between 8.3% and 12.1% were reported for recycled fine aggregates [8]. In terms of hardened concrete properties, the weak bond between RCA and cement matrix leads to remarkable reduction in compressive strength, the degree of which depends on the quality of RCA and grade of new concrete [6,9]. In the worst scenario, complete RCA replacement of natural aggregates can cause the reduction of compressive strength by as much as 87% [10].

Both high water absorption and weak bond of RCA to new matrix are associated with the thin layer of old mortar attached to the original aggregates. The attached mortar is composed of cement paste and loose sands. Both the micro porosity in cement past and the meso porosity between sand particles enhance RCA's water

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absorption. When the old concrete fails, micro-crack usually penetrates through a high-porosity matrix band close to the aggregate, which is referred as interfacial transition zone (ITZ). Such failure mechanism of concrete implies that the mortar attachment on RCA surface is likely to contain part of the old ITZ with micro cracks. When RCA are cast into new concrete, such old ITZ merges with the new ITZ between RCA and new matrix, forming an extended weak band between matrix and aggregate and subsequently decreasing concrete strength [9,11].

In order to reduce RCA water absorption and to improve RCA-matrix bond, researchers have investigated various methods. Otsuki et al. [9] and Tam et al. [12] proposed a two-stage mixing method to produce RCA concrete. In this method, RCA are firstly mixed with cement and part of water for 60 to 90 s (stage one). The remaining water is then added into the fresh mixture and mixed for another 120 s (stage two). Premix in stage one allows stronger cement paste, i.e. paste with lower water-to-cement ratio, to fill the porosity and micro cracks on RCA surface. 28-day compressive strength of the resulting RCA concrete is enhanced by up to 20%. Li et al. [13] and Kong et al. [14] extended the two-stage mixing method by pre-coating RCA with different pozzolanic powders such as fly ash, slag, and silica fume before mixing RCA with the cement paste. Pozzolanic powder pre-coating not only fills the porosity in old ITZ but also forms new pozzolanic products by consuming calcium hydroxide in the old cement paste. As a result, the compressive strength of RCA concrete is further enhanced.

Methods have been proposed to enhance the removal of attached mortar from RCA. Shima et al. [15] developed a heating and rubbing method to produce high-quality aggregate from demolished concrete. The performance of the resulting RCA concrete is comparable to that of ordinary concrete. Ong et al. [5] used microwave to separate mortar attachment and original stone in RCA. Concrete incorporating microwave-treated RCA achieves remarkable increase in terms of compressive strength, elastic modulus, and flexural strength. Katz [16] used ultrasonic cleaning technique to treat RCA, which led to increase of RCA concrete compressive strength up to 7%. Most of these methods, however, are energy intensive and resulting in higher cost. In addition, those methods can potentially cause aggregates damage due to high heat and intense mechanical stress.

Another approach is to modify RCA surface with coating. Kou and Poon [17] impregnated RCA with polyvinyl alcohol (PVA) solution, successfully reducing water absorption and enhancing crushing value (10%) of RCA. Tsujino et al. [18] used oil-type improving agent, a mineral oil with emulsifying agent and Lanolin fatty acid slat in emulsion state, and silane-type improving agent, a silicon analogue with emulsifying agent and water in emulsion state, to pre-coat RCA. This approach significantly reduces RCA water absorption. However, the compressive strength of the resulting RCA concrete is also reduced.

Microbial carbonate precipitation (MCP) refers to the technique of using microorganism, especially bacteria, to induce calcium carbonate crystal precipitation. This technique has been used to improve properties of soil and construction materials [19,20]. Such improvements were realized in two methods, namely bio-deposition and bio-cementation. Bio-deposition refers to bacteria-induced $CaCO_3$ precipitation on surface of stones or cementitious materials [19]. Though it was used originally as a protective layer to prevent stones decaying [21,22], it has been applied recently on mortar to increase its resistance towards carbonation, chloride penetration and freeze–thaw cycles [23,24]. Bio-cementation refers to the bacteria-induced $CaCO_3$ precipitation functioning as binder material [19]. It increases concrete compressive strength [25] and heals cracks in concrete [26].

Formation and precipitation of $CaCO_3$ are governed by four key factors: calcium ion concentration, dissolved inorganic carbon, pH, and availability of crystal nucleation site [27]. While many bacteria can alter all of the four factors, their primary role in inducing precipitation has been ascribed to increasing pH by different metabolisms [19] and attracting metal ion, e.g. Ca^{2+}, as crystal nucleation sites [28]. *Sporosarcina pasteurii* (previously known as *Bacillus pasteurii*), an ureolytic bacterium widely used to induce carbonate precipitation [29–31], was used in current research. It hydrolyzes urea and creates alkaline environment [27].

Although MCP has been well investigated and used to modify stones and concrete, MCP on RCA was rarely investigated. The only study was reported by Grabiec et al. [32], in which they investigated and confirmed the feasibility of MCP to modify the surface of RCA. In this paper, factors influencing MCP on RCA treatment were studied. The results form the basis for optimizing the culturing condition to enhance MCP on RCA.

2. Experimental program

Experiments were carried out in two stages. Parametric studies as shown in Table 1 were conducted first to experimentally determine the effects of pH (7.0–10.5), temperature (25 °C and 35 °C), calcium content (5.6–50.4 g/L of CaCl) and bacteria concentration (10^6–10^8 cells/ml) on microbial carbonate precipitation (MCP) in the growth media, i.e. without RCA, or MCP on RCA. Based on the insights obtained from the parametric studies, conditions were chosen to maximize MCP on RCA as shown in Table 2. Materials and methods used in this study are summarized below.

2.1. Recycled concrete aggregates

Recycled concrete aggregates (RCA) were obtained from Pan-United Corporation Ltd, Singapore. The RCA were screened by a 20-mm sieve to exclude any big particles. The properties of RCA used in this research were determined under the guidance of ASTM C127-12 Standard Test Method for Density Relative Density (Specific Gravity) and Absorption of Coarse Aggregate, and are given in Table 3. As can be seen, the RCA have higher water absorption and lower density indicating the presence of attached mortar on the surface of RCA. In Singapore, RCA are produced by normal crushing, separation, and screening. Heat treatment and/or mechanical rubbing are not used in the production of RCA locally.

2.2. Bacteria and culture media

S. pasteurii (DSM No. 33) obtained from the German Collection of Microorganism and Cell Cultures (DSMZ), Braunschweig, Germany, was adopted in this research. Bacteria were inoculated into autoclaved liquid culture media and grew in a 30 °C incubating shaker (150 rpm) for 48 h. The composition of liquid culture media was 20 g/L peptone, 5 g/L NaCl, and 20 g/L urea, which is recommended by DSMZ. After growing for two days, the bacterial cell concentration was determined by optical density test (λ = 600 μm). The bacteria culture media was then centrifuged at 10,000 rpm for 30 min to obtain highly concentrated bacterial cells.

2.3. Test of microbial carbonate precipitation

Tests were carried out to study the effects of pH and temperature on MCP. The concentrated bacteria cells obtained from centrifuge were diluted to 10^6 cells/ml with liquid growth media which consist of nutrient broth (3 g/L), urea (20 g/L), NH_4Cl (10 g/L), Na_2CO_3 (2.12 g/L) and $CaCl_2$ (5.6 g/L). Sodium hydroxide (1 N solution) and hydrochloric acid (1 N) were used to adjust the pH of the solution between 7.0 and 10.5. The growth media containing bacteria were stored at either 25 °C or 35 °C for 72 h for calcium carbonate precipitation. After three days of MCP, the medium was filtered through a filter paper.

Prior to filtering, the filter paper was oven-dried at 105 °C for an hour to remove any humidity. The weight of the oven-dried filter paper was recorded as M_1. An analytical balance with 0.0001 g in resolution was used in this study. After filtering, the filter paper containing calcium carbonate precipitates was oven-dried again at 105 °C for an hour and the new weight was taken as M_2. The amount of calcium carbonate precipitates was calculated as M_2-M_1.

2.4. Test of microbial carbonate precipitation on RCA

Tests were carried out to study the effects of calcium content and bacteria concentration on MCP on RCA. The growth media were prepared in a similar way as described in Section 2.3. The concentrated bacteria cells obtained from centrifuge were diluted to pre-determined cell concentration (10^6–10^8 cells/ml) with liquid growth media which consist of nutrient broth (3 g/L), urea (20 g/L), NH_4Cl (10 g/L), Na_2CO_3 (2.12 g/L) and $CaCl_2$ (5.6–50.4 g/L). Sodium hydroxide (1 N solution) was