OPTIMIZING PRECIPITATION METHOD OF MICROBIAL CARBONATE PRECIPITATION ON MODEL RECYCLED AGGREGATE

Welai Zeng[1]*, Yuxi Zhao[1], Zhangyao Feng[1], Zhenmei Lv[2], Surendra P. Shah[3]

[1] Institute of Structural Engineering, Zhejiang University, Hangzhou 310058, Zhejiang, PR China

[2] College of Life Science, Zhejiang University, Hangzhou 310058, Zhejiang, PR China

[3] Center for Advanced Cement-Based Materials, Northwestern University, Evanston, IL 60208, USA

*Corresponding author; e-mail: 3110101092@zju.edu.cn

Abstract

This paper improves the properties of recycled aggregate (RA) with the optimum precipitation methods by employing *Sporosarcina pasteurii* bacteria to induce microbial carbonate precipitation (MCP). To study the modification effect on different specimen surfaces, concrete cube specimens were prepared as model RAs. To optimize precipitation method, two influencing factors on precipitation, namely the calcium-source addition method and rotation treatment, were considered. An optimum precipitation method was proposed: RAs were dried, and submerged with precipitation culture media containing bacteria first; then the bacteria-saturated RAs were submerged into another precipitation culture media without bacteria, where the calcium-source solution was added in batches.

Keywords:

Recycled aggregate; Model aggregate; Microbial carbonate precipitation; Water absorption.

1 INTRODUCTION

Nowadays recycled aggregates (RAs) produced from construction and demolition waste have been promoted and used worldwide, to solve the environmental problem and scarce natural resource problem. However lots of researches find that recycled aggregate concrete (RAC) has the worse mechanical properties and durability, such as its lower compressive and tensile strength [Xiao J 2012, N. Kisku 2017, İlker Bekir Topçu 2004, Gómez-Soberón J M V 2002, Silva R V, Brito J D 2015], worse resistance to chloride ion penetration [Xiao J 2012, N. Kisku 2017, Buyle-Bodin F 2002, Shicong K 2006] and freeze-thaw [N. Kisku 2017, İlker Bekir Topçu 2004]. This is caused by the old mortar attached, which increases the water absorption of RAs and induces more interfacial transition zones (ITZs) within RAC [Poon C S 2004, Hongru Zhang 2015].

There have been many researchers trying to improve the properties of RA and RAC by either removing the old mortar or enhancing the old mortar. Akbarnezhad A et al [Akbarnezhad A 2011] used microwave heating to remove the cementitious mortar effectively. But they also found that the incorporation of up to 40% of microwave-treated RA seemed to have negligible effect. Tam V W Y et al [Tam V W Y 2007] revealed that RA's water absorption reduced and mechanical properties improved after acid solution treatment. But there were some potential adverseness on durability of RAC. Chi-Sun Poon et al [Kou S C 2014] cured RA with CO_2 and found the improvement of old mortar attached on RA. But it was necessary to discover whether the carbonation of old mortar would influence the steel corrosion in RAC. Li W et al [Li W 2017] improved the mechanical properties of RAC with the incorporation of nano silica. And more types of nano material had been used to modify RAs.

These days microbial mineralization has been increasingly popular for application in engineering, e.g., the repair and protection of cementitious materials and concrete structure [Muynck W D 2008, Qian C 2011, Verma 2015, Qian C 2009, Khaliq W 2016, Srinivasa Reddy 2012]. It is attributed to the microbial carbonate precipitation (MCP) produced with the effect of bacterial, which has been proved to have the ability of healing cementitious material and improving their durability. Based on the existed studies, bacteria such as *Bacillus sphaericus*, *Bacillus psychrophilus*, *Planococcus okeanokoites*, and *Sporosarcina pasteurii* are often chosen to induce the precipitation of calcium carbonate. This precipitation process can be described as follow:

$$CO(NH_2)_2 \rightarrow 2NH_4^+ + CO_3^{2-}$$

$$\text{cell} + Ca^{2+} \rightarrow \text{cell-}Ca^{2+}$$

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3\downarrow$$

$$\text{cell-}Ca^{2+} + CO_3^{2-} \rightarrow \text{cell-}CaCO_3\downarrow$$

In this study, *S. pasteurii* is selected considering that it is an aerobic, alkaliphilic, common soli bacterium with no pathogenicity.

Meanwhile the attempt on the RA's modification with MCP has been carried out. Grabiec et al. [Grabiec A M 2012] reduced RA's water absorption by modifying the surface of RA with calcium carbonate bio-deposition. Qiu et al. [Qiu, Jishen 2014] investigated the factors that influenced MCP on RA, and found that enhanced MCP on RA could be achieved with proper control of culturing and precipitating conditions. Pan et al. [Pan, Zhong-Yao 2015] added microbial modified RAs into asphalt mixtures and found the better adhesion behavior than that of the untreated RAs. Jianyun Wang et al. [Wang J 2017] tested the compressive strength and water absorption of concrete after using the bio-treated RAs with different two methods, spraying and several immersion. They found the very close or even slightly better properties of bio-treated RAC than normal concrete.

In this study, the optimum microbial precipitation of RAs were investigated. Different precipitation methods including calcium-adding methods and rotation methods were considered to optimize precipitation method by modifying RAs more efficiently; the distribution of MCP on the surface of the specimens, the mass increase, and the reduction in water absorption of the samples were measured.

2 EXPERIMENTAL PROGRAM

2.1 Material

2.1.1. Bacterial and culture media

S. pasteurii (DSM No. 33), purchased from the German Collection of Microorganism and Cell Cultures (DSMZ), Braunschweig, Germany, was used in this study. The bacteria were activated in a culture media over a period of 2 days, within an incubator shaker at 30 °C. The culture media was composed of 20 g/L tryptone, 5 g/L NaCl, and 20 g/L urea, as recommended by DSMZ. Following a two-day activation period and a one-day growth period, the bacteria culture media was centrifuged to obtain a high concentration of bacterial cells.

2.1.2 Aggregate

A series of 30 × 30 × 30 mm concrete cubes were cut from 100 × 100 × 100 mm natural aggregate concrete specimens with a 28-d compressive strengh of 30 MPa, as model RAs, to optimize the precipitation methods and observe the modification effect on different RA's surfaces. The natural aggregates are ranging from 5 to 20 mm.

2.2 Precipitation method

2.2.1. Preparation

Before modification, model RAs were dried by using an oven at a temperature of 40 °C for 24 h, rather than 105 ± 5 °C, considering that the high temperature might change the internal pore structures [Ma Q 2015]. The 10^8 cell/mL concentrations of bacterial cells were diluted and obtained from the centrifuge. The precipitation culture media consisted of 3 g/L tryptone, 10 g/L NH_4Cl, 20g/L urea, and 2.12 g/L Na_2CO_3. The pH of the solution was adjusted to 9.5 by using sodium hydroxide (1 N solution) and hydrochloric acid (1 N).

The entire MCP modification process occurred over 7 days. On the first day, the model RAs were placed into a precipitation culture media containing the bacteria, and were stored within an incubator for 1 day at a temperature of 30 °C.

2.2.2. Calcium addition methods

To enhance the effect of microbial modification and realize the high-efficient use of MCP, three different calcium addition methods were performed, from 2 d to 7 d, as show in the Fig. 1.

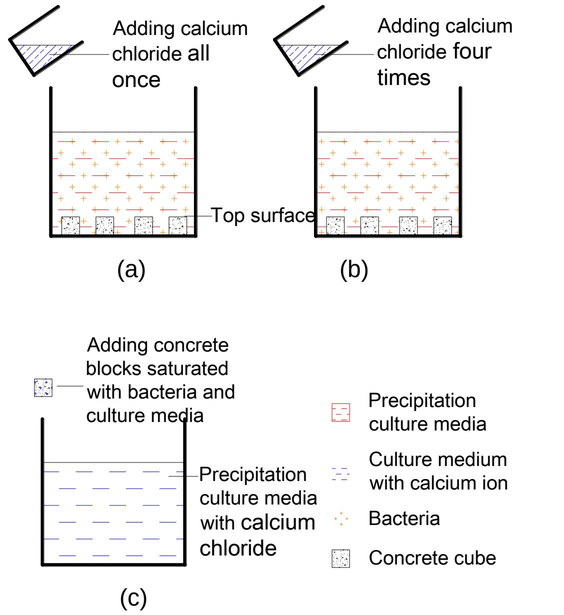


Fig. 1. Schematic diagrams of precipitation methods: (a) Precipitation method 1, (b) Precipitation method 2, (c) Precipitation method 3.

(1)Precipitation method 1: All the $CaCl_2$ solution with a pH of 9.5 was poured into the precipitation culture media on the second day. The calcium-ion concentration of the entire solution was 0.55 mol/L, as shown in Fig. 1 (a).

(2)Precipitation method 2: $CaCl_2$ solution same as that of Precipitation method 1 was divided into four parts, which were poured into the precipitation culture media each day from 2 d to 5d. The $CaCl_2$ solution and culture media were identical to those used in Precipitation method 1, as shown in Fig. 1 (b).

(3)Precipitation method 3: The model RAs were taken out from the precipitation culture media, and placed into another precipitation culture media, which received an addition of a solution with a calcium-ion concentration of 0.55 mol/L at the pH of 9.5, as shown in Fig. 1 (c).

2.2.3. Rotation methods

To study the influence of the rotation treatment on the accumulation of MCP on the different RA's surfaces, two model RAs were rotated. Meanwhile, the other two model RAs remained static throughout the entire MCP modification process, as a control experiment. During the rotation process, each side of the model RA would be the top surface for 1-day precipitation, as shown in Fig. 2.

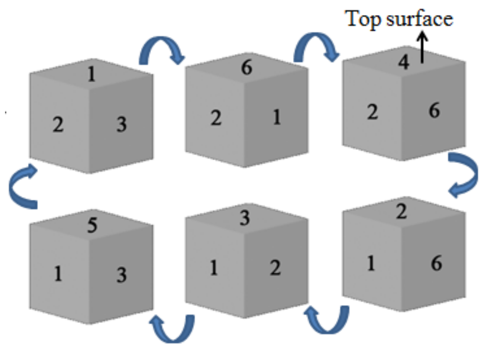


Fig. 2. Schematic diagrams of the rotation treatment.

2.3 RA's properties testing

According to *Chinese Standard for technical requirements and test method of sand and crushed stone (of gravel) for ordinary concrete* (JGJ 52-2006), water absorption was determined. However the drying temperature was modified at 40 °C, rather than the recommended 105 ± 5 °C. The reason for this was explained in Section 2.2.1.

Before modification, the model RAs were immersed in water for 24 h, and the weight of each saturated-surface-dry (SSD) model RA was recorded as M_1. Subsequently, the model RA was oven-dried at 40 °C until they attained a constant weight. The weight of each oven-dried model RA was then recorded as M_2. The oven-dried model RA was saturated in the precipitation culture media with bacterial for 1 d as discussed in Section 2.2. After modification, the SSD model RA was removed from the precipitation culture media and weighed as M_3. Finally, the modified model RAs were oven dried at 40 °C again until a constant weight, M_4. Mass increase ΔM, water absorption w, and water absorption reduction Δw, were calculated using the equations that follow:

$$\Delta M = M_4 - M_2 \tag{1}$$

$$w_1 = (M_1 - M_2)/M_2 \tag{2}$$

$$w_2 = (M_3 - M_4)/M_4 \tag{3}$$

$$\Delta w = (w_1 - w_2)/w_2 \tag{4}$$

where w_1 is the water absorption of the model RAs before modification, and w_2 is the water absorption of the model RAs after modification.

3 EXPERIMENTAL PROGRAM

3.1 MCP distribution

MCP distributions on the external surfaces of the model RAs treated with different precipitation methods are shown in Fig. 5.

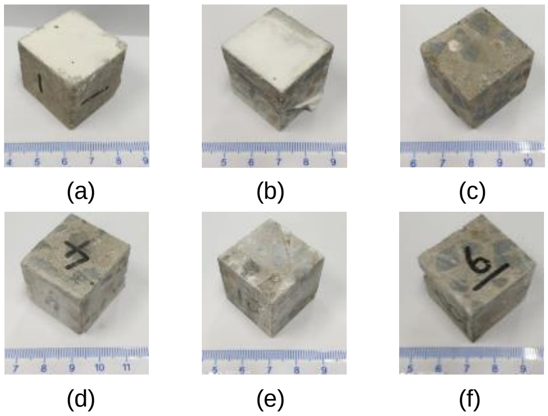


Fig. 5. MCP Distributions treated with different precipitation methods: (a) Precipitation method 1-Control, (b) Precipitation method 2-Control, (c) Precipitation method 3-Control, (d) Precipitation method 1-Rotation, (e) Precipitation method 2-Rotation, (f) Precipitation 3-Rotation.

As shown in Fig. 5, there are white-colored precipitations, mainly consisting of $CaCO_3$, on the surfaces of the all model RAs. Especially, for Precipitation method 1-Control and Precipitation method 2-Control group, MCPs on the top surface are thicker than other groups as shown in Fig. 5 (a) and Fig. 5 (b). Moreover this thick layer MCP just can be found on the top surfaces of the samples. While for Precipitaiton method 1-Rotation group and Precipitation method 2-Rotation group, no thick layer can be found on the top surfaces.

For Precipitation method 3, there is not an apparent difference on MCP distribution between Precipitation method 3-Rotation and Precipitation method 3-Control group as shown in Fig. 5 (c) and Fig. 5 (f). It means that rotation treatment has no influence on the Precipitation method 3.

3.2 Mass increase and water absorption

Mass increases after different precipitation methods are shown in Fig. 6.

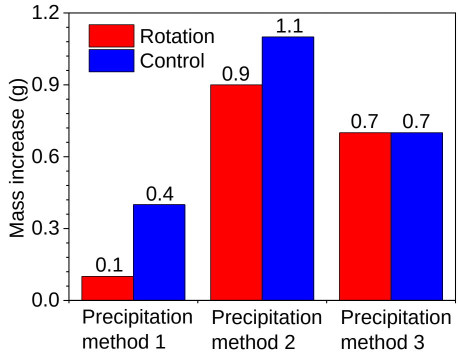
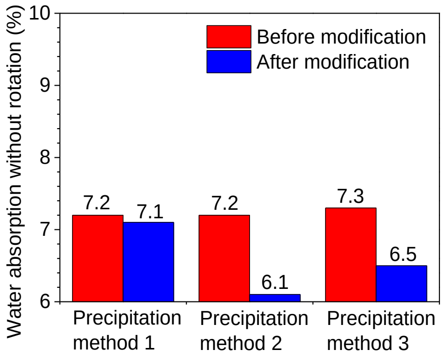


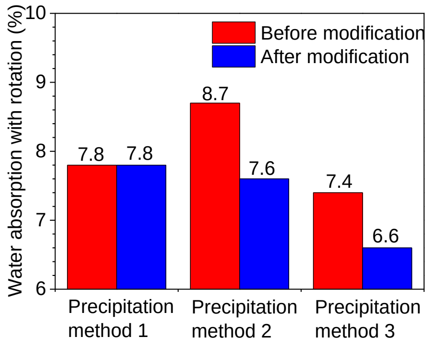
Fig. 6. Mass increases after different precipitation methods.

As shown in Fig. 6, the mass of each model RA increases because of MCP attached. For Precipitation method 2 and Precipitation method 3 group, the mass increases are greater than that of Precipitation method 1 group. For Precipitation method 1 and Precipitation method 2, after rotation treatment the mass increases are lower. While for Precipitation method 3, the rotation treatment seems to have no influence on the mass increase. These results are in accordance with the findings discussed in Section 3.1.

Fig. 7 shows the water absorption before and after modification.



(a)



(b)

Fig. 7. Water absorption before and after modification: (a) Control, (b) Rotation.

From Fig. 7, after modification, water absorption of the model RAs decreases, except the Precipitation method 1-Rotation group. The decrease of water absorption is due to the formation of MCP on the surface of model RAs, which fills the cracks and pores, and covers the old cement mortar. Considering the differences in the initial water absorption of model RAs, the reduction in water absorption is calculated, to further compare the modification effects of the different precipitation methods, as shown in Fig. 8.

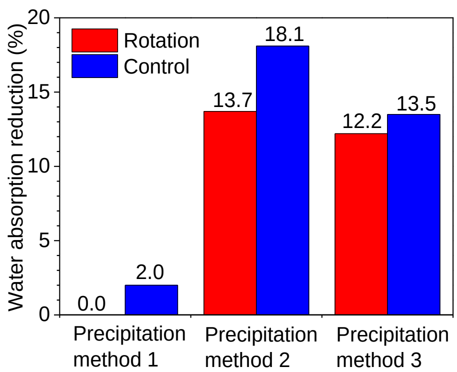


Fig. 8. Water absorption reduction with different precipitation methods.

As shown in Fig. 8, the water absorption reduction of Precipitation method 2 and Precipitation method 3 are much higher than that of Precipitation method 1. It means that Precipitation method 2 and Precipitation method 3 have the better modification effect. Moreover results show that for the all precipitation methods, water absorption reduction decreases with the rotation treatment, because the rotation treatment has a negative effect on the MCP agglomeration.

For the Precipitation method 1-Rotation, the mass of model RAs increases, but the water absorption does not change. The Precipitation method 3-Control group and Precipitation method 3-Rotation group have identical mass increase, but they have different water absorption reduction. This indicates that the results of mass increase cannot be used to accurately reveal the modification effect.

3.3 Optimum precipitation method

Based on the results discussed in Section 3.2, the optimum precipitation method is proposed:

(1) Before the modification, RAs are dried to absorb more bacteria.

(2) Dried RAs are placed into the precipitation culture media containing bacteria, and stored within an incubator at an appropriate temperature.

(3) Calcium-source solution is divided equally into several parts.

(4) RAs saturated with bacteria are taken out, and directly immersed into another new precipitation culture media without bacteria.

(5) Divided calcium-source solution is added into the new precipitation culture media in batches.

(6) Finally RAs are dried at an appropriate temperature for test or use.

4 SUMMARY

In this study, precipitation method optimization of MCP modification is experimentally investigated and summarized. as follows:

(1) The optimum precipitation method is proposed:

- Before modification RAs are dried.
- Dried RAs are immersed into a precipitation culture media containing bacteria.
- Calcium-source solution is divided.
- RAs saturated with bacteria are placed into another new precipitation culture media without bacteria.
- Divided calcium-source solution is added into the precipitation culture media in batches.
- Modified RAs are dried for use.

(2) After MCP modification, model RAs have the increasing mass and decreasing water absorption.

5 ACKNOWLEDGMENTS

Financial supports from the *National Natural Science Foundation of China* (51578489) is gratefully acknowledged.

6 REFERENCES

[Akbarnezhad A 2011] Akbarnezhad A, Ong K C G, Zhang M H, et al.; Microwave-assisted beneficiation of recycled concrete aggregates. Construction & Building Materials, 2011, 25(8), 3469-3479.

[Buyle-Bodin F 2002] Buyle-Bodin F, Hadjieva-Zaharieva R; Influence of industrially produced recycled aggregates on flow properties of concrete. Materials & Structures, 2002, 35(8), 504-509.

[Gómez-Soberón J M V 2002] Gómez-Soberón J M V; Porosity of recycled concrete with substitution of recycled concrete aggregate: an experimental study. Cement & Concrete Research, 2002, 32, 1301–1311.

[Grabiec A M 2012] Grabiec A M, Klama J, Zawal D, et al.; Modification of recycled concrete aggregate by calcium carbonate biodeposition. Construction & Building Materials, 2012, 34(34), 145-150.

[Hongru Zhang 2015] Hongru Zhang, Yuxi Zhao; Integrated interface parameters of recycled aggregate concrete. Construction & Building Materials, 2015, 101, 861-877.

[İlker Bekir Topçu 2004] İlker Bekir Topçu, Şengel S; Properties of concretes produced with waste concrete aggregate. Cement & Concrete Research, 2004, 34(8), 1307–1312.

[Khaliq W 2016] Khaliq W, Ehsan M B; Crack healing in concrete using various bio influenced self-healing techniques. Construction & Building Materials, 2016, 102, 349-357.

[Kou S C 2014] Kou S C, Zhan B J, Poon C S; Use of a CO_2 curing step to improve the properties of concrete prepared with recycled aggregates. Cement & Concrete Composites, 2014, 45(1), 22-28.

[Li W 2017] Li W, Long C, Tam V W Y, et al.; Effects of nano-particles on failure process and microstructural properties of recycled aggregate concrete. Construction and Building Materials, 2017, 142, 42-50.

[Ma Q 2015] Ma Q, Guo R, Zhao Z, et al.; Mechanical properties of concrete at high temperature—A review. Construction & Building Materials, 2015, 93, 371-383.

[Muynck W D 2008] Muynck W D, Debrouwer D, Belie N D, et al.; Bacterial carbonate precipitation improves the durability of cementitious materials. Cement & Concrete Research, 2008, 38(7), 1005-1014.

[N. Kisku 2017] N. Kisku, H. Joshi, M. Ansari, et al.; A critical review and assessment for usage of recycled aggregate as sustainable construction material. Construction & Building Materials, 2017, 131, 721–740.

[Pan, Zhong-Yao 2015] Pan, Zhong-Yao, et al.; Modified recycled concrete aggregates for asphalt mixture using microbial calcite precipitation. RSC Advances, 2015, 5(44), 34854-34863.

[Poon C S 2004] Poon C S, Shui Z H, Lam L; Effect of microstructure of ITZ on compressive strength of concrete prepared with recycled aggregates. Construction & Building Materials, 2004, 18(6), 461-468.

[Qian C 2009] Qian C, Wang J, Wang R, et al.; Corrosion protection of cement-based building materials by surface deposition of $CaCO_3$ by Bacillus pasteurii. Materials Science & Engineering C, 2009, 29(4), 1273-1280.

[Qian C 2011] Qian C, Wang R, Wang J; Bio-deposition of a calcite layer on cement-based materials by brushing with agar-immobilised bacteria. Advances in Cement Research, 2011, 23(4), 185-192.

[Qiu, Jishen 2014] Qiu, Jishen, David Qin Sheng Tng, En-Hua Yang; Surface treatment of recycled concrete aggregates through microbial carbonate precipitation. Construction and Building Materials, 2014, 57, 144-150.

[Shicong K 2006] Shicong K, Poon C S; Compressive strength, pore size distribution and chloride-ion penetration of recycled aggregate concrete incorporating class-F fly ash. Journal of Wuhan University of Technology, 2006, 21(4), 130-136.

[Silva R V 2015] Silva R V, Brito J D, Dhir R K; Tensile strength behaviour of recycled aggregate concrete. Construction & Building Materials, 2015, 83, 108-118.

[Srinivasa Reddy 2012] Srinivasa Reddy, V., Achyutha Satya, K., Seshagiri Rao, V. M, & Azmatunnisa, M; A biological approach to enhance strength and durability in concrete structures. International Journal of Advances in Engineering & Technology, 2012, 4(2), 392-399.

[Tam V W Y 2007] Tam V W Y, Tam C M, Le K N; Removal of cement mortar remains from recycled aggregate using pre-soaking approaches. Resources, Conservation and Recycling, 2007, 50(1), 82-101.

[Verma 2015] Verma, Rajesh K., et al.; Biomineralization and bacterial carbonate precipitation in mortar and concrete. Biosci Bioeng, 2015, 1, 5-11.

[Wang J 2017] Wang J, Vandevyvere B, Vanhessche S, et al; Microbial carbonate precipitation for the improvement of quality of recycled aggregates. Journal of cleaner production, 2017, 156, 355-366.

[Xiao J 2012] Xiao J, Li W, Fan Y, et al.; An overview of study on recycled aggregate concrete in China (1996–2011). Construction & Building Materials, 2012, 31(6), 364-383.