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Properties of Mortar Containing Recycled Fine Aggregate Modified by Microbial Mineralization

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Abstract: Microbial-induced mineralization deposition was used to improve the quality of the recycled fine aggregate (RFA) in this paper. In order to obtain a better improvement effect, the microbial mineralization conditions were first optimized. The effect of the pH value, temperature, bacterial concentration and calcium ion concentration on the mineralization ability of bacteria were investigated. The optimal microbial mineralization conditions were selected for the treatment of RFA and the microbial mineralization modification effect of RFA was evaluated based on the water absorption and crushing index. In addition, the natural fine aggregate (NFA), unmodified RFA and modified RFA were made into ordinary mortar, recycled mortar and modified recycled mortar, respectively. The workability, mechanical properties and chloride ion penetration resistance of mortars was investigated. Meanwhile, the precipitations formed by microbial mineralization were characterized using a scanning electron microscope (SEM) with an energy dispersive spectrometer (EDS) and X-ray diffraction (XRD). The pore structure of mortars was analyzed using the mercury intrusion porosimeter (MIP). The results showed that the bioprecipitations were mainly calcite calcium carbonate and the quality of the RFA was improved by microbial-induced calcium carbonate deposition. The water absorption and crushing index of the modified RFA decreased by 25.7% and 4.2%, respectively. Compared with the crushing index, the water absorption of the RFA was improved more obviously. The workability, mechanical performance, chloride ion penetration resistance and pore structure of the modified recycled mortar was improved. Compared with the recycled mortar, the fluidity of the modified recycled mortar was 7.3% higher, the compressive strength of 28 d was 7.0% higher and the 6 h electric flux was 18.8% lower. The porosity of the ordinary mortar, recycled mortar and modified recycled mortar was 16.49%, 20.83% and 20.27%, respectively. The strengthening of the modified recycled mortar performance may be attributed to the improvement of the mortar microstructure due to the enhancement of the RFA quality after the biotreatment.

Keywords: recycled mortar; recycled fine aggregate; microbial mineralization; compressive strength; chloride ion penetration resistance

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1. Introduction

The demolition and reconstruction of buildings generate a large amount of construction and demolition (C&D) waste, which not only invades the land, but also causes serious environmental pollution. Therefore, the disposal of C&D waste is an important issue and has received extensive attention. One of the effective ways is to make C&D waste into recycled materials and reuse it in concrete and mortar production [1–3]. As two kinds of important cement-based materials, concrete and mortar are widely used in civil engineering and play an important role in the disposal of C&D waste and other solid wastes [4,5]. C&D wastes contain various materials, such as concrete waste, glass waste, metal waste, etc. Concrete waste is usually made into recycled aggregates to replace natural aggregates. Powder made from waste glass could be utilized as a partial cement replacement in normal concrete [6]. However, compared with natural aggregates, recycled aggregates made of

concrete waste have a large amount of cement paste attached to the surface, which has defects such as high porosity, high water absorption, high crushing index and weak interface transition zones, resulting in the performance decline of the prepared recycled aggregate concrete [7,8]. Thus, the quality of the recycled aggregate needs to be improved to promote its application in actual projects.

At present, there are two main technical ways to improve the quality of the recycled aggregate, namely, removing the paste attached to the surface of the recycled aggregate (such as mechanical grinding, heat treatment, acid immersion, etc.) and strengthening the paste attached to the surface of the recycled aggregate (such as polymer immersion, sodium silicate treatment, carbonization treatment, etc.) [9–13]. These methods have improved the performance of the recycled aggregate to some extent, but there are still some shortcomings, such as mechanical grinding may cause secondary damage of the recycled aggregate, acid treatment may cause secondary pollution and polymer impregnation may have the compatibility problem of polymer and concrete. Therefore, researchers are actively optimizing the existing methods to improve the quality of the recycled aggregate or exploring new more efficient and environmentally friendly treatment methods.

Microbial-induced mineralization is a common natural phenomenon [14]. Many microorganisms in nature can use their own life activities to induce calcium carbonate deposition under suitable conditions. In recent years, this phenomenon has been used for the consolidation of loose particles [15,16], the restoration of stone cultural relics and concrete crack healing [17–20], etc. Inspired by the repair of concrete and stone damages, researchers have begun to explore the use of microbial-induced calcium carbonate deposition in-situ to repair the surface defects of the recycled aggregate and improve the performance of the recycled aggregate [21,22]. The results of several studies in the literature [9,23–30] show that microbial mineralization deposition can significantly reduce the water absorption of the recycled aggregate and improve the strength of the recycled aggregate, thus enhancing the performance of recycled aggregate concrete. The microbial modification process of the recycled aggregate is a milder reaction which neither consumes a large amount of energy nor causes secondary pollution. This low-energy consumption, pollution-free method is in line with the environmental protection of the recycling of C&D waste.

The recycled aggregate includes recycled coarse aggregate (RCA) with a particle size greater than 4.75 mm and recycled fine aggregate (RFA) with a particle size less than 4.75 mm, of which RFAs will account for a large part. Compared with RCA, RFA has higher water absorption and porosity, which has a greater impact on the properties of concrete and is difficult to utilize in engineering [31]. Therefore, it is of great significance to improve the quality of RFA and make it meet the requirements of engineering applications for making full use of C&D waste and alleviating the shortage trend of natural fine aggregates (NFAs). However, for the improvement of the recycled aggregate by microbial deposition technology, the current research mainly focuses on the modification and application of RCA [9,21–30]. The RCA after microbial modification is generally made into concrete and the performance improvement of the concrete is widely evaluated. Wang et al. [9] found that the compressive strength of the recycled aggregate concrete improved by 40% and the water absorption decreased by 27% after microbial-induced mineralization modification. The research results of Zeng et al. [26] showed that the microhardness of the interface transition zone of biomodified recycled aggregate concrete was improved, thus emerging higher compressive strength. Compared with the RCA, relatively little attention is paid to the RFA treatment by microbial mineralization. Wu et al. [24] treated RCA and RFA by microbial deposition at the same time, and the results showed that the water absorption of the RFA decreased more significantly. The water absorption of the RFA was reduced by 23%. Feng et al. [28] prepared mortar from microbial-modified RFA and studied the mechanical properties of mortar. The results showed that the flexural and compressive strength of the mortars cast by the modified RFAs were improved. To sum up, microbial-mineralization treatment has a good potential in improving the quality of RFA. However, the research on the properties of mortar containing biotreated RFA is still limited. In addition to the

mechanical properties, the working performance, the durability and the microstructure of mortar containing biotreated RFA need to be further studied.

In this study, microbial-induced mineralization deposition was used to improve the quality of the RFAs. In order to obtain a better improvement effect, the microbial-mineralization conditions were first optimized. The effect of the pH value, temperature, bacterial concentration and calcium ion concentration on the mineralization ability of bacteria were investigated. The optimal microbial mineralization conditions were selected for the treatment of the RFAs and the microbial mineralization modification effect of the RFAs was evaluated by the water absorption and crushing index. Then, the RFAs modified by microbial mineralization was made into recycled mortar. The workability, mechanical properties and chloride ion penetration resistance of the recycled mortar were investigated. In addition, the precipitations formed by microbial mineralization were characterized using a scanning electron microscope (SEM) and X-ray diffraction (XRD). The pore structure of the recycled mortar was analyzed using the mercury intrusion porosimeter (MIP). The performance improvement mechanism of the recycled mortar was also discussed.

2. Materials and Methods

2.1. Bacteria Strain

Sporosarcina pasteurii was selected for the modification of the RFAs in this study. This bacterium is nonpathogenic, does no harm to the ecological environment and can produce a large amount of urease in the metabolic process to induce $CaCO_3$ deposition. Table 1 shows the compositions of culture medium for this bacterium. Figure 1 shows the process of bacteria cultivation. After sterilization of the medium, bacteria were inoculated and the inoculated medium was cultured in a constant temperature oscillator (30 °C, 180 rpm) for 48 h. More detailed information about this bacterial strain can be found in our previously published studies [32,33].

Table 1. The compositions of culture medium.

Compositions	Content (g/L)
Yeast extract	20
Urea	20



(**a**) (**b**) (**c**)

Figure 1. The process of bacteria cultivation: (**a**) autoclave sterilization; (**b**) inoculation in ultra-clean workbench; (**c**) incubation in constant temperature oscillator.

2.2. Recycled Fine Aggregate (RFA)

The recycled fine aggregates used in this study were provided by a local renewable resource company and were produced by the waste cement concrete panels from an old road reconstruction and expansion project, as shown in Figure 2. The particle gradation of the RFA is shown in Figure 3. The fineness modulus of the RFAs is 2.95.



Figure 2. The recycled fine aggregates.

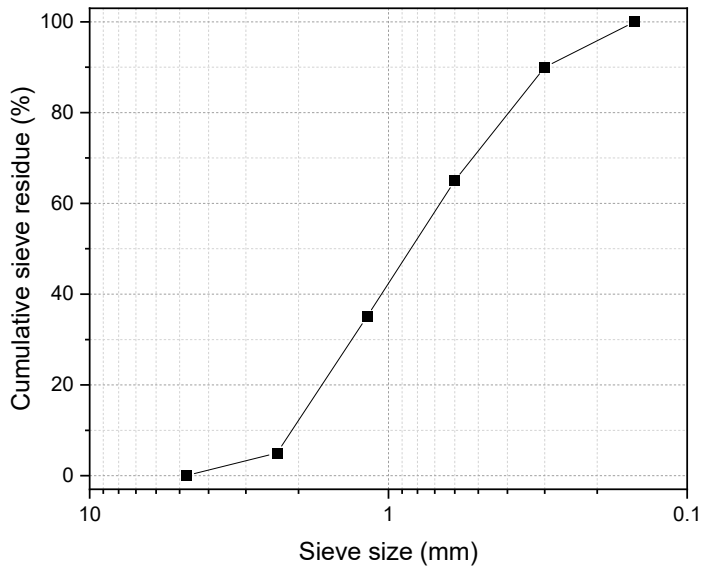


Figure 3. Particle gradation of recycled fine aggregate.

2.3. Optimization of Microbial Mineralization Conditions

Microbial-induced mineralization deposition was used to improve the quality of the RFA. In order to obtain better improvement effect, microbial mineralization conditions were first optimized. The effect of the pH value, temperature, bacterial concentration and calcium ion concentration on the mineralization ability of bacteria were investigated. Calcium chloride and urea were added into the bacterial solution to prepare a mixed solution, in which the concentration of calcium ion was 0.05 mol/L, the concentration of urea was 0.5 mol/L and the concentration of bacteria was 1.0×10^8 cells/mL. The pH value of the above mixed solution was adjusted to 8.0 and packed into conical flasks, each of which was 300 mL. The conical flasks with mixed solution were placed in a constant temperature oscillating incubator set at 30 °C for 7 days of mineralization and the weight of bioprecipitations produced were measured to evaluate the microbial mineralization capacity. When studying the effect of the pH value on microbial mineralization ability, the pH values of the mixed solutions were adjusted to 7.5, 8.0, 8.5, 9.0, 9.5, 10.0, 10.5 and 11.0, respectively. For the effect of temperature, the temperature was set as 20, 25, 30 and 35 °C, respectively. For the effect of bacterial concentration, the bacterial concentration was varied

to 10^6, 10^7, 10^8 and 10^9 cells/mL, respectively. For the effect of bacterial concentration, the calcium ion concentration was adjusted to 0.05, 0.10, 0.30, 0.50, 0.70 and 1.00 mol/L, respectively.

2.4. Microbial-Mineralization Modification Process of RFA

The most appropriate microbial-mineralization conditions was used to modify the RFA. The modification process is as follows: after soaking in acetic acid–chitosan solution (the concentrations of glacial acetic acid and chitosan are both 10 g/L) for 2 h, the RFA was taken out and dried. Then, the RFA with chitosan was immersed in the bacterial solution with the concentration of 1.0×10^8 cells/mL for 48 h. After that, the RFA loaded with bacteria was transferred to the mineralization solution (pH value is about 9.5) containing urea, calcium chloride and yeast extract for modification of 7 d. The urea concentration and calcium ion concentration in the mineralization solution are both 0.5 mol/L.

2.5. Performance Evaluation of Modified RFA

2.5.1. Water Absorption

The water absorption test was performed on the RFA before and after biodeposition treatment according to the method described in the literature [9]. After soaking in water for 24 h, the RFA was taken out from the container and the surface water was removed by a wrung out wet towel. Thus, the saturated-surface dry weight of RFA (M_1) was obtained. Then, the RFA in the saturated-surface dry condition was dried in an oven at 105 °C for about 24 h to constant weight and the dry weight of RFA (M_2) was obtained. The water absorption (W) was determined according to Equation (1).

$$W = \frac{M_1 - M_2}{M_2} \times 100\% \quad (1)$$

2.5.2. Crushing Index

The crushing index is a parameter indicating the strength and resistance to crush of the aggregate. The test was carried out in accordance with the regulations of the Chinese specification (GB/T 25176-2010). The RFA was sieved into four particle grades: 0.3~0.6 mm, 0.6~1.18 mm, 1.18~2.36 mm and 2.36~4.75 mm. The crushing index of each particle grade was measured and the largest one was taken as the crushing index of RFA. For the crushing index test of single particle grade, 330 g of RFA were put into the round steel mold of the crushing index tester and the surface was leveled. Subsequently, the mold with the RFA was placed on the loading machine and loaded it evenly to 25 kN at a speed of 500 N/s and stabilized for 5 s. After unloading, the RFA in the mold was poured out. The rushed particles were removed using a sieve with an aperture of the lower limit of each particle grade. The crushing index of single particle grade (Y_i) is calculated by Equation (2), where G_1 is the weight of RFA remaining on the sieve after sifting (g), and G_2 is the weight of RFA passing through the sieve (g).

$$Y_i = \frac{G_2}{G_2 + G_1} \times 100\% \quad (2)$$

2.6. Mortar Specimen Preparation

The NFA, unmodified RFA and modified RFA by microbial mineralization were made into ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX), respectively. The proportion of ordinary mortar is as follows: 450 g of Portland cement, 1350 g of NFA and 225 g of water. For recycled mortar and modified recycled mortar, all of the NFA was replaced by the RFA in equal volume. Meanwhile, because the water absorption of the RFA is higher than that of the NFA, additional water consumption was considered for recycled mortar and modified recycled mortar. The additional water was used to prewet the RFA before mixing. Two sizes of specimens were cast for each mortar, 40 mm × 40 mm × 40 mm prismatic mortar specimens were used for the flexural and compressive strength tests and φ100 mm × 50 mm cylindrical mortar specimens were

prepared for the chloride ion penetration resistance test. All test specimens were demolded 24 h after casting and then transferred to the standard curing room (20 ± 2 °C, RH > 95%) for continuous curing until the specified age.

2.7. Performance Evaluation of Mortars

The fluidity, flexural and compressive strength and resistance to chloride ion penetration of ordinary mortar, recycled mortar and modified recycled mortar were investigated in the study. The specific operation method of mortar fluidity test was in accordance with the Chinese standard (GB/T 2419-2005). The flexural and compressive strength tests were carried out in accordance with the regulations of the Chinese specification (GB/T 17671-2021) on mortar specimens at standard curing ages of 3 d, 7 d, 14 d and 28 d, respectively. The flexural strength tests were carried out at a loading speed of 50 N/s and that of compressive strength tests were at a loading speed of 2.4 kN/s. The chloride ion penetration resistance tests were conducted in accordance with the relevant provisions in Chinese standard (GB/T 50082-2009). After curing for 28 d, mortar specimens were taken out and carried out on concrete chloride ion electric flux meter.

2.8. Microstructure Investigation

2.8.1. Characterization of Biodeposition Precipitation

Gemini SEM 300 field emission scanning electron microscope (Carl Zeiss Company, Oberkochen, Germany) with an energy dispersive spectrometer (EDS) (Carl Zeiss Company, Oberkochen, Germany) and D8-Advance X-ray diffractometer (Bruker-AXS company, Karlsruhe, Germany) was used to investigate the morphology and chemical composition of biodeposition precipitation. The precipitations formed by microbial mineralization were dried in a vacuum desiccator and grinded to pass a 75 μm mesh sieve for SEM/EDS and XRD tests.

2.8.2. Pore Structure Analysis of Mortars

The porosity and pore size distribution of ordinary mortar, recycled mortar and modified recycled mortar were analyzed using AutoPore IV 9500 mercury intrusion porosimeter (Micromeritics Instrument Corporation, Atlanta, America). The maximum intrusion pressure is 33,000 psia and the pore diameter range that can be tested is from approximately 360 to 0.005 μm. At the age of 28 d, mortar specimen was taken out from the standard curing room and crushed into blocks suitable for MIP test. After that, the selected test samples were soaked into absolute ethanol for 24 h to stop further hydration. Then, the samples were put into the vacuum drying oven for drying until MIP test.

3. Results and Discussion

3.1. Optimization of Microbial Mineralization Conditions

Figure 4 shows the effect of pH value, temperature, bacterial concentration and calcium ion concentration on the mineralization ability of bacteria. It can be seen from Figure 4a that, with the increase of pH value from 7.5 to 11.0, the weight of bioprecipitations first increases and then decreases. When the pH values are 9.5 and 10, the mineralization ability of bacteria is good. However, when the pH value exceeds 10, the mineralization ability of the bacteria starts to decrease. This phenomenon may be due to the reduction of bacterial growth rate and urease activity due to the high pH value [28,34]. It can be seen from Figure 4b that when temperature is 25~35 °C, the weight of bioprecipitations increases with the increase of temperature. This may be due to the increase of urease activity and urea hydrolysis rate at a higher temperature [35]. However, the increase of bioprecipitations with temperature is not significant. This is because the growth of bacteria and urease activity are both very good in the temperature range of 25~35 °C.

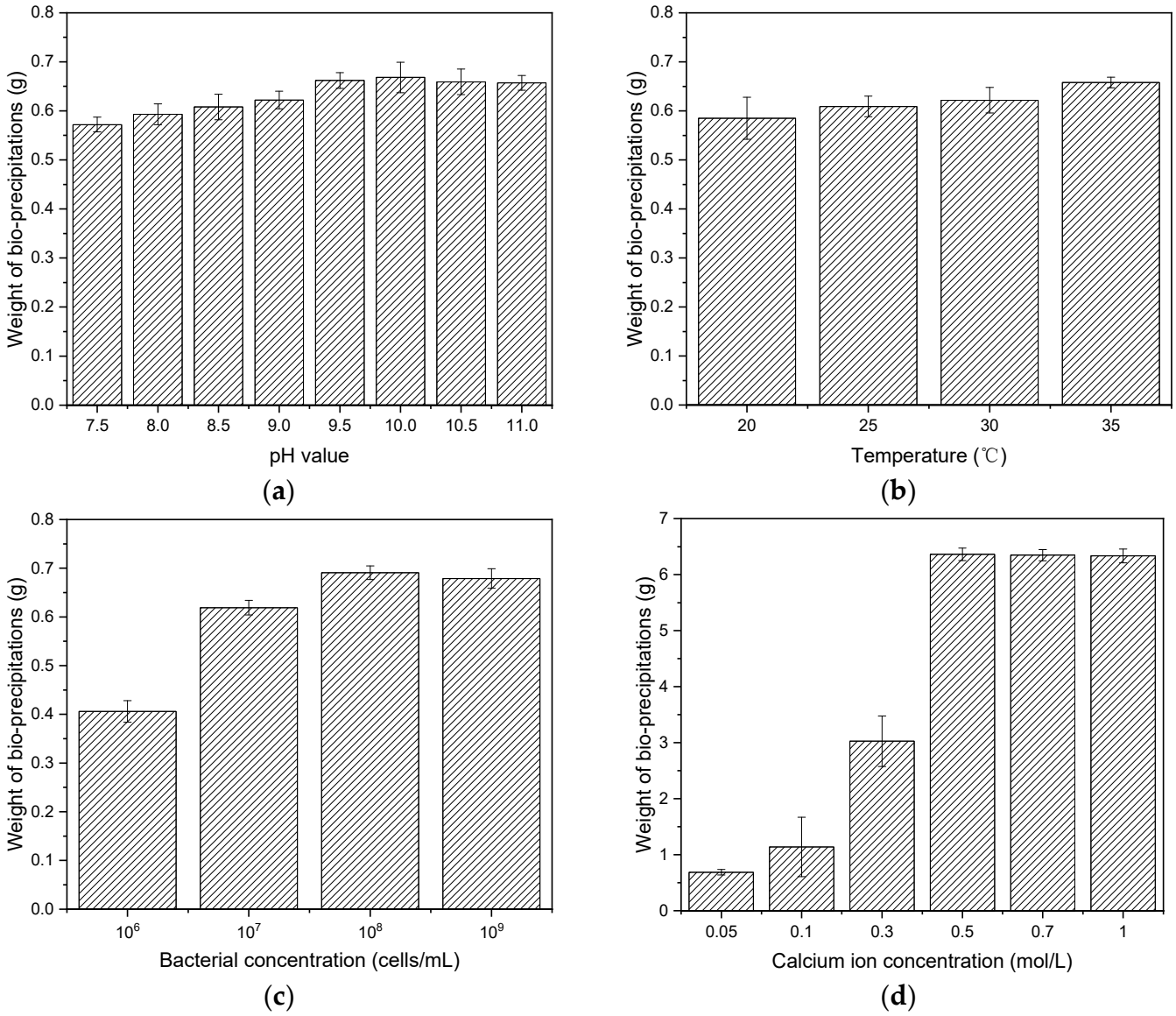


Figure 4. The effect of pH value, temperature, bacterial concentration and calcium ion concentration on the mineralization ability of bacteria. (**a**) pH value, (**b**) Temperature, (**c**) Bacterial concentration, (**d**) Calcium ion concentration.

It can be seen from Figure 4c that, when the bacterial concentration is less than 10^8 cells/mL, the increase of bacterial concentration will lead to the increase of bioprecipitations weight. When the bacterial concentration is greater than 10^8 cells/mL, the bioprecipitations weight will not increase with the further increase of bacterial concentration. More bacterial cells can not only hydrolyze more urea to produce a higher pH, but also provide more nucleation sites for microbial mineralization, thus increasing the weight of bioprecipitations. It can be seen from Figure 4d that the weight of bioprecipitations is significantly related to the concentration of calcium ion. when the calcium ion concentration is 0.5 mol/L, the weight of bioprecipitations reaches the maximum. After this peak, the weight of bioprecipitations decreases slightly with the further increase of calcium ion concentration. This could be attributed to saturation effect of calcium ion and inhibited bacteria growth at a higher calcium ion concentration [36]. Based on the above results, the pH value of 9.5, temperature of 30 °C, bacterial concentration of 10^8 cells/mL and calcium ion concentration of 0.5 mol/L were selected as the optimal microbial mineralization conditions for the treatment of the RFA.

3.2. Microbial-Mineralization-Modification Effect of RFA

The water absorption and crushing index of the RFA before and after microbial-mineralization modification are shown in Table 2. After microbial-mineralization modification, the water absorption and crushing index of the RFA decreased by 25.7% and 4.2%, respectively, which indicated that the quality of treated RFA was enhanced. Compared with crushing index, the water absorption of the RFA was improved more obviously. The improvement of the RFA performance is due to the bioprecipitations deposited on the surface and in the pores of RFA. Qiu et al. [22] and Wang et al. [9] found that a large amount of calcium carbonate precipitations was deposited on the aggregate surface after microbial treatment. Garcia-Gonzalez et al. [37] observed that the bioprecipitations not only covered on the aggregate surface, but also filled some superficial pores. The bioprecipitations formed will act as a barrier for the penetration of water and thus reduce the water absorption of recycled aggregate. Qiu et al. [22] pointed out that the decrease of water absorption may also be related to the organic matter produced in the process of microbial-induced calcium carbonate, which may reduce the wettability of recycled aggregate surface. Microbial-deposition treatment strengthens the recycled aggregate, thus reducing the crushing index. However, the strengthening effect is mainly concentrated on the surface, and the interior of recycled aggregate is still weak [9]. Therefore, the reduction of the crushing index of the recycled aggregate is not obvious.

Table 2. Water absorption and crushing index of RFA before and after microbial-mineralization modification.

Recycled Aggregate Type	Water Absorption/%	Crushing Index/%
Untreated RFA	7.0 ± 0.3	24.6 ± 0.4
Treated RFA	5.2 ± 0.2	23.6 ± 0.5

Figure 5 shows the SEM images and EDS spectrum of bioprecipitations scraped from treated RFA. As shown in Figure 5a–c, the morphology of bioprecipitations is mostly spherical and the size of precipitation particles is about 5–20 μm. Meanwhile, a lot of bacterial imprints can be observed on the surface of bioprecipitations, which indicates that bacteria are involved in the precipitation process. It can be seen from the EDS spectrum of bioprecipitations in Figure 5d that the bioprecipitations contain three elements C, O and Ca, which are analyzed as calcium carbonate. Figure 6 shows the XRD pattern of bio-precipitations scraped from the treated RFA. It can be found that the bioprecipitations are mainly calcite calcium carbonate. Sporosarcina pasteurii was used for the modification of the RFA in this study. This bacterium can produce a large amount of urease in the metabolic process, which can be used for urea hydrolysis to produce CO_3^{2-}, and then reacts with Ca^{2+} from mineralization solution to produce $CaCO_3$ precipitation, as shown in Equations (3)–(5). The cell wall of the bacteria is negatively charged and can attract positively charged Ca^{2+} and act as a nucleation site.

$$CO(NH_2)_2 \rightarrow CO_3^{2-} + NH_4^+ \quad (3)$$

$$Cell + Ca^{2+} \rightarrow Cell\text{-}Ca^{2+} \quad (4)$$

$$Cell\text{-}Ca^{2+} + CO_3^{2-} \rightarrow Cell\text{-}CaCO_3 \quad (5)$$

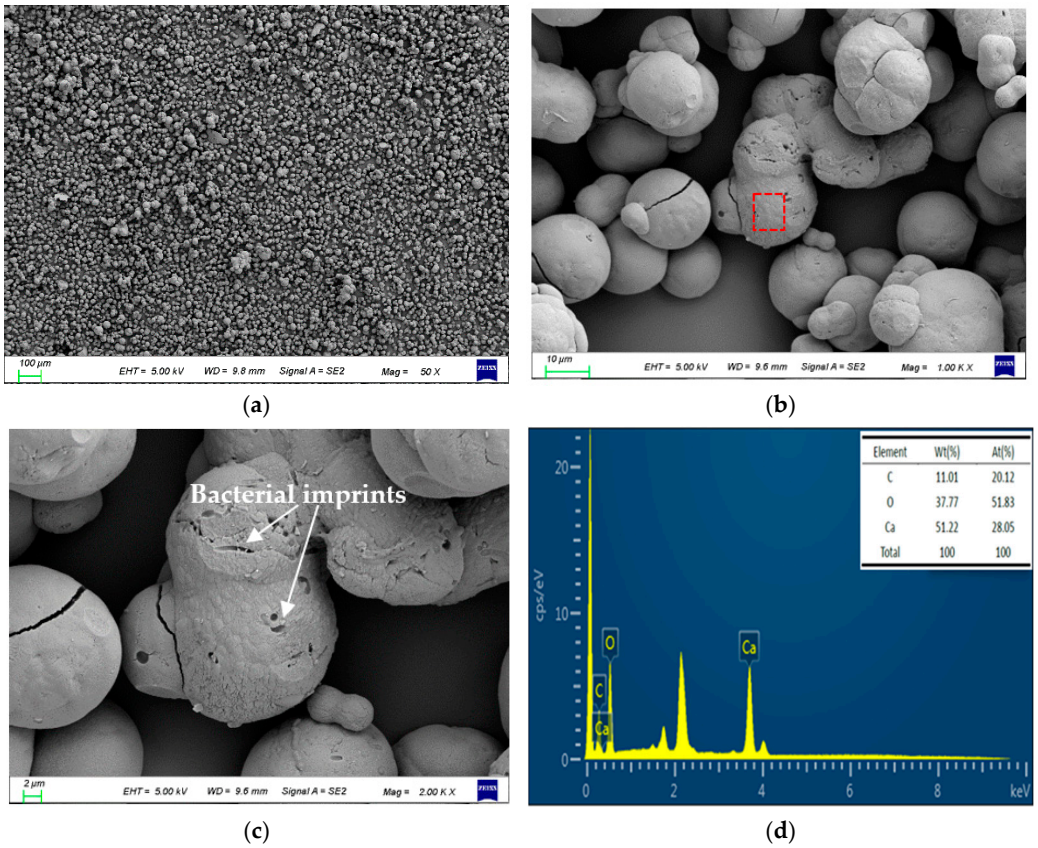


Figure 5. SEM images and EDS spectrum of bioprecipitations scraped from treated RFA: (**a**) at 50× magnification; (**b**) at 1000× magnification; (**c**) at 2000× magnification; (**d**) EDS spectrum of bioprecipitations in the red rectangle in image (**b**).

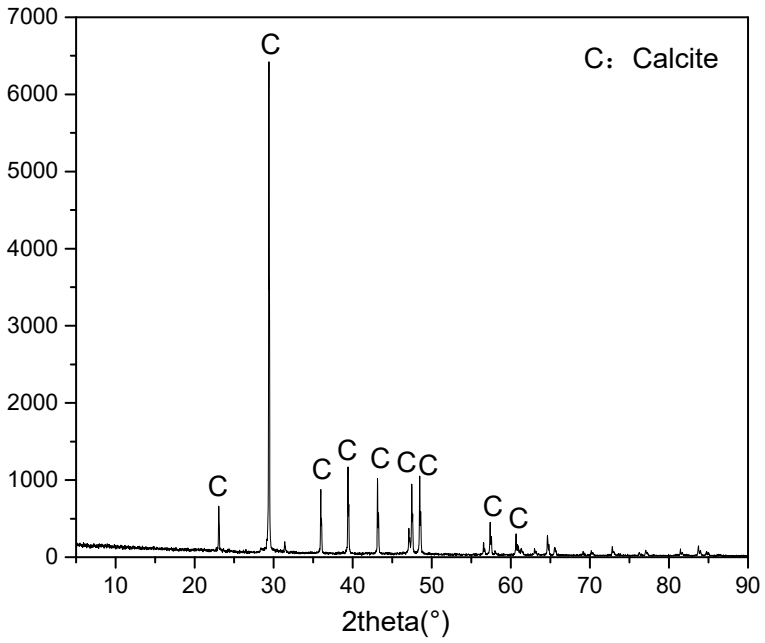


Figure 6. XRD pattern of bioprecipitations scraped from treated RFA.

3.3. Properties of Modified Recycled Mortar

3.3.1. Fluidity

Fluidity is an important index of the mortar workability. The fluidity of ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX) is shown in Table 3. It can be found that despite the additional water consumption is considered, the fluidity of recycled mortar is still not as good as that of ordinary mortar. This may be due to the irregular particle shape of the RFA obtained after crushing, and the increased friction between the aggregates due to more edges and corners on the surface. However, the fluidity of the modified recycled mortar is 7.3% higher than the recycled mortar, indicating that the modified RFA can improve the fluidity of the recycled mortar. This is because the surface of the modified RFA is coated with a layer of calcium carbonate crystals, which not only reduces the water absorption of the aggregate itself, but also improves the particle morphology of the aggregate.

Table 3. The fluidity of ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX).

Mortar Type	Fluidity/mm
Ordinary mortar (M-JZ)	220
Recycled mortar (M-WG)	182
Modified recycled mortar (M-GX)	198

3.3.2. Flexural and Compressive Strength

Figure 7 shows the flexural and compressive strength of ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX) cured for 3 d, 7 d, 14 d and 28 d, respectively. As shown in Figure 7a, the compressive strength of the recycled mortar (M-WG) was obviously lower than that of the ordinary mortar (M-JZ). When the curing age is 28 days, the compressive strength of the ordinary mortar reaches 45.6 MPa, while the compressive strength of the recycled mortar is just 30.7 MPa. This is because the quality of the RFA is inferior to that of the NFA. After microbial mineralization, the quality of the RFA was improved. The water absorption and crushing index of the modified RFA decreased. Therefore, the compressive strength of the modified recycled mortar (M-GX) made with the modified RFA have been improved. The compressive strength of the modified recycled mortar cured for 3 d, 7 d, 14 d and 28 d is 16.9%, 16.1%, 7.3% and 7.0% higher than that of the recycled mortar.

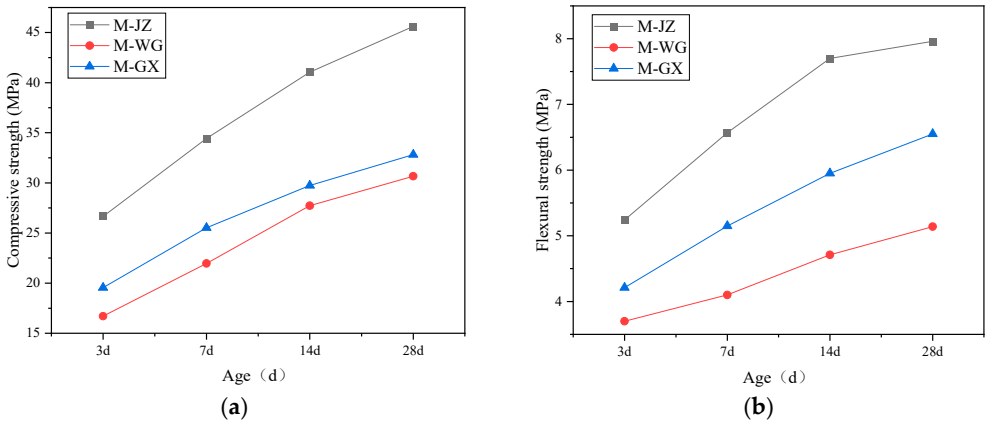


Figure 7. The flexural and compressive strength of ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX). (**a**) Compressive strength, (**b**) Flexural strength.

It can be seen from Figure 7b that the change trend of flexural strength is consistent with that of the compressive strength. The flexural strength of the recycled mortar (M-WG)

at 3 d, 7 d, 14 d and 28 d is 70.6%, 62.4%, 61.2% and 64.6% of that of the ordinary mortar (M-JZ), respectively, while the flexural strength of the modified recycled mortar (M-GX) at 3 d, 7 d, 14 d and 28 d is 80.3%, 78.4%, 77.3% and 82.3% of that of the ordinary mortar (M-JZ), respectively, which is 13.7%, 25.6%, 26.3% and 27.4% higher than that of the recycled mortar (M-WG) at 3 d, 7 d, 14 d and 28 d. It is worth noting that the flexural strength of the recycled mortar seems to be improved more significantly than the compressive strength after microbial-mineralization modification. The same results were obtained in the research by Feng et al. [28].

3.3.3. Resistance to Chloride Ion Penetration

Chloride ion penetration resistance is usually an effective index to evaluate the durability of cement-based materials. The electric flux method was used to obtain the resistance of mortar to chloride ion penetration. Figure 8 shows that the 6 h electric flux of the ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX). The 6 h electric flux of the recycled mortar was significantly higher than that of the ordinary mortar, indicating that the chloride ion penetration resistance of the recycled mortar was obviously lower than that of the ordinary mortar. Compared with the recycled mortar, the 6 h electric flux of the modified recycled mortar was 18.8% lower, indicating that microbial mineralization treatment of the RFA improved the chloride ion penetration resistance of the recycled mortar. This may be attributed to the improvement of the microstructure of the RFA itself after microbial-mineralization treatment and the denser interface transition zone between the modified RFA and the cement matrix. Wang et al. [9] found that after microbial treatment, the porosity of the recycled aggregate decreased. Wu et al. [24] observed a denser ITZ in the biotreated recycled mortar and speculated that the calcium carbonate particles adhering to the aggregate surface may provide nucleation sites for hydration products, thus promoting the hydration reaction in the ITZ. The research results of Zhao et al. [23] showed that the microhardness of the interface transition zone of the recycled aggregate was improved by the microbial-mineralization treatment.

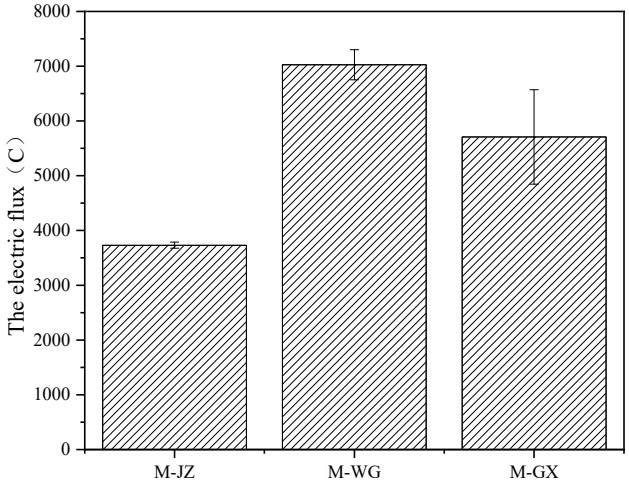
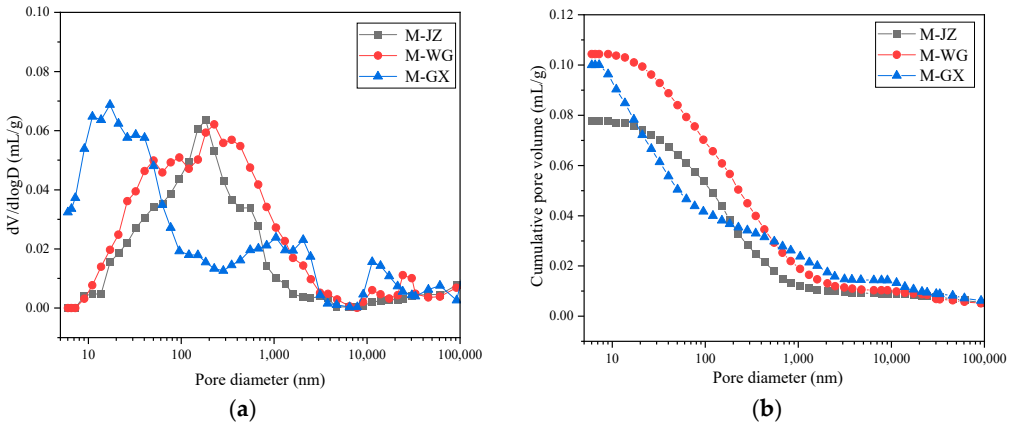


Figure 8. The electric flux of ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX).

3.3.4. Pore Structure Analysis of Modified Recycled Mortar

The pore structure of the ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX) was analyzed by MIP test. Figure 9 shows the pore size distribution and cumulative pore volume of different types of mortar. It can be found from Figure 9a that the pore size distributions of the ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX) were different. According to the Figure 9b, the total porosity of the ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX) can be calculated as 16.49%, 20.83% and 20.27%, respectively. The

total porosity of the unmodified recycled mortar (M-WG) is obviously higher than that of the ordinary mortar (M-JZ). Compared with the unmodified recycled mortar, the porosity of the modified recycled mortar was only slightly reduced, but the pore diameter had a clear trend of refinement. The results can well explain the mechanical properties and chloride ion penetration resistance of the recycled mortars, which are correlated closely with the porosity and the pore size distribution [38,39]. For the recycled mortar, the significantly higher porosity resulted in its flexural and compressive strength was significantly lower than that of the ordinary mortar (Figure 7). At the same time, its electric flux was obviously higher than that of the ordinary mortar (Figure 8). For the modified recycled mortar, reduction of porosity and refinement of pores may be attributed to decrease of porosity of the recycled aggregate after the microbial treatment [9], and denser interface transition zone (ITZ) between the modified recycled aggregate and the mortar matrix paste [24]. As a result, the mechanical properties and chloride ion penetration resistance of the modified recycled mortars were improved compared to the ordinary mortar.



(**a**) (**b**)

Figure 9. Pore size distribution and cumulative pore volume of ordinary mortar (M-JZ), recycled mortar (M-WG) and modified recycled mortar (M-GX): (**a**) pore size distribution; (**b**) cumulative pore volume.

4. Conclusions

The optimal microbial mineralization conditions were used for the treatment of the RFA in this paper. The microbial mineralization modification effect of the RFA was evaluated based on water absorption and crushing index. In addition, the NFA, unmodified RFA and modified RFA were made into ordinary mortar, recycled mortar and modified recycled mortar, respectively. The workability, mechanical properties and chloride ion penetration resistance of the mortars were investigated. Meanwhile, the precipitations formed by microbial mineralization were characterized using SEM and XRD, and the pore structure of the mortars was analyzed using MIP. The following conclusions can be drawn:

- The quality of the RFA was improved after microbial mineralization modification. The water absorption and crushing index of the modified RFA decreased by 25.7% and 4.2%, respectively. Compared with crushing index, the water absorption of the RFA was improved more obviously.
- The workability, mechanical performance and chloride ion penetration resistance of the recycled mortar were obviously inferior to those of the ordinary mortar. Microbial-mineralization pretreatment of the RFA enhanced the performance of the recycled mortar. Compared with the recycled mortar, the fluidity of the modified recycled mortar was 7.3% higher, the compressive strength of 28d was 7.0% higher and the 6h electric flux was 18.8% lower.
- The pore structure of the modified recycled mortar was improved. Compared with the unmodified recycled mortar, the porosity of the modified recycled mortar was reduced and the pore diameter had a clear trend of refinement.

5. Future Recommendations

For the durability of mortar containing RFA modified by microbial mineralization, only the resistance to chloride ion penetration was studied in this paper, and the frost resistance and carbonation resistance can be further investigated later. In addition, in order to better explain the mechanism of performance improvement of modified recycled mortar, it is also necessary to investigate the improvement of ITZ between the modified RFA and the cement matrix.

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References

1. Wang, R.J.; Yu, N.N.; Li, Y. Methods for improving the microstructure of recycled concrete aggregate: A review. *Constr. Build. Mater.* **2020**, *242*, 118164. [CrossRef]
2. Tam, V.W.Y.; Wattage, H.; Le, K.N.; Buteraa, A.; Soomro, M. Methods to improve microstructural properties of recycled concrete aggregate: A critical review. *Constr. Build. Mater.* **2021**, *270*, 121490. [CrossRef]
3. Kisku, N.; Joshi, H.; Ansari, M.; Panda, S.K.; Nayak, S.; Dutta, S.C. A critical review and assessment for usage of recycled aggregate as sustainable construction material. *Constr. Build. Mater.* **2017**, *131*, 721–740. [CrossRef]
4. Marvila, M.T.; Azevedo, A.R.G.; Monteiro, S.N. Verification of the application potential of the mathematical models of lyse, abrams and molinari in mortars based on cement and lime. *J. Mater. Res. Technol.* **2020**, *9*, 7327–7334. [CrossRef]
5. Azevedo, A.R.; Marvila, M.T.; Zanelato, E.B.; Alexandre, J.; Xavier, G.C.; Cecchin, D. Development of mortar for laying and coating with pineapple fibers. *Rev. Bras. Eng. Agr. Amb.* **2020**, *24*, 187–193. [CrossRef]
6. Raydan, R.; Khatib, J.; Jahami, A.; El Hamoui, A.K.; Chamseddine, F. Prediction of the mechanical strength of concrete containing glass powder as partial cement replacement material. *Innov. Infrastruct. Solut.* **2022**, *7*, 311. [CrossRef]
7. Kou, S.C.; Poon, C.S.; Etxeberria, M. Influence of recycled aggregates on long term mechanical properties and pore size distribution of concrete. *Cem. Concr. Compos.* **2011**, *33*, 286–291. [CrossRef]
8. Shi, C.; Li, Y.; Zhang, J.; Li, W.; Chong, L.; Xie, Z. Performance enhancement of recycled concrete aggregate: A review. *J. Clean. Prod.* **2016**, *112*, 466–472. [CrossRef]
9. Wang, J.; Vandevyvere, B.; Vanhessche, S.; Schoon, J.; Boon, N.; Belie, N.D. Microbial carbonate precipitation for the improvement of quality of recycled aggregates. *J. Clean. Prod.* **2017**, *156*, 355–366. [CrossRef]
10. Katz, A. Treatments for the Improvement of Recycled Aggregate. *J. Mater. Civil. Eng.* **2004**, *16*, 531–535. [CrossRef]
11. Kou, S.C.; Poon, C.S. Properties of concrete prepared with PVA-impregnated recycled concrete aggregates. *Cem. Concr. Compos.* **2010**, *32*, 649–654. [CrossRef]
12. Tsujino, M.; Noguchi, T.; Tamura, M.; Kanematsu, M.; Maruyama, I. Application of Conventionally Recycled Coarse Aggregate to Concrete Structure by Surface Modification Treatment. *J. Adv. Concr. Technol.* **2007**, *5*, 13–25. [CrossRef]
13. Tam, V.W.Y.; Tam, C.M.; Le, K.N. Removal of cement mortar remains from recycled aggregate using pre-soaking approaches. *Resour. Conserv. Recy.* **2007**, *50*, 82–101. [CrossRef]
14. Boquet, E.; Boronat, A.; Ramos-Cormenzana, A. Production of calcite (calcium carbonate) crystals by soil bacteria is a general phenomenon. *Nature* **1973**, *246*, 527–529. [CrossRef]
15. Yu, X.N.; Rong, H. Seawater based MICP cements two-phase/one-phase cemented sand blocks. *Appl. Ocean. Res.* **2022**, *118*, 102972. [CrossRef]
16. Yu, X.N.; Yang, H.Q.; Wang, H. A cleaner biocementation method of soil via microbially induced struvite precipitation: A experimental and numerical analysis. *J. Environ. Manage.* **2022**, *316*, 115280. [CrossRef] [PubMed]
17. Qian, C.X.; Yu, X.N.; Zheng, T.W.; Chen, Y.Q. Review on bacteria fixing CO2 and bio-mineralization to enhance the performance of construction materials. *J. CO2 Util.* **2022**, *55*, 101849. [CrossRef]
18. Muynck, W.D.; Belie, N.D.; Verstraete, W. Microbial carbonate precipitation in construction materials: A review. *Ecol. Eng.* **2010**, *36*, 118–136. [CrossRef]

19. Wiktor, V.; Jonkers, H.M. Quantification of crack-healing in novel bacteria-based self-healing concrete. *Cem. Concr. Compos.* **2011**, *33*, 763–770. [CrossRef]
20. Tittelboom, K.V.; Belie, N.D.; Muynck, W.D.; Verstraete, W. Use of bacteria to repair cracks in concrete. *Cem. Concr. Res.* **2010**, *40*, 157–166. [CrossRef]
21. Grabiec, A.M.; Klama, J.; Zawal, D.; Krupa, D. Modification of recycled concrete aggregate by calcium carbonate biodeposition. *Constr. Build. Mater.* **2012**, *34*, 145–150. [CrossRef]
22. Qiu, J.S.; Tng, D.; Yang, E.H. Surface treatment of recycled concrete aggregates through microbial carbonate precipitation. *Constr. Build. Mater.* **2014**, *57*, 144–150. [CrossRef]
23. Zhao, Y.X.; Peng, L.G.; Zeng, W.L.; Poon, C.S.; Lu, Z.M. Improvement in properties of concrete with modified RCA by microbial induced carbonate precipitation. *Cem. Concr. Compos.* **2021**, *124*, 104251. [CrossRef]
24. Wu, C.R.; Zhu, Y.G.; Zhang, X.T.; Kou, S.C. Improving the properties of recycled concrete aggregate with bio-deposition approach. *Cem. Concr. Compos.* **2018**, *94*, 248–254. [CrossRef]
25. Zhu, Y.G.; Li, Q.Q.; Xu, P.Z.; Wang, X.R.; Kou, S.C. Properties of Concrete Prepared with Recycled Aggregates Treated by Bio-Deposition Adding Oxygen Release Compound. *Matererials* **2019**, *12*, 2147. [CrossRef]
26. Zeng, W.L.; Zhao, Y.X.; Chi, S.P.; Zhang, Y.F.; Lu, Z.M.; Shah, S.P. Using microbial carbonate precipitation to improve the properties of recycled aggregate. *Constr. Build. Mater.* **2019**, *228*, 116743. [CrossRef]
27. Singh, L.P.; Bisht, V.; Aswathy, M.S.; Chaurasia, L.; Gupta, S. Studies on performance enhancement of recycled aggregate by incorporating bio and nano materials. *Constr. Build. Mater.* **2018**, *181*, 217–226. [CrossRef]
28. Feng, Z.Y.; Zhao, Y.X.; Zeng, W.L.; Lu, Z.M.; Shah, S.P. Using microbial carbonate precipitation to improve the properties of recycled fine aggregate and mortar. *Constr. Build. Mater.* **2020**, *230*, 116949. [CrossRef]
29. Ouyang, J.Z.; Liu, K.W.; Sun, D.S.; Xu, W.Y.; Wang, A.G.; Ma, R. A focus on Ca2+ supply in microbial induced carbonate precipitation and its effect on recycled aggregate. *J. Build. Eng.* **2022**, *51*, 104334. [CrossRef]
30. Wang, R.X.; Jin, P.; Ding, Z.C.; Zhang, W. Surface modification of recycled coarse aggregate based on Microbial Induced Carbonate Precipitation. *J. Clean. Prod.* **2021**, *328*, 129537. [CrossRef]
31. Duan, Z.; Hou, S.; Xiao, J.; Li, B. Study on the essential properties of recycled powders from construction and demolition waste. *J. Clean. Prod.* **2020**, *253*, 119865. [CrossRef]
32. Luo, M.; Liu, Y.; Li, X.; Dai, J. Crack self-healing of cement mortar containing ureolytic bacteria immobilized in artificial functional carrier under different exposure environments. *Buildings* **2022**, *12*, 1348. [CrossRef]
33. Luo, M.; Li, X.; Liu, Y.; Jing, K. Application of artificial functional aggregates encapsulated bacteria for self-healing of cement mortars. *J. Mater. Civil Eng.* **2022**, *34*, 04022291. [CrossRef]
34. Cuzman, O.A.; Rescic, S.; Richter, K.; Wittig, L.; Tiano, P. Sporosarcina pasteurii use in extreme alkaline conditions for recycling solid industrial wastes. *J. Biotechnol.* **2015**, *214*, 49–56. [CrossRef] [PubMed]
35. Muynck, W.D.; Verbeken, K.; Belie, N.D.; Verstraete, W. Influence of temperature on the effectiveness of a biogenic carbonate surface treatment for limestone conservation. *Appl. Microbiol. Biot.* **2013**, *97*, 1335–1347. [CrossRef]
36. Muynck, W.D.; Verbeken, K.; Belie, N.D.; Verstraete, W. Influence of urea and calcium dosage on the effectiveness of bacterially induced carbonate precipitation on limestone. *Ecol. Eng.* **2010**, *36*, 99–111. [CrossRef]
37. Garcia-Gonzalez, J.; Rodriguez-Robles, D.; Wang, J.; De Belie, N.; Ma Moran-del Pozo, J.; Ignacio Guerra-Romero, M.; Juan-Valdes, A. Quality improvement of mixed and ceramic recycled aggregates by biodeposition of calcium carbonate, Constr. *Build. Mater.* **2017**, *154*, 1015–1023. [CrossRef]
38. Wang, L.; Huang, Y.; Zhao, F.; Huo, T.; Chen, E.; Tang, S. Comparison between the Influence of Finely Ground Phosphorous Slag and Fly Ash on Frost Resistance, Pore Structures and Fractal Features of Hydraulic Concrete. *Fractal Fract.* **2022**, *6*, 598. [CrossRef]
39. Wang, L.; He, T.; Zhou, Y.; Tang, S.; Tan, J.; Liu, Z.; Su, J. The influence of fiber type and length on the cracking resistance, durability and pore structure of face slab concrete. *Constr. Build. Mater.* **2021**, *282*, 122706. [CrossRef]