Preparation of artificial aggregate using waste concrete powder and CO₂ fixed by microorganisms

Xiao Zhang1,2,3,4 · Chunxiang Qian1,2,3,4 · Dengmin Xie1,2,3,4

Received: 22 August 2021 / Accepted: 12 December 2021 / Published online: 12 January 2022
© The Author(s), under exclusive licence to Springer-Verlag GmbH Germany, part of Springer Nature 2022

Abstract
High carbon emissions, shortages of natural aggregates and environmental pollution of waste concrete powder (WCP) have become open issues for the traditional concrete industry. Aggregates prepared by crushing and screening waste concrete usually possess poor mechanical properties. Meanwhile, the WCP cannot be effectively utilized. This paper proposes a novel approach based on microorganisms for strengthening mechanical properties and improving CO₂ sequestration of ‘newly’ artificial aggregates prepared by cold-bonding pelletization of WCP. Specifically, the microorganisms enhanced the artificial aggregates, resulting in their apparent density, crushing strength and water absorption increasing to 2620 kg/m³, 9.1 MPa and 4.8%, respectively. With the increase of well-crystallized mineralization products, the artificial aggregates exhibited a denser microstructure where the porosity decreased from 20.9 to 13.9%. The CO₂ fixation of artificial aggregates increased from 7.4 to 16.0 wt. % due to the existence of microorganisms. The compressive strength of concrete indicated that artificial aggregate could partially substitute the natural aggregates without affecting its strength, and a better substitution rate should be controlled within 50%. This method improves waste resource utilization and CO₂ emission reduction, showing good potential for future applications.

Graphical abstract
Preparation process of the artificial aggregate and its application.

Keywords Waste concrete · Artificial aggregate · Microorganism · CO₂ fixation

Introduction
Concrete is the most widely used building material in the world (Ahmed Shaikh et al. 2019). At present, China’s foundation and building construction are in the stage of rapid development, and the amount of concrete is enormous. As an indispensable component of concrete, sand and gravel have gradually become the mineral resource with the most significant production and consumption (China in 2019), more than 20 billion tons. The increasing demand for sand and
gravel resources has led to many environmental problems, such as overexploitation, the environmental pollution and high energy consumption in production and transportation (Jiang et al. 2020a, b). At the same time, due to restrictions on the mining area, sand and gravel are often in short supply. Environmental pollution and insufficient supply lead to a resource crisis, which seriously impacts construction and economic development. In addition, with the demolition of old concrete buildings, a large amount of alkaline waste concrete will be produced (Xiao et al. 2018), causing environmental pollution. Most of them are directly stacked or buried, which occupy land and consume many financial resources. On the other hand, the concrete industry has become an essential source of carbon dioxide emissions. New buildings increase the demand for concrete, and the construction industry will face tremendous pressure to reduce carbon dioxide emissions (Li et al. 2021). Therefore, to maintain the sustainable development of the construction industry, there is an urgent need to solve the problems of shortages of natural aggregates, inefficient treatment of waste concrete and huge carbon dioxide emissions.

Based on the above reasons, the preparation of recycled aggregate from waste concrete as building materials effectively alleviates the pressure of shortage of natural aggregate. This can reduce environmental pollution and promote the recycling of waste solid waste. However, the traditional recycled aggregates prepared by crushing and screening processes are mainly composed of broken brick and concrete aggregate, which have some problems, such as the low strength grade, high water absorption and unstable quality (Kisku et al. 2017; Mistri et al. 2021). Many fine concrete powders are difficult to use effectively and have a low utilization rate in preparing recycled aggregate. In order to improve the quality of recycled aggregate and make full use of all components of waste concrete, new technical processes and methods should be proposed to prepare high-quality aggregate. Recently, more studies have focused on using the cold-bonding pelletization technique to fabricate artificial aggregates by various industrial by-products, such as fly ash (Narattha and Chaipanich 2018), steel slag powder (Pang et al. 2015), solid waste incineration bottom ash (Tang and Brouwers 2017), blast furnace slag (Fca et al. 2015), solid waste fine powder (Job et al. 2019) and other solid waste to prepare aggregates. The cold-bonding pelletization method polymerizes powdered material into aggregate particles with a specific particle size and then strengthens the particles to achieve the expected strength. This method can provide a new idea for the utilization of waste concrete fine powder. Because minerals containing calcium and magnesium have good carbonization activity, mineral carbonation can provide strength. Waste concrete is also a material with a high calcium mineral content. Using the CO$_2$ curing method to refine the pores of artificial aggregate and improve aggregate performance has gradually become the research focus. The method avoids the problems of cost and environmental pollution caused by a high binder content, chemical impregnation and strong alkali excitation. It can also realize carbon sequestration and reduce carbon emission, which is conducive to the sustainable development of the construction industry. In the literature, artificial aggregate treated by carbonation leads to improved density, water absorption and strength (Shi et al. 2015). Zhan et al. (2014) showed that artificial aggregates with smaller particle sizes were more easily carbonated. The carbonation process proceeded rapidly within the first 2 h. Jiang et al. (Jiang et al. 2020a, b) used waste concrete to prepare artificial aggregates. After adding calcium hydroxide and CO$_2$ curing, the aggregate strength reached 3.0 MPa. Kou et al. (2014) reported that carbonation decreased the water absorption of aggregates by approximately 40% and greatly increased the strength. Pan et al. (2017) indicated the optimal CO$_2$ curing condition was identified as a CO$_2$ concentration of 70% and moisture content of 5%. Xiao et al. (2016) suggested that the carbonation effect of aggregates with a high water-to-binder ratio is more prominent. To obtain a better carbonization effect of aggregates, high specific surface area, high pressure, prolongation time and other conditions are usually used to accelerate and improve carbonization. In addition to the above methods, microbial enzyme catalysis can improve the carbon capture capacity of minerals and accelerate the carbonation reaction. It can also be considered an environmentally friendly alternative to improve the performance of artificial aggregates, which have high efficiency and low energy consumption.

MICP (microbial-induced carbonate precipitation) technology has been applied in the fields of civil engineering, such as self-healing concrete cracks (Ashraf 2016) and surface treatment of cement-based materials (Chunxiang et al. 2009). Inspired by the above application, MICP treatment has been investigated to modify artificial aggregates (Sonmez et al. 2021; Zhao et al. 2021). However, these studies mainly focused on the coating treatment of recycled concrete aggregate using MICP technology. Few studies have been aimed at artificial aggregates prepared by cold-bonding pelletization.

In this paper, the artificial aggregate was prepared to use a core–shell structure with waste concrete powder (WCP) as the shell component material and waste concrete aggregate particles (WCAP) as the core, making full use of WCP. The microorganisms added as shell components could enhance the performance of artificial aggregates through microbial mineralization. The MICP effect was realized in the shell structure. This method can solve the difficult utilization of WCP and effectively realize the dual purposes of artificial aggregate enhancement and carbon fixation. At the same time, it will also provide new preparation technical ideas.
for using waste concrete to alleviate the shortage of natural aggregate.

**Materials and methods**

**Raw materials**

Waste concrete powder and particle

WCP and WCAP were made by crushing concrete with a jaw crusher, grinding by a centrifugal mill, and sieving. The waste concrete was prepared to consist of cement, concrete admixture, water sand, and aggregate by the proportion of 1: 0.45: 0.40: 1.58: 3.04, which were cured more than one year with the compressive strength of 60.5 MPa. WCP was screened out by a 0.3 mm square hole sieve, and its particle-sized was less than 0.3 mm. WCAP was screened out with a particle size of 2.36 ~ 4.75 mm. The main component of WCP was cement stone. The main components of WCAP were sand and aggregate. The main phase compositions of WCP were quartz, calcium hydroxide and hydration products of cement such as calcium silicate hydrate; the XRD analysis is shown in Fig. 1. And the chemical compositions and particle size distribution of WCP and Portland cement (P·II 42.5) were measured using an energy-dispersive type X-ray fluorescence (XRF) spectrometer (ARL Perform’ X-4200, America) and laser particle sizer (Microtrac S3500, America). The results are shown in Table 1 and Fig. 2, respectively. The major chemical composition is mainly CaO and SiO₂ of waste concrete. The bulk and apparent densities of WCP were approximately 985 kg/m³ and 2631 kg/m³, respectively (according to Chinese standard GB/T 8074–2008).

**Microbial additive**

The microorganism used in this paper was *Bacillus mucilaginosus*, which could produce carbonic anhydrase. As a kind of metalloenzyme combining Zn²⁺ as its active centers, it can effectively accelerate the reaction of CO₂ and water to form H₂CO₃ through catalysis. Under ideal conditions, a single enzyme molecule can catalyze 10⁶ CO₂ molecules per second. In our previous work (Qian et al. 2015; Zheng et al. 2020), the *Bacillus mucilaginosus* with strong adaptability to the alkaline environment was obtained through laboratory microbial domestication and screening. This bacillus is a facultative gram-positive bacterium with a spindle structure, harmless to the human body and the environment. In this experiment, *Bacillus mucilaginosus* was inoculated in a sterilized medium solution. The medium was incubated at 25 °C in a shaking incubator at 170 rpm for 72 h and centrifuged to prepare a concentrated bacterial solution. Then a spray dryer was used to transform liquid microbial spores into powdery spores as the microbial additive (MA).

**Preparation and curing**

In general, cold-bonding disk pelletization refers to powder materials under the action of a liquid bridge and capillary tube to form micronuclei, which continuously rotate and grow in the powder layer, and finally form spherical particles of a specific

---

**Table 1** Chemical composition (wt.%) of WCP and cement

|       | SiO₂  | CaO  | Al₂O₃ | Fe₂O₃  | SO₃  | K₂O  | Na₂O  | MgO  | LOI  |
|-------|-------|------|-------|--------|------|------|-------|------|------|
| WCP   | 39.85 | 30.84| 6.34  | 2.03   | 1.93 | 1.76 | 1.13  | –    | 12.18|
| P·II 42.5 | 21.01 | 63.35| 4.23  | 3.15   | 2.31 | –    | –     | 2.19 | 3.16 |
size. However, this study improved the technology by preparing waste concrete sand particles as the core, which were more conducive to formatting and improving performance. The method of cold-bonding disk pelletization was slightly different from other methods reported in the literature, and the specific steps were as follows: The WCAP was placed in the granulator in advance with the wetting surface. Subsequently, setting the given rotation speed, the powdery materials were continuously fed to the granulator, and an appropriate amount of water was sprayed on the dry powders. While evenly adding powder and spraying water, the particles rotate continuously. The surface was gradually coated with powder, and the particles grew to form a certain core–shell structure artificial aggregate continually. The artificial aggregates can be divided into two groups based on the raw material of the shell: For the non-bacterial group (NB), the powder materials included pre-mixed WCP, cement; for the bacterial group (B), MA was added as another component of powder materials. The mixed proportion of core–shell raw materials is listed in Table 2.

After pelletization, all artificial aggregates were placed in the standard curing room for one day with the relative humidity was more than 90% RH. And the temperature was 20 ± 2 °C, and CO₂ concentration was about 0.03%. The initial curing time can ensure the germination and growth of microorganisms and obtain a small strength due to the hydration of cement. Then, two curing systems were designed to investigate the properties of artificial aggregates under different CO₂ concentrations at standard atmospheric pressure: (a) curing in the room environment condition (RC): the CO₂ volume fraction was about 0.03%, the temperature was 20 ± 2 °C, and relative humidity was 70 ± 5%; (b) cuing in a carbonation box: the CO₂ volume fraction was 95 ± 1%, the temperature was 20 ± 2 °C, and the relative humidity was 70 ± 5%, as shown in Table 2.

For example, Fig. 3 shows the preparation process and raw material of artificial aggregates as the B-C group. In the first step, waste concrete was crushed, ground and screened into waste concrete powder (WCP, diameter less than 0.3 mm) and waste concrete aggregate particles (WCAP, diameter within 2.36 ~ 4.75 mm); in the second step, mix cement, MA and WCP evenly in advance as powdery materials for standby; then WCAP was placed in the granulator in advance with the wetting surface. The powder was continuously fed to the granulator, with a set rotation speed at 35 r/min. While evenly adding powder and spraying water, the WCAP particles were gradually wrapped by powder to form the artificial aggregates with a core–shell structure of the sizes of 16 ± 2 mm (WCAP was used as the core, and the cement, MA, WCP were used as shell material and for cold-bonding disk pelletization); in the third step, after placement in the standard curing room for 1 day, the artificial aggregates were cured in the carbonated box of the curing system (b) for 24 h.

**Methods**

**Mechanical properties**

To study microorganisms' enhancement of artificial aggregate after different curing times, the apparent density, 24-h

| Table 2 | Mix proportion of artificial aggregates and curing conditions (g) |
|---------|---------------------------------------------------------------|
|         | WCP | Cement | WCAP | MA | Water | Curing condition |
| NB      | 1000 | 160    | 100  | 0  | 187    | (a)              |
| B       | 1000 | 160    | 100  | 10 | 187    | (a)              |
| NB–C    | 1000 | 160    | 100  | 0  | 187    | (b)              |
| B–C     | 1000 | 160    | 100  | 10 | 187    | (b)              |

**Fig. 3** The preparation process and raw material of artificial aggregate

 Springer
Preparation of artificial aggregate using waste concrete powder and CO₂ fixed by...

Water absorption and crushing strength were investigated to reflect the mechanical properties of artificial aggregates after curing for 24 h under curing system (a) or (b). The apparent density and 24-h water absorption were measured following Chinese standards GB/T14685-2011 and GB/B17431.2-2010. A California Bearing Ratio tester was used to determine the crushing strength value of the prepared artificial aggregates. Before the test, a Vernier caliper was used to test the diameter of aggregates in two vertical directions, and the average value was taken as the result. The aggregates were placed between two parallel plates, and the stiffener's loading rate was kept at 0.5 mm/min. The maximum pressure value was recorded when the aggregates were broken. The average value of 50 aggregates was taken for each group of data. The principles of testing are shown in Fig. 4. According to the literature (Arslan and Baykal 2006), the strength of aggregates can be measured by testing the crushing strength of a single aggregate by Eq. (1):

$$\sigma = \frac{2.8P}{\pi d^2}$$  

where $\sigma$ is the crushing strength (MPa), $P$ is the failure load (N), and $d$ is the diameter of artificial aggregates (mm).

Micro-morphology and structure of aggregate shell

Furthermore, to study the effect of microorganisms on the micro-morphology of artificial aggregate, the NB and NB-C groups after curing for 24 h (a or b curing system) were broken, and the shell parts were taken from the aggregates particles with the sizes of 16 ± 2 mm for testing. Tiny particles with a length size of 1 ~ 2 mm from the shell structure were dried for three days in a drying oven and subjected to a gold coating process to ensure good electrical conductivity. Then, the surface micro-morphologies of the samples were observed under scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM–EDX, Sirion, Netherlands). Mercury intrusion porosimetry (MIP, Auto pore IV 9510, Micromeritics USA) was adopted to test the porosity of artificial aggregate shells. Significantly, the samples were dried in a vacuum drying oven at 60 °C for seven days to meet the vacuum requirements before testing.

Microstructure of artificial aggregate

To observe the microstructure and composition of artificial aggregates with and without microorganisms, the NB, B, NB-C and B-C artificial aggregate specimens cured for 24 h under a curing system (a) or (b) were adopted. Aggregate particles with the sizes of 16 ± 2 mm were chosen and dried for three days. Then, the aggregates were tested by an X-ray tomography imaging system (Xradia 510, Zeiss, Germany). Defected analysis modules in VG Studio Max software (using the gray threshold algorithm to extract pore information) were used to analyze the interface area condition and the 3D pore structure of whole artificial aggregates.

CO₂ fixation of artificial aggregate

CO₂ weight loss and CO₂ fixation of artificial aggregates after curing for 24 h were quantified by thermogravimetric analysis (TG, STA 449 F3, Germany) as per Eqs. (2) and (3), respectively (Jiang and Ling Jiang et al. 2020a, b). Aggregate particles with the sizes of 16 ± 2 mm were chosen, and about 10 mg of the homogeneously ground sample was weighed and heated from 25 °C to 950 °C at a heating rate of 10 °C/min in a nitrogen atmosphere.

$$\Delta\text{CO}_2(\text{wt.} \%) = \left( W_{600} - W_{800} \right) \times 100\%$$  

$$\text{CO}_2 \text{ fixation (wt.} \%) = \frac{\Delta\text{CO}_2(\text{wt.} \%)\text{after} - \Delta\text{CO}_2(\text{wt.} \%)\text{before}}{100 - \Delta\text{CO}_2(\text{wt.} \%)\text{after}} \times 100$$  

where $W_{600}$ and $W_{800}$ represent the mass loss of the aggregate at 520 °C and 780 °C, respectively. The $\Delta\text{CO}_2$ (wt. %) after means the $\Delta\text{CO}_2$ (wt. %) value of artificial aggregate after curing stage, and the $\Delta\text{CO}_2$ (wt. %) before means the $\Delta\text{CO}_2$ (wt. %) value of fresh-formed artificial aggregate without the curing stage.

Compressive strength of concrete

The natural aggregate and artificial aggregate of the B-C group were used to prepare the concrete. The compressive strength of the samples with a size of 100 mm × 100 mm × 100 mm was based on the Chinese standard GB/T 50081–2019. Every group with three samples was calculated based on the following formula Eq. (4), and the result was accurate to 0.1 MPa.

$$f_{cc} = \frac{F}{A}$$  

Fig. 4 Schematic diagram of artificial aggregates strength measurement
where the $f_{cc}$ refers to the compressive strength of the concrete samples (MPa), the F refers to the failure load of the samples (N), the A refers to the loaded area of the samples (mm$^2$), and the $\alpha$ refers to dimension conversion factor (0.95).

**Results and discussion**

**Mechanical properties of artificial aggregate**

The apparent density, crushing strength and water absorption of artificial aggregates at different times are shown in Fig. 5. It can be observed that with the presence of microorganisms, the mechanical properties of artificial aggregates (B or B-C group) are better than those of NB or NB-C. As for the group of NB and B (under curing (b) condition), the mechanical properties slightly change with increasing of curing time. For the group of NB-C and B-C, the apparent density and crushing strength gradually increase and then remain stable.

As for the NB group, the mechanical properties were improved because during the hardening process of aggregates, cement hydration products (mainly calcium hydroxide (CH) and calcium silicate hydrates (C–S–H)) were gradually produced (Bye 1999), which improved the performance of the aggregates. Even with the addition of microorganisms, the improvement effect was not apparent because of the low CO$_2$ concentrate. When cured in the CO$_2$ concentration above 95% ((b) condition), the mechanical properties of artificial aggregates were greatly improved. For example, after curing for 24 h, the apparent density of the NB-C group was 2520 kg/m$^3$, the crushing index was 4.4 MPa, and the water absorption was 9.7%, while those of NB were 2400 kg/m$^3$, 2.3 MPa, and 11.5%, respectively. The enhancement was consistent with other studies, because more carbon dioxide could react with calcium hydroxide, calcium silicate hydrates and other calcium substances in aggregates leading to more precipitation of calcium carbonate (Pu et al. 2021). The produced calcium carbonates were stable and high-hardness, with a higher elastic modulus and solid-phase volume.
than hydration products (Fang and Chang 2015). The porosity decreases, which contributes to the improvement of artificial aggregate properties. More carbonated products also compacted the microstructure of aggregates. Furthermore, the microorganisms show a huge promotion effect at high CO$_2$ concentrations. Compared with the NB-C group, the apparent density of aggregates in B-C was 2620 kg/m$^3$ increased by 4%, crushing strength was 9.1 MPa increased by 106.8%, and water absorption was 4.8% decreased by 50.5% after cured 24 h. When the CO$_2$ curing time exceeds 24 h, the performance of B-C artificial aggregates remains stable with the extension of time. The microorganisms accelerate the carbonization reaction process of minerals, making the shell structure of aggregates denser. The reaction degree may reach a certain threshold, and enough products fill the pores of the aggregates. It changed the microstructure of artificial aggregates, which made the strengthening effect more conspicuous.

3D pore structure of artificial aggregate

X-CT is reviewed as the most advanced nondestructive test methodology for evaluating cement-based materials at the microstructural level (Prasad 2020). The pore' distribution and connectivity concerning their location can be visualized by reconstructing three-/two-dimensional sections. The pore structure of 4 types of artificial aggregates was investigated by X-ray computed tomography to explore the effect of mineralization by microorganisms. Figure 6 shows the X-CT 3D reconstruction of artificial aggregates. From blue to red, the defect volume gradually increased. The whole structure showed many properties for the NB group, with a maximum detection volume of 1.84mm$^3$. After curing with 95% vol CO$_2$ (curing (b) condition), it is worth noting that the NB-C porosity showed a gradient porosity structure. From the edge to the center of the pellet, the colored area gradually became denser, which indicated that the porosity gradually increased. Because the CO$_2$ concentration at the edge was higher than that inside, with the increasing depth, the concentration of carbon dioxide gradually decreased.

Furthermore, the precipitation of carbonate products generated at the edge hindered carbon dioxide transmission, resulting in a slower rate and lower degree of internal carbonation reaction. However, the presence of microorganisms in artificial aggregates increased the CO$_2$ concentration inside. More CO$_2$ penetrated deep into the shell structure through the pores in the early stage, accelerated by microbial catalysis. Carbon dioxide dissolved in the pore water and produced carbonic acid. For the B group, the blue-colored area was sparse than that of NB group because the
microorganism uniformly distributed in the whole aggregate in advance accelerated the capture of CO$_2$. In high CO$_2$ concentration, the microorganisms increased the carbonation depth. For the B-C group, a gradient structure in a shell of artificial aggregate was not detected, and deeper distances from the surface showed fewer pores. This means microorganisms could make the carbonation reaction more complete and the aggregate structure more uniform.

In X-CT, quantitative analysis of the void content and size of pores was determined by obtaining the two-dimensional images as the result of the various intensities correlated to the material density (Wei et al. 2011). Figure 7 shows the 2D of artificial aggregates, and the black spots were identified as the voids in the cementitious matrix. It can be found that the B-C was denser than the other groups, also reflecting the improvement of microorganisms on the microstructure of artificial aggregates. Microorganisms evenly distributed in the shell of artificial aggregate mineralized and deposited calcium carbonate to fill and bond fine powder, making the aggregate structure more uniform and denser.

**Porosity of artificial aggregate shell**

In order to analyze the improvement of microorganisms on the porosity of artificial aggregate, specimens of shell structure were tested by MIP. The porosity content was obtained as the total volume of the mercury intruded into the aggregates pellet. The total volume of the sample was used to accurately analyze the micropores of aggregates ranging from 3 to 5000 nm. The integral curves and differential curves of the pore size distribution of artificial aggregates with and without bacteria under different curing conditions are shown in Fig. 8. Furthermore, the calculation of the results data is listed in Table 3. It can be found that the mechanical porosities of NB and B group samples are similar, which are 30.5% and 29.8%, respectively. The enhancement of microbial mineralization is not significant under the condition of low concentration of CO$_2$. But the average pore diameter and most probable aperture decreased because mineralization products originated by microorganisms and WCP/cement filled the pore. However, after CO$_2$ curing with a concentration above 95%, it is worth mentioning that the porosity of the B-C group had the most significant decline, down to 13.9%. Meanwhile, the average pore diameter and most probable diameter also decrease. That can be concluded that the microorganism can significantly modify the micro-pore structure of artificial aggregates. The greatest aperture of the B group was only 9.0 nm, which was classified as a harmless pore (Wu 1979; Wang 2022), which was incredibly beneficial for aggregates to resist the invasion of foreign harmful substances.
Preparation of artificial aggregate using waste concrete powder and CO₂ fixed by...

Micro-morphology of carbonated and mineralized products in shell

The artificial aggregates after CO₂ curing showed better mechanical properties and denser microstructures. Therefore, the shell parts of artificial aggregates without and with microorganisms (NB-C and B-C groups) were adopted to study the effect of microorganisms by SEM. Figures 9 and 10 show the SEM images of the micro-morphology of carbonated and mineralized products in the shell, respectively. Products related to the carbonated and microbial mineralized reactions are also visible in the waste concrete powder. They are crystal depositions, mainly in the form of rhombohedral-/cubic-shaped and granular-texture boning together. From the EDS analysis (Fig. 9d), the main elements NB-C are C, Ca and O, which proves that carbonated products are calcium carbonate. The other products may be calcium silicate hydrate because of the detection of tiny Si elements. The crystal morphology of products deposited of NB-C is mainly ellipsoidal and massive and relatively sparse. Figure 10d also proves that the mineralized products are calcium carbonated. In contrast, the crystals on the surface shown in Fig. 10c were mainly irregular with crisscross growth and denser, which was just the favorable evidence of microbial promoting calcium carbonate deposition. The mineralized products are cohesively connected and well bonded to the substrate. More calcium carbonates filled the micromesopores and air voids, and the dense precipitate acted as a film to decrease water absorption.

Analysis of the mechanical properties of artificial aggregate concrete

To prepare concrete, the artificial aggregate was replaced by 10%, 30%, 50%, 70% and 100% of natural aggregate. The apparent densities of natural and artificial aggregate are 2730 kg/m³ and 2600 kg/m³. The water absorptions of that are 0.7% and 3.0%. There is a particular gap in the apparent density and water absorption of both aggregates. The specific mix proportion of concrete was calculated based on the volume method. The proportions of various materials with 1 m³ are shown in Table 4.

Figure 11 shows the strength of natural aggregate concrete (NC) and artificial aggregate concrete (AC) with different replacements under curing for seven days and 28 days. The compressive strength of NC or AC shows a similar strength-development trend. The compressive strength increases with age, and curing for 7 days can reach above 70% of the 28-day strength. When the replacement rate of artificial aggregate is less than 30%, the compressive strength of AC has little change and slightly increases compared with that of NC. However, when the replacement rate reaches 50%, the compressive strength of concrete reaches that of the NC concrete. When substitution quantity exceeds more than 50%, the compressive strengths decrease sharply with the maximum reduction of 22.4% at the AC-100 group. The low replacement rate has little effect on the concrete strength because the water absorption rate of aggregate is higher than that of natural aggregate. Increasing
the replacement material within a specific range reduces the effective water–cement ratio of concrete, which reduces the porosity. In the long-term curing, the moisture absorbed by artificial aggregate will be released to the matrix gradually with the hydration process of concrete cement, which is conducive to the development of strength. In addition, part of the cement slurry is easily attached to the surface micropores of coarse aggregate, making the mosaic structure between aggregate and matrix. However, there is no doubt that the compressive strength of concrete mainly depends on the coarse aggregate. The high replacement rate inevitably leads to a significant reduction in the strength of concrete due to the low strength of artificial aggregate. Therefore, the artificial aggregate prepared in this study can replace the natural aggregate in a particular proportion to prepare concrete, and the full content should be controlled within 50%.

**CO₂ fixation of artificial aggregate**

The thermogravimetric (TG) and differential thermogravimetric (DTG) curves of artificial aggregates are plotted in Fig. 12. The peaks observed in the DTG curve (600 ~ 800 °C) mainly denote the decomposition of calcium carbonates (Shah et al. 2018). According to Eqs. (2) and (3), CO₂ change (ΔCO₂) and fixation of artificial aggregate are listed in Table 5. It should be pointed out that CO₂ fixation in the table refers to the result after deducting the CO₂ content of WCP itself (2.34 wt. % detected). The CO₂ content in WCP was derived from the absorption of CO₂ in the atmosphere by the waste concrete matrix. The CO₂ fixation of NB and B groups was similar and less than 1 wt. % due to the low CO₂ concentration, indicating that the CO₂ fixation ability of is weak. However, under 95% CO₂ concentration, the CO₂ fixation increases considerably. With the presence of microorganisms, the CO₂ fixation of B-C increased from 7.39 wt. % to 16.00 wt. %, illustrating that microorganisms further enhanced the carbon fixation capacity of artificial aggregates under high CO₂ concentration. The artificial aggregates mixed with microorganisms show great carbon fixation potential. The per-ton artificial aggregate product can fix 160 kg CO₂, which has remarkable economic and social significance for the construction industry to reduce the carbon footprint.
The production cost of artificial aggregate mainly includes raw material and process costs. The raw materials of artificial aggregate come from waste concretes, mainly obtained by crushing and classifying demolished buildings. Moreover, considering CO₂ curing, the artificial aggregate processing site is suitable for building near high carbon emission plants (such as cement and iron steel factories). Industrial CO₂ is obtained from the carbon collection and enrichment equipment and then transported to the curing device by pipeline. Carbon fixation by artificial aggregate helps factories realize carbon emission reduction benefits. Many factories have carried out the collection and purification of low-purity CO₂ from flue gas. Therefore, the electric fee of the equipment should not be calculated. In our economic evaluation, when calculating the cost of artificial aggregate, the cost of CO₂ raw material is not considered, but only the cost of pipelining CO₂ process. The raw materials of artificial aggregate
include WCP, WCAP, cement, MA and water. WCP and WCAP are by-products produced during the crushing and screening of waste concrete, which are difficult to use effectively (treated by stacking and landfill). Therefore, these raw materials costs are negligible. The production process of artificial aggregate includes pelletization and CO$_2$ curing processes, calculated according to power consumption. The cost of one ton of artificial aggregate was calculated according to the proportion of artificial aggregate, and the estimation results are shown in Table 6. The calculation basis is as follows: Portland cement was CNY400/ton; the concentrated microorganism solution was obtained by strain culture, and the cost was estimated to be CNY30/kg. After water dilution, batching and processing, the cost of MA powder was calculated as CNY5/kg; industrial tap water was CNY4/ton; the average capacity of industrial pelletization was 2 tons/h, and the motor power was 7 kW/h; the cost of CO$_2$ curing mainly considered the fees of collection and pipeline transportation. The CO$_2$ transportation capacity of the fan is 1 ton/h, and the power is 5.5 kW/h; the average market price of industrial power was CNY0.64/kWh. The results showed that the production cost of artificial aggregate was estimated to be about CNY74.2/ton. The cost will be reduced with the large-scale production of artificial aggregates, such as reducing microorganisms, cement dosage and process cost.

Natural aggregate mainly comes from rivers, lakes and mines, and the cost mainly comes from mining and transportation costs. The local market price of natural aggregate is usually CNY100~150/ton. According to the relevant tax laws of China, the production and sales of natural sand and gravel resource need to pay 6% or 13% value-added tax (VAT) on goods and 1%~5% resource tax. In addition, a certain additional tax is paid, usually 11% of the VAT amount. However, as the artificial aggregate uses waste concrete, it can get the support and preferential policies of the government. This enjoys preferential tax policy advantages, such as exemption from resource tax, and VAT reduction. Artificial aggregate has a cost and commercial sales advantages. Only in terms of tax, artificial aggregate products have a price advantage of about 10%. As a result, the cost and profit are considerable.
Environmental analysis

Previous results emphasize that the performance of artificial aggregates is excellent and stable, which can replace natural aggregates to prepare concrete. This can benefit from construction and industrial activities; at the same time, CO₂ emissions can be reduced by carbon sequestration of artificial aggregate. The CO₂ fixation content per ton of artificial aggregate with added microorganisms can reach 154.3 kg, calculated based on the CO₂ fixation of shell parts 16.0%. China’s annual output of waste concrete will be about 1.5 billion tons in 2020 (Cao et al. 2021). The WCP has reached 10% ~ 20%, above 150 million tons in 2020. If they are used to prepare artificial aggregate, about 200 million tons of artificial aggregate can be produced, and 31 million tons of CO₂ can be fixed. According to the carbon emission report of “carbon dioxide emission report of China building materials industry (2020),” the CO₂ emission of building materials industry in 2020 is 1.48 billion tons. The rate of CO₂ reduction by produced artificial aggregate can reach 2.09%, which is better for the construction industrial carbon reduction. In the process of producing artificial aggregate, it can not only use the waste concrete but also fix CO₂ from the cement production process. This method can effectively realize the utilization of waste concrete resources, reduce the exploitation of natural aggregates, and reduce concrete carbon emissions in the whole life cycle, which provides significant economic and environmental benefits.

Conclusion

In this paper, the artificial aggregate with core–shell structure was prepared by pelletization of waste concrete materials, and CO₂ was fixed by microorganisms in the shell to strengthen the aggregate. The influences of microorganisms on the basic properties and microstructure of artificial aggregate were studied. The mechanism of the improving mechanical properties and microstructures of artificial aggregates by microorganisms was analyzed. The compressive strength of concrete prepared by partial substitution of artificial aggregate was studied. Finally, the economic and ecological benefits of artificial aggregate were analyzed. The specific conclusions are as follows:

1. Artificial aggregates (NB-C group) have excellent mechanical properties: the sample’s apparent density, crushing strength and water absorption of the samples after curing (b) for 24 h were 2620 kg/m³, 9.1 MPa and 4.8%, respectively.

2. The presence of microorganisms promotes the carbonization of WCP and calcium silicate hydrate in artificial
aggregates. More mineralized products compact the structure and reduce the porosity. The calcite crystals with more compact and complete crystal growth were formed in the shell part due to the addition of microorganisms. The porosity of artificial aggregate decreased from 20.98% to 13.88%, and the CO₂ fixation decreased from 7.39 wt. % to 16.00 wt. %.

(3) The artificial aggregates can replace the natural aggregates in a certain proportion to prepare concrete. When the replacement rate is less than 50%, the mechanical properties of concrete have no adverse change.

(4) The artificial aggregate prepared by microbial CO₂ fixation can make full use of waste concrete powder and reduce the exploitation of natural aggregate. It can also be used for industrial CO₂ fixation and reduce the carbon footprint of the life cycle concrete. Microbial mineralization technology has mild reaction conditions and a friendly environment. Artificial aggregate products have good market prospects and obvious economic, social and environmental benefits.

Acknowledgements The authors appreciate the financial support from the National Nature Science Foundation of China (Grant No. 51738003).

Author contributions In this paper, XZ assisted in the preparation samples and test, analyzed data and revised and modified the manuscript. CQ contributed to the methodology and funding acquisition, conceived the central idea and directed the experiments of this study, and was involved in writing—review. DX contributed to the investigation, conceptualization, methodology, experiments, data curation, analysis of data and writing—original draft.

Declarations

Conflict of interest The authors declared that they have no conflicts of interest in this work.

References

Ahmed Shaikh FU, Nath P, Hosan A, John M, Biswas WK (2019) Sustainability assessment of recycled aggregates concrete mixes containing industrial byproducts. Mater Today Sustain 5:100013. https://doi.org/10.1016/j.mtsustain.2019.100013

Arslan H, Baykal G (2006) Analyzing the crushing of granular materials by sound analysis technique. J Test Eval 34(6):464–470. https://doi.org/10.1520/JT010032

Ashraf W (2016) Carbonation of cement-based materials: challenges and opportunities. Const Build Mater 120(1):558–570. https://doi.org/10.1016/j.conbuildmat.2016.05.080

Bye GC (1999) Portland cement. In: 8. The nature of hardened cement paste, 2nd edn. pp. 131–162. https://www.icevirtuallibrary.com/doi/abs/10.1680/pccpap.27664.0008

Cao Y, Wang S, Wang Y, Zhu Z, Wang S (2021) Present situation and future development trend of comprehensive utilization of construction waste in China. China Urban Economy Doi: https://doi.org/10.16291/j.cnki.gzjc.2021.09.043

China, M.o.N.R.o.t.P.s.R.o (2019) Fully promote the high-quality development of mechanism sand and gravel industry. http://www.mnr.gov.cn/ftc/201911/20191129_2485004.html

Chunxiang Q, Jianyun W, Ruixing W, Liang C (2009) Corrosion protection of cement-based building materials by surface deposition of CaCO₃ by Bacillus pasteurii. Mater Sci Eng C 29(4):1273–1280. https://doi.org/10.1016/j.msec.2008.10.025

Fang Y, Chang J (2015) Microstructure changes of waste hydrated cement paste induced by accelerated carbonation. Constr Build Mater 76:360–365. https://doi.org/10.1016/j.conbuildmat.2014.12.017

Fca B, Fma B, Rca B (2015) Recycling of MSWI fly ash by means of cementitious double step cold bonding pelletization: technological assessment for the production of lightweight artificial aggregates. J Hazard Mater 299:181–191. https://doi.org/10.1016/j.jhazmat.2015.06.018

Jiang Y, Ling TC (2020) Production of artificial aggregates from steel-making slag: influences of accelerated carbonation during granulation and/or post-curing. J CO2 Util 36:135–144. https://doi.org/10.1016/j.jcou.2019.11.009

Jiang Y, Ling TC, Shi M (2020) Strength enhancement of artificial aggregate prepared with waste concrete powder and its impact on concrete properties. J Clean Prod 257:120515. https://doi.org/10.1016/j.jclepro.2020.120515

Job T, Harilal B (2019) Sustainability evaluation of cold bonded aggregates made from waste materials. J Clean Prod. https://doi.org/10.1016/j.jclepro.2019.117782

Kisku N, Joshi H, Ansari M, Panda SK, Nayak S, Dutta SC (2017) A critical review and assessment for usage of recycled aggregate as sustainable construction material. Const Build Mater 131:721–740. https://doi.org/10.1016/j.conbuildmat.2016.11.029

Kou SC, Poon CSM, Etxeberria (2014) Residue strength, water absorption and pore size distributions of recycled aggregate concrete after exposure to elevated temperatures. Cem Concr Com 53:73–82. https://doi.org/10.1016/j.cemconcomp.2014.06.001

Li S, Wu Q, Zheng Y, Sun Q (2021) Study on the spatial association and influencing factors of carbon emissions from the Chinese construction industry. Sustainability 13(4):1728–1728. https://doi.org/10.3390/SU13041728

Mistri A, Dhami N, Bhattacharyya SK, Barai SV, Mukherjee A, Biswas WK (2021) Environmental implications of the use of bio-cement treated recycled aggregate in concrete. Res Conserv Recy 167:105436. https://doi.org/10.1016/j.resconrec.2021.105436

Narathna C, Chaipanich A (2018) Phase characterizations, physical properties and strength of environment-friendly cold-bonded fly ash lightweight aggregates. J Clean Prod 171(2):1094–1100. https://doi.org/10.1016/j.jclepro.2017.09.259

Pan G, Zhan M, Fu M, Wang Y, Lu X (2017) Effect of CO2 curing on demolition recycled fine aggregates enhanced by calcium hydroxide pre-soaking. Constr Build Mater 154:810–818. https://doi.org/10.1016/j.conbuildmat.2017.07.079

Pang B, Zhou Z, Xu H (2015) Utilization of carbonated and granulated steel slag aggregate in concrete. Constr Build Mater 84(1):454–467. https://doi.org/10.1016/j.conbuildmat.2015.03.008

Prasad C (2020) High-efficiency techniques and micro-structural parameters to evaluate concrete self-healing using X-ray tomography and mercury intrusion porosimetry: a review. Constr Build Mater 252(2020):119030. https://doi.org/10.1016/j.conbuildmat.2020.119030

Pu Y, Li L, Wang Q, Shi X, Luan C, Zhang G, Fu L, El-Fatah Abo-mohra A (2021) Accelerated carbonation technology for enhanced treatment of recycled concrete aggregates: a state-of-the-art
Preparation of artificial aggregate using waste concrete powder and CO₂ fixed by…

Qian C, Chen H, Ren L, Luo M (2015) Self-healing of early age cracks in cement-based materials by mineralization of carbonic anhydrase microorganism. Front Microbiol. https://doi.org/10.3389/fmicb.2015.01225

Shah V, Scrivener K, Bhattacharjee B, Bishnoi S (2018) Changes in microstructure characteristics of cement paste on carbonation. Cem Concr Res 109:184–197. https://doi.org/10.1016/j.cemconres.2018.04.016

Shi C, Li Y, Zhang J, Li W, Chong L, Xie Z (2015) Performance enhancement of recycled concrete aggregate—a review. J Clean Prod. https://doi.org/10.1016/j.jclepro.2015.08.057

Sonmez M, Ilcan H, Dundar B, Yildirim G, Ersan YC, Sahmaran M (2021) The effect of chemical—versus microbial-induced calcium carbonate mineralization on the enhancement of fine recycled concrete aggregate: a comparative study. J Build Eng 44:103316. https://doi.org/10.1016/j.jobe.2021.103316

Tang P, Brouwers H (2017) Integral recycling of municipal solid waste incineration (MSWI) bottom ash fines (0–2 mm) and industrial powder wastes by cold-bonding pelletization. Waste Manage 62:125–138. https://doi.org/10.1016/j.wasman.2017.02.028

Wang C, Xiao J, Zhang G, Li L (2016) Interfacial properties of modeled recycled aggregate concrete modified by carbonation. Constr Build Mater 105(15):307–320. https://doi.org/10.1016/j.conbuildmat.2015.12.077

Wang Y, Zhang S, Niu D, Fu Q (2022) Quantitative evaluation of the characteristics of air voids and their relationship with the permeability and salt freeze–thaw resistance of hybrid steel-polypropylene fiber–reinforced concrete composites. Cem Concr Compos 125:104292. https://doi.org/10.1016/j.cemconcomp.2021.104292

Wei Q, Leblon B, Rocque AL (2011) On the use of X-ray computed tomography for determining wood properties: a review. Can J Forest Res 227(41):2120–2140. https://doi.org/10.1139/x11-111

Wu Z (1979) An approach to the recent trends of concrete science and technology. J Chin Ceram Soci 7(3):82–90

Xiao J, Ma Z, Sui T, Akbarnezhad A, Duan Z (2018) Mechanical properties of concrete mixed with recycled powder produced from construction and demolition waste. J Clean Prod 188:720–731. https://doi.org/10.1016/j.jclepro.2018.03.277

Zhan B, Poon CS, Liu Q, Kou S, Shi C (2014) Experimental study on CO₂ curing for enhancement of recycled aggregate properties. Constr Build Mater 67:3–7. https://doi.org/10.1016/j.conbuildmat.2013.09.008

Zhao Y, Peng L, Feng Z, Lu Z (2021) Optimization of microbial induced carbonate precipitation treatment process to improve recycled fine aggregate. Clean Mater 1:100003. https://doi.org/10.1016/j.clema.2021.100003

Zheng TW, Su YL, Zhang X, Zhou HY, Qian CX (2020) Effect and Mechanism of Encapsulation-Based Spores on Self-Healing Concrete at Different Curing Ages. ACS Appl Mater Interf 12(47):52415–52432. https://doi.org/10.1021/acsami.0c016343

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.