Improving the quality of various types of recycled aggregates by biodeposition

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Abstract

Demand for construction materials has been rising in recent decades in many countries around the world, placing a heavy burden on the environment in terms of both the natural resources consumed and the enormous flow of waste generated. In order to obtain a more sustainable construction, it is often suggested to reintroduce the industry's own waste as input for the manufacture of new materials. In this study, the use of construction and demolition waste of concrete or mixed concrete/ceramic nature is investigated as a replacement of natural aggregates in concrete. The greater affinity of recycled aggregates for water directly affects the workability and/or the concrete strength and durability. One possible solution to reduce the aggregates water absorption is to apply a biogenic treatment with calcium carbonate-precipitating bacteria that consolidate the aggregate surface or the adhering mortar.

Experimental results show that the biodeposition treatment reduced the recycled aggregate water absorption by generating precipitation in the pores and an impermeable outer layer, most effectively on the roughest particle surfaces. The largest decrease happened in the aggregates with the highest porosity. The biogenic layer had a good cohesion with the aggregates. The results of sonication indicated that the most effective treatment was on recycled concrete aggregates (RCA) instead of mixed aggregates (MA). Therefore, the treated RCA was used to make concrete for further investigation. The concrete made with bio-treated RCA had a denser structure, a decreased water absorption (around 1%) and an improved compressive strength (25%).

Introduction

Construction and demolition waste (C&DW) is the collective term for waste materials generated by construction, renovation and demolition of buildings. The waste originating form construction and demolition of roads and pavements is also included. From the viewpoint of sustainable resource management, concrete and demolition waste is sorted and reduced to applicable sizes. From there on the material can be given a meaningful function. One of the most often used applications is as an aggregate. The granular material obtained from the crushing of construction and demolition waste is called a recycled aggregate (RA). Due to its origin, it is often referred to as a secondary raw material.

Recycled aggregates can be used to replace a primary raw material (sand, limestone, etc.). In theory this can be done for fine and coarse materials. In practice however, when they are used in concrete, it is mostly limited to only the coarse fractions. For fine fractions the aggregate has to go through a much more intensive breaking process. This is not only more expensive, it also seems to have a negative influence on the properties of the aggregates. Finer recycled particles seem to have a higher porosity than larger ones. The porosity may undesirably affect the properties of the concrete. The use of fine recycled aggregates is therefore not recommended [1, 2]. Mostly, the coarse fraction is investigated for secondary use. However, around 95% is used in low quality application (foundations of roads, rip-rap, etc.). This is caused by the questionable performance of these aggregates. Increasing active research is being conducted for the application of this material in higher grade application. The focus is first to improve the properties of the recycled aggregates, more specific, to decrease the water absorption capacity.

One very promising method is by bacterially induced precipitation of calcium carbonate in and around the aggregates. This is known as the so-called 'biodeposition method'. Under suitable conditions, most bacteria can mediate $CaCO_3$ precipitation [3]. This biogenic material is regarded as an environmentally friendly and economical material for engineering applications. Furthermore, it has a good compatibility with the inorganic materials matrix. So far, the biodeposition treatment has been investigated mostly for surface protection and consolidation of limestone and concrete [4-7]. The calcium carbonate could fill the pores and/or form a continuous water-proof coating to hinder the penetration of corrosive substances. The dense surface layer and decreased permeability thus result in an improved durability. Inspired by the successful application on surface protection, biodeposition treatment has recently been investigated for improving the properties of recycled aggregates [8,9]. In this study, a carbonate precipitating bacterium, *Bacillus sphaericus*, was used to induce the formation of calcium carbonate. This strain can decompose urea ($CO(NH_2)_2$) into ammonium (NH_4^+) and carbonate (CO_3^{2-}). The latter subsequently promotes the bacterial deposition of $CaCO_3$ in a calcium rich environment. Through this process, biogenic $CaCO_3$ can be formed. The aim of this study is to use this bio-$CaCO_3$ to decrease the water absorption by filling the pores and/or forming an extra dense layer on the surface of recycled aggregates, and hence improve the quality of the recycled aggregates which can then be used for concrete making.

Materials and Methods

Recycled aggregates. Three types of recycled aggregates were used in this study. One crushed concrete aggregate (RCA) from the processing site of AC Materials nv (Belgium) and two mixed aggregates (MA-TEC and MA-ANT) from construction and demolition waste plants (Tecnología y Reciclado S.L., Madrid, and Antwerp Recycling Company, Belgium). The apparent density (ρ_a), the oven-dried density (ρ_{rd}) and the saturated surface-dried density (ρ_{ssd}) of each kind of aggregates were determined according to the EN 12620+A1(2009) standard [10], and are shown in Table 1.

Table 1 Particle densities of different aggregates

	ρ_a (kg/m^3)	ρ_{rd} (kg/m^3)	ρ_{ssd} (kg/m^3)
RCA	2660	2350	2460
MA-TEC	2530	2080	2260
MA-ANT	2510	2060	2240

Bacterial strain and cultivation. *Bacillus sphaericus* LMG 22257 (Belgian Coordinated Collection of Microorganisms, Ghent) was used in this research. Selection of this bacterial strain was based on our earlier research [11,12], which show that this strain has a high urease activity (40 mM urea hydrolyzed. OD^{-1} h^{-1}) and a high calcium carbonate production. The bacteria were grown in a sterile medium consisting of yeast extract (20g/L) and urea (20g/L) for 24 hours on a shaker (120 rpm, 28°C). The grown culture was then used for the following bio-deposition treatment. The concentration of the bacteria in the culture was around 10^8 cells/mL.

Bio-deposition treatment. Each type of dry aggregates (each portion around 200g, n=3) was first immersed in the bacterial grown culture for 24 hours. After that, the aggregates were taken out from the bacterial culture and were submerged into a deposition medium which consisted of 0.5 M urea and 0.5 M Ca-nitrate for another 3 days.

Water absorption. The water absorption of the aggregates before treatment and after treatment was determined according to EN 1097-6:2014 [13]. For all aggregates, the pycnometer method was used. The aggregates were submerged in the pycnometer for 24 hours. The entrapped air was removed by gently shaking the pycnometer. After 24 hours, the pycnometers were weighed. Then the aggregates were taken out and the surface water was removed with dry cloths. Hence, the saturated surface dry mass was known. At last, the aggregates were dried in an oven at 75°C until constant mass was achieved (less than 1% within 24 h). According to the standard EN 1097-6 (2014) [13], it was suggested to maintain the temperature at 75°C in order to avoid removal of interlayer water from hydrated calcium silicates, which occurs at 78-90°C and which could have distorted the results [14]

$$\rho_{ssd} = \rho_w \frac{M_1}{M_1 - (M_2 - M_3)} \tag{1}$$

$$WA_{24} = \frac{M_1 - M_4}{M_4} \times 100\% \tag{2}$$

Where:

M_1 = mass of the saturated and surface-dried aggregate in air (g)
M_2 = mass of the pycnometer containing the sample of saturated aggregate and water (g)
M_3 = mass of the pycnometer filled only with water (g)
M_4 = mass of the oven-dried aggregates (g)
ρ_w = density of water at the test temperature (kg/m^3)
WA_{24} = Water absorption after 24 hours (m%)

The test was performed in a controlled environment. The air temperature was around 20°C. The water temperature was also checked on a regular basis to ensure it was constant during the test duration.

Weight increase. After the bio-treatment, the aggregates were taken out from the deposition medium and were gently rinsed with tap water to remove the loose precipitates from the surface of the aggregates. Subsequently, they were dried in an oven at 75°C until constant mass was achieved (less than 1% within 24 h). The weight difference before and after the treatment can therefore be obtained.

Resistance to ultrasonic pulse. The dried treated recycled aggregates were then subjected to an ultrasonic pulse test. Samples (around 70 g) of each recycled aggregate were placed on a 1 mm sieve and were submerged in the demineralized water container of the ultrasonic bath (Haver USC 200-76). The samples were sonicated for 5 min at a frequency of 46 kHz with ultrasonic waves. After that, samples were collected and dried in the oven at 75°C until constant mass was achieved (less than 1% within 24 h). The weight loss of the treated aggregates after ultrasonic pulse was used as a parameter to evaluate the cohesion between the precipitation and the aggregate surface. As a control, the untreated aggregates were also subjected to the same ultrasonic pulse test.

Characterization of the bio-precipitation. The morphology of the precipitates was investigated by use of a scanning electron microscope (SEM, JEOL JSM-7600F). The aggregates were first coated with carbon and were subjected to SEM analysis. Energy dispersive spectroscopy (EDS) spectra were used to verify the chemical composition of the precipitates on the aggregate surface.

Concrete made with treated and untreated recycled aggregates. Among the three treated aggregates, RCA was selected for concrete making to further investigate the effect of bio-treated aggregates on concrete properties. Three concrete mixtures were made: mixture with normal limestone aggregates (NAC), with untreated (RCAC/U) and treated RCA (RCAC/T). The composition is shown in Table 2. The aim was to create a concrete which met the criteria for exposure class EE3. The minimal cement content for this class is 320 kg/m^3 and maximum w/c ratio is 0.5.

Table 2 The composition of concrete mixtures

	NAC	RCAC/U	RCAC/T
CEM III/A 42.5LA [kg/m^3]	320	320	320
Water [kg/m^3]	165	200	200
Sand 0/4 [kg/m^3]	722	699	699
Limestone 4/6 [kg/m^3]	456	448	448
Limestone 6/20 [kg/m^3]	786	-	-
RCA 6/20 [kg/m^3]	-	805	805

Each concrete mixture was produced in the same way. The sand and limestone 4/6 were air-dried by spreading them on the floor. The coarse 6/20 aggregates were pre-wetted before mixing. The water absorption of the coarse aggregates was accounted for in the mixing water. Pre-wetting was done by submerging the coarse aggregates in 90% of the mixing water. This amount of water was added to the aggregates which were then stored in a closed container for 24 hours. Sediments of fines were flushed from the container by using the remaining 10% of the mixing water. The further concrete mixing and casting were performed according to NBN EN 12390-2 (2009). For each concrete mixture, four 150×150×150 mm^3 cubes were made to determine the compressive strength and water absorption. The specimens were then stored in a moist room with a temperature of 20±2 °C and a relative humidity higher than 95% for 14 days.

To determine the water absorption of the concrete, the four cubes were submerged in water for 7 days after the 14 days storage in the moisture room. Subsequently, the saturated mass of the concrete was determined according to NBN B 15-215. The specimens were then dried in the oven at 105 °C for another 7 days or until constant weight was achieved, to determine the dry mass of the concrete. The water absorption by submersion can be calculated with the following formula:

$$A = \frac{m_1 - m_2}{m_2} \times 100\% \qquad (3)$$

Where:

A = water absorption of concrete by submersion (m%)
m_1 = saturated weight of concrete (g)
m_2 = oven-dried weight of concrete (g)

The compressive strength of concrete was determined on the oven-dried specimens after the water absorption test according to NBN EN 12390-3 (2009) [15].

Results

Weight increase. All bio-deposition treatments resulted in weight increase of the recycled aggregates due to the biodeposits, as shown in Fig.1 (the circles). Much more precipitates (4~4.5%) were formed on the mixed recycled aggregates, both MA-TEC and MA-ANT, than on the recycled concrete aggregates (around 0.7%).

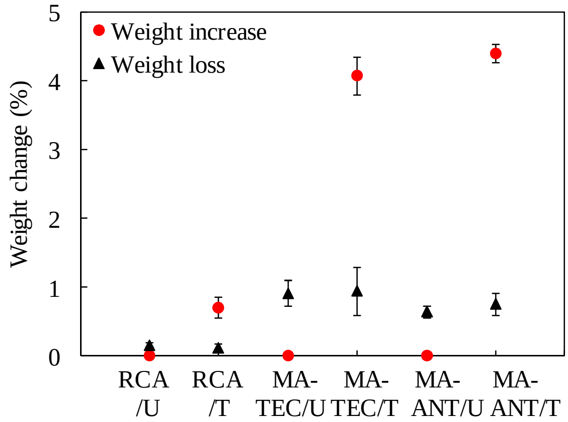

Fig.1 Weight changes after bio-treatment and sonication test

Resistance to ultrasonic pulse. Weight loss occurred in all aggregates, both treated and untreated ones, during the sonication test. As shown in Fig.1 (the triangles), RCA has the least amount of weight loss, around 0.1%. Much more weight loss (about 1%) happened in the mixed aggregates, MA-TEC and MA-ANT. The difference between treated and untreated aggregates was not significant.

Decrease of water absorption. The water absorption of the aggregates was decreased after bio-treatment. As shown in Fig.2, the WA_{24} of RCA was in the range of 5~5.5 % before treatment, and was decreased to 4.5~5% after treatment. More pronounced decrease was observed in MA-TEC, of which the WA_{24} was decreased from 6.3~6.9 % to 5.6~5.9 %. However, the water absorption of MA-ANT was not decreased so much. Almost no decrease occurred in two of the three replicates. For another replicate, the WA_{24} was decreased from 7.5% to 7.1%.

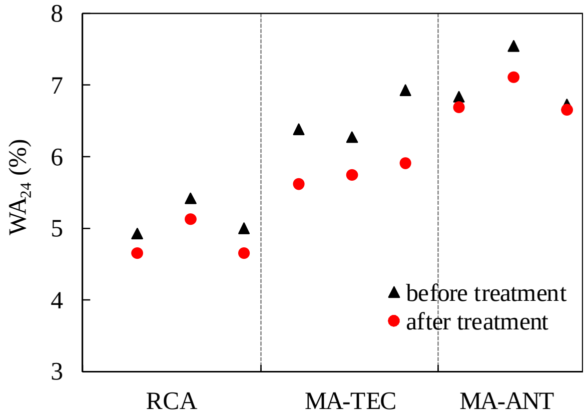

Fig.2 Water absorption after 24 hours of the recycled aggregates before and after the bio-treatment

Morphology of bio-precipitates. Compared to the untreated aggregates (Fig.3a), large amount of precipitates were clearly seen on the surface of the bio-treated aggregates (Fig.3b). The size of the particles ranged from 5 to 20 µm with various shapes (spherical, hexagonal, etc.). Bacterial imprints can be clearly seen on the particles (Fig.3c). The particles were calcium carbonate according to the

EDS analysis. Figure 3 shows the precipitates on the RCA samples. The precipitates on MA-TEC and MA-ANT showed similar morphology (images not shown).

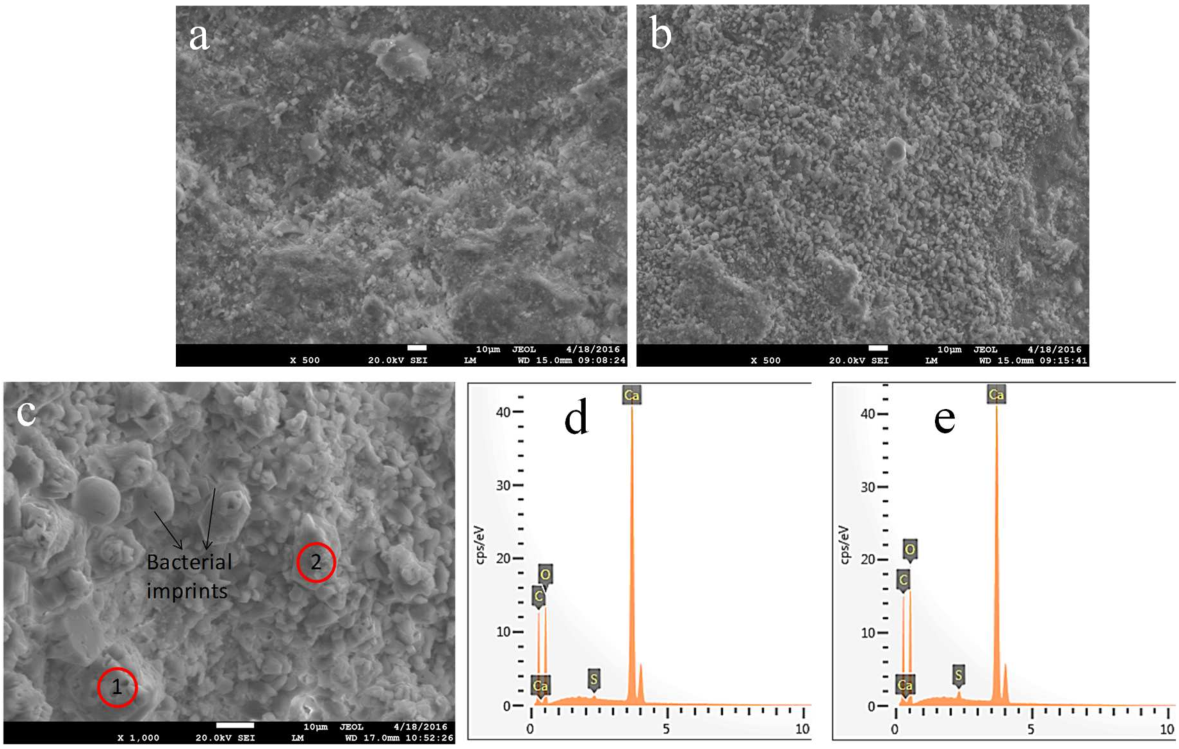

Fig.3 SEM images of the surfaces of untreated (a) and treated (b, c) RCA aggregates and EDS of the precipitated particles (d and e corresponding to 1 and 2 marked in c, respectively).

Concrete properties. Concrete made with normal aggregates had a mass density of 2350 kg/m^3. When the normal aggregates were fully replaced (the coarse fraction) by RCA, the mass density was sharply decreased to 2250 kg/m^3, around 4% (Fig.4a). However, this was improved when using the bio-treated RCA. No significant difference in mass density between the concrete made with normal aggregates and treated RCA was observed. Similar result was also observed in saturated water absorption. As shown in Fig.4b, the saturated water absorption of the concrete made with untreated RCA was about 1% higher than that of the concrete with normal aggregates and treated RCA. While there was no significant difference in saturated water absorption between NAC and RCAC/T. Use of RCA affected the compressive strength of concrete. As shown in Fig.4c, the strength was decreased by 12% when using untreated RCA and was increased by 25% when using bio-treated RCA. Bio-treatment on RCA has a positive effect on improving concrete properties.

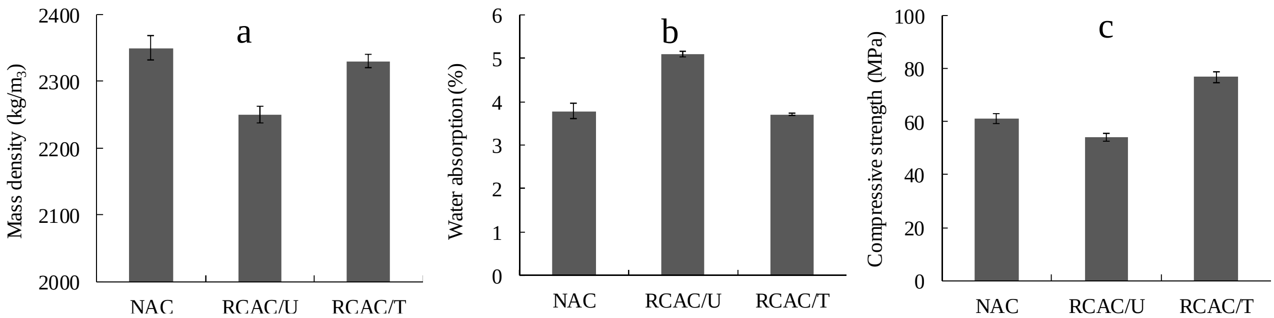

Fig.4 Properties of concrete made with normal aggregates, untreated RCA and bio-treated RCA (a: mass density; b: saturated water absorption; c: compressive strength)

Discussion

Biodeposition resulted in weight increase in both recycled concrete aggregates and two kinds of mixed aggregates. Under the same conditions, more precipitation was formed on/in the mixed aggregates (MA-TEC and MA-ANT) than on recycled concrete aggregates (RCA). This is attributed to a higher porosity in MA, indicated by a larger water absorption capacity. The formed calcium carbonate precipitates could fill the pores or form a coating, which can effectively hinder the penetration of water and/or other substances. And hence, the water absorption was decreased after the treatment. However, the decrease was not proportional to the amount of the $CaCO_3$ formed. The MA-TEC had about 4% weight increase, leading to a decrease around 1% in water absorption. Similar weight increase occurred on MA-ANT, yet the decrease in water absorption was not so significant. Though showing a lower weight increase (0.7%), the water absorption of RCA was decreased around 0.5%. Ultrasonication tests show that the precipitation has a stronger bonding with RCA than with MA. The weight loss on treated RCA was really limited (0.1%) while much more weight loss occurred on MA (1%). Therefore, it can be concluded that an effective and functional treatment more depends on the bonding or cohesion with the aggregates than the amount of the precipitates. This is logic because the treatment is meaningless if the deposited particles easily release. It seems that RCA has a better affinity with calcium carbonate precipitation than MA. This could be due to the complex composition of MA including not only crushed concrete but also masonry, glass, wood, etc, which has less affinity than concrete aggregates with calcium carbonate precipitation.

The efficiency of biotreatment was also seen in the improved properties of the concrete. The treated aggregates had a reduced porosity and water absorption, which resulted in a decrease of water absorption for the respective concrete. What is unexpected is that the compressive strength was even increased by 25% after using the treated RCA. The reasons could be that the pre-wetted aggregates still absorbed water from the mixture which decreased the effective w/c ratio of concrete. A lower w/c ratio resulted in an increased strength. The strength of the aggregates could also be improved after treatment, which needs further demonstration.

Conclusions

Bacterially induced calcium carbonate precipitation was demonstrated to have an positive effect on recycled aggregates regarding the aspects of reducing porosity and water absorption. The bio-deposition treatment is more effective on recycled concrete aggregates than on mixed aggregates; the former has a better affinity with calcium carbonate precipitation. The properties of concrete were also improved after using the bio-treated aggregates. The water absorption was decreased by 1% and compressive strength was increased by 25%.

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