Biocement treatment for upcycling construction and demolition wastes as concrete aggregates

Abhijit Mistri · Navdeep Dhami · Sriman Kumar Bhattacharyya · Sudhirkumar V. Barai · Abhijit Mukherjee

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Abstract Reutilisation of the construction and demolition (C&D) wastes as aggregate in concrete is a vital step towards sustainability as it prevents depletion of natural resources as well as alleviates wastes. However, the attached mortar on the aggregate surface renders certain shortcomings like excessive water absorption, high porosity, and weak interfaces. Recycled aggregates can be treated to improve these shortcomings. However, the minimisation of the drawbacks involves huge energy, materials, and cost. Moreover, the efficacy of such adopted method is sometime questionable, and which needs further research. This study demonstrates bio-treatment of recycled coarse aggregate (RCA) as a means of upcycling and compares it with conventional cement slurry treatment. A novel spraying technique has been applied that significantly economises biocement treatment. The experimental results show that biocement treatment reduced the water absorption by 70%. The treatment has filled the pores of RCA and has prevented water absorption. In contrast, cement slurry coating treatment shows increase in water absorption of RCA by 19%. The compressive strength of concrete with 100% biocement treated RCA surpasses that of concrete with natural coarse aggregates. The genesis of this dramatic improvement in case of biocement has been established through micro-scale studies including scanning electron microscopy and energy-dispersive X-ray spectroscopy. The cost analysis demonstrates that RCA upcycled with biocement treatment is more economical than natural aggregates or cement treated ones. Findings of the present study led to the conclusion that 100% replacement of natural coarse aggregates can be achieved by upcycling C&D wastes as coarse aggregate through bio-treatment.

Keywords Microbial carbonate precipitation · Concrete recycling · Recycled aggregate · Microstructural analysis · Construction and demolition waste · Compressive strength
1 Introduction

With global growth in construction industries, consumption of concrete has increased rapidly. It has created two problems: rapid depletion of natural resources and build up of C&D wastes that can have severe hazardous consequence. Aggregates occupy more than 75% of the volume of concrete [1, 2]. To meet this demand, natural resources such as stone and sand have been overexploited all over the world. In order to prevent further depletion, recycling of C&D wastes is imperative [3, 4]. Globally, about 40% of the total municipal solid waste comes from C&D [5, 6]. The most common use of these C&D wastes is in landfills that occupy vast areas of land and in addition, there is a risk of contamination of soil as well as water [6]. Removal of toxic pollutants from C&D waste is difficult and expensive. Encapsulation of the pollutants back into concrete by using C&D wastes as aggregate is both economical and convenient [7]. However, a few shortcomings of recycled aggregate (RA) have restricted this endeavour [8–10]. It is noticed that RA obtained from high strength concrete such as 80 MPa, perform close to that of natural coarse aggregates (NCA) [11]. However, a vast majority of recycled concrete is of poor grade and is often contaminated with other building materials such as mortar, tiles, and wood chips [8, 12]. Thus, commercial RA needs more focus to solve the real C&D waste problem.

To combat this problem, a test method for evaluation of the quality of the RA is imperative. A robust technique of petrographic characterisation of RAs to determine their suitability for construction can be found in literature [8]. It is observed that the main weakness is the old mortar that remains attached on the aggregate surface. The attached mortar (AM) has significantly higher porosity than the natural aggregates that leads to higher water absorption [13–15]. Again, micro-pores and cracks within RA may lead to weak interfacial properties [16–18]. Moreover, the failure of recycled aggregate concrete (RAC) under the compression loading is mainly owing to the coalescence of micro-cracks in the interfacial transition zone (ITZ) [18–20].

Attempts have been made to upgrade recycled coarse aggregate (RCA) through pre-processing [6, 12, 14, 21]. Methods for strengthening RCA can be subdivided into two categories: removal of the AM or strengthening of it [6, 21]. AM can be detached from the aggregate surface using mechanical [22], thermal [23] and chemical process [13]. Wei et al. [24] reported that a specific heating treatment on RCA results in high quality aggregate. However, it is also observed that a combination of heat and mechanical treatment on RCA was not enough to achieve the properties similar to that of NCA [25]. In addition, these methods require high energy or aggressive chemicals to remove mortar from the aggregate surface [6, 15, 21]. Moreover, the treatment can damage the aggregate surface affecting its performance. In another class of treatment, the AM is allowed to remain on the aggregates but strengthened using a variety of treatments. Cement slurry [26], polymers [27], nano materials [19, 28], carbonation [29] have been attempted. Essentially, these techniques make an effort to fill the pores and microcracks within RCA or provide a coating layer on surface of the aggregate. For instance, polyvinyl alcohol treatment on RCA has been showed 50% reduction in water absorption which is due to an impervious layer on the aggregate surface [30]. Although this treatment improves water repellence, improvement in strength properties (compressive) is marginal. In another technique, carbonation of the AM to densify its matrix has been reported [29]. As this treatment fills the pores by making CO₂ react with unhydrated lime, it is dependent on the amount of lime present in the AM. In another technique, a two-stage mixing approach has been adopted to strengthen the AM [31]. The first stage treats the RCA by churning it with cement and water followed by a regular concrete mixing. Both the mechanical as well as durability properties of RCA is improved by the method as reported [31–33]. Moreover, pozzolanic admixtures (for example, fly ash) have been reported to improve RCA properties [9, 34]. Further discussion on different treatment methods on RCA to improve its properties, advantages and constrains can be found in literature [6, 21].

It has been demonstrated that more sustainable and cost-effective concrete could be achieved by strengthening of AM compared to the removal of it [6, 35]. However, success of the treatment is dependent on its ability to reduce water absorption as well as densifying the pores and cracks. The treatment fluid should be able to penetrate deep inside the pores of microscopic scale as well as deposit enough solids to fill them. However, these two requirements are contradictory as
The presence of higher solid content would make the fluid more viscous. The biomineralisation process is particularly suitable for this application [36]. In this process, the solids remain dissolved in the fluid keeping its viscosity low at the time of transport, but they mineralise when they come in contact with the microbes [36]. It is noteworthy to mention that the efficacy of the calcite precipitation depends on types of bacteria used, cell concentration, environmental condition, nutrients, number of doses, etc. [36, 37, 39, 40]. This MICP technology is based on a natural process wherein bacterial cells produce inorganic minerals as part of their basic metabolic activities [36, 37, 39, 40]. In the process of urea hydrolysis, via metabolic activity of bacteria, urea is converted into ammonia (NH₃) and carbon dioxide (CO₂). This ammonia (NH₃) reacts with water (H₂O) within the microenvironment to produce ammonium (NH₄⁺) and hydroxyl ions (OH⁻) increasing the pH of the microenvironment. At high pH, carbon dioxide (CO₂) reacts with hydroxyl ions (OH⁻) to form bicarbonate (HCO₃⁻). Finally, in the presence of calcium source (for example, calcium chloride (CaCl2)), bicarbonate (HCO₃⁻) reacts with hydroxyl ions (OH⁻) to precipitate calcium carbonate (CaCO₃). The whole process of calcium carbonate precipitation is represented in Eq. 1–4 [36, 37, 39, 40].

\[ \text{NH}_2 - \text{CO} - \text{NH}_2 + \text{H}_2\text{O} \rightarrow 2\text{NH}_3 + \text{CO}_2 \]  
\[ 2\text{NH}_3 + 2\text{H}_2\text{O} \rightarrow 2\text{NH}_4^+ + 2\text{OH}^- \]  
\[ \text{CO}_2 + 2\text{OH}^- \rightarrow \text{HCO}_3^- \]  
\[ \text{Ca}^{2+} + \text{HCO}_3^- + \text{OH}^- \rightarrow \text{CaCO}_3 + \text{H}_2\text{O} \]

Although there are some evidences of ability of biomineralisation in improving RCA, for its industrial application, several systematic steps must be taken. Moreover, most of the previous biocement methods follows immersion process which involves a relatively larger quantity of chemicals and water [41]. Thus, an alternative to the immersion process is essential [41]. Porter et al. [42] has demonstrated the superiority of spraying over immersion in rammed earth blocks. It is observed spraying makes the bacteria settle inside the pores and they nucleate mineralisation on their cell membranes [43]. Thus, the entire quantity of biocement is deposited inside the aggregate. Spraying can also cut down the water requirement to a great extent.

A very limited numbers of study has been reported to improve RCA using biocement and which needs more research to understand the applicability and efficacy. Moreover, biocementation technology must be benchmarked against the existing processes such as cement slurry treatment.

In this paper, we evaluate biocementation on RCA to mitigate its drawbacks and compare it with conventional cement slurry treatment. The aim of the study is upcycling of C&D waste as aggregate into concrete. Spraying method for biocementation is used for the treatment of commercial RCA. Thus, this paper shifts the existing immersion techniques of biocement to newly spraying method, which is more feasible and economical in possible future field application. Performance of both treated aggregates and treated aggregate concrete are recorded and compared with the control one. Both the systematic conceptualised and microstructural study to analyse the phenomenon is reported. The findings lead to significant improvement in both physical properties of treated RAC and concrete strength compared with to control aggregate. Finally, a cost analysis of concrete produced from natural and recycled aggregates has been presented. Overall, the present research contributes to the understanding of insights of C&D waste problem, weakness of RCA and mitigation strategies, establishment of efficacy of the applied method and benchmarking, and finally, possible disposal solution via 100% replacement of NCA with treated RCA supporting sustainability in the concrete industry.

2 Experimental work

2.1 Recycled and natural aggregates

Commercial RCA was obtained from Capital Recycling Plant located in Perth, Western Australia. In this plant, concrete from different sources is recycled. Therefore, strength of parent source concrete is unavailable. In absence of available information, a detailed characterisation of the aggregate was performed. The aggregates were supplied in two size grades: 4.75–9.5 mm and 9.5–19 mm. The NCA consisted of crushed granite rock. It can be observed that the aggregates have similar grading pattern. Gradation curve of NCA and RCA is given the supplementary materials (SF1). Further, separation...
and classification of different RCA were conducted by basic petrographic analysis of RCA [8]. The detailed petrographic characterisation can be found elsewhere [8]. The RCA was a mixture of materials ranging from almost pure aggregate to almost pure mortar and even impurities such as brick bats, wood chips and paper. A schematic representation of four major aggregate categories is shown in Fig. 1. The four categories of aggregates were: a) aggregates partially covered with a thin AM (Type A); b) aggregates with a thin AM covering almost the entire surface (Type B); c) more than one aggregate bound by mortar (Type C); and d) aggregate comprising of mortar only (Type D). Understandably, the performance of different types of aggregates is bound to be different. Therefore, aggregates of a representative volume were segregated into the four major classes. Figure 2 presents the composition of the different classes of aggregates in RCA. It can be seen that both grades are composed of type A to D aggregates. Moreover, there is about 5% impurity such as pieces of tiles, marbles brickbats and even wood chips and paper. The compositions are different in the two size grades. In larger aggregates, 48% were sound belonging to type A or B. Type C and D consisted of nearly 48%. In smaller grades sound aggregates (Type A or B) are only 20%. Type C and D are nearly 74%. Thus, aggregates of smaller grade sizes are weaker. The aggregates as received have been used in this investigation without any effort of screening the weak components as that will add to the cost of processing.

Both the physical and mechanical characteristic of different aggregates was estimated using Australian Standards [44–48] and presented in Table 1. Based on the obtained results on RCA, the aggregate can be classified as Class II according to RILEM recommendation [49]. Class II type of RCAs is limited to absorb a maximum of 10% and maximum allowable strength class is C50/60. Water absorption of RCA is found comparatively higher than NCA. The specific gravity of RCA is slightly lower and flakiness index higher. However, mechanical properties of RCA demonstrated that it was much more fragile than NCA. This is due to the mortar particles and impurities present in RCA. Clearly, this RCA would require strengthening to produce concrete of adequate strength.

2.2 Conventional cement treatment

Ordinary Portland cement (Type GP according to Australian standard [50]) was used for the treatment as well as concrete production. The chemical composition of cement is given in the supplementary materials (ST1). Following Martirena et al. [26], a cement slurry was produced by mixing 40% water with the cement. The RCA was maintained in saturated surface dry condition. Then it was rolled in the slurry in a rotating drum for at least 3 min. After rolling, the aggregates were kept on a hard platform (thick plastic) for drying for 24 h in ambient condition. Then, it was cured for 7 days by immersing in water. Subsequently, they were allowed to dry for 24 h in ambient condition. The quantity of slurry was calculated and prepared as per Li et al. [51]. A picture of the aggregates before and after the cement slurry treatment is given in the supplementary materials (SF2) and visible cement slurry layer can be seen. Before mixing the concrete, water absorption and specific gravity tests of the treated RCA were conducted.

2.3 Biocement treatment

Present biocement treatment consisted of three steps: fixing, bacterial treatment and cementation. The protocol as in [43] was followed. Figure 3 shows the steps in the biocement treatment. The aggregates were placed in a tray in one layer. They were mixed thoroughly at every step to ensure that all the
aggregates receive the treatment uniformly. The aggregates were dried for 24 h. All the aggregates were placed in a fume hood. The volume of treatment fluid was estimated as 90% of water absorbed by the RCA in 24 h. A fixing solution consisting of 25 mM CaCl2 was sprayed on the RCA. After three hours, the bacterial solution was sprayed on the RCA. The bacterial solution consisted of *Bacillus pasteurii* (ATCC 11,859) strain. The microbe was grown in a specific media solution that contains 10 g/l Ammonium Sulphate, 10 g/l Yeast Extract, 7.85 g/l Tris Base, and distilled water. The working pH of the media solution was 9.08 and adjusted by adding 0.5 M HCl. During the incubation period (at 180 rpm for 24 h) for the bacterial growth, the bacteria culture was kept at a temperature of 37 ± 1 °C. Spectrophotometer was used to measure the concentration of the bacterial culture in terms of optical density (OD600) at 600 nm and the obtained value was 1.60. After spraying the bacterial solution, the RCA was left in ambient condition for 24 h. Cementation solution consisting of 30 g/l Urea, 55.5 g/l CaCl2, and 1 g/l Yeast Extract was sprayed on the RCA. The cementation spray was repeated every 12 h for 8 days. The water absorption of the RCA was checked every day. After 8 days of treatment the water absorption was close to that of NCA.

A separate set of RCAs was treated to note the change in weight. Six representative aggregates of size range 16–19 mm were chosen (total weight of approximately 100 g). The dry weight was noted after keeping them at 105 °C temperature for 24 h. They were treated as above and at the end of 8 days they were dried for 24 h. Their weight was noted.

### 2.4 Concrete production

Four different aggregate types were used in the concrete mixes. NCA, untreated RCA consisted of the control batches. Remaining two were RCAs treated with conventional cement and biocement. All aggregates were maintained in saturated surface dry condition. It may be noted that the specific gravity of different classes of aggregates were different. To maintain the concrete volume same, the weights of the coarse aggregates have been adjusted accordingly. Natural fine sand (grading size 75 μm-4.75 mm) was used as fine aggregates. The water absorption of sand was 1.02%. The concrete mix design consisted of different aggregates, 400 kg/m³ cement (OPC), 640 kg/m³ sand, and water of 160 kg/m³. Four different concrete like NAC (with control NCA), RUC (with untreated RCA), RCCC (with cement slurry treated RCA), and RBCC (with biocement treated RCA) were produced in which the different aggregates were used.
The quantity of coarse aggregates is 1180, 1045, 980, and 1062 kg/m$^3$, respectively. For mixing of concrete, a 70L pan mixer was used. For both the compressive strength and tensile strength tests \[52\], cylinder samples were prepared. The dimension of cylinder sample was 200 mm height with 100 mm diameter. Compressive strength of cylinder sample was aimed to achieved 40 MPa at 28 days and the mix design was prepared accordingly. After 24 h of casting, the cylinder samples were demolded. Next, the samples were kept immersed in lime water to prevent the leaching of lime from the test samples during the curing period \[53\]. The samples were tested after 7 and 28 days of curing. For testing of the samples, a UTM of the capacity of 2 MN was used. A load rate of 0.25 MPa/s was applied during both the compressive and tensile strength tests as per ASTM standard \[54\].

3 Results and discussion

Performance of the treatment methods has been compared at two phases: aggregate and concrete. At aggregate level, the basic physical properties (density and water absorption) of the aggregates were compared. Moreover, the efficacy of the present method was benchmarked against the previously reported biotreated RCAs. On the other hand, at concrete level, strength properties (compressive and tensile strength) were measured. Microstructure of different concrete were assessed to establish the relationship between different properties.

3.1 Conventional and bio-treatment

In this section, recycled biotreated coarse aggregates (RBC) are compared with natural aggregates (NCA), recycled untreated aggregates (RUC) and recycled cement treated aggregates (RCC). Figure 4a presents the water absorption in different aggregates. It can be seen that NCA absorbs less than 1% water while RUC absorbs more than 5% water. Clearly, this RUC is not suitable for replacing NCA unless it is treated to drastically bring down its water absorption. The conventional cement treatment on RUC increased the water absorption further by about 19%. Clearly, this RUC is not suitable for replacing NCA unless it is treated to drastically bring down its water absorption. The conventional cement treatment on RUC increased the water absorption further by about 19%. Clearly, the present cement treatment did not fill the pores. Instead, it created an additional porous layer of cement that absorbed more water. In contrast, Martirena et al. \[26\] have reported a reduction in water absorption due
to cement treatment. In that case the specific gravity of recycled aggregates (2.37) is just 0.23 less than of the natural aggregates. While in the present case, the difference is 0.29. It seems that the present RCA has relatively higher proportion of mortar which might have resulted in poor resistance to water absorption. RBC, on the other hand, has about 70% lower the water absorption than RUC. It demonstrates the ability of RBC in improving RUC even when it contains substantial highly absorbing materials. It is demonstrated earlier that the bacteria are able to penetrate even small pores of diameter of about 1 μm and seal them. Thus, instead of forming a surface layer, bacterial treatment has densified the outer layer of the aggregates. However, RBC absorbed nearly 1.6% water which is higher than NCA. It may be due to the residual pores left unfilled by the treatment. It may be filled by further spraying, or it is also possible that these pores are too narrow for the bacteria to enter. The effect of the residual water absorption on concrete is tested later in the paper.

3.2 Specific gravity

Figure 4b presents the specific gravity measured with different samples. NCA has the specific gravity of 2.7, which is in the range of that of granite. Thus, the NCA is sound. The specific gravity of RUC is lower around
2.4, which is expected as it is a heterogeneous mixture of rock and mortar. RCC had a specific gravity of 2.25, marginally below that of RUC. Evidently, RCC has not densified the aggregate. RBC, on the other hand, had increased the specific gravity from 2.4 to 2.5. This demonstrates that RBC has indeed densified the surface of the aggregate.

On an average, about 2.05% weight gain was observed after bio-treatment. Aggregates with higher proportion of attached mortar had a higher weight gain. The result is given in the supplementary materials (ST2). Prior literature has reported 1–2% weight gain due to bio-treatment [35, 55]. The weight gain is dependent on the fraction of mortar in the RUC. The present RUC is likely to have a higher fraction of mortar than the previous investigations.

3.3 Biotreatment techniques

The present spraying technique is compared with the immersion technique used hitherto [35, 55]. For immersion of 100 g of RCA approximately 150 ml of water is required. The materials requirement of prior methods vis-à-vis the present technique is indicated in Table 2. In [35], the RCA has been immersed twice in cementation solution while in [55], it is immersed only once. Thus, water requirement approximately 300 ml and 150 ml respectively. In the present method, water requirement was 96 ml. The requirement of urea in present method too is considerably lower than [35] and marginally lower than [55]. The source of Calcium for the present method and [55] is CaCl₂ while [35] uses Ca(NO₃)₂. However [55], uses much lower concentration of CaCl₂. Thus, while present consumption is higher than that of [55] it is considerably lower than that of [35]. It is prudent to mention that these indicators can be greatly affected by the quality of the RAC. Thus, the comparison presented here is of specific experimental values only. There are other pros and cons of immersion and spraying. While immersion method may need a large container for the treatment, spraying would require space to spread the aggregates. Again, the final result from biocement treatment depends on several factors (like type of bacteria, concentration of bacteria, chemical used, environmental condition, etc.) and that was not consider under the present scope of work.

Figure 5 presents the reduction in water absorption achieved by different techniques. While the immersion techniques achieved 15–20% reduction, spraying achieved 70%. It may be recalled that less quantity of chemicals was used in spraying. Therefore, the amount of carbonate cannot be higher. In case of immersion, the bacteria can leave the aggregates and nucleate carbonate deposition anywhere within the solution. While in spraying, the bacteria and the cementation media are restricted to remain in the pores. Thus, deposition can be targeted inside the pores by spraying. A micrograph of the growth of carbonates after spraying is given in supplementary materials (SF3).

3.4 Compressive strength

Concrete mix design was prepared with a target strength of 40 MPa for the NAC. Figure 6 presents the compressive strength of concrete after 7 and 28 days. NAC achieved the target strength. The compressive strength of RUC was 14% and 13% lower than NAC at 7 and 28 days respectively. RCCC shows 17%

| Table 2 Consumption of materials in cementation fluid |
|-----------------------------------------------|
| Cementation potential chemicals | Doses | Method of application | Spraying [Present] |
|---------------------------------|-------|----------------------|-------------------|
|                                 |       | Immersion            |                   |
|                                 |       | Twice [35]           | Once [55]         |
| Water                           | Volume (ml) | 300 (+ 212.5%)* | 150 (+ 56.25%) | 96 |
| Urea                            | Concentration (g/l) | 30 | 20 | 30 |
|                                 | Weight (g) | 9 (+ 212.5%) | 3 (+ 4.17%) | 2.88 |
| Ca source                       | Concentration (g/l) | 82.04 | 16.80 | 55.5 |
|                                 | Weight (g) | 24.61 (+ 361.72%) | 2.52 (− 52.72%) | 5.33 |
|                                 |         |                      |                   |
| *Represents percentage with respect to the present spraying technique |
improvement over RUCC in compressive strength at 28 days. The strength of RCCC was close to that of NAC validating the hypothesis that recycled aggregates can replace natural ones 100%. The reason for this marked improvement is possibly a denser ITZ due to the cement rich outer surface of the aggregates. It corroborates the observation in [51]. The RBCC shows 24.22% higher compressive strength than RUCC at 28 days. It surpasses the compressive strength of even the NAC. Wang et al. [35] reported about 16% higher compressive strength with biotreatment of mixed recycled aggregates. Feng et al. [56] reported that bio treatment on recycled fine aggregate (RFA) can improve compressive strength of about 14% than the control RFA. Clearly, biotreatment can valorise recycled aggregates mixed with impurities to replace natural aggregates by 100%. However, a long-term test of would be necessary to confirm its durability properties. Moreover, structural application of such treated aggregate concrete could be considered as future goal.

Figure 7 show schematic diagrams of the concrete with its aggregates. Regular interface between coarse aggregate and cement mortar is formed in NAC.
In case of RUCC (Fig. 7b), there are existing interfaces within the aggregate. Additionally, when untreated RCA is used in concrete, another interface with new cement mortar is formed. Moreover, it can be expected that the interface with untreated RCA is comparatively weaker than interface with NCA. Thus, lower strength can be expected in concrete with untreated RCA. Similarly, predictions in lower strength of concrete with untreated RCA is observed by several researchers [12, 15]. Cement slurry treatment creates a cement rich layer over the RUC this layer interacts with the cement matrix in concrete resulting in a stronger bond. However, it does not penetrate deep into the aggregate. In case of RBCC, the weak AM on the aggregate surface is densified through carbonate deposition rather than forming a coating (like RUCC). The prime reason behind this is the low viscosity of the cementation solution that allows it to penetrate deep into the aggregate and fill the micro-pores and cracks. The biocement treated aggregate would also minimise an abrupt change in the aggregate properties. As a result, the concrete strength with RBCC surpasses even that with the NAC.

3.5 Tensile strength

Figure 8 shows the tensile strength test results at 7 and 28 days. The tensile strength of NAC is about 20% and 19% higher than that of RUCC at 7 and 28 days, respectively. The tensile strength of RCCC and RBCC is about 7% and 8% higher respectively than that of RUCC at 28 days. However, unlike compressive strength, the tensile strength of concrete with NAC was higher than RBCC. The possible reason for this phenomenon is explored through the observation of the damage patterns of the samples.

3.6 Damage progression

The initiation and progression of damage in the samples were monitored at the time of both compression and tensile tests. Figure 9 shows the pictures of the samples at the time of failure. NAC developed a number of tensile cracks in the axial direction of the cylinder. As the loading progressed, the cracks coalesced, and the sample failed in large chunks. The aggregates remained generally intact, and the cracks passed through the cement matrix. In case of RUCC, a larger number of spalling locations were observed that were relatively smaller in size. It was also observed that the cracks passed through the AM of the RUCC. This indicates that relatively weaker aggregates leading to delamination of the surface layer of the cylinder. With RCCC, the number of spalling location has reduced although they can be clearly identified. Failure through cracking of AM still persisted. In case of RBCC, the failure of the AM is largely avoided although occasional AM locations still failed. Thus, the crack pattern is closest to that of the NAC.

3.7 Microscopic investigation

Microscopic investigations were performed both on the aggregates and the concrete. Figure 10 presents the optical microscopic observation of RBC. Rich deposit
of carbonate on the surface as well as in the cracks is clearly discerned.

Prior research has concluded that the improvement in the mechanical properties of RAC is directly related to the improvement of the microstructure [19, 35, 57].
Scanning electron microscopy (SEM) was used to study the concrete microstructure. Samples were collected from the concrete cylinders and care was taken to include the aggregate-concrete interface. The samples were polished but left uncoated. They were examined using a TESCAN, model Mira3 SEM with variable pressure condition. The magnification was adjusted to obtain images that best reveal the specimen microstructure.

Figure 11 presents the micrograph of different samples. Figure 11a shows the interface between NAC and cement matrix. Figure 11b reveals that in RUCC there is aggregate, attached mortar and the new mortar. Thus, multiple interfaces are formed. It also reveals a crack in the AM signifying that it is the weakest component in the material system. Therefore, the aggregate is weak and not suitable for high grade concrete. Figure 11c presents the materials map in RCCC. The RCA with AM and a thin coating of the cement treatment is visible. Figure 11d presents the magnified image of the boxed area of Fig. 11c. Clearly the cement treatment did not penetrate into the AM. Also, its thickness is variable. Moreover, a crack along the interface of the cement layer and the RCA is observed. It reveals the relatively weak interface between the cement layer and the RCA, which has resulted in the lower strength. Figure 11e presents an interface of RBCC. In this case, unlike RCCC, no separate layer of treatment is seen. Thus, the bacterial system has fused into the AM of the RCA densifying it. The treatment does not create another interface. Evidently, the interfacial cracking between the treatment and the AM is avoided in this case. The crack is at the interface of the new cement matrix and the RBCC. Thus, it can be concluded that the RBCC has been strengthened enough to avoid the failure of the aggregate. Figure 11f presents a magnified view of the boxed area of Fig. 11e. A dense matrix is clearly seen. However, the proof of densification through biotreatment would need further investigation.

3.8 EDS analysis

The chemical elements within the treated system (RCCC and RBCC) were analysed by Energy-dispersive X-ray spectroscopy (EDS). The details can be found in the supplementary materials (SF4 and SF5). However, the presence of carbonate is not conclusively proved in these spectra.

The difficulty in visualising calcium carbonate through elemental mapping in presence of cement is that they both have a common element, Calcium. To circumvent the problem, an elemental map of the RBC and the new matrix is developed. Figure 12 shows the image of the area. In this map, spatial distribution of the key elements is depicted. First Si is plotted. Clearly Si rich sand particles stand out in the image. However, Si is also present in cement albeit in smaller proportion. Therefore, a lighter distribution of Si is seen throughout the domain confirming the presence of cement. In the map of Ca, dark patches are seen at the location of sand particles, as there is no Ca in sand. However, Ca is present both in cement and calcium carbonate. A denser concentration of Ca is observed in the RBC region than in the matrix region. Thus, there are more contributors of Ca in the RBCC region. This extra Ca is the result of bio-treatment. Clearly the biotreatment has penetrated deep inside the RA. Another way of confirming the presence of calcium carbonate is to map aluminium, which is present in cement but not in calcium carbonate. In the composite image, distinct patches of Ca without Al are discernible. It can be concluded that these are the voids that are filled by calcium carbonate. This analysis clearly depicts that the presence of Ca is denser in the RBCC, and it is clustered in pockets where there were voids prior to the treatment. Thus, densification of the AM has clearly been evidenced in the case of RBCC.

3.9 Cost analysis

Materials that were used to make concrete along with treatment processes are summarised in Table 3. The costs were as paid for supply to the concrete laboratory of Curtin University. Labour and manufacturing costs were not included as they are nearly the same for all the processes. NA is understandably more expensive than RA. However, RA has additional cost of treatment. In totality, NAC (USD 229.97/m³) was found to be most expensive. RUCC (182.58) was 20% cheaper per m³. Even after the cost of treatment was added, RCCC (196.66) and RBCC (197.54) were found to be economical than that of NAC. Clearly, treated RA scores higher over NA both on account of environment and economy. It may be recalled that the concrete with different aggregates had variable compressive strength. To account for that effect, cost in terms of USD per MPa of compressive strength of concrete was
Fig. 11 SEM image of the different concrete interfaces
Fig. 12 Mapping of the element present in RBCC
calculated where the total materials cost is divided by compressive strength (in MPa) at 28 days. On this account again, the NAC was found to be most expensive. RUCC was in the second position. RCCC was cheaper than RUCC due to its higher strength. RBCC was found to be the cheapest. Thus, upcycling of C&D waste using biocement treatment is found to be both economical and environmentally friendly. It is noteworthy to mention that the cost of materials varies based on region and performance. Labour and manufacturing costs may be included for large scale production. Moreover, environmental benefits can be analysed further, and which needs a separate study.

### 4 Conclusions

The present study demonstrates upcycling of C&D wastes as RCA in concrete. The objective of the investigation was to examine if it is possible to treat the RCA to get rid of its weaknesses and match the strength of NAC. Recently developed biocement treatment on RCA is found effective to mitigate its drawbacks as well as to make fresh concrete. It is observed that up to 70% reduction in water absorption can be achieved compared to the control RCA through the biocement treatment by spraying method. The present bio-treatment technique by spraying was compared with the immersion technique used hitherto. It was observed that spraying technique greatly economises material consumption. The surface porosity, microcracks and micro-meso pores within RCA can be minimized considerably with the bacterial induced CaCO₃. Therefore, the water absorption of the RCAs can be almost similar to NCAs after adoption of a suitable surface improvement technique. In contrast, the cement slurry coating can improve the surface properties, but it actually increases the water absorption of RCAs. The water absorption has increased by about 19% compared to the untreated RCA. This is because of the pervious layer on the aggregate surface.

Concrete strength after treatment of RCAs has increased compared to the untreated RAC. The number of ITZ is decreased in case of bacterial treatment compared to cement slurry coating where the same number is increased. Moreover, if the number of weak ITZ increases, more complex behaviour can be expected based on its character.

Optical microscopic images revealed rich deposits of carbonate on the surface as well as in the cracks of RCA. Further, the SEM images revealed clear existence of multiple interfaces in RUCC: aggregates, attached mortar and the new mortar. In RCCC, interfacial cracking along the cement layer was

| Materials | NAC  | RUCC | RCCC | RBCC |
|-----------|------|------|------|------|
| Aggregate (10 mm) | 26.18 | 6.27 | 5.61 | 6.25 |
| Aggregate (20 mm) | 38.76 | 11.29 | 10.10 | 11.25 |
| Sand | 23.50 | 23.50 | 23.50 | 23.50 |
| Cement (GP) | 141.20 | 141.20 | 157.02 | 141.20 |
| Water | 0.32 | 0.32 | 0.44 | 2.24 |
| Urea | 8.89 | 8.89 | 8.89 | 8.89 |
| Calcium Chloride | 1.50 | 1.50 | 1.50 | 1.50 |
| Yeast extract | 1.85 | 1.85 | 1.85 | 1.85 |
| Ammonium Sulphate | 0.01 | 0.01 | 0.01 | 0.01 |
| Tris Base | 0.85 | 0.85 | 0.85 | 0.85 |
| Total materials cost (USD/m³) | 229.97 | 182.58 | 196.66 | 197.54 |
| Compressive strength (MPa) | 43.51 | 37.62 | 44.22 | 46.73 |
| USD/MPa | 5.29 | 4.85 | 4.45 | 4.23 |

The unit rates (in USD) are: NCA-10 mm (55.47/tonne), NCA-20 mm (54.75/tonne), RCA-10 mm (15/tonne), RCA-20 mm (18/tonne), sand (36.72/tonne), cement (GP) (353/tonne), urea (400/tonne), Calcium Chloride (165/tonne), yeast extract (6000/tonne), ammonium sulphate (125/tonne), tris base (100/100 kg), water (2/kL)
observed. In the case of RBCC, no separate layer of treatment is seen. Thus, the bacterial treatment had fused into the AM of the RCA densifying it. The cracking was shifted to the interface of the new cement matrix and the RBC. Thus, it can be concluded that the RBC has been strengthened enough to avoid the failure of the aggregate. Through elemental mapping using EDS, the analysis clearly depicted that the presence of Ca is denser in the RBCC, and it is clustered in pockets where there were voids prior to the treatment. Therefore, densification of the AM has clearly been evidenced in the case of RBCC.

The analysis of costs demonstrates that upcycled C&D wastes offer the lowest cost per MPa of compressive strength of concrete. Although the cost can vary with geographical locations, the study demonstrates the competitiveness of upcycled C&D wastes, especially considering its environmental benefits. However, long term performance of biocement treated RCA in concrete needs further research.

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Declarations

Conflict of interest The author declared that there is no conflict of interest.

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