

Recycled concrete aggregate properties improvement

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ABSTRACT

In recent years, there has been increasing interest in the use of recycled concrete aggregate (RCA) as a sustainable alternative to natural aggregates in construction. RCA is obtained by crushing and reusing old concrete, which contains both the original aggregate and a cement mortar layer. However, the quality of RCA can vary depending on the characteristics and quantity of the leftover mortar. In this study, we aim to investigate different methods for improving the chemical, physical, and mechanical features of RCA, with a focus on untreated RCA, RCA exposed to a pre-soaking treatment in HCl, and RCA exposed to rapid carbonation. The overall results show that if RCA is obtained by crushing compact, high-quality concrete, the procedures for improving aggregate quality are not necessary.

KEYWORDS

Recycled concrete aggregate, Accelerated carbonation, Hydrochloric acid

1. INTRODUCTION

In order to reduce the amount of natural resources used in construction and the environmental impact of concrete manufacturing, Europe implemented a regulation that actively encourages the use of recycled aggregates in concrete production. "The necessary measures to promote the re-use of products and the preparing measures for re-use activities, particularly by promoting the establishment of economic tools and criteria about tenders, quantitative targets or other measures," are required of the states that have ratified European Directive nb.98 as of November 19, 2008 [1]. It is stressed that at least 70% more weight will be recovered through recycling and other comparable recovery materials, including building waste [2]. Recycled Concrete Aggregate (RCA) is a material made of natural, unpolished aggregate with a high percentage of attached mortar. It is derived from concrete and demolition trash, giving it a porous, uneven texture and a lower density [3, 4]. Depending on the size of the aggregate, the volume of residual mortar in RCA might vary from 25% to 60% [5]. Some articles have reported that, for particle sizes ranging from 20 to 30 mm, over 20 percent of cement paste is attached to the RCA's facet [6, 7]. Existence of a few interfacial transition zone (ITZ) types between the "old" and "new" composites is an unusual characteristic of RCA, and it may play a significant role in the internal microstructure of the concrete. Therefore, an easier application of RCA will be enabled if the attached cement mortar can be enhanced. Reinforcing and removing the attached mortar are the two basic methods used to intensify the RCA characteristics.

2. THE IMPROVEMENT METHODS

2.1. Mechanical grinding

A straightforward and often applied procedure for removing attached mortar from natural aggregate is ball-milling. The primary drawback is the possibility of micro-fractures developing in RCA during the grinding process. Endogenous cleaning [2] involves putting RCAs in a revolving mill drum and letting them strike against one another to scrape off fragments of the mortar that is adhered. The mill drum (Figure 1), which measured 30 cm in diameter and 50 cm in depth, rotating at a speed of 60 rotations per minute and contains 33 percent "raw" recycled aggregates. Aggregates were water washed and then dried to remove any tiny particles and contaminants following the endogenous cleaning procedure. The water absorption capacity decreased over time, with protracted periods ranging from 2 to 10-15 minutes, according to the cleaning outcomes. As the endogenous cleaning process was carried out, the amount of absorbed water decreased by 50% and 20%, respectively, while the amount of fine particles increased.



Figure 1: The mill drum [2]

Additionally, the attached mortar content of the cleaned aggregates was 15% lower than that of the uncleaned recycled aggregates, which had a 30% attached mortar content. Coarse RCA samples were heated up for an hour at four different temperatures –250°C, 350°C, 500°C, and 750°C – in a traditional electric oven as part of the heat treatment method [8,9]. Since higher temperatures clearly have an adverse influence on coarse RCA properties, this approach successfully improves numerous physical parameters, including water absorption, specific gravity, porosity, and freezing and thawing at 300°C – 350°C. Internal strains as a result of exposure to high temperatures (400°–600°C) are observed, followed by thermal expansion. In contrast, material subjected to a greater temperature range (600°C - 800°C) experiences significant micro fracturing of the cement matrix, which causes disintegration, mass loss, and degradation of the material.

2.2. Pre-soaking in water (pre-saturation)

The study conducted by García-González et al. [10] demonstrated that a brief submersion of recycled aggregates in water improves the consistency of fresh recycled concrete, albeit at the expense of a minor reduction in compressive strength. The decline varied between 11% and 13% for the 3 and 5 minute soaking and pre-saturation intervals, respectively.

2.3. Pre-soaking in acid

Cement hydration products are dissolved in hardened paste using an acid solution in the following manner: first, soak the RCA in an acidic solution for a full day at a temperature of around 20°C. Afterwards, use distilled water to wash away the acidic solvents. RCA must be submerged in water for 24 hours before the new concrete can be mixed. Three acidic solvents, namely hydrochloric acid (HCl), phosphoric acid (H₃PO₄), and sulfuric acid (H₂SO₄), were utilized at a concentration of 0.1 mol each. This created a suitable chemical environment that allowed the RCA to remove the old cement mortar without compromising the quality of the aggregate. While the mechanical qualities of the RCA were enhanced and its alkalinity, chloride, and sulphate portions remained unaffected, the pretreated RCA water absorption values demonstrated a notable decrease.

Ismail et al. [12] employed an unrefined RCA that was submerged in 0.5 mol HCl for 24 hours a day. The weak mortar disintegration was accelerated by shaking the container occasionally. This improved the efficiency of the acid reaction. Subsequently, the aggregates were irrigated with distilled water and allowed to drain. An impregnation with a

calcium metasilicate (CaSiO_3) solution was then applied for a full day with the objective of covering the unrefined RCA surface with particles of calcium metasilicate, which would replenish the pores and fractures throughout the aggregate's physical face. Simultaneously, the RCA surface is coated with calcium metasilicate particles, which dissolve during mixing and are intended to serve as a filler. The cement hydration product acts as an agent for densification of the interface structure, thereby strengthening the contact bond between the aggregate facet and the cement matrix. In a study conducted by Al-Bayati et al. [9], coarse rice ash (RCA) was soaked for 24 hours at room temperature, approximately 20°C , in an acidic solution that contained 37% hydrochloric acid (HCl) and 99.7% acetic acid ($\text{C}_2\text{H}_4\text{O}_2$) at one mole concentration.

In a study conducted by Guneyisi et al. [13], aggregates were submerged for 24 hours at 20°C in a 0,5mol hydrochloric acid (HCl) solution. Submersion in distilled water was then used to eliminate the acidic solution. Within the same study, an additional technique involved submerging the aggregates for 30 minutes in a $\text{Na}_2\text{O}\cdot n\text{SiO}_2$ sodium silicate water glass. After that, they were held in suspension for 10 minutes to allow any remaining water to seep out of the aggregates that had been removed from the solution. To avoid connecting the aggregate particles, the last step was to dry them in an oven. The SEM study showed that the loose attached mortar and other loose materials may be removed from the RCA surface by using a 0,1 mol solution of HCl. As opposed to the untreated RCA, certain characteristics of the treated RCA, such as density and water absorption, improved after the HCl treatment. When compared to other applied treatments, the aggregates treated with water glass, however, dramatically reduced the water absorption. New ITZs in SCCs with treated RCA produce a microstructure that is less porous but denser and more bound, according to SEM analysis (Figure 2).

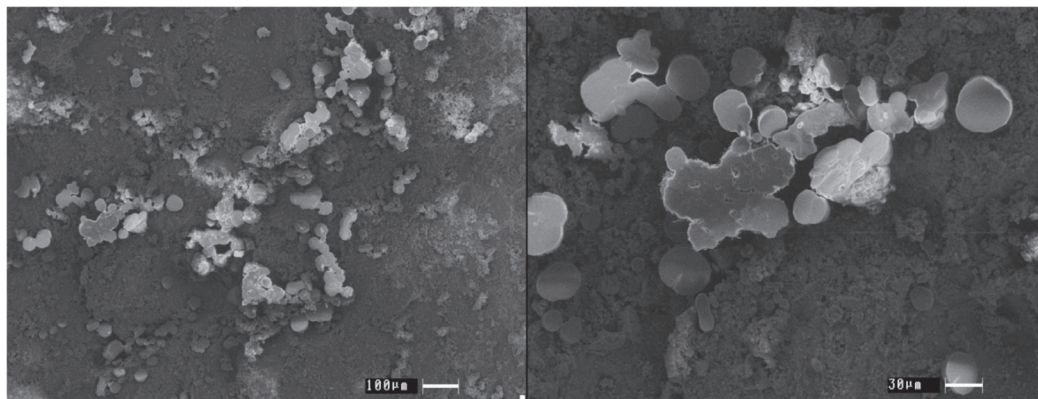


Figure 2: Scanning electron micrograph of recycled aggregate grain ($w/c = 0.45$, fraction 12/16 mm) after biodeposition treatment [16]

Along with estimates of the effects of treated aggregates on compressive concrete strength, this study [14] also examines the effects of different acid concentrations and treatment times on the mechanical and physical characteristics of unrefined RCA. In the investigation, different HCl types with mol acid molarities of 0.1, 0.5, and 0.8 were utilized.

For one, three, and seven days, the aggregates were submerged in acidic solutions. As demonstrated by the linear relationship between the quantity of fatal loss and the rise in acid molarity, low HCl concentrations have the ability to remove loosely adhered mortar from the RCA facet. Nevertheless, there was no discernible effect of the RCA's acid soak period on the quantity of the mortar loss. According to study findings, the ideal concrete compressive strength is produced when treated RCA is blended with concrete at a ratio of 45%.

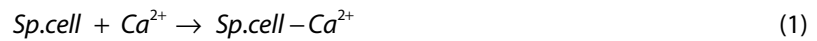
2.4. Two – stage mixture approach

In order to enhance the RCA quality, Tam et al. [15] created a mixing technique called the two-stage mixing approach (TSMA). The procedure is split into two sections, with the water being proportionately separated into two parts. These portions are then introduced after one part is blended with fine aggregate and the other with unrefined aggregate and cement. The standard technique of blending involves mixing all of the constituent parts of the concrete. Half of the water used for the first step of TSMA is used to cover the RCA facet with a thin layer of cement residue, which then seeps into the old cement mortar that is permeable and fills in the existing fractures. To complete the cement's hydration process, the remaining water is added in the following TSMA stage. In conclusion, for 20% of RCA, TSMA can increase strength by up to 21.19% throughout the course of 28 days of treatment.

2.5. Biodeposition of calcium carbonate

A different approach from TSMA is the biodeposition of calcium carbonate, as shown in Figure 2, used with *Sporosarcina pasteuni* bacteria. Unlike other techniques, biodeposition is natural and less damaging to the environment because the strain and all of the ingredients needed to produce the substrates are already present in the natural

world. This technique is predicated on the bacteria's capacity to produce calcium carbonate on the exterior facet of the cell wall as a result of the subsequent appearance of an appropriate negative zeta potential. The following is an illustration of the biodeposition process:



When carbonate ions $CaCO_3^{2-}$ from the breakdown of urea ($CO(NH_2)_2$) are attracted to the *Sporosarcina pasteurii* cell (Sp. cell), interactions with Ca ions Ca^{2+} are initiated. Ammonia ions NH^{4+} concurrently raise the pH of the surrounding medium, which enhances the calcite precipitation process's inherent efficiency. The results demonstrated a decrease in the aggregate's water absorption, and the drop was considerably more pronounced when finer fractions from the lower-quality concrete were employed [16].

2.6. Carbonation

An expedited carbonation process could partially enhance the bad quality of RCA given the components of the current cement mortar affixed to the RCA facet. It is conceivable that the old cement mortar connected to the RCA facet, which contains calcium hydroxide as one of its necessary byproducts of hydration, causes a reaction with carbon dioxide that has a noticeably larger volume, as the reaction demonstrates:



Other hydration products, such calcium silicate hydrate gel (CSH), can also be changed, yielding calcium carbonate, water, and an increased amount of polymerized silica gel or a lower Ca/Si ratio. The process of natural carbonation lowers the concrete's alkalinity, which leads to the corrosion of the steel reinforcement when reinforced concrete constructions are taken into consideration. As a result, the reinforced concrete constructions' durability is reduced. The most obvious effect of carbonation, however, is still the reduction in pore volume of concrete.

As demonstrated by the testing results, the RCA's mortar becomes denser following the CO_2 treatment process. Water soaking and porousness significantly decreased after the operation, according to RCA. More surface area allowed for easier carbonation of RCA with a smaller particle sizes. The RCA moisture level had a significant impact on the carbonation % because the saturated matrix's pores were saturated with water, which prevented CO_2 from penetrating, and there was insufficient water in the dry matrix to start carbonation reactions. In addition, the carbonation process proceeded quickly during the first two hours before clearly slowing down [17–22].

3. OWN EXPERIMENTAL RESEARCH

The recycled concrete aggregate utilized in our experiment came from the same concrete that was initially used to build the foundation of tram rails. Thus, there was no direct environmental exposure for the initial concrete. According to EN 206/2013, testing of the basic samples revealed that the concrete's compressive strength was in the C35/45 class. In order to obtain RCA with a maximum aggregate size of 22.4 mm, the original concrete has to be crushed. Original concrete made up 98% of the substance of the RCA, followed by asphalt at 2% and brick detritus at 0.8%.

Four fractions were identified from the recycled concrete aggregate that was obtained using the conventional sieving method: 0/4mm, 4/8mm, 8/16mm, and 16/22.4mm. An other estimate states that three-fraction concrete makes up more than 80% of Serbia's concrete production. Because of this, testing is limited to unrefined RCA (4/8mm and 8/16mm fractions), with the particle size distribution displayed in Figure 3.

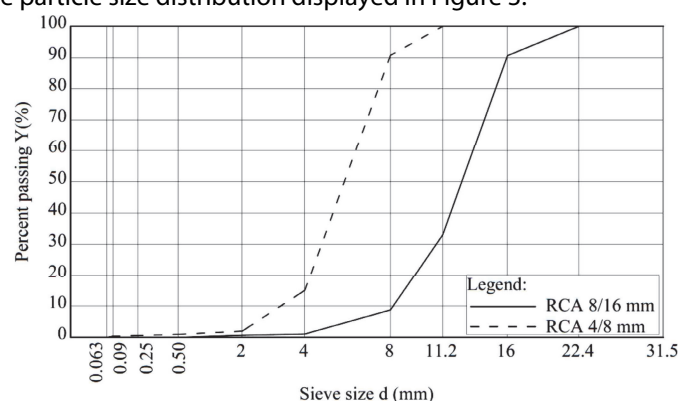


Figure 3: Particle size distribution of RCA

Table 1. contains data about chemical composition of the recycled concrete aggregate: both for primary aggregate and bonded mortar. It is clear from the Table that the natural aggregate in the primary concrete consists mostly of SiO₂ grains.

Table 1: Chemical composition of a representative sample of RCA

	Composition [%]						
	O	Mg	K	Ca	Al	Si	Fe
Natural aggregate	67.64	-	-	0.48	-	31.88	-
Adhered cement mortar	67.88	0.4	0.21	28.25	0.72	2.17	0.37

To enhance the quality of the recycled concrete aggregate, two processes were used: one including HCl and the other involving CO₂. RCA was immersed in a 0.1 mol/dm³ solution for a day as part of the HCl experiment, after which it was cleaned and dried at 100°C. The other RCA sample is used to determine the accelerated carbonation reaction.

Under constant conditions (CO₂ - 4%, temperature - 20°C, relative air humidity - 55%), the accelerated carbonation was carried out in compliance with the pre-standard (prCEN/TS 12390-12) until the sample mass became constant (after three weeks) [22]. Over the course of the entire experiment, the change in mass of the recycled concrete aggregate was monitored every 72 hours. By using the following calculation [24], it was possible to determine that the duration of the laboratory tests roughly equated to a year of carbonation under natural settings (CO₂ concentration up to 0.3% [23]), equation 5:

$$t_{NCT} = \frac{[CO_2]_{ACT}}{[CO_2]_{NCT}} \cdot t_{ACT} = \frac{4}{0.3} \cdot 21 = 280 \text{ days} \approx 1 \text{ year} \quad (5)$$

In which:

- t_{ACT} - the exposure time during the rapid test [days]
- t_{NCT} - the exposure time for samples exposed to a natural concentration of CO₂ [days]
- $[CO_2]_{ACT}$ - the concentration of CO₂ within the rapid test [%]
- $[CO_2]_{NCT}$ - the concentration of CO₂ under natural conditions [%]

The following tests are performed on all aggregate samples: pH value (EN 16192:2011/EN 12457 (1-4):2002), sulfate and chloride concentration (EN 16192/EPA 9056:2007), water absorption (EN 1097-6:2013), resistance to fraying by the Los Angeles method (EN 1097-2:2013), observable particle density, and the density of saturated and oven-dried aggregate.

4. RESULTS AND DISCUSSION

The obtained results within this research are shown in Table 2.

Table 2: Untreated and treated RCA: physical and mechanical properties

		RCA		RCA _(HCl)		RCA _(CO₂)	
		4/8 mm	8/16 mm	4/8 mm	8/16 mm	4/8 mm	8/16 mm
ρ_a	(kg/m ³)	2618	2699	2681	2654	2667	2657
ρ_{ssd}	(kg/m ³)	2499	2530	2547	2525	2522	2523
ρ_{rd}	(kg/m ³)	2426	2431	2466	2448	2435	2441
WA	(%)	3.0	4.1	2.9	4.0	2.6	3.3
LA	(%)	31.5		32.0		28.4	
Δm_1	(%)	-	-	-2.3	-2.2	0.9	1.4
Δm_2	(%)	-	-	-4.3	-2.9	0.9	1.4

In Table 2, the following symbols are used:

- ρ_a (kg/m³) – apparent particle density,
- ρ_{ssd} (kg/m³) – saturated and surface-dried particle density,
- ρ_{rd} (kg/m³) – oven-dried particle density,
- WA (%) – water absorption,
- LA (%) – Los Angeles coefficient,
- Δm_1 (%) – percentage of change in mass after treatment (acid or carbonation),
- Δm_2 (%) – percentage of change in mass after treatment and sieving.

After the HCl treatment, the physical characteristics of the RCA revealed a reduction in water absorption of 33.3% and 2.50% for the portions 4/8mm and 6/16mm, respectively. Concurrently, there was an increase in RCA density (by 1.65% for fraction 4/8mm and 2.90% for fraction 8/16mm) when compared to the untreated aggregate. As a result, following HCl treatment, the aggregate's mass loss was 4.30% for fraction 4/8mm and 2.90% for fraction 8/16mm, which is similar to the findings of study [25]. The water soaking and aggregate density reversely depended upon the partial extraction of the permeable cement mortar, but the carbonation process produced noticeably better results, with water soaking increasing by 13.3% for the 4/8mm fraction and 20% for the 8/18mm fraction—again, in line with the findings of the study [26]. Additionally, an increase of 1.40% and 0.40%, respectively, in the oven-dried densities for fractions 4/8mm and 8/16mm was found. After the carbonation process was completed, fraction 4/8mm recorded a 0.90% rise in the aggregate mass overall, but fraction 8/16mm showed a 1.40% increase. The carbonation products produced in the microcracks in the cement mortar led to the creation of these unique RCA characteristics.

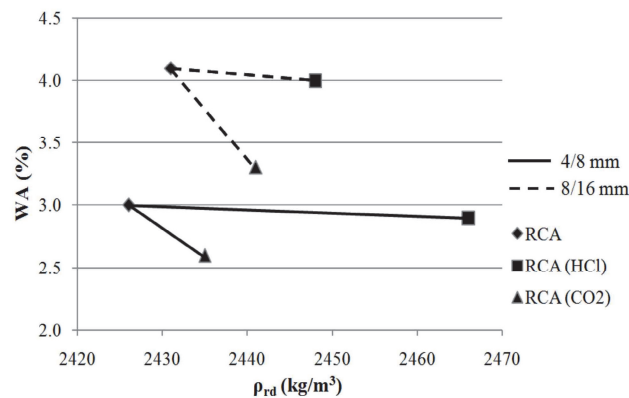


Figure 4: The relationship between RCA density and water absorption trends

When the treatment is applied, Figure 4 illustrates the relationship between the density of RCA and water soaking. The inference could be that, whether or not treatments are used, the water soaking increases as the aggregate fraction grows. The propensity of water absorption to diminish is the same for both portions.

The results demonstrate that all three RCA types have acceptable quality (LA coefficient below or close to 30%) after assessing the mechanical characteristics of the RCAs based on measuring the resistance down to fragmentation. However, there was no discernible difference between the acid-treated and untreated aggregate, indicating that the natural aggregate's grains and the old cement mortar had a good level of adhesion. Comparing the carbonated RCA to the untreated RCA, it was found that the carbonated RCA had aggregate resistance to fragmentation that was 10% higher. The chemical characteristics of RCA determined both before and after the treatment are shown in Table 3.

Table 3: Untreated and treated RCA: chemical properties

	RCA		RCA _(HCl)		RCA _(CO2)	
	4/8 mm	8/16 mm	4/8 mm	8/16 mm	4/8 mm	8/16 mm
pH value	10.7	11.8	10.6	11.5	9.5	9.8
Concentration of Cl ⁻ (mg/kg)	177	242	214	533	175	271
Concentration of SO ₄ ²⁻ (mg/kg)	1212	605	514	330	1008	576

Potentially harmful composites with concentrations of sulfate and chloride ions were managed. These composites have the ability to significantly deteriorate the quality of concrete by causing steel reinforcing to corrode. As expected, the HCl treatment significantly increased the chloride concentration in the aggregate, rising to 20.9% for fraction 4/8mm and even 120.2% for fraction 8/16mm. Simultaneously, this treatment resulted in a noticeable 50% decrease in sulfate concentration; the reason for this can be attributed to the dissolution and extraction of the old cement mortar. This was also validated by other researchers [5]. Regarding the carbonation process, it essentially had no impact on any variations in the RCA's concentration of sulfate or chloride. The pH value did not significantly alter throughout the HCl experiment; nevertheless, the carbonation process decreased it for both the 4/8mm and 8/16mm portions [27].

CONCLUSIONS

This study investigated the mechanical, chemical, and physical characteristics of three unrefined RCA types: RCA treated to fast carbonation, RCA pre-soaked in HCl, and RCA without any treatments. The carbonation treatment was intended to tighten the cement matrix, while the HCl procedure was intended to remove the bonded cement mortar

piece by piece. The unrefined RCA experienced a 2-3% drop in water soaking after being treated with HCl acid, while the density increased by 1% and the mechanical characteristics stayed the same. In terms of the chemical testing, the treatment with HCl resulted in an increase in the concentration of chloride, a decrease in the concentration of sulfate, and an unaltered pH level. Conversely, a quick carbonation process produced superior results in terms of the mechanical characteristics (~10%) and the decreased water soaking feature. The aggregate's contents of sulfate and chloride did not change, density increased negligibly, and pH level dropped. The SEM results validated the experimental research findings, and the photos demonstrated that the RCA fractures were no greater than 2 μm after the carbonation process was completed.

Regarding how RCA density and water absorption interacted, the greater the aggregate fraction (8/16mm) was relative to the smaller fraction (4/8mm) in terms of water absorption, regardless of the treatment used. With both RCA fractions, the drop trend lines for water absorption were comparable.

It should be noted that the Los Angeles coefficient values were utilized to validate the study's usage of unprocessed RCA, which was obtained from compressing high-quality concrete (C35/45 class of compressive strength). The study also demonstrated that there is no need for quality improvement measures when treated RCA is utilized. All of which points to the general conclusion that this kind of RCA could be easily applied while making cement or asphalt concrete without the need for pretreatment of the concrete.

In conclusion, the study sought to enhance the quality of RCA and facilitate its broader application in the construction sector, considering the perspective of environmental appreciation. There has been increasing pressure in recent years to increase the use of alternative building materials throughout Europe, including Serbia. RCA is a great way to save natural resources, store CO₂, and reduce its emissions while also having a very positive and aesthetically pleasing impact on the environment.

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