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Investigating the Mechanical and Durability Properties of Carbonated Recycled Aggregate Concrete and Its Performance with SCMs

Shimza Jamil [1], Maria Idrees [2,3,*], Arslan Akbar [4,*] and Wisal Ahmed [5]

[1] Department of Building and Architectural Engineering, Faculty of Engineering and Technology, Bahauddin Zakariya University, Multan 60600, Pakistan; shimzajamil@bzu.edu.pk
[2] Department of Civil, Construction and Environmental Engineering (CCEE), University of Alabama, Tuscaloosa, AL 35401, USA
[3] Department of Architectural Engineering, University of Engineering and Technology, Lahore 54000, Pakistan
[4] Department of Civil and Environmental Engineering, Pennsylvania State University, 231M Sackett Building, University Park, PA 16802, USA
[5] Department of Architecture and Civil Engineering, City University of Hong Kong, Hong Kong, China; wahmed7-c@my.cityu.edu.hk
* Correspondence: mariaidrees@uet.edu.pk or midrees3@ua.edu (M.I.); aba6457@psu.edu (A.A.)

Abstract: Utilizing recycled aggregates (RAs) in concrete production represents a promising path toward sustainability; however, it often results in reduced physical and durability properties. The weak interfacial transition zone (ITZ) and the adhered mortar in recycled aggregate concrete (RAC) contribute to lower mechanical strength and limit its application in demanding environments. This study investigates an accelerated carbonation technique to enhance the mechanical and durability properties of RA and RAC. Recycled aggregates, with a particle size of 10–20 mm, were subjected to carbonation at 1 bar for 2 h in a controlled carbonation chamber. Results demonstrate substantial improvements in the compressive and split tensile strengths of the carbonated recycled aggregate concrete (CRAC), with increases of 30% and 42% compared to conventional RAC, respectively. The CRAC mix also exhibited a 1.5% increase in dry density and reduced water absorption (6%) compared to RAC (7.58%). After 90 days of acid exposure, compressive strength reductions of 48.85% and 37.9% were observed for RAC and CRAC mixes, respectively, from their 28-day strength, while weight loss in CRAC was limited to 5.4%, compared to 10.92% in RAC. In sulfate exposure tests over 150 days, RAC and CRAC showed compressive strength reductions of 31.4% and 19.7% and weight losses of 3.6% and 2.2%, respectively, indicating enhanced resistance of CRAC to harsh environments. However, CRAC blended with supplementary cementitious materials (SCMs) showed diminished mechanical properties, likely due to a reduced alkaline environment caused by rapid calcium hydroxide absorption in RA pores during carbonation. Overall, the findings highlight the practical potential of accelerated carbonation to improve the performance of RAC, offering a viable pathway for sustainable construction applications.

Keywords: recycled aggregate concrete (RAC); carbonated recycled aggregate concrete (CRAC); mechanical properties; durability properties; supplementary cementitious materials (SCMs)

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1. Introduction

Concrete is considered the most widely utilized building material, with an annual production of approximately 30 billion tons worldwide [1]. Generally, concrete contains between 45% and 65% natural coarse aggregates, and unsurprisingly, natural aggregates

(NAs) rank as the second most employed natural resource [2]. Nevertheless, the excessive exploitation of such naturally occurring resources has great environmental and ecological consequences, such as the overuse of natural aggregates, which has led to the substantial consumption of energy and carbon dioxide (CO_2) emissions [3–5]. Aggregate mining has a profound environmental footprint, leading to the depletion of finite resources and causing ecosystem degradation through erosion, deforestation, and riverbed damage [2,6]. Typically, conventional concrete structures have a specific lifespan, necessitating reconstruction and demolition [7]. There has also been a significant rise in the generation of construction and demolition waste (CDW) through accelerated urbanization and economic development, particularly in developing regions and countries [8]. In 2016, the worldwide production of CDW reached 2.01 billion tons annually, a figure that may exceed 3.4 billion tons by 2050 [9]. Employing recycled waste materials in construction promotes economic benefits and supports eco-friendly building practices [10]. Utilizing recycled concrete aggregates to produce normal concrete represents a method for effectively consuming construction and demolition waste (CDW). Reusing and recycling aggregates can offer substantial benefits for sustainability [11]. However, property levels of recycled aggregate concrete (RAC) are generally lower due to the adhered mortar stuck to the surface of recycled concrete aggregates, limiting its application to some extent [12]. Usually, they have lower density and strength than natural aggregates (NAs) [13]. The hardened mortar on recycled aggregate (RA) particles, being more porous than natural aggregates, results in higher water absorption [14]. As per previous studies, for 100% replacement, the compressive strength of RAC can be 10–25% [15,16] reduced compared to that of normal concrete. Split tensile strength may be reduced by 10–35% [17,18] and 33% [19] when using 100% RCA. In addition, global warming has recently become a significant concern due to the ongoing temperature rise, which is linked to increased greenhouse gas emissions. Over the past two centuries, the concentration of carbon dioxide (a major greenhouse gas) has increased by approximately 50% [20]. The construction industry has been recognized as a major contributor to carbon dioxide (CO_2) release, accounting for about 33% of global CO_2 emissions. As stated in a UNEP report [21], carbon dioxide generated by the construction industry reached its highest level in 2022, at around ten gigatons. This represents a 5% increase compared to 2020. Cement is one of the leading sources of construction materials, releasing CO_2, and has a massive carbon footprint [22,23]. Researchers suggested that the most practical approach to minimizing the carbon footprint of cement is by increasing the substitution percentage of supplementary cementitious materials (SCMs) [24]. Sustainability initiatives should prioritize minimizing the carbon footprint and lowering the depletion of natural resources [25]. The primary objective of sustainable development is reducing environmental degradation and conserving natural resources, which are depleting rapidly. Green and eco-friendly construction can be accomplished by minimizing the utilization of natural resources to the greatest possible extent [26].

From the above discussions, employing the accelerated carbonation technique on RAs is one of the new methods that is a sustainable and environmentally friendly approach to alleviating inferior properties of RAs. The carbonation mechanism involves the formation of calcium carbonate ($CaCO_3$), a product of carbonation reaction produced when CO_2 reacts with hydrates like portlandite or calcium silicate hydrate found in the cement paste bonded to the natural aggregates [27], and precipitation within the pores and cracks of RAs, improving its microstructure. Enhancing the individual properties of RAs also leads to better overall performance of RAC. Recently, there has been a growing need to utilize carbon dioxide to strengthen RA and sequester it [28], which enhances both old and new ITZ. By natural carbonation, CO_2 emissions associated with concrete can be reduced by approximately one-fifth. Meanwhile, around 0.5 tons of carbon dioxide can be sequestered through 1 ton of cement

adhered to RAs [29]. Moreover, the accelerated carbonation on RAs can offset around 9% of carbon released through concrete production as it can capture CO_2 of about 7.9 kg/ton [30].

Meanwhile, durability issues of concrete structures can lead to severe damage and deterioration [31]. Durability plays a crucial role in determining the lifespan of concrete structures. Durable concrete maintains its appearance, strength, and integrity throughout its service life when subjected to adverse environmental conditions. Alongside the mechanical properties of RAC, it is crucial to evaluate the durability properties of RAC to determine their potential for use. The durability of RAC is compromised due to old mortar on the RA surface. Water can be readily released and absorbed during the hydration process of cement due to the increased porosity of RAs [32]. The porous nature of RA particles leads to more water absorption that can harm concrete's durability properties. Pores and cracks in RCA may introduce chlorides, sulfates, and other organic materials into the concrete mix [33,34]. Therefore, it is crucial to investigate methods to improve the quality of RAs.

The current research aims to provide sustainable alternatives to conventional construction materials, addressing environmental and resource scarcity challenges. Carbon capture is vital to enable the utilization of RAs in structural concrete to support sustainability and ensure environmental protection. Therefore, the study will focus on implementing accelerated carbonation treatment on RAs to develop sustainable concrete. Calcium carbonate ($CaCO_3$) formation in cementing materials like adhered mortar offers significant potential to enhance the properties of RAs. Moreover, research also highlights the long-term effects of the carbonation treatment of RAs on concrete properties, an area that has been insufficiently explored in prior studies. Therefore, this paper contributes to the current literature addressing the durability performance of carbonated recycled aggregate concrete under harsh environments which assists in filling the research gap. The present study introduces an innovative blend of carbonated recycled aggregates and supplementary cementitious materials (SCMs) to investigate the performance of concrete. It intends to examine the physical properties of carbonated and non-carbonated recycled aggregates. Furthermore, the mechanical and durability properties of NAC, RAC, and CRAC mixes were also explored. The mechanical properties, including compressive strength, split tensile strength, and concrete dry density, were evaluated for NAC, RAC, and CRAC mixes. The durability properties of the under-study concrete samples related to water absorption (WA), sulfuric acid attack (SAA), and sulfate attack resistance (SAR) were experimentally explored. Performance regarding residual compressive strength and change in weight were assessed against sulfuric acid and sulfate environments. Additionally, the current study also emphasized examining the mechanical properties of concrete blended with partially incorporated supplementary cementitious materials (SCMs), including silica fume (SF), zeolite powder (ZP), and limestone powder (LSP), either individually or in combination with CRAs.

2. Experimental Methodology

The following subsections present details of the materials used, concrete production procedures, and concrete tests conducted. Figure 1 shows a flow diagram of the proposed work of this paper.

2.1. Materials

2.1.1. Binders

Commercially accessible, ordinary Portland cement (OPC) named Bestway (specific gravity = 3.15) with ASTMC150 Type1 (42.5 N) was used. Silica fume (SF), a pozzolanic supplementary cementitious material (SCM), grayish black, produced as a by-product of the ferrosilicon alloys industry, was utilized as a 10% cement replacement material. Fine zeolite powder (ZP), a naturally existing pozzolanic material, was employed as a

5% cement replacement material, whereas coarser ZP was added as a 5% fine aggregate replacement material in concrete preparation. Limestone powder (LSP), a ground limestone, was incorporated as a 10% replacement material for cement. The chemical properties of OPC, SF, ZP, and LSP are mentioned in Table 1.

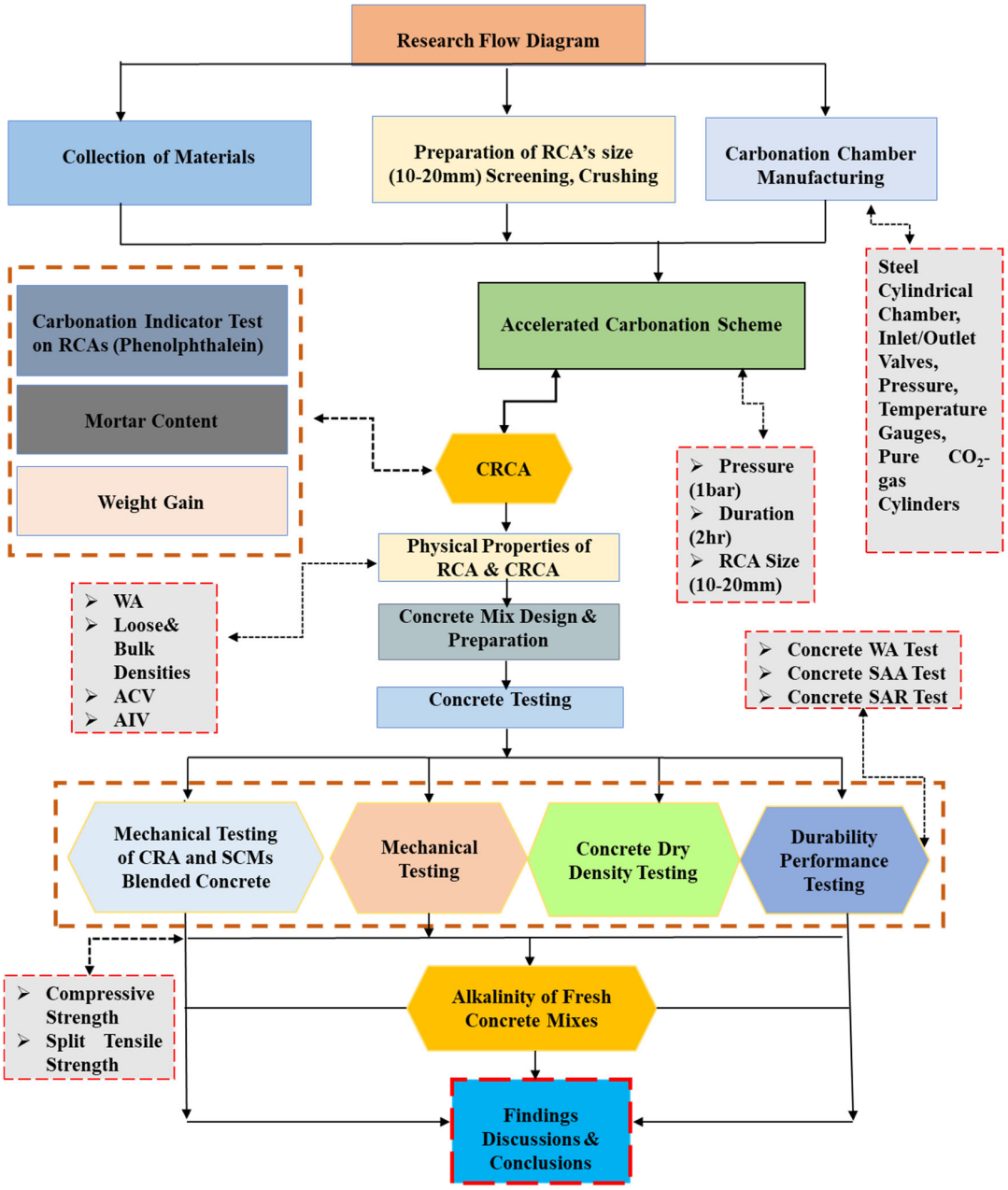


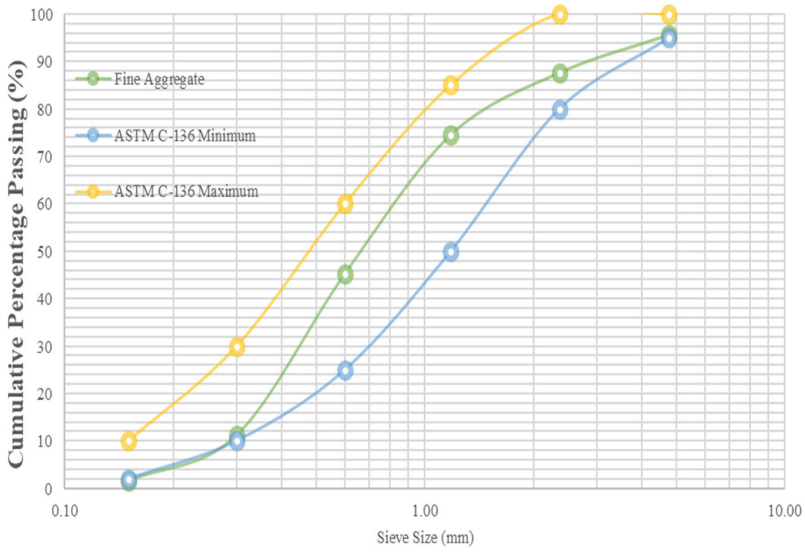
Figure 1. Overview of the research methodology: Step-by-step process illustrating the experimental design.

Table 1. Chemical composition of OPC, SF, ZP, and LSP.

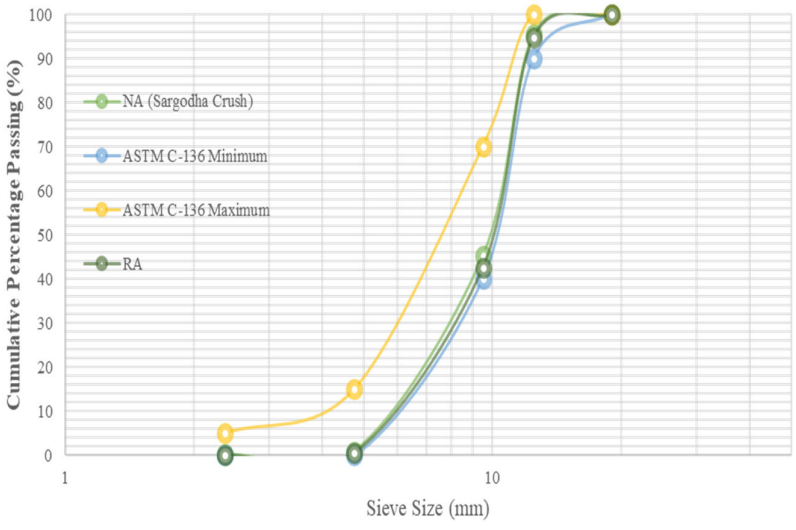
Chemical Composition (Oxides)	OPC%	SF%	ZP%	LSP%
Lime (CaO)	60.30	0.95	1.25	39
Silica (SiO_2)	19.70	92.17	82.15	1.89
Alumina (Al_2O_3)	5.17	0.45	2.25	0.85
Iron Oxide (Fe_2O_3)	3.48	0.22	1.12	-
Magnesia (MgO)	2.70	0.10	0.80	0.55
Sulfur Trioxide (SO_3)	2.48	0.28	0.031	-
Loss on Ignition (LOI)	4.95	4.59	3.75	35.2

2.1.2. Aggregates

A pure siliceous sand named Lawrencepur, with a fineness modulus of 2.8, was used as a fine aggregate, and its gradation curve is shown in Figure 2. A 1020 mm limestone aggregate was obtained from the Sargodha quarry in Pakistan and utilized as natural aggregate (NA). Moreover, RAs were produced from cracked or discarded pre-cast members. First, those members gathered and stored outdoors in unprotected conditions for some weeks. Meanwhile, those were broken into small chunks manually and were crushed and screened to achieve RAs with a size of 10–20 mm. Then, RAs were kept in sealed plastic bags in the laboratory at room temperature for months before being used for carbonation and concrete production. Figure 3. presents the granulometry of FA, NA, and RA.



(**a**)



(**b**)

Figure 2. Granulometry of (**a**) fine aggregate and (**b**) coarse aggregates.

The physical properties of NA, RA, and CRA are mentioned below in Table 2. For carbonated recycled aggregates (CRAs), water absorption, aggregate impact, and crushing values were decreased, and an increase in bulk density was noticed compared to non-carbonated RAs owing to the precipitation of calcium carbonate ($CaCO_3$) within the pores of RA. $CaCO_3$ had a greater density than portlandite, resulting in improved physical properties of CRA, as concluded in previous studies [35,36].



Figure 3. Accelerated carbonation chamber for RAs.

Table 2. Properties of aggregates.

Sr.#	Material Property	Fine Aggregate	Coarse Aggregate		
		-	NA	RA	CRA
1	Max. aggregate size (mm) (ASTM C-136) [37]	4.75	19	19	19
2	Min. aggregate size (mm) (ASTM C-136) [37]	-	9.5	9.5	9.5
3	Fineness modulus (ASTM C-136) [37]	2.8	6.56	6.61	-
4	Water Absorption (%) (ASTM C-127/128) [38]	1.26	0.72	5.8	3.0
5	Loose bulk density (kg/m^3) (ASTM C-29) [39]	1670	1490	1377	1443
6	Rodded bulk density (kg/m^3) (ASTM C-29) [39]	1850	1630	1489	1567
7	Aggregate impact value (%) (BS 812-112) [40]	-	13.8	20.1	16.5
8	Aggregate crushing value (%) (BS 812-110) [41]	-	20.9	26.8	22.8

2.2. Accelerated Carbonation of RCA

2.2.1. Accelerated Carbonation Chamber

In the present work, a cylindrical carbonation chamber was manufactured, made up of stainless steel, with a size of 250 mm by 375 mm (Figure 3). The carbonation chamber was equipped with a gas inlet and outlet valves along with eight bolts at its outer lid. Pressure and temperature gauges were also mounted to it. The chamber was designed to withstand a pressure range of up to 7.0 bar. Before initiating the carbonation into RAs, any unwanted

gas or air was expelled from the chamber by introducing 2–3 cycles of CO_2 gas at a rate of 3 bar pressure. The pneumatic valve connected to the CO_2 cylinder controlled the gas pressure within the chamber. Current research maintained a carbonation pressure in the chamber at 1 bar for the duration of 2 h. Temperature and relative humidity were kept at 23 ± 5 °C and 60 ± 5%, respectively. Cylinders containing pure CO_2 gas (99.9%) were connected to the chamber. Continuous monitoring was performed during carbonation to ensure that the pressure, temperature, and humidity levels were in said ranges. The phenolphthalein indicator test was performed before and after RA carbonation and can be shown in Figure 4.



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

Figure 4. RA's pH test (**a**) before and (**b**) after carbonation.

2.2.2. Accelerated Carbonation Scheme

A recent study determined that the optimal carbonation regime for RAs was a pressure of 1 bar for 2 h. This was more effective than pressures of 0.5 and 2 bars applied for either 2 h or 4 h [36]. Thus, an accelerated carbonation scheme as mentioned in Table 3 was performed on RAs to evaluate the mechanical and durability properties of concrete mixes and their performance along with blends of SCMs.

Table 3. Accelerated carbonation scheme.

Carbonation Pressure (bar)	Carbonation Duration (h)	Relative Humidity (%)	Temperature (°C)
1	2	60 ± 5	25 ± 3

3. Experimentation

3.1. Mortar Content

The below procedure was performed to calculate the mortar content adhered to with RAs.

Initially, RA samples were immersed in clean water for 2 h, denoted as (M0). Once fully saturated, these samples were subsequently dried in an oven at 250 °C for 3 h. Following this, all RA samples were promptly submerged in cold water. Consequently, this rapid cooling induced thermal stresses in RA samples, causing cracks between the original aggregate and adhered mortar. This mortar was removed by rubbing the RAs against a 2.36 mm steel mesh, and the weight of the RAs obtained above the steel mesh after this procedure was designated as (M1). The following equation calculated the percentage of adhered mortar content on RAs.

$$\text{Adhered mortar content}(\%) = \frac{\text{M0} - \text{M1}}{\text{M0}} \times 100 \quad (1)$$

It has been found that around 39.5% of the mortar content was adhered to RA samples.

3.2. Weight Gain

Table 4 demonstrates the change in weight of recycled aggregates (RAs) before and after accelerated carbonation. The initial weight of RAs before carbonation (w_{bc}) and the final weight of RAs after carbonation (w_{ac}) were utilized to calculate the CO_2 absorption as presented in Equation (2) [42]; around 0.21% of the mass was increased due to the formation of calcium carbonate in pores.

$$CO_2 \text{ Uptake} = \frac{\text{Wbc} - \text{Wac}}{\text{Wbc}} \quad (2)$$

Table 4. Weight gain of CRAs.

Sample	Original Weight Before Carbonation (gm)	After Carbonation Weight (gm)	Change in Weight	Percentage Change %
CRA	225.0	225.49	0.49	0.21

3.3. Concrete Mix Composition

The concrete mix composition was prepared to have a w/b ratio of 0.42 and 40 MPa targeted concrete strength to investigate the mechanical and durability properties of RAC and CRAC samples, as presented in Table 5. Moreover, the concrete samples, including carbonated recycled aggregates blended with SCMs, were prepared following the design mix with 0.55 and 0.42 w/b ratios mentioned in Table 6. Both concrete mix compositions were within the guidelines of ACI 211.1. According to the concrete strengths and water-to-binder ratios (0.55 and 0.42), the amount of mixing water was 195 kg/m^3 and 216 kg/m^3, respectively. To improve the workability of recycled aggregate concrete (RAC), a superplasticizer (SP), Chemrite SP-303, was used to attain the desired workability, supplied by a company; Imporient Construction Chemicals Pvt. Ltd. (Lahore, Pakistan). It was a pale whitish liquid due to its carboxylic acid-based composition SP dosage was varied between 0.5% to 1.5% by weight of cement.

Table 5. Concrete mix composition.

Sr.#	Nomenclature	RA	*w/b*	Cement	Water	Fine Aggregate	Coarse Aggregate	RA	SP
	-	(%)	-	(kg/m^3)					(%)
1	NAC	0	0.42	515	216	684	967	0	0.75
2	RAC	100	0.42	515	216	684	0	967	0.75
3	CRAC	100	0.42	515	216	684	0	967	0.75

Table 6. Concrete mix composition of CRAC blended with SCMs.

Sr.#	Nomenclature	*w/b*	Cement	SF	ZP	LSP	Water	Fine Aggregate	RA	SP
	-	-	(kg/m^3)							(%)
1	RA-0.55	0.55	355	0	0	0	195	505	940	1
2	CRA-OPC-0.55	0.55	355	0	0	0	195	505	940	1
3	CRA-10SF-0.55	0.55	319.5	35.5	0	0	195	505	940	1
4	CRA-10SF-10ZP-0.55	0.55	301.75	35.5	17.75	0	195	487.25	940	1
5	RA-0.42	0.42	515	0	0	0	216	684	967	0.5
6	CRA-OPC-0.42	0.42	515	0	0	0	216	684	967	0.5
7	CRA-10SF-10LSP-0.42	0.42	412	51.5	0	51.5	216	684	967	0.5

3.4. Testing Procedures and Sample Preparation

3.4.1. Mechanical Properties

Mechanical properties of NAC, RAC, and CRAC mixes were investigated after 28 curing days using a universal testing machine (UTM). Compressive strength was evaluated by testing 100 mm × 100 mm × 100 mm cubes in compliance with BS EN 12390-3 [43]. Tensile strength was determined by testing cylinders with a diameter of 100 mm and a height of 200 mm following ASTM C-496 [44].

3.4.2. Concrete Dry Density

Concrete dry density was calculated after 28 days of curing. It was calculated by averaging the weights of four cubes and then dividing it by the volume of one cube. This test assesses the compactness of concrete. Having higher compactness corresponds to increased density and less air voids.

3.4.3. Durability Properties

Details of the durability tests of NAC, RAC, and CRAC mixes regarding water absorption (WA), sulfuric acid attack (SAA), and sulfate attack resistance (SAR) are given below.

Concrete Water Absorption (WA)

The concrete WA test determines the permeable and porous nature of concrete by measuring the amount of water absorbed through its surface. This test is essential for evaluating the durability of concrete under different environmental conditions. WA tests on NAC, RAC, and CRAC samples were performed in the laboratory following ASTM C-642 [45]. As shown in Figure 5. concrete discs with a 50 mm thickness and 100 mm diameter were used.



Figure 5. Concrete water absorption (WA) test.

Sulfuric Acid Attack (SAA)

To investigate the resistance of concrete samples against sulfuric acid attack (SAA). An exposure solution was prepared by mixing 5% dilute sulfuric acid (H_2SO_4) with 900 mL of water. Cube samples were then immersed in the solution for 90 days. Residual compressive strength was determined after 90 days of immersion in an acid environment, as shown in Figure 6. The percentage weight loss for each concrete mix was evaluated by comparing the weight of 28 days of cured samples before immersion in acid solution with the weight of the sample after 90 days of exposure to H_2SO_4 solution.



Figure 6. Sulfuric acid attack test after 90 days.

Sulfate Attack Resistance (SAR)

The sulfate attack resistance (SAR) was conducted on 100 mm blocks of NAC, RAC, and CRAC in compliance with ASTM C-1012-04, as depicted in Figure 7. An exposure solution was prepared with 50 g sodium sulfate (Na_2SO_4) dissolved in 900 mL of water and further diluted with additional water to acquire 1.0 L of solution. Cube samples were immersed in the solution, and the residual compressive strength and percentage of weight change in concrete samples were calculated after 150 days of exposure.



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

Figure 7. Concrete samples for sulfate attack (**a**) before and (**b**) after 150 days.

3.4.4. Mechanical Properties of CRA and SCM Blended Concrete

Mechanical properties for CRA and SCM blended concrete, including compressive and tensile strength, were evaluated after 28 curing days with the assistance of a UTM.

3.4.5. Alkalinity of Fresh Concrete Mixes

Alkalinity or acidity for NAC, RAC, and CRAC, readily mixed samples were assessed using pH strips (paper).

4. Findings and Discussion

4.1. Mechanical Properties

The mechanical properties of NAC, RAC, and CRAC were investigated. The following sections present the findings and discussions regarding compressive and tensile strengths.

4.1.1. Compressive Strength

The compressive strengths of NAC and RAC mixes were 40 MPa and 28 MPa, respectively. RAC showed a 30% strength reduction compared to NAC (Figure 8). This

reduction was likely owing to the inferior properties of RAs that may have been produced during demolition and crushing processes. However, the CRAC (1 bar-2 h) exhibited a 30% improvement approximately in compressive strength (36.4 MPa) compared to RAC (28 MPa). It can be attributed to $CaCO_3$, which is formed in the carbonation process. Consequently, the interfacial transition zone (ITZ) is enhanced between the cement matrix and aggregates, which leads to improved compressive strength, as concluded in recent research [36]. CRAC's performance also depicted comparable results other than RAC when compared to other advanced concretes. A concrete mix prepared with the shared incorporation of carbon fibers and silica fume-treated aggregate concrete showed a 30% improvement in compressive strength [46].

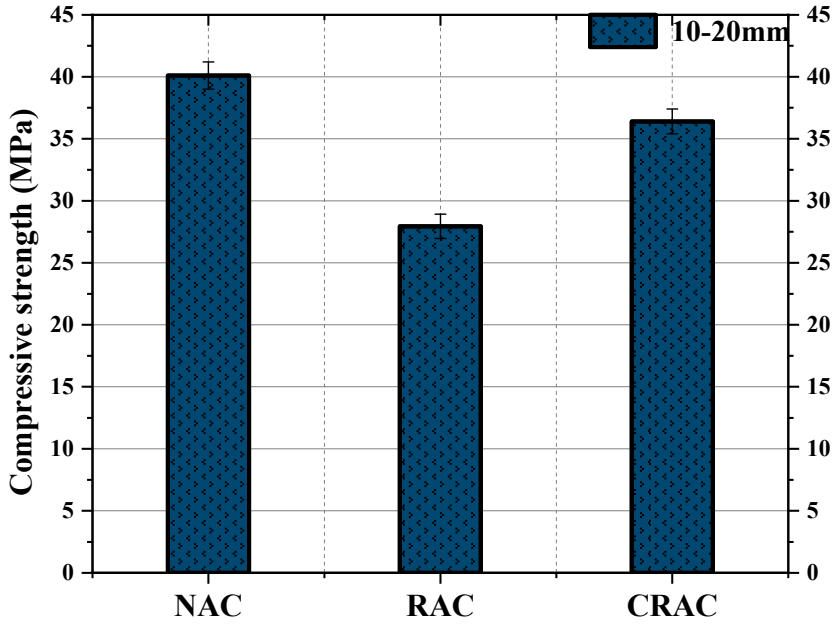


Figure 8. Compressive strength test results of NAC, RAC, and CRAC.

4.1.2. Tensile Strength

The tensile strength of the NAC mix showed a maximum strength of 4.81 MPa, whereas the strength of the RAC mix was 3.05 MPa (Figure 9). RAC found a tensile strength reduction of approximately 36.59% compared to NAC. The lower strength of RAC was because of the porous nature and weak interfacial transition zone (ITZ) of RAs. In contrast, the tensile strength of the CRAC mix was 4.35 MPa, indicating an improvement of strength by 42.62% compared to the RAC mix. The better tensile strength of the CRAC mix confirmed that the porous structure of RA was enhanced through the carbonation process. The presence of $CaCO_3$ refined the pores and consequently, better tensile strength was achieved. This implied that the carbonation treatment of RAs compensated for its tensile strength limitations. Moreover, the performance of CRACs was found to be equal to or more than that of other advanced concretes in terms of split tensile strength. About 40% of improvement was shown by both carbon fibers and the silica fume-treated slurry aggregate concrete [46].

4.2. Concrete Dry Density

The dry density of normal concrete generally ranges from 2400 kg/m^3 to 2900 kg/m^3, depending upon the water–binder ratio, the type of aggregates being used, and the curing conditions. Previous studies have found that the dry density of NAC, RAC, and CRAC mixes was between 2500 and 2600 kg/m^3, 2300 and 2500 kg/m^3, and 2500 and 2590 kg/m^3, respectively [36].

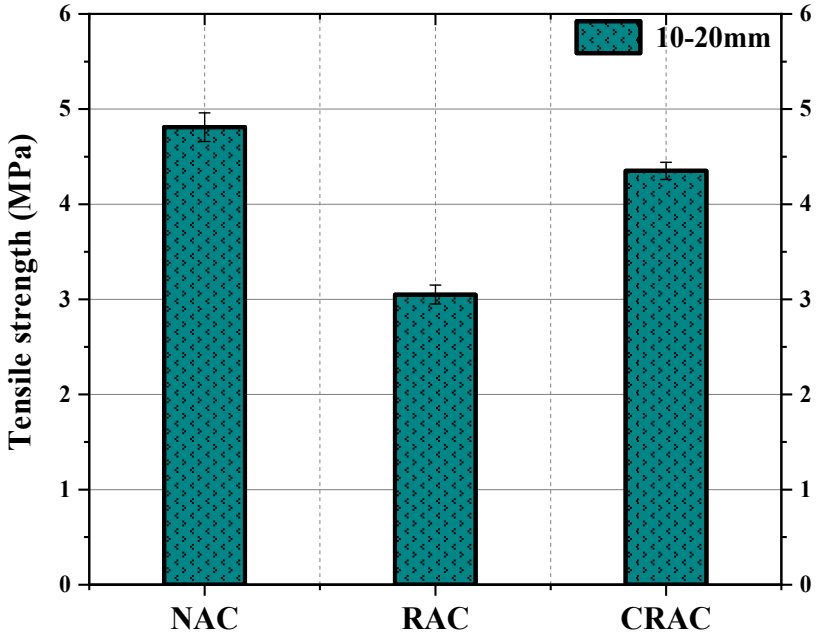


Figure 9. Tensile strength test results of NAC, RAC, and CRAC.

In the current study, the RAC mix revealed a dry density of 2502.3 kg/m^3, about 3.6% less than the NAC mix (2598 kg/m^3) owing to its porous structure. It can be shown in Figure 10. that the CRAC mix (1 bar-2 h) had comparatively about 1.5% more dry density, measuring 2540 kg/m^3 when compared to the RAC mix (2502.3 kg/m^3). Calcium carbonate was produced during the carbonation process and had comparatively more density than calcium hydroxide. Consequently, it resulted in the concrete matrix that became denser and compacted. It is clear from the above results that carbonation can homogenize the surface of RAs, leading to an improved packing of particles thus developing denser concrete.

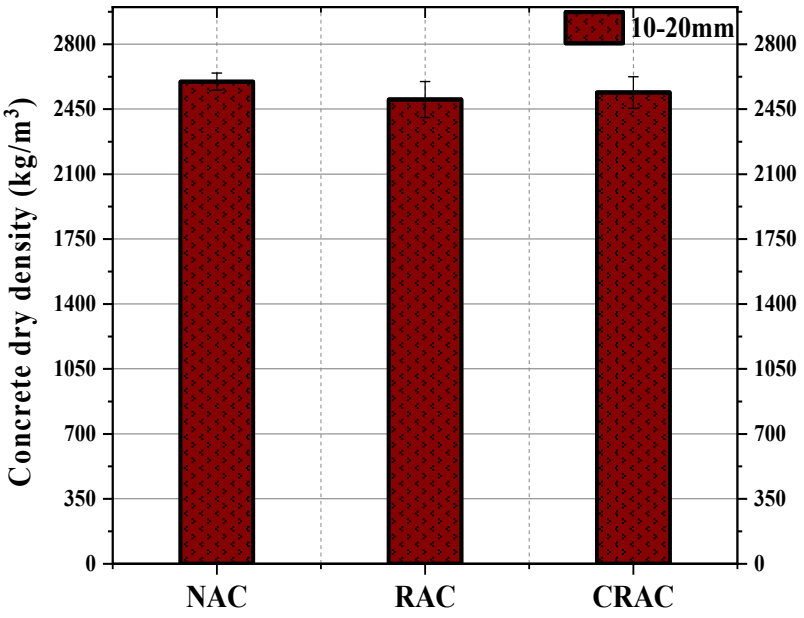


Figure 10. Concrete dry density test results of NAC, RAC, and CRAC.

4.3. Durability Properties

The durability properties of NAC, RAC, and CRAC mixes were evaluated. The following sections present the findings and discussions.

4.3.1. Concrete Water Absorption (WA)

The water absorption of concrete mixes, i.e., NAC, RAC, and CRAC, was 4.6%, 7.58%, and 6.06%, respectively, as shown in Figure 11. This indicated that the formation of $CaCO_3$ after the carbonation of RAs reduced porosity and improved the new ITZ, thus reducing the water absorption capacity of the CRAC mix. Therefore, it can be stated that the properties of RAs were enhanced by employing accelerated carbonation.

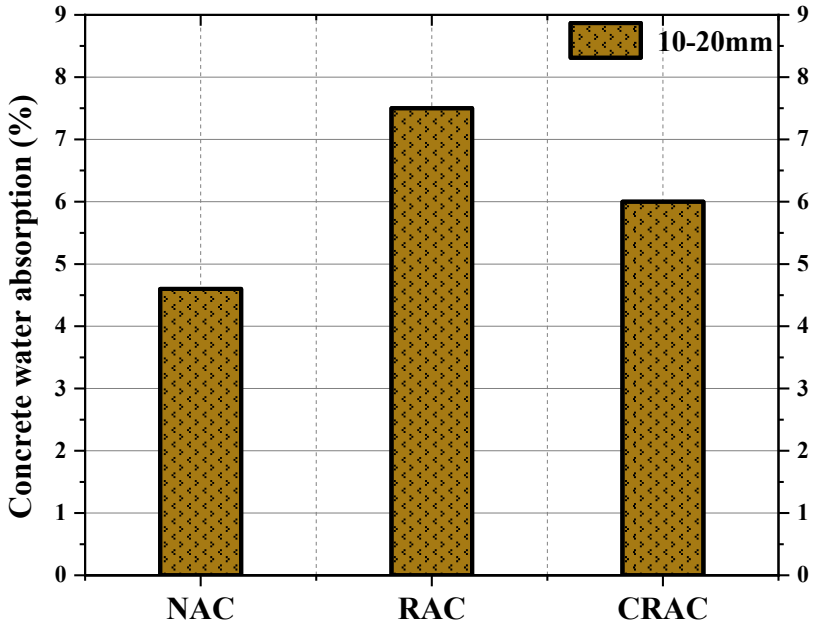


Figure 11. Concrete water absorption test results of NAC, RAC, and CRAC.

4.3.2. Concrete Sulfuric Acid Attack (SAA)

Loss in Compressive Strength

Acid exposure can endanger concrete service life due to various alkaline by-products from cement hydration. The most destructive acid reacts with these alkaline substances to produce salts, rapidly deteriorating concrete structures [46]. The percentage loss of compressive strength for NAC, RAC, and CRAC due to 90 days of exposure to sulfuric acid (H_2SO_4) is demonstrated in Table 7. Residual compressive strength for all under-study concrete samples is presented in Figure 12. A significant percent of loss in compressive strength was observed for the RAC mix as per previous studies [46,47], around 48.8% due to the inferior quality of RAs. CRAC mix had comparatively more residual compressive strength (22.57 MPa) after the sulfuric acid attack than the RAC mix (14.32 MPa). It was attributed to the precipitation of calcium carbonate in the porous structure of RAs, which may have caused the reduction in the pore size of RAs. Improved microstructure and smaller pores of CRAC (1 bar-2 h) depicted resistance to an acid attack.

Table 7. Loss of percentage in compressive strength after SAA.

Sr.#	Sample	Original Compressive Strength (After 28 Days)	Residual Compressive Strength After SAA (After 90 Days)	Percentage Loss of Compressive Strength (After 90 Days)
	-	MPa		%
1	NAC	40.1	27.40	31.67
2	RAC	28	14.32	48.85
3	CRAC	36.4	22.57	37.9

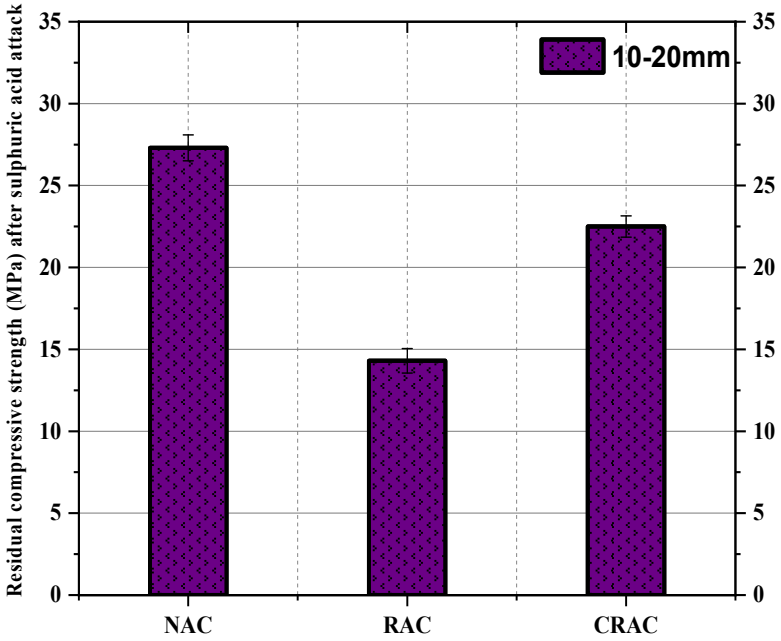


Figure 12. Residual compressive strength of NAC, RAC, and CRAC after SAA.

Change in Weight Due to SAA

Figure 13 displays the percentage of weight loss of all concrete mixes. Weight loss of concrete samples up to 3.38%, 10.92%, and 5.28% was observed for NAC, RAC, and CRAC, respectively. Results revealed that RAC mix is more susceptible to sulfuric acid attack than NAC and CRAC mixes. This loss of strength and weight in RAC occurs because of the porous nature of recycled aggregates (RAs) and more content of free lime in adhered mortar on its surface [46]. This free lime reacts readily with acid, causing concrete deterioration. To produce a recycled concrete mix that resists acid attack, it is crucial to minimize the free lime content in RAs [48]. Meanwhile, CRAC mix deterioration is slower than RAC mix due to calcium carbonate formation in the pores of RAs, which hindered its degradation.

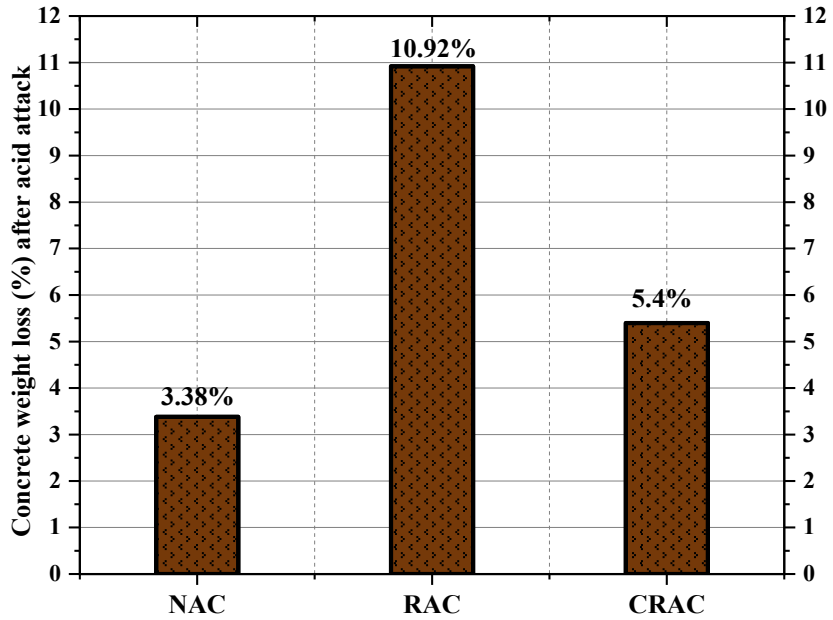


Figure 13. Percentage of weight loss of NAC, RAC, and CRAC after SAA.

4.3.3. Concrete Sulfate Attack Resistance (SAR)

Loss in Compressive Strength

Table 8 demonstrates the percentage loss of compressive strength for NAC, RAC, and CRAC mixes due to 150 days of exposure to sodium sulfate solution (Na_2SO_4). Figure 14 presents the residual compressive strength for all under-study concrete samples.

Results indicated that sulfate ions accumulate more rapidly in RAC due to multiple ITZs, cracks, and voids around its surface. As a result, RAC shows poorer resistance to long-term external sulfate attacks than NAC, which previous researchers also confirmed [49–51]. The sulfate resistance of RAC is crucial for ensuring serviceability and long-term longevity. However, Figure 10 shows that the CRAC mix (1 bar-2 h) had comparatively more residual compressive strength (29.2 MPa) after the sulfate solution attack than the RAC (19.2) mix.

Table 8. Loss of percentage in compressive strength after SAR.

Sr.#	Sample	Original Compressive Strength (After 28 Days)	Residual Compressive Strength After SAR (After 150 Days)	Percentage Loss of Compressive Strength (After 150 Days)
-		MPa		%
1	NAC	40.1	32.4	19.2
2	RAC	28	19.2	31.4
3	CRAC	36.4	29.2	19.7

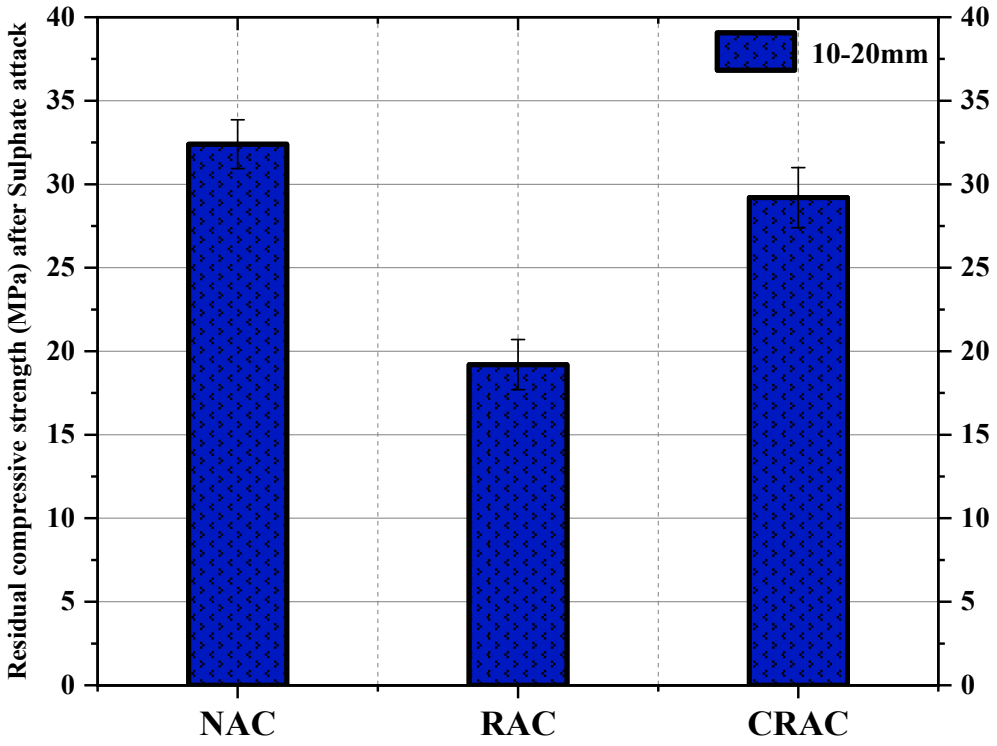


Figure 14. Residual compressive strength of NAC, RAC, and CRAC after SAR.

Change in Weight Due to Sulfate Attack

After 150 days of immersion in Na_2SO_4 solution, the resistance of NAC, RAC, and CRAC mixes was observed regarding weight change. The weight loss percentage for each mix was determined by comparing the weight of 28 days of cured samples before immersion and after exposure to Na_2SO_4 solution. It can be shown in Figure 15. The weight loss percentage for NAC, RAC, and CRAC was measured as 1.1%, 3.6%, and 2.2%, respectively.

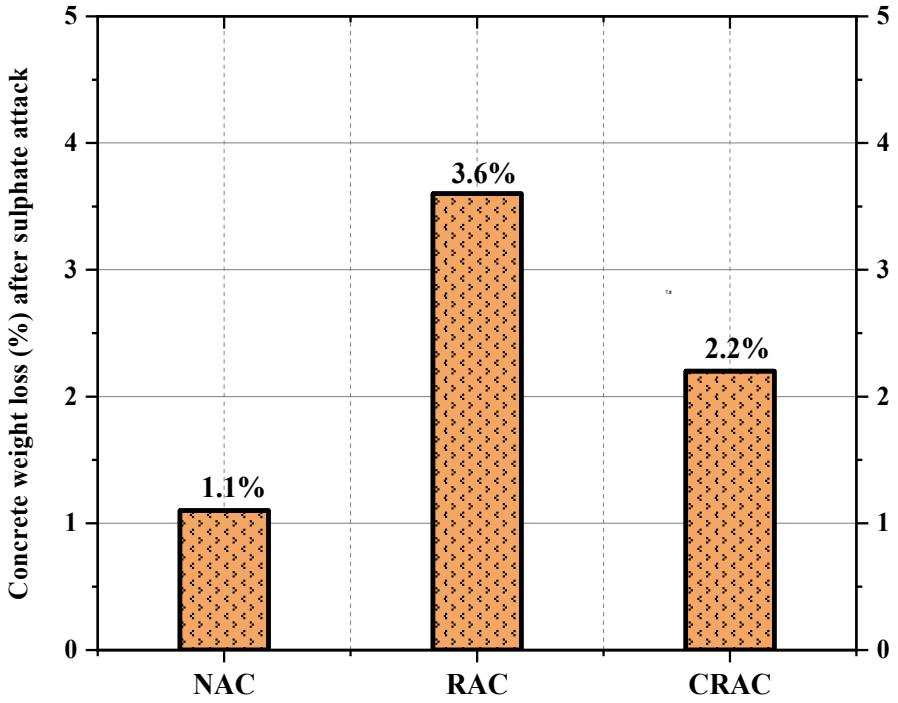


Figure 15. Percentage of weight loss of NAC, RAC, and CRAC after SAR.

The CRAC mix exhibited a lower percent weight loss than RAC. The accelerated carbonation on RA resulted in a densified concrete mixture and provided a barrier to slow the deterioration process of the CRAC mix. Calcium carbonate acted as a protective shield, reducing the pores and permeability of the CRAC mix and, consequently, preventing the infiltration of harmful substances like sulfate ions. Therefore, the CRAC mix had more resistance to sulfate solution than RAC. The findings demonstrate that incorporating carbonated recycled aggregates into concrete blocks the intrusion of harmful substances, reducing the weight change loss due to the Na_2SO_4 attack.

4.4. SEM Analysis

Figure 16a presents RA's microstructure, revealing pores and cracks that confirm it is weak and poor quality. Therefore, these porous and weak RAs and their cracked and rough surfaces contribute to lesser mechanical and durability properties of the RAC mix Figure 16b shows an improved, less porous, and compacted CRAC structure due to the carbonation product calcium carbonate, which fills the microcracks in RA. Hence, this has resulted in an improved mechanical and durability performance of the CRAC mix.

4.5. Mechanical Properties of CRA and SCM Blended Concrete

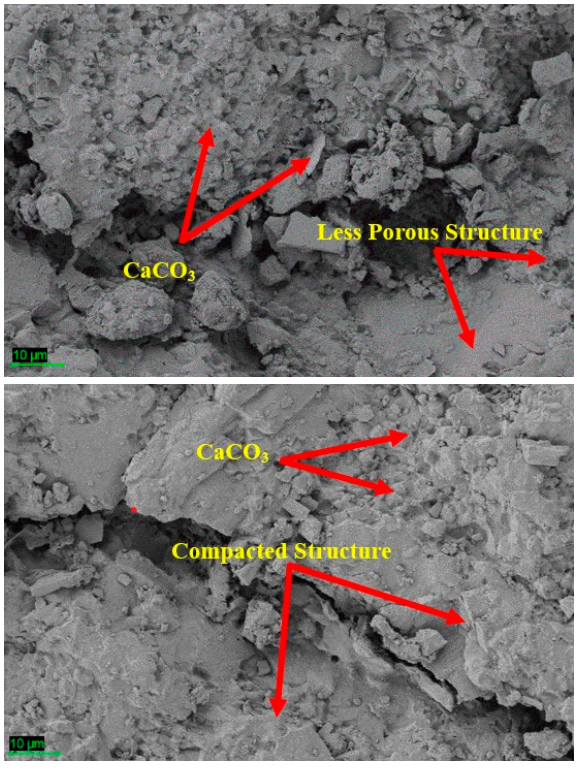
4.5.1. Compressive Strength

In the preparation of the CRAC mix, supplementary cementitious materials were partially incorporated, i.e., 10% silica fume (SF), 5% zeolite powder (ZP), and 10% limestone powder (LSP), to replace cement. Furthermore, 5% zeolite powder was used as a substitute material for fine aggregate. Compressive strength results for CRAC mixes blended with SCMs at a water–cement ratio (w/c) of 0.42 and 0.55 are depicted in Figure 16. This can be observed from Figure 17. The CRAC sample (CRA-OPC-0.55) containing ordinary Portland cement (OPC) showed 31.26 MPa strength, which was 17.96% greater than that of the RA-0.55 mix (26.50 MPa). The compressive strength of the mixes, i.e., CRA-10SF-0.55 and CRA-10SF-10ZP-0.55, were 27.78 MPa and 18.91 MPa, respectively; these values correspond to approximately 11.1% and 39.5%, had lower compressive strength than CRAC

(CRA-OPC-0.55) sample. Calcium hydroxide is a crucial component likely responsible for providing an alkaline environment to pozzolanic waste materials. Thus, the reduced alkalinity of carbonated aggregates could impede the pozzolanic reactivity of silica fume and zeolite powder. This reduced alkalinity of carbonated aggregates may not be conducive to facilitating the silica fume and zeolite powder for the pozzolanic reactivity to occur.



(**a**)



(**b**)

Figure 16. SEM images of (**a**) RAC and (**b**) CRAC.

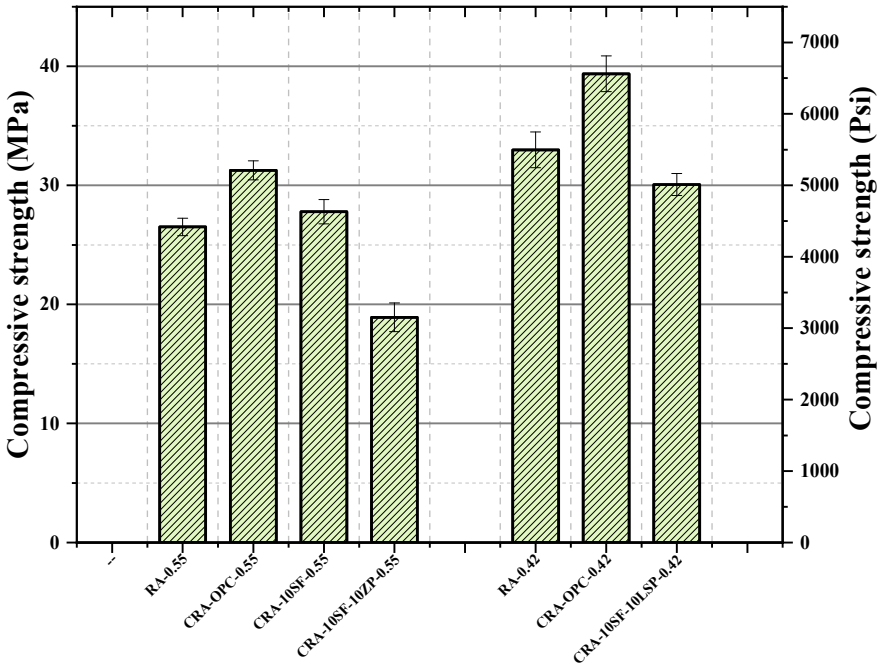


Figure 17. Compressive strength of CRA and SCM blended concrete.

The concrete produced by the blends of 10% silica fume (SF) and 10% limestone powder (LSP) with carbonated aggregates (CRA-10SF-10LSP-0.42) also exhibited 23.64% lower compressive strength (30.06 MPa) than the reference CRA-OPC-0.42 mix (39.37 MPa). This may be because of the slower rate of the hydration process, which can be attributed to the reduced amount of cement and lesser quantity of calcium hydroxide resulting from the carbonation reaction.

4.5.2. Tensile Strength

It can be shown from Figure 18 that the tensile strength for the mixture CRA-0.55 was 3.78 MPa, indicating 15.24% higher strength than non-carbonated RAC (3.28 MPa). Better strength can be attributed to carbonation; it strengthens the bond at the interface between cement matrix and aggregates. The tensile strength of the sample CRA-10SF-0.55, which contained 10% Silica Fume (SF) and carbonated aggregates, was 3.58 MPa. However, it was slightly less than the CRAC mix. The tensile strength of this mixture CRA-10SF-10ZP-0.55 was 3.24 MPa. Although, it was lower to some extent when compared to other mixes. As pozzolanic chemical reactions of SCMs require (an alkaline environment), calcium hydroxide is produced from cement hydration. The limited availability of calcium hydroxide could be due to its consumption during the carbonation process. In the blend of SF and ZP in the CRA-10S-10Z-0.42 mix, the tensile strength was reduced compared to that of the other mixes.

The tensile strength of the mixture RA-0.42, produced using RCA, was 3.90 MPa. The tensile strength was enhanced by 4.24 MPa, presenting an improvement of around 9.23% over the non-carbonated RAC mix. Improved tensile strength resulted from better interfacial bonding and densified microstructure brought about by carbonation. The tensile strength of the CRAC mix (CRA-10SF-10LSP-0.42) was 3.94 MPa, which was 7.16% lower than that of the CRAC mix. As stated earlier, SCMs require an alkaline environment for pozzolanic reaction, whereas calcium hydroxide may be consumed during carbonation reaction; thus, SF and LSP blends with CRCAs could not provide better results.

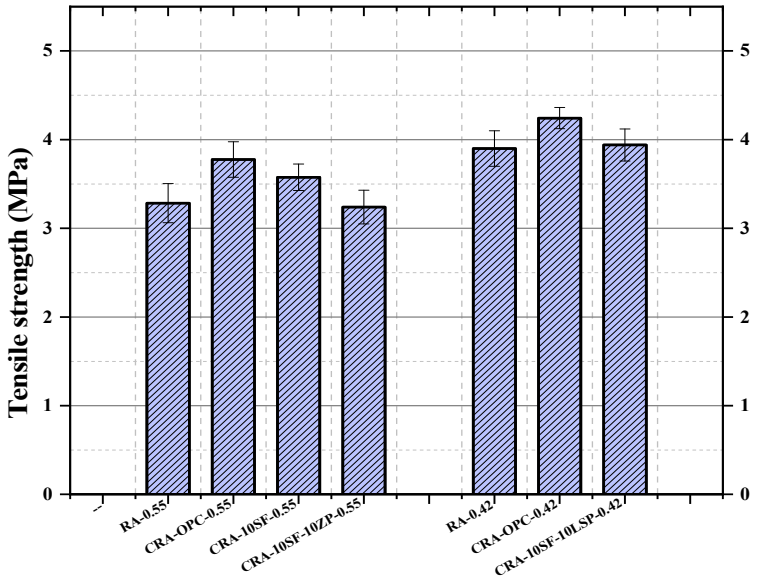


Figure 18. Tensile Strength of CRA and SCM blended concrete.

4.6. Alkalinity of Fresh Concrete Mixes

Figure 19a–c shows that both NAC and RAC mixes had higher alkalinity (pH value = 12). However, the CRAC mix exhibited lower alkalinity (pH = 10) than the RAC mix, suggesting that CRAC can be used effectively in plain cement concrete (PCC). The carbonation process in concrete, although improving durability and enhancing the mechanical properties of RAs, may also lead to some environmental side effects. Carbonation can alter the pH of concrete, potentially compromising its structural integrity by causing the corrosion of the steel reinforcement over time. However, with proper management, carbonation can assist in reducing the overall carbon footprint of concrete by capturing CO_2 during its lifecycle.



(**a**)



(**b**)

Figure 19. *Cont.*



(**c**)

Figure 19. Alkalinity of fresh concrete mixes (**a**) NAC, (**b**) RAC, and (**c**) CRAC.

5. Conclusions

1. Physical property tests revealed lower WA, aggregate impact, and crushing values, and improved bulk density for carbonated CRAs compared to non-carbonated RAs.
2. Compressive strength for the RAC mix was lower than for the NAC mix. Around 30% of strength loss was observed for the RAC mix compared to normal aggregate concrete. However, carbonated CRAs depicted a compressive strength increment of 30% compared to the RAC mix, thus overcoming compressive strength loss. The splitting tensile strength of RAC was 36% less than when compared to NAC. However, accelerated carbonation (1 bar, 2 h) on RAs caused a significant rise of around 42% in the tensile strength of the concrete mix. The dry density of the CRAC mix was improved by 1.5% compared to the RAC mix, which can be attributed to the calcium carbonate, a by-product of the carbonation process that develops a denser and compacted concrete mix.
3. Water absorption (WA) of recycled aggregate concrete mix is higher than that of normal concrete. In total, 7.58% of water absorption was found for mixed RAC than NAC. This was due to the adhered mortar content in RAs, which raised its porosity. However, accelerated carbonation assisted in reducing the water absorption (WA) of the concrete mix. Calcium carbonate precipitates into pores and cracks, thus resulting in an enhanced microstructure of RAs. Therefore, the CRAC sample presented a lower percentage of water absorption, around 6.06%, than the RAC sample.
4. Recycled aggregate concrete mixes are more vulnerable to acid and sulfate attack than normal aggregate concrete. However, the CRAC mix was still durable under acid and sulfate environments owing to its higher residual compressive strength and lower percent weight change.
5. When the concrete samples were exposed to a 5% acid (H_2SO_4) attack for 90 days, the losses in compressive strength for RAC and CRAC were measured around 48.85% and 37.9%, respectively. Percentage losses in weight were 10.92% and 5.4% for RAC and CRAC mixes, respectively. Thus, CRAC exhibited better resistance against acid attacks. Calcium carbonate behaved as a defensive product around RAs that may not allow the passing of harmful substances.
6. When the concrete samples were immersed in a 5% Na_2SO_4 solution for 150 days, the losses in compressive strength for RAC and CRAC were observed at about 31.4% and 19.7%, respectively. Percentage losses in weight were 3.6% and 2.2% for RAC and CRAC mixes, respectively.
7. The compressive and split tensile strength of concrete blended with CRAs and SCMs was lower than that of an individual carbonated recycled aggregate concrete mix. SCMs, i.e., SF, ZP, and LSP at both water-to-binder ratios (0.55 and 0.42), showed

lesser mechanical properties. CRA-10SF0.55 and CRA-10SF-10ZP-0.55 mixes revealed 11.1% and 39.5% less compressive strength than the CRA-OPC-0.55 mix. Furthermore, CRA-10SF-10-LSP-0.42 depicted a 23.64% decrease in compressive strength than CRA-OPC-0.42 mix. For tensile strengths, parallel trends were followed. It can be attributed to the extent of the hydration reaction, which became slower as the amount of cement was decreased and less calcium hydroxide was present due to the carbonation reaction, which is crucial for pozzolanic reactions. It can be said that the selection of OPC is essential to attain maximum strength in carbonated recycled aggregate concrete.

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Nomenclature

CRA	Carbonated recycled aggregate
NA	Natural aggregate
RA	Recycled aggregate
ITZ	Interfacial transition zone
OPC	Ordinary Portland cement
WA	Water absorption
AIV	Aggregate impact value
ACV	Aggregate crushing value
SAA	Sulphuric acid attack
SAR	Sulfate attack resistance
PCC	Plain cement concrete

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