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Effect of Carbonation Curing on the Performance of Concrete Containing Recycled Aggregates from Returned Concrete

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ABSTRACT

The stringent quality control measures on the properties of concrete result in the production of a large quantity of returned concrete in many countries. Waste management strategies include extracting the aggregates from this returned concrete to reduce the consumption of natural aggregates. This study aims to extract and characterize the recycled coarse aggregates (RCAs) obtained from returned concrete by using superabsorbent polymer (SAP) additives (RCA-SAP), and by washing (RCA-W), and to compare the properties with local specification for recycled aggregates. Furthermore, the comparative performance of the concrete containing those two types of differently treated recycled aggregates as partial replacements for natural aggregates (NCAs) under accelerated carbonation and water curing was analyzed through strength, water absorption and gas adsorption tests. Quality testing revealed that RCA-W outperforms RCA-SAP in terms of water absorption and abrasion resistance. RCA-W showed a water absorption of 1%, compared to a value of 4.5% for RCA-SAP. Also, a Los Angeles abrasion value of lower than 30% was observed for RCA-W, indicating better abrasion resistance compared to RCA-SAP. The concrete containing polymer-modified recycled aggregates at a 30% replacement showed a maximum strength of 60.44 MPa under carbonation curing and 51.93 MPa under water curing, indicating 16.4% higher strength under carbonation curing. In summary, it was confirmed that carbonation curing enhances the concrete performance with polymer-modified recycled aggregates.

Keywords: Absorption, Accelerated carbonation, Returned concrete, Superabsorbent polymer, Pore structure.

INTRODUCTION

Construction and demolition wastes (C&DWs) are increasing due to rapid urbanization, and governments worldwide are trying to manage these wastes as alternative resources for natural aggregates effectively. Large quantities of concrete wastes as returned concrete are generated from ready-mix concrete plants due to dumping the excess unused concrete from trucks to the plants (Ferrari et al., 2014). The construction industry in Kuwait has witnessed a remarkable boom in recent years and, at the same time, faces a huge shortage of construction raw materials. Over a period of five years, from 2015 to 2020, Kuwait government has allocated KD 34 billion for construction (New Kuwait Report, 2019). In 2019, the market demand for ready-mix concrete reached 11.85 million cubic meters (Industrial Bank of Kuwait Report, 2020). This thriving construction creates a significant demand for construction materials, which challenges Kuwait's resources. In 1997, the local authorities in Kuwait banned the production of coarse aggregates from local quarries due to the environmental impact of the production (Rahal, 2006). Therefore, all construction companies and ready-mix plants heavily rely on imported natural aggregates from nearby countries, such as the United Arab Emirates (UAE) and Iran. This necessitates investigating the potential of using waste as a sustainable source of aggregates in construction.

Globally, it is estimated that over 125 million tons of returned concrete are produced yearly, confirming as a main part of C&DW and becoming a burden for ready-mix concrete plants (Ferrari et al., 2014). It is estimated that approximately 2.3 % of the volume of concrete that is produced from ready-mix plants is returned to the plants in the UK, while 3 % and 1.4 % were estimated to be returned to the plants in Brazil and the US, respectively (Sealey et al., 2001; Obla et al., 2007; Vieira et al., 2019). It has been estimated that returned concrete waste consists of 70 % or more recoverable aggregates and 30 % or less non-recoverable cementitious paste, which can be recycled into aggregates and used in the formation of new construction projects (Xuana et al., 2018; De Brito Prado Vieira et al., 2020). This problem of returned concrete is also escalating in Kuwait due to an increase in the sheer mass production of concrete and the stringent governmental policies towards dumping solid wastes in landfills for environmental protection.

An earlier study reported that C&DW in Kuwait represents about 15%-30% of all solid waste by weight, compared to 29% in USA, and 40% in China (Al-Rifai & Amoudi, 2016). According to recent reports, Al Asima governate and Mubarak al-Kabeer governate in Kuwait had shown an increase in construction and demolition waste sent to landfills of 15 % and 8 %, respectively (Annual Statistical Report of Municipality of Kuwait, 2023-2024). Also, C&DW received to all landfill sites increased from 11.3 million tons in 2018 to 13.7 million tons in 2023 (Al-Raqeb et al., 2023; Kuwait Population Estimation, 2023).

Many waste management strategies are followed to deal with the returned concrete. One is the recycling of returned concrete when it is fresh into non-structural elements or new downgraded products, such as blinding or backfilling. Another strategy is the reuse of returned concrete into the new concrete mixture using retarders and activators. A common waste management strategy to deal with the returned concrete is to reclaim it through a washing-out process to recover aggregates by using 150 to 300 gallons of water for each mixing truck (Abdol & William, 1996). However, this washing-out process generates large quantities of grey water as hazardous waste due to the presence of heavy metals and high pH, and this grey water must be treated before being used in new batches of concrete mixes (Xuana et al., 2018).

One more strategy for recycling returned concrete into coarse aggregate is by adding chemical superabsorbent polymer (SAP) into the concrete truck mixer without the requirement of any special equipment. The addition of the required quantity of SAP, as per the manufacturer’s recommendations, can transform 1 m^3 of returned concrete into 2.4 tons of new recycled aggregates (Kazaz & Ulubeyli, 2015). Studies conducted in Brazil and Japan have demonstrated the potential of using recycled aggregates generated from returned concrete for various applications with slight differences in quality, low density, and high water absorption, as adding SAP accumulates the sand and fines in returned concrete waste (RCW) around the aggregates (De Brito Prado Vieira et al., 2020; Ferrari et al., 2012). It has been observed that crushed returned concrete aggregates are better than recycled aggregates obtained from C & DW as it is free of contaminants and with a significant residual value (Kim & Goulias, 2015).

In addition, previous studies have reported the advantage of short-term accelerated carbonation curing of concrete containing RCA in strength development (Rostami et al., 2012; Silva et al., 2015; Zhang et al., 2015; Liang et al., 2020). The properties of recycled concrete aggregates can be improved through the carbonation of the attached cement paste, as during the carbonation reaction, CO_2 reacts with calcium hydroxide ($Ca\,(OH)_2$) and calcium silicate hydrate (C-S-H) to form $Ca\,(CO)_3$ and silica gel that fill the pores (Zhang et al., 2015). A recent study has confirmed that the carbonation curing of recycled aggregates reduces water absorption from 12.6 % to 9.9 % compared to untreated recycled aggregates (Liang et al., 2020). Also, carbonation curing has improved the quality of new recycled concrete aggregate from crushing designed concrete, as it reduced the water absorption by 16.7 % and increased the compressive strength by 22.6 % (Xuan et al., 2016). In addition to its potential to reduce the water required for moist and steam curing of concrete products, carbonation curing helps recycle CO_2 emitted from cement production (El-Hassan, 2021).

Furthermore, accelerated carbonation curing is considered a sustainable method to improve the mechanical and durability properties of the concrete under specific operating conditions (El-Hassan, 2021; Rostami et al., 2012; Li et al., 2019; Ikumapayi et al., 2012; Rahmani & Mohammadzade, 2023). The influence of carbonation on the micro-structure depends on the period of curing. Hence, the time of exposure to CO_2 is crucial for optimum capturing without affecting the cementitious matrix (Neves Junior et al., 2015; Neves Junior et al., 2019). The effect of carbonation curing on concrete performance will be more predominant when replacing normal coarse aggregates (NCAs) with recycled coarse aggregates (RCAs) due to the higher porosity of RCAs compared to NCAs, resulting in the reduction of carbonation resistance of RCA concrete (Silva et al., 2015). In addition, it is reported that mortar and concrete mixes containing mineral additions show lower carbonation resistance in accelerated conditions (Leemann et al., 2015). Accelerated carbonation causes a reduction in the permeability of the concrete surface, as the volume of the reaction products ($CaCO_3$) is more than that of the original reactants, resulting in a change in the micro-structure and coarsening of the pore structure (Claisse et al., 1999; Shah et al., 2018). Considering the advantages of recycled aggregates in carbon capturing, the comparative performance of concrete containing two types of differently processed recycled aggregates under accelerated carbonation curing and water curing was investigated and reported in this paper.

RESEARCH SIGNIFICANCE

This study utilizes different waste management approaches to recycle aggregates from returned concrete to solve the problem of accumulated returned concrete and deal with the illegal dumping of concrete. The recycling of returned concrete using two different methods and the assessment of the quality of the recycled aggregates obtained from returned concrete according to the local specifications are investigated and reported in this paper.

In addition to washing and sieving the returned concrete using water to reclaim aggregates, an innovative technology is used to recycle the returned concrete to coarse aggregates using an SAP-based additive. Moreover, the paper reports on the effect of CO_2 curing on enhancing the mechanical performance of concrete mixes containing two types of recycled aggregates.

MATERIALS AND EXPERIMENTAL METHODS

Materials

The two types of recycled aggregates used in this research study were produced from returned concrete: recycled coarse aggregates processed from returned concrete using SAP additives, a proprietary product obtained from the manufacturer (RCA-SAP), and recycled coarse aggregates obtained from returned concrete by washing (RCA-W). A two-component powder product was used as SAP and added to the concrete truck mixer while rotating at a high speed of 15 rpm. Component A acts as a water absorber/viscosifier, and component B acts as a setting accelerator. The bulk density of component A is 0.8 g/cm^3, and that of component B is 1.1 g/cm^3. The alkali content for both components is $\leq 1\%$, and that of chlorides is $\leq 0.1\%$. The polymer absorbed the free mixing water and the returned concrete consolidated through chemical sintering during the process. Thus, the returned concrete was transformed into a granular material made with coarse aggregates, which was covered by a composite material consisting of cement, sand and SAP (Figure 1). No special equipment or water was needed to recycle the returned concrete. The treated recycled aggregates were spread on the ground for adequate curing. Light handling and crushing were carried out using digger excavators to break the conglomerated mass.

Figure (1): Extraction of granular aggregates (a) discharge of treated aggregates and (b) spreading for adequate curing

The quality of the recycled aggregates obtained from both processes was characterised and evaluated in comparison to the specification limits locally recognized for coarse aggregates by GSO1809, 2007, GSOASTM C33, 2016, ASTM C33, 2018, and GSO 2489, 2015. The quality tests are detailed in Table 1.

Table 1. Quality testing program for recycled coarse aggregates

Property	**Standard Test Method**
Grading	ASTM C136, 2019
Materials finer than 75-µm	ASTM C117, 2017
Abrasion loss	ASTM C131, 2017
Density	ASTM C29, 2017
Absorption and specific gravity	ASTM C-127, 2015
Particle shape	BS 812Part105.1/105.2, 989&1990
Soundness	ASTM C88, 2018
Clay lumps and friable particles in aggregates	ASTM C142, 2017
Acid soluble chloride test	BS EN1744, part 5, 2006
Acid soluble sulphates	BS EN1744, part 1, 2009

Also, ordinary Portland cement type 1 tested following ASTM C150, 2017, and fine aggregates, which were local siliceous sand conforming to ASTM C33, 2018, were used in the study.

Concrete Mix Preparation, Curing, and Testing Sample Preparation

The effect of replacing normal coarse aggregates (NCAs) with recycled aggregates on plain cement concrete properties was evaluated by preparing concrete mixes with a w/c ratio of 0.4 and a designed strength of 45 MPa. NCAs were replaced with both types of RCAs in two different percentages of 30 % and 50 % by weight.

These two percentages were selected based on the inferences from previous studies, which showed that a remarkable reduction in strength could not be observed up to 30 % replacement (Awadh et al.,2021). In contrast, a reduction in strength could be observed for 50 % replacement. RCA-SAP and RCA-W were used in saturated surface dry (SSD) conditions, so that the water absorption of RCA was accounted for in the mix design. A polycarboxylate-based superplasticizer was added to control the workability of the mixes. The details of the mix design of concrete containing the processed returned aggregates are given in Table 2. A control mix and four test mixes were prepared with two categories of processed recycled aggregates.

Table 2. Mix design of concrete containing processed returned aggregates

Materials	**Mix 1**	**Mix 2**	**Mix 3**	**Mix 4**	**Mix 5**
	Control (C)	**PRA30**	**PRA50**	**WRA30**	**WRA50**
Cement (kg)	450	450	450	450	450
Free water (kg)	180	180	180	180	180
Sand (kg)	640	640	640	640	640
NCA 20 mm (kg)	540	378	270	378	270
SSD RCA 20 mm (kg)	-	162	270	162	270
NCA 12.5 mm (kg)	334	234	168	234	168
SSD RCA 12.5 mm (kg)	-	100	168	100	168
NCA 10 mm (kg)	276	193	138	193	138
SSD RCA 10 mm (kg)	-	83	138	83	138
Superplasticizer (L)	5-7.5	5-7.5	5-7.5	5-7.5	5-7.5
Free w/c	0.40	0.40	0.40	0.40	0.40

Curing

Two different curing conditions, (1) water curing and (2) carbonation curing (CO_2 supply up to 30%) were employed in this study. After demolding, water curing was carried out by immersing the samples in a water curing tank for the required ages of curing of 7 days and 28 days.

In the case of carbonation curing, the carbonation cycle included pre-carbonation, carbonation, and post-carbonation to achieve the maximum benefit of carbonation curing on concrete properties as suggested in previous research (El-Hassan, 2021). Samples were subjected to air curing inside the moulds on the casting day. After demolding, the samples were kept in an accelerated carbonation chamber under the controlled conditions of a temperature of 20ºC, a relative humidity of 60 %, and CO_2 of a dosage of 30 % for 24 hours. After that, the samples were kept in water for 7 days, taken out and kept for air curing in the laboratory conditions until the age of testing. The 7-day water curing is employed as a post-carbonation hydration step to restore the lost water during the pre-conditioning and the exothermic carbonation reaction and to promote further hydration of unreacted hydraulic cement. The benefit of post-carbonation hydration on compressive strength has been reported in previous studies (Klemm and Berger, 1972; El-Hassan et al., 2021).

Maximum carbonation occurs at a relative humidity of 50%-70% and hence, the carbonation chamber had been set at a relative humidity of 60 %. The accelerated carbonation chamber was using a single stage refrigeration system utilizing a refrigerant with an air cooled condenser, and atomizing humidifiers for controlling the required temperature and humidity. The chamber was equipped with a CMP control system and a proprietary dual wavelength infrared sensor for continuous monitoring and control of CO_2 concentration level. The gas concentrations were monitored continuously during the 24-hour carbonation process using the digital display in the color touch screen.

Test Methods for the Determination of the Properties of Concrete

The fresh state properties of the concrete, such as slump, air content, and unit weight of concrete, were determined following ASTM C143, 2020, ASTM C231, 2022, and ASTM C138, 2020, respectively. The hardened state properties of concrete were determined, and these properties include compressive strength according to BS EN 12390-3, 2019 at 7 days and 28 days, as well as splitting tensile strength according to ASTM C496, 2017 at 28 days. To understand the effect of replacing NCAs with RCAs on durability, water absorption according to ASTM C642, 2021, rapid chloride permeability according to ASTM C1202, 2017, and gas adsorption analysis were also conducted.

RESULTS AND ANALYSIS

Quality Characterization of the RCA-SAP and RCA-W

The quality characterization of RCA-SAP was evaluated and compared to the quality of RCA-W, as given in Table 3. The comparison showed a better quality of RCA-W, especially in terms of absorption, which showed a value lower than 1 %. Both types of recycled coarse aggregates had shown a specific gravity of more than 2, satisfying the limit of specific gravity according to GSO2489, 2015. In addition, RCA-W showed a Los Angles abrasion value of lower than 30%, indicating better abrasion resistance compared to RCA-SAP.

Furthermore, RCA-W was the only recycled aggregate that satisfied the specified limit for abrasion resistance to be lower or equal to 30 % (ASTM C136, 2019). However, compared to the limit specified for natural aggregates to be used in concrete, the studied RCA samples achieved values less than 50 %, the maximum limit specified for natural aggregates in concrete (GSO1809, 2007). Moreover, all RCA samples showed satisfactory performance to the specified limits (GSO2489, 2015) in terms of the amount of clay lumps and friable particles and total flakiness index as given in Table 3.

Table 3. Physical and chemical properties of processed recycled coarse aggregates

Physical Property	GSO 2489 Limit	(RCA-SAP)	(RCA-W)
Bulk Density (kg/m^3)	--	1352	1595
Specific Gravity	2 % minimum	2.84	2.713
Water Absorption (%)	3-4 % maximum	4.53	0.34
% of Voids (%)	--	43.6	39.2
Los Angles Abrasion (%)	30 % or less	36.35	26.66
Materials Finer than 75-µm (%)	--	0.64	0.43
Clay Lumps and Friable Particles in Aggregates (%)	Maximum 2 %	0.26	0.30
Total Flakiness Index (%)	40 %	7.2	9.54
Elongation (%)	--	7.3	17.4
Element Concentration (%)			
Sulfate (SO_4) (%)	1.5 % or less	0.31	0.24
Chloride (%)	1.0 % or less	0.001	0.004

The test results of grading of coarse recycled aggregates showed a limited quantity of materials finer than 75 µm for RCA-SAP and RCA-W. Results were less than 1 % for both samples. RCA-W showed the best grading when compared to grading limits specified for the 20-mm aggregate size specified in GSO 1809, 2007 and GSO 2489, 2015, which follow the limits of ASTM C33, 2016. The chemical analysis results detailed in Table 3 showed that all samples of recycled aggregates met the limits specified in GSO2489, 2015. Results also showed that RCA-W exhibited the highest chloride content compared to RCA-SAP. Also, when comparing the results with the maximum limit specified for natural aggregates in concrete (GSO1809, 2007), all samples were within the recommended limits of 0.8 % for chloride content and 4 % for sulfate content.

Properties of Concrete in the Fresh State

The slump value of the different mixes showed that workable mixes were produced in all cases. The slump and the unit weight of mixes in the fresh state were satisfactory to the requirements of normal-weight concrete. A workable slump of 90-120 mm and a normal unit weight of 2430 kg /m^3 could be produced for all mixes. The air content of the prepared mixes varied from 1% to 3%, which was also within the recommended range.

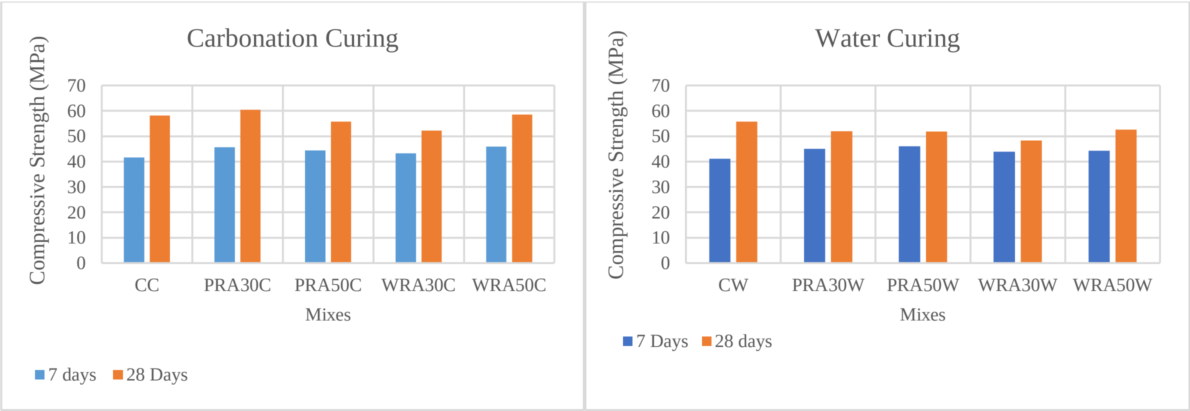
Properties of Concrete in the Hardened State

The effects of replacing NCAs with polymer-modified recycled aggregates (PRAs) and washed recycled aggregates (WRAs) on strength and durability properties were evaluated.

Compressive Strength Analysis

The compressive strength of concrete mixes tested at different ages is shown in Figure (2). Compared to control mixes, after 7 days of curing, mixes with both types of recycled aggregates had shown an increase in compressive strength under both water curing and carbonation curing, which may be because of the increased hydration due to higher water absorption of the RCA. As expected, the compressive strength at 28 days was higher than the compressive strength at 7 days under both curing methods. The control mixes showed comparable strength under both curing methods, whereas, after 28 days of curing, recycled aggregates showed a higher strength under carbonation curing than under water curing. The higher mechanical strength under accelerated carbonation curing may be due to the precipitation of calcium carbonate deposits in the open pores, resulting in the modification of the micro-structure (Shah et al., 2018).

In addition, mixes with both recycled aggregates at 28 days had shown higher strengths than the control mixes, indicating the better capability of recycled aggregates to absorb CO_2 than normal aggregates and resulting in a reduction in porosity. After 28 days, the difference in strength between water curing and carbonation curing was higher for mixes with polymer-modified recycled aggregates than the control mixes and mixes with washed recycled aggregates. This is because the PRA with super absorbent polymers (SAP) can absorb more water during mixing, resulting in an increased hydration reaction and formation of more hydrated products. Maximum strength development of 60.44 MPa was observed for mix with PRA at 30 % replacement under carbonation curing (PRA30C). This mix had shown 16.4 % higher strength than the mix under water curing. One of the differences that could be observed between the two types of recycled aggregates was that PRA had shown higher strength at 30 % replacement whereas WRA had shown higher strength at 50 % replacement under both curing conditions at 28 days.



(a) (b)

Figure (2): Compressive strength of concrete mixes under (a) carbonation curing and (b) water curing

Splitting Tensile Strength

The splitting tensile strength of concrete mixes at 28 days with both types of recycled aggregates under both methods of curing is shown in Figure (3). A comparable strength could be obtained for the control mixes under both methods of curing. However, consistent performance could not be obtained for splitting tensile strength measurements for mixes with both types of recycled aggregates. This may be because, it is an indirect test for determining the tensile strength, which is influenced by the heterogenous nature of concrete, including aggregate interlocking, micro-structure, pore size distribution in the interfacial transition zone, and bonding of aggregate with cement matrix. As in the case of compressive strength, PRA had shown a higher strength than WRA under carbonation curing.

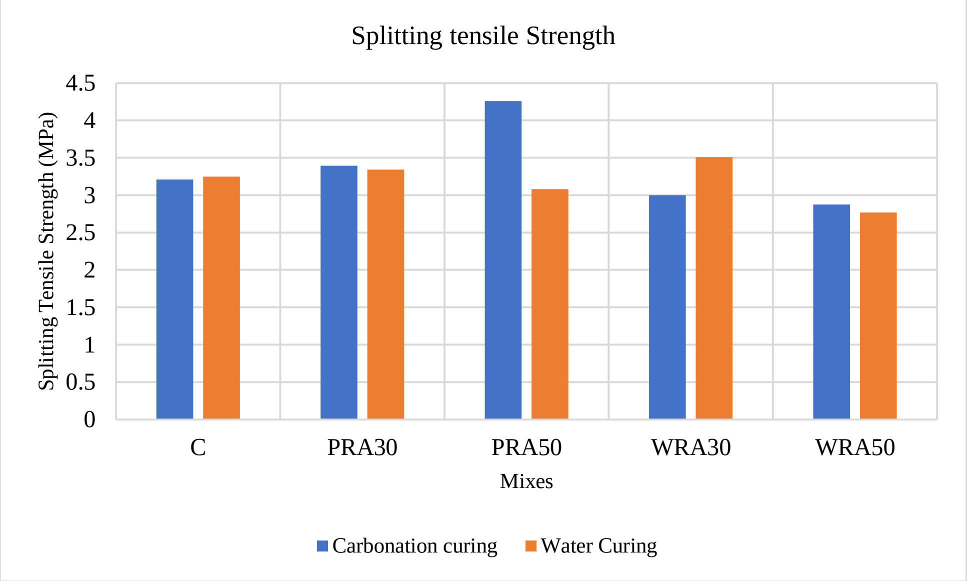


Figure (3): Splitting tensile strength of concrete mixes

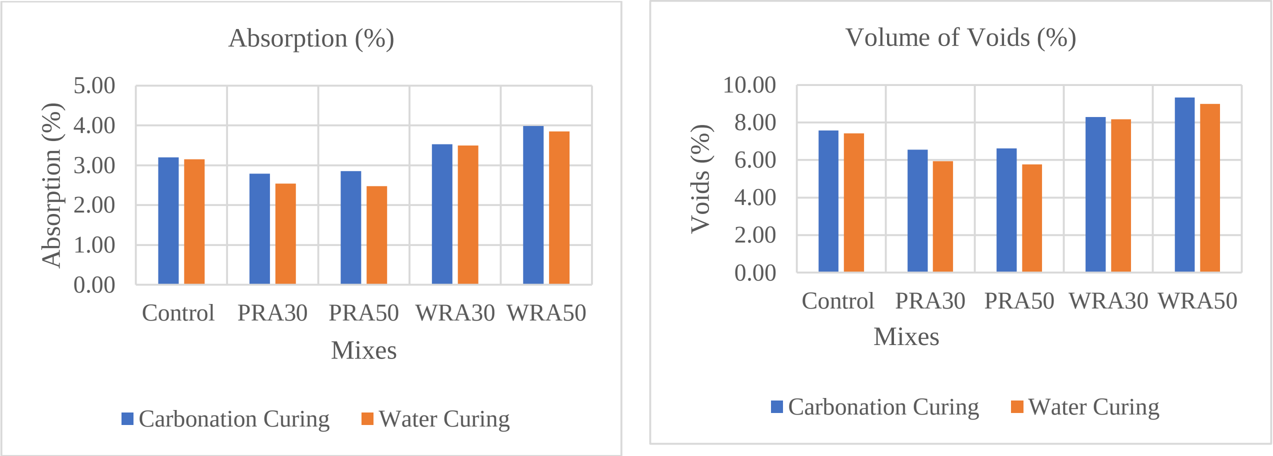
Water Absorption and Percentage of Voids

The water absorption and volume of voids of different concrete mixes were determined according to ASTM C642, 2021, and the test results are shown in Figure (4). The percentages of water absorption and voids were comparable for control mixes under both methods of curing. It can be inferred from the test results that the use of PRA improves the performance of concrete mixes, as the water absorption and volume of voids of such mixes were lower than in the control mixes. This is confirmed through the compressive strength results of mixes with PRA. Compared to the control mix, PRA50W showed a maximum reduction of 22.2 % in the volume of voids, and therefore, the lowest water absorption of 2.47 % could be observed for the mix PRA50W.

The reported results also have confirmed that the curing conditions can affect the water absorption of concrete. Different curing conditions can vary the permeability of concrete, and obviously, air conditioning causes a higher water absorption due to more voids, as shown in Figure (4), which is in line with earlier results (Zhang & Zong, 2013). In general, concrete mixes with both recycled aggregates had shown slightly higher water absorption and volume of voids under carbonation curing than under water curing. This is predicted, as the samples under carbonation curing have more voids, whereas samples under water curing are already saturated with water. Even though the water absorption of polymer-modified recycled aggregates (4.96 %) was higher than that of washed recycled aggregates (0.34 %), the percentage of absorption of water was less for concrete mixes with PRA than with WRA under both curing conditions.

Compared to control mixes, PRA mixes at 30 % and 50 % replacement under both methods of curing had shown a lower water absorption, whereas WRA mixes at both percentages of replacements had shown a higher water absorption and a higher percentage of voids. This is expected, as the PRA is already saturated with the water absorbed by the SAP under water curing, resulting in a reduction in further water absorption. Also, the percentage of voids was less for the mixes with PRA.

In addition, previous studies have reported that accelerated carbonation tends to produce a higher water absorption only in the absence of additives (Carvajal et al., 2006). As mentioned earlier, the mixes with RCA had shown a higher compressive strength under carbonation curing than under water curing, which cannot be correlated with water absorption. This is also justified, as the strength of concrete depends on the surface properties as well as on the internal structure.



(a) (b)

Figure (4): (a) Water absorption of concrete mixes after 28 days of curing, and (b) percentage of voids of concrete mixes after 28 days of curing

Rapid Chloride Permeability Test (RCPT)

The ability of concrete to resist chloride ion penetration was studied as per ASTM C1202-17 (2017), after 28 days of curing. The test results given in Table 4 indicated that the permeability class of all mixes falls under the category of "moderate" permeability under both methods of curing. However, none of the mixes had shown low permeability. One of the reasons for the concrete containing RCA fails to achieve "low" permeability classification may be due to chemical limitation, as the RCA-SAP mixes contain additional chemical ions resulting in increased permeability while using the RCPT method. Also, returned RCA contains a more porous surface due to the presence of adhered mortar, resulting in higher permeability.

Except for PRA 30, other mixes had shown a higher resistance in carbonation curing than in water curing, and some of the earlier studies have reported that carbonated concrete can have a higher resistance to chloride ion penetration (Rostami et al., 2012). This improved durability of the carbonated concrete is due to the conversion of Ca (OH)$_2$ into Ca (CO)$_3$ deposits, resulting in the reduction of the calcium hydroxide at the surface and the reduction in the connectivity of pores (Rostami et al., 2012). This can be correlated with RCPT results given in Table 4, as carbonated concrete has a lower value of charge passed in Coulombs, indicating a higher resistance to chloride penetration compared to water cured samples. However, this test cannot be considered a quantitative indication of chloride ion penetration, as the charge passed depends on the presence of other ions, especially in the case of mixes containing PRA.

Table 4. Chloride ion permeability of concrete mixes after 28 days of curing

Sample	Charge (Coulombs)	Permeability Class	Charge (Coulombs)	Permeability Class
	Carbonation Curing		Water Curing	
Control	3207	Moderate	3306	Moderate
PRA30	3236	Moderate	3103	Moderate
PRA50	2590	Moderate	3361	Moderate
WRA30	2899	Moderate	3231	Moderate
WRA50	2943	Moderate	3467	Moderate

Determination of Carbonation Depth and pH of Concrete Samples

The samples subjected to carbonation curing and water curing were subjected to a phenolphthalein test after 6 months to understand whether there were any adverse effects due to carbonation curing. In addition, the pH of concrete samples was determined after grinding the samples. From each cylinder, the samples were collected from at least three areas after performing the phenolphthalein test. Then, they were powdered for homogenization and the required quantity was used for pH testing. Table 5 gives the details of the carbonation depth and pH of the different samples.

The concrete samples can be classified as non-carbonated, partially carbonated and fully carbonated, based on the phenolphthalein test (Chang & Chen, 2006). It can be observed from the test results that the samples under carbonation curing had indicated a carbonation-affected zone from 1 mm to 10 mm, whereas the presence of carbonation was not shown for samples under water curing. The pH of the mixes was measured after the phenolphthalein test. In the case of carbonated concrete, the pH at the surface was less than that at the center of the sample. The pH of the mixes studied varied from 11-12, indicating partly carbonated and non-carbonated samples in the studied cases (Chang & Chen, 2006).

Table 5. Carbonation depth and pH of the samples

Mix Details	Carbonation Depth (mm)	pH
CC	2	11.93
CW	Nil	11.39
PRA30C	5	11.83
PRA30W	Nil	12.03
PRA50C	2	11.16
PRA50W	Nil	11.00
WRA30C	10	11.12
WRA30W	Nil	11.13
WRA50C	1	11.11
WRA50W	Nil	11.36

Nitrogen Adsorption Analysis

Porosity and pore size distribution of concrete have a predominant effect on the strength and durability characteristics. Therefore, gas adsorption analysis was conducted using nitrogen as an adsorbent to evaluate the effect of replacing natural coarse aggregates with recycled aggregates on the pore size, pore distribution and pore structure of concrete under two different curing conditions. After 28 days of curing, concrete chips were taken from the sample and kept in acetone for one week to stop hydration. The samples were taken out, vacuum saturated, and concrete chips of approximately 0.6 g were taken and used for analysis after degassing. Based on the results, the adsorption-desorption isotherm, BET surface area and pore volume were measured and calculated through various models.

The adsorption-desorption isotherms were developed for different mixes to understand the effects of different types of RA in different dosages on the gas adsorption measurements. As concrete is a porous material, Brunauer, Emmett and Teller's (BET) model for multi-layer surface adsorption was used to compare the BET surface area of different samples, as given in Table 6. Horwath-Kawazoe model was used to analyze the pore-volume development corresponding to different pore widths. The different pore sizes were designated as micro-pores of <2 nm, meso pores of 2-50nm, and macro pores of greater than 50 nm.

Except in the case of PRA30, all other mixes had shown a higher BET surface area under water curing than under carbonation curing. As expected, approximately 28 % reduction in the BET surface area of the control sample cured under carbonation was noticed compared to the sample cured under water. The decrease in BET surface area under carbonation curing may be due to the deposit of hydrated products due to the carbonation on the pore entrances, resulting in the reduction in the entry of gases into the pores (Kupwade-

Patil et al., 2016). The increase of PRA from 30% to 50% resulted in a maximum reduction of 63% in the BET surface area in the case of carbonation curing compared to water curing. This supports the compressive strength results, as PRA30 under carbonation curing had shown a higher strength than PRA 50 due to better hydration reaction, resulting in higher BET surface area. However, an increase of WRA from 30 % to 50 % resulted in a remarkable percentage increase in BET surface area of 160.5 % and 149 % in the case of carbonation curing and water curing, respectively. However, a direct comparison between different concrete mixes was not possible, as the surface area depends on several factors.

Table 6. BET surface area of different concrete mixes

Mix	BET Surface Area (m^2/g)
CC	9.686
CW	12.390
PRA30C	15.044
PRA30W	11.617
PRA50C	9.317
PRA50W	15.231
WRA30C	5.713
WRA30W	7.029
WRA50C	14.886
WRA50W	17.502

Furthermore, it was observed that the pore width varied from 0.7 nm to 18 nm for the studied mixes, which inferred that the pores in our samples represented both micro-pores and meso-pores. The isotherm shown in Figure (5) represents the type-II isotherm with H3 hysteresis loop as per IUPAC, and as mentioned in previous research publications (Kupwade-Patil et al., 2016; Kupwade-Patil et al., 2018; Joseph et al., 2019). All samples in this case showed the same type of isotherm, indicating open-slit or parallel-plate mesoporous pores. Initial mono-layer adsorption (Point A) followed by multi-layer adsorption was shown in this type of isotherm (Kupwade-Patil et al., 2018). A slow increase in nitrogen adsorption was observed after point A due to multi-layer adsorption. A sudden increase in adsorption observed after point B is due to capillary pore condensation, as shown in Figure 5. The nitrogen adsorption-desorption isotherm for control mix and mixes with polymer-modified and recycled coarse aggregates under water curing and carbonation curing are shown in Figure (6).

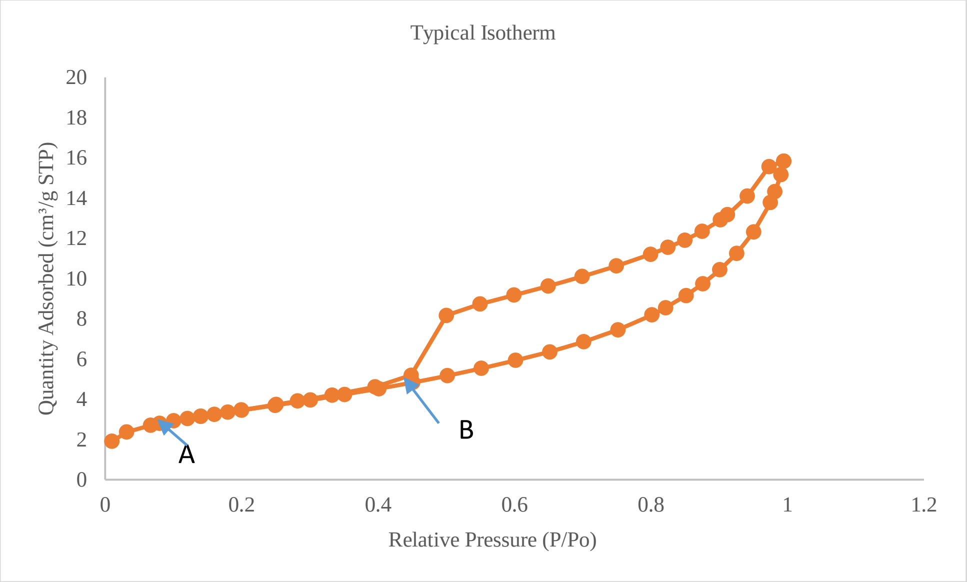


Figure (5): Typical adsorption-desorption isotherm

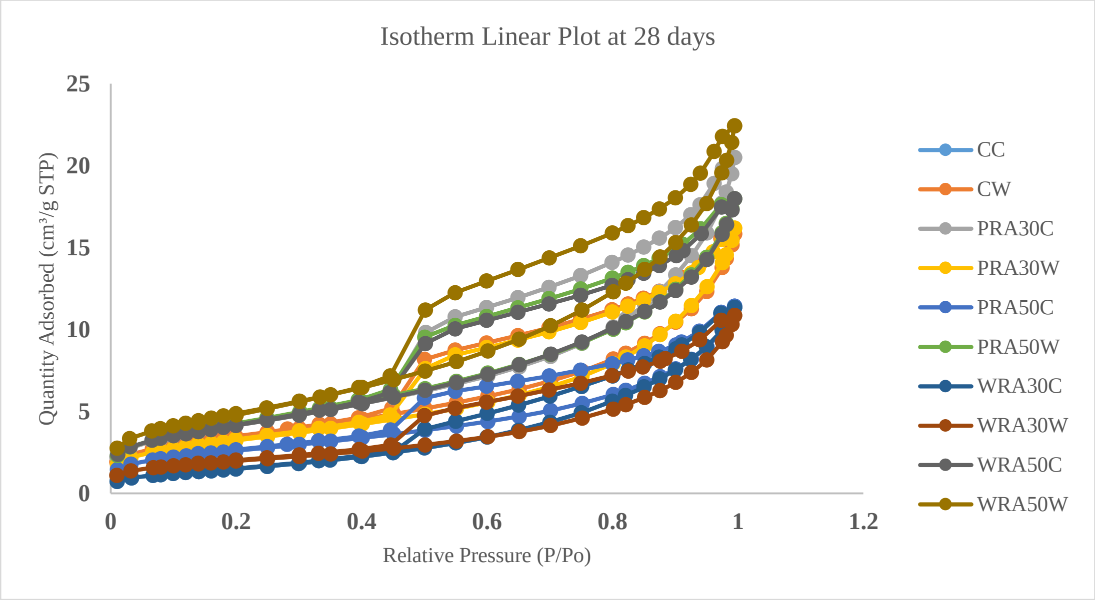


Figure (6): Adsorption-desorption isotherm for concrete mixes

Except in the case of PRA30, all other mixes had shown a higher amount of gas adsorption in the case of water curing, compared to carbonation curing, which is in line with the higher BET surface area observed in the case of water curing. $CaCO_3$ occupies more volume than $Ca(OH)_2$, resulting in a decrease in the porosity of carbonated concrete. Also, water released by $Ca(OH)_2$ due to carbonation results in an increase in internal humidity and further hydration of unhydrated cement. This is in line with the increased strength and reduction in surface permeability observed in the case of carbonated concrete. At 30% replacement, PRA samples showed a higher adsorption than that of WRA samples. Also, PRA30C had shown a higher adsorption than that of PRA30W, may be due to the formation of more hydration products or due to the formation of coarser pores, which was confirmed through a higher BET surface area. This is in line with the higher compressive strength observed in the case of PRA30C.

Further, the area between the adsorption-desorption curve was more in the case of PRA30C compared to PRA30W, indicating a greater number of larger pores. As in the case of PRA30C, a larger number of coarser pores could also be observed in the case of WRA50W. The cumulative pore volume was plotted against the pore width using the data obtained through the nitrogen adsorption analysis using the Horvath-Kawazoe model for slit or parallel-shaped pores as shown in Figure (7). An increase in cumulative pore volume observed for control mixes under water curing than under carbonation curing may be due to an increase in the formation of porous cement hydration products. As in the case of adsorption-desorption isotherm, except for PRA30C, all other mixes had shown a higher cumulative pore volume under water curing, compared to carbonation curing, indicating a reduction in porosity due to carbonation.

PRA30C showed more cumulative pore volume as well as more pores of size larger than 100 nm compared to PRA30W, which was confirmed through the adsorption-desorption isotherm. WRA30W showed the least cumulative pore volume among all mixes. Even though WRA30W had shown the least pore volume for >100 nm pores, WRA30C had shown a lower pore volume for pores of size less than 50 nm. This indicates that in the case of WRA, carbonation curing had resulted in the formation of a smaller number of micro-pores and meso-pores, whereas more macro-pores were formed. It was confirmed through previous studies that a marked increase in the volume of pores of size greater than 100 nm was observed with carbonation (Leemann et al., 2015). However, with the increase in WRA from 30 % to 50 %, carbonation curing resulted in a reduction in the cumulative pore volume compared to water curing, which is in line with the higher compressive strength observed in the case of WRA50C. In general, it was detected that carbonation curing improves the pore structure and micro-structure of concrete, as confirmed through previous studies (Leemann et al., 2015; Shah et al., 2018).

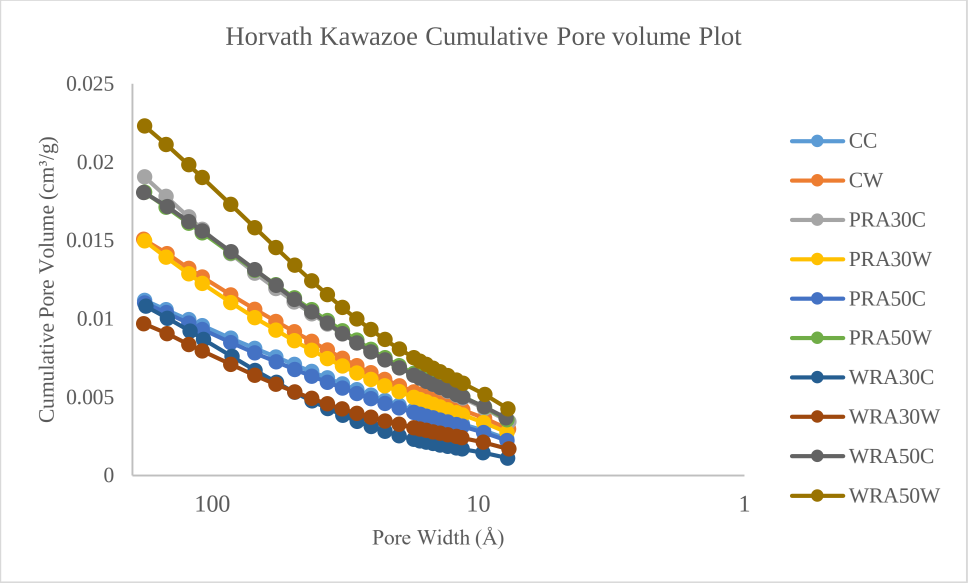


Figure (7): Pore size distribution plot for cumulative pore volume *vs.* pore width

CONCLUSIONS AND RECOMMENDATIONS

The characterization of the recycled coarse aggregates processed from returned concrete using SAP additives (RCA-SAP) and by washing (RCA-W) confirmed that RCA-W is having a superior quality compared to RCA-SAP in terms of water absorption and abrasion resistance.

However, concrete containing RCA-SAP outperformed concrete containing RCA-W, making it a viable option for replacing up to 30% of natural coarse aggregates (NCAs).

In addition, the performance evaluation of concrete containing both types of recycled aggregates showed that carbonation curing improved the compressive strength compared to water curing, despite a slightly higher water absorption and void volume. Also, the enhanced performance of PRA mixes was due to the reduced porosity and modification of the concrete micro-structure, including the formation of a greater number of coarser pores.

Furthermore, the carbonation curing of concrete containing RCA has a positive environmental impact by reducing natural aggregate consumption, minimizing landfill waste, capturing CO_2 during curing, and thus lowering greenhouse gas emissions. However, due to experimental limitations (30% carbonation for 24 hrs), the degree of carbonation may not be sufficient to achieve optimum carbonation benefit. Hence, future research is recommended on achieving 100% carbonation, optimum carbon uptake, and conducting detailed micro-structural analysis using scanning electron microscopy.

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Conflict of Interests

The authors have no conflict of interests to declare.

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