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Optimization of gas-solid carbonation conditions of recycled aggregates using a linear weighted sum method

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ABSTRACT

Optimization of pretreatment and various gas-solid interaction conditions is necessary for effective carbonation of recycled coarse aggregates derived from concrete wastes (RCA). This study demonstrated the use of linear weighted sum method to simultaneously optimize the pretreatment and carbonation test conditions. The optimization of the test conditions produced simplified, and eco-friendly pretreatments and drastically reduced the carbonation duration of RCA from 72 h to 3 weeks in previous studies to 18 h in this study. In addition, the optimum experimental conditions were validated by casting concrete using the carbonated RCAs. The optimized pretreatment and carbonation conditions were spraying Ca^{2+}-rich wastewater at a level of 60% of the 24 h water absorption of RCA, 12 h air drying and 6 h carbonation at 60 °C. This study also demonstrated that wastewater derived from ready-mix concrete batching plants can be utilized for spraying the RCAs to improve carbonation efficiency. Also, the mechanical and durability properties of concrete prepared with RCAs produced from the optimized conventional carbonation and pressurized carbonation were similar. Therefore, optimized conventional carbonation can be utilized as an alternative to pressurized carbonation. The improved mechanical and durability properties of concrete prepared with carbonated RCAs were attributed to the improved physical properties of carbonated RCAs and improved microhardness of the interfacial transition zone of the new mortar. The simplified, faster, ecofriendly, optimized pretreatment and carbonation conditions opens a new vista for industrial carbonation applications and optimization of gas-solid interactions.

1. Introduction

Much interests are now being paid on recycling recycled aggregates derived from construction and demolition wastes (CDW) due to sustainability concerns (Bao et al., 2020a; Silva et al., 2019; Xuan and Shui, 2011). Environmental sustainability concerns with respect to CDW includes leaching of hazardous chemicals from CDW, rapid natural resource depletion, land depletion due to landfill-based waste management, greenhouse gas emission and risk of landslides of landfilled CDW by rainfall (Bao et al., 2020b; Behera et al., 2014; Bonoli et al., 2021; Lu et al., 2017). The economic sustainability concerns includes increased production costs and time, creation of new jobs and industries, increased landfilling operation and maintenance costs as well as increased government expenditure on new landfills (Alsheyab, 2021; Zhang et al.,

Abbreviations: RCA, Recycled coarse aggregates; RAC, Concrete prepared with recycled coarse aggregates; CDW, Construction and demolition wastes; NA, Natural aggregates; NAC, Concrete prepared with natural aggregates; RMA, Regular mortar aggregates; CS, Compressive strength; PC, Pressurized carbonation; FCSE, Optimized conventional carbonation; FC, Conventional carbonation; RAC-FC, Concrete prepared with 24-h carbonated recycled coarse aggregates (RCA); RAC-PC, Concrete prepared with pressurized carbonated RCA; RAC-FCSE, Concrete prepared with optimized carbonated RCA; NSC24-25, Regular mortar aggregates non-sprayed and carbonated for 24 h @ 25 °C; SC24-40, Regular mortar aggregates wastewater-sprayed, non-dried and carbonated for 24 h @ 40 °C; SC12-60, Regular mortar aggregates wastewater-sprayed, non-dried and carbonated for 12 h @ 60 °C; SA12C6-60, Regular mortar aggregates wastewater sprayed 12-h air dried and carbonated for 6 h @ 60 °C; SE6C6-60, Regular mortar aggregates wastewater sprayed, 6 h dried in environmental chamber and carbonated for 6 h @ 60 °C; $MH_{new\ mortar}$, Microhardness of new mortar surrounding the regular mortar aggregate; MH_{RMA}, Microhardness of the carbonated and uncarbonated regular mortar aggregate; BD, Bulk density; WA, Water absorption; BEC, Bulk electrical conductivity.

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Table 1
Pre-treatment and carbonation conditions of RCA in previous researches.

References	Pre-treatment	Carbonation conditions	Benefits and drawbacks
Li et al. (2019)	• Drying in a chamber to obtain appropriate moisture content	• 20% CO_2 • 70±5% RH • 23 ± 2 °C • 3 days	• Drying to enhance carbonation. • Long carbonation time.
Kazmi et al. (2020)	• C Lime-treated, acid-treated and direct carbonation. • Drying at 25 °C and 50% RH for 3 days.	• 100% CO_2 • +0.8 bar • 24 h	• Lime-treatment before carbonation resulted in lowest WA and porosity of RCA.
Wang et al. (2020a)	• Air-dried before direct carbonation. • Impregnation with hydrated lime for 1 h. • Dried at 50 °C.	• 20±3% CO2 • 20±2 °C • 70 ± 5% RH • No pressure.	• RAC exhibited good properties.
Zhan et al. (2018)	• Presoaking in a saturated limewater for 3 days. • Pre-drying for 3 days at 26 °C & 50% RH.	• 100% CO_2 • +0.1 bar • 24 h • Room temperature	• Lower WA, higher CO_2 uptake. • Increased microhardness recorded by lime-soaked RCA. • High carbonation pressure and long time
Zhan et al. (2020)	• Soaking in limewater for 56 days. • Drying at 25 °C & 50±2% RH for 14 days.	• 100% CO_2 • +1 bar • 25 °C • 7 days	• Improvement in microhardness of RCA • Improvement of their interfacial bonding (ITZ) of RAC • High carbonation pressure and long time.

2019a, 2019b). Beside these, the other major issues of CDW management are attributed to rapid urbanization, low recovery and recycling rates, ineffective government policies, management deficiencies and inadequate capacity to manage the wastes (Zhang et al., 2019b; Aslam et al., 2020; Bao and Lu, 2020; Kabirifar et al., 2020; Ruiz et al., 2020). Recycled concrete aggregates from CDW (RCA) has thus been identified as an alternative source of aggregates to mitigate the negative impact of natural aggregate (NA) exploitation, as well as the waste management and environmental issues arising from CDW disposal (Muduli and Mukharjee, 2020).

Recycling of CDW offers potential opportunities for waste reduction, resource efficiency and resource conservation, aversion of environmental pollution and risks, reduction of government expenditure on landfill operations and management and creation of new jobs and industries (Silva et al., 2019; Zhang et al., 2019b; Bao and Lu, 2020; Agrela et al., 2021; Liu et al., 2021; Yazdani et al., 2021). To reap the benefits of environmental and economic benefits of CDW recycling, governments of various countries have increased landfilling costs and set up environmental programmes and policies to encourage recycling (Ghaffar et al., 2020; Jain et al., 2015; Kurniawan et al., 2021; Li et al., 2020a; Turkyilmaz et al., 2019; Wang et al., 2021). In addition, several companies have embraced in-house recycling/upcycling to achieve project efficiency, sustainable construction and improve their profits (Wang et al., 2021).

Despite numerous studies confirming the feasibility of using RCA in concrete for applications (Ceia et al., 2016; Majhi et al., 2018; Pereira-De-Oliveira et al., 2014; Rao et al., 2011), its use is largely limited owing to its inferior properties compared to NA, leading to reduction in mechanical and durability properties of recycled aggregate concrete (RAC) compared to natural aggregate concrete (NAC). Even though some studies reported that RCA application rate can reach up to 100% depending on the quality (Le and Bui, 2020; Rockson et al., 2020; Xuan et al., 2012), most standards limit RCA utilization in different civil engineering works.

In recent years, carbonation treatment has attracted much interest to enhance the properties of RCA as shown in Table 1. It generally involves two steps, namely pre-treatment by using other foreign admixtures (i.e. Ca-rich foreign admixture, $Ca(OH)_2$ solution) and adjustment of proper carbonation conditions (i.e. temperature, pressure, duration, drying condition) to enhance carbonation. Recent studies have revealed that pre-treatment prior to carbonation had beneficial effects such as reduction of water absorption of the carbonated RCA, improved their strength and CO_2 uptake (Ouyang et al., 2020; Zhan et al., 2018). The significant improvement of RAC was attributed to the enhancement of the interfacial transition zone (ITZ) between the new and old mortar (Zhan et al., 2018; Shaban et al., 2019). This is due to the formation of a dense and compacted ITZ (Zhan et al., 2018; Hanif et al., 2020; Kim et al., 2018). Since the total aggregates occupy 55–80% of concrete by volume (Mindess et al., 2002; Saxena and Pofale, 2017; Tufail et al., 2017; Wang et al., 2018) and coarse aggregates occupy between 40 and 50% of concrete by volume (Saxena and Pofale, 2017; Li et al., 2016), therefore, utilization of the optimized carbonated RCA is a low-cost and eco-friendly alternative to improve the mechanical performance and durability of recycled concrete products.

Previous studies reported that the efficiency of the gas-solid carbonation of RCA is influenced by various experimental conditions, including pre-treatment (spraying/soaking) methods, temperature, carbonation duration, and type of pre-drying conditions (Fang et al., 2020),. In most of the studies, the pressurized carbonation of RCA resulted in effective carbonation and improved strength development of RAC (Xuan et al., 2016). Several processing variables including carbonation pressure, moisture contents of samples, CO_2 concentrations were also reported to affect carbonation efficiency (Bao et al., 2017; Huijgen et al., 2005; Zhan et al., 2016; Zhao et al., 2015). In addition, porosity and permeability reduction alongside strength development of concrete have been linked to pressurized carbonation (Bao et al., 2017; Hay et al., 2021; Hyvert et al., 2010). However, the industrial operations of pressurized carbonation is more challenging compared to conventional carbonation by applying a CO_2 gas stream flowing through the recycled aggregates continuously under ambient pressure conditions.

Furthermore, recent studies have also identified moderate elevated temperature carbonation as an effective means to accelerate the rate of carbonation reaction, increase CO_2 uptake and promote the formation of carbonation products both on the surface and interior layers of RCA (Wang et al., 2019, 2020b). Moderate elevated temperature carbonation promotes the formation of $CaCO_3$ (calcite), enhances CO_2 molecular mobility and prevents pore blockage by a dense protective carbonated layer (Liang et al., 2020; Liu et al., 2018). In contrast, excessive elevated temperature carbonation promotes leaching of Ca^{2+} ions, reduces CO_2 solubility leading to high pH of the water and lowers dissolution rate of Ca^{2+} (Liang et al., 2020). In addition, there is lack of consensus on the optimum elevated temperature to be utilized to carbonate recycled aggregates (Azdarpour et al., 2017; Qian et al., 2016).

Therefore, this study aims to optimize the conventional carbonation process as stated above for effective carbonation of RCA using regular mortar aggregates (RMA) prepared in the laboratory and validated the optimal conditions by casting concrete using RCA collected from a local recycling plant concrete. The objectives of this study are:

- To investigate the influence of carbonation conditions on the physical properties of regular mortar aggregates
- To optimize the carbonation conditions including pretreatment by using wastewater sourced from concrete batching plants as a calcium-rich additive (Fang et al., 2020) and carbonation conditions using a linear weighted sum method
- To investigate the mechanical and durability properties of RAC containing the optimally treated RCA collected from a local recycling plant.

Table 2
Chemical compositions of conventional and white OPC (% by mass).

Type	CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	Na_2O	K_2O	SO_3
Conventional OPC	63.15	19.61	7.52	3.32	2.14	0.13	0.32	2.03
White OPC	67.60	20.20	4.78	0.21	1.48	–	0.73	4.90

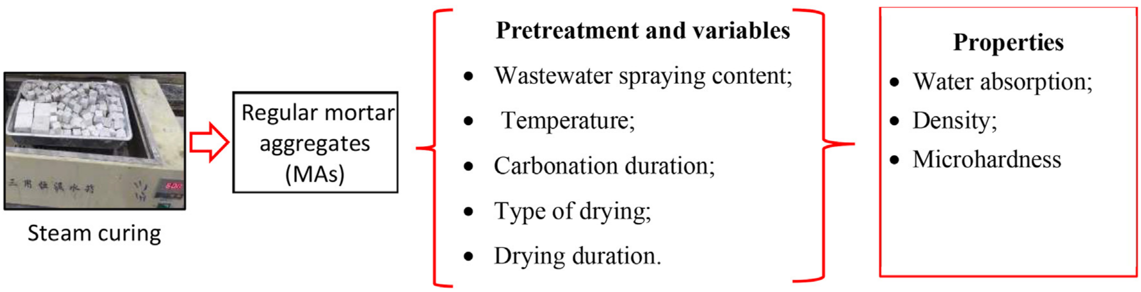


Fig. 1. Conventional gas-solid carbonation conditions of RMAs and properties tested.

This study is significant by revealing the optimum carbonation conditions for improving the properties of RCA using the linear weighted sum method. The optimized pretreatment and carbonation time were reduced significantly which makes it attractive to the construction industry. This study also demonstrated the usefulness of Ca-rich wastewater from ready-mixed concrete batching plant for improving the properties of RCA. This study also revealed the correlations between the mechanical and durability properties of concrete prepared with carbonated RCA and conventional coarse aggregates.

2. Materials and methodology

2.1. Materials and preparation of specimens

Regular mortar aggregates (RMAs) prepared in the laboratory were used in this study to investigate the influence of various pretreatment conditions and gas-solid carbonation test conditions in order to overcome inherent variability of recycled concrete aggregates (RCA) collected from recycling plants (Ouyang et al., 2020). White Ordinary Portland cements (OPC) provided by Green Island Cement Limited complying with ASTM Type 1 were used for preparing RMAs in combination with a siliceous river sand and water, while conventional OPC was used for preparing the new mortar. The white cement helped to easily delineate/differentiate the interface between the old mortar (RMAs) and the new mortar in RAC. The chemical compositions of the conventional and white cements are listed in Table 2. The mix proportion for producing RMAs was 0.5:1:2 (water: cement: sand). After homogeneously mixing the proportioned constituents in a mixer, casting of RMA was done in 20 × 20 × 20 mm steel moulds. The fresh RMAs were demoulded after 24 h and then steam cured for 7 days at 60 °C using a steam bath. White OPC was used in casting the regular mortar aggregates while conventional OPC was used in casting the mortar surrounding the regular mortar aggregates to easily delineate the regular mortar aggregate (representing the old mortar) from the surrounding new mortar. The OPC is called white because of its white colour and it is white because of its negligible ferric oxide content. In addition, conventional OPC was used in the concrete mix designs. The chemical compositions of both conventional OPC and white OPC were presented to show their similarity/differences, calcium oxide (CaO) content and the potential major ions contributing to carbonation. As shown in Table 2, the main difference is that conventional OPC has ferric oxide (Fe_2O_3) while white OPC has negligible ferric oxide. The similarity is that both conventional OPC and white OPC have similar CaO and SiO_2 contents, both of which enhance carbonation and also have similar pozzolanic content. Likewise, the major oxide in both conventional OPC and white OPC is CaO. Therefore, white OPC can be utilized to replace conventional OPC to simulate carbonation experiments similar to conventional OPC.

Table 3
Chemical compositions of wastewater (ppm).

Items	Ca^{2+}	K^+	pH
Wastewater	1211.8 ± 69.3	606 ± 59.6	12.8 ± 0.1

Meanwhile, the specimens for testing the microhardness of the carbonated RMAs and the new mortar were prepared by casting the mortars into 40 × 40 × 40 mm plastic moulds in two layers. First, the fresh new mortar prepared with the conventional OPC was cast into the moulds to reach approximately one-quarter of the height and vibrated for 10 s. Afterwards, respective RMAs (simulating the old mortars) were placed into the middle part of the moulds. Then, the fresh new mortar was further cast to fully cover the RMAs and vibrated again for 10–20 s. These specimens were demoulded after 24 h and steam cured at 60 °C for 7 days in a steam bath as shown in Fig. 1.

The chemical compositions of wastewater collected from a local ready-mixed concrete plant for pretreating the RMA and RCA are shown in Table 3. It was an alkaline wastewater with a rich Ca^{2+} content. The precondition and carbonation treatments optimized for the regular mortar aggregates were: (i) amount of wastewater sprayed on the mortar aggregates, (ii) the temperature of carbonation, (iii) drying duration and drying type as shown in Fig. 1. The properties of the regular mortar aggregates investigated were water absorption, density and microhardness as shown in Fig. 1.

2.2. Conventional gas-solid carbonation conditions of RMAs and preparation of recycled aggregate concrete (RAC)

2.2.1. Conventional gas-solid carbonation conditions of RMAs

The RMAs were placed in one layer inside a flat container as shown in Fig. 2 (a) and sprayed with wastewater with an inclined hand-held spraying device similar to previous studies (Fang et al., 2020, 2021). Before commencement of spraying, the container with the RMAs was placed on the weighing balance and the balance was zeroed. The RMAs were manually sprayed from the top and to ensure even spraying, the RMAs were turned manually after spraying a quarter of wastewater mass required for spraying. The wastewater quantities sprayed were 20%, 40%, 60% and 80% of total water absorption of RMAs while another RMA was soaked for 5s for comparison. The wastewater used was obtained from a local ready-mixed batching plant in Hong Kong, which was used to wash the returned concrete or concrete trucks (Xuan et al., 2018).

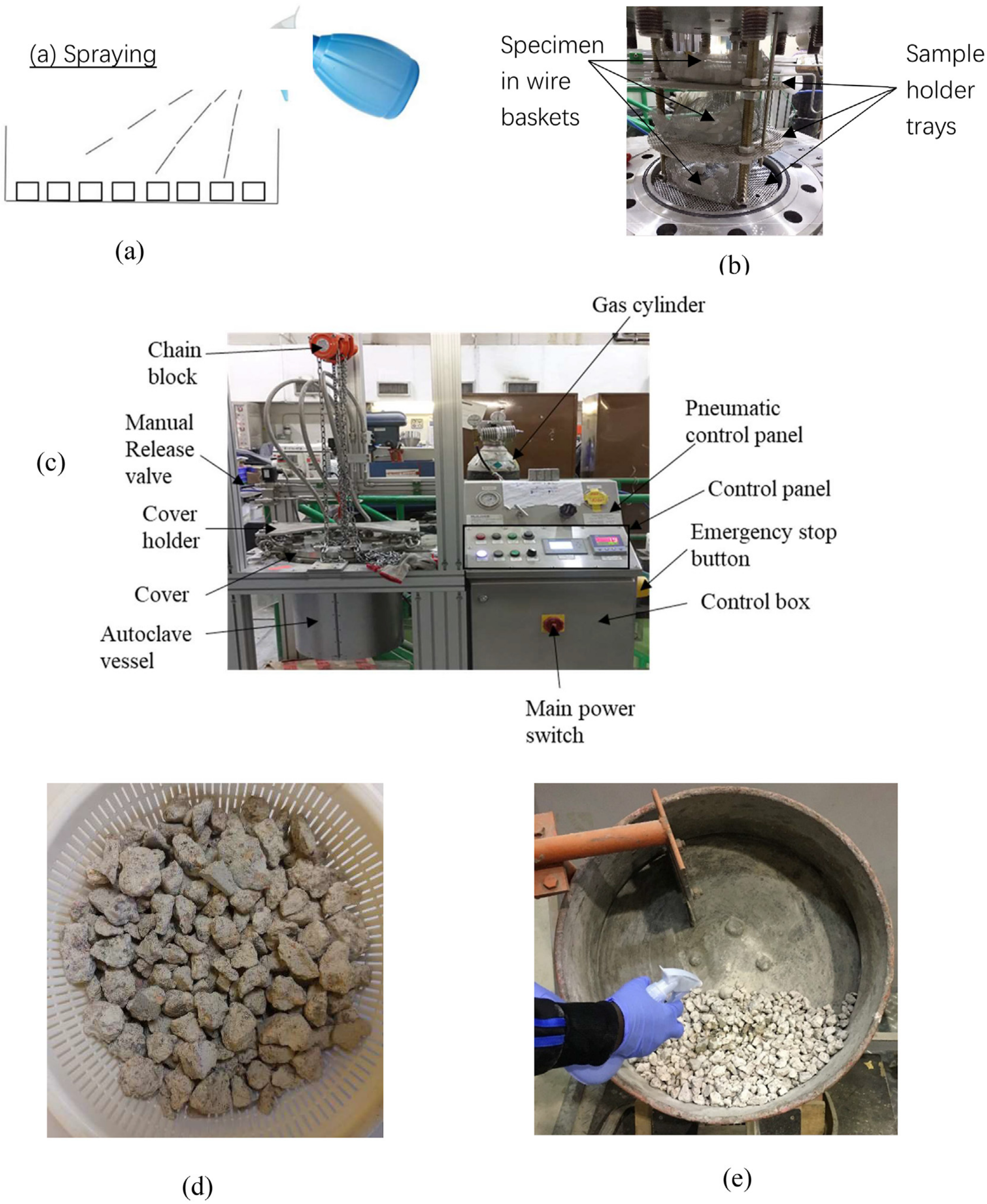


Fig. 2. (a) Spraying of regular mortar aggregates (b) Placement of regular mortar aggregates in layered autoclave chamber (c) Autoclave chamber for conventional and pressurized carbonation (d) RCA from pure demolished waste (e) Spraying of RCA with wastewater in inclined mixer using hand-held spraying device.

The wastewater is alkaline with a rich Ca^{2+} content. An industrial grade CO_2 source (>99% purity CO_2 gas) was used for carbonation of RMAs and RCA in this study. Before placement of RMAs and RCAs into the autoclave chamber, the manual release valve was opened to make sure there is zero pressure in the autoclave. Thereafter, the cover of the autoclave was opened first by loosening the nuts with the torque wrench followed by lifting the autoclave cover with the chain block in combination with the cover holder. To ensure uniform carbonation, the sprayed RMAs were placed in three wire baskets and each basket was placed in each of the three layers of the autoclave as displayed in Fig. 2(b).

With the aid of the chain block and cover holder, the specimens were lowered into the autoclave chamber as shown in Fig. 2 (c). The cover holder was removed and the nuts were tightened with the torque wrench. For conventional carbonation, the manual release valve was opened and there was no need to set pressure while for pressurized carbonation the valve was closed and the pressure of 1 bar was utilized for pressurized carbonation which is below the 14 bar pressurized capacity of the autoclave equipment. The main power was switched on and with the aid of the temperature and pressure regulators on the control panel, the experimental condition was set and CO_2 gas was injected into the chamber at a flow rate of 1±0.5 lit/min. The specimen for air drying conditions were air dried at ambient temperature of 25 °C before carbonation inside the laboratory while the specimen for environmental chamber drying were dried at ambient temperature of 25 °C and 50% relative humidity in the environmental chamber and the specimen for direct carbonation were immediately placed in the autoclave after wastewater spraying. The experimental carbonation conditions of RMAs as well as study framework are shown in Fig. 1. The temperature investigated were 25 °C, 40 °C, 60 °C and 80 °C while the carbonation duration investigated were 3 h, 6 h, 12 h, and 24 h. In addition, the type

Table 4
Water absorption and bulk density of RCAs after subjection to different carbonation conditions.

Test conditions	Water absorption (%)		Bulk density (kg/m^3)	
	5–10 mm	10–20 mm	5–10 mm	10–20 mm
Uncarbonated	7.73	6.66	2193	2223
FC	7.02	6.39	2220	2330
PC	6.64	5.64	2260	2290
FCSE	6.53	5.90	2250	2270

of drying investigated were air drying before carbonation, environmental chamber drying before carbonation and direct carbonation (no drying). In addition, the effects of drying durations of 3 h, 6 h, 12 h and 24 h for both air drying and environmental chamber drying were investigated.

2.2.2. Preparation of recycled aggregate concrete (RAC)

Construction waste materials were collected from a local construction waste recycling company in Hong Kong. The pure concrete waste materials, without stone, were crushed in the laboratory with the aid of automated crusher to produce the RCAs utilized in this study, displayed in Fig. 2 (d). After crushing, the RCAs were sieved into two size ranges namely 5–10 mm and 10–20 mm in line with the mix design. To achieve an even spraying, the RCAs were sprayed in an inclined mixer at a rotating speed of 10 rev/min for every 5 kg batch as shown in Fig. 2 (e). The RCAs were stored in plastic bags and sealed prior to carbonation. After carbonation, they were stored again in plastic bags prior to concrete casting. The RCAs were carbonated using the optimized conditions obtained in RMA experiments. For comparison, the same batch of RCAs were carbonated at different conditions, namely: conventional carbonation at ambient temperature for 24 h (FC), pressurized carbonation at ambient temperature for 24 h (PC) and the optimized elevated temperature carbonation (FCSE) shown in Table 5. Table 4 lists the water absorption and bulk density values of RCAs after subjecting to different carbonation conditions.

Based on dry bulk densities for uncarbonated and carbonated RCAs displayed in Table 4, the RCAs can be described as normal weight aggregates (NWA) since the apparent bulk density range of 2193–2260 kg/m^3 for 5–10 mm RCA and 2223–2330 kg/m^3 for 10–20 mm RCA were within 2100–3000 kg/m^3 required for NWA (BS-EN-206-1, 2000). In addition, it was observed that similar bulk densities were obtained by both optimized conventional carbonation and pressurized carbonation.

Also, it was observed that RAC prepared at optimized conventional carbonation conditions (RAC-FCSE) recorded lower water absorption compared to conventional carbonation (RAC-FC) and achieved comparable water absorption with RCA subjected to pressurized carbonation (RAC-PC). The water absorption obtained in our study was comparable to results from a previous study (Xuan et al., 2017). This implies improvement in RCA properties by conventional carbonation was limited due to the rapid densification of the surface (Fang et al., 2021) and limited supply of moisture. Therefore, optimized carbonation is preferable owing to reduced water absorption of carbonated RCAs and slightly improved bulk densities. Recent studies also reported similar improvements in RCA properties due to carbonation and wastewater pretreatment (Li et al., 2019; Fang et al., 2020, 2021). However, pre-treatment with wastewater prior to optimized conventional and pressurized carbonations supplied extra Ca^{2+} ions in the pore solution which provided extra carbonation products resulting in improved and comparable bulk density and lower water absorption of RC (Fang et al., 2021). Therefore, optimized conventional carbonation and pressurized carbonation have similar carbonation efficiency.

For validation purposes, NAC and four types of RACs prepared with real RCAs from a local recycling plant were cast and the mix proportions are shown in Table 5. The four types of RACs were prepared using 100% replacement of NAs with the carbonated RCAs. Based on their respective water absorptions, water adjustments were made for each batch to ensure the same effective water to cement ratio of 0.6. For each mix, four concrete cylinders with dimensions of $\Phi 100 \times 200$ mm and four cubes with the dimensions of $100 \times 100 \times 100$ mm were prepared. After casting, the specimens were covered with a plastic sheet and cured in the laboratory environment for 24 h before demoulding and then cured by full immersion in a water tank at ambient temperature for 7 day and 28 days in the laboratory. The mechanical and durability properties of NAC and RACs, including compressive strength, elastic modulus, sorptivity and bulk electricity conductivity, were tested on specific days and evaluated for comparison.

2.2.3. Linear weighted sum method to optimize testing conditions

Optimization studies have attracted a lot of interest in diverse disciplines to reduce production cost, improve production process efficiency and product quality and thereby reduce maintenance expenses. Two commonly utilized optimization techniques in concrete design are Taguchi and Response Surface Methodology. Recent Taguchi optimization studies have shown that processing of concrete materials/concrete at optimized conditions is crucial to achieve best mechanical and durability properties (Chen et al., 2017; Mubina et al., 2019; Onoue et al., 2019). Taguchi design of experiment utilizes fractional factorial design approach to accomplish single, dual and multi-objective optimization of concrete composition and processing conditions in an efficient and systematic manner. Though Taguchi has the potential to reduce experimental runs with L^9 orthogonal arrays, the time and cost-effectiveness of Taguchi approach depends on the array type selected which can range between L9 to L36. Taguchi optimization method utilizes the control factors to eliminate the noise caused by the uncontrollable factors to achieve desired target performance while minimizing the associated loss. Based on desired response, optimum control factors are identified and selected from consideration of the interactive effects of the signal-to-noise ratios.

Taguchi optimization applications are diverse and has been utilized in composition optimization of alternative binders, tertiary blended cements, artificial fly ash aggregates, fly-ash based geopolymer mortar, sandcrete block and lightweight aggregates to mention a few (Chen et al., 2017; Onoue et al., 2019; Ikeagwuani et al., 2020; Peyronnaard and Benzaazoua, 2012; Shivaprasad and Das, 2018; Teirmotashlu et al.,

Table 5
Mix proportions for natural/recycled aggregate concrete (kg/m^3).

Items	Treatment methods of RCAs	Cement	Sand	Agg. 5–10 mm	Agg. 10–20 mm	Effective W/C
Control	Natural aggregates	325	752	282	846	0.6
RAC	No carbonation treatment					
RAC-FC	Conventional carbonation for 24 h[a]					
RAC-PC	Pressurized carbonation for 24 h at 1 bar					
RAC-FCSE	•Spray wastewater (60% of water absorption) •Conventional carbonation •Temperature: 60 °C •Duration: 6 h					

[a] carbonation by applying a CO_2 gas stream flowing through the recycled aggregates continuously under ambient pressure conditions.

2018). Specifically, Taguchi optimization method has been utilized in reduction of production duration of concrete pavement, reduction of production temperature of asphalt mix, eco-friendly and economic design of self-compacting cement composite through 64% cement replacement with supplementary cementitious materials (SCM), improve corrosion durability of RC structures, composition optimization of pervious concrete pavement, permeable asphalt concrete, lightweight concrete, alkali-activated fly ash binders and identify the optimum processing temperature for binders (Cabrera-Luna et al., 2018; Ghanei et al., ; Hinislioglu and Bayrak, 2004; Hoseinpour-Lonbar et al., 2020; Jalal et al., 2019; Joshaghani et al., 2015; Panagiotopoulou et al., 2015; Slebi-Acevedo et al., 2020; Suttaphakdee et al., 2016). The major disadvantages of Taguchi optimization approach are limited visualization of multiple interactive effects of input factors on desired product properties, pre-screening requirement of variables to select the most important factors and their suitable ranges as well as accurate interpretation of results to select optimum conditions.

RSM (response surface methodology) is a mathematical and statistical technique for experimental design, which utilizes desirability optimization approach to select the optimum process conditions or composition (Chelladurai et al., 2020). Most RSM optimization studies focused mainly on material composition optimization and was applied to plastic fibre-reinforced recycled concrete with improved cracking properties and strength, hybrid fibre-reinforced concrete, clay-based self--compacting rigid concrete pavement, mortar, steel fibre reinforced ultra-high performance concrete, rapid-repair cement mortar, blended concrete, design eco-efficient UHPC and pervious recycled concrete (Ahmed et al., 2020; Bankir and Sevim, 2020; Busari et al., 2019; Delarami et al., 2021; Dingqiang et al., 2020; Hassan et al., 2020; Rivera et al., 2019; Zhang et al., 2020). By specifying the desirable minimum and maximum ranges of output properties (quality ranges), the composition or process conditions that gives the highest desirability value is selected as the optimum (Chelladurai et al., 2020; Bankir and Sevim, 2020; Dingqiang et al., 2020). With the aid of in-built ANOVA, input parameters that have significant effect on specified product qualities are found while regression model equations are generated for the responses (Zhao et al., 2015; Bankir and Sevim, 2020). With the aid of the 2D and 3D visualization graphs it generates, RSM aids understanding of the effects of the input parameters on the output responses (product quality) (Delarami et al., 2021; Hassan et al., 2020; Aydemir and Ozkul, 2020; Carrion et al., 2020; Usman et al., 2020).

RSM has enormous capabilities and has been applied to identify significant parameters to optimize both fresh and hardened properties of concrete and varied composition of concrete mixtures to achieve different target slumps and different target strengths (Fardin and dos Santos, 2021; Li et al., 2020b; Mohammed et al., 2018). In addition, RSM can assess the suitability of alternative fine aggregates, SCMs and fibres in concrete, and rank their effectiveness for different construction applications (Rivera et al., 2019; Mohammed et al., 2019a, 2019b; Noroozi et al., 2019; Pinheiro et al., 2020; Qi et al., 2020). RSM has also been applied to study the effect of porous aggregate properties on autogenous shrinkage reduction (Sun et al., 2019) and Los Angeles value of aggregates on concrete strength (Tunc and Alyamac, 2020). In summary, RSM can be utilized in both material selection and process configuration to replace conventional construction materials with new eco-friendly materials without sacrificing desired product qualities. Based on literature review, multi-stage optimization using RSM is scarce as well as its application to concrete durability. The major disadvantage of RSM is large experimental runs requirements which is time-consuming and expensive. To minimize the drawbacks of both optimization methods, a recent study advocated coupling both RSM and Taguchi optimization methods to maximize their benefits (Zhang et al., 2019b).

In this study, linear weighted sum optimization method was applied because it is suitable for one-factor at a time (OFAT) experiments utilized in this research. Linear weighted sum is a simplified optimization method recommended by some researchers (Sojobi, 2019; Sojobi and Liew, 2020) and has several advantages such as simplified computation and elimination of bias, allows consideration of several properties concurrently and facilitates ranking of the outcome of experimental conditions to determine the optimized experimental condition. In addition, it can also be utilized to rank the effectiveness of the various pretreatment conditions on the physical properties of the aggregates. Optimizing the pretreatment conditions of RCA before usage has the potential to effectively carbonate RCA, shorten the accelerated carbonation process and improve durability of recycled concrete (Xuan et al., 2018; Lin et al., 2015; Zhan et al., 2019). The testing condition with the highest rank is selected as the optimum testing condition which can be utilized in RAC production.

2.3. Determination of physical properties of RMAs

The water absorption and bulk density of RMAs before and after carbonation treatments were determined in accordance to BS-EN-1097-6 (BS-EN-1097-6, 2013) as well as the microhardness. The water absorption and bulk density were determined using the wire basket method because it gave more consistent results compared to the pycnometer method, it is less prone to experimental errors, faster and more convenient. Based on previous study (Fang et al., 2020), improvements from carbonation and spraying with wastewater are reflected in reduction of water absorption, increase in bulk density and improvement in microhardness of the RMAs. The variables tested for RMAs were wastewater spraying content, temperature, carbonation duration, type of drying and drying duration.

2.4. Determination of mechanical and durability properties of RACs

2.4.1. Compressive strength and elastic modulus

The compressive strength test of RAC was conducted using 100 × 100 × 100 mm cubes at a loading rate of 0.6 MPa/s at 7 and 28 days. The elastic modulus was determined in accordance with BS-EN-12390 (BS-EN-12390, 2013). The stress-strain curve of concrete was firstly determined at the loading speed of 0.6 MPa/s. The displacement of each concrete specimen was measured by two linear variable differential transformers (LVDTs). The average value was used to calculate the strain of the specimen. The elastic modulus E was then determined as the secant modulus of the stress–strain curve at the range of 5% to 1/3 peak stress.

2.4.2. Sorptivity (Capillary water absorption)

The sorptivity test of RAC was carried out by using three concrete discs (Ø 100 × 50 mm) in accordance with ASTM-C1585 (ASTM-C1585, 2013). The sorptivity was calculated by using the increase in mass due to water uptake, divided by the cross-sectional area of the specimen exposed to the water. The weight variations of the specimens were measured at 0, 1, 5, 10, 20, 30 min and 1, 2, 3, 4 5, 6 h for the first stage and 1, 2, 3, 4, 5, 6 and 7 days for the second stage. A recent study reported reduction in sorptivity of carbonated and wastewater-sprayed RCA compared to carbonated and uncarbonated RCA (Fang et al., 2021).

2.4.3. Bulk electrical conductivity

Measurement of bulk electrical conductivity of RAC was performed on three concrete discs (Ø 100 × 50 mm) according to the procedures described in ASTM-C1760 (ASTM-C1760, 2012). This test measured the electrical current through a saturated specimen with a potential (U) of 60 V DC across the ends of the specimen. The current (I) was measured at 1 min after the voltage was first applied. The electrical conductivity (σ, S/m) was then calculated by an equation described in a publication (Xuan et al., 2017). A recent study reported 15.1% reduction in bulk electrical conductivity of concrete with carbonated RCAs (Xuan et al., 2017).

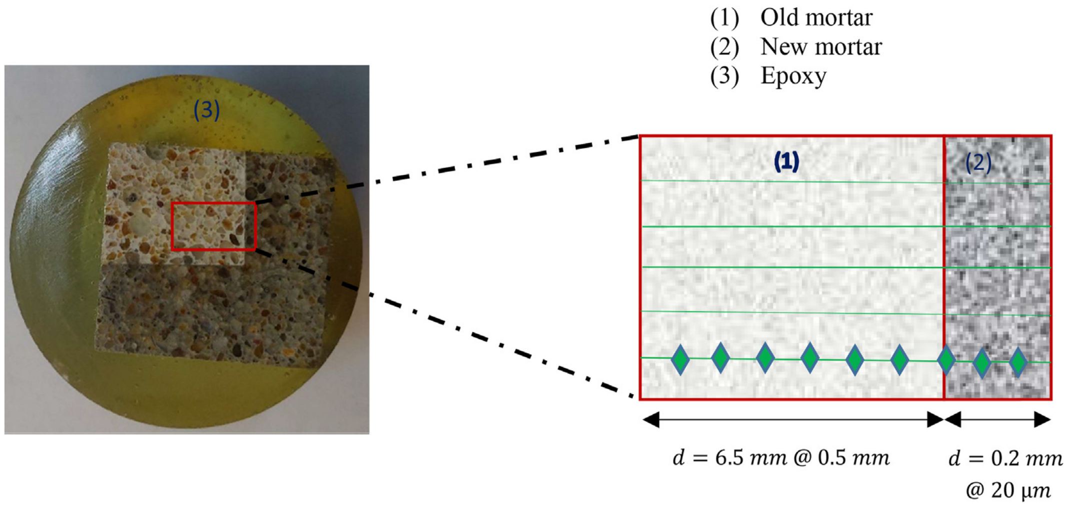


Fig. 3. Test protocol of microhardness indentation around the interface between carbonated RCA (old mortar) and new mortar. Green diamonds represent the testing points.

Table 6
Experimental conditions and physical properties of RMA.

Items	Conditions	Bulk density (kg/m^3)	Water absorption (%)
NSC 24-25	-Non-sprayed -Carbonation: Conventional -Carbonation duration: 24 h -Temperature: 25 °C	2108 ± 5.00	6.86±0.15
SC24-40	-Wastewater-sprayed -No drying (Direct carbonation) -Carbonation: Conventional -Carbonation duration: 24 h - Temperature: 40 °C	2089 ±5.95	6.27±0.02
SC12-60	-Wastewater-sprayed -No drying (Direct carbonation) -Carbonation: Conventional -Carbonation duration: 12 h - Temperature: 60 °C	2220±21.92	5.12±0.50
SA12C6-60	-Wastewater-sprayed -Drying type: Air drying at 25 °C -Drying duration: 12 h -Carbonation: Conventional -Carbonation duration: 6 h - Temperature: 60 °C	2167 ±14.13	4.89±0.68
SE6C6-60	-Wastewater sprayed -Drying duration: 6 h -Drying type: Environmental chamber at RH: 50% -Carbonation duration: 6 h - Temperature: 60 °C	2199±14.00	5.06±0.72

2.4.4. *Microhardness measurement of ITZ between carbonated aggregates and new mortar*

The carbonated RMA and the new mortar were polished by a grinding and polishing equipment (Buehler AutoMet 250) after fixing in an epoxy resin according to the previous established procedures (Zhan et al., 2018). The samples were stored in a vacuum chamber for 24 h before testing the microhardness. The microhardness measurements were performed by using a digital optical Vickers microhardness tester (HVX-1000A, China) equipped with 40 x measurement lens and 10 x objective lens. For the new mortar, the microhardness was conducted up to 160 μm away from the surface of RMA at an interval of 40 μm. For the carbonated RMA, microhardness test was conducted up to 6.5 mm inside the surface of RMA at an interval of 0.5 mm. The microhardness test protocol is shown in Fig. 3. The number of microhardness measurements at one point is between 6 and 8 and the standard deviation was determined using excel spreadsheet. A recent study reported increase in ITZ of RCA by carbonation treatment (Zhan et al., 2020).

RCA is covered by old mortar and during carbonation of RCA, it is the old mortar that is carbonated. When RCA is used in production of new concrete, a new interfacial zone is formed between the old mortar and the new mortar. Our experiment simulates the old mortar and the new mortar using the carbonated regular mortar aggregates that was surrounded with new mortar. Therefore, the microhardness results of RMA and the surrounding new mortar represents the microhardness of the real RCAs and the new mortar. The advantage of this method is that it avoids the variability of attached mortar of RCAs (Jayasuriya et al., 2018).

3. Results and discussion

3.1. *Influence of test conditions on properties of RMAs*

3.1.1. *Bulk density and water absorption*

Table 6 shows the experimental conditions and physical properties of the carbonated RMAs. The 12-h direct carbonation of the wastewater-sprayed RMAs at 60 °C (SC 12–60) exhibited the highest bulk density, followed by the wastewater-sprayed RMAs dried in an environmental chamber for 6 h, and carbonated for 6 h at 60 °C (SE6C6-60). The wastewater-sprayed RMA air-dried for 12 h and carbonated for 6 h at 60 °C (SA12C6-60) had the third highest bulk density. All the three RMAs mentioned above recorded higher bulk densities than that of the non-sprayed RMAs prepared using the conventional carbonation at the ambient temperature (control) (NSC24-25). However, the wastewater-sprayed RMAs carbonated at 40 °C (SC24-40) recorded the lowest bulk density compared to the control (NSC24-25). This implies the wastewater spraying combined with an appropriate elevated temperature and drying condition is beneficial and crucial to improve the density of RMA and carbonation efficiency.

It was also found that the wastewater-sprayed RMAs air-dried for 12 h prior to carbonation (SA12C6-60) recorded the lowest water absorption of 4.89%, which represented a reduction of 28.7% in the water absorption when compared to the control (NSC24-25). This implies that the pre-drying process of the wastewater-sprayed RMAs prior to carbonation is essential to reduce the water absorption of the carbonated RMAs. A low water absorption is desirable for RCA to achieve significant improvement in the mechanical and durability properties of RAC.

Table 7
Optimization of wastewater spraying quantity for the carbonated RMA.

Spray quantity	Final sum	Ranking
20%	1.08	2
40%	0.77	4
60%	1.39	1
80%	1.00	3
Soaking	0.75	5

Table 8
Optimization of temperature for the carbonated RMA.

Temperature (°C)	Final sum	Ranking
40	0.57	2
60	1.94	1
80	0.49	3

Table 9
Optimization of drying condition for the carbonated RMA.

Test conditions	Normalized Final sum	Ranking
3 h direct carbonation	0.81	11
6 h direct carbonation	0.86	10
12 h direct carbonation	0.87	9
24 h direct carbonation	0.89	8
3 h air drying before carbonation	1.08	4
6 h air drying before carbonation	1.07	5
12 h air drying before carbonation	1.11	3
24 h air drying before carbonation	1.05	6
3 h environmental chamber drying before carbonation	0.97	7
6 h environmental chamber drying before carbonation	1.14	2
24 h environmental chamber drying before carbonation	1.15	1

3.2. Optimization of test conditions to enhance the properties of RMA

3.2.1. Optimization of test variables using the linear weighted sum method

The test variables (amount of wastewater spraying; temperature; carbonation durations; pre-drying method) were optimized using the linear weighted sum method to optimize the testing conditions. The amount of wastewater sprayed on the surface ranged from 20% to 80% of the water absorption value of the RMA while a soaking treatment in wastewater for 5 s was also used for comparison. The elevated temperature investigated ranged from 40 °C to 80 °C. The optimization results are shown in Table 7. Similarly, for the optimization of the temperature, the results are displayed in Table 8. Subsequently, based on the results, the optimum wastewater spraying amount of 60% and the optimum temperature of 60 °C were recommended to improve the quality of the recycled aggregates.

Table 9 shows the optimization of carbonation durations and drying methods (namely direct carbonation, pre-air drying before carbonation, and pre-environmental-chamber drying before carbonation). It was found that the 24-h pre-environmental-chamber drying attained the best results, followed by the 6 h pre-environmental-chamber drying and the 12 h air drying before carbonation. Since the difference between the 24 h and 6 h pre-environmental-chamber drying (228.25 and 227.81) is insignificant, the 6 h pre-environmental-chamber drying process was preferable to save time and energy costs. Furthermore, to improve the efficiency of the carbonation process, the air-drying processes can also be utilized. Therefore, in this study, a 12-h air drying period, and the 6 h elevated temperature carbonation was selected for treatment of wastewater-sprayed recycled aggregates.

3.2.2. Ranking of various test conditions

The relative importance of all the tested conditions listed in Table 6 was also investigated using the linear weighted sum method and the normalized results are displayed in Table 10. The pre-drying process recorded the highest impact on the bulk density, followed by carbonation duration. In addition, wastewater spraying recorded the highest influence on the water absorption, followed by carbonation duration alone. In terms of the overall physical properties of RCA, a combination of pre-drying and carbonation duration recorded the highest influence, followed by carbonation duration alone and wastewater spraying.

In summary, the optimized carbonation conditions are: (i) spraying with wastewater containing additional Ca^{2+} at the level of 60% of the 24-h water absorption of recycled aggregates, (ii) followed by a 12-h air drying period and (iii) 6-h elevated temperature carbonation at 60 °C.

4. Mechanical properties and durability of RAC prepared with the optimized carbonated RCA

4.1. Mechanical properties of NAC and RACs

After finding the optimized carbonation conditions, and by using the carbonated RCAs listed in Table 4, different types of RACs were prepared as shown in mixture proportions displayed in Table 5. Table 11 lists the mechanical properties of RACs. The optimized RAC-FCSE recorded the highest 7-day compressive strength (CS_7) of 29.1 MPa, while RAC-PC (pressurized carbonation) recorded the highest 28-day compressive strength (CS_{28}), followed by RAC-FCSE with CS_{28} of 35.6 MPa. The increase in 28-th day compressive strength (CS_{28}) was 9.1%, 5.8% and 3.9% for RAC-PC, RAC-FCSE and RAC-FC, respectively, compared with the uncarbonated counterpart (RAC). Also, utilization of 100% RCA in concrete resulted in 23.4% reduction in elastic modulus. However, a slight improvement in elastic modulus of concrete with carbonated RCA was observed. The elastic modulus was higher than elastic modulus of 19.9–21.9 GPa obtained in another study with 0% and 3% steel fibre at 100% RCA (Kachouh et al., 2019). Although the mechanical properties in terms of compressive strength and E values did not differ much, higher improvements were noticed in the durability properties.

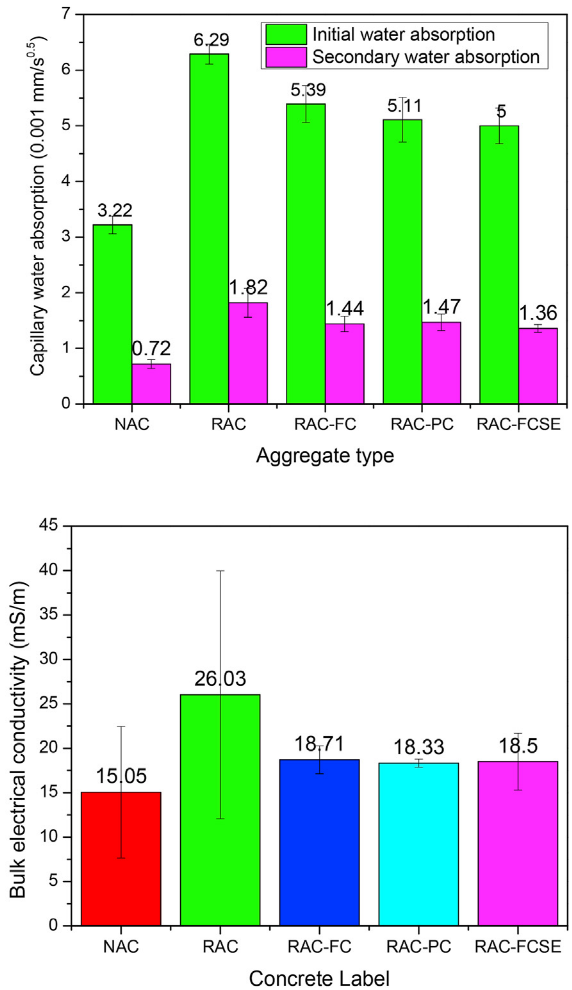
Table 11
Mechanical properties of RAC prepared with different carbonated RCAs.

Label	CS_7 (MPa)	CS_{28} (MPa)	Elastic modulus (GPa)
NAC (control)	36.9±1.1	46.8 ±0.9	29.5±0.7
RAC	27.7 ± 0.7	33.6±1.6	22.6± 0.4
RAC-FC	27.2 ± 0.3	34.9 ± 0.1	23.1 ± 0.8
RAC-PC	28.6± 0.4	36.7±0.1	23.1± 0.8
RAC-FCSE	29.1± 0.4	35.6±0.6	23.2± 0.4

Table 10
Ranking of influence of test conditions on the physical properties and overall physical properties.

Test conditions	Bulk density	Ranking	WA	Ranking	Overall properties	Ranking
Wastewater spraying	0.95	3	1.07	1	2.82	3
Carbonation temperature	0.64	4	0.97	3	2.90	4
Carbonation duration	1.18	2	1.03	2	3.04	2
Drying process	1.23	1	0.93	4	3.24	1

(a)



(b)

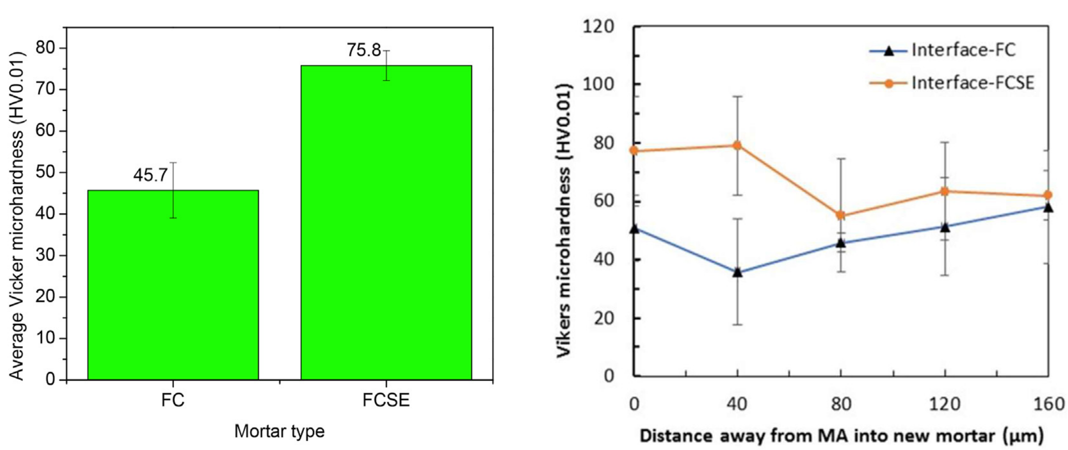
Fig. 4. Durability properties of different types of concrete (a) capillary water absorption (b) bulk electrical conductivity. NAC = concrete prepared with natural coarse aggregates; RAC = uncarbonated RCAs; RAC-FC = concrete prepared with conventional carbonated RCAs; RAC-PC = concrete prepared with pressurized carbonated RCAs; RAC-FCSE = concrete prepared with optimized wastewater sprayed and carbonated RCAs.

4.2. Sorptivity and bulk electrical conductivity of RAC

Sorptivity is the ability of concrete to absorb and transmit water and other liquids by capillary suction and provides an engineering measure of microstructure, total porosity and pore diameter which affect the durability properties of concrete (Bao et al., 2020a; Alexandridou et al., 2018; Bravo et al., 2015; Uzoegbo et al., 2016). Sorptivity is a function of the permeability of the pore system of concrete and is a useful durability indicator (Shaikh et al., 2018). On the other hand, bulk electrical conductivity (BEC) provides rapid indication of resistance of concrete to chloride ion penetration and indirectly measures the interconnected porosity of concrete (Habibi et al., 2021; Kurda et al., 2019). Increasing RCA beyond 30% content in concrete tend to accelerate steel corrosion due to the porous nature of RCA (Arredondo-Rea et al., 2019). This situation makes measuring bulk electrical conductivity very important especially where 100% RCA applications is used. Therefore, to avert the accelerated corrosion problem, strengthening the RCA prior to usage in concrete is important.

The durability of concrete was investigated using sorptivity and bulk electrical conductivity and the results are displayed in Fig. 4 (a) and (b). Sorptivity was divided into two types namely initial water absorption (IWA) and secondary water absorption (SWA). As shown in Fig. 4 (a), initial water absorption is higher than secondary water absorption because it measures capillary water absorption within the first 6 h of the experiment and the pores are not yet filled up. But as the experiment progresses, the pores get filled up and the capillary water absorption reduced drastically. Compared to uncarbonated RAC, concrete prepared with optimized carbonation (RAC-FCSE) exhibited 20.5% reduction and 25.3% reduction in initial and secondary water absorption respectively. In addition, concrete prepared with optimized carbonated RCAs and pressurized carbonated RCAs (RAC-PC) recorded similar initial and secondary water absorption. These results demonstrated that the optimized conventional carbonation was as effective as the pressurized carbonation to enhance the properties of RAC. In addition, similar reduction trend was observed in both the initial and secondary capillary water absorption. Surface pretreatment of RCA increases surface homogeneity, improves ITZ microstructure, and reduces porosity (Al-Bayati et al., 2016). Another study also reported that both capillary water absorption and chloride diffusion were sensitive to the quality of RCA and water-binder ratio and were closely related to the ITZ microstructure (Bao et al., 2020a).

Furthermore, the bulk electrical conductivities of RAC-FC, RAC-PC and RAC-FCSE were similar as shown in Fig. 4 (b), indicating that RAC-



(a) (b)

Fig. 5. Microhardness comparison of (a) conventionally carbonated (FC) and optimized wastewater-sprayed and carbonated RMA (FCSE) (b) interfacial zone of the new mortar surrounding the RMAs produced by conventional carbonation (FC) and optimized wastewater-sprayed and carbonated RMA (FCSE).

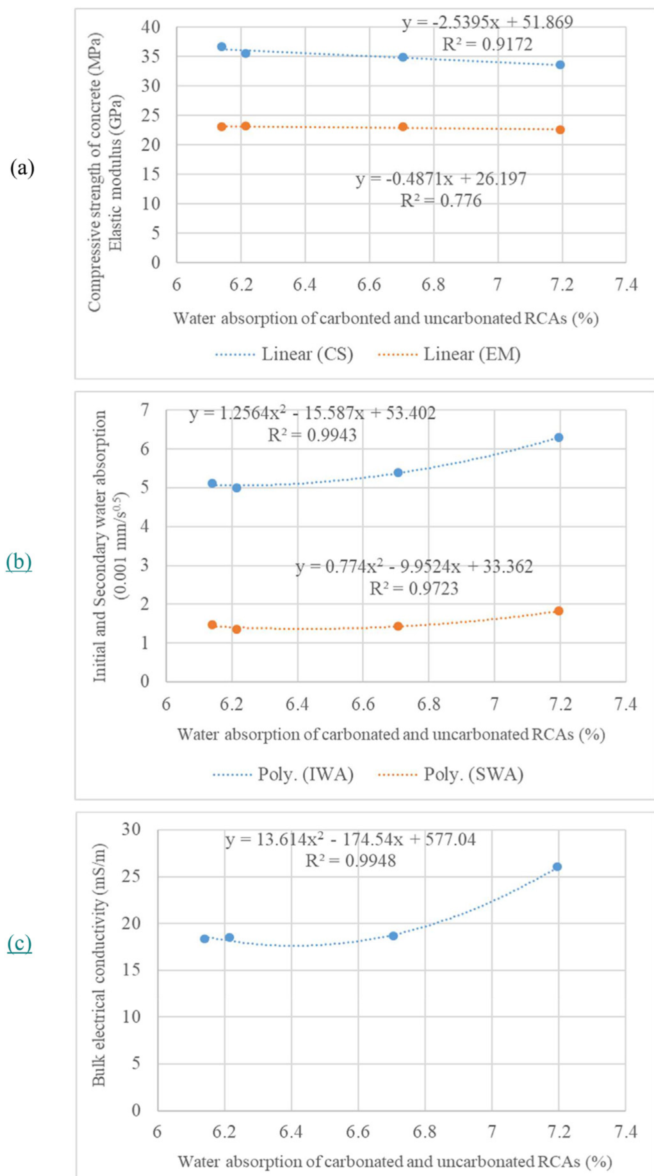


Fig. 6. Relationships between water absorption of carbonated and uncarbonated RCAs and (a) compressive strength and elastic modulus (b) initial and secondary water absorption (c) bulk electrical conductivity of concrete prepared with carbonated and uncarbonated RCAs.

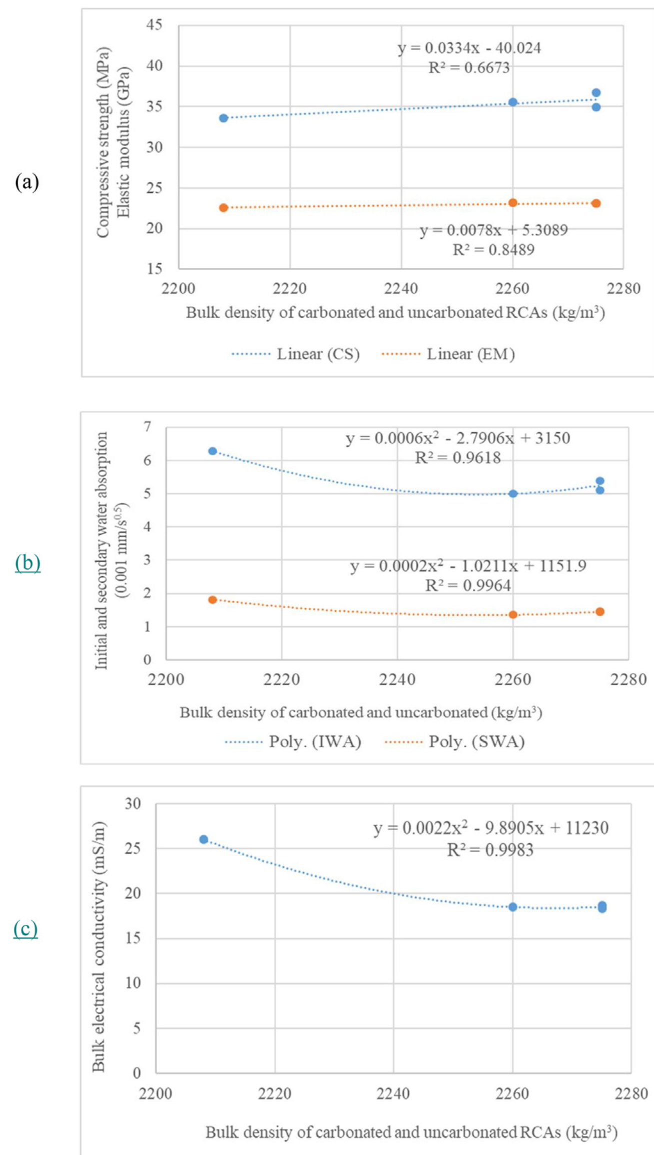


Fig. 7. Relationships between bulk density of carbonated and uncarbonated RCAs and (a) compressive strength and elastic modulus (b) initial and secondary water absorption (c) bulk electrical conductivity of concrete prepared with carbonated and uncarbonated RCAs.

FCSE achieved similar bulk electrical conductivity compared with RAC-PC. Bulk electrical conductivity of concrete tended to increase with increase in both initial and secondary water absorption. Conversely, compressive strength tended to decrease with increase in bulk electrical conductivity. Therefore, bulk electrical conductivity is related to the compressive strength and sorptivity of concrete prepared with natural and carbonated RCAs.

4.3. Microhardness of carbonated RMA and new mortar

Microhardness testing is the hardness of a material surface at the microscopic level. Microhardness of aggregate is important because it gives a measure of the strength of the aggregate and the quality improvement derived from different pretreatment methods. Knowledge of the microhardness gives valuable information on the optimum pretreatment conditions in production of high-quality RCA. Microhardness can also reveal the mechanism governing the durability and mechanical properties of the aggregates. RMAs were utilized because their properties were more consistent compared to RCA (Jayasuriya et al., 2018).

Fig. 5 showed the microhardness of the interfacial area of the specimen prepared with the carbonated RMA and new mortar. As shown in Fig. 5 (a), it was observed that adopting the optimized conventional carbonation (FCSE) can enhance the microhardness of RMA itself. Fig. 5(b) showed the improvement of the interface can be up to 160 μm away from the carbonated RMA into the new mortar. A recent study corroborated the microhardness findings reported in (Fang et al., 2020), where the authors attributed the improved microhardness at the ITZ to both porosity reduction and the growth of C–S–H gel due to calcite grains serving as nucleation sites (Zhan et al., 2020).

4.4. Correlation between properties of carbonated RCAs and the mechanical and durability properties of resulting concrete

As shown in Fig. 6 (a), linear decreasing relationship was observed between water absorption of carbonated and uncarbonated RCAs and

Table 12
Correlation coefficients between RCA properties and resulting concrete.

	Properties of concrete prepared with carbonated and uncarbonated RCAs				
Properties of RCA	Compressive strength	Elastic modulus	Initial water absorption	Secondary water absorption	Bulk electrical conductivity
WA	−0.96	−0.88	0.96	0.85	0.88
BD	0.82	0.92	−0.89	−0.9	−0.97

Table 13
Comparison of pretreatment and carbonation effects on RCA in previous research and current study.

Treatment method	RCA type	Water absorption (%)			Reference
		Untreated	After treatment	Reduction (%)	
Direct carbonation	5–20 mm	6.85	5.65	17.5	Kazmi et al. (2020)
Acid-treated & carbonated	5–20 mm		6.43	6.1	
Lime-treated & carbonated	5–20 mm		5.47	20.1	
Direct carbonation	2.25–6.25 mm	4.93	4.25	13.8	Wang et al. (2020a)
Air-dried, lime-treated and carbonated	2.25–6.25 mm		3.91	20.7	
Environmental-chamber dried & carbonated for 3 days	20–30 mm	4.62	3.14	32.0	Li et al. (2019)
Optimized ww spray & carbonation	5–10 mm	7.73	6.53	15.5	Current study
	10–20 mm	6.66	5.90	11.4	

compressive strength and elastic modulus of concrete prepared with carbonated and uncarbonated RCAs. This implies reduction of water absorption of carbonated RCAs leads to improvement in compressive strength and elastic modulus of resulting concrete. However, much better relationship was observed between water absorption of RCAs and compressive strength than elastic modulus (EM). In contrast, a positive polynomial relationship was observed between water absorption of RCAs and concrete prepared with carbonated and uncarbonated RCAs and both initial and secondary water absorption as shown in Fig. 6 (b). This implies reduction in water absorption of carbonated RCAs results in reduction of both initial and secondary water absorption of resulting concrete as well. However, slightly better relationship was observed between water absorption of RCAs and initial water absorption than secondary water absorption of resulting concrete. Likewise, a positive polynomial relationship was observed between WA of carbonated and uncarbonated RCAs and bulk electrical conductivity of concrete prepared with carbonated and uncarbonated RCAs as shown in Fig. 6 (c). This implies reduction in water absorption of carbonated RCAs results in reduction in bulk electrical conductivity of concrete prepared with carbonated and RCAs. Positive polynomial relationship was also observed between bulk electrical conductivity and water absorption of concrete prepared with carbonated and uncarbonated RCAs by another study (Xuan et al., 2017).

With respect to the bulk density of RCAs, a positive linear relationship was observed with both compressive strength and elastic modulus of concrete prepared with carbonated and uncarbonated RCAs as shown in Fig. 7 (a). However, a better relationship was observed between bulk density of RCAs and elastic modulus than compressive strength. Furthermore, a negative polynomial relationship was observed between bulk density of RCAs and both initial and secondary water absorption of concrete prepared with carbonated and uncarbonated RCAs as depicted in Fig. 7 (b). A slightly better relationship was observed with secondary water absorption compared to initial water absorption. Also, a negative polynomial relation was observed between bulk density of RCAs and concrete prepared with carbonated RCAs as shown in Fig. 7 (c). This implies carbonation of RCAs results in increased bulk density of RCAs and reduction in bulk electrical conductivity of concrete prepared with carbonated RCAs. The result of the regression coefficients was also corroborated by the correlation efficients shown in Table 12. While negative correlation was recorded between water absorption of RCAs and both compressive strength and elastic modulus of concrete, a positive correlation was obtained with initail and secondary water absorption and bulk electrical conductivity of concrete prepared with carbonated RCAs as shown in Table 12. Futhermore, a positive correlation was observed between bulk density of RCAs and both compressive strength and elastic modulus of concrete prepared with carbonated and uncarbonated RCAs while a negative correlation was observed with initial and secondary water absorption and bulk electrical conductivity. These results imply water absorption and bulk density of carbonated and uncarbonated RCAs have opposite effects on mechanical and durability properties of concrete.

Table 13 shows the comparison of water absorption reductions of RCAs from previous studies which utilized different pretreatment methods. It was observed that water absorption reductions from current study is better than acid-treated, carbonated RCAs. However, it seems less effective comparable to air dried, lime-treated carbonation and those carbonated for longer h and treated with highly concentrated lime. Compared to current study, those methods are more energy demanding and utilizes chemical agents which is not required in wastewater spraying. Therefore, optimized wastewater spraying is an environmentally friendly method for enhancing the carbonation of RACs and improving the mechanical and durability properties of concrete prepared with carbonated RACs.

5. Conclusions

Optimization of conventional gas-solid carbonation conditions is important for effective carbonation of RCA. A linear weighted sum method was used to optimize the carbonation conditions of recycled aggregates in this study. The following conclusions can be drawn:

- The adopted linear weighted average sum method was helpful in simultaneously optimizing both the pretreatment and carbonation test conditions involving several input factors and corresponding output responses.
- The optimized pretreatment and gas-solid carbonation conditions: spraying Ca^{2+}-rich wastewater at a level of 60% of the 24-h water absorption of the recycled aggregates, followed by a 12-h air drying and 6-h carbonation at 60 °C.
- Concrete prepared by using the optimized conventional carbonated RCAs exhibited improved mechanical and durability properties similar to concrete prepared with pressurized carbonated RCAs.
- The improvement in the mechanical and durability properties of concrete prepared with carbonated RCAs is attributed to the improvement in the microhardness of both the wastewater-sprayed RCAs and the interfacial zones (ITZ) of the new mortar facilitated by optimized pretreatment and carbonation conditions. The

improvement in the microhardness of the new mortar was observed up to 160 μm in the new mortar.
- Although the mechanical properties in terms of compressive strength and E values did not differ much, higher improvements were noticed in the durability properties compared to mechanical properties. For instance with the use of the optimized carbonation conditions, 20.5% reduction in initial water absorption, 25.3% reduction in secondary water absorption and 28.9% reduction in bulk electrical conductivity were recorded for the concrete prepared with the carbonated RCAs compared to uncarbonated RCAs.

Novelty of this study

This study demonstrated the use of a linear weighted sum method to simultaneously optimize the pretreatment and carbonation test conditions. The optimization of the test conditions produced simplified, and eco-friendly pretreatments and drastically reduced the carbonation duration of RCA from 72 h in previous study (Zhan et al., 2020) to 18 h in this study. In addition, the optimum experimental conditions were validated by casting new concrete using the carbonated RCAs. This study also demonstrated that wastewater derived from ready-mix concrete batching plants can be utilized for spraying the RCAs to improve the carbonation efficiency. Also, the mechanical and durability properties of concrete prepared with RCAs produced from the optimized conventional carbonation and pressurized carbonation were similar. Therefore, optimized conventional carbonation can be utilized as an alternative to pressurized carbonation.

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CRediT author contribution statement

Adebayo Sojobi: Investigation, Validation, Data curation, Formal Analysis, Writing - original draft. Dongxing Xuan: Conceptualization, Methodology, Supervision, Writing - review & editing. Long Li: Resources, Validation. Songhui Liu: Resources, Methodology. Chi Sun Poon: Project administration, Resources, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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