

AQUEOUS CO₂ SEQUESTRATION FOR LOW-CARBON READY-MIX CONCRETE

Yujie, Jamie Chong^{*,1}, Guan Feng Chua¹, Mingshan Zhao¹, Colin Yip², Saradhi Babu Daneti³, Fei Jin⁴

*jamie.chong@ singaporetech.edu.sg

¹Singapore Institute of Technology, 10 Dover Drive, Singapore 138683

²Woh Hup (Private) Limited, 217 Upper Bukit Timah Road, Singapore 588185

³Alliance Concrete Singapore Pte. Ltd., 72 Sungei Kadut St 1, Singapore 729372

⁴Cardiff University, Queen's Buildings, 5 The Parade, Newport Road, Cardiff, CF24 3AA

Abstract

The cement industry accounts for 8% of global energy- and process-related greenhouse gas emissions. To achieve global net-zero emission targets by 2050, the need for commercially ready low-carbon construction materials is becoming increasingly urgent. The fixation of captured carbon dioxide in concrete through CO₂ sequestration is a crucial area of study to reduce concrete embodied carbon. This paper discusses the development of a low-carbon ready-mix concrete (RMC) with aqueous CO₂ sequestration, and the synergy between carbon dioxide and other constituent materials. The effectiveness of this approach was evaluated through mineralogical composition analysis using TGA, and the mechanical and rheological properties of various concrete mixes were studied. Aqueous CO₂ sequestration using carbonated mixing water can stably fix up to 0.84% of CO₂ by weight of cement within the cement matrix as CaCO₃. The poor workability and incompatibility with GGBS that results from this approach were addressed by the inclusion of RCA as an external source of alkalinity and lubrication. This mix of low-carbon RMC has similar strength and rheological properties to conventional RMC and achieved an embodied carbon reduction of approximately 47%.

Keywords

CO₂ sequestration, cement, carbonated water, supplementary cementitious materials, recycled concrete aggregates

1 INTRODUCTION

Since the Paris Climate Agreement came into effect after the 21st Conference of Parties (COP21) in 2015, many green initiatives have been initiated and commercialised internationally in a bid to reduce levels of global greenhouse gas emissions to keep the global average temperature rise below 1.5 °C and prevent catastrophic and potentially irreversible changes to the environment [1]. Concrete is one of the most consumed materials globally. However, its key constituent, ordinary Portland cement (OPC), single-handedly contributes to almost 8% of global carbon emissions [2]. During the production of OPC, emissions are primarily generated by the combustion of fossil fuels to produce the heat required for clinkerisation at 1400 to 1500 °C and the decomposition of carbonates in the form of quarried limestone [3], with an estimated 37% and 63% contribution to the carbon footprint of cement, respectively [2]. With economic development and accelerated urbanisation, the demand for green innovations to reduce the embodied carbon of concrete has increased, giving rise to solutions such as electrification [4], full and partial replacement of OPC with low-carbon materials [5], [6], and carbon dioxide (CO₂) sequestration [7].

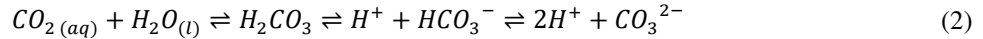
State of the Art

The state of the art in CO₂ sequestration technology concerns the introduction of CO₂ during the mixing and curing stages of concrete production [8], [9]. The injection of CO₂ into fresh concrete allows for stable fixation of CO₂ through the almost instantaneous formation of calcium carbonate in the cement matrix [10], [11], [12], resulting in higher compressive strengths. This reduces the cement demand and subsequently the embodied carbon of an equivalent concrete mix [13], [14], [15]. Unlike concrete carbonation, this process does not impact the alkalinity of hardened concrete, and thus it does not inhibit the passivation of reinforcement steel in concrete [16], [17].

Beyond the direct sequestration of CO₂ into concrete, CO₂ treatment of raw materials for use within concrete has also been studied. These solutions, such as the carbonation of recycled concrete aggregates (RCA) from construction and demolition wastes (CDW) [18], [19] and aqueous CO₂ sequestration, i.e. the carbonation of mixing water [20], [21], [22], [23], aim to maximise the dosage of CO₂ used for sequestration by either improving

the physical and mechanical properties of waste materials [18], [19] or improving efficiency through controlled carbonation conditions [24]. However, there are limited studies available on aqueous CO₂ sequestration due to the poor concrete workability associated with the process [23], [24], [25], [26], and its incompatibility with other constituents [27], [28], [29].

It is well known that the dissolution of gaseous CO₂ into water will reduce its pH value. This is due to the dissociation of the resultant carbonic acid (1), (2), which raises the concentration of hydrogen ions in the solution.



This reduction in pH hinders the activation of pozzolanic materials, such as fly ash and ground granulated blast-furnace slag (GGBS), which are commonly used in low-carbon concrete in high percentages as a replacement for OPC. As CO₂ sequestration is not a standalone solution for the decarbonisation of concrete, its ability to work harmoniously with other low-carbon solutions is essential to its application. Therefore, to raise the pH of carbonated water, neutralisation via sources of hydroxide ions or buffering via sources of bicarbonate and carbonate ions should be implemented [22].

As previously mentioned, the presence of CO₂ in the cement matrix results in the formation of calcium carbonate during the early stages of concrete mixing, hampering the workability of fresh concrete. It is hypothesised that this is a result of the nucleation of nano-calcium carbonate around cement particles, increasing their surface area and inter-particle friction. Therefore, to improve the workability of CO₂ concrete, the introduction of a source of hydroxide ions would enable their reaction with free hydrogen ions in the carbonated water to chemically form water molecules in the cement matrix (3), thus providing extra lubrication between particles while still promoting the formation of calcium carbonate.



In this study, carbonated mixing water is used at a 100% replacement rate in low-carbon ready-mix concrete. The effectiveness of aqueous CO₂ sequestration in a cement-based binder was investigated by thermogravimetric analysis (TGA). In addition, its impact on the mechanical and rheological properties of concrete was investigated.

2 METHODOLOGY

Materials

CEM I Ordinary Portland Cement (OPC) from Engro and Ground Granulated Blast-furnace Slag (GGBS) provided by Alliance Concrete Singapore were used for this study. They comply with standardised specifications from SS EN 19701 and SS EN 15167-1, respectively. Crushed granite with a maximum size of 20 mm and concrete sand provided by Singapore Institute of Technology (SIT) were used as the primary coarse and fine aggregates, respectively. They comply with standardised specifications from SS EN 12620 and the relevant standards listed. A batch of coarse recycled concrete aggregate (RCA), Rc90, with a maximum size of 20 mm obtained from the demolition of the former Port of Singapore Authority (PSA) was used in this study. They comply with standardised specifications from SS EN 12620. Dynamon NRG 1030 from Mapei was used as the superplasticiser.

The carbonated mixing water used for this study was produced by carbonating tap water using an off-the-counter household soda carbonator, SodaStream. The system utilises the pressurised injection of gaseous CO₂ into a proprietary water container. Due to the effect of temperature on the degree of carbonation of water, the temperature of the pre-carbonated water was controlled at room temperature. The effectiveness of carbonation is quantified and regulated using the mass gain of carbonated water: the mass loss of the CO₂ cylinder and the pH of the water before and after carbonation are shown in Fig. 1.

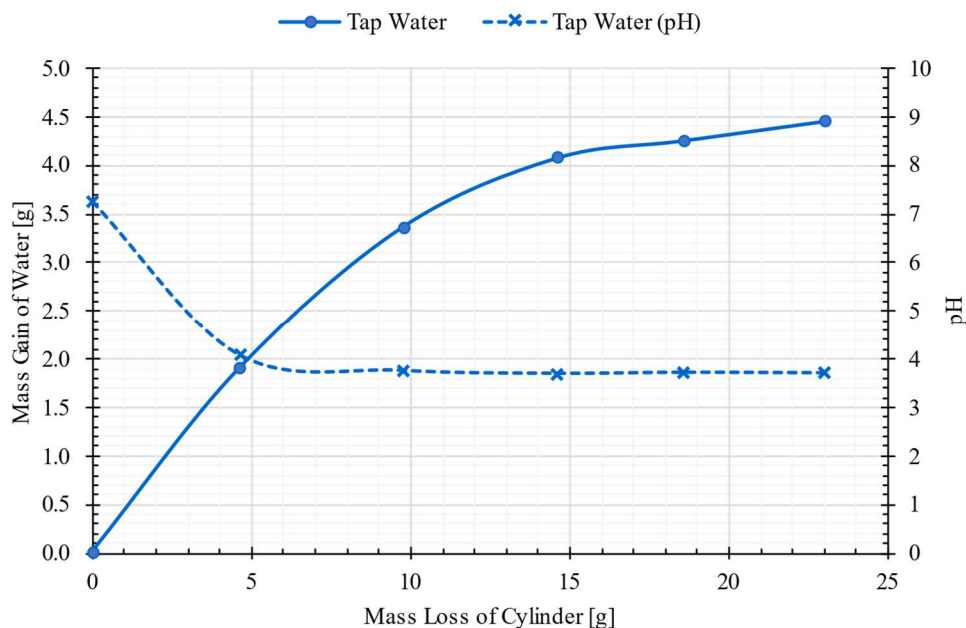


Fig. 1 Correlation of mass gain and pH loss of carbonated water against mass loss of the CO₂ cylinder.

Thermogravimetric Analysis of Cement Paste

Thermogravimetric analysis (TGA) is a widely used method for the quantification of portlandite, Ca(OH)₂, and calcium carbonate, CaCO₃, in cementitious samples. This method involves the measurement of mass changes within a sample as it is heated. In CO₂ sequestration, CO₂ is stably fixed into concrete through the formation in the cement matrix of nano-CaCO₃, which will decompose at temperatures ranging from 530 °C to 760 °C into solid calcium oxide, CaO, and gaseous CO₂. Therefore, the mass loss of the tested sample within this temperature range will represent the amount of CO₂ successfully sequestered into the sample.

Cement paste samples were produced using OPC and mixing water, with a water-cement ratio of 0.4 to emulate the binder properties of the concrete mix design used in this study. The cement paste samples were prepared in accordance with the procedure from EN 196-1, cast into 50 mm cube moulds, demoulded after at least 16 hours and submerged in tap water to cure until the date of testing. The cement paste samples were crushed with a ZwickRoell Z600E material testing machine, filed, and sieved through an 80 µm sieve and immediately tested in a Netsch F3 Tarsus thermogravimetric analyser.

The temperature of the furnace was programmed to rise at a constant rate of 20 °C per minute to 105 °C, where the temperature was maintained for 10 minutes to facilitate the evaporation of moisture from the sample and prevent further hydration. After this, the temperature was raised to 1000 °C at the same rate. The tests were performed under an airflow of compressed air of 40 mL per minute.

Rheological and Mechanical Properties of Concrete

Tab. 1 Concrete Mix Design.

Materials	Quantity kg/m ³
OPC CEM I	395
Mixing Water	169
Fine Aggregate	784
Coarse Aggregate	990
Superplasticiser	3.95

The base concrete mix design for this study is shown in Tab. 1. The water-cement ratio of the concrete mix was slightly reduced from 0.43 to 0.41 for all mixes where GGBS was used as a partial replacement for OPC. Based

on the water absorption on the aggregates used, an adjusted amount of additional water was included; this part of the water was not included in the mix design.

Tab. 2 Experimental Matrix.

Mix	OPC	GGBS	Water	Carbonated Water	Fine Aggregate	Coarse Aggregate	Recycled Concrete Aggregate
Control	100%	-	100%	-	100%	100%	-
CW	100%	-	-	100%	100%	100%	-
GGBS	50%	50%	100%	-	100%	100%	-
CW-GGBS	50%	50%	-	100%	100%	100%	-
RCA	100%	-	100%	-	100%	90%	10%
CW-RCA	100%	-	-	100%	100%	90%	10%
CW-GGBS-RCA	50%	50%	-	100%	100%	90%	10%

To investigate the rheological and mechanical properties of concrete with carbonated mixing water and its compatibility with other low-carbon constituents, an experimental matrix isolating and combining different combinations of materials was adopted, as shown in Tab. 2. The concrete was prepared according to the standard procedure specified in SS EN 206, cast into 100 mm cube moulds, demoulded after at least 16 hours and submerged in tap water to cure until the date of testing.

Immediately after mixing, the workability of fresh concrete was evaluated via the slump test in accordance with the equipment and procedure specified in EN 12350-2. On the 3rd, 7th and 28th day of curing, the concrete cubes were removed for compressive strength testing in accordance with a procedure specified in EN 12390-3 using a SANS YAW 4306 servo hydraulic 3000 kN compression machine.

Environmental Impact of Concrete

To study the environmental impact of concrete, its embodied carbon was assessed. The embodied carbon of individual raw materials was obtained from the Inventory of Carbon and Energy (ICE) database and presented in Tab. 3.

Tab. 3 Embodied Carbon of Raw Materials.

Materials	Embodied Carbon kg CO ₂ /kg
OPC CEM I	0.912
GGBS	0.042
Fine Aggregate	0.0044
Coarse Aggregate	0.0044
RCA	0.0061

The embodied carbon of concrete is calculated using (4).

$$EC_{concrete} = \sum (EC_{material} \times MD_{material}) - (\%CO_2 \times MD_{OPC}) \quad (4)$$

where $EC_{concrete}$ is the embodied carbon of concrete in kg CO₂ / m³ concrete, $EC_{material}$ is the embodied carbon of raw materials in kg CO₂ / kg material, $MD_{material}$ is the mix design quantity of raw materials in kg / m³ concrete and $\%CO_2$ is the percentage of sequestered CO₂ per unit weight of OPC.

3 RESULTS

Thermogravimetric Analysis of Cement Paste

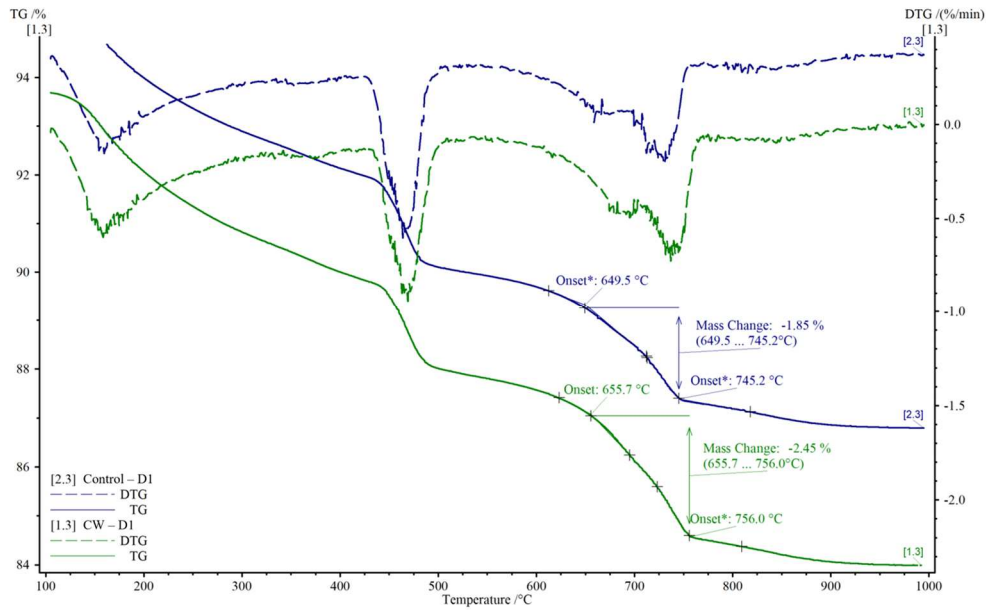


Fig. 2 Derivative thermal gravimetric (DTG) and thermal gravimetric (TG) curves for 1-Day Cement Pastes.

Thermogravimetric analysis (TGA) was used to determine the mass of CO₂ sequestered into the cement matrix. Derivative thermal gravimetric (DTG) and thermal gravimetric (TG) curves of the cement paste samples are shown in Fig. 2. In the case of the DTG curve, the peak at ~650 °C to ~750 °C represents the temperature of decomposition of calcite (CaCO₃). Referring to the sample's corresponding TG curve, the mass loss as CO₂ from CaCO₃ decomposition can be obtained.

The mass losses due to CaCO₃ decomposition in the two types of 1-day cement paste samples are shown in Fig. 2. The CaCO₃ decomposition mass loss of tap water samples was 1.85%, which is attributed to the filler present in the OPC and to natural carbonation occurring within the sample, and is taken to be the baseline. The mass loss of carbonated water samples shows a 32.4% increase to 2.45%. Therefore, the net mass of CO₂ sequestered aqueously is 0.60% of the weight of the cement binder or 0.84% of the weight of the cement.

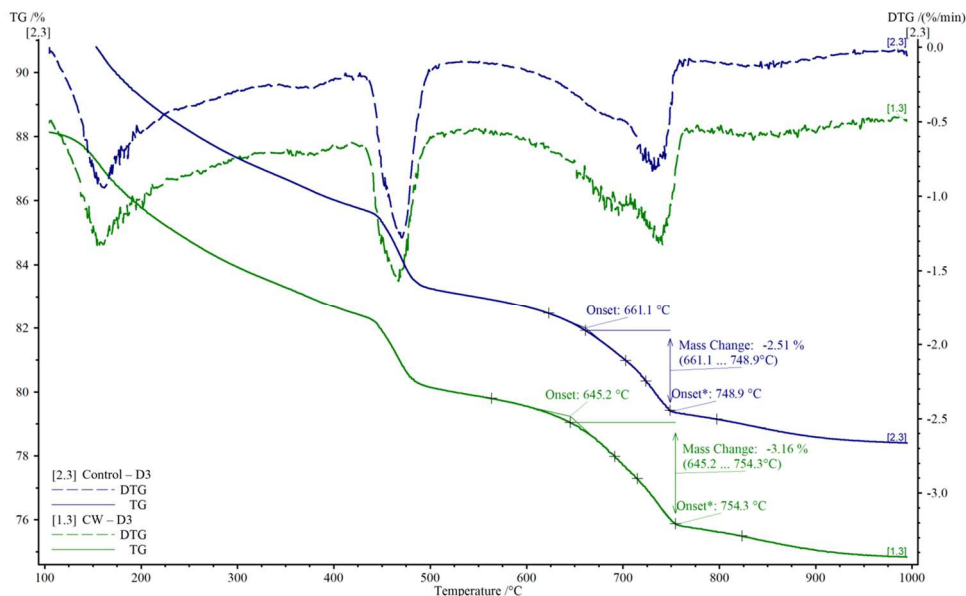


Fig. 3 Derivative thermal gravimetric (DTG) and thermal gravimetric (TG) curves for 3-Day Cement Pastes.

In addition, 3-day cement paste samples were analysed, and the results are shown in Fig. 3. The mass loss of tap water samples was 2.51%, while the mass loss of carbonated water samples was 3.16%. A similar net mass of 0.65% was observed, verifying that the aqueous sequestration of CO₂ occurred within less than 1 day after mixing, and that the CO₂ remains stably fixed in the cement matrix.

Rheological and Mechanical Properties of Concrete

Tab. 4 Workability and compressive strength test results.

Mix	Slump mm	Compressive Strength MPa		
		3-Day	7-Day	28-Day
Control	170	29.23	39.85	51.51
CW	25	32.78	41.04	53.62
GGBS	50	25.98	40.29	55.69
CW-GGBS	50	23.04	33.52	42.64
RCA	110	23.46	30.52	40.04
CW-RCA	130	35.19	39.52	50.24
CW-GGBS-RCA	170	30.11	38.74	49.33

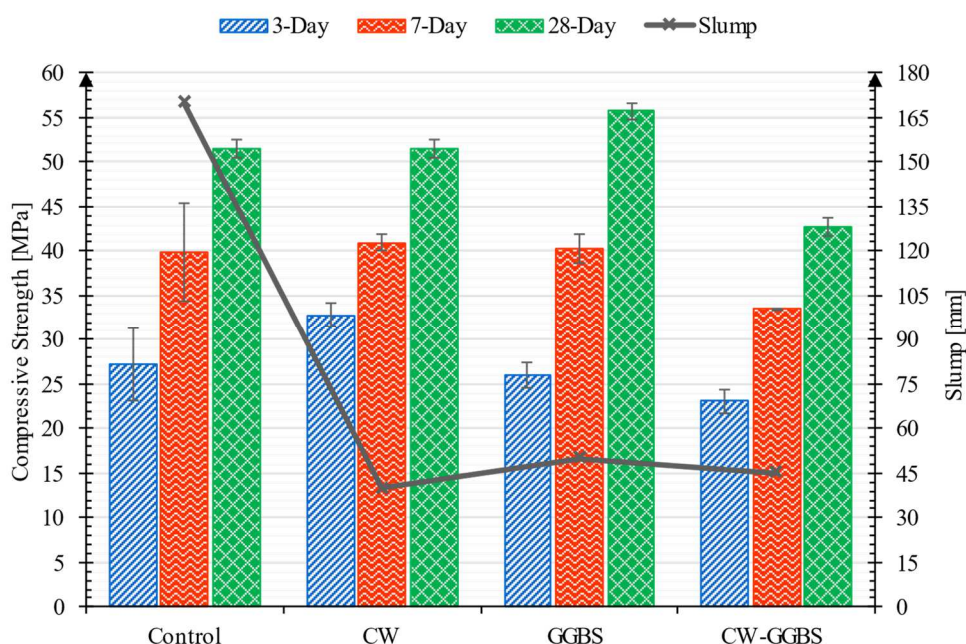


Fig. 4 Workability and compressive strength comparison between the Control, CW, GGBS and CW-GGBS mixes.

Tab. 4 presents the compressive strength and workability of the concrete mixes. Fig. 4 depicts a comparison of the compressive strengths of the tap water and carbonated water concrete mixes. It is observed that the 3-day compressive strength of the CW mix was 32.78 MPa, which is 12.1% higher than the Control mix, which has a compressive strength of 29.39 MPa. This accelerated early development of compressive strength in the CW mix appears to plateau at a similar magnitude to the Control mix, as shown through a similar 28-day compressive strength of 51.51 MPa and 53.62 MPa, respectively. It is also observed that the workability of the CW mix was significantly stunted, with a slump of 40 mm, which is 76.5% lower than the Control mix slump of 170 mm.

The compressive strengths of concrete with a 50% replacement of OPC with GGBS with and without carbonated mixing water, the GGBS mix and the CW-GGBS mix, are shown in Fig. 4. It is observed that both the compressive strength and the strength development of the CW-GGBS mix are lower than those of the GGBS mix, with a 3-day compressive strength of 23.04 MPa and 25.98 MPa, respectively and a 28-day compressive strength of 33.52 MPa and 40.29 MPa, respectively. This reduction in strength by 11.3% and 23.4% shows the incompatibility between carbonated mixing water and GGBS.

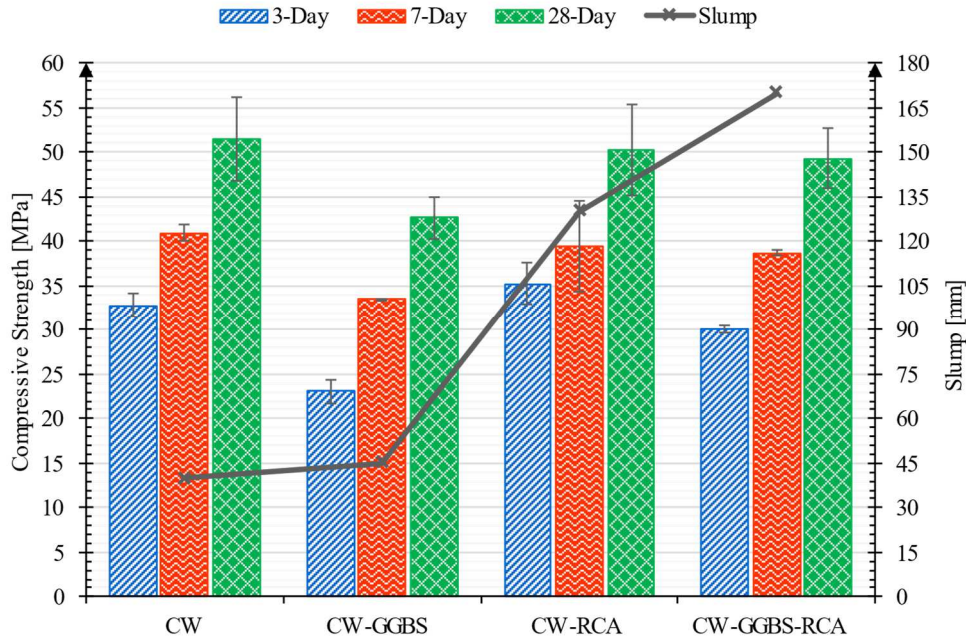


Fig. 5 Workability and compressive strength comparison between the CW, CW-GGBS, CW-RCA and CW-GGBS-RCA mixes.

The compressive strengths of concrete with recycled concrete aggregates (RCA), both with and without carbonated mixing water, the CW-RCA mix and CW-GGBS-RCA mix, are shown in Fig. 5. It is observed that the addition of RCA resulted in a significant increase in workability for both the CW-RCA mix and the CW-GGBS-RCA mix, achieving slumps of 130 mm and 170 mm, which is comparable to the Control mix. It is also observed that the compressive strength of both the CW-RCA mix and the CW-GGBS-RCA mix followed a similar strength development trend to the CW, with 3-day strengths of 35.19 MPa and 30.11 MPa, respectively and 28-day strengths of 50.24 MPa and 49.33 MPa, respectively, with the CW-GGBS-RCA mix achieving a mildly lower overall strength of 1.8%, which is likely to be a result of the slow strength development properties of GGBS.

Environmental Impact of Concrete

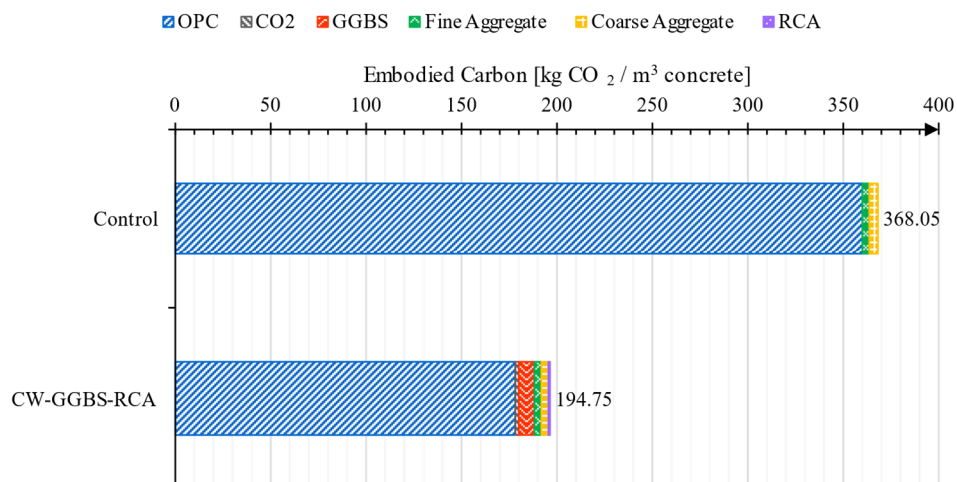


Fig. 6 Embodied carbon comparison for different concrete mix.

The comparison between the embodied carbon of the conventional concrete mix (Control) and an equivalent-strength low-carbon concrete mix with aqueous CO₂ sequestration (CW-GGBS-RCA) is presented in Fig. 6. It was

determined that the embodied carbon CW-GGBS-RCA mix was 194.75 kg CO₂ / m³ concrete, which is 47.09% lower than the Control mix, which was 368.05 kg CO₂ / m³ concrete.

4 DISCUSSION

In this study, carbonated mixing water was used to study the effects of aqueous CO₂ sequestration on the mechanical and rheological properties of low-carbon ready-mix concrete, as well as the method's effectiveness at sequestering CO₂ into an OPC-based binder. A net CO₂ dosage of 0.60% of the weight of the binder was successfully sequestered in cement paste with a water-cement ratio of 0.4: this is equivalent to 0.84% of the weight of the cement. When compared to the state of the art for direct CO₂ sequestration in concrete, the dosage of CO₂ was higher than is typically acceptable, resulting in a concrete mix that is prone to poor workability [25], [30].

It was found that concrete produced with carbonated mixing water exhibited an increase in early strength of 12.1% as compared to the Control mix, with both ultimately achieving similar strengths at 28 days. However, a significant reduction in workability by 76.5% was also observed in the carbonated mixing water concrete mix. It is hypothesised that CO₂ sequestration leads to the formation of nano-CaCO₃ in the cement matrix almost instantaneously upon the introduction of CO₂ [10], [11]. This is often characterised by an increase in concrete strength and a decrease in concrete workability [25], which is in agreement with the results obtained.

When utilised in a concrete mix where 50% of OPC was replaced with GGBS, carbonated mixing water showed a further reduction in the 28-day compressive strength of concrete by 17.2% with no improvements in workability when compared to the Control mix. It is widely studied that the use of GGBS in concrete relies heavily upon the presence of high alkalinity of above pH 9.5 (in the form of Ca(OH)₂ from the reaction between OPC and mixing water) for the activation of a pozzolanic reaction [27], [28], [29]. Therefore, it is hypothesised that the low pH carbonated mixing water hinders this pozzolanic reaction, resulting in the poor compatibility of aqueous CO₂ sequestration and pozzolanic SCMs at high replacement rates, as reflected in the results obtained.

To overcome these challenges faced by this method of aqueous CO₂ sequestration, the use of RCA in the concrete mix at 10% replacement of coarse aggregates was studied. It is hypothesised that the attached old mortar present on the RCA would act as a source of alkalinity for the activation of GGBS and provide chemically formed water molecules which improve workability through lubrication between particles without negatively affecting the formation of nano-CaCO₃ in the cement matrix. It was found that the addition of RCA into low-carbon concrete produced with carbonated mixing water showed promising results, achieving an equivalent slump of 170 mm as compared to the Control mix while retaining the benefits of high early strength from a carbonated mixing water concrete mix of 30.11 MPa and an acceptable 28-day compressive strength of 49.33 MPa, despite the replacement of a high percentage of OPC with GGBS.

From embodied carbon calculations, it was determined that the primary contributor to the CO₂ emissions of concrete is OPC. Therefore, the replacement of a high percentage of OPC with SCMs results in significant reductions to the overall embodied carbon of 47.09%. Meanwhile, aqueous CO₂ sequestration and RCA are primarily used to improve the early strength and workability of low-carbon concrete without incurring significant additional carbon emissions [20], [22].

5 CONCLUSION

In this study, aqueous CO₂ sequestration was applied for low-carbon ready-mix concrete. The effectiveness of this approach was evaluated through mineralogical composition analysis using TGA. Various concrete mixes were produced to isolate and study the cross-compatibility of aqueous CO₂ sequestration with other low-carbon constituents.

- Aqueous CO₂ sequestration using carbonated mixing water can stably fix up to 0.84% CO₂ by weight of cement within the cement matrix in the form of CaCO₃. As a result, the early compressive strength of concrete is increased by 12.1%, while the 28-day compressive strength is comparable to conventional OPC-based concrete.
- When used in conjunction with GGBS, aqueous CO₂ sequestration reduces the compressive strength of concrete by 23.4% and reduces workability by 76.5%. This is in agreement with the hypothesised incompatibility between GGBS activation and the acidic nature of carbonated water.
- The poor workability and compatibility issues with GGBS and CO₂ are addressed with the use of 10% RCA, which is introduced as an external source of alkalinity and lubrication. The CW-GGBS-RCA concrete mix successfully achieves similar workability and compressive strength as compared to an

equivalent conventional concrete. Through a combination of multiple decarbonisation solutions, a 47.09% reduction in embodied carbon for low-carbon ready-mix concrete (RMC) was achieved.

Further microstructure and mineralogical composition studies regarding the specific mechanism of aqueous CO₂ sequestration and its reaction with RCA can be performed to better understand the reactions that resulted in the behaviour observed. A better understanding of the mechanism would allow for additional optimisations to the concrete mix to maximise the CO₂ reduction potentially through the replacement of raw materials with waste by-products and increased CO₂ dosage for CO₂ sequestration. The use of low pH mixing water seems nonideal for structural-use reinforced concrete. Therefore, further studies should be carried out to investigate the use of alternative high pH carbonated solutions as mixing water for concrete.

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