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Durability performance of self-compacting concrete produced using CO₂ as an admixture

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ABSTRACT

This research investigates the fresh characteristics and durability of self-compacting concrete (SCC) made using CO₂ as an admixture. In the existing practice, CO₂ is added to concrete mainly through accelerated carbonation curing. That has many limitations, such as low diffusion rate, requirements of the large airtight chamber, and use in precast concrete only. To overcome these limitations, the current study employed a CO₂ mineralization approach. As cementitious materials hydrate during mixing, CO₂ reacts with it and produces more hydration products. In addition, calcium carbonate particles are formed in situ, filling the minute pores and densifying the SCC matrix. The present research results reveal that a small quantity of CO₂ mineralization improves the SCC compressive strength and durability; the best durability is achieved at 0.3% mineralization of CO₂ by the weight of the cement used. Compare to normal SCC, a 4.3% higher compressive strength of CO₂ mineralized SCC was noticed at 28 days of testing. 0.05 % lowers weight gain and 1.88 % higher compressive strength was noticed in CO2 mineralized SCC at 180 days of 5% sodium sulfate exposure condition. Similarly, the rapid chloride penetration test result shows that 11.35% lesser chloride ions pass from CO_2 mineralized SCC compared to normal SCC. The fresh characteristics of the SCC as a result of CO₂ mineralization behave similarly to reference SCC with small degradation that is considered acceptable. Hence, the present study demonstrates that CO₂ can be used beneficially in concrete to enhance its properties and lower CO₂ emissions into the atmosphere. Copyright © 2024 Elsevier Ltd. All rights reserved.

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1. Introduction

Self-compacting concrete (SCC) was developed in Japan in 1988 by a team of researchers as a practical method to produce durable concrete with high performance [1]. SCC can also help in reducing noise pollution on the job site caused by concrete vibration [2]. SCC can readily fill formwork without compaction, and vibrating and at the same time it is having flowability through the reinforcing [3]. These Properties of the SCC speed up the building construction process and enhance the working environment. However, because of the high energy and resource requirements for its production, the higher usage of Portland cement in these specialized concretes has a substantial adverse environmental impact, resulting in higher CO₂ emissions [4].

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About 0.8 tons of CO₂ are emitted during the production of each ton of Portland cement. [5]. One of the best solutions to the issue of rising emissions of CO₂ is the storage of carbon in cementitious materials [6]. Additionally, the use of industrial waste makes the construction industry sustainable and greener [7]. Concrete carbonation is a durability issue because it reduces concrete passivity, resulting in reinforcement corrosion. However, early-age carbonation does not have the same impact; instead, it reacts with the hydrating phase, producing additional hydration products and micro-level calcium carbonate (CaCO₃), filling the minute pores of the concrete [8]. Hence, densifying the concrete matrix, and improving its durability and strength [9].

The first study on the carbonation of freshly hydrated cement was done in the 1970s by the University of Illinois [6]. The major calcium silicate phases of cement were found through experimentation to react with CO_2 to create $CaCO_3$ and C-S-H gel. as shown in Equations (1) and (2) [6]:

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$$(3-x)CO_2 + 3CaO \cdot SiO_2 + yH_2O$$

$$\rightarrow (3-x)CaCO_3 + xCaO \cdot SiO_3 \cdot yH_2O$$
(1)

$$(2-x)CO_2 + 2CaO \cdot SiO_2 + yH_2O$$

$$\rightarrow (2-x)CaCO_3 + xCaO \cdot SiO_3 \cdot yH_2O$$
 (2)

In addition, it was found through experimentation that the calcium hydroxide $\{Ca(OH)_2\}$ found in cement paste also reacts with CO_2 , as presented in Equation (3), [6]:

$$CO_2 + Ca(OH)_2 \rightarrow CaCO_3 + H_2O \tag{3}$$

The current research aims to identify the optimal CO_2 dose by the weight of cement used to be mineralized in SCC to obtain the best outcomes. A varying quantity of CO_2 by weight of cement used was mineralized, and the best results of resistance to sulfate attack, compressive strength, and chloride ion penetration were tested, and the change in test outcome was compared with control SCC (Mix-0) with zero % CO_2 mineralized.

2. Experimental program

2.1. Materials and mix proportions

Liquefied CO₂ obtained from industrial emission, with a specific gravity of 1.1, was used. 53 grade OPC, confirming to IS 269: 2015 [10], was used. The fineness and the specific gravity of the OPC were 305 m²/kg and 3.13, respectively. Class F fly ash, confirming to IS 3812 (Part-2): 2013 [11] with a specific gravity of 2.2, was utilized. Locally sourced standard river sand conforming IS-2386 (Part-3): 1963 belonging to zone-II [12], was utilized as fine aggregate. The coarse aggregate has a fineness modulus of 7.086 and a specific gravity of 2.85. Water that satisfied IS 456: 2000 [13] standards was used. A commercially available superplasticizer with a specific gravity of 1.1 was used.

Following the recommendations of the EFNARC [14] specification, an SCC mix considering a target compressive strength of 40 MPa was designed. The mix used passed the basic SCC test and the strength criterion. Three mixes were applied with variable amounts of CO₂. 0.15% CO₂ mineralized in SCC (Mix-I), 0.3% CO₂ mineralized in SCC (Mix-II), and 0.45% CO₂ mineralized in SCC (Mix-III). The weight of ingredients per cubic meter of concrete for each mix ID is presented in Table 1. To create a homogeneous mixture with good consistency, the high-range water-reducing admixture namely, CHRYSO Fluid Optima K- 15, was used in the present experimental work. Fresh property tests were conducted after it had been properly mixed to assess the concrete capacity for flowing, filling, and passing.

2.2. CO₂ mineralization into SCC

The mineralization of CO_2 into the SCC was done in two steps; in the first stage, CO_2 was sequestrated in a cement slurry and then it was combined with the other ingredients to produce SCC. Fig. 1 depicts the CO_2 mineralization equipment that was employed in this study. Fig. 2 shows a schematic diagram of the same. The following is the step-by-step procedure to sequestrate CO_2 into the



Fig. 1. CO₂ mineralization setup.

cement slurry: cement slurry is created in the vessel with a water-cement ratio of 0.5. The slurry vessel is air-tightly packed using the bolts after the slurry has been prepared. The slurry cylinder is then set on the weighing balance once the flexible pipe connecting the $\rm CO_2$ cylinder and the slurry vessel has been connected. In the second stage, the $\rm CO_2$ -sequestrated cement slurry is combined with the other ingredients to produce $\rm CO_2$ - mineralized SCC. The required amount of $\rm CO_2$ is injected into the slurry cylinder, followed by the closing of the ball valve and vigorous shaking for a minute.

2.3. Experimental design

In the fresh condition, the following tests were performed: slump flow and flow time of SCC; J-ring test; L-box, V-funnel, and the U-box test in accordance with the EFNARC guidelines

Table 1Mix proportion for control and CO₂ mineralized SCC m³/kg.

Mix ID	Cement (kg)	Fly ash (kg)	Water-cementitious ratio	Water (kg)	Superplasticiser (kg)	Coarse aggregate (kg)	Fine aggregate (kg)	CO ₂ (gram)
Mix-0	430	140	0.35	200	5.7	680	890	_
Mix -I	430	140	0.35	200	5.7	680	890	645
Mix -II	430	140	0.35	200	5.7	680	890	1290
Mix -III	430	140	0.35	200	5.7	680	890	1935

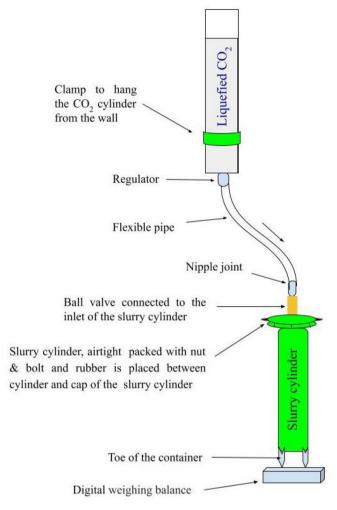


Fig. 2. Diagram of the CO₂ mineralization setup.

[14]. A compression testing equipment with a 3000 KN capacity is used to perform the tests on three 150 mm cube specimens following the IS 516 (Part-1 Sec-1): 2021 [15]; each testing day for each mix and average is taken into consideration. The maximum load placed on the specimen was noted in the machine. Using the formula $f_c = P/A$, the specimen's compressive strength was calculated.

Where fc stands for compressive strength, P is the highest load on the specimen, & A is the specimen's cross-sectional area.

The change in compressive strength and weight change of specimens immersed in 5% Na₂SO₄ solution for 7, 28, 90, and 180 days were used to assess sulfate resistance. 48 specimens were submerged in 5% sodium sulfate (Na₂SO₄) solutions after 28 days of water cure to simulate sulfate exposure conditions. 50 g of Na₂SO₄ and 950 ml of water were combined to produce a litre of 5% Na₂SO₄ solution. On the testing day, the specimens were taken out from the sulfate solution, rinsed 3 times with tap water, and wiped clean. After that, the specimens were allowed to dry for 30 min $(20 \pm 2 \text{ C}, \text{ and RH } 50 \pm 10\%)$ and measured the weight change was with an accuracy of 0. 01% before the compressive strength test, which was carried out as per ASTM C 267: 2001 [16] guidelines. On each testing day, a percentage variation in weight and compressive strength was calculated for 3 samples of each mix with an accuracy of 0.01%, and the result is reported as an average of the tested three specimens. According to ASTM C: 1202 [17], the rapid chloride penetration test (RCPT) was performed on 95 mm dia and 50 mm thickness disc specimens.

3. Results and discussion

3.1. Fresh properties

The necessary level of filling capacity, resistance to segregation and passage ability, must all be present in fresh SCC. All these properties of the SCC were measured according to EFNARC guidelines [14]. Fresh concrete tests were performed within 5 min after the addition of the mixing water. Figs. 3–7 show testing of slump flow, J-ring, U-box, L-box, and V-funnel tests, respectively. The results of fresh properties are given in Table 2. It can be noticed that marginal reductions in all the fresh properties increase as the doses of CO₂ increase; furthermore, in all the mix, ID, the change in all the fresh properties is very small, which is deemed to be acceptable.

3.2. Compressive strength

The compressive strength test is performed for cubes and the obtained results are depicted in Fig. 8. The test was conducted after 7, 28, and 56 days of water curing. The result shows that Mix-II gains maximum compressive strength at 7, 28, and 56 days of testing. It shows that a small optimum dose of CO₂ mineralization, in the range of 0.3 to 0.45% by the weight of cement used, into SCC improved the compressive strength. In the present research, Mix-II gained 7.74%, 4.3%, and 3.9% higher compressive strength than Mix-0 at 7, 28, and 56 days of testing, respectively. Furthermore, the percentage rise in compressive strength of all mixes of CO₂ mineralized SCC at 7 and 28 days are more than that of 56 days Mix-0; this is due to the early carbonation increase in the rate of hydration leading to the higher early strength in CO₂ mineralized SCC than Mix-0 [18]. It is clear from equations (1) and (2) that CO₂ mineralization into concrete produced CaCO₃, which contributes to strength gain. Furthermore, the reactions consumed water from the concrete matrix; this can be the reason behind obtaining the highest benefit of compressive strength at small doses of CO₂ mineralization into SCC. A higher dose than 0.3% of CO₂ by weight of cement consumes more water and can create deficiencies of water in the concrete matrix for the continuous hydration process.



Fig. 3. Slump flow test.



Fig. 4. J-ring test.



Fig. 5. U-box test.

3.3. Variation in compressive strength and weight in sulfate exposure condition

The variation in compressive strength, when the samples were stored in 5% Na₂SO₄ solution is shown in Fig. 9. The compressive



Fig. 6. L-box test.

strength variation was calculated by comparing the strength of 28 days of water curing and sulfate exposure conditions at 7, 28, 90, and 180 days. Mix-II shows the highest gain in compressive strength, 1.82%, 3.58%, 5.74%, and 7.63% higher compressive strength than Mix-II 28 days water cured at 7, 28, 90, and 180 days of testing, respectively. Whereas Mix-0 gained 1.14%, 2.65%, 4.26%, and 5.75% more compressive strength than Mix-0 at 28 days water cured tested at 7, 28, 90, and 180 days of sulfate exposure conditions, respectively. Hence Mix-II gains 0.68%, 0.93%, 1.48%, and 1.88% higher compressive strength than Mix-0 at 7, 28, 90, and 180 days of sulfate exposure condition, respectively. The strength variation in sulfate exposure conditions for all mixes shows similar trends that continuously increase with time. This is due to the hydration of cement during the given exposure period. It can be concluded that a small percentage of CO₂ by the weight of cement used can increase the SCC's capability to resist the loss of compressive strength under the aggressive environment of sodium sulfate.

The result of the change in weight of the sample exposed in the sulfate environment curing is presented in Fig. 10. The weight variation was calculated by comparing the weight of 28 days of water curing and different exposure conditions at 7, 28, 90, and 180 days. The Mix-II change in weight is the least, 0.05%, 0.11%, 0.18%, and 0.26% increase in weight of Mix-II was observed at 7, 28, 90, and 180 days respectively. The Mix-0 gain in weight tested at 7, 28, 90, and 180 days of exposure condition is 0.07%, 00.16%, 0.24%,



Fig. 7. V funnel test.

and 0.31%, respectively. Hence, the variation in the weight of Mix-II is 0.02%, 0.05%, 0.06%, and 0.05% lower than Mix-0 at 7, 28, 90, and 180 days, respectively. In the sulfate exposure condition, the weight of the specimen increased due to salt precipitation on the surface and inside of the specimens [19]. The weight change followed the same trend of change in the compressive strength; a small optimum mineralization of CO_2 , in the range of 0.3% to 0.45% by weight of cement in SCC gives lesser weight change. Less weight change of concrete under an aggressive environment indicates its higher capacity to resist degradation under the given condition.

3.4. The rapid chloride penetration test

The values of RCPT are conducted at 180 days of sulfate exposure condition, and the result is shown in Fig. 11. The maximum number of chloride ions is passing from Mix-0 which is 1365 C, and the least passes from Mix-II, that is 1290 C. 155 Coulombs lesser chloride ions pass from Mix-II than Mix-0, hence 11.35% lower

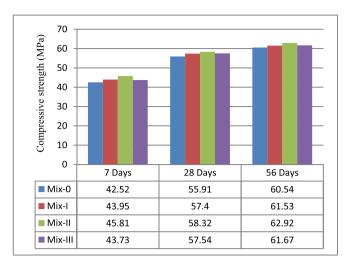


Fig. 8. Compressive strength.

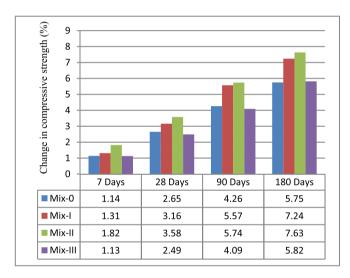


Fig. 9. Variation in compressive strength under a sulfate exposure condition.

chloride ions pass from Mix-II than Mix-0. All of the mixes utilized in this investigation fall into the category of "low" penetration, with chloride penetration ranging from 1000 to 2000 C [17]. A small optimum dose of CO_2 in the range of 0.3% to 0.45% by the weight of cement used in SCC, improves its capacity to resist chloride ions penetrability. As this optimum dose of CO_2 mineralization gives maximum density by filling the minute pores with $CaCO_3$ and producing more hydration products, resulting in lesser chloride ions penetration.

Table 2Fresh properties of control and control and CO₂ mineralized SCC.

Test method	Mix-0	Mix-I	Mix-II	Mix-III 680
Slump flow (mm)	710	695	685	
Slump flow -T ₅₀ cm (sec)	3.62	3.75	3.83	3.91
J-ring- diameter (mm)	695	685	680	670
V-Funnel-T ₀ (sec)	6.56	6.63	6.67	6.72
V-Funnel-T ₅ (sec)	7.88	8.01	8.08	8.18
L-box (h_2/h_1)	0.95	0.93	0.92	0.91
U-box (mm)	13	14	15	15

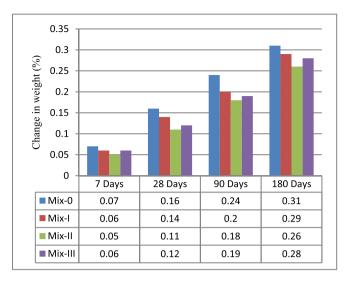


Fig. 10. Variation in weight because of sulfate exposure condition.

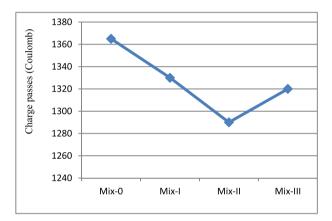


Fig. 11. The rapid chloride penetration at 180 days.

3.5. Cost analysis

The economy index is calculated by dividing the total cost of all raw materials by the 28 days compressive strength. [20]. Table 3 provides a thorough examination of the economy index, together with the price of all the ingredients. 3. Fly ash and CO2 are industrial byproducts and emissions, respectively. Hence, the cost required to transport the fly ash from the industry to the utilization site has been considered. Similarly, the cost to capture and transport the CO2 from emitted source to the utilization site has been considered. The economy index

shows that Mix-II highest value, which indicates that Mix-II is having highest 28 days compressive strength per unit price of SCC. The lowest economy index was found in Mix-0, it shows that the Mix-0 having least 28 days of compressive strength per unit price of SCC.

4. Conclusion

The current study explains the beneficial utilization of CO₂ in concrete production. The present research finding shows that Mix-II gains 4.3% higher compressive strength at 28 days of water curing compared to Mix-0. The results of tests on resistance to sulfate attack and rapid chloride penetration show that a low amount of CO₂ mineralization in the range of 0.3 to 0.45 % by weight of cement can increase the SCC's durability. A 0.3% mineralization of CO₂ by the weight of cement lowers 0.05% weight gain and 1.88 % higher compressive strength was noticed in Mix-II, at 180 days of exposure to 5% Na₂SO₄ condition than in Mix-0. Similarly, the rapid chloride penetration test result shows that 11.35% lesser chloride ions pass in Mix-II than in Mix-O at 180 days of testing. The fresh characteristics of the SCC as a result of CO₂ mineralization behave similarly to Mix-0 with minor degradation that is regarded acceptable. Hence, the suggested approach effectively utilizes CO₂ and significantly lowers CO₂ emissions. The mineralization of CO₂ in concrete resulted in environmentally friendly building materials improving the mechanical and durability performance of concrete.

CRediT authorship contribution statement

Md Athar Kazmi: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, original draft. Lakshmi Vara Prasad Meesaraganda: Methodology, Project administration, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. P. Suresh Chandra Babu: Data curation, Formal analysis, Resources, Software, Validation, Visualization, Writing – original draft, Writing – review & editing.

Data availability

Data will be made available on request.

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.

Table 3 Economy index analysis.

Mix ID	Cost of ingredients in US \$ (kg/m ³)						Total cost/m ³ in US \$	28 Days	Economy index
	Cement	Fly ash	Coarse aggregate	Fine aggregate	Superplasticiser	CO ₂		Compressive strength (MPa)	(strength/cost)
Mix-0	41.46	1.5	8.73	9.38	9.03	0	70.01	55.91	0.8
Mix -I	41.46	1.5	8.73	9.38	9.03	0.24	70.34	57.4	0.82
Mix -II	41.46	1.5	8.73	9.38	9.03	0.48	70.58	58.32	0.83
Mix -III	41.46	1.5	8.73	9.38	9.03	0.72	70.82	57.54	0.81

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