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Mechanical and durability properties of concretes produced by two-step CO_2 mineralization

Lakshmi Vara Prasad Meesaraganda[a, *], *Md Athar* kazmi[a, 1], and *Ashok* Muruganantham[b]

[a]Department of Civil Engineering, National Institute of Technology Silchar, Assam -788 010, India
[b]Corrosion and Materials Protection Division, CSIR – Central Electrochemical Research Institute, Karaikudi, Tamil Nadu - 630 003, India

Abstract. The current ways of using CO_2 in concrete include carbonation conditioning of recycled aggregates and carbonation curing alone. These procedures have several drawbacks, including the need for a large carbonation chamber and the low diffusion rate applied to the outer surfaces only. CO_2 mineralization during concrete mixing is an innovative CO_2 utilization method; in this process, CO_2 reacts with calcium-rich cementitious materials and beneficially impacts cement hydration, producing more hydration products and forming nano-scale calcium carbonate, thus filling the minute pore. Hence, utilizing CO_2, a major greenhouse gas, in manufacturing concrete provides a two-fold benefit. The characteristics of CO_2-mineralizing concrete are investigated in this work; a two-step CO_2 mineralization process in concrete was used. The outcome of the work reveals that incorporating a small proportion of CO_2 into concrete substantially improves its performance. There was a considerable increase in mechanical characteristics at a CO_2 dosage of 0.2% of cement utilized; an 18.7%, 8.3%, and 7.4% increase in compressive, tensile and flexural strength was observed at 28 days of testing. Additionally, CO_2 mineralization significantly improves the long-term durability of concrete. Hence, the present approach improves concrete performance and helps reduce CO_2 emissions.

1 Introduction

Around 0.6 tons of CO_2 are released while producing each ton of ordinary Portland cement (OPC) [1]. CO_2 capture and beneficial use can minimize anthropogenic greenhouse gas emissions. In the present process, CO_2 is captured from the emitting source, compressed and stored in liquefied form, and then transported to where it will be utilized. Carbon storage in cementitious materials is the preeminent resolutions to the problem of rising anthropogenic CO_2 releases [2]. Additionally, using industrial waste as a component makes

* Corresponding author: mlvprasad@civil.nits.ac.in

[1]Present Address: Corrosion and Materials Protection Division, CSIR – Central Electrochemical Research Institute, Karaikudi, Tamil Nadu - 630 003, India

the construction industry more sustainable and environmentally friendly [3]. Concrete carbonation is a durability concern due to the diminished passivity of concrete, causing reinforcement corrosion. Instead of having the same effect, early-age carbonation combines with the hydrating stage to produce micro-level calcium carbonate ($CaCO_3$) and more hydration products that fill the concrete's micro pores [4], strengthening and prolonging the concrete by compacting it [5]. The University of Illinois began investigating the carbonation process of OPC in the 1970s [2]. It was discovered that the calcium phase of the hydrating OPC reacted with CO_2 and water to produce $CaCO_3$ and calcium silicate hydrate (C-S-H).

In the existing practice of CO_2 mineralization into concrete, it is reacted to concrete during mixing in the concrete mixer [6]. The novelty of the present work is that a simplified two-step mineralization process was followed, which further improved efficiency and cost-effectiveness. In the current work, CO_2 was first added into a slurry of OPC; after that, the CO_2-mineralized OPC is mixed with the concrete constituents to form CO_2-sequestrated concrete. Although the mineralization of CO_2 into concrete during mixing enhances the concrete performance, however the reaction of CO_2 with the hydration product of OPC is water harvesting; hence, a slight decrease in the workability is observed by various researchers [6-8]. However, the workability reduction scale is small, which can be regarded tolerable. Furthermore, CO_2 is in a gas phase at atmospheric temperature, and pressure; hence, during mixing with cementitious materials, careful attention must be paid to the airtightness of the mixing drum in order to achieve higher efficiency.

To determine the optimal amount of CO_2 for the maximum strength of concrete, three concrete mixes were treated with varying doses of CO_2, and a control mix that did not include any CO_2 mineralization was examined. The relative strength change was noted compared to the control concrete. Slump value testing was used to evaluate the workability, and compressive, tensile, and flexural strength tests were used to find mechanical properties. To increase the mechanical strength of concrete, the current research established a new technology that uses CO_2 as an additive. This offers a viable path for the valuable application of captured CO_2 in the construction industry, which has two benefits.

2 Materials and methodology

2.1 Materials mix proportioning

Concrete was made using 53 grade OPC that was confirmed to ASTM C150/C150M [9] and IS 269-2015 (Reaffirmed-2020) [10]. Table 1 lists the OPC's physical properties. The OPC's fineness and specific gravity were 3.15 and 304 m2/kg, respectively. Fly ash with a specific gravity of 2.21 that was procured from a local thermal power plant and confirmed to ASTM C618 −12a, [11] and IS 3812- (Part 1)-2013 (Reaffirmed-2017), [12] was used.

Table 1. Physical characteristics of OPC

Properties		Result
Fineness (m^2/kg)		304
specific gravity		3.15
The residue (%) on 75 µm		4.5
Consistency (%)		31
Setting time (min)	Initial	53
	Final	291
Compressive strength (N/mm^2)	7 days	35.3
	28 days	53.6

20 mm of readily available crushed stone aggregate was used as a coarse aggregate. The coarse aggregate had a specific gravity of 2.84 and a fineness modulus of 7.084. Zone II river sand that was easily accessible in the area was used as fine aggregate for this project. The specific gravity and fineness modulus of fine aggregate are 2.63 and 2.51, respectively. Potable water was used to mix and cure concrete specimens. CO_2 was mineralized using 99% purified CO_2 stored from an ethanol distillery and a commercially available polycarboxylate-based superplasticizer with a specific gravity of 1.1 from the Sudanta company.

Table 2. Weight of ingredients of concrete (kg/m^3)

Particular	M-0	M-1	M-2	M-3
OPC	380	380	380	380
Fly ash	76	76	76	76
Water	180	180	180	180
Superplasticizer	3.1	3.2	3.3	3.4
Water/binder	0.39	0.39	0.39	0.39
Fine aggregate	705	705	705	705
Coarse aggregate	1050	1050	1050	1050
CO_2 (gram)	-	380	760	1140

Following the recommendations of the IS 456-2000 (Reaffirmed 2021) [13] and IS 10262-2019 [14] specification, the design of the concrete mix took into account the intended compressive strength of 40 MPa. Four batches of concrete considered in this work are control concrete with no CO_2 (M-0). Three batches of concrete were prepared with different CO_2 dosages of 0.1%, 0.2%, and 0.3% by weight of OPC, referred to as M-1, M-2, and M-3, respectively. The quantities of all ingredients used per cubic meter of concrete for each batch are presented in Table 2.

2.2 CO_2 mineralization into concrete

A two-step process of CO_2 mineralization into concrete was followed, in which the required quantity of CO_2 to be mineralized is first sequestrated into cement slurry separately in a closed cylindrical container. Subsequently, to form CO_2-mineralized concrete, it was combined with the concrete ingredients. A cylindrical container was used to prepare the OPC slurry. Once the slurry is prepared, a slurry cylinder is firmly packed with bolts. The flexible pipe joining the slurry cylinder and CO_2 cylinders is then linked, and it is directly kept onto the weight machine. The specified weight of CO_2 is then added to the slurry cylinder, and the inlet valve is quickly closed.

2.3 Experiment programs

According to IS 1199 (Part-2): 2018 [15], the workability of concrete was evaluated using the slump test, as shown in Figure 1 (a). Compressive strength was measured after 7, 28, and 56 days of water curing, while splitting tensile and flexural strengths were tested after 28 and 56 days as per IS 516 (Part-1 Sec-1): 2021 [16]. All results represent the average of three specimens tested at each age. A 3000 kN compression testing machine applied a uniform load of 14 N/mm²/min on 150 mm cubes for compressive strength. Splitting tensile strength was determined using 300 ×150 mm cylindrical specimens at a loading rate of 0.7 N/mm²/min on a 2000 kN universal testing machine. Flexural strength was evaluated on 100×100×500 mm prisms at the same loading rate. Figures 1 (b) to (d) illustrate these tests.

(a) Slump test (b) Compressive strength test

(c) Splitting tensile test (d) Flexural test

Figure 1. The slump and strength test

Durability performance was examined through bulk resistivity, depth of water penetration, and the rapid chloride penetration tests (RCPT) after 56, 120, and 180 days of curing. RCPT was conducted as per ASTM C1202-2012 [17] using 100 mm diameter and 50 mm thick discs. Bulk resistivity, determined using ASTM C1876-23: 2023 [18], was measured on saturated 100 × 200 mm cylinders using side electrodes and calculated from Equation (1):

$$\rho = \frac{RA}{\ell} \tag{1}$$

where ρ = resistivity (kΩ-cm), R = resistance (kΩ), A = cross-sectional area (cm²), and ℓ = length (cm). The depth of water penetration was tested on 150 mm cubes as per IS 516 (Part-2 Sec-1): 2018 [19].

3 Results and discussion

3.1 Slump value

The mix M-0 has the maximum slump; adding CO_2 caused a slight decline in the slump. The slump value of the regular and CO_2 mineralizing concrete mix is presented in Figure 2. A 114-, 112-, 109-, and 107-mm slump value was found in the mix having zero, 0.1%, 0.2%, and 0.3% of CO_2 respectively. The mix M-2 slump value was 3 mm lower than M-0, with the M-0 slump value being 109 mm and M-2 at 106 mm, with the observed decrease compared to M-0. It indicates that CO_2 mineralized concrete has a satisfactory slump value. The change in slump was minimal, suggesting that CO_2 mineralized concrete behaves similarly to M-0. Consequently, incorporating CO_2 into concrete does not alter its inherent qualities. Because CO_2 mineralization causes the concrete to lose water during the early phases of carbonation, the slump value of CO_2-mineralized concrete decreases [20].

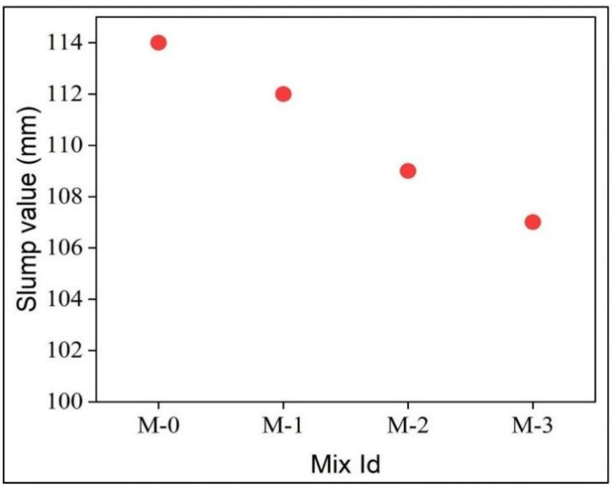

Figure 2. Slump value test result

3.2 Compressive strength

The mix M-2 exhibited the most significant improvement, showing a modest amount of CO_2 mineralisation at a dose of 0.2% by cement weight. Figure 3 presents the compressive strength results for control and CO_2 mineralizing concrete. The compressive strengths of mix M-2 were found as 38.42 MPa, 59.23 MPa, and 62.81 MPa, which are 18.2%, 18.7%, and 19.1% higher than the control mix at 7, 28, and 56 days of testing, respectively. The improvement in strength is due to the formation of $CaCO_3$ microcrystals, which leads to densification of the concrete volume and sealing of micro-pores [21]. Compressive strength has increased due to a higher amount of cement hydration product created by CO_2 mineralization. Calcium carbonate precipitation reinforces the cement paste matrix, improving its cohesion and strength. The nucleation effect plays a crucial role in hydration by significantly increasing compressive strength by filling in the tiny voids left by hydration products [22]. A CO_2 dose larger than 0.2 may cause insufficient water for continuous cement hydration, resulting in incomplete hydration inside the concrete matrix. Carbonation activates silica particles, promoting additional cementitious reactions and strengthening the concrete. Additionally, the interfacial transition zone is strengthened by CO_2 mineralization [23].

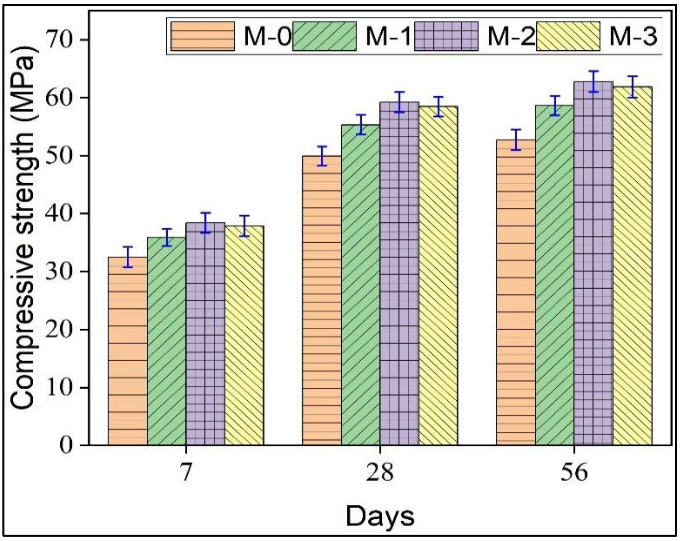

Figure 3. Compressive strength test result

3.3 Cost analysis

The economy index is calculated by taking the ratio of the 28-day compressive strength to the total cost of the raw materials used. [24]. Table 3 presents a detailed analysis of the economic index and ingredient prices. CO_2 and fly ash are emissions and by-products of industry, respectively. Since fly ash is a by-product from industry, which is a waste material, only the cost associated with its transportation is considered. Similarly, the cost of transporting CO_2 from the source of emission to the site of usage has been evaluated. M-2 has the highest economy index rating, indicating the maximum 28-day compressive strength achieved per unit cost of concrete. The minimum economy index, M-0, indicates that the concrete's compressive strength can be maintained for at least 28 days for every unit price.

Table 3. Economy index calculation

Particular	M-0	M-1	M-2	M-3
OPC (US$/kg·m³)	43.5	43.5	43.5	43.5
Fly ash (US$/kg·m³)	1.6	1.6	1.6	1.6
Coarse aggregate (US$/kg·m³)	8.86	8.86	8.86	8.86
Fine aggregate (US$/kg·m³)	9.57	9.57	9.57	9.57
Superplasticizer (US$/kg·m³)	9.2	9.2	9.2	9.2
CO_2 (US$/kg·m³)	0	0.22	0.44	0.66
Total cost (US$/m³)	72.73	72.95	73.39	74.05
28-day compressive strength (MPa)	50.47	56.47	60.64	58.81
Economy index (strength/cost)	0.68	0.77	0.83	0.79

3.4 Splitting tensile strength

Figure 4 presents the splitting tensile strength results; like compressive strength, a 0.2% addition of CO_2 by weight of OPC yields the optimum splitting tensile strength on all days of testing. When tested at 28 and 56 days, the mix M-2 splitting tensile strength is 6.44 and 6.56 MPa, respectively. The splitting tensile strength of mix M-2 was 8.3% and 8.5% higher than that of the control mix. The cement paste becomes more compact and tensile stress-resistant due to the CO_2 mineralization' reduced porosity. By precipitating $CaCO_3$, CO_2 mineralization strengthens the bond between aggregates and cement paste, enhancing the cement paste-aggregate bond. Further, CO_2 mineralization promotes the formation of smaller, more uniform pores, refining the microstructure [8].

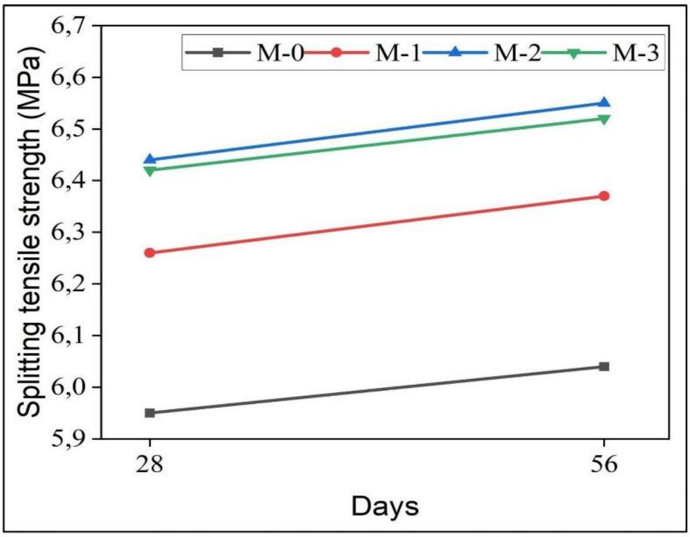

Figure 4. Result for the splitting tensile strength test

3.5 Flexural strength

Figure 5 presents the flexural strength results for both conventional and CO_2-mineralized concretes. It can be seen from the Figure 5 that the concrete flexural strength increased when a small dose of CO_2 mineralized, with mix M-2 showing the maximum improvement; the mix M-2 flexural strength is 4.78 and 5.43 MPa at 28 and 56 days, respectively. At 28 and 56 days, the flexural strength of mix M-2 increases by 7.4% and 7.5%, respectively; furthermore, the addition of $CaCO_3$ to C-S-H may have resulted in a more crucial calcium-silicate-hydrate binding phase [5]. Table 4 lists the findings for the strength test.

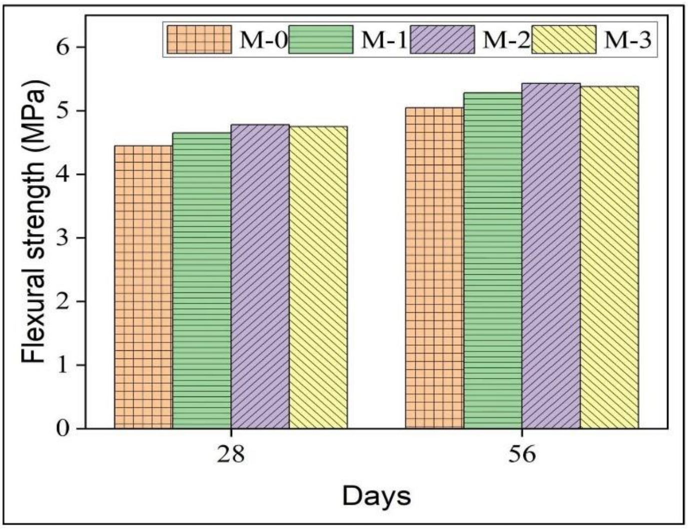

Figure 5. Result for the flexural strength test

Table 4. Result of the mechanical test on control and CO_2 mineralized concrete

Test	Dating day	M-0	M-1	M-2	M-3
Compressive strength (MPa)	7	32.5	35.91	38.42	37.9
	28	49.92	55.34	59.23	58.46
	56	52.74	58.65	62.81	61.86
Splitting tensile strength (MPa)	28	5.95	6.26	6.44	6.42
	56	6.04	6.37	6.55	6.52
Flexural strength (MPa)	28	4.45	4.65	4.78	4.75
	56	5.05	5.28	5.43	5.38

3.6 The durability study

At 56, 120, and 180 days, the M-2 mix recorded charge passes values of 1426, 1276, and 1089 Coulombs, respectively, showing reductions of 18%, 16.52%, and 17% compared with M-0. Figure 6 illustrates the RCPT outcomes, revealing that partial CO_2 mineralization by cement weight effectively lowers chloride ion penetration. This improvement is attributed to increased density and the formation of $CaCO_3$ within micro-pores, resulting from additional hydration products at the optimum CO_2 dosage [22].

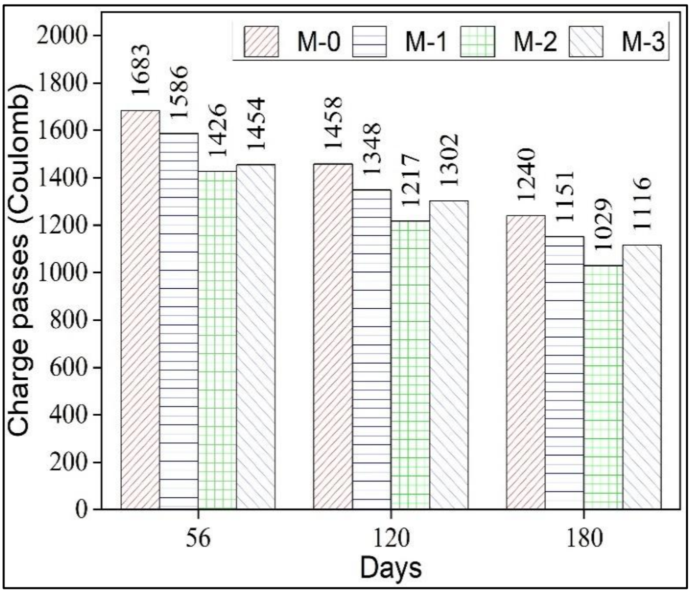

Figure 6. The result of the rapid chloride resistance test

Bulk resistivity and water penetration depth tests, shown in the Figure 7, and Figure 8, further confirmed enhanced durability of CO_2-mineralized concrete. The bulk resistivity of M-2 was 19.48%, 18.56%, and 18.97% higher than that of M-0 at 56, 120, and 180 days, respectively, indicating reduced ionic conductivity. Similarly, water penetration depth in M-2 was 14.69%, 15.42%, and 16.13% lower than in M-0, demonstrating superior impermeability. These findings highlight that fine $CaCO_3$ formation within the pore structure significantly limits water ingress, improving performance in water-exposed structures such as dams, tanks, and basements.

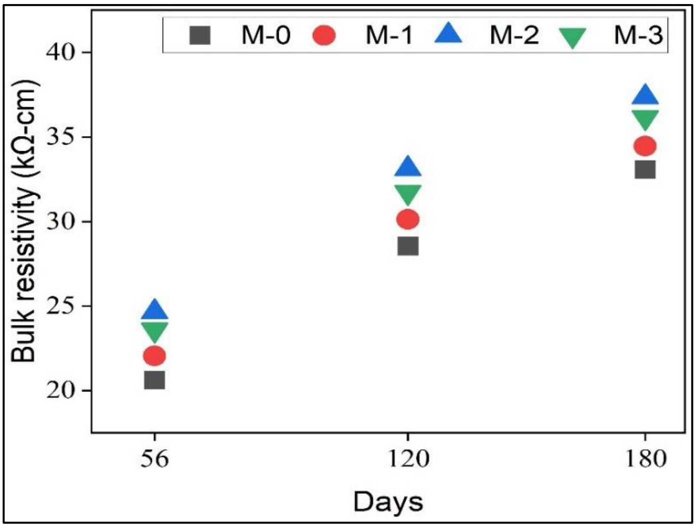

Figure 7. The result of the bulk resistivity test

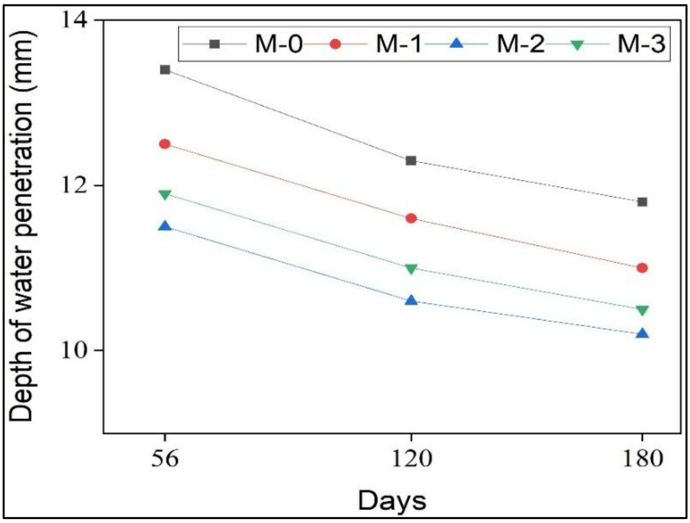

Figure 8. The depth of the water penetration test result

4 Conclusions

The durability and strength of concrete were studied to assess the impact of CO_2-mineralizing on the concrete; the results indicate that 0.2% of CO_2 mineralization, by weight of OPC improves concrete performance. Taking into consideration the current research, the following inferences are drawn:

1. The method of CO_2 mineralization into concrete followed here in this work involves a two-step procedure in which CO_2 is mineralized in cementitious materials initially, then combined with the rest of the concrete ingredients during mixing. The current method overcomes the limitation of utilizing CO_2 for early-age concrete carbonation by carbonation curing only.
2. The proposed method effectively utilize CO_2 while lowering CO_2 emissions. The ability of concrete to mineralize CO_2 has made concrete sustainable building material.

3. The workability of CO_2 mineralizing concrete is similar to that of regular concrete, with nominal acceptable deterioration. The best outcomes for strength were achieved through an optimal dosage of CO_2, ranging from 0.2 to 0.3 by weight of the OPC, as indicated by current research.
4. An 18.2%, 8.3%, and 7.4% higher compressive strength, splitting tensile strengthand flexural strength were found in M-2 than in M-0 at 28 days of water curing.
5. The CO_2-mineralization improve durability compared to conventional concrete, as evidenced by RCPT, bulk resistivity, and water penetration tests. At 180 days, mix M-2 showed 17% lower chloride ion penetration, 18.97% higher bulk resistivity, and 16.13% reduced water penetration depth compared to mix M-0.
6. The strength per unit cost of concrete is significantly reduced due to the mineralization of CO_2 during mixing, giving a double-fold benefit.

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