



Properties and microstructure of concrete and cementitious paste with liquid carbon dioxide additives

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ABSTRACT

This study verified the effect of liquid CO₂ admixture on fresh, hardened, and durability properties of concrete in-situ. It was found that the overall compressive strength of the concrete increased by approximately 5% after 7 days and by about 3% after 28 days. The *t*-test, however, revealed no statistically significant difference between the two groups of samples in terms of 28-day compressive strength for concrete mixes. Therefore, using liquid CO₂ admixture may not help with significant strength improvement and consequently, with reducing cement content within the mix design. In addition, results showed that liquid CO₂ does not significantly affect the fresh, hardened, or durability properties of concrete. In addition, XRD and SEM techniques were performed on paste samples to evaluate the chemical effect of CO₂ admixture on hydrated cement, and hydration products.

1. Introduction

A major component of concrete construction is Ordinary Portland Cement (OPC), and the manufacture of OPC produces significant amounts of greenhouse gases. In the cement industry, there is about 5% of global carbon dioxide emissions attributed to the production of one ton of cement clinker (Xin et al., 2018). The goal of reducing emissions 50% below 2006 levels by 2050 has been recognized as a challenging one to attain (Miller, 2017; Monteiro and Roussanally, 2022). There are already existing improvements in cement replacement and production efficiency, but without new ideas, the future emission improvements will be incremental. Therefore, it makes sense to seek innovative approaches that can be incorporated into a portfolio strategy. An approach to this problem involves binding carbon dioxide as an additive while making concrete products (Gregorya et al., 2021; Monkman et al., 2016; How Green Concrete Can Reduce). Carbonation of freshly hydrating cement occurs when calcium silicate phases react with carbon dioxide in the presence of water and produce calcium carbonate and hydrated calcium silicate gel. However, the reaction of carbon dioxide with a mature concrete micro-structure is conventionally acknowledged to be a durability issue due to reduced pore solution pH, and carbonation-induced corrosion. In contrast, a carbonation reaction in freshly mixed concrete enables CO₂ to react with hydrating cement, rather than the hydration phases present in mature concrete. Because of

adding gaseous CO₂ to freshly mixed concrete, the carbonate reaction products are expected to form in situ, are at the nanoscale, and are homogeneously distributed. (Monkman and Peng; Young et al., 1974).

Qian et al. (Xin et al., 2018) studied the performance of ordinary Portland cement concrete with carbon dioxide as an additive. Fresh concrete was pre-carbonated by incorporating gaseous carbon dioxide directly into the mix. The process creates nano to submicron sized CaCO₃ particles in situ, providing more nucleation sites for OPC hydration. In consequence, the proposed method can significantly improve the compressive strength of cement mortar samples.

In addition, Shao et al. (2015) tested the ability of masonry blocks, paving stones, cement boards, and fiberboards to absorb carbon dioxide during the curing process. With this method, carbon dioxide can be sequestered in concrete products safely and permanently by using either recovered carbon dioxide or flue gas, which would reduce energy costs and offset CO₂ recovery costs.

Monkman et al. (Monkman et al., 2016) investigated adding carbon dioxide to truck-mixed concrete. By reacting CO₂ with cement, calcium carbonate reaction products are formed. In the experiment, optimizing the carbon dioxide dosage decreased the initial setting time by 40%. It also increased the 1-day and 3-day compressive strength by 14% and 10%, respectively. Results of concrete durability tests revealed that carbon dioxide treatment had no adverse impact on durability. Carbon dioxide can enhance concrete's performance. A further study by

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Monkman et al. (2022) looked at the carbonation of general-use Portland cement and calcium tri-silicate powder pastes with optimum water-to-cement ratios of 1.2 and CO₂ contents of 0.3% per weight of cement. Mineralization of CO₂ in situ resulted in more rapid hydration of both systems, with cement being more affected, and TGA analysis confirmed that both systems contained increased carbonate content.

Li and Wu (An overview of utilizing CO₂, 2022) examined the potential benefits of accelerating concrete carbonation. Increasing CO₂ uptake in cement-containing composites did not significantly improve mechanical properties. Early carbonation-accelerated curing affects cement-based composites, and it compromises their strength.

Studies discussed above investigated the effect of CO₂ as an admixture in concrete and cement paste using various experimental methods on laboratory scale. However, there is limited evidence that calcium carbonate (CaCO₃) is generated while hydrated cement is mixing using the liquid CO₂ injection method on large scale field applications, which may complicate quantifying the impact of CO₂ admixture on concrete microstructure and mechanical performance (compressive strength). Therefore, this research aims to: 1) use statistical analysis to verify the improvement of compressive strength using liquid carbon dioxide liquid admixture, and 2) detect calcium carbonate crystalline phases produced from the reaction between CO₂ and cementitious phases, using chemical and microstructural analysis techniques such as X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM). The research also studies the effect of CO₂ admixture on other mechanical and durability properties of concrete samples such as creep, shrinkage, and sulfate attack, as compared to control samples without CO₂ injected.

2. Materials and methods

2.1. Materials

A liquid carbon admixture pure CO₂ gas¹ was used. Concrete and paste samples were made with type I/II cement² and its chemical compositions is shown in Table 1. Fine aggregates were from Hawaii Kaapa quarry and British Columbia, Canada with a bulk specific gravity of 2.65 and 2.75, respectively. The coarse aggregate was crushed basalt from Hawaii Kaapa quarry with a specific gravity of 2.7. Also, the aggregate combined gradation was followed by ASTM C-33 upper and lower limits. The sieve analysis of aggregates is shown in Fig. 1. The concrete mix design is shown in Table 2, and the design strength is 21 MPa (3000 psi) for a w/c ratio of 0.65, and the CO₂ admixture dosage is

Table 1
Cement chemical composition.

Item (%)	Value
Silicon Dioxide (SiO ₂)	20.3
Aluminum Oxide (Al ₂ O ₃)	3.9
Ferric Oxide (Fe ₂ O ₃)	3
Calcium Oxide (CaO)	62.9
Magnesium Oxide (MgO)	4.4
Sulfur Trioxide (SO ₃)	2.5
Sodium Oxide (Na ₂ O)	0.24
Potassium Oxide (K ₂ O)	0.45
<i>Potential Phase Compositions (%)</i>	<i>Value</i>
C ₃ S	63.8
C ₂ S	10.2
C ₃ A	5.4
C ₄ AF	9
Alkali Equivalent (NaEq.)	0.54

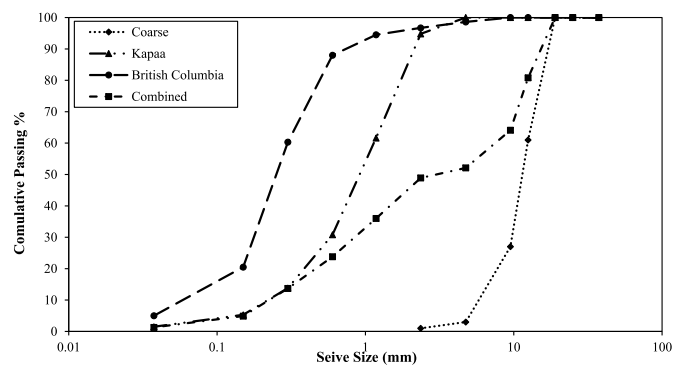


Fig. 1. Sieve analysis of coarse, fine, and combined aggregates.

Table 2
Concrete mix constituents.

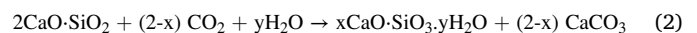
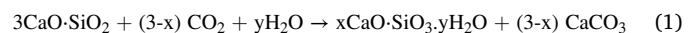
Materials	Quantity (lb.)	Volume (yd ³)
Fine Aggregate (Basalt)	1204	0.27
Fine Aggregate (British Columbia)	473	0.102
Coarse Aggregate (Basalt)	1625	0.357
Cement	442	0.083
Water	288	0.171
Super Plasticizers (fl oz) *	48	
Air Content	-	0.017
Total		1
CO ₂ Liquid Admixture (fl oz) **	30	

*, ** 7.0 and 6.7 fl oz Per 100 lb of Cement.

0.5% by weight of cement³ (). The samples were collected from Island Ready Mix company, where the concrete and paste mixes were casted for concrete pavements. The cement paste (slurry) mix had the same mix proportions as the concrete with a w/c ratio of 0.65 and a CO₂ dosage of 0.5% by weight of cement.

2.2. Hypothesis of liquid CO₂ carbonation

Carbon dioxide was injected into concrete during batching. Pressurized liquid CO₂ is supplied through a metering system to discharge the liquid CO₂. Liquid CO₂ is then converted to a gaseous mixture of finely divided CO₂ particles after it has been discharged. During the initial mixing of the concrete, carbon dioxide may react with the hydrating cement. Calcium carbonate is produced by a rapid reaction between carbon dioxide and calcium ions, which is generated by hydrating cement. The reactions are shown in equations (1) and (2). As a solid phase, the CO₂ reacted in the concrete is chemically bound, so no gaseous CO₂ remains in concrete (Monkman et al., 1016; Shao et al., 2010; CarbonCure's 500 Megatonne CO₂ Reduction; Monkman, MacDonald, Hooton).



2.3. Experimental program

Evaluation and testing were conducted on concrete and paste samples. Freshly mixed concrete was tested for slump. Compressive strength of hardened concrete was evaluated at various ages, as well as shrinkage, creep, alkali silica reaction and sulfate attack tests to evaluate

¹ Hawaii Gas Company.

² Hawaiian Cement Inc.

³ Recommended dosage by CarbonCure Technologies company.

long-term durability. Analyses of chemical compositions and microstructure were carried out using X-Ray Powder Diffraction (XRD) on cement paste samples at various hydration ages to track calcium carbonate formation and hydration products.

3. Results and discussion

3.1. Fresh properties

As per ASTM C143 (ASTM C143-78), the slump of control and carbonated concrete mixes were 4 ± 1 . Both mixes had an air content (ASTM C231 (ASTM C231-09a, 2010)) of 1.7% and a unit weight (ASTM C138 (ASTM C138/C138M-17a)) of 2393.2 kg/m³ (149.4 lb./ft³). Adding liquid CO₂ to concrete did not significantly affect slump flow, air content, or unit weight. Therefore, injecting CO₂ admixture does not negatively affect fresh properties of concrete.

3.2. Compressive strength

Compressive strength of control and carbonated samples were tested according to ASTM C39 (ASTM C39/C39M-21, 2021) on three replicas for each group of samples at 7 and 28 days of hydration, under moist curing condition. As shown in Fig. 2, the control mix design had an average compressive strength of 14.35 MPa at 7 days. In contrast, the carbonated concrete had an average compressive strength of 15.13 MPa showing a 5% improvement. The 28 days compressive strength for carbonated concrete was 23.8 MPa, while for the control mix was 23 MPa, showing a 3% improvement for the carbonated mixes. The average compressive strength of carbonated concrete is generally higher than that of control concrete mix, and injecting CO₂ into the fresh concrete mix may enhance the compressive strength.

3.2.1. Statistical analysis of compressive strength

A statistical analysis was applied to 10 concrete samples of each mix to assess if it is statistically stronger than the control mix after 28 days, and whether it can help reduce cement content. The compressive strength results of each group of concrete were tested using the small sample test method based on the *t*-test distribution. *T*-tests are used to check for significant differences between two sets of data. Initially, the null hypothesis is assumed to be true (no significant difference between the mean values of the two groups) and the results are normally distributed. Typically, this test can be used when the sample size is too small, and the groups are independent (Navidi, 1259; Kim, 2015; Emmert-Streib and Dehmer, 2019). In a population $N(\mu, \sigma^2)$ with n samples, samples should have a normally distributed mean \bar{X} with $N(\mu, \sigma^2/n)$. In the presence of a null hypothesis, standardizing statistics requires using a normal distribution $= \frac{\bar{X} - \mu_0}{\sigma/\sqrt{n}}$. for the unknown population variance, a sample variance s^2 can be substituted. Therefore, the

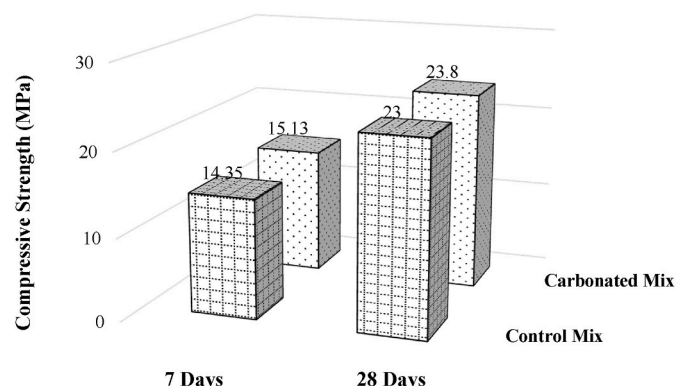


Fig. 2. Compressive strength of carbonated and control concrete mixes.

statistics are based on the *t*-distribution with $n - 1$ degrees of freedom (Navidi, 1259). The following formula was used to calculate standard deviation (s) for small samples (Navidi, 1259).

$$S = \sqrt{\frac{(x_1 - \bar{X})^2 + \dots + (x_n - \bar{X})^2}{n - 1}} \quad (4)$$

In addition, the following formulation allows for the calculation of degrees of freedom based on the standard deviation and number of samples for both dataset (Navidi, 1259):

$$\nu = \frac{\left(\frac{s_1^2}{n_1} + \frac{s_2^2}{n_2}\right)^2}{\frac{\left(\frac{s_1^2}{n_1}\right)^2}{n_1 - 1} + \frac{\left(\frac{s_2^2}{n_2}\right)^2}{n_2 - 1}} \quad (5)$$

Then, the test static is calculated using equation (6) (Navidi, 1259):

$$t = \frac{(\bar{X}_1 - \bar{X}_2) - (\mu_1 - \mu_2)}{\sqrt{\frac{s_1^2}{n_1} + \frac{s_2^2}{n_2}}} \quad (6)$$

For the null hypothesis $\mu_1 - \mu_2 \leq 0$ and for the alternate hypothesis $\mu_1 - \mu_2 > 0$. The test static probability can be calculated from *t*-tables, indicating whether the null hypothesis is true (Navidi, 1259). To determine whether the *t*-distribution applies to samples in this study, a box plot of their compressive strength is shown in Fig. 3. When the samples are normally distributed, the *t*-test works well. Small sample sizes make it difficult to determine the distribution of samples. It would therefore be better to use a box plot to check for asymmetry or outliers. Based on the box plot in Fig. 3, both types of concrete mixes have approximately symmetric data with no outliers. As a result, they can be tested using the *t*-test method. The sample mean and standard deviation for both mix types are shown in Table 3. The average compressive strength for carbonated samples is 23.18 MPa and for the control mix is 21.91 MPa after 28 days, with the deviation of 6%. Similarly, Equations (5) and (6) were used to calculate the degree of freedom and static test for null hypothesis, and the results are shown in Table 3.

According to the degree of freedom, *t*-test results, and *t*-table (Navidi, 1259), for concrete samples the probability values (*p*-values) are 0.25 and 0.1 and the *t*-values are 0.765 and 1.638, while the observed value ($t = 1.384$) from the samples falls between those values. A smaller *p*-value indicates stronger evidence against a null hypothesis, whereas a larger *p*-value indicates stronger evidence for the hypothesis. Considering the *p*-value exceeds 10%, the null hypothesis cannot be refuted, and there is no statistically significant evidence to conclude that carbonated samples are stronger than control samples in this study. Thus, liquid CO₂ admixture cannot reduce cement content within concrete mix design or improve the compressive strength of concrete.

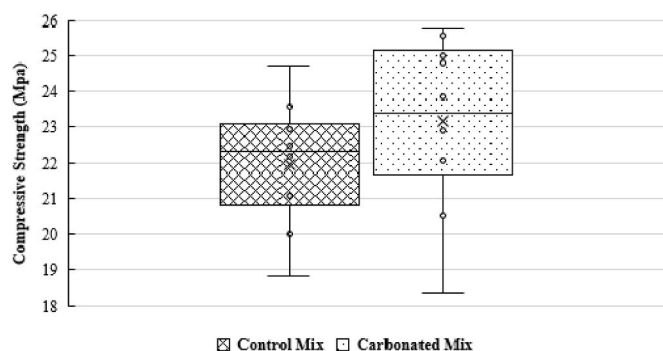


Fig. 3. Boxplot of compressive strength data for control and carbonated concretes.

Table 3
Statistical values for *t*-test analysis.

Values	Control Mix	Carbonated Mix
Sample Mean	21.91	23.18
Standard Deviation	1.68	2.366
Degree of Freedom	16.24	
Static Test	1.384	

3.3. Drying shrinkage

Drying shrinkage for mortar bars were tested according to ASTM C157 (ASTM C157-75), under moist curing condition, and the samples were stored in lime-saturated water storage for 64 weeks. Fig. 4 shows similar shrinkage between these two mixes from the beginning to up to 64 weeks. There was a lower length change in the CO₂ sample after 28 days, 8 weeks, and 16 weeks than in the control sample. Nevertheless, CO₂ samples showed greater shrinkage after 32 and 64 weeks. Generally, both samples shrank less than 0.05% after 28 days, and less than 0.2% after 64 weeks. Thus, injecting liquid CO₂ seems not negatively or positively affect drying shrinkage of concrete.

3.4. Creep

The volume of concrete changes throughout its service life due to applied loads and shrinkage and long-term creep tests were conducted on cylindrical specimens based on ASTM C512 (ASTM C512/C512M-15, 2016). Fig. 5 compares the creep of carbonated concrete and normal concrete over time. The responses of both concrete mixes were similar since the first days of constant loading. Normal concrete showed 397.45 micro strains of creep after 28 days, while carbonated concrete had 426.4 micro strains. In CO₂-treated concrete, the strain rate after 120 days was 450.8 micro strains, while in normal concrete, it was 458.3 micro strains. After one year, the strain of control concrete was 553.9 micro strains while the strain of carbonated concrete was 561.5 micro strains. Cement paste creep results from capillary networks within the cement gel, which increases with higher water to cement ratios. Therefore, it is reasonable to observe relatively high concrete creep due to the high water to cement ratio. However, the results indicate that injecting carbon dioxide into concrete during mixing may not significantly change its creep behavior.

3.5. Alkali silica reaction

Fig. 6 shows sample expansion due to alkali-silica reactions. Readings were taken from a comparator dial in relation to a reference bar and the length change of the specimen after continuous soaking in sodium hydroxide solution at various ages. As per ASTM C1260 (ASTM C1260-22, 2022), expansions less than 0.10% at 28 days after casting indicate innocuous behavior in most cases; expansions greater than 0.20% at 28 days after casting indicate potentially deleterious behavior;

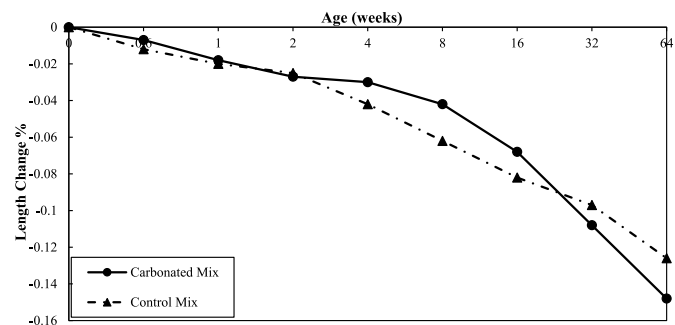


Fig. 4. Drying shrinkage of carbonated and control samples.

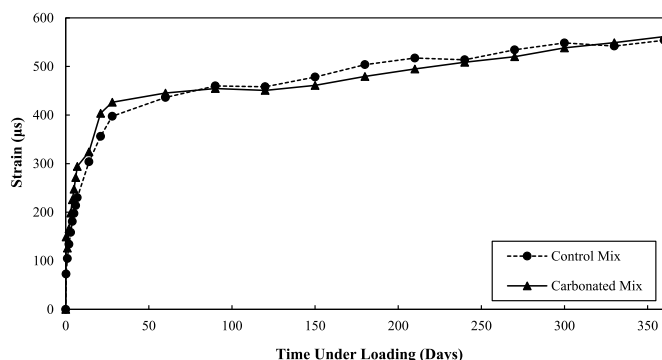


Fig. 5. Creep of carbonated and control concrete over a year.

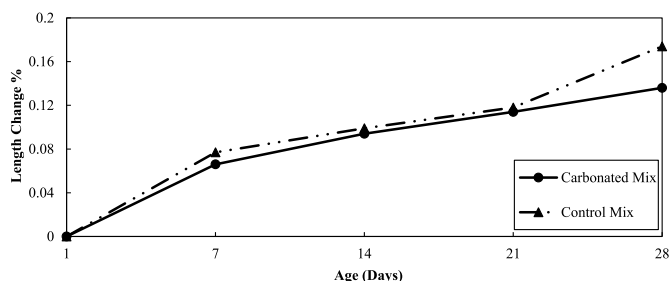


Fig. 6. Length change of concretes due to alkali silica reaction.

and expansions between 0.10 and 0.20% include both innocuous and deleterious aggregates in most cases. Considering the experimental expansions were below 0.20 percent, the aggregates used in this experiment are determined not to be potentially detrimental to internal expansion due to alkali-silica reactions. Because the expansions of both test specimens were similar, it can be concluded that utilizing carbon dioxide in the concrete mix may not affect expansion attributed to alkali-silica reaction.

3.6. Sulfate attack

Fig. 7 depicts the measured bar expansions for carbon-cured and control mortar samples subjected to sulfate attack after 1, 2, 3, 4, 8, 13, and 15 weeks, as well as 4–12 months. Following ASTM C1012 (ASTM C1012/C1012M-18b), three samples from both normal and carbon-cured were immersed in a sodium sulfate solution with concentration of 50 gr dissolved in 900 mL of water. The average length change after one year of monitoring is 0.019% for normal samples and 0.0127% for CO₂ samples. Concrete specimens' expansion was relatively low, at less than 0.05 percent, indicating that they can withstand severe sulfate environments. Moreover, the expansion rates of both specimens were similar, so incorporating carbon dioxide into the concrete mix does not seem to compromise the concrete's sulfate resistance in this study.

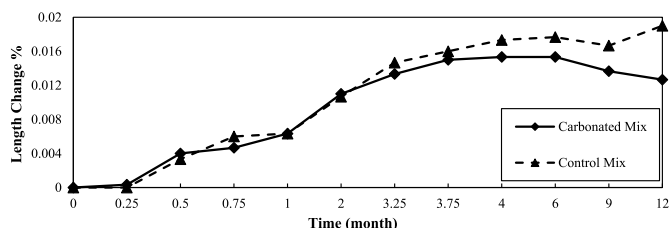


Fig. 7. Length change of concretes due to sulfate attack.

3.7. X-Ray Diffraction

Cement paste powder samples were analyzed using high-resolution X-ray powder diffraction on a Bruker D8 Advance diffractometer with a 3 kW CuK α source. Rietveld analysis was conducted using Bruker TOPAS 5 (Coelho, 2018). The Crystallographic information about mineral phases of portlandite, ettringite, alite (C₃S), and calcite were taken from the ICDD Powder Diffraction File PDF-4+ database: PDF 00-004-0733, PDF 04-013-3691, PDF 00-055-0740, and PDF 00-047-1743, respectively (Gates-Rector and Blanton, 2019). Refinement included optimization of background, sample displacement, unit cell parameters and peak profiles (controlled by grain size and strain models) and peak type. Additionally, single crystal XRD analysis of individual grains or small grain aggregates of optically distinct phases was performed for unambiguous detection of pure crystalline phase of calcite, using a Bruker D8 Venture single crystal diffractometer equipped with a PHOTON-II CPAD detector and an Ag K α I μ S microfocus source (0.56089 Å), and the data collection was conducted using APEX3 software (Version, 2018.1-0, Bruker AXS Inc., Madison, WI, USA) (APEX3, 2016). The tests and analysis were conducted at the University of Hawai'i at Manoa X-Ray Atlas Diffraction Laboratory.

The XRD patterns of crystalline phases for control and CO₂ injected paste samples are shown in Figs. 8 and 9. The patterns are for samples after hydration of 7 and 28 days. Diffraction peaks of the major hydration products, such as portlandite, ettringite, and calcium carbonate (calcite); and major crystalline phases of cement, such as the tricalcium silicate (alite) are indicated in Figs. 8 and 9.

For CO₂ injection and control mix samples after 7 and 28 days, the crystalline phase of carbonate was fitted to the pattern along with portlandite, alite and ettringite as shown in Figs. 9–12. In Figs. 9 and 10 the green curve is the experimental pattern, grey below is a difference curve between experimental and calculated pattern, red is fitted profile and purple is calculated profile for the selected phase (calcite). Similarly, in Fig. 11 the blue curve is the experimental pattern, grey below is a difference curve, red is fitted profile and purple is calculated profile for the selected phase (alite). And, in Fig. 12 the black curve is the experimental pattern, grey below is a difference curve, red is fitted profile and blue is calculated profile for the selected phase (calcite). Although the XRD pattern of control samples are very similar to the carbonated ones, the phase fractions of hydration products in these two mixes are different. The lower percentage of portlandite and ettringite in carbonated samples for both 7 and 28 days after hydration, compared to the control mix, may be due to binding CO₂ admixture within concrete mix design and its effect on hydration process. The carbonated system had an increase in

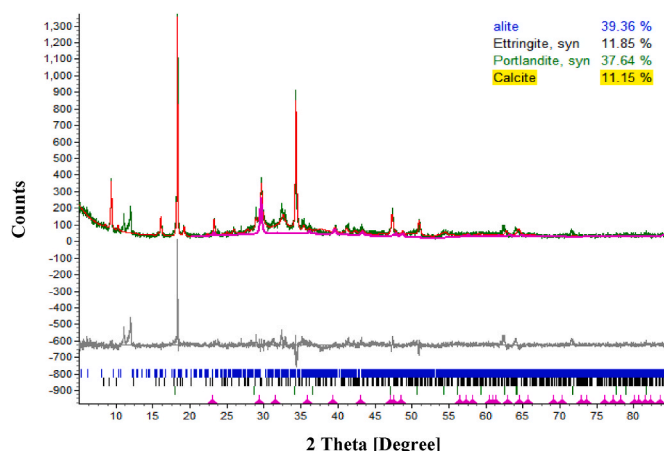


Fig. 9. Rietveld analysis of control sample after 7 days of hydration.

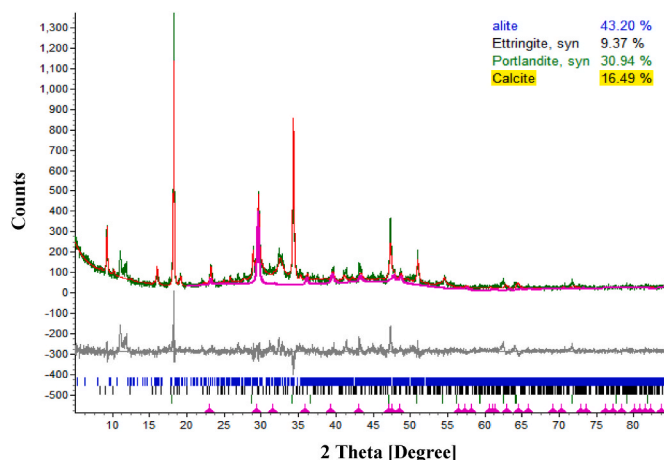


Fig. 10. Rietveld analysis of carbonated sample after 7 days of hydration.

the amount of carbonate and a corresponding decrease in the amount of portlandite and ettringite formed. The CO₂ injected mix after 7 days has about 16.5% of carbonate, 31% of portlandite and 9.4% of ettringite formed, while the control mix contains 11% of calcium carbonate, 37% portlandite and 12% of ettringite. The same results were derived from

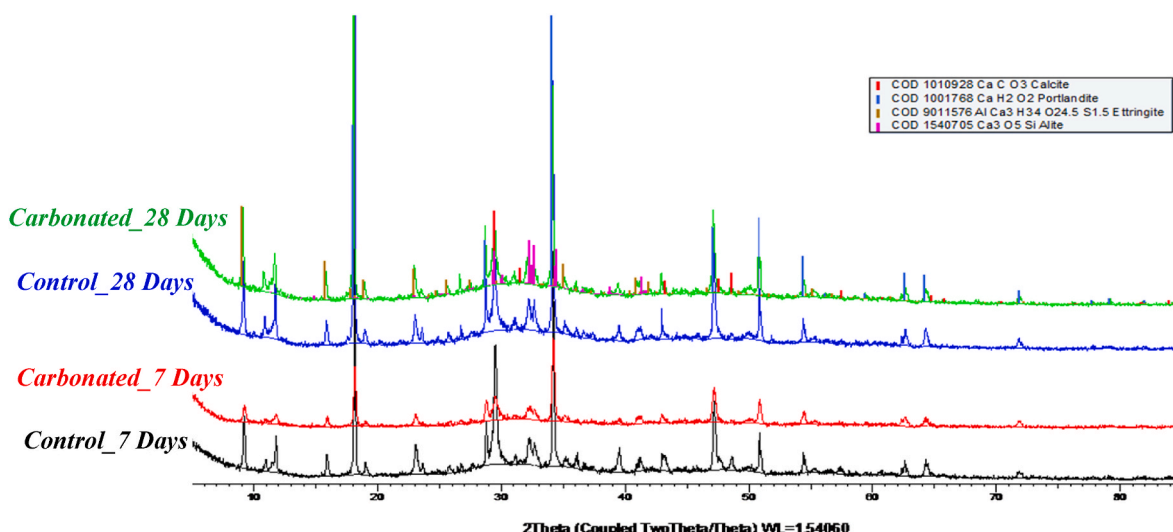


Fig. 8. XRD patterns of pastes for carbonated and control mixes after 7 and 28 days of hydration.

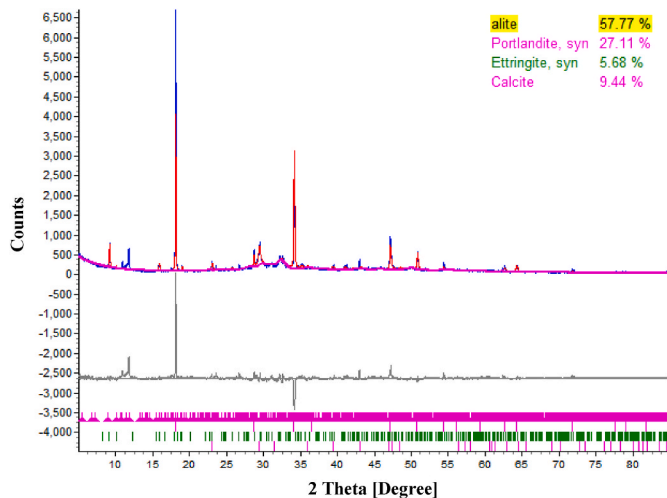


Fig. 11. Rietveld analysis of control sample after 28 days of hydration.

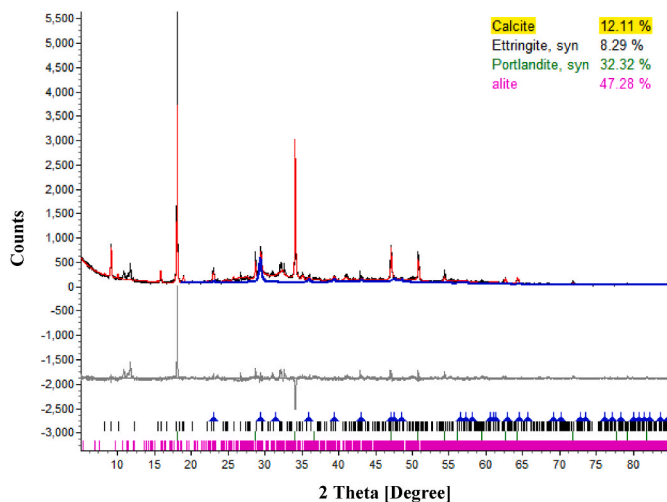


Fig. 12. Rietveld analysis of carbonated sample after 28 days of hydration.

the structural refinement analysis for sample after 28 days of carbon dioxide injection and control mixes, respectively.

Further, single grain analysis was carried out using optical microscope to select randomly different grains within small sample of the paste powder of control and carbonated mixes to verify weather pure crystalline phase of calcite is within the system or no. Results for diffraction patterns and XRD patterns of portlandite, alite, and calcite are shown in Figs. 13 and 14. The grains were retrieved from the samples for both 7 and 28 days of hydration and mounted on a 10- μm MiTeGen mesh needle (Fig. 15). As a result, the pure calcite crystalline phase was successfully detected in carbonated samples for 7 and 28 days of hydration, which would be an indication of the reaction between CO_2 admixture and hydrated cement compounds such as alite. However, it should be noted that cement paste powder can easily be carbonated within the environment, since there is carbon dioxide in the atmosphere. Therefore, it is not possible to verify by XRD analysis, whether the carbonate phases are from the reaction of liquid CO_2 with hydrated cement phases or from the reaction of carbon dioxide in the atmosphere with paste powder.

3.8. Scanning Electron Microscopy

Samples were prepared for SEM analysis technique at the Advanced Electron Microscopy Center (AEMC) of the University of Hawaii at

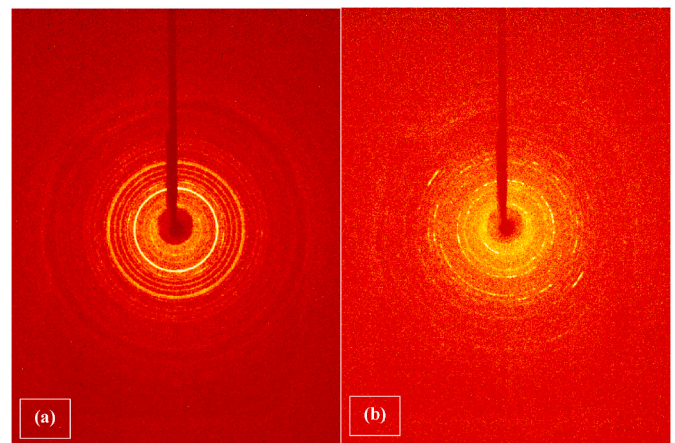


Fig. 13. Single grain aggregate diffraction pattern for calcite/portlandite (a) and portlandite/alite (b) grains.

Manoa. The CO_2 injected samples were gold coated instead of carbon coated to eliminate the effect of carbon coating on detection of carbonates during EDS mapping. The analysis of samples for finding the carbonate in cement powder samples was carried out using SEM-EDS (energy-dispersive X-ray spectroscopy) mapping. The SEM-EDS mapping was conducted with the FEI Helios Nano Lab 660 Extreme High-Resolution Dual Beam FIB instrument equipped with variable landing energy (20 V–30 kV), high electron beams spatial resolution, and variable incident ion beam acceleration (500 V–30 kV) which allows low-kV surface cleaning. Fig. 16 shows the backscattered SEM images of hydrated products within the matrix of cement paste samples of CO_2 injected, after 7 and 28 days. SEM micrographs exhibited a multitude of hexagonal-shaped portlandite and needle-shape of ettringite crystals, along with calcium silicate hydroxide (C–S–H) in the CO_2 -injected system. However, no cubic shape of pure carbonate crystals was spotted in the matrix of carbonated cement paste. This may be due to the duration of data collection to map the carbonate rich regions not being as much as would be ideal to obtain a high signal to noise map. In addition, the low abundance of pure carbonate phase from the possible reaction of liquid CO_2 with hydrated cement within the paste matrix, would be another reason for not being detected in cement paste (see Fig. 17).

4. Conclusions

This research evaluated the effect of using liquid CO_2 admixture in concrete mix design as a method to reduce carbon footprint of cement and concrete industries. As a result of injecting liquid CO_2 into fresh concrete, the overall compressive strength of the concrete increased by about 5% after 7 days and 3% after 28 days. However, statistical analysis using *t*-test revealed no statistically significant difference between both groups of samples in terms of 28 days of compressive strength for concrete mixes. Even though adding liquid CO_2 may positively affect compressive strength, it could be challenging to reduce the amount of cement within concrete mix design to reach to the same compressive strength of control sample. Moreover, it shows that adding liquid CO_2 to the fresh concrete mix has no significant effect on the concrete's long-term and short-term mechanical properties, such as shrinkage and creep, as well as durability tests such as sulfate attack and alkali silica reaction. The finding that adding CO_2 admixture does not positively or negatively affect concrete performance may help address the scientific questions on whether sequestering CO_2 in concrete would negatively influence concrete properties. Further, microstructural analysis of paste samples was carried out using XRD and SEM to evaluate the formation of calcite from the possible reaction between hydrated cement and CO_2 , as well as its effect on hydration products within the paste matrix. Pure

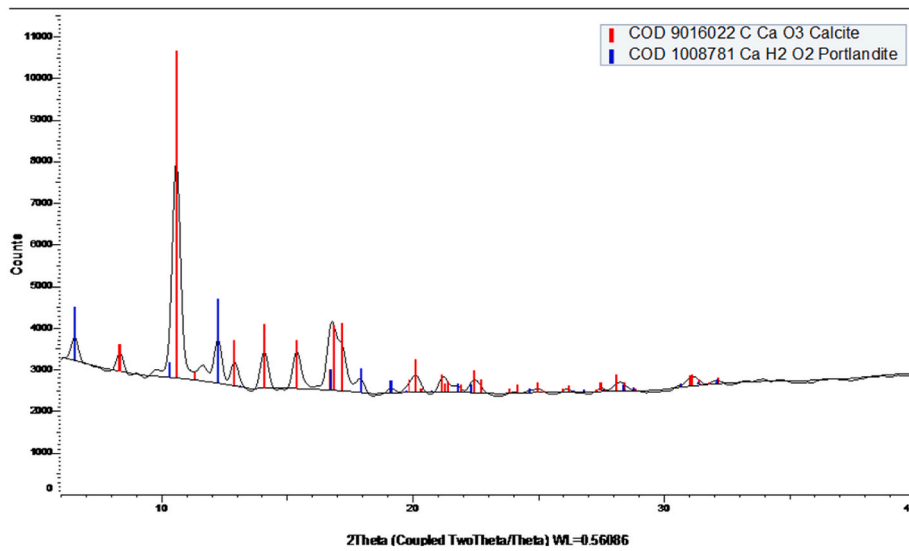


Fig. 14. Single grain aggregate XRD pattern of calcite and portlandite in carbonated samples.

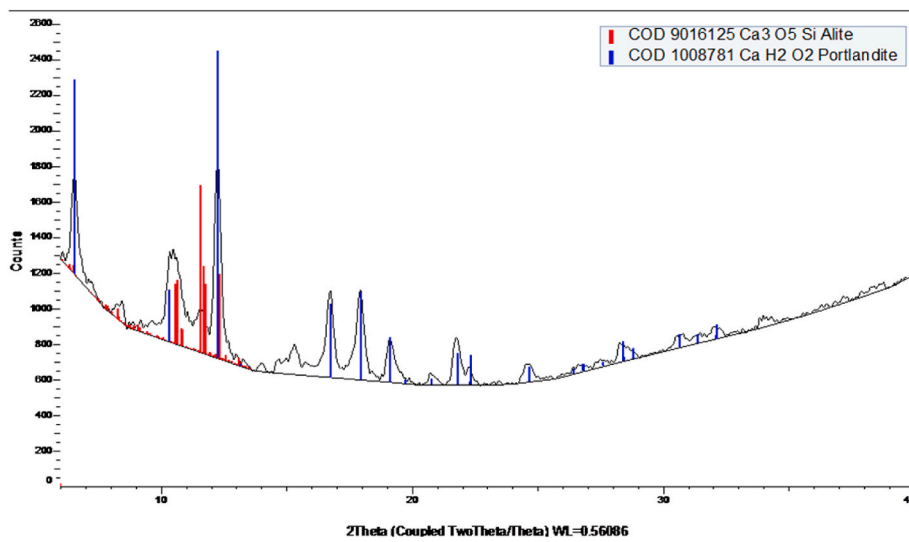


Fig. 15. Single grain aggregate XRD pattern of alite and portlandite in carbonated samples.

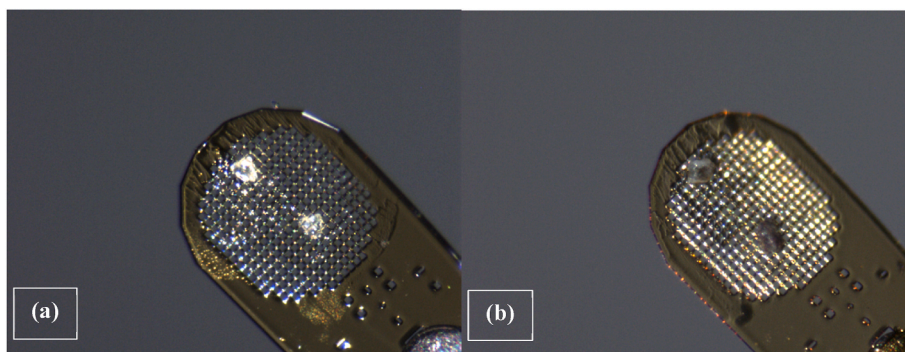


Fig. 16. Calcite and portlandite (a), portlandite and alite (b) grains selected for single grain analysis and mounted on a MiTeGen mesh.

crystalline phase of calcite was successfully identified with XRD single grain analysis in carbonated samples, however there is not enough evidence that the detected phase is from the rapid reaction of injected CO₂ with cement, or it is from the carbonation of paste powder with CO₂ in

the atmosphere. Finally, SEM analysis of CO₂-injected samples did not show any distinctive features of pure cubic carbonate phase within the systems.

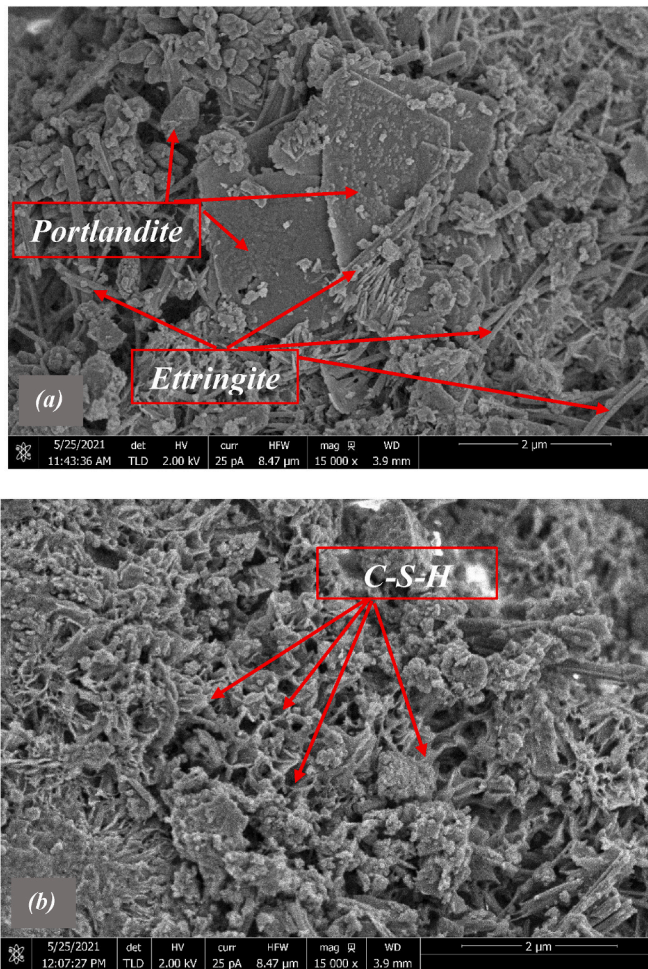


Fig. 17. SEM images (a) and (b) of hydration products of carbonated samples.

CRedit authorship contribution statement

Azadeh Hosseinpanahi: Methodology, Validation, Formal analysis, Investigation, Writing – original draft. **Lin Shen:** Conceptualization, Validation, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Przemyslaw Dera:** Formal analysis, Investigation, Writing – review & editing. **Reza Mirmoghtadaei:** Investigation.

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.

Data availability

Data will be made available on request.

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