



# Carbon dioxide upcycling into industrially produced concrete blocks



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## HIGHLIGHTS

- Industrial trial investigated injection of CO<sub>2</sub> in concrete mixer at masonry plant.
- Blocks became less dense and harder to compact; mix water increased in response.
- Concrete block compressive strength and absorption was improved.
- The carbon dioxide was efficiently absorbed into the fresh concrete.
- Approach offers net sequestration of carbon dioxide into useful building products.

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## ABSTRACT

A novel carbon utilization trial produced carbon dioxide treated concrete blocks by adding carbon dioxide gas to the concrete during the mixing and forming stages of the block production. The gas was supplied at up to 1.5% by weight of cement. The carbon dioxide was absorbed into the concrete with an average efficiency of about 88% of the gas delivered. The carbon dioxide was shown to reduce the block compaction (and thereby the density) but increasing the mix water was an effective countermeasure. The carbonation process was shown to increase compressive strength (13–33% at ages from 7 to 56 days) and decrease absorption (by 18–36%). The CO<sub>2</sub> upcycling reduced the carbon footprint of the blocks by 1.4% thereby demonstrating a simple approach to utilizing carbon dioxide to make useful construction materials.

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## 1. Introduction

Carbon dioxide emissions are recognized as a significant issue for the cement and concrete industry. It is estimated that 5% of the world's annual CO<sub>2</sub> emissions are attributable to cement production [1]. Portland cement clinker typically has embodied CO<sub>2</sub> on the order of 866 kg CO<sub>2e</sub>/t of clinker [2]. Cement production releases CO<sub>2</sub> due to the calcination of limestone that is heated to drive off CO<sub>2</sub> and yield reactive CaO phases. Carbon dioxide emissions are further associated with the energy required to operate the cement kiln. About 40% of the process emissions are associated with the energy consumption and 60% are associated with the calcination.

A number of approaches have been identified to reduce the emissions intensity of the cement produced and used [3]. The thermal and electrical efficiency of cement production can be improved by deploying the best available technology in new cement plants and retrofits. Alternative and less carbon-intensive fuels can be used as the energy source. The rate of substitution in blended

cements can be maximized. Finally, carbon capture and storage (CCS) can capture cement industry CO<sub>2</sub> emissions before they are released and store them permanently.

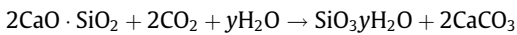
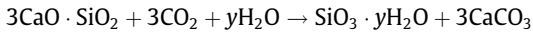
It is clear, however, that practical limits on the impacts of these measures mean that it will be difficult [4] to attain the industry goal to reduce emissions 50% below 2006 levels by 2050 that is outlined in the IEA & WBCSD roadmap [3]. The limitation of conventional approaches is highlighted when Carbon Capture and Storage, a developing but uncertain technology, has been proposed to account for 56% of the sought reductions. Thus, a range of further approaches will also have to be pursued.

One potential method is to upcycle carbon dioxide into concrete products by treating them with CO<sub>2</sub> prior to the end of their processing, such as during the curing stage [5–9]. If an industrial process could successfully use carbon dioxide as a feedstock in the production of concrete blocks there would be widely distributed carbon utilization that would effectively ‘close the loop’ for some of the carbon dioxide emitted during the cement production while simultaneously producing useful building products.

The carbonation of freshly hydrating cement involves the reaction of CO<sub>2</sub> with the main calcium silicate phases to form calcium carbonate and silicate hydrate gel [10]:

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The reaction occurs in the aqueous state when  $\text{Ca}^{2+}$  ions from the cementitious phases react with  $\text{CO}_3^{2-}$  ions from the applied gas. The carbonation reaction is exothermic evolving 347 kJ/mol for  $\text{C}_3\text{S}$  and 184 kJ/mol for  $\beta\text{-C}_2\text{S}$  [10]. When the calcium silicates carbonate, the formed  $\text{CaCO}_3$  is understood to be co-formed with calcium silicate hydrate (C-S-H) gel which itself can lose CaO and water to convert to silica gel [10]. Gel formation has been observed even in the model cases of reacting  $\beta\text{-C}_2\text{S}$  and  $\text{C}_3\text{S}$  exposed to a 100%  $\text{CO}_2$ . It was found that the amount of calcium silicate that reacted exceeded the amount that would be attributable to the formation of the carbonate products alone [10].

The reaction of carbon dioxide with a mature concrete microstructure is acknowledged as a durability issue given effects such as shrinkage, reduced pore solution pH and carbonation induced corrosion. In contrast, a carbonation reaction integrated into concrete production reacts  $\text{CO}_2$  with freshly hydrating cement, rather than the hydration phases present in mature concrete, and does not have the same effects. The potential to integrate beneficial carbon dioxide utilization into industrial block production was investigated.

An industrial scale experiment was performed whereby gaseous carbon dioxide was injected into mixing concrete that was then used to create concrete masonry units. The goal was to assess the potential of using the concrete to sequester carbon dioxide during production albeit via a universally deployable retrofit approach that involves minimal disruption to the conventional production process. Secondly, the experiment determined the attendant material science impacts. Finally, the carbon dioxide uptake was assessed to determine how much gas became locked in the concrete as stable reaction products.

## 2. Materials and experimental

### 2.1. Mix design and procedure

A tank of liquid  $\text{CO}_2$  was connected to a gas control system and manifold. The liquid was expanded to a gas and metered for injection into the mixer. The gas was delivered, at a specified flow rate over a fixed injection interval, whereupon it reacted with the hydrating cement before the concrete mix was finished.

A medium weight mix design was used to make standard 8" (200 mm) concrete blocks. The mix design contained 8.8% (by mass, excluding mix water) gravel, 55.7% sand, 16.1% silica sand, and 12.9% cement. The mix design also contained an admixture to improve the water repellency of the block. Carbon dioxide was injected at a rate of 0.5 to 1.5% by weight of cement (bwc).

The cement used in the trial was an ordinary Portland cement from the Illinois Cement Company with a Blaine fineness of  $501 \text{ m}^2/\text{kg}$ . The composition as deduced from analyses done using X-ray Fluorescence (XRF) is shown in Table 1.

### 2.2. Gas supply

The carbon dioxide used in the experiment was purchased as liquid  $\text{CO}_2$  from an industrial supplier. Such carbon dioxide, almost

**Table 1**  
Composition (in wt%) of the ordinary Portland cement found using the Bogue formulas (ASTM C150) and deduced from XRF analysis.

$\text{C}_3\text{S}$	$\text{C}_2\text{S}$	$\text{C}_3\text{A}$	$\text{C}_4\text{AF}$	MgO	$\text{K}_2\text{O}$	$\text{Na}_2\text{O}$
52.7	15.1	10.7	6.8	2.7	1.0	0.3

without exception, is captured as a byproduct from an industrial process (e.g. ammonia, fertilizer or ethanol production). Consequently, carbon dioxide purchased from an industrial supplier is effectively carbon dioxide that has been diverted from an emission stream and captured to meet market demand. Any carbon dioxide captured in the concrete treatment process using industrially supplied carbon dioxide can represent a net sequestration benefit. Any carbon dioxide from the carbonation process that is lost to the atmosphere (as opposed to being absorbed by the concrete) is taken to be returning to the destination it had had prior to the industrial capture.

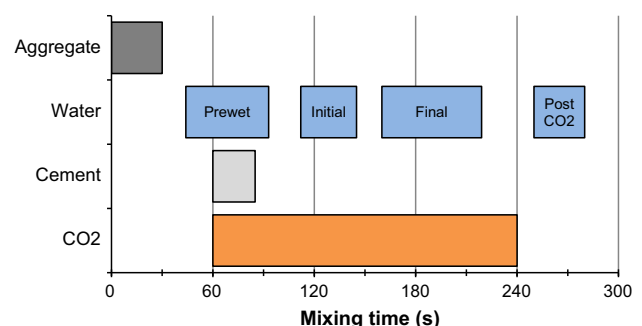
The use of commercial carbon dioxide does come with an environmental cost given that the gas must be captured, liquefied, and transported. Therefore environmental considerations would have to consider all of the dosed  $\text{CO}_2$  (not just the absorbed  $\text{CO}_2$ ) while the net sequestration would have to be assessed on a case-by-case basis according to the specific capture scheme and transportation requirements.

### 2.3. Experimental approach

The carbon dioxide injection equipment was retrofit onto the conventional block production line (a Besser Ultrapac). The carbon dioxide was injected into the concrete mixer (a Besser Rapid Pan Mixer) through an access hatch in the mixer lid. The mixer was effectively enclosed and ensured that the injected gas was contained.

The mixing scheme, as outlined in Fig. 1, started with delivering the aggregates over 30 s. This was followed by the addition of pre-wet water and cement. The carbon dioxide injection was synchronized with the addition of the cement and lasted 3 min which adhered to the default mixing time given the imposed constraint that the process cycle time remained unchanged. The initial water addition took place between 112 and 145 s after mixing started. The final water addition took place across at 160 s after mixing began and lasted 59 s. If needed, a post- $\text{CO}_2$  water addition was added to the end of the cycle. After the batching was complete the concrete would mix for a few minutes prior to being discharged. In a typical mix, the free water associated with the aggregates accounted for 59% of the total mix water, while the prewet, initial and final water additions were 14%, 5% and 23% respectively.

An example consideration (shown in Fig. 2, as per a control sample) of the change in w/c in the concrete mix over the three distinct water additions shows that at the time of cement addition (60 s) the nominal w/c would be 0.27 before increasing to 0.30 when the cement input is complete. When the carbon dioxide injection is finished the mix has reached a final w/c of 0.43. The motivation of injecting the  $\text{CO}_2$  alongside the cement is two fold – to take advantage of the low w/c conditions at that time in the mix cycle (the reaction between hydrating cement and carbon dioxide is known to be accelerated at low w/c [11]), and to maxi-



**Fig. 1.** Schematic representation of batching and  $\text{CO}_2$  injection sequence.

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