More Sustainable Concrete Produced Using Waste Cement Industry CO_2

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ABSTRACT

Concrete is the world's most important and most widely used building material. Cement production is one of the most significant industrial sources of CO_2 emissions. The capture of cement industry CO_2 for upcycling in downstream value-added concrete applications is a a viable, synergistic and beneficial approach.

The beneficial use of carbon dioxide in concrete production has been investigated through lab and industrial studies. Retrofit applications have been developed for masonry block production, ready mix concrete production, and concrete wash water beneficiation.

Concrete masonry blocks produced using a CO_2 injection applied during mixing means that CO_2 is locked into the concrete, the block compressive strength increases and the absorption decreases. A ready mix implementation uses an optimum dose of carbon dioxide to increase the compressive strength of the concrete thereby allowing producers to optimize their mix designs (for example, reducing the cement loading and using the CO_2 to restore the reduced strength) to create reduced carbon footprint of concrete without compromising performance. High solids concrete wash water can be beneficiated for reuse through treatment with CO_2. The process eliminates issues associated with reusing waste water in concrete (accelerated set, increased water demand), leads to reduced use of fresh water and offers sequestration of carbon dioxide.

INTRODUCTION

Concrete is the world's most important building material whose production has steadily increased, particularly in the last 20 years in response to rising demand from emerging economies (Figure 1). The annual global cement production has surpassed an estimated 4.1 Gt (U.S. Geological Survey 2016) . For a generic concrete mix that contains 300 kg cement per cubic meter and a global population of 7.2 billion (U.S. Census Bureau 2016), it is evident that the annual global production of concrete is currently around 1.9 m^3 per person. At a density of 2.3 tonnes per cubic meter there would be 4.3 tonnes of concrete produced annually for each person.

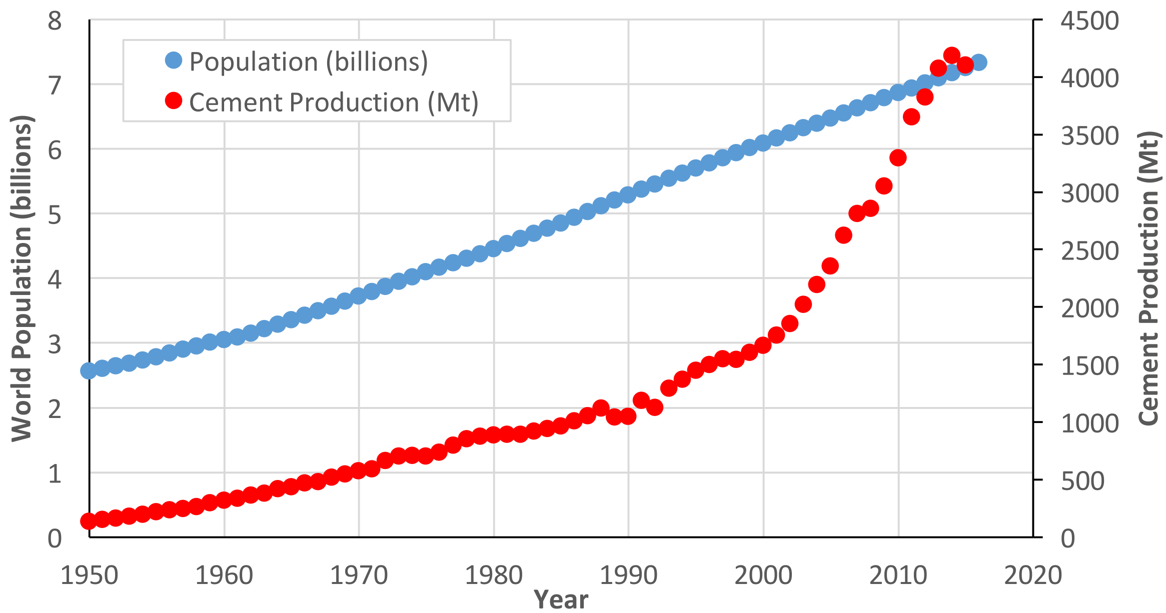

Figure 1. Global Population Growth and Global Cement Production (adapted from (U.S. Census Bureau, 2016) and (U.S. Geological Survey, 2016))

The ever-increasing demand for concrete, combined with the impetus to reduce greenhouse gas emissions, has driven the search for ways to reduce the specific carbon footprint of concrete. If concrete can be made with a lower carbon footprint per unit of production, then there is the potential to increase output without increasing the overall environmental impact.

About 85% of the greenhouse gas emissions associated with concrete are attributable to the cement (Marceau, Nisbet, and VanGeem 2007). Concrete production has long embraced circular production principles especially practices which have additionally served to allow for more efficient usage of cement. Many waste materials have been beneficially used to make concrete (Chandra 1997; Siddique 2008). The most widely used examples are blast furnace slag (a by-product of iron and steel-making), fly ash (a by-product of coal fired power generation), and silica fume (a by-product of silicon metal production). These materials can be diverted from landfills for beneficially reuse in concrete.

New solutions are required to meet the industry goal for lower carbon footprint concrete. One emerging technology segment that has been developed is the concept of carbon dioxide utilization to create concrete products (Ashraf 2016; Jang et al. 2016). The use of carbon dioxide to produce concrete potentially connects one industry waste (carbon dioxide) with the industry's main commercial output thereby offering a potentially attractive upcycling solution that works within the principles of a circular economy (Figure 2).

In order for a CO_2 utilization solution to be accepted and adopted by industry it must a be readily integrable into existing technologies and production modes. Three different technologies have been developed to allow concrete producers to make more sustainable concrete through the beneficial utilization of waste carbon dioxide. The technology segments are masonry block production, ready mixed concrete production, and concrete waste water beneficiation. The carbon dioxide utilization concepts involve the reaction of CO_2 with cement in ways that adhere to otherwise conventional production practices. When the carbon dioxide reacts with cement at the earliest stages of hydration there will be the formation of calcium carbonate. The carbon dioxide is stored permanently in the concrete and can impart performance benefits.

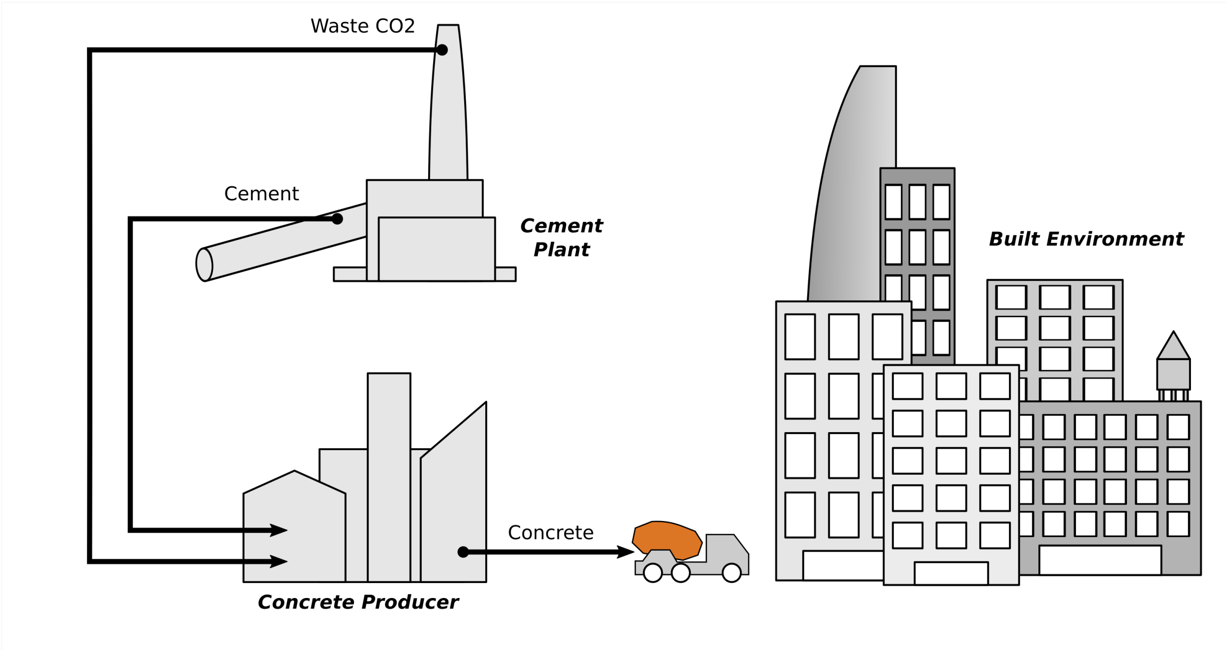

Figure 2. Circular economy principles for concrete production – Use waste CO_2 from cement plants to create better concrete

The present carbon dioxide utilization work began with the development of a technology for masonry block production. The approach involved injecting carbon dioxide into mixing concrete before it was molded into blocks. The carbon dioxide injection was centralized thereby allowing one hardware retrofit to address any concrete made in the production setup. The goal was to maximize the amount of carbon dioxide absorbed by the concrete while maintaining the existing cycle time and production rate.

The injection of carbon dioxide into a concrete mixer was extended for applications in ready mix concrete production. Preliminary work concluded that a small dose of carbon dioxide could feasibly be used to provide performance benefits in ready-mixed concrete. A pilot program of limited scope suggested that the CO_2 addition could produce a strength benefit. A permanent system installation permitted a study of the performance outcomes and the potential to leverage the strength benefit to produce lower carbon footprint mix designs and provide a net environmental benefit.

A final approach to addressing the environmental impact of concrete production revolves around industry waste management practices. A large volume of wash water is generated through the cleaning and maintenance of concrete mixing trucks. The handling and disposal of this water represents a large operational and financial burden to concrete producers. A common solution within the industry is to reuse a portion of the water generated from washing as batch water in subsequent loads of concrete, thereby creating a closed loop of water use. While viable, this reuse strategy is limited by negative impacts including unacceptable acceleration to time of initial set and reduced workability of fresh concrete. A carbon dioxide treatment of the wash water can reduce the scale of the negative properties associated with wash water reuse to the point where they may be easily managed by QA/QC personnel at extreme specific gravity via conventional strategies. Additional benefits in compressive strength may also be achieved, allowing for the creating of additional value from wash water reuse

CO2 UTILIZATION CONCEPTS

Laboratory and Industrial scale research was conducted to examine each of the carbon dioxide utilization approaches. The masonry block and ready mix concrete approaches were developed into industrial production approaches. The wash water beneficiation concept was assessed at a proof-of-concept lab scale.

Masonry Blocks

Research Outline The carbon dioxide injection equipment was installed as a retrofit onto the conventional block production line. CO_2 gas was injected once all the dry materials had been loaded. The duration was ultimately limited by the constraint of avoiding any extension of the process cycle time. A medium weight mix design was used to make standard 8” (200 mm) concrete blocks. A carbon dioxide injection at a rate of 1.5% by weight of cement was found to be achievable without impacting the production rate.

The fresh concrete was assessed visually and via feedback from the machine (i.e. compaction time) and production personnel. The finished blocks were transported to a curing room heated with forced humid air where they were held for 3 or 4 days.

Concrete performance was assessed through compressive strength testing at 7, 28 and 56 day testing at the producer’s testing laboratory with 5 blocks for each condition at each test age. Water absorption and density testing was also conducted on three blocks per condition. Testing was conducted according to ASTM C90 - Standard Specification for Loadbearing Concrete Masonry Units.

Masonry Results The production observed that the CO_2 had a perceivable drying effect and the carbonated mix required additional water (about 10%) to make visually acceptable blocks. The detectable carbon uptake of the treated batches was found to reach 93% of the supplied CO_2. The strength development of the test batches is shown in Figure 3. The carbon dioxide resulted in a strength increase in the range 18-19% at all three ages. Furthermore, the water absorption was reduced by 18%.

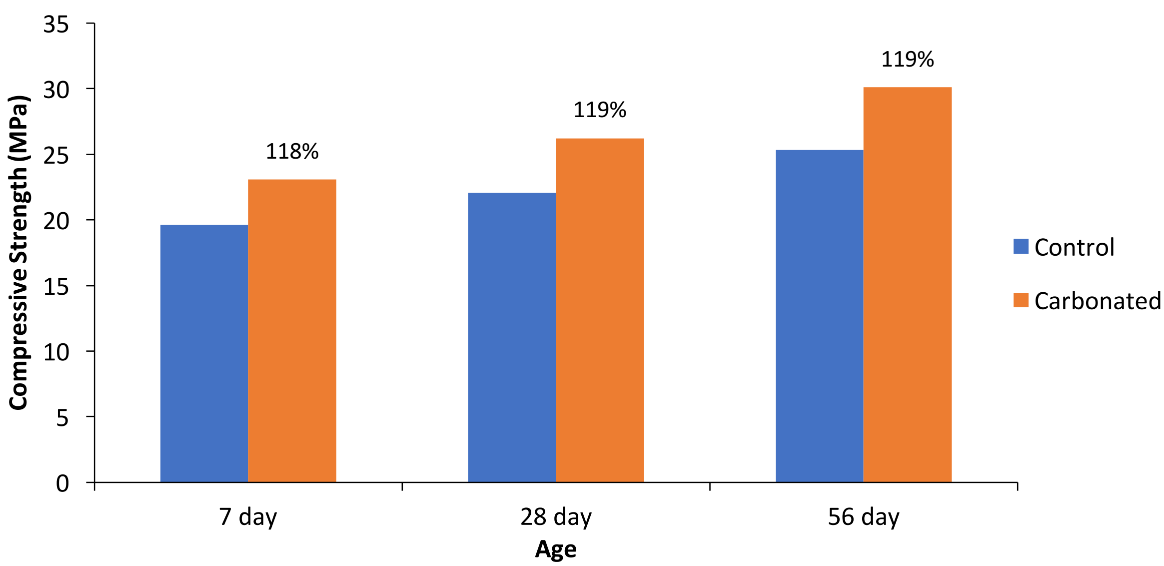

Figure 3. Compressive Strength of concrete masonry units produced with and without a CO_2 addition

The performance of the CO_2 batch is not attributable to any difference to an improved block density; the block density for the two conditions was equivalent (2093 kg/m^3 for the control vs 2088 kg/m^3 for the carbonated set). The addition of the CO_2 may have had a chemical impact that contributed to a strength increase, and in a general sense it may have allowed a higher water content in the block and increased the overall hydration. The conclusion is that the carbon dioxide injection can be simply integrated as a retrofit into a conventional block production line and unlock material benefits alongside achieving permanent storage of CO_2 into the blocks.

The CO_2 absorption, after consideration of the energy required to carry out the process (Monkman and MacDonald 2016), means that a net of 30.8 kg of carbon dioxide would be permanently locked within a 100 m^2 concrete block wall.

Ready-mixed Concrete

Research Outline Ready mixed concrete was produced whereby an optimal dose of carbon dioxide was injected into the central mixer during batching. A gas metering system fed a controlled supply of liquid CO_2 through to a discharge conduit. The liquid was converted into a mixture of CO_2 gas and finely divided solid carbon dioxide particles (commonly referred to as CO_2 "snow") The carbon dioxide was delivered into the fresh concrete, at a specified flow rate over a fixed injection interval, whereupon it reacted with the hydrating cement during initial mixing. The concrete was then delivered to the ready mix truck for assessment and testing.

The trial focused on a three way comparison between a control mix design, a reference mix design with reduced cement compared to control, and the reference mix design produced using a CO_2 addition. The concrete had a design strength 35 MPa. The modified mix design included a 5% reduction in the cement loading which was a 3.75% reduction of overall binder. The sand was increased in the modified mix design to maintain yield. The carbon dioxide was added at 0.10% by weight of cement.

Ready-mixed concrete results The air content of the control batch and the CO_2 batch were similar (5.5 vs 5.7%) whereas it was increased for the reduced cement reference batch (6.6%). The slump (workability) was 80 mm for the control, 100 mm for the reduced cement reference and 120 mm for the CO_2 treated concrete. While the fresh properties were acceptable in all cases it is likely that the variation was associated with the further need to optimize the admixture loading on the modified mix design.

The 28 day compressive strength results are summarized in Figure 4. The concrete was shown to have an 11% lower strength when the binder was removed (though the increased air content likely contributed to some of this difference). In turn, the addition of the CO_2 restored the lost strength. Relative to the reduced cement mix design with CO_2, the carbon dioxide increased the concrete strength by 23%. Compared to the unadjusted mix the CO_2 contributed to a 9% gain at 28 days – despite having 5% less cement.

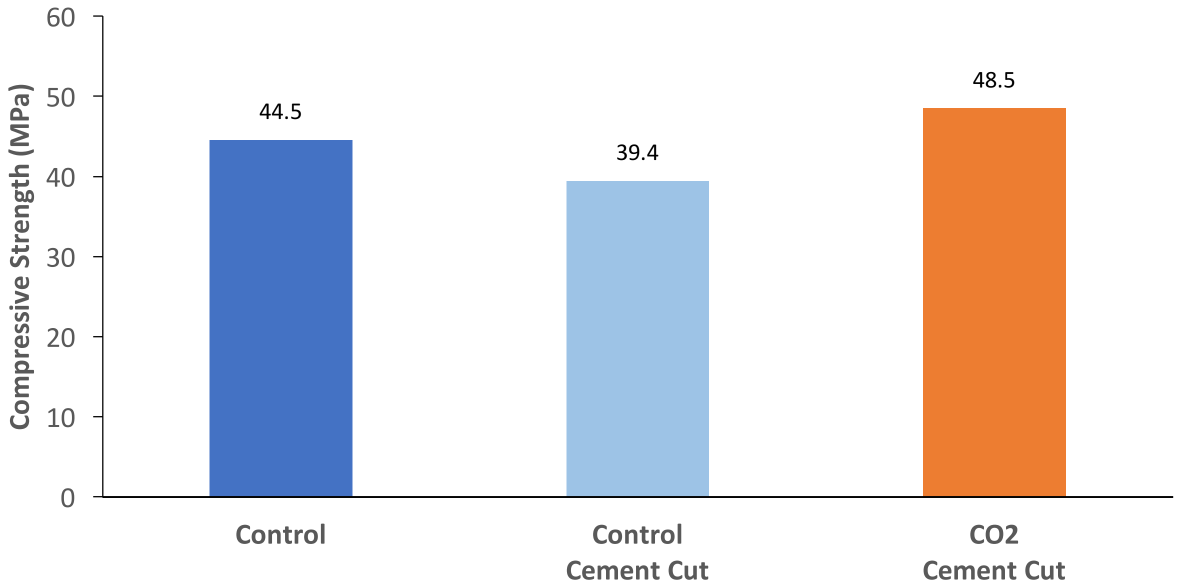

Figure 3. Ready Mixed Concrete Production – 28 day compressive strength of control batch, batch with reduced cement, and batch with reduced cement produced using CO_2

A concrete producer can pursue the carbon dioxide utilization with three goals in mind.

- Improve the concrete performance – an increase in early strength development can lead to increased use of slag and fly ash that would otherwise contribute to lower early strength. Increased proportioning of these materials is often pursued to improve concrete durability.
- Improve the concrete economics – a strength benefit can be the basis to reduce the overall amount of binder. If there is a particular component (e.g. cement or slag) that is the most expensive then its reduction can be prioritized.
- Improve the concrete environmental characteristics - A reduction in the cement usage will further result in avoided CO_2 emissions. The approach can lead to a lower carbon footprint for the mix design.

The cement reduction has a net environmental impact on the process. The implementation of the technology would result in some CO_2 emissions associated with capture and transportation of the CO_2. These emissions are sensitive to the electrical grid emissions where the capture takes place and the distance of transport but are on the order of about 14% of the CO_2 (Monkman and MacDonald 2016). A dose of 0.1% CO_2 by weight of cement required 342 g per cubic meter of concrete. The emissions to implement the technology totaled about 48 g of CO_2/m^3 concrete.

The cement reduction results in avoided CO_2 emissions that are far greater than the process emissions. Generic specific emissions for cement are 0.927 tonnes CO_2/tonne finished cement) (Athena Sustainable Materials Institute 2016). The 5% reduction of cement amounted to 18 kg/m^3 and resulted in an avoided carbon dioxide emission of 16.7 kg/m^3 concrete. The US National Average carbon footprint for a 35 MPa mix design is 485 kg $CO2_e/m^3$ concrete (Athena Sustainable Materials Institute 2016). A reduction in the carbon footprint by 16.7 kg CO_2/m^3 concrete would represent a 3.4% decrease.

Concrete Wash Water

Research Outline The impact of wash water on concrete properties is largely linked to the specific gravity and age of the wash water. The specific gravity is generally used to describe the amount of solid materials contained in the water; the recommended limit for wash water when it is reused as mix water is to remain below a solids content of 50,000 ppm (an approximate specific gravity of 1.03). The suspended solids are the binder phase from the concrete.

As the specific gravity of the water increases, the impact of the wash water on fresh properties worsens. In an operational environment, the impacts of specific gravity make for a "moving target" for QA/QC personnel when trying to counteract the negative properties associated with wash water reuse, imposing a practical limit on the extent that wash water can be successfully reused. Conventional practices involve clarification of the water for safe disposal, and periodic landfilling of cementitious sludge.

Samples of concrete wash water were simulated by adding 23 g of cement, 14 g slag, and 9 g of fly ash to 268 g of water. In all cases the wash water was prepared at a specific gravity of 1.10 (more than triple the recommended solids limit) and allowed to age for 1 day. The wash water was prepared in a batch format in bottles containing enough water to produce one batch of mortar. The suspended solids comprised 50% cement, 30% slag and 20% class F fly ash to simulate the mix of components that might appear in a concrete production environment. The wash water samples were either untreated or included a CO_2 treatment. The CO_2 treatment comprised vigorous mixing and aging of the wash water under a CO_2 atmosphere. Typically, the exposure to CO_2 was initiated in the timeframe of 30 to 120 minutes after preparation of the wash water and continued until the water was used for mortar preparation. The treatment would result in carbon dioxide uptake on the order of 10 to 40% by weight of cement.

The lab-produced wash water was used in the preparation of mortar by combining the required mix water with 535 g of Type I/II cement and 1350 g of EN 196-1 sand. No adjustment was made to compensate for change in mix volume due to the addition of powder to the mix water, rather the total amount of potable mix water was held constant in each test. Mortar samples were assessed in terms of set time (ASTM C403), workability and compressive strength. The testing compared samples produced potable water, untreated wash water and CO_2 treated wash water.

Beneficiated Concrete Wash Water Results The test data for the mortar samples is presented in Table 1. The use of the untreated wash water resulted in unacceptably large reduction in the time of initial set (defined as exceeding 60 minutes as outlined in the ASTM C1602 Standard Specification for Mixing Water Used in the Production of Hydraulic Cement Concrete). The carbon dioxide treatment completely eliminated the set time acceleration. The mortar made with wash water showed a lower workability (slump) than for mortar made with potable water. This is due to effectively lowering the water to cement ratio when considering the impact of the suspended solids on the binder system. The

Table 1. Beneficiated Wash Water Results

Property	Control	Untreated Water	CO_2 Treated
Set Time (min)	388	306	412
Slump (mm)	108	80	50
Strength (MPa) – 1 day	14.4	21.2	15.3
Strength (MPa) – 7 days	38.0	45.8	46.6
Strength (MPa) – 28 days	44.5	57.3	61.6

CO_2 treatment did not improve the workability but for both wash water mortars it is feasible that the workability could be easily manipulated through conventional methods such as admixtures.

Compressive strength tests were performed in duplicate on 2″ × 2″ × 2″ mortar cubes at 1, 7 and 28 days after mixing. The use of untreated wash water lead to higher strengths than using potable water. Likewise, the samples made with the carbon dioxide treated wash water were stronger than samples made with the potable water at all ages. The strength of the treated wash water samples was behind the untreated wash water case at 1 day, equivalent at 7 days and ahead at 28 days. The CO_2 treatment has acted on the cement in the wash water thereby reducing the early cementitious contribution that the suspended solids can provide. However, the later strength contribution suggests that the CO_2 treatment provided an additional improvement above and beyond the inherent cementitious properties of the additional binder.

Both the fresh property and strength results suggest that the optimized use of untreated wash water could support a reduction in the binder loading of concrete made therewith (maintaining the water to binder ratio in the former case, taking advantage of latent cementitious capacity in the latter case). However, an undesirable set acceleration would still be an issue. The carbon dioxide treatment could allow the beneficial reuse of high solids wash water without the undesired set acceleration. The concept additionally realizes the beneficial use of carbon dioxide and the reduction in waste outputs (clarified waste water and waste water sludge).

The implementation of a wash water beneficiation technology would have a positive impact on fresh water consumption. An average cubic meter of concrete requires 129 L of batching water and produces 118 L of wash water (average US national values from (Athena Sustainable Materials Institute 2016)). If 100% of the wash water could be reused as batch water then, for a given mix, the amount of fresh water used in batching the concrete could be reduced by 92%. Further benefits may arise if the potential strength benefit is leverage to reduce the cement loading in the mix design.

CEMENT PLANT CO_2 – THE SUSTAINABLE SOLUTION

The ideal source of carbon dioxide for a CO_2 utilization technology in concrete production is from the emissions stream of a cement plant. This approach would be the ideal representation of the circular economy within the cement and concrete industry. The carbon impact of the cement and concrete industry has been the subject of increasing attention. The production of cement is responsible for 5.6% of carbon emissions from fossil fuel and industry and is the largest industrial emitter (Le Quéré et al. 2016). If the industry's waste carbon dioxide could be repurposed to make better, cheaper and/or lower carbon concrete then the opportunity is not only enticing, but obvious and strategically adroit.

Industrially-sourced carbon dioxide is typically the byproduct of an industrial process (examples include hydrogen plants, fertilizer plants, titanium pigment processes, ethanol production). The concept of capturing CO_2 from cement plant flue gas is not yet practiced. An assessment of industry readiness for cement kiln carbon dioxide capture was summarized in a 2016 journal article (Hills et al. 2016). Five promising carbon capture technologies were compared but none of them are projected to be widely available for the next 10 to 25 years. While these technologies would aim to capture the majority, if not the entirety, of the carbon emissions from a cement plant, the reality is that a smaller scale approach may be appropriate to service the CO_2 utilization needs for concrete production applications.

A masonry producer making 4 million blocks per year would use about 140 tonnes of carbon dioxide. A ready mix concrete producer of medium size (50,000 m^3/year) would utilize about 25 tonnes of carbon dioxide. A wash water application treating the wash water from a 50,000 m^3/year plant would utilize about 225 tonnes. It is useful to consider this in the context of one cement plant.

In 2015, the United States had 99 cement plants with a total output of 75.7 million tonnes of cement (U.S. Geological Survey 2016). With an average output of 764,000 tonnes cement and a generic emissions rate of a 927 kg CO_2/tonne of finished cement then each cement plant emitted about 708,000 tonnes of CO_2.

There are an estimated 5,550 ready mixed concrete plants in the United States (NRMCA 2016). Therefore, each cement plant serves about 55 concrete producers. If a single cement plant could find, among their existing customers, 20 adopters for each of the masonry, ready mix concrete and wash water technologies then the required CO_2 would be about 7,800 tonnes per year.

The rollout of the CO_2 utilization technologies around a generic cement plant would require the capture of about 1.1% of the plant's emissions. Whereas fully integrated capture technologies would measure their success by addressing a large fraction of a cement plant's emitted carbon dioxide the actual requirements for the utilization technologies could be served by a small capture system with a lower capital cost.

CONCLUSIONS

Concrete is the world's most widely consumed construction material. Producers have successfully adopted circular economy principles in the past through the reuse of industrial by-products. Recent research directions have established both the reality and possibility of adding cement industry carbon dioxide emissions to the beneficially reused wastes in concrete.

A masonry technology has been installed at 18 locations that allows concrete block producers to inject carbon dioxide into their blocks. The CO_2 can potentially improve the compressive strength of the blocks while the absorbed CO_2 serves to reduce the carbon footprint of the block.

Industrial scale integration of a carbon dioxide injection into ready mixed concrete has demonstrated the ability to leverage CO_2 as a new tool in mix design optimization. The combination of the strength enhancing properties of an optimized dose of carbon dioxide and reduced binder loadings allows a concrete producer to achieve equivalent 28-day compressive strength performance with a reduced environmental footprint. Systems have been installed with 13 producers.

The use of carbon dioxide to treat concrete wash water can mitigate or reduce some of the problems associated with reusing wash water as concrete mix water. The acceleration of the time to initial set is eliminated. The carbon dioxide treatment potentially allows the use of concrete wash water with higher solids contents than can typically be achieved thereby reducing the disposal of waste water sludge, waste water, and beneficially absorbing carbon dioxide. If the beneficiated wash water can be used to replace fresh water used for batching, then a significant savings of fresh water is feasible.

Sourcing CO_2 from cement plants is not yet feasible insofar as full scale capture is concerned. However, for beneficial CO_2 utilization it is likely that a small-scale slipstream capture technology can be employed to meet the carbon dioxide needs. Cement producers would then able to able to put their waste CO_2 to beneficial use in concrete thereby upcycling a portion of their primary waste product and using resources in a manner consistent with circular economy principles.

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