EXECUTIVE SUMMARY

Concrete is the world’s most important and widely used construction material. Carbon dioxide utilization in the production of ready mixed concrete was investigated through the injection of an optimal amount of CO$_2$ during batching and mixing. The carbon dioxide improved the concrete compressive strength with minimal impact on fresh air content or workability. Three-way comparisons between a reference batch, reduced binder batch and reduced binder batch with CO$_2$ addition, confirmed that the carbon dioxide could allow for a 5-8% reduction in binder loading without compromising strength. A model case shows that integrating a CO$_2$ utilization step into conventional concrete production can, net of process emissions, reduce the carbon footprint of the concrete by 4.6%. The direct utilization is amplified to attain a carbon footprint improvement that is more than 35 times larger than the amount of carbon dioxide required. One year production at a medium sized producer would use about 24 tonnes of carbon dioxide to achieve nearly 897 tonnes of CO$_2$ absorbed and avoided.

Keywords: concrete, carbon footprint, construction materials, environmental impact; carbon dioxide utilization
1. INTRODUCTION

Concrete is the world’s most widely used material. Annual cement production has surpassed an estimated 4.6 Gt (CEMBUREAU, 2016). It is a versatile and economical building material and demand is ever increasing (Scrivener, 2014). Cement production in 1950 amounted to 133 million tonnes (Fig. 1) thereby creating about 0.4 tonnes of concrete per person. By 2015, global cement production had seen a nearly 35-fold increase while the per capita concrete production had increased more than 12-fold. Considering that a generic concrete mix contains about 300 kg cement per cubic meter and global population exceeds 7.4 billion (Population Reference Bureau (PRB), 2016), it is evident that the annual global production of concrete is currently around 2.1 m³ per person. At a density of 2.3 tonnes per cubic meter there are about 4.8 tonnes of concrete produced annually for each person.

The carbon impact of the cement and concrete industry has been the subject of increasing attention. The production of cement involves the calcination of limestone (CaCO₃) to create reactive calcium silicates; carbon dioxide is a by-product (Barcelo et al., 2014). Cement production is responsible for 5.6% of emissions from fossil fuel and industry and is the largest industrial emitter (Le Quéré et al., 2016). The industry is facing a challenge to meet demand yet address carbon emissions. The cement and concrete industry worked with the International Energy Agency to outline the ambitious effort that would be required to reduce industry emissions to 50% below 2006 levels by 2050 (IEA, 2009); the target is consistent with the “blue map scenario” (International Energy Agency, 2008) wherein atmospheric CO₂ is limited to a level commensurate with atmospheric warming of þ3 C (IPCC, 2007). It was concluded that a projected 0.79 Gt of CO₂ reduction from the BAU baseline 2050 emissions needed to be achieved and would be addressable by four approaches:

1. Reducing CO₂ emissions for the manufacture of Portland based cements through the increased use of alternative fuels and/or alternative raw materials (potentially 24% of the required reduction)
2. Improving the energy efficiency of cement kilns (10%)
3. Increasing clinker substitution through the increased use of low-carbon supplementary cementitious materials (SCMs) (10%)
4. Capture and sequestration (CCS) of the carbon dioxide emissions released from cement plants (56%)

Traditional levers to reduce the carbon footprint of concrete (approaches 1 to 3) have practical limits whether it is the finite supply of viable supplementary cementitious material (SCMs) (Scrivener, 2014) or the realistic understanding of the potential to reduce the energy required to produce cement (Madlool et al., 2013). As a result, the greatest proportion of the projected carbon reduction depends upon the implementation of carbon sequestration technologies that are as-yet undefined solutions developed outside the industry. The time that has elapsed since the IEA roadmap was published in 2009 has allowed thinking to evolve and finds that, arguably, CCS is no longer the most promising technology for the reduction of CO₂ emissions related to cement based materials (Scrivener et al., 2016). Research has demonstrated that CO₂ utilization concepts can contribute to the creation of lower carbon concrete products (Ashraf, 2016; Jang et al., 2016). Methods rooted in CCS-style approaches focus upon maximizing the amount of CO₂ that can be sequestered and stored within useful building products. The necessity to contain the supplied CO₂ gas during the reaction has meant that efforts have largely focused on precast and/or masonry concrete applications wherein closed curing is feasible (El-Hassan and Shao, 2015; Wang et al., 2017; Zhan et al., 2016). Further applications have focused on the development of building products produced using carbon dioxide activated binder systems (Ashraf and Olek, 2016; Mahoutian and Shao, 2016; Vance et al., 2015), partly due to the limited amount of CO₂ that Portland cement can absorb. The potential for such applications to achieve large sustainability improvements within the niche
of addressable compatible concrete is promising, but carbon dioxide utilization solutions are required for the ready mixed concrete market segment, which consumes about 70% of the cement produced in the United States (U.S. Geological Survey, 2016) and is associated with 60% of concrete industry revenue.¹

Recent work has identified the potential for CO₂ to unlock a performance benefit in cast-in-place concrete without impacting durability (Monkman et al., 2016). A carbon dioxide utilization method that can be integrated as a retrofit into typical concrete production and use normal Portland cement presents an attractive route to lessen the environmental impact of concrete while upcycling one industry waste (carbon dioxide) with the main industry output (ready mixed concrete).

The objective of this research was to test the hypothesis that the carbon dioxide utilization could improve the compressive strength of concrete so-produced and whether said improvement could be leveraged to improve the carbon footprint of the concrete and produce a more sustainable concrete mix. The work involved a retrofit CO₂ injection system installed at a ready mix concrete producer. Carbon dioxide was injected into the concrete while it was being batched and mixed. A model examines the potential process benefits and the net impact on the carbon footprint of concrete so-produced.

2. MATERIALS AND EXPERIMENTAL

A carbon dioxide utilization approach for ready mix concrete production was designed to be implementable as a retrofit technology. If the concept is shown to be technically viable through the realization of performance benefits and improvements in concrete sustainability then successful integration must respect incumbent practices and conform with existing equipment, sequences and process. A mixer injection approach demonstrated for masonry block production (Monkman and MacDonald, 2016) established a template for a central CO₂ injection approach for ready mix concrete.

Concrete was produced whereby carbon dioxide was injected during batching. A metering system fed a controlled supply of pressurized liquid CO₂ through to a discharge conduit. The liquid was converted into a mixture of CO₂ gas and finely divided solid carbon dioxide particles (commonly referred to as CO₂ “snow”) once it reached the atmosphere upon discharge (liquid carbon dioxide is not stable at atmospheric temperature and pressure; the phase transition is spontaneous upon depressurization of the liquid). 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. CO₂ was injected directly into the central mixer prior to the discharge of the concrete into the truck. The carbon dioxide rapidly reacts with calcium ions, produced by the hydrating cement, to form calcium carbonate. The reacted CO₂ is chemically bound in the concrete as a solid phase; no gaseous carbon dioxide persists in the concrete.

The concrete was then subjected to assessment and testing. Industrially produced concrete was tested in the fresh state in terms of slump (ASTM C143 Standard Test Method for Slump of Hydraulic-Cement Concrete), air content (ASTM C231 Standard Test Method for Air Content of Freshly Mixed Concrete by the Pressure Method), unit weight and temperature. Hardened concrete cylinders were assessed in terms of compressive strength (ASTM C39 Standard Test Method for Compressive Strength of Cylindrical Concrete Specimens) at various ages and test conditions.

Preliminary proof-of-concept testing established that the addition of the carbon dioxide could produce a compressive strength benefit. The present investigation paired the strength-boosting CO₂ injection with a mix design optimization wherein the concrete mix was redesigned to have a reduced binder content. The intention was to produce a concrete with the same target strength but with a reduced carbon footprint. The cement reduction testing considered five cases across which comparisons could be made with both historical data (quality control data provided by the producer) and reference data produced at the same time. The dose of CO₂ varied slightly from batch to batch but can be generalized as 0.15% by weight of cement. The five cases were:

¹ The concrete industry can be divided into three segments. An expected total 2016 US revenue of $44.5B can be divided according to $26.9B for ready mix (Ulama, 2016), $10.6B for precast (Masterson, 2016a) and $7.0B for concrete pipe and block (Masterson, 2016b).
1. A three-way comparison using mix 30RT - a 3000 psi (21 MPa) residential mix design with a ternary blend of 50% cement, 25% slag, 25% class F fly ash. Batches representing a control (reference), reduced binder, and reduced binder + CO2 were compared both with and without air entrainment. The nominal binder adjustments were a reduction of 7% for the non-air entrained comparison, and 8% for the air entrained comparison.

2. Extended production using Mix 30RT wherein batches using CO2 included a cement reduction of about 5.7% or 11 kg/m^3. The binder further had a 1.4% increase in class F fly ash and 7.2% reduction in slag for an overall binder reduction of 4.3%.

3. Mix 30CF - a 3000 psi (21 MPa) general commercial use non-air-entrained fly ash mix, with a total binder loading of 320 kg/m^3 comprised of 74% cement and 26% class F fly ash. The modified batch used an average cement reduction of 4.5% or 14 kg/m^3. The fly ash loading was not changed. The reduction as a proportion of the total binder reduction was 3.5%.

4. Mix 50HE - a 5000 psi (35 MPa) non-air entrained high early strength mix design with a total binder loading of 419 kg/m^3. The entirety of the binder was Type III cement. The modified batch used an average cement reduction of 6.7% or 28 kg/m^3.

5. Mix 80T - an 8000 psi (55 MPa) structural mix design, with a total binder loading of 575 kg/m^3 comprised of 52% cement, 12% class F fly ash and 36% slag. The modified batch used an average cement reduction of 6.0% or 18 kg/m^3. The fly ash and slag loadings were not changed.

The binder reductions were accompanied by small adjustments of fine aggregate loadings to ensure that yield was maintained.

2. MATERIALS AND EXPERIMENTAL

3.1. Three way comparison - mix 30RT

The binder adjustments for the batches produced in this study are detailed in Table 1. The modified mix design for the non-air entrained concrete reduced the overall binder by 7% via a 3% reduction in cement and a 19% reduction in slag. The modified mix design for the air entrained concrete used an overall binder reduction of 8% with the cement reduced 12%, slag reduced 21% and fly ash increased 18%.

An overview of the fresh properties for the loads produced during the three-way comparison is presented in Table 2. In general, the fresh properties were found to be comparable and within the range of normal batch-to-batch variation. No adjustments in mix water volume, admixture dose or batching process were made to accommodate the modified binder loadings, nor in response to the use of CO2. Thus, in addition to a reduction in paste volume, the reduced binder mixes were observed to have slightly increased water to cementitious ratios and admix loadings per unit of binder. The former is expected to have a negative impact on strength development while the latter is expected to have a neutral impact.

The average compressive strength measured for each non-air entrained batch at three test ages is summarized in Fig. 2. The binder modification lead...
to a 17% drop in 28 day compressive strength. The strength of the reduced binder batch 18% at 1 day, 17% at 7 days and 16% at 28 days. Ultimately the strength of the batch with CO2 and the 7% reduced binder content was within 4% of the reference at 28 days. The trial represented the first attempt at an optimized mix design and it was concluded that further tweaks to the binder loading and/ or CO2 dose should establish that the CO2 addition can achieve at least equivalent performance at all ages.

The average compressive strength measured for each air entrained batch at three test ages is summarized in Fig. 3. The binder reduction in the air entrained batch resulted in an 11-13% drop in compressive strength. The addition of the carbon dioxide improved the strength of the reduced binder concrete by 15% at 1 day, 10% at 7 days and 13% at 28 days. The strength of the batch with 8% less binder and the CO2 addition was equivalent to the reference at all three test ages.

A useful assessment of the mix design modifications can be developed using the concept of binder intensity and CO2 intensity (Damineli et al., 2010). These metrics allow broad comparisons to be made between mix designs in terms of their functional and environmental performance. For the non air entrained batches the binder intensity relevant to compressive strength (bics) for the reference condition was 10.2 kg binder m-3 MPa-1, 11.4 for the reduced cement batch and 9.9 for the batch produced with CO2. The binder modification initially increased the bics by 11% but the addition of the carbon dioxide however, the addition of carbon dioxide improved resulted in a net 9% decrease. With the air entrained batches the three calculated indices were 11.4, 11.9 and 10.4 kg binder m-3 MPa-1. The binder modification increased the factor slightly (4%) whereas the addition of CO2 decreased the factor by 9%.

An approximate CO2 intensity calculation uses an emission factor of 915 kg CO2/tonne of cement (as communicated by the cement supplier) and no emissions associated with the SCMs. For the non air entrained batches the carbon intensity (ci) is 4.5, 5.2 and 4.5 kg CO2 m3 MPa-1 for the reference, cement cut and cement cut with CO2 batches respectively. The ci increased 16% due to the cement cut, but the carbon dioxide restored it to be equivalent to the baseline. For the air entrained batches the carbon intensities for the three conditions were 5.4, 5.4 and 4.8 kg CO2 m-3 MPa-1. The cement cut had no impact but the addition of CO2 caused a 12% reduction.

The results of the three way comparison tests prompted subsequent mix design modifications to be made with only a two way comparison – reference control mix and modified mix design that included the CO2 injection.

3.1.1. Production case - mix 30RT

A production run was conducted employing mix 30RT. Ten batches were produced during the run; eight batches were made using the CO2 injection alongside two complementary control batches. The average slump was 145 mm for the reference data
(total 9 batches including two created within the same production, standard deviation 19 mm, coefficient of variation 13%) and 165 mm for the CO₂ batches. The compressive strength was tested at 7 and 28 days and compared against the reference data. A chart presents the average values for each condition at each age with error bars indicating the overall range of the collected data (Fig. 4). The control mix design averaged 16.4 MPa at 7 days (and ranged between a minimum of 12.2 MPa and a maximum 21.2 MPa, standard deviation 3.5 MPa, coefficient of variation 21%). The eight batches produced with the 5.7% reduced binder loading and a CO₂ addition averaged 15.5 MPa (ranging between a minimum of 11.9 MPa and maximum 17.3 MPa psi). At 28 days the historical performance was an average of 27.1 MPa, with a minimum of 21.6 MPa and maximum of 32.6 MPa (standard deviation 3.5 MPa, coefficient of variation 13%). The CO₂ production data was observed to average 27.9 MPa, and ranged between 22.1 and 31.1 MPa.

The average strength of the CO₂ treated batches with reduced binder was 95% of the typical historical strength at 7 days, and 103% of the typical historical strength at 28 days. The production variation was comparable to what was observed with the regular production data. The bi₃ and ci were reduced by 7% (from 11.5 to 10.6 kg binder m⁻³ MPa⁻¹) and 9% (from 5.3 to 4.8 kg CO₂ m⁻³ MPa⁻¹) respectively.

3.1.2. Production case - mix 30CF

A production using Mix 30CF created four CO₂ treated batches with reduced binder. The 7 and 28 day compressive strength results are plotted in Fig. 5 with the control data comprising the performance of 31 historical reference batches (including the one batch created within the same production). The unaltered mix design typically averaged 19.7 MPa at 7 days (ranging between 15.0 MPa and 25.2 MPa, standard deviation 2.7 MPa and coefficient of variation 13%). The four batches produced with the reduced binder loading and a CO₂ addition averaged 19.1 MPa (minimum 16.7 MPa and maximum 23.5 MPa). At 28 days the historical performance was an average of 28.3 MPa, with tests as low as 23.7 MPa and as high as 34.6 MPa, standard deviation 3.0 MPa and coefficient of variation 11%. The CO₂ production data was observed to average 26.1 MPa, and range between 23.7 and 31.1 MPa. The binder and carbon intensity indices slightly increased. The bi increased 5% (11.3 to 11.9 kg binder m⁻³ MPa⁻¹) and the ci increased 3% (7.6 to 7.9 kg CO₂ m⁻³ MPa⁻¹).

The average strength of the reduced binder CO₂ batches was slightly lower than the historical averages (within 3% at 7 days and 8% at 28 days) but the overall variation was consistent with performance of the unmodified mix. The acceptable production of batches using the CO₂ and a reduced binder loading was assured.
3.1.3. Production case - mix 50HE

The compressive strength results suggested compressive strengths that clearly exceeded the historical performance (Fig. 6). The historical data set comprised 30 batches. The baseline performance was an average strength of 36.5 MPa, standard deviation of 3.1 MPa, and coefficient of variation 8% at 7 days and an average strength average of 44.9 MPa, standard deviation of 2.1 MPa, and coefficient of variation 5% at 28 days. The strength of the CO2 batches averaged 39.0 MPa at 7 days (overall 7% increase). At 28 days the batches produced using CO2 were 8% stronger at 48.4 MPa. The increased strength accompanying a cement decrease of nearly 7% may indicate that the carbon dioxide has a particular synergy with the chemistry and/or high fineness of the Type III cement. Both the binder and carbon intensity indices decreased 22%. The $\text{bi}_{\text{c}}$ decreased from 9.9 to 7.8 kg binder m$^{-3}$ MPa$^{-1}$ while the $\text{ci}$ decreased from 9.1 to 7.1 kg CO$_2$ m$^{-3}$ MPa$^{-1}$.

3.1.4. Production case - mix 80T

The compressive strength of the batches using CO$_2$ compared favourably to the historical data (Fig. 7). The historical data set comprised 30 batches. The baseline performance was an average strength of 52.1 MPa, standard deviation of 6.0 MPa, and coefficient of variation 12% at 7 days and an average strength of 67.5 MPa, standard deviation of 6.5 MPa, and coefficient of variation 10% at 28 days. The strength of the CO$_2$ batches averaged 57.9 MPa at 7 days (overall 11% increase) and 73.9 MPa at 28 days (10% increase) albeit using 6% less cement.

The 80T mix had the lowest binder and carbon intensities at 8.5 kg binder m$^{-3}$ MPa$^{-1}$ and 4.0 kg CO$_2$ m$^{-3}$ MPa$^{-1}$ but the addition of the carbon dioxide allowed for a 12% reduction to the binder intensity (to 7.5) and 14% reduction to the carbon intensity (to 3.5).
4. DISCUSSION

4.1. Process benefits

The compressive strength benefit results confirmed two major outcomes.

1. A reduction in the binder loading contributed to a reduction in the compressive strength

2. The strength reduction could be offset through the introduction of an optimized dose of CO₂ into the concrete while it is batched and mixed

The conclusions were confirmed in three-way comparison testing and supported through extended production.

The carbon dioxide utilization can be a platform for further producer actions according to their specific motivation whether it be to improve the economic, performance, or environmental characteristics of the concrete so produced. A producer that is economically motivated may seek to reduce the overall amount of binder to save money. The reduction of the most expensive components can be prioritized. If performance benefits are the most important consideration, then an improvement in early strength development can allow for increased proportioning of slower-hydrating slag and/or fly ash and thereby target the durability benefits that may arise. If the motivation is environmental, then a strength boost that allows for a reduction in the cement usage will consequently lead to avoiding the associated CO₂ emissions and thereby result in a reduction in the carbon footprint of the mix.

4.2. Model environmental impact

The Sustainable Concrete Plant Guidelines (RMC Research and Education Foundation, 2011), published in 2011 in conjunction with the National Ready Mixed Concrete Association, outlined industry targets for improving the sustainability of ready mixed concrete. The stated goal to improve the carbon footprint of concrete, relative to 2007, was a 20% reduction by 2020, and a 30% reduction by 2030. In this context, the carbon footprint of concrete is the total amount of CO₂ emitted due to concrete production and encompasses raw material extraction, production, delivery to job sites, and eventual disposal or reuse. A 2016 industry survey concluded that the NRMCA benchmark carbon footprint for a generic 4000 psi (27.6 MPa) concrete was 393 kg CO₂e/m³ (Athena Sustainable Materials Institute, 2016). On average, the cement used in a concrete mix represents over 85% of the embodied energy and up to 96% of greenhouse gas (GHG) emissions per unit volume of concrete produced (Marceau et al., 2007). Thus, the most important lever to reducing the carbon footprint of concrete will be to reduce the carbon contribution of the cement. The impact of the described approach to improve the sustainability characteristics of concrete can be assessed through an environmental analysis.

The calculations are considered in terms of one m³ of concrete. The baseline process considers the raw materials required to make the concrete, the operational inputs to produce concrete, and greenhouse gas outputs associated with transporting materials, using electricity, and burning fuels. A generic model concrete mix can be based upon benchmark data compiled by the National Ready Mixed Concrete with a target compressive strength...
of 4000 psi (27.6 MPa) a baseline cement loading of 570 lb/yd³ (3.38 kg/m³). Additional process elements are included when the concrete is produced with the addition of carbon dioxide: the capture and liquefaction of the carbon dioxide, the carbon dioxide transport, and the production and operation of the carbon dioxide injection hardware. Additionally, a model 5% cement reduction is achieved (a reduction of 17 kg to 321 kg/m³). The environmental footprint of the process considers:

1. The baseline CO₂ emissions related to conventional concrete production
2. The net impact of mix design changes on CO₂ emissions
3. CO₂ emitted during the capture and compression of the CO₂
4. CO₂ emitted during the transportation of the CO₂
5. CO₂ emitted during the production of the gas injection equipment
6. CO₂ emitted during the transportation of the gas equipment
7. CO₂ absorbed through the utilization step
8. CO₂ emissions avoided due to the reduction in the cement content

The calculations are reported in terms of creating one cubic meter of concrete. The CO₂ dosage used in the model is 0.15% by weight of cement or 482 g CO₂ per cubic meter of generic concrete.

4.2.1. Gas production and gas transport impacts

The industrial gas processing to create liquid CO₂ from an emissions stream requires on the order of 200 kWh/tonne CO₂ (Haring, 2008) with emissions that are dependent upon the electrical grid emissions rate at the location where the work is performed. The 2014 average CO₂ emission rate for electrical power produced in the United States was 1130.2 lb CO₂/MWh (512.6 g CO₂/kWh) (US Environmental Protection Agency, 2017). Thus, the industrial processing to produce a tonne of liquid CO₂ would involve the emissions of 102.5 kg of carbon dioxide.

Given a carbon dioxide dose of 482 g/m³ concrete, the CO₂ emissions associated with gas processing are estimated to be 49.4 g/m³ concrete. This compares favourably to the GHG emissions impact of producing a typical plasticizing concrete admixture; 1.88 kg CO₂ per kg of admixture produced (European Federation of Concrete Admixtures Associations Ltd. (EFCA), 2015) or 160 g CO₂/m³ concrete according to the generic loading of 3 oz/m³ concrete reported in the NRMCA benchmark data.

The carbon dioxide would be transported from the industrial source to the concrete producer. Transport emissions can be modeled using an emissions rate of 1.430 kg CO₂/vehicle-mile of freight using medium to heavy duty trucks (EPA Center for CEMBUREAU, 2016) and a single bulk load mass of 25 tons (22.7 tonnes). The emissions are then 0.063 kg CO₂/tonne-mile. If a roundtrip transport distance of 200 miles (322 km) is used then the gas transport emissions are calculated to be 6.1 g CO₂/m³ concrete produced.

4.2.2. Equipment production, transport and operation impacts

The gas injection equipment is comprised of steel, brass and plastic. Considering the amounts used and CO₂ emissions factors associated with the production of these materials (calculated as generic examples of OHF steel, generic brass, and polypropylene plastic) results in an estimated CO₂ emission of 80.7 kg (Table 3). The transportation of the equipment is estimated as the truck freight transport of 61.2 kg (equipment totaling 43.1 kg and contained within a crate of 18.1 kg) over a generic single trip distance of 1250 miles. The transport emissions would be a partial load shipment with an emissions rate of 146 g CO₂/tonne-mile (EPA Center for CEMBUREAU, 2016). The transport emissions associated with the delivery of one gas injection system total 12.3 kg CO₂.

Table 3

<table>
<thead>
<tr>
<th>Component</th>
<th>Amount (kg)</th>
<th>Emissions Factor (kg CO₂/kg)</th>
<th>Emissions (kg CO₂)</th>
</tr>
</thead>
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<tr>
<td>Steel</td>
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<td>1.72 (PCIC, 2016)</td>
<td>5.46</td>
</tr>
<tr>
<td>Brass</td>
<td>2.3</td>
<td>4.66 (Mathis, 2014)</td>
<td>10.6</td>
</tr>
<tr>
<td>Plastic</td>
<td>9.1</td>
<td>1.71 (EPA, 2015)</td>
<td>15.5</td>
</tr>
<tr>
<td>Total</td>
<td>43.1</td>
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<td>80.7</td>
</tr>
</tbody>
</table>

If the total equipment production and transport emissions (93.0 kg CO₂) are amortized over a 20-year operational life and an annual production of 50,000 m³ concrete, then the associated emissions are 0.09 g of CO₂ per m³ concrete (comprising 0.08 g from the production and 0.01 from the transport). The power demand of the hardware has been estimated to be 0.037 kWh/kg CO₂ injected. For the carbon dioxide dosed into a cubic meter of concrete the corresponding power consumption is 0.018 kWh. According to the generic carbon intensity for power generation there is an anticipated 9.2 g CO₂ emitted. The overall emission for the production, transport and operation of the equipment is 9.3 g CO₂/m³ concrete.
4.2.3. Direct CO₂ absorption

Direct quantification of the absorbed carbon dioxide is difficult (the dose of 482 g is applied to concrete with a nominal density of 2300 kg/m³ which is equivalent to an abundance on the order of about 200 ppm). The carbon dioxide applied to the concrete is about 50% solid and 50% gas. If the solid fraction, directly observed to adhere to the wet concrete, is incorporated into the concrete with a high efficiency (say 90%) while the gas, which is heavier than air but otherwise above the mixing concrete, is incorporated at a low efficiency (say 30%) then the combined overall absorption efficiency can be estimated at 60%. The 482 g total dosage/m³ concrete absorbed at a rate of 60% would result in about 289 g of CO₂ being fixed. This would mean an estimated 2.3 kg of CO₂ are absorbed in an 8 m³ truck load of concrete, and 14.4 tonnes over an annual production of 50,000 m³ concrete.

4.2.4. Changes to material flows

The addition of the carbon dioxide allows for a reduction in the cement loading in the concrete. The cement in turn has a carbon impact that is directly avoided both through the material reduction and the associated transportation that is not required. Additionally, the fine aggregate (sand) loading in the mix design may be increased to compensate for the volume of the removed cement. If the specific gravities of cement and sand are taken to be 3.15 and 2.61 respectively, then for a given unit mass of cement removed then the equivalent volume would be filled by 0.85 units of sand.

The 5% cement reduction means that 16.9 kg of cement are removed per m³ of concrete. Conversely, the sand would be increased by 14.0 kg. This would be a relative sand increase of 1.8% in the model mix design.

The transportation distances of the raw materials can be used to model a total GHG impact for materials transport. The NRCMA reports average values for distances between the suppliers of each of the mix components and the concrete producer (Athena Sustainable Materials Institute, 2016). Applying the previously identified emissions rate for bulk freight transport can determine the transport GHG impact for the components of the baseline (Table 4) and modified (Table 5) mix designs. It was calculated that the modified mix design would result in a net reduction in transport emissions of 124 g CO₂e/m³ concrete, or a 1.6% reduction. The plant operations are not anticipated to change in response to the mix design modification. The inputs of fuel and electricity, and the output of concrete wash water are unchanged.

<table>
<thead>
<tr>
<th>Component</th>
<th>Distance (km)</th>
<th>Freight (kg)</th>
<th>Emissions (g CO₂)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cement</td>
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<tr>
<td>Fly Ash</td>
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<tr>
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<td>15</td>
<td>61</td>
</tr>
<tr>
<td>Course Aggregate</td>
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<tr>
<td>Fine Aggregate</td>
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<td>1899</td>
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4.2.5. Overall process flow

A process flow diagram (Fig. 8) outlines the overall inputs and outputs of concrete production using the carbon dioxide injection approach. Compared to the baseline approach there is the addition of the CO₂ capture and utilization portion, a reduction in the cement usage (17 kg), an increase in the sand usage (14 kg) and a reduction in the overall material transport emissions (124 g CO₂).

4.2.6. Total net process impacts

The environmental impacts are summarized in Table 6. The total process emissions (CO₂ processing, CO₂ transport, equipment production, equipment transport and equipment operation) are estimated to be 64.7 g CO₂/m³ concrete. This equates to 22% of the modeled absorbed CO₂. By this metric an estimated 78% of the modeled absorbed CO₂ would represent a net storage of CO₂.

Industrially-sourced CO₂ is typically the byproduct of an industrial process. Consequently, CO₂ captured from such a source would have been atmospheric carbon emissions if not for having been captured and transported to address a market demand in an economically viable fashion. The environmental analysis considers that any CO₂ leakage at the injection site does not represent a net increase of CO₂.

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in the atmosphere since the injected carbon dioxide is comprised of industrial process emissions that were displaced prior to being utilized/absorbed/lost.

### 4.2.7. Impact model - sensitivity analysis

A sensitivity analysis is presented in Fig. 9 to consider the impact of changes to different inputs. The analysis considers factors that may vary with location and factors associated with technologies employed: electrical grid carbon intensity, the energy to capture CO₂, the transport distance of the CO₂, and the energy to operate the gas injection hardware. The analysis considers alternative cases for the energy use from electric grids (US National average of 513 kg CO₂/kWh), industrial gas energy requirements (200 kWh/tonne CO₂), CO₂ gas transport distances (100 miles one way), and injection hardware energy requirement (0.037 kWh/kg CO₂). The plot shows reasonable variation ranges for the various factors. The grid emissions are considered between two United States examples – the second highest (Wyoming) and second lowest (Idaho) carbon intensity power grids in the US (US Environmental Protection Agency, 2017). The gas processing energy requirement was changed ±25%, the gas transport distance was considered from 50% to 100%, while the hardware energy requirement was changed ±50%.

It is shown that the overall process emissions are most sensitive to the electrical grid CO₂ emissions associated with the CO₂ capture and processing. A 25% change in the electrical grid emissions results in a 23% change in the overall emissions. This factor can vary widely according to location. If the capture took place in the second highest carbon intensity US electrical grid then the overall emissions would increase 69% to 109.7 g CO₂/m³ concrete. Conversely, if in a location with the second lowest carbon intensity power grid, then the emissions would be 79% lower at 13.8 g CO₂/m³ concrete.

The process emissions were next most sensitive to the CO₂ capture energy; a 25% change in the gas capture energy resulted in a 19% change in the overall emissions. While considering an alternate electrical grid intensity is simple considering the breadth of grid emissions rates, proposing alternate gas capture strategies is less clear. The analysis is open to the possibility that a newer technique (e.g. membrane technology (Giordano et al., 2016) or cryogenic carbon capture (Jensen et al., 2015)) may achieve a sizably lower specific energy than the incumbent industrial gas processing approach.

Changing the CO₂ transport distance and injection hardware energy had the least effect with a 25% variation to each factor resulting in an impact to the
overall process emissions of ±3.4% and ±2.4% respectively.

4.2.8. Overall impact on carbon footprint

The strength increase produced by the small amount of CO₂ utilized can be leveraged into reductions in cement loading. The environmental benefit associated with using less cement is an order of magnitude greater than the calculated direct CO₂ absorption. A generic cement in the United States has an emissions intensity of 1040 kg CO₂/tonne of finished cement (Portland Cement Association, 2016). The cement reduction has a net environmental impact on the process given the avoided carbon dioxide emissions associated with cement production. There are 17.6 kg of CO₂ associated with the 16.9 kg of cement removed from each cubic meter of concrete. The overall net environmental impact would also include 289.1 g of CO₂ absorbed, 123.6 g of CO₂ avoided (materials transport) and 64.7 g of CO₂ emitted to result in a net CO₂ footprint reduction of 17.933 kg CO₂/m³ concrete. The avoided cement emissions would be responsible for 97.7% of the net environmental impact. The environmental impact of the cement removal is over 60 times the net direct CO₂ absorption. In comparison to the baseline carbon footprint of 393 kg CO₂/m³ concrete the overall impact would be a 4.6% reduction. A facility with an annual production of 50,000 m³ of concrete could see an annual direct CO₂ utilization of about 24 tonnes, of which about 14 tonnes would be absorbed and leveraged to achieve a total net absorbed and avoided CO₂ of 897 tonnes.

The small scale of the process emissions (64.7 g CO₂/m³ concrete) can be placed into context by comparing them to the environmental impact of a cement reduction. The process emissions would be offset by the avoided emissions for a cement reduction of 0.062 kg cement/m³ concrete or 0.02% of the baseline loading of 338 kg/m³.

If carbon accounting would conclude that the CO₂ that is dosed but unreacted represents emissions associated with the concrete plant rather than the original industrial emitter then the process emissions are increased by 193 g to 258 g CO₂/m³ concrete. (While the small amount of carbon dioxide absorbed suggests this the concept is not accurately positioned as a sequestration technology it is likely that the emissions would be counted this way if adhering to carbon accounting guidelines for geological storage approaches (IPCC, 2006)). The process becomes environmentally neutral if the cement removal is over 0.248 kg/m³ concrete, or 0.07% of the baseline cement loading.

The upper bound of the process emissions would occur in a case where both the absorption of CO₂ is minimal (i.e. assumed in calculation as 0% of the dose) and the carbon accounting classed the unabsorbed CO₂ as concrete plant emissions. The process emissions would increase to 547 g CO₂/m³ concrete. This is equivalent to the emissions from 526 g of cement and thus the process is carbon neutral once the cement reduction reaches 0.16% of the baseline loading. The relative scale of the process emissions to the impact of the cement loading means that only miniscule cement reductions are required to produce a net carbon benefit.
The equipment emissions of 93 kg CO₂ have been amortized over 20 years of production, but alternately can be considered as a one-time implementation penalty. The equipment emissions are equivalent to the CO₂ avoided through a cumulative cement reduction of 89 kg. This amount is less than the cumulative cement reduction observed after producing one truckload of concrete (8 m³ load with a 17 kg/m³ cement reduction). The implementation emissions are rapidly surpassed by the accumulating environmental benefit.

The required dosage of carbon dioxide is small and, thusly, so is the direct CO₂ absorption. However, the utilization approach can be leveraged to achieve the cement reduction. In a comparative sense, the dosage is about 3% of the net carbon impact. Alternatively, the utilization of one unit of CO₂ can unlock a carbon benefit 36 times greater.

4. CONCLUSIONS

Industrial scale integration of a carbon dioxide injection into ready mixed concrete has demonstrated a means to beneficially use carbon dioxide to improve concrete performance and create more sustainable concrete. The performance improvement can be the basis to reduce binder loadings without compromising on compressive strength. Three way comparisons confirmed that removing 7-8% of binder from a concrete mix would lead to a reduction in strength, but the addition of CO₂ had the potential to restore the compressive strength performance.

The modified binder loadings result in a concrete with a reduced carbon footprint. A small amount of carbon dioxide is absorbed directly but a larger amount of CO₂ emissions would be avoided by reducing the cement loading. A generic case suggests that a 4.6% reduction in the carbon footprint is feasible. The energy and materials required to implement the approach (building the equipment, capturing the carbon dioxide, transporting the equipment and the carbon dioxide) result in a small emission of CO₂ that is less than the amount of absorbed CO₂ or otherwise quickly outstripped by the environmental impact associated with the mix optimization.

Cement producers would then be able to put their waste CO₂ to beneficial use in concrete production thereby upcycling a portion of their primary waste product and using resources in a manner consistent with circular economy principles.

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Compliance with ethical standards.

Conflict of interest

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References

http://dx.doi.org/10.1016/j.conbuildmat.2015.10.080.
Athena Sustainable Materials Institute, 2016. NRMA Member National and Regional Life Cycle Assessment Benchmark (Industry Average) Report e Version2.0 [WWW Document]. URL.
http://dx.doi.org/10.1016/j.cemconcomp.2010.07.009.
http://dx.doi.org/10.1016/j.cemconcomp.2015.07.004.
European Federation of Concrete Admixtures Associations Ltd (EFCA), 2015. Environmental Product Declaration - Concrete Admixtures - Plasticisers and Superplasticisers.


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