Field demonstration of the Reversa™ mineral carbonation process using natural gas flue gas streams at the National Carbon Capture Center, AL, 2024

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Executive summary:
Concrete, a mixture composed of a cementation agent, mineral aggregates, and water has the potential to serve as a gigaton-scale sink for carbon dioxide (CO_2). This could make concrete the world's largest CO_2 utilization opportunity. CarbonBuilt's Reversa^{TM} process, developed at UCLA's Institute for Carbon Management exploits simple acid-base chemistry to mineralize CO_2-dilute flue gas emissions into mineral carbonate-based cementation agents at ambient pressure, at flue gas temperatures, and without a need for carbon capture. The approach leverages innovations in the use of portlandite ($Ca(OH)_2$: calcium hydroxide, or slaked lime) which carbonates readily, and produces limestone ($CaCO_3$: calcium carbonate) – a potent cementation agent – upon its carbonation. Within the scope of a project sponsored by the US Department of Energy's Office of Fossil Energy, the Reversa technology was upscaled and demonstrated using a modularized pilot-plant at the National Carbon Capture Center (Wilsonville, AL) using natural gas (~7-8.5 vol. % CO_2) flue gas streams. During the course of the demonstration, 31.34 tonnes of CMU, 51.31 tonnes of SRW and 40.06 tonnes of concrete manhole were produced and carbonated: (1) test carbonation process on 3 separate concrete products, (2) produce >10 t/d of concrete of each product, (3) absorb > 0.2 $gCO_2/g_{reactant}$, and (4) ensuring compliance of carbonated concrete with industry standard specifications. The success of this demonstration suggests that the pioneering Reversa technology is ready for further product development commercialization.

1 Introduction

Anthropogenic sources of carbon dioxide are generated from a number of sources, but key among these are ordinary Portland cement (OPC) production and combustion of fossil fuels [1]. Cement production is the largest global CO_2 source from the mineral decomposition of carbonates [1]. This is due to the clinkering process whereby limestone (mainly consisting of $CaCO_3$) is decomposed into CaO and CO_2, and combined with silica rich clays at high temperatures to form clinkers (i.e. the four key minerals that comprise cement) [2]. The high temperature range of 1400 – 1550 °C required for this process accounts for up to 60% of the generated CO_2 from cement production [3]. Combination of the limestone decomposition and thermal requirements of the clinkering process causes cement production to contribute 8-9% of annual global CO_2 emissions [1,2,4–6]. Combustion of fossil fuels (coal, oil, and gas) was shown to contribute a much larger portion of global CO_2 emissions. As of 2018, combustion of fossil fuels accounted for 65% of global CO_2, where 41% was derived from stationary sources for electricity and heat generation and the other 24% was related to transport [7]. To reduce these contributions, key steps forward in CO_2 utilization technologies are required. Therefore, a CO_2 mineralization technology (CO_2 mineralization concrete) to reduce the OPC content in concrete, while utilizing flue gas emissions from fossil fuel combustion has been developed to address both areas simultaneously.

This Reversa technology utilizes low-carbon cementation agents produced by *in situ* CO_2 mineralization ("mineral carbonation reactions") to offer a promising alternative to OPC [8–12]. CO_2 mineralization relies upon the reaction of dissolved CO_2 with inorganic alkaline reactants to precipitate mineral carbonates (e.g., $CaCO_3$), which bind proximate particles and achieve cementation [9,12–14]. Herein, a concrete *green body*, that is composed of a mixture of binder, water, and mineral aggregates, is exposed to CO_2 borne in industrial flue gas streams. This manner of CO_2 mineralization allows the production of construction components that feature equivalent engineering attributes as their OPC based counterparts while featuring a much smaller embodied carbon intensity (eCI).

The reliability of the Reversa technology has proved effective for the production of concrete masonry units (CMUs) at bench scale, where the units exceeded the required 13.8 MPa compressive strength requirements [14,16]. This report demonstrates the applicability of this technology at an industrial scale and expands the testing to include two additional concrete products; segmental retaining walls (SRW) and concrete manholes. In total, 9 production runs were completed at the National Carbon Capture Center (NCCC), Wilsonville, AL, using natural gas (NG) flue gas as the CO_2 source. This was the third site demonstration performed using this technology following 12 successful production runs at the Integrated Test Center (ITC), Gillette, WY in 2020 and 6 successful runs at the NCCC in 2021. Over the course of the production runs at NCCC, the CO_2 utilization as a function of time, 18-h CO_2 uptake, electricity usage, and 28-d net area compressive strength was recorded for each run. Collection of this data was used to determine the success of the demonstration goals: (1) test carbonation process on 3 separate

concrete products, (2) produce >10 t/d of concrete, (3) absorb > 0.2 $gCO_2/g_{reactant}$, and (4) ensuring compliance of carbonated concrete with industry standard specifications.

2 Materials and methods

The field demonstration at NCCC required the CMUs to be produced off-site at the Blair Block (CMU and SRW) and Alcrete (manhole) concrete plants. The concrete products were transported to NCCC, then loaded into a custom-built carbonation chamber. A mixture of inorganic reactants (e.g., the *binder*), inert aggregates (concrete sand, fine and coarse aggregates), and water was used to produce "dry-cast" formulations suitable for the fabrication of CMU and SRW. Formulations suitable for "wet-cast" concrete were composed for the manhole product. The binder used consisted of commercially available portlandite ($Ca(OH)_2$) powder (Standard Hydrated Lime, Mississippi Lime Co.), ASTM C150-compliant ordinary portland cement (PLC),[17] and an ASTM C618-compliant class C fly ash [18].

Blair Block produced 8" CMUs and SRWs. After forming, the fresh concrete blocks were pre-cured to gain sufficient strength (compressive strength σ_c = 6 ± 1MPa) to enable transport, handling and loading into the carbonation reactor. Alcrete produced 4' length and 4' ID concrete manholes. The concrete manholes were cured overnight to achieve sufficient demolding strength (compressive strength σ_c = 13.8 ± 1MPa) before being palletized and transported to site.

The curing chamber at the NCCC, consisted of a modified open-sided 40' shipping container with steel racks (40 in. wide x 98 in. deep x 76 in. tall), each capable of holding 96 and 72 CMU and SRW, respectively, blocks (around 1.4-to-2.1 metric tons of carbonated concrete), comprising a total of 10.9-to-16.3 metric tons of carbonated concrete per production batch. For the manholes, additional racks were fabricated to hold each of the concrete manholes. The manholes weighed ~1.67 metric tons, totaling 13.3 tonnes of wet-cast concrete.

Up to 4 process cycles were used to complete the carbonation process: 1) drying, 2) humidification, 3) carbonation cycle where conditioned flue gas was introduced to the curing chamber, and 4) purge cycle to remove the flue gas from the chamber. During the carbonation cycle, the inlet flue gas stream of [CO_2] = 7-8.5 v/v% for NG flue gas was used. The total curing cycle was set at 24 hours including 6-h pre-curing and 18-h carbonation to match commercial block production cycle time under ambient pressure and sub-boiling temperature (<75 °C). The concrete manhole underwent 12-16 hour pre-curing at the concrete plant followed by a 0-to-24 hour drying cycle before undergoing an 18-hour carbonation cycle.

CO_2 utilization as a function of time was calculated based on the change of CO_2 mass fraction of the outlet flue gas as compared to the inlet flue gas over time. Over the course of an 18-h production run, this value can be averaged to provide the 18-h CO_2 uptake value. Electricity usage of the chiller and process skid (heater, fans, and control HMI) was recorded to detail the energy usage of the process.

The performance of carbonated products produced during system operations was assessed against the relevant industry standard. The net area compressive strength of the concrete units were measured after 28 days of age.

Embodied carbon intensity was calculated via the sum of the equivalent CO_2 for producing the raw materials, transport, energy used in the concrete making process, energy for the carbonation process and finally a reduction based on the CO_2 uptake.

3 Test procedures for NCCC demo

The accelerated concrete carbonation production process incorporates the following steps (see Figure 1): **Step 1:** At a concrete block plant, concrete mixtures are batched from their raw materials (i.e., aggregates, binders, and water) and homogenized. This mixture is subsequently compacted using a concrete block machine to form fresh, concrete blocks as CMU or SRW. These blocks will remain at the concrete block plant for a pre-curing period. The manholes will be poured into their forms and cured at ambient conditions for 24 hours. All three products will be curing until they achieve sufficient *green-strength* for handling and transportation. The semi-cured concrete blocks will be palletized at the concrete block plant for transportation.

Step 2: The semi-cured concrete units are then transported from the manufacturing plant to the demonstration site. **Step 3:** the products are loaded into a curing chamber for the following sequential processing steps: flue gas injection and carbonation/CO_2 mineralization. The cured concrete will then be removed from the curing chamber and stored. **Step 4:** The fully cured concrete will then be sampled and selected per a pre-determined scheme for off-site testing and evaluation of products.

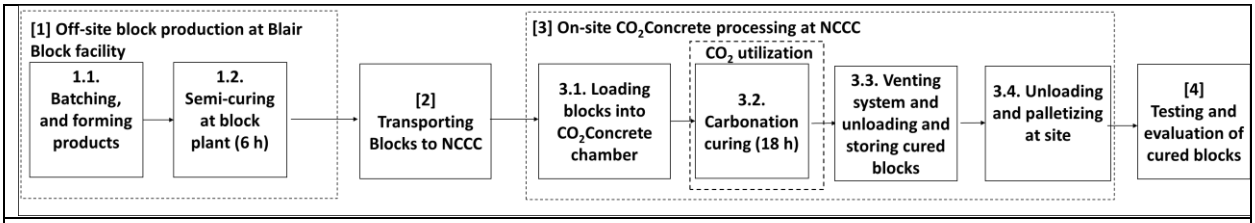


Figure 1: A schematic illustration of the different steps involved in the CO_2Concrete block production process.

The test plan for operations at the National Carbon Capture Center (NCCC) has been detailed in Table 1. Utilization of steady state process models have guided the development of the system design test plans. Three main process operation variables will be considered during the demonstration:

1) Effect of recycle ratio,
2) Effect of gas processing conditions,
3) Drying time required for wet cast units.

Variables 1) and 2) will be applied to the dry cast concrete units of concrete masonry units (CMU) and segmented retaining walls (SRW). The learnings from these tests will be applied to the wet cast concrete units of the manholes. Variable 3) will only be applied to the wet cast unit as drying of the saturated pores will be more necessary to improve the carbonation mechanics.

The suggested test plan is shown in Table 1. Trial IDs C1 to C3 will focus on the effect of the recycle ratio on the system and be tested against the CMU product to test Variable 1). The effect of the gas processing conditions at a low recycle ratio will be tested on the SRW product based on trial ID S1 to S3 to test Variable 2). Variable 3) will be tested using trial IDs M1 to M3.

Table 1: Design of experiments for the scale-up demonstration at the National Carbon Capture Center. 'RR' = recycle ratio.

Trial ID	Product ID	Drying time (h)	Carbonation time (h)	Single run target	Variable tested
C1	CMU	0	18	No RR	Testing low-to-high recycle ratio
C2		0	18	Medium RR	
C3		0	18	High RR	
S1	SRW	0	18	High energy	Testing low-to-high energy requirements
S2		0	18	Medium energy	
S3		0	18	Low energy	
M1	Manhole	0	18	No drying	Testing wet-cast concrete product
M2		12	18	Medium drying	
M3		24	18	High drying	

4 System installation

The system was delivered to NCCC site at the end of February 2024 and was completed with commissioning by the end of April 2024. Figure 1 presents photographs of the system in various stages of installation.

(a)

(b)

Figure 2: Photographs of (a) delivery of equipment, and (b) the CO_2 mineralization curing chamber.

Once the system was installed and the pre-startup safety review (PSSR) meeting was completed, commissioning of the equipment began. Commissioning consisted of applying improvements to the system based on the NCCC HAZOP and, instrument and sensor installation.

5 Results

The pilot scale demonstration of the Reversa system at the NCCC host site was completed by the end of May 2024. In total, 9 operational runs were completed fulfilling an operational duration of nearly 219 hours. Around 123 tonnes of concrete was produced while achieving 1.3 tonnes of CO_2 uptake.

The CO_2 uptake (i.e., CO_2 removed from the gas stream and mineralized within the CO_2Concrete blocks) over an 18-h carbonation duration was determined via the system's own instrumentation (i.e. flow and CO_2 sensors). Thermogravimetric analysis was not performed on the concrete as the producers used limestone aggregate which would contaminate the TGA results.

The CO_2 utilization efficiency % was determined as the average CO_2 uptake divided by the CO_2 input over the 18-h carbonation period. The CO_2 input into the system was calculated by calculating the mass flow rate of CO_2 into the system, from the measurements of CO_2 concentration and flow rate at the system inlet. The measured CO_2 concentration was corrected for the concentration of water vapor in the flue gas, which was determined based on the system inlet relative humidity/temperature sensor readings and flow rate measurements. The gas flow rate calculations were accounted for the dependence of flow rate on gas density and its composition. Similar calculations were performed for various locations in the system.

Figure 3a), b) and c) display the CO_2 conversion performance of CMU, SRW and manhole concrete units, respectively. The CO_2 uptake was determined in two ways: (1) from the difference in CO_2 mass flow rates between the system inlet and system exhaust, and (2) from

the difference in chamber inlet and outlet mass CO_2 mass flow rates. The CO_2 utilization efficiency was calculated as the CO_2 uptake into the CO_2Concrete divided by the CO_2 input into the system. Each batch demonstrated CO_2 uptake trends similar to that of Figure 3a-c), with some variations due to changes in system inlet flow rates, mixture compositions, and the specific processing conditions that were applied.

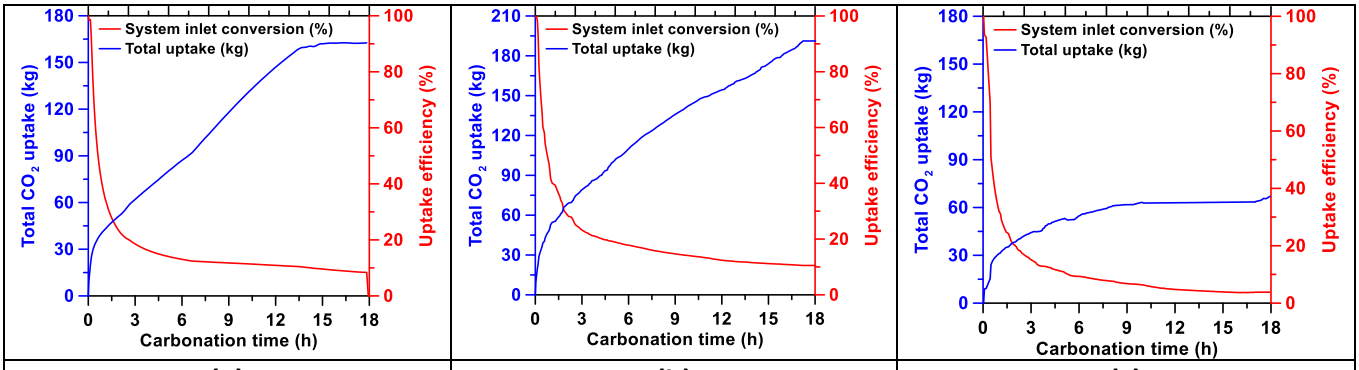
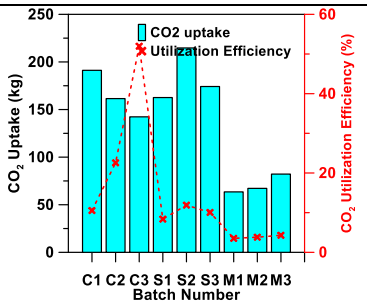


Figure 3: Representative plot of cumulative CO_2 uptake into the CO_2Concrete products, and CO_2 utilization efficiency as a function of 18-h carbonation duration in a) S1 (SRW), b) C1 (CMU) and c) M1 (manhole).

The cumulative CO_2 uptake into the CO_2Concrete products and CO_2 utilization efficiency as a function of the 18-h carbonation period for all 9 demonstration runs are shown in Figure 4(a).

- C1-C3: Higher RR (C3) resulted in lower CO_2 uptake but greater CO_2 utilization as evidenced by C1-3, as expected by the process modeling. C1 observed the highest CO_2 uptake for the CMU products due to the higher throughput of CO_2 into the reactor.
- S1-S3: The SRW data used a slight variation on the system's heater settings which led to minimal variation in CO_2 uptake except for S2 which showed a spike.
- M1-M3: The manhole units displayed the lowest uptake and conversion. This was expected due to the wet cast nature of the concrete and the greater wall thickness. Including a drying cycle before carbonation (M2 and M3) showed minor improvement but vastly increased the energy intensity of the CO_2 uptake.

As a result of the mixture design, CO_2 processing and carbonation conditions, the target 0.2 $gCO_2/g_{reactant}$ was exceeded for each of the production runs as shown in Figure 4b).



a)

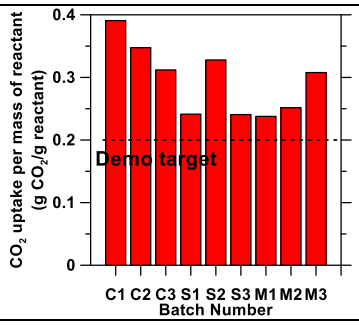


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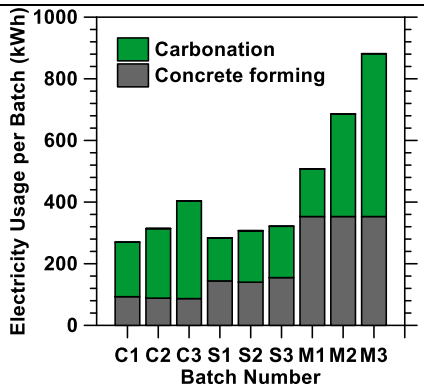
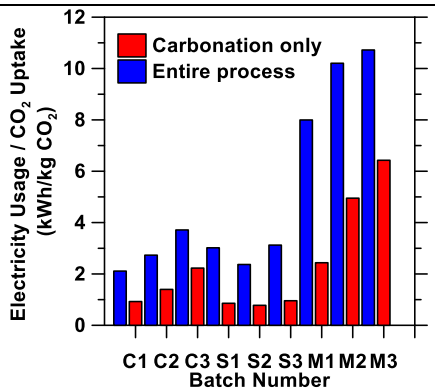
Figure 4: a) CO_2 uptake and conversion of the various production runs, and b) CO_2 uptake per mass of reactant.

Energy usage as shown in Figure 5a) was used to determine the energy intensity of the carbonation and entire process as shown in Figure 5b).

- The low recycle ratio of C1 proved to be most energy-efficient, - and the SRW runs appeared to have similar energy intensity compared to C1. This is likely due to SRW containing ~39% more concrete in the reactor compared to CMU which could account for a comparable energy intensity.
- The concrete manhole did not perform well at all due to the low CO_2 uptake and high energy penalties.



a)



b)

Figure 5: a) the energy usage for carbonation and entire production, and b) energy intensity of the production runs.

The performance/compliance of CO_2Concrete units produced during system operations are assessed against the relevant industry standard for each product. The key output for the concrete products are the required compressive strengths as shown in Figure 6a-c).

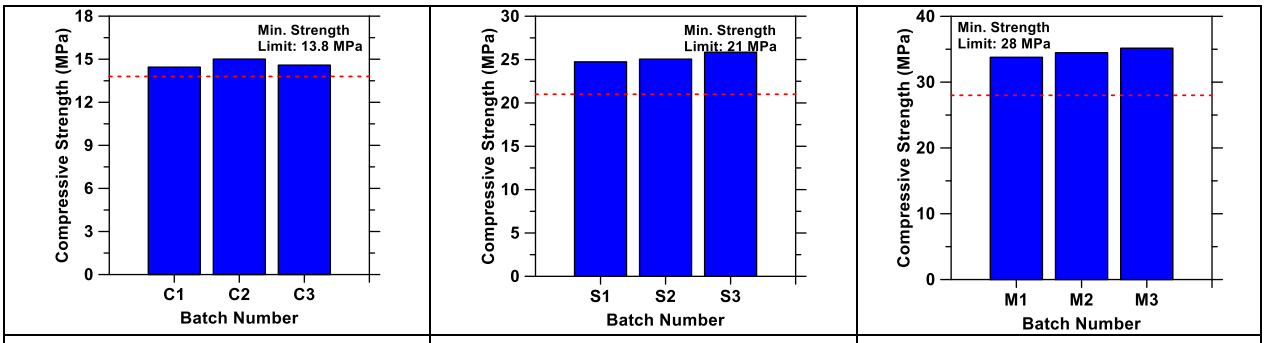


Figure 6: The average compressive strengths of CO_2Concrete units for a) CMU (28 d), b) SRW (28 d) and c) manhole (28 d).

A separate full Life Cycle Assessment (LCA) report was submitted to DoE, however a summary of the results can be seen here. The life cycle impact assessment results for the proposed and comparison product systems including the future ideal scenarios are shown in **Error! Reference source not found.**. The emissions from the CO_2Concrete products calculated for each product with the best performing Pilot-scale data shown and the ideal inputs as proposed by the sensitivity analysis. The net emissions from the production of one cubic meter of traditional CMU, SRW and manhole are 314, 365 and 466 kg CO_2e. Using CO_2concrete technology, the net emissions were lowered to 220 (30.1 % reduction), 251 (31.3 % reduction), and 373 (19.8 % reduction) kgCO_2e, respectively.

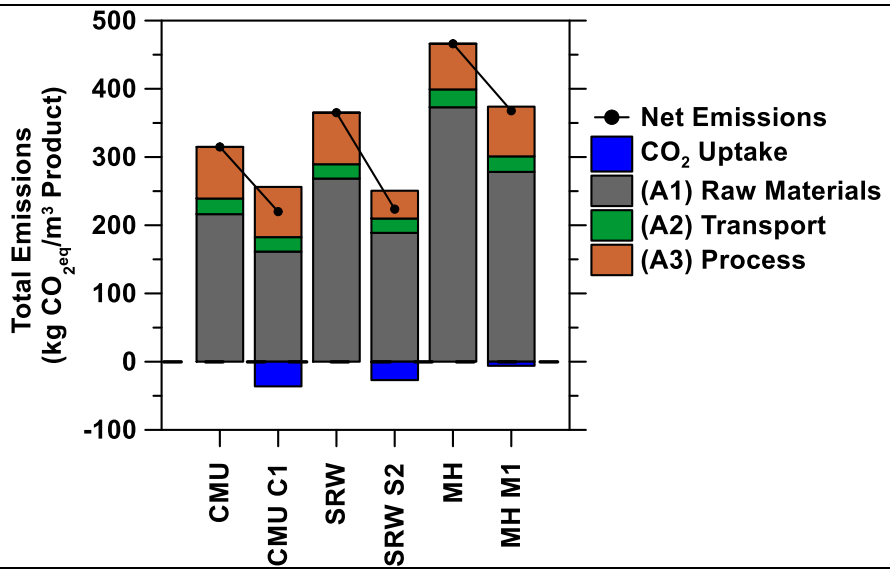


Figure 7: The global warming potential (GWP, kg CO_2e/m^3 concrete) for carbonated concrete products (estimated) and conventional OPC-concrete products. 'MH' = manhole.

6 Conclusion

The system's performance fulfilled all design specifications: (1) Test carbonation process on 3 separate concrete products, (2) Produce >10 t/d of concrete, (3) absorb > 0.2 $gCO_2/g_{reactant}$, and (4) ensuring compliance of carbonated concrete with industry standard specifications. Furthermore, the Reversa process conditions were optimized to reduce the energy input as compared to conventional cement-based concrete products. The Reversa process addresses the largest opportunity for CO_2 utilization (i.e., construction materials) by exploiting chemical reactions that do not require extrinsic energy inputs (i.e., thermodynamically downhill) even while using CO_2-dilute feedstocks (flue gases). The concrete produced via Reversa processing and formulation achieved price parity to traditional cement-based concrete products.

7 References

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