

A Scalable Process for Upcycling Carbon Dioxide (CO₂) and Coal Combustion Residues into Construction Products

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Motivation and project objectives

The problem at hand – CO₂ emissions from cement/concrete: Concrete, a mixture of portland cement, aggregate, and water is indispensable in construction (> 30 billion tons produced / year). But nearly 1 ton of CO_2 is emitted for each ton of portland cement produced (> 4 billion tons / year). As the vast concrete market provides an impactful sink for CO_2 emissions, the CO_2 mineralization process can enable scalable and cost-effective decarbonization in construction.

1. Upcycle industrial wastes and CO₂ - Produce low-carbon CO₂Concrete products from coal combustion residues, flue gas CO₂, and low-grade waste heat

- 2. Design CO₂ mineralization system Develop process models to inform scale-up design of a "bolt-on" system at coal-fired power plants
- 3. Field test system using real flue gas Fabricate and field test CO₂ mineralization system to capture around 100 kg of CO₂ per day from coal-fired flue gas
- 4. Product compliance Ensure CO₂Concrete product compliance with industry standards; demonstrate potential utilization in construction applications

Process flow for developing low-carbon concrete by CO₂ mineralization



• Portlandite $(Ca(OH)_2)$ is a highly efficient reactant for CO_2 mineralization $(CO_2 \text{ uptake } 0.59 \text{ g/g})$ that is also abundant and near cost parity to cement

- Carbonation occurs rapidly at ambient temperature and pressure without carbon capture step, pressurization or gas clean-up (insensitive to SOx and NOx)
- "Green bodies" are shape-stable components that are exposed to flue gas in a carbonation reactor
- Process is flexible: Simple integration at any CO₂ emissions source ("stack-tap") which enables co-location and low-cost processing



Carbonation kinetics, system performance, and product compliance



Portlandite carbonation at dilute CO_2 concentrations:

- Reaction kinetics are largely independent of CO₂ concentration for flue gas concentrations ($\geq 2\%$)
- Activation energy is rather low: initial surface reaction (3 kJ/mol) and (22 kJ/mol) when transport barriers may form; confirming that no pressurization, CO_2 enrichment,



Effects of microstructure and pore saturation on carbonation:

- CO₂ diffusion through pore structure limits reaction rates which scales with scale with body's moisture diffusivity
- Liquid water saturation (S_w) in porous cementing microstructures influences carbonation kinetics; S_w $\approx 0.1 - 0.2$: critical level for CO₂



Computational fluid dynamics (CFD) modeling for reactor design:

- Performed CFD to inform design of flue gas handling and distribution equipment within the CO_2 mineralization reactor
- Spatial distribution of CO₂ uptake is significantly affected by gas flow configuration. Greater gas velocity and homogeneity offer higher CO_2

Pilot scale system demonstration at Integrated Test Center, Wyoming:

12

Period of Flue Gas Exposure (h)

16

20

— Inlet

— Outlet

– Uptake

150

50

- Produced around ~200 tonnes of CO₂Concrete blocks over 12 demonstration runs that featured nearly 4 tonnes of CO₂ uptake
- System performance fulfilled all design specifications: (1) achieved in excess of $75\% CO_2$ utilization efficiency and (2) utilized greater



Performance of CO₂Concrete products:

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20

- CO₂Concrete products complied with industry standard specifications: strength > 13.8 MPa and water absorption < 208 kg/m³
- Preliminary lifecycle analysis (LCA) indicated ~ 65 % CO_2 emissions reduction relative to conventional CMUs















• Falzone, G.; et al. New insights into the mechanisms of carbon dioxide mineralization by portlandite. AIChE Journal. Accepted for publication.

• Mehdipour, I.; et al. How Microstructure and Pore Moisture Affect Strength Gain in Portlandite-Enriched Composites That Mineralize CO₂. ACS Sustainable Chem. Eng. 2019, 7 (15), 13053–13061.