The carbonation kinetics of alkaline solids at conditions relevant to direct flue gas utilization

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(Session 11: Laboratory Investigations)

Presentation outline – The three questions

Where do you come from?	• CO_2 mineralization processes in history
Where are you going?	• Process overview and development pathway
What have you learned?	• Investigations into portlandite carbonation

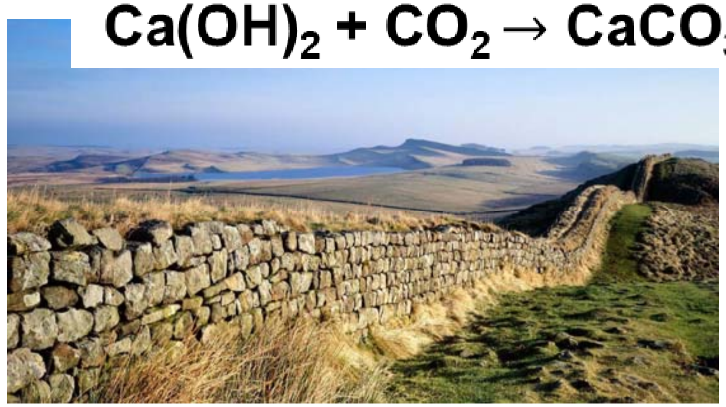
Where do you come from? CO_2 mineralization in history

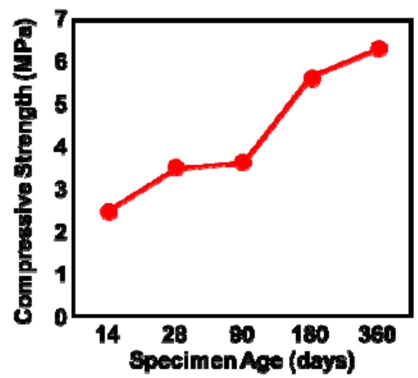
- CO_2 mineralization (carbonation) reactions
- Geo. / bio. processes
- Lime mortars used in ancient civilizations
- Reaction in ambient air

Limitations:

- Slow uptake kinetics
- Lack of control
- Poor strength

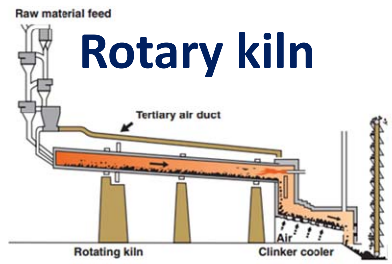
$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$$

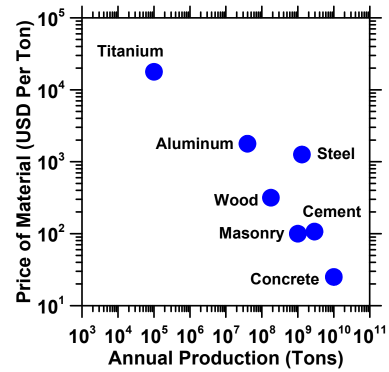


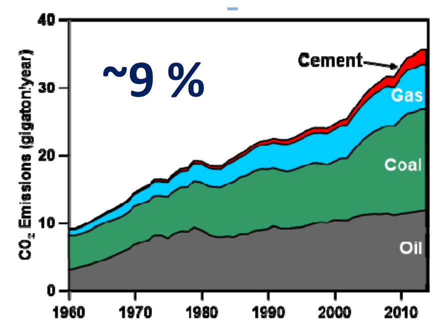


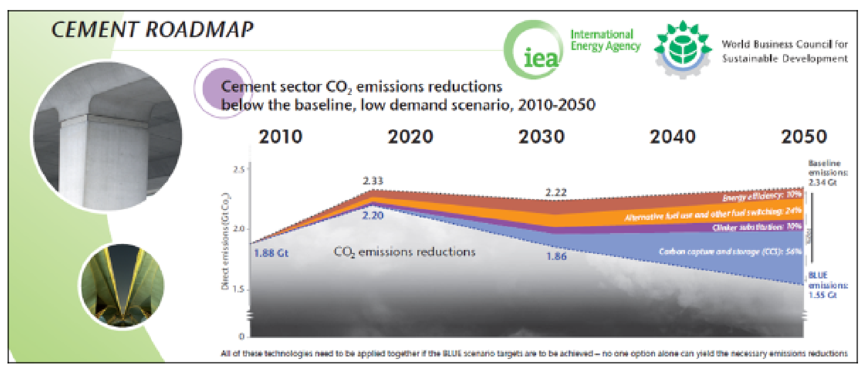
Cement/concrete – Problem and opportunity

- Late 1800s: OPC (portland cement)
- OPC concrete = the foundation of modern infrastructure
- Cheap, abundant, formable, resilient
- 9 % of annual global CO_2
- Large potential sink for CCU (gigaton level)



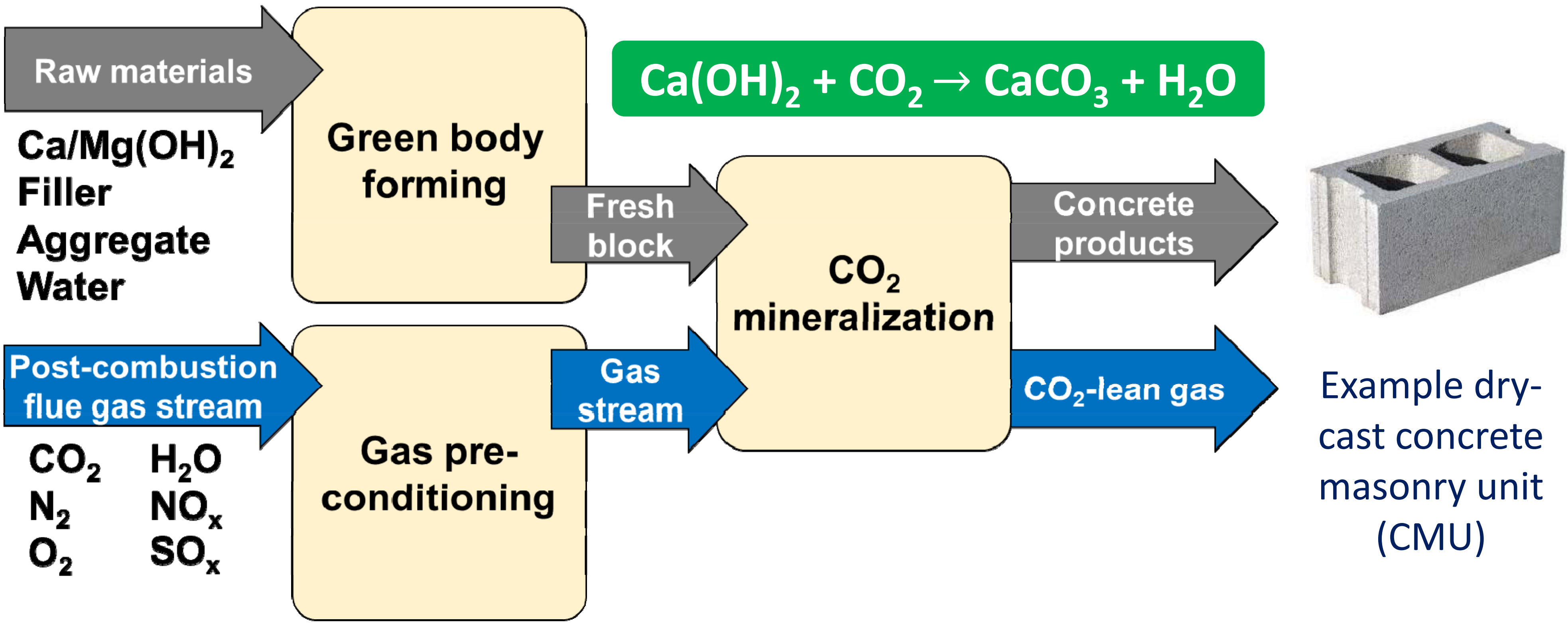






Limited options for reducing OPC's CO_2 footprint

Where are you going? CCU by CO_2 mineralization and cementation



Example dry-cast concrete masonry unit (CMU)

CO_2 emissions reduction potential – Preliminary lifecycle analysis

- Potential to impactfully reduce greenhouse gas reductions vis-à-vis concrete production
- Absolute emissions reduction scales with binder content (main factor for determining strength)

ASTM International. *Environmental Product Declaration: Normal-Weight and Light-Weight Concrete Masonry Units as Manufactured by Members of the Canadian Concrete Masonry Producers Association (CCMPA)*; p 16.

Estimated global warming potential (GWP, kg CO_2e/m^3 concrete)

Category	Conventional CMU	CO_2 mineralized CMU
A1 (Raw Material)	170	103
A3 (Manufacturing)	63	26
CO_2 uptake	0	-47
GWP (A1 + A3)	233	82

~ 65 % emissions reduction relative to conventional CMU

A1: CO_2 avoidance from substitution of OPC by fly ash and $Ca(OH)_2$

A3: CO_2 utilization from the gas stream

CO_2 mineralization technology development pathway

TRL 1	TRL 2	TRL 3	TRL 4	TRL 5	TRL 6	TRL 7	TRL 8	TRL 9
Concept	Formulation	Proof of concept	Lab prototype	Lab-scale system	Pilot plant	Demonstration	Commercial refinement	Commercial

- Current project raising Technology Readiness Level (TRL) of CO_2 mineralization technology from ~ 3 to ~ 5 (by end of 2020)
- Design, fabrication, and field testing of mineralization system using > 100 kg CO_2/day from coal-fired power plant flue gas
- Demonstrate product compliance to construction product standards (ASTM C90)
- Assess system performance and scalability (TEA & LCA)

U.S. DEPARTMENT OF ENERGY | NATIONAL ENERGY TECHNOLOGY LABORATORY

https://netl.doe.gov/project-information?p=FE0031718

What have you learned? Investigations into CO_2 mineralization

- Laboratory studies initiated in 2015
- Simulated demonstration (200 kg CO_2 input in 24 h)
- Uptake from simulated NG and coal flue gases
- System achieved > 65 % CO_2 utilization efficiency per 24 h
- *Increasing mineralization efficiency*

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Article

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Direct Carbonation of $Ca(OH)_2$ Using Liquid and Supercritical CO_2: Implications for Carbon-Neutral Cementation

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Clinkering-free cementation by fly ash carbonation

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Article

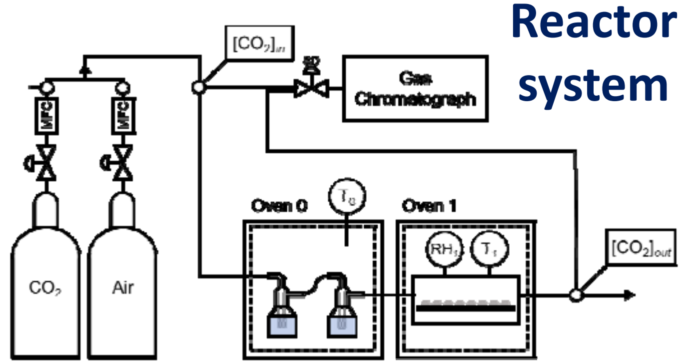
How microstructure and pore moisture affect strength gain in portlandite-enriched composites that mineralize CO2

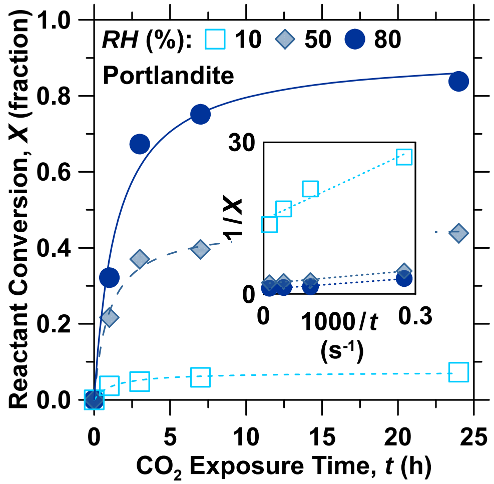
Iman Mehdipour, Gabriel Falzone, Erika Callagon La Plante, Dante A Simonetti, Narayanan Neithalath, and Gaurav N. Sant

Influences of processing conditions on portlandite carbonation

- Controlled RH, T, & [CO_2]
- Continuous gas flow (semi-batch)
- Conversion (% of max. CO_2 uptake) by TGA
- Data best-fit by equation of Lee (2004); used to estimate X_f and k

Reactor system





$$X(t) = (X_f \cdot t)/(X_f/k + t)$$

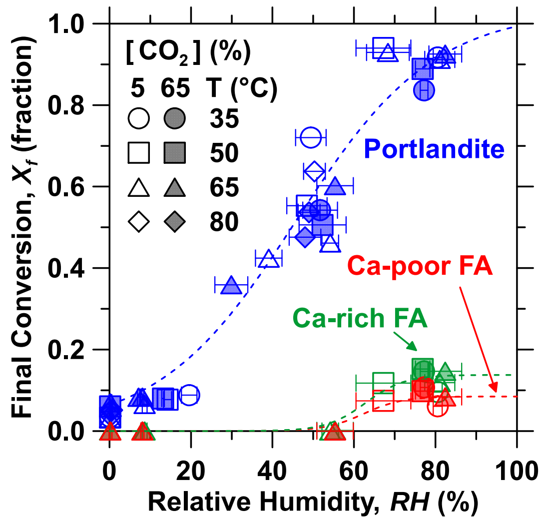
X_f = Final conversion (fraction) i.e., conversion limit

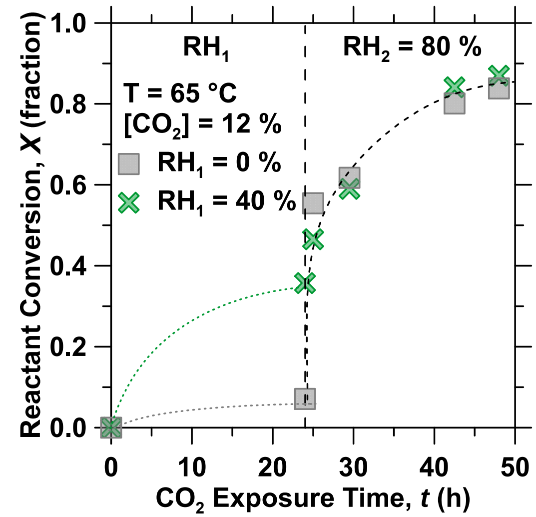
k = Apparent carbonation rate constant (s^{-1})

Lee, D. K. *Chem. Eng. J.* **2004**, *100* (1), 71–77. DOI:10.1016/j.cej.2003.12.003.

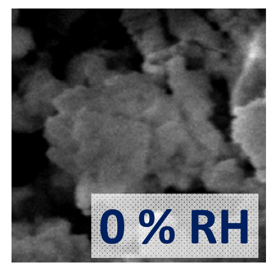
Conversion limits in portlandite carbonation

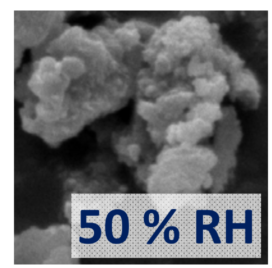
- Limited carbonation in dry gases*
- Dissolution-precipitation at higher RH
- No appreciable effect of T and [CO_2]
- Reaction at low *RH* is not passivating to further carbonation

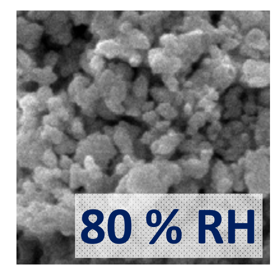




SEM (5 µm width) 24 h



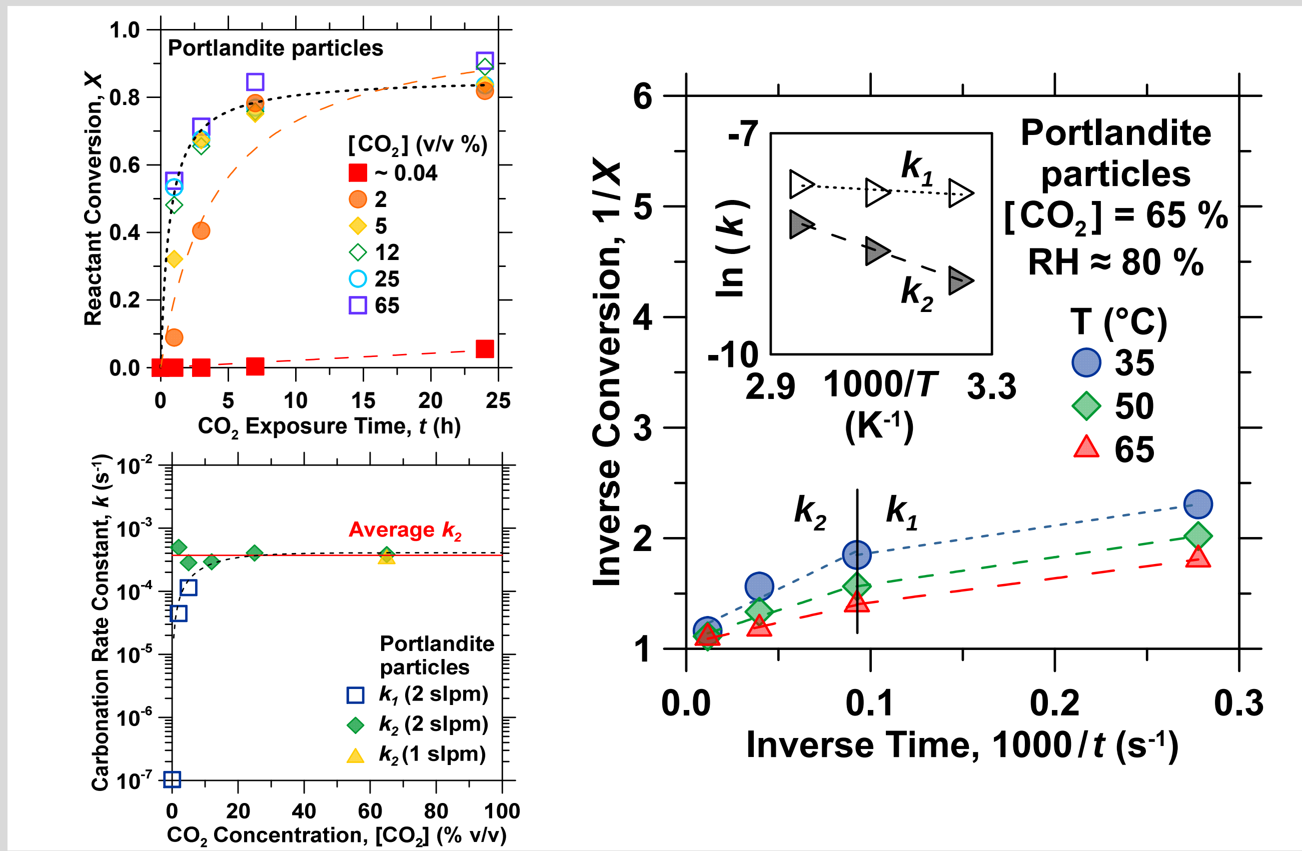




* Beruto, D. T.; Botter, R. *J. Euro. Cer. Soc.* **2000**, *20* (4), 497–503. DOI: 10.1016/S0955-2219(99)00185-5.

Portlandite carbonation kinetics – Sensitivity to [CO_2] and temp.

- Near-complete reaction within 24 h gas contact
- Largely independent of CO_2 concentration
- Low activation energy (~3 to 22 kJ / mol)
- Pressurization, CO_2 enrichment, or significant heating is not required



Presentation summary and future directions

- CO_2 mineralization is seeing new life in modern low-carbon construction
- Improving understanding of CO_2 mineralization reactions in relevant conditions – direct use of post combustion flue gas streams (no CO_2 capture step)
- Scaling and field demonstration of CO_2 mineralization processes for production of dry-cast concrete masonry from flue gas through 2020
- Presenting further research into microstructure effects on CO_2 mineralization reactions in tomorrow's Session 15: Carbon utilization (II)