Achieving low-carbon concrete with high mechanical properties using $CaCO_3$ suspension produced by CO_2 sequestration

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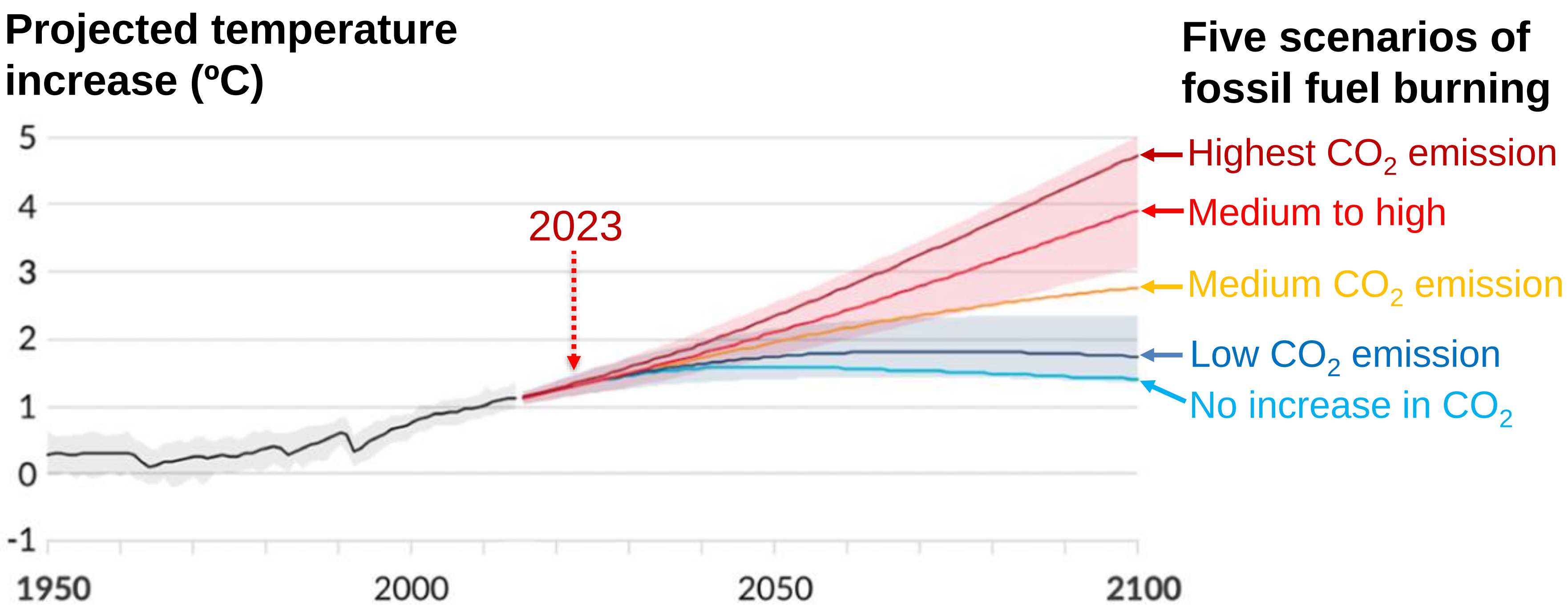
Climate change is impacting the earth

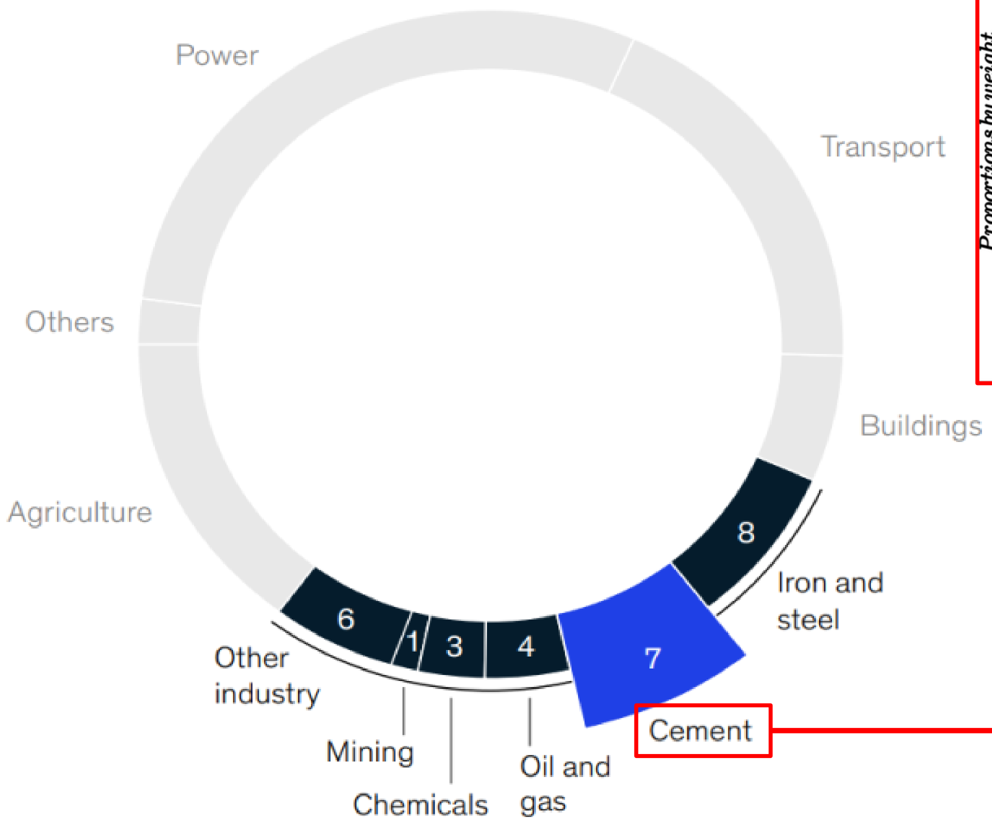


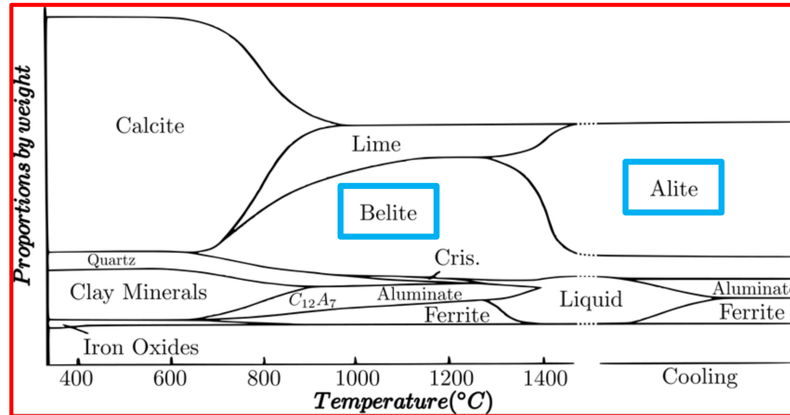
Temperature will continue increasing before the carbon neutrality goal is achieved

https://climateknowledgeportal.worldbank.org/overview

Cement production involves high CO_2 emission

High-temperature calcination for producing cement clickers is energy intensive





Cement production is responsible for 7% global CO_2 emissions in 2021

https://ourworldindata.org/CO_2-emissions

Strategy to decarbonize concrete

Through integrating low carbon ingredients, advanced processing methods, and new construction techniques

Low-carbon cost-effective high-performance concrete

Low carbon ingredients	Advanced processing	New construction
Binders Aggregates Fillers Fibers Admixtures	Pre-treatment Mixing Casting Curing	Controlled casting 3D printing Robotic spraying Robotic assembly
Solid wastes (fly ash, biochar)	Carbonation (CO_2) sequestration	

Intelligent design based on machine learning

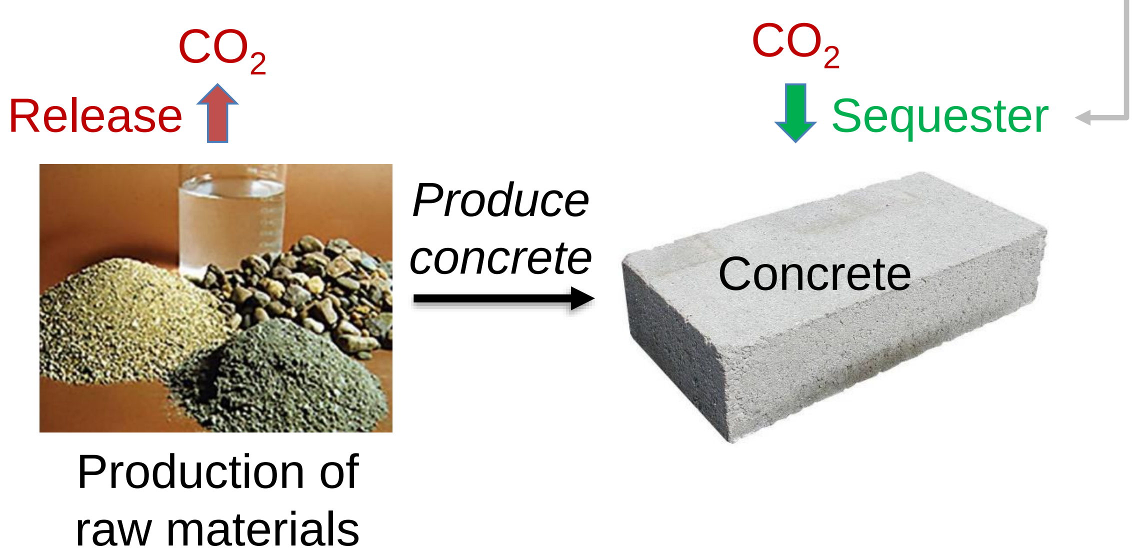
This presentation focuses on carbonation sequestration

Concrete can sequester CO_2

Cementitious materials-based concrete contains calcium ions (Ca^{2+}), which can induce the following reaction:

$$Ca^{2+} + CO_3^{2-} = CaCO_3$$

Ca^{2+} ← Concrete; CO_3^{2-} ← H_2O+CO_2; $CaCO_3$ ← Concrete



CO_2 sequestration can take place in three stages

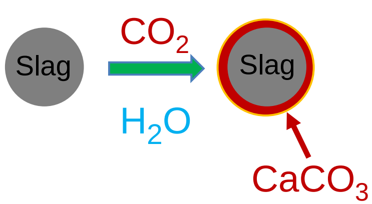
Manufacturing of concrete

Pre-treatment

Raw materials of concrete are cured in a CO_2-rich chamber

As-received steel slag

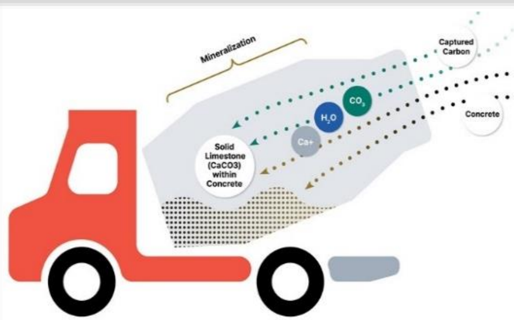
Carbonated steel slag

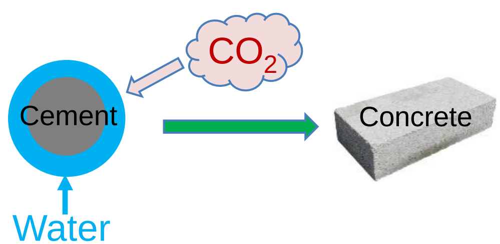


https://www.carbon8.co.uk/

Mixing

CO_2 is injected into the fresh concrete during the wet mixing process

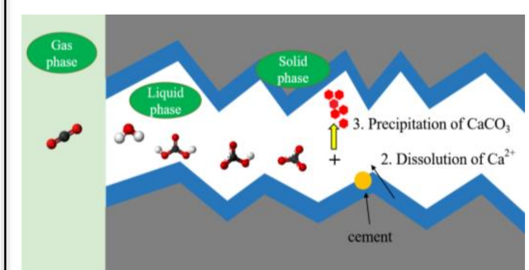




https://www.carboncure.com/

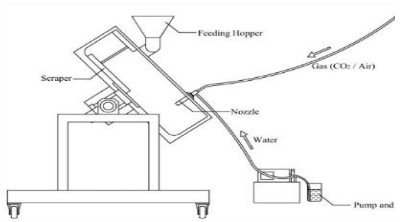
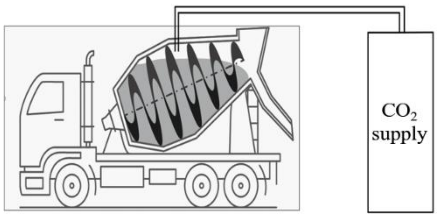
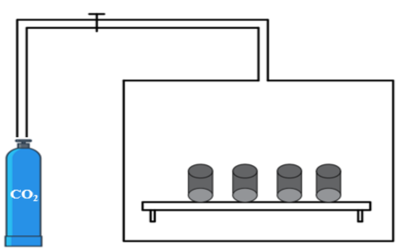
Curing

Concrete is cured in a CO_2-rich chamber with a controlled environment



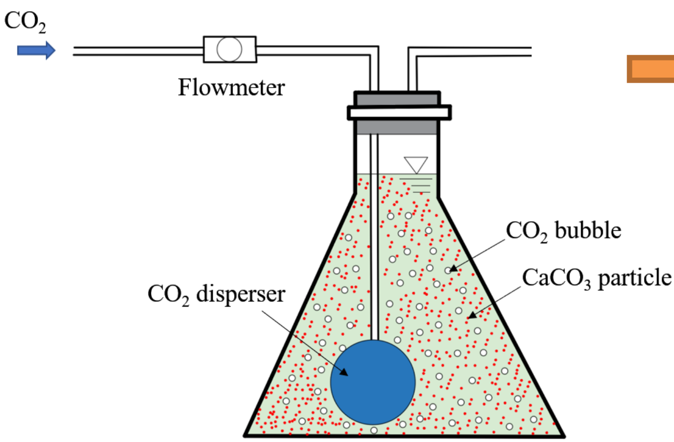
Liu and Meng, 2021, 2022

Comparison of the three technologies

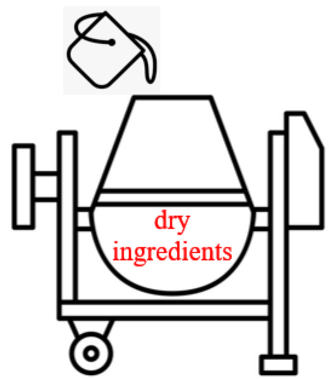
Methods for CO_2 sequestration	Advantages	Disadvantages
Pre-treatment stage: 	+ Recycle industrial waste + High sequestration rate (5-30%)	- Difficult for large-scale implementation as it requires large reactor and can be energy extensive.
Mixing stage: 	+ Applicable to cast-in-place construction	- Low sequestration rate (0.3%, by mass of cement by *Sean et.al., 2017*) - Negative impact on workability
Curing stage: 	+ High sequestration rate (10-20%) + Time-efficiency and energy-saving	- Difficult for cast-in-place implementation

A new method for CO_2 sequestration in concrete

- The method was proposed to enable cast-in-place concrete application with enhanced CO_2 sequestration rate.
- CO_2 is bubbled in calcium rich solution for preparation of uniformly distributed $CaCO_3$ suspension, which is then added into mixing water for concrete batching.



$CaCO_3$ suspension production



Concrete mixing

Concrete products

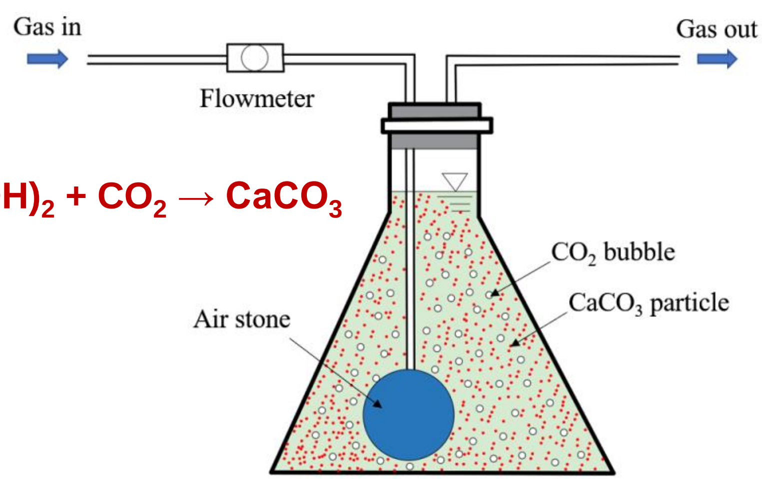
Compatible with existing industrial facilities and convenient for large-scale production

Preparation of $CaCO_3$ suspension

- CI suspension: is prepared by injection of CO_2 into different concentration of $Ca(OH)_2$ with controlled speed at 30 L/min to form $CaCO_3$ suspensions. The injection was continued until the mass of suspension became stable, indicating that no more CO_2 was absorbed. An air stone was used to distribute CO_2.

- CP suspension: is prepared with different content of nano-$CaCO_3$ powders.

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



Properties of nano-$CaCO_3$ powder

Particle size	30-60 nm
Bulk density	0.68 g/ml
pH	8.0-9.0
Moisture content	0.5%
$CaCO_3$ content	>97.5%
MgO content	<0.5%

Investigated mixtures

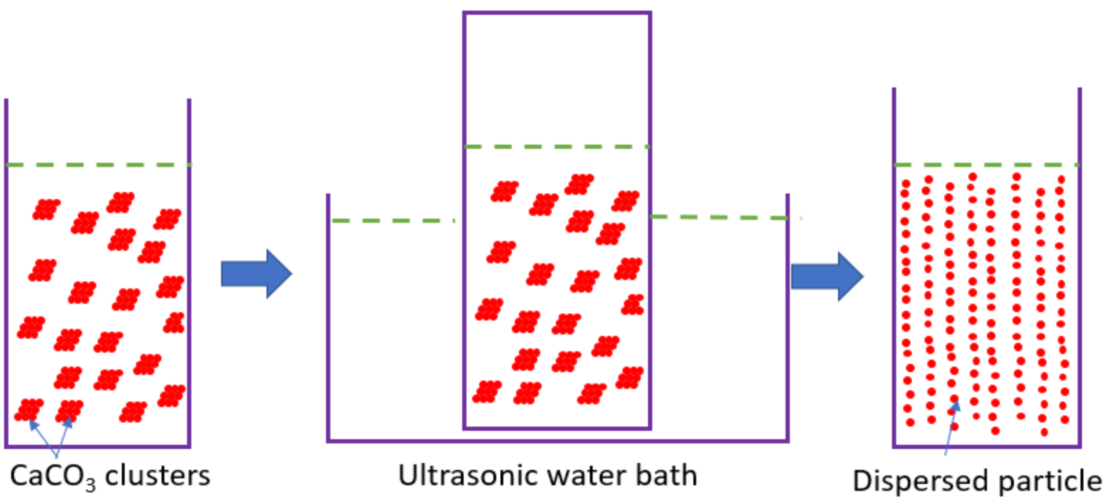
- Ten mixtures with different concentrations of $CaCO_3$ suspension prepared by the CO_2 injection (CI) method and nano-$CaCO_3$ powder (CP) method were investigated.

Designation	Cement	Synthesized $CaCO_3$	Nano-$CaCO_3$ powder	Water	HRWR
Control	1000	0	-	400	2.5
CI-0.5%	998	2	-	400	2.5
CI-1%	996	4	-	400	2.5
CI-2%	992	8	-	400	2.5
CI-4%	984	16	-	400	2.5
CI-6%	976	24	-	400	2.5
CP-1%	996	-	4	400	2.5
CP-2%	992	-	8	400	2.5
CP-4%	984	-	16	400	2.5
CP-6%	976	-	24	400	2.5

Note:
CI-1% represents the mixture with 1% $CaCO_3$ suspension using the proposed method.
CP-1% represents the mixture with 1% $CaCO_3$ suspensions using nano-$CaCO_3$ powder.

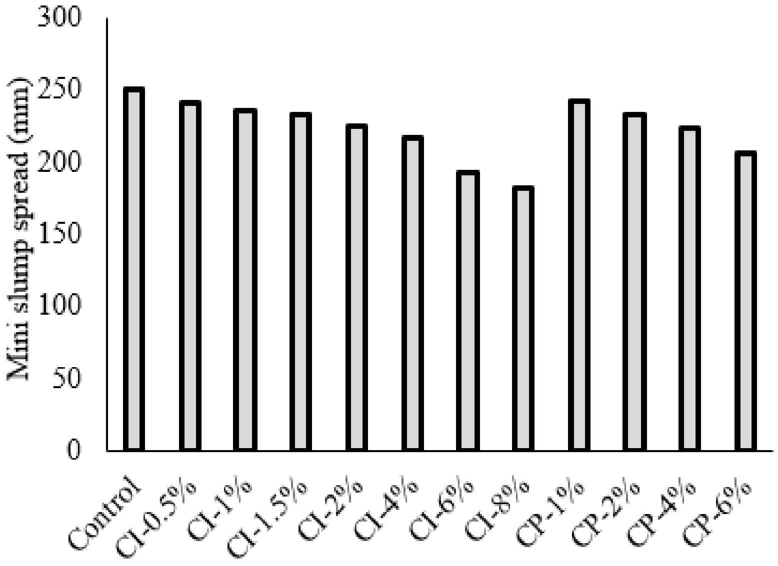
Dispersion of nano-$CaCO_3$ powders

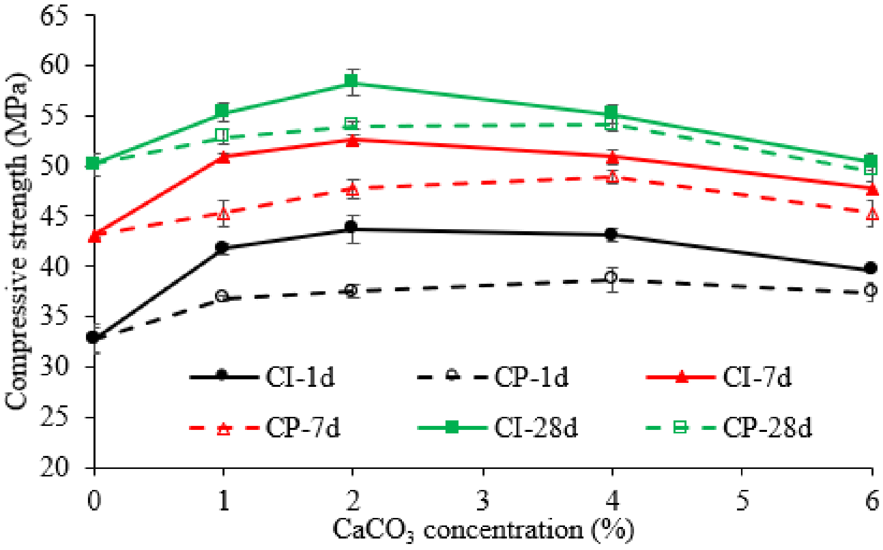
- When the nano-$CaCO_3$ powders to prepare concrete, the proper method to uniformly disperse the nano particles is critical, as nano particles are prone to agglomerate. The agglomeration significantly compromises the mechanical and durability of cementitious materials.
- In this study, the $CaCO_3$ suspension prepared with nano $CaCO_3$ powder was dispersed in the ultrasonic bath at 20k Hz for 10 min before use.
- This process makes the nanomaterials hard to be utilized in large-scale concrete production.



Fresh and hardened properties

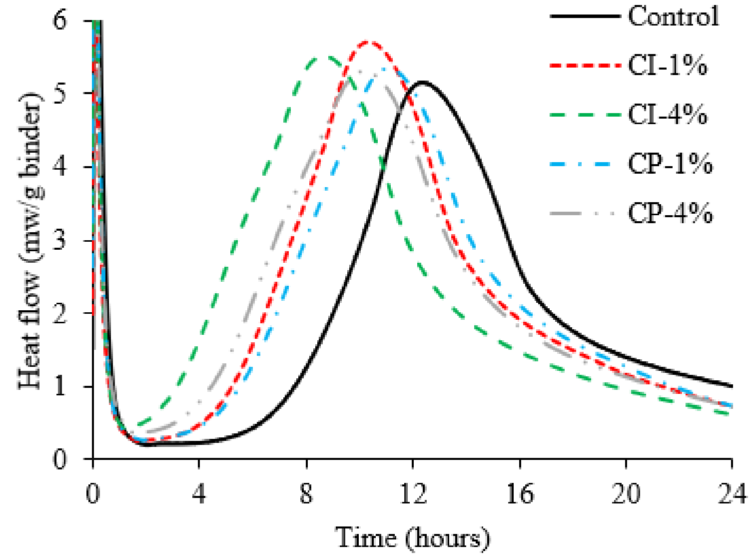
- Adding $CaCO_3$ suspension decreases the flowability, and the reduction rate by the CI method was higher than CP method.
- Compared with the reference mixture, the 28-day compressive strength was increased by 16% by the CI method (2% of nano-$CaCO_3$) and by 6% by the CP method (2% of nano-$CaCO_3$).

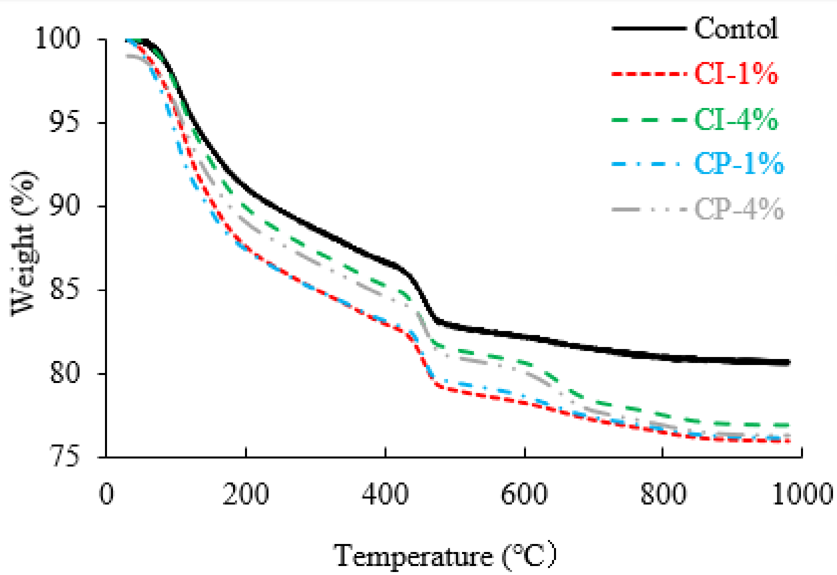


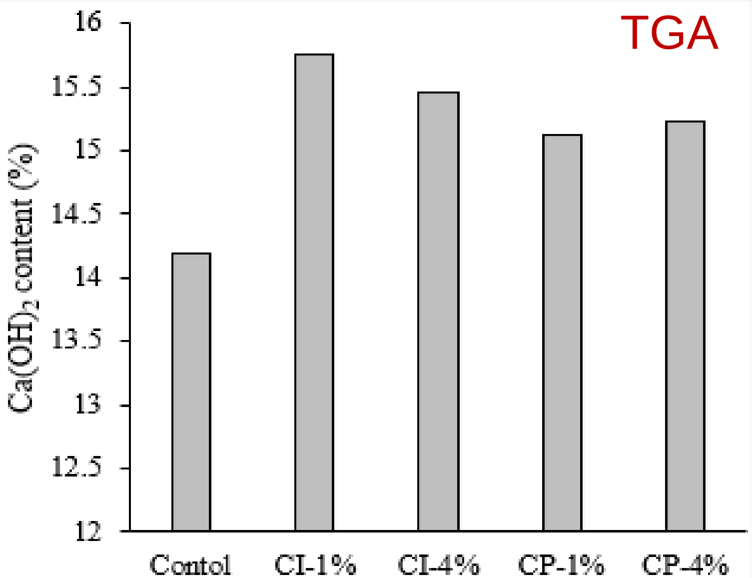


Hydration kinetics and thermal analysis

- Both the CI and CP methods promote cement hydration. Compared with the reference mixture,
  - ➢ the presence of peak is accelerated;
  - ➢ more hydration products are produced

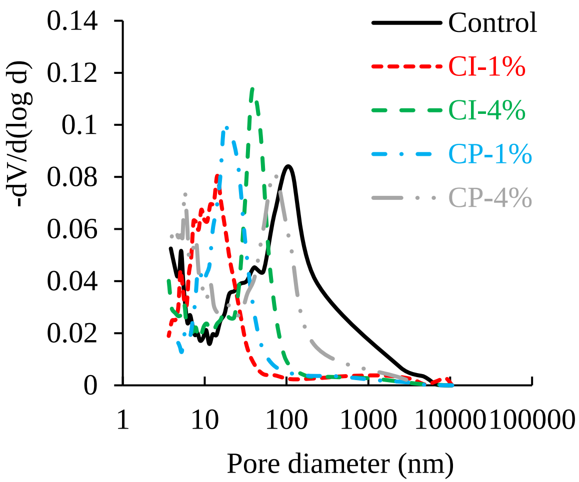


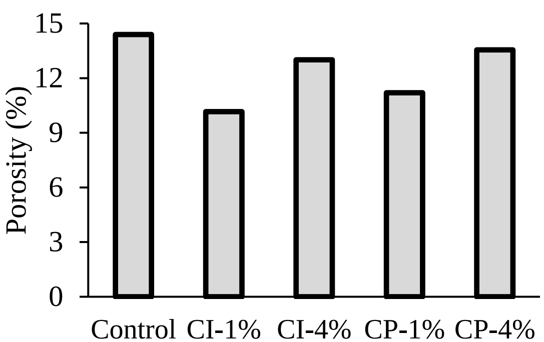




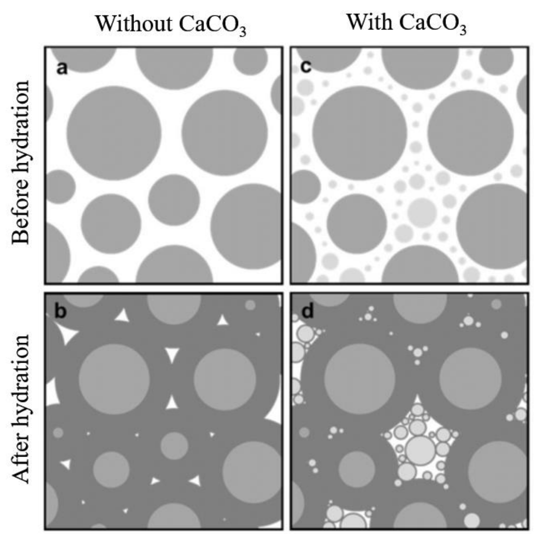
Porosity

- The use of nano-$CaCO_3$ reduced the capillary pores and pore sizes.
- The porosity of the CI mixtures was 8%-20% lower than that of the CP mixtures when the nano-$CaCO_3$ concentration was the same.

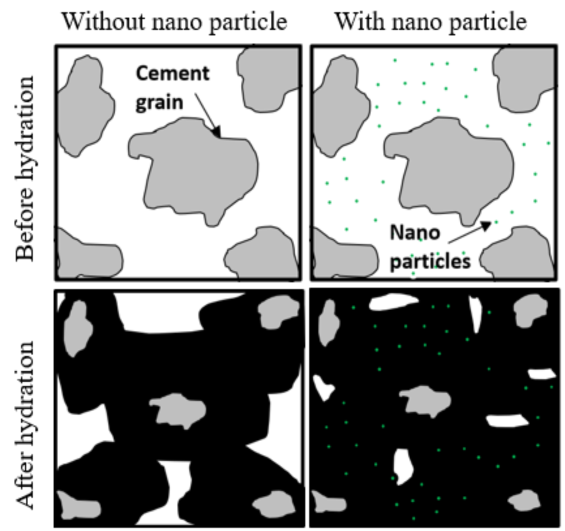




Filler effect



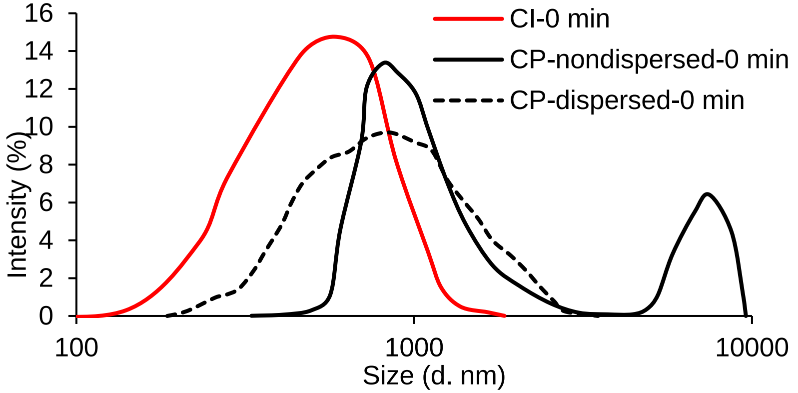
Chung et.al., 2020

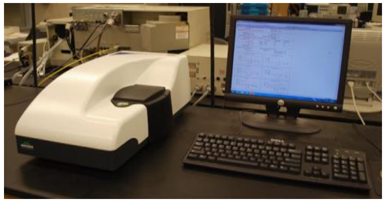


Meng, 2019

Particle size distribution of $CaCO_3$ in suspension

- $CaCO_3$ particles in the suspensions produced by the CI have finer particle size, better dispersion and homogeneity than that produced by the CP method with or without ultrasonic dispersion.

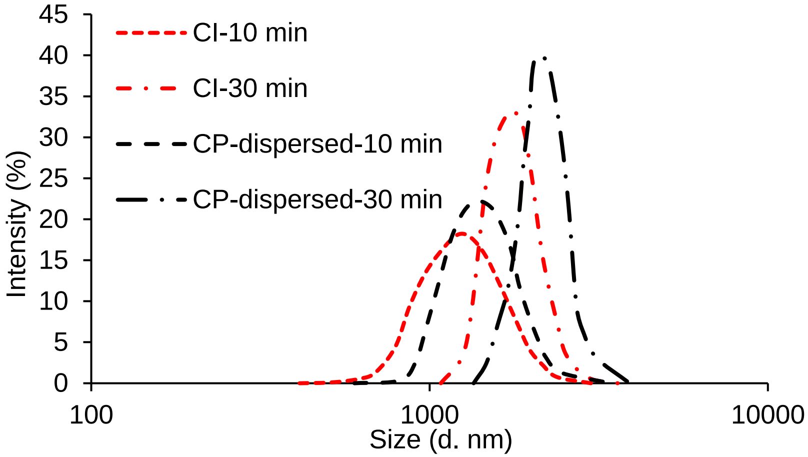




Dynamic light scattering analyzer

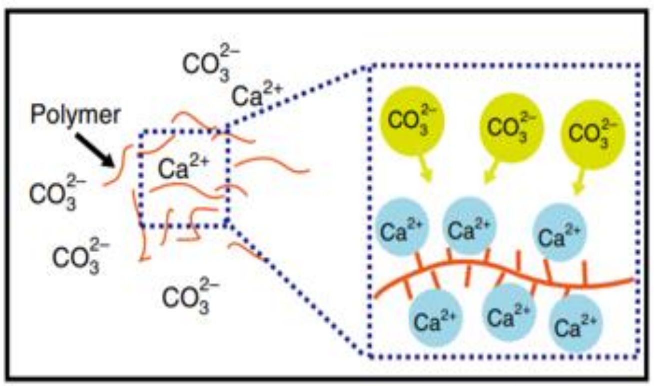
Note: both suspensions were diluted to 0.5%

- $CaCO_3$ particles tend to agglomerate with retention time.

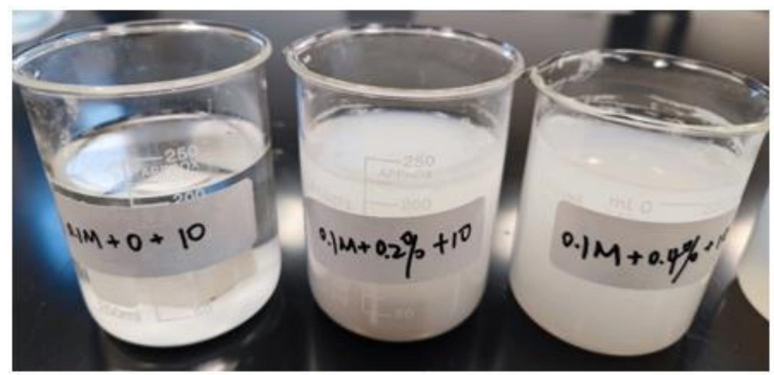


Stabilization of $CaCO_3$ suspension using polymer

- polyacrylic acid (PAA) can be used for stabilizing the $CaCO_3$ suspension for long period as the hydroxy groups on the surface of PAA can interact with calcium ions via ion coupling and thus prevent the agglomeration of $CaCO_3$.
- Preliminary studies showed that the suspension dispersion level is stable in 3 days using 0.2-0.4% PAA.



Xu et.al., 2019

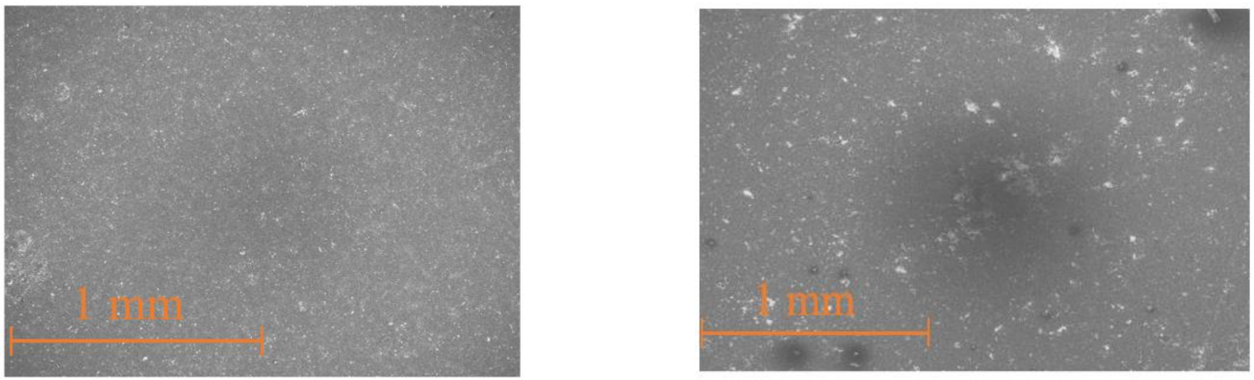


Without PAA   With 0.2% PAA   With 0.4% PAA

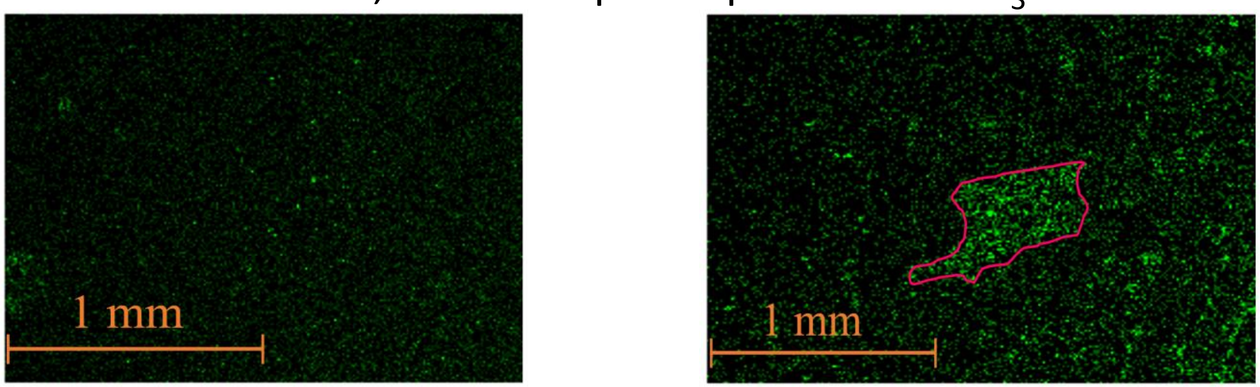
SEM observations of $CaCO_3$ in concrete

- The SEM-EDS images show that the proposed CO_2 injection method can lead to better dispersion of the nano-$CaCO_3$ particles in concrete.

CI – 2% | CP – 2%



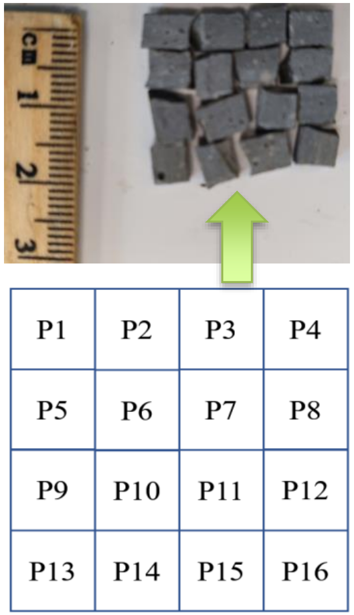
SEM, the white part represents $CaCO_3$

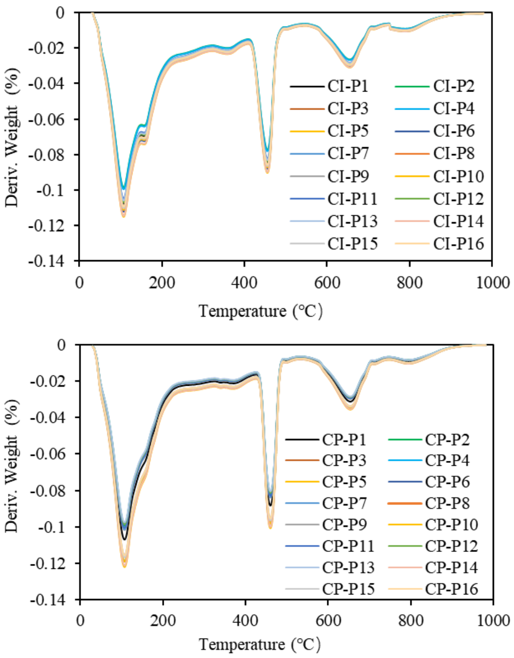


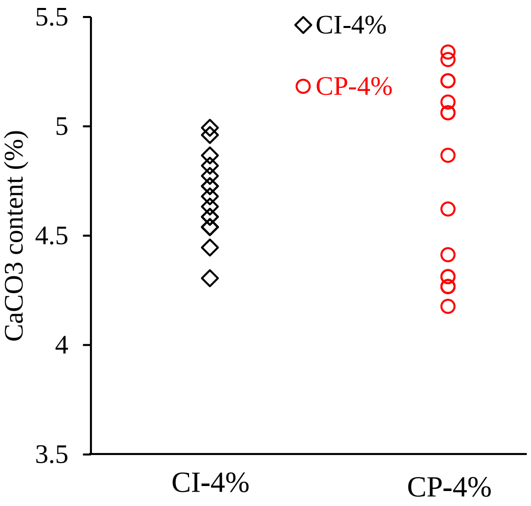
SEM-EDS, Green color represents $CaCO_3$

Dispersion level of $CaCO_3$ in concrete

- To validate the dispersion level of $CaCO_3$, sample was divided into 16 pieces for measuring $CaCO_3$ content using TGA.
- It is observed that the data points for CI-4% sample are less dispersive than those of CP-4% sample, demonstrating that the CI method leads to better $CaCO_3$ dispersion level than the CP method.







Life-cycle carbon footprint

- The carbon footprint of investigated mixtures was calculated using the below equation.

$$C = \sum_{i=1}^{n}(ce_i - cs_i)m_i/\mathrm{Fcu}$$

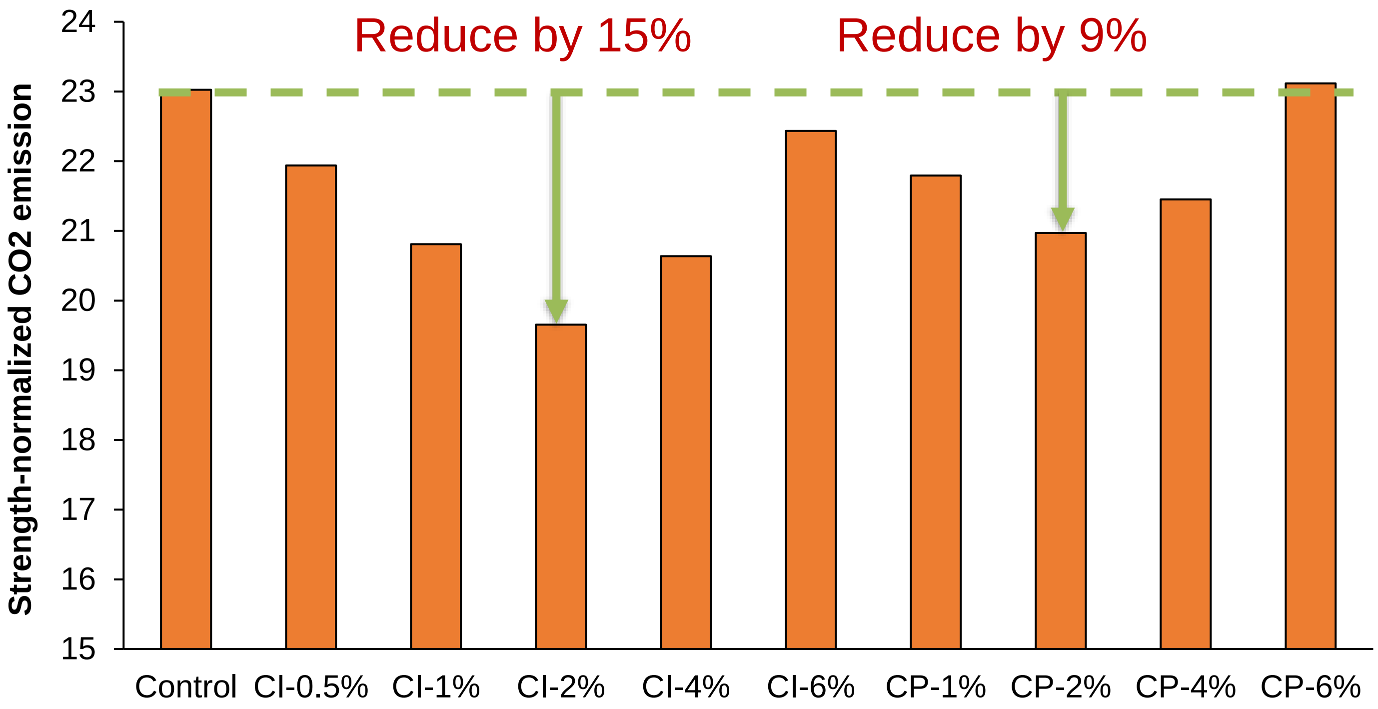
C is the strength normalized carbon emission of a mixture;
ce_i is the unit carbon emission of the i-th ingredient of the mixture (i = 1, 2, 3, ..., n);
cs_i is the unit carbon sequestration of the i-th ingredient of the mixture (i = 1, 2, 3, ..., n);
m_i is the mass of the i-th ingredient of the mixture;
Fcu is the 28-d compressive strength of the mixture.

- The unit carbon emission and carbon sequestration of the raw materials are listed in below table.

No.	Ingredient	Carbon emission (kg/kg)	CO_2 sequestration (kg/kg)
1	Cement	0.83	-
2	HRWR	0.72	-
3	Water	0.00	-
4	Calcium hydroxide	0.683	0.595
5	Nano-$CaCO_3$ powder	0.505	-

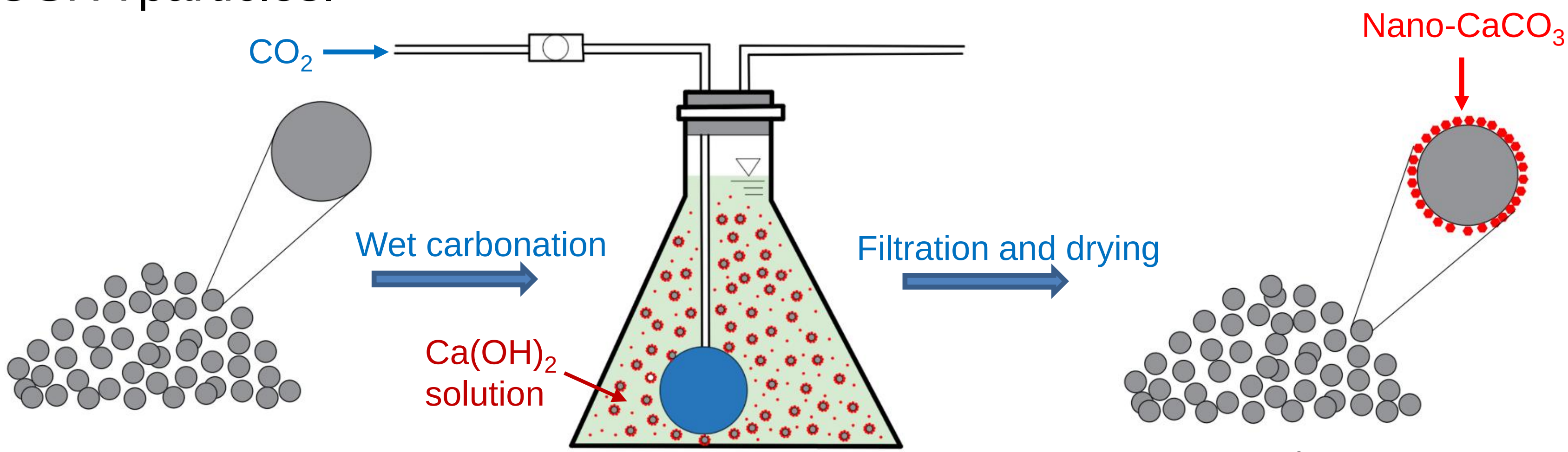
Strength normalized carbon emission

- Life-cycle carbon footprint analysis results showed that the CI method reduces the strength-normalized carbon emission by up to 15%. The reduction rate is 40% higher than the CP method at the same $CaCO_3$ concentration.



Coat wastes with nano-$CaCO_3$ on the surface

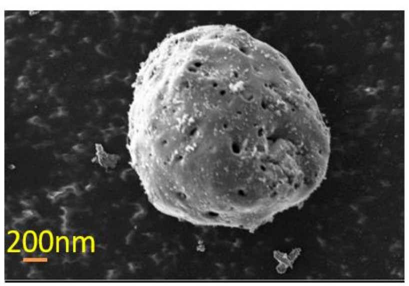
- Procedure: (1) mix off-specification fly ash (OSFA) with $Ca(OH)_2$ solution; (2) inject CO_2 into the mixture; and (3) filtrate the coated OSFA particles.



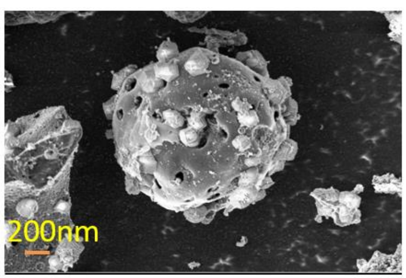
Original OSFA

Coating process

Coated OSFA



Porous (water absorption), low density, low strength



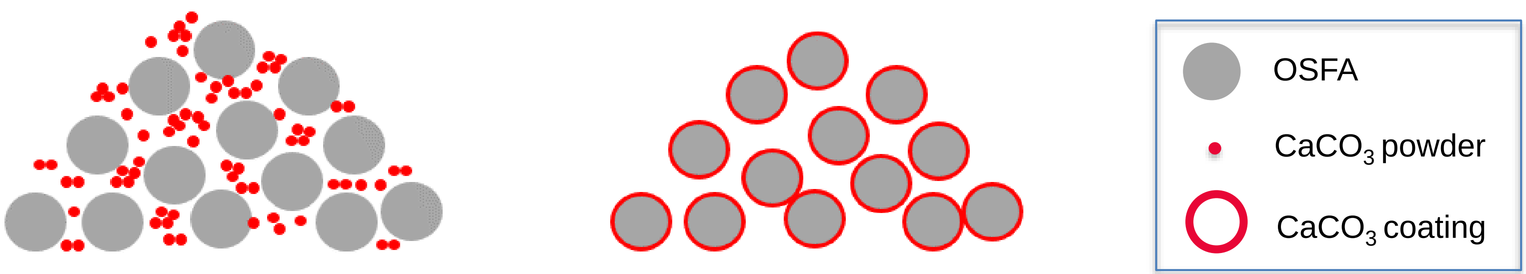
Denser surface, higher density, higher strength

Mixture design

- Four mortar mixtures were designed and tested

Mixtures	Cement	Fly ash	CC	FA@CC	Sand	Water	HRWR
FA0	1	0	0	0	1.00	0.36	0.01
FA20	0.80	0.09	0	0	1.00	0.36	0.01
CC-FA20	0.80	0.09	0.02	0	1.00	0.36	0.01
CC@FA20	0.80	0	0	0.11	1.00	0.36	0.01

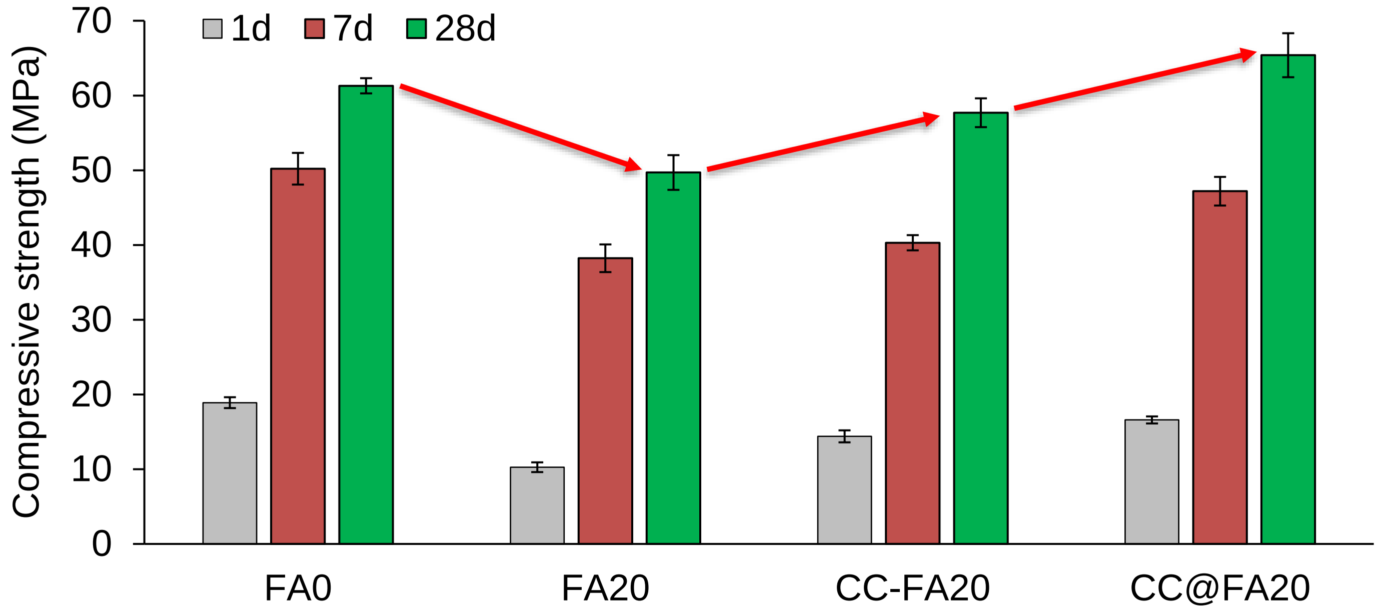
FA0 (the reference or control mixture): without OSFA
FA20: with 20% OSFA (uncoated original OSFA)
CC-FA20: with 20% OSFA and nano-$CaCO_3$ powder
CC@FA20: with 20% OSFA coated by nano-$CaCO_3$



CC-FA CC@FA

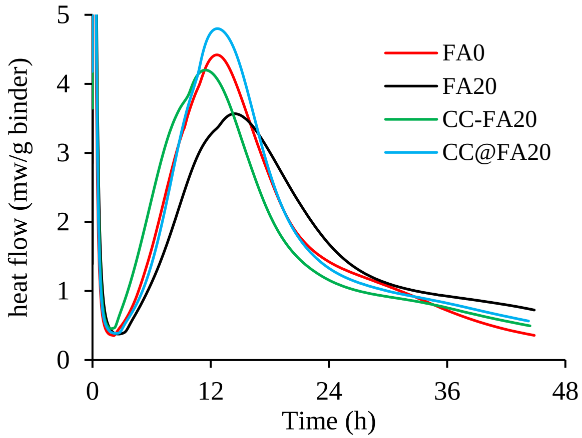
Compressive strength

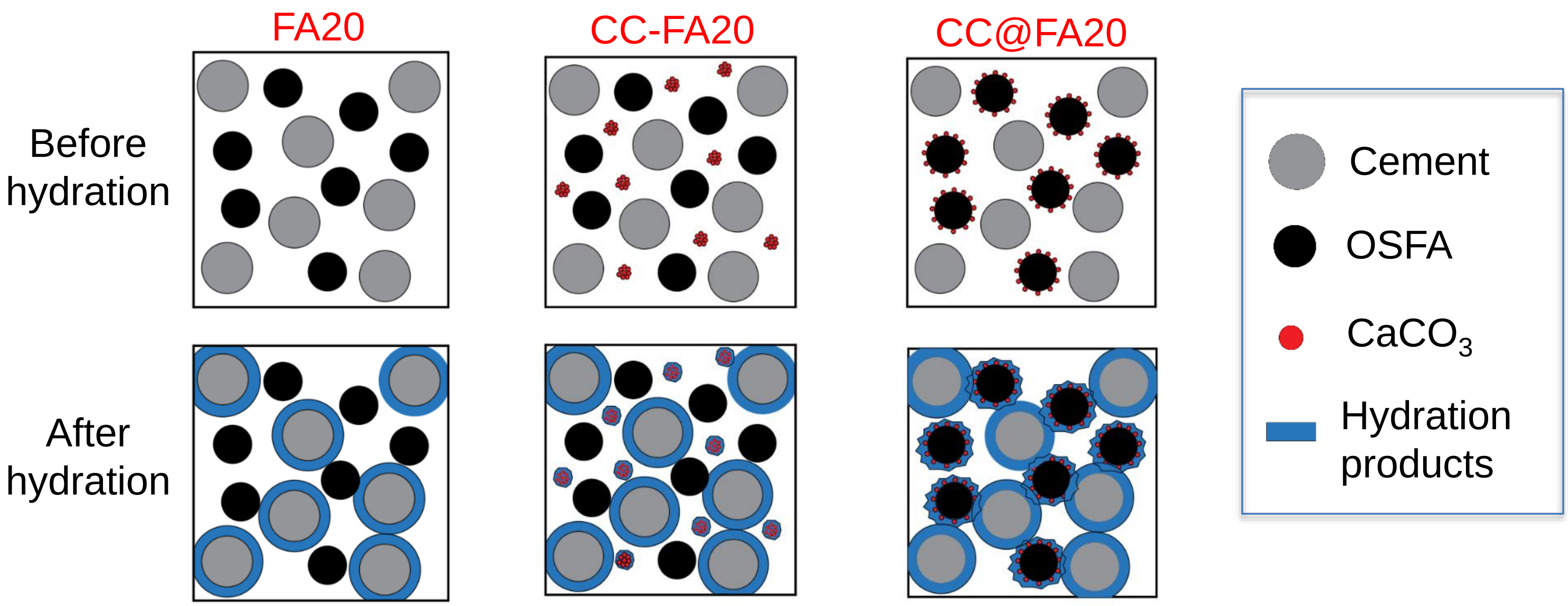
- Compared with the reference mixture (FA0), replacing cement with 20% OSFA (FA20) reduced the 28-day compressive strength by 20%
- Incorporating nano-$CaCO_3$ powder (CC-FA20) enhanced the 28-day compressive strength of mixture FA20 by 16%
- Using the OSFA with the nano-$CaCO_3$ coating (CC@FA20) increased the 28-day compressive strength of mixture FA20 by 32%



Hydration kinetics

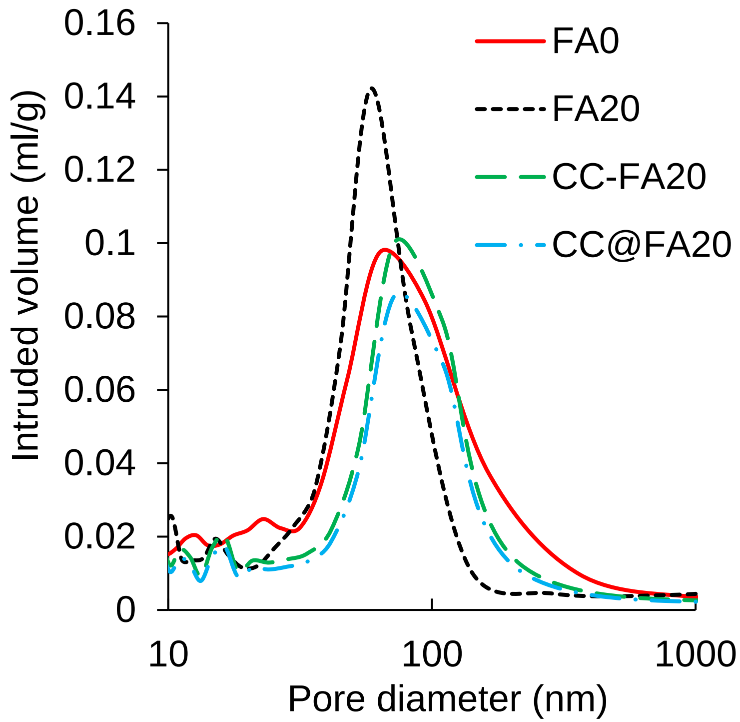
- Direct incorporation of nano-$CaCO_3$ powder accelerated and increased the hydration peak compared with the reference mixture.
- Replacing the nano-$CaCO_3$ powder with the OSFA with the nano-$CaCO_3$ coating highly increased the hydration peak.

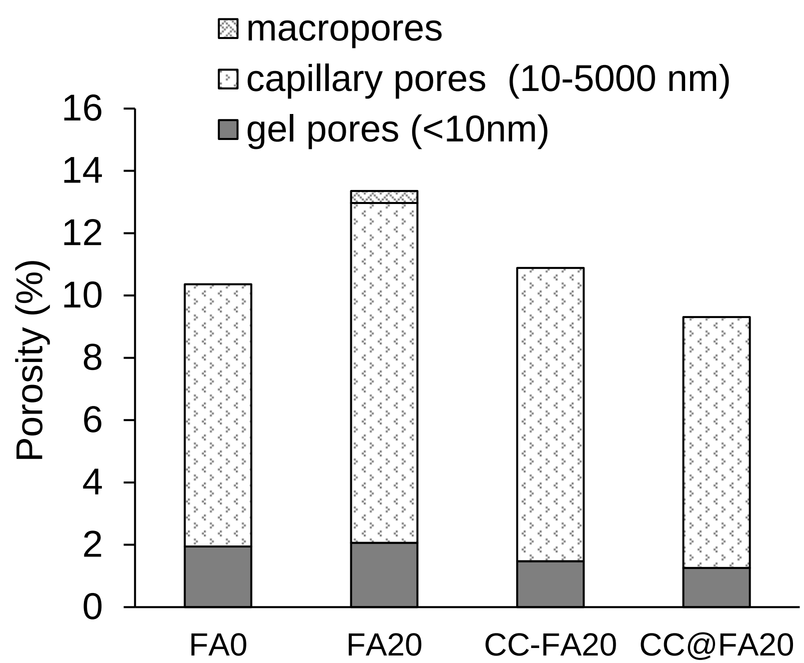




Pore structure

- Compared with the reference mixture (FA0), replacing cement with 20% OSFA (FA20) increased the porosity by 30%
- Using the nano-$CaCO_3$ powder (CC-FA20) reduced the porosity by 17%
- Using the nano-coated OSFA (CC@FA20) reduced the porosity by 26%





Conclusion

- ❑ This research proposed an alternative approach to utilize CO_2 to prepare low-carbon cementitious materials with multiple advantages over the existing approaches:

  - High CO_2 sequestration efficiency and reaction rates
  - Produced uniformly dispersed nano-$CaCO_3$ particles with low energy consumption
  - High mechanical properties
  - Availability for cast-in-place and precast concrete applications

- ❑ Through CO_2 pre-treatment, the porous waste is nano-coated, and using the nano-coated waste in concrete enhanced the mechanical properties. This method is more effective than directly adding nano-$CaCO_3$ powder into concrete.

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Thanks for watching!