Biomolecular Regulated Carbonation Pathway to Process Calcium-Rich Alkaline Industrial Waters Into Supplementary Cementitious Materials (BioCarb)

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Project Overview

– Funding (DOE and Cost Share)
  - DOE: $2,000,000.
  - Cost Share: $500,002.

– Overall Project Performance Dates
  - 07/01/23-06/30/2025.

– Project Participants

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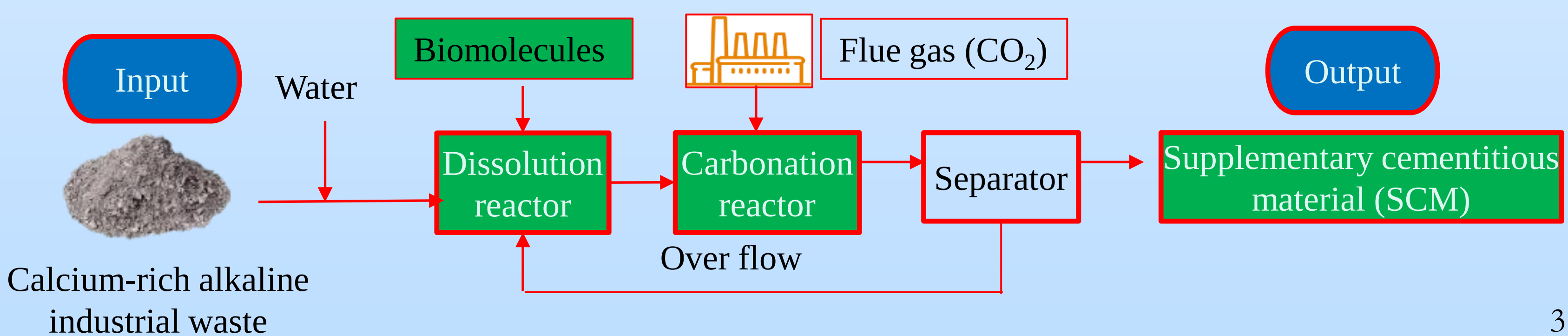
Overall Project Objective

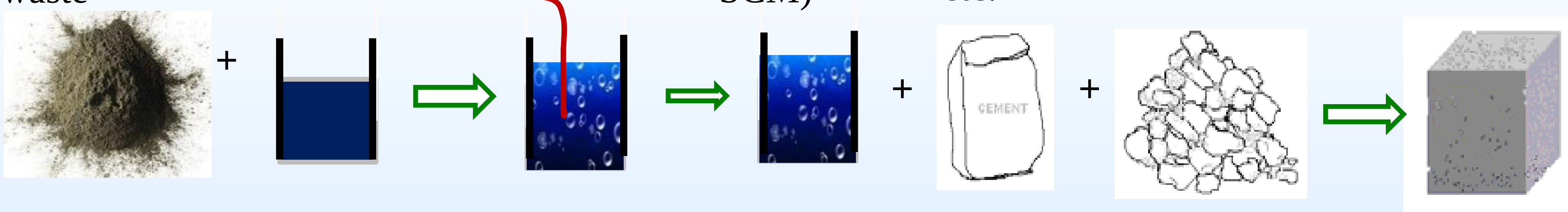
- To test a lab-scale bio-regulated CO_2 mineralization (BioCarb) system that converts calcium-rich, alkaline industrial wastes (recycled cement fine (RCF), cement kiln dust (CKD), high calcium fly ash) into a carbon-negative supplementary cementitious materials (SCMs) and permanently stores CO_2 in the produced SCMs.

- The massive volume of calcium-rich alkaline industrial wastes offers one of the largest sinks for $CO_{2.}$
    - Recycled concrete fines(RCF) (CO_2 uptake > 25%)
    - Class C fly ash (>15% CaO)
    - Cement kiln dust (>50% CaO)



BioCarb Process

Calcium-rich industrial waste | Mixing water + Biomolecule | Carbonating slurry | Carbonated slurry (New SCM) | The rest ingredients of concrete: OPC, aggregate, etc. | Concrete product



I: Carbonating calcium-rich industrial waste with the presence of the biomolecule (BioCarb)

II: Carbonated slurry is directly used as the supplementary cementitious material (SCM) without drying

$3(3CaO \cdot SiO_2) + (3\text{-}x)CO_2 + yH_2O \rightarrow xCaO \cdot SiO_2 \cdot yH_2O + (3\text{-}x)CaCO_3$

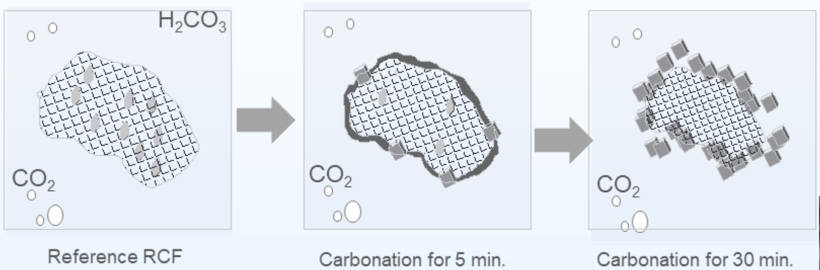
$2(2CaO \cdot SiO_2) + (2\text{-}x)CO2 + yH_2O \rightarrow xCaO \cdot SiO_2 \cdot yH_2O + (2\text{-}x)CaCO_3$

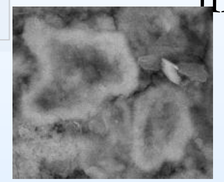
$Ca(OH)_2(s)+CO_2(aq) \rightarrow CaCO_3(s)+H2O(aq)$

$(CaO)x(Al_2O_3)y(SiO_2)(H_2O)z(s)+xCO_2(aq) \rightarrow xCaCO_3(s)+(Al_2O_3)y(SiO_2)(H_2O)l(s)+(z\text{–}l)$
$H_2O(aq)$

- Sequester CO_2 in the carbonated waste
- Activate pozzolanic reactivity
- Disperse produced $CaCO_3$ particles

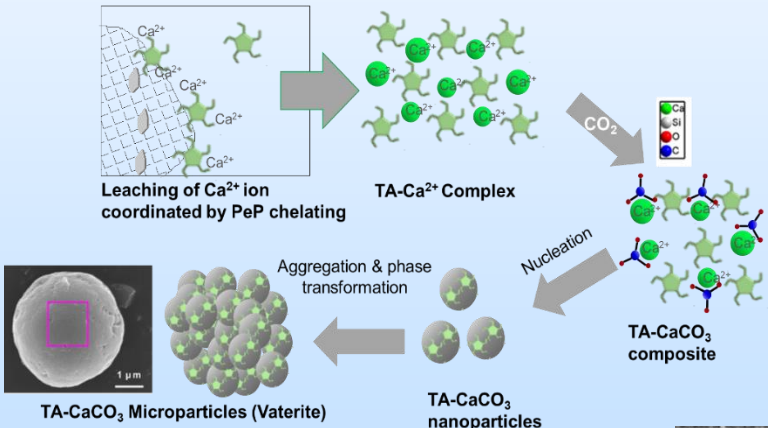
Regulating the formation of $CaCO_3$





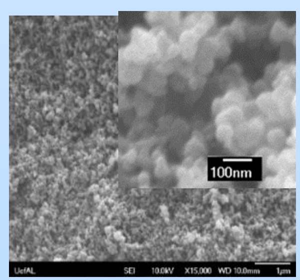
Carbonation of RCF without molecular control:

- The formation of a dense calcite layer limits the Ca^{2+} leaching and CO_2



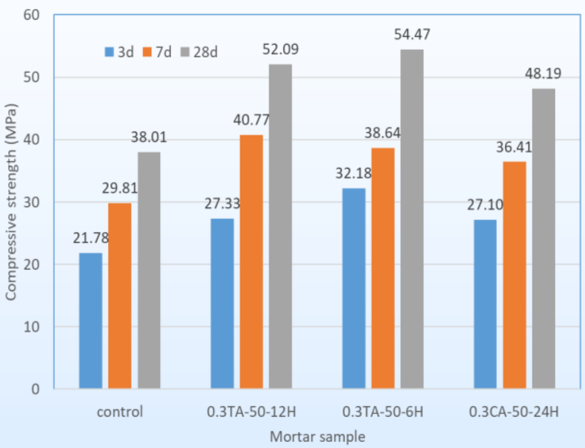
Carbonation of RCF with the biomolecular control:

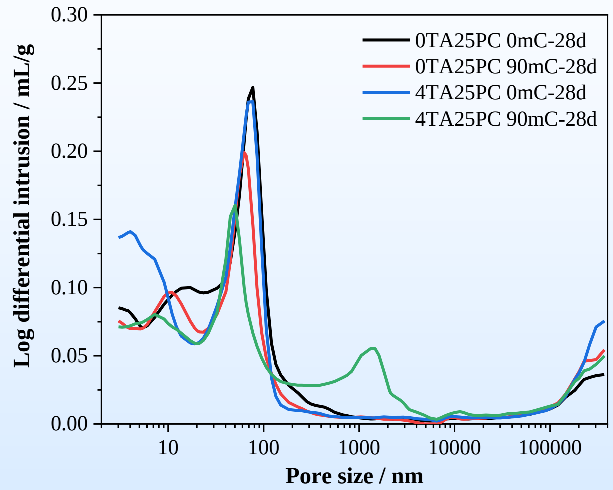
- Chelating of tannic acid to Ca^{2+} to promote the carbonation
- Controlling the morphology and polymorph of $CaCO_3$
- Dispersing $CaCO_3$ particles



Carbonation can be drastically improved with admixtures

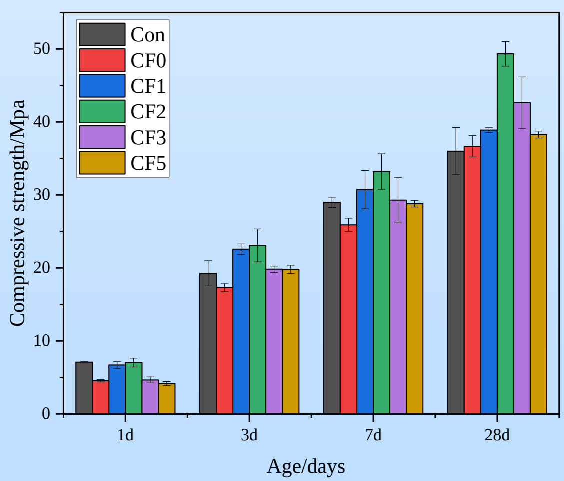
Previous Lab Testing





Initial success was achieved by carbonate cement using the bioCarb process.

- Store over 20lb/yd3 CO2 in concrete
- Enhancing the strength of the concrete over 26%.
- It is critical to keep nanosize CaCO3 particles in the carbonated slurry



Carbonated recycled concrete fine with the bioCarb method exhibits high reactivity.

sampl e	OPC	RCF	Water	sand	TA	Carbonation
Ctrl	31.05	3.45	16.75	50	0	No
CF0	31.05	3.45	16.75	50	0	
CF0.1	31.05	3.45	16.75	50	0.1%cement	
CF0.2	31.05	3.45	16.75	50	0.2%cement	Yes
CF0.3	31.05	3.45	16.75	50	0.3%cement	
CF0.5	31.05	3.45	16.75	50	0.5%cement	

Advantages and Challenges

Advantages:

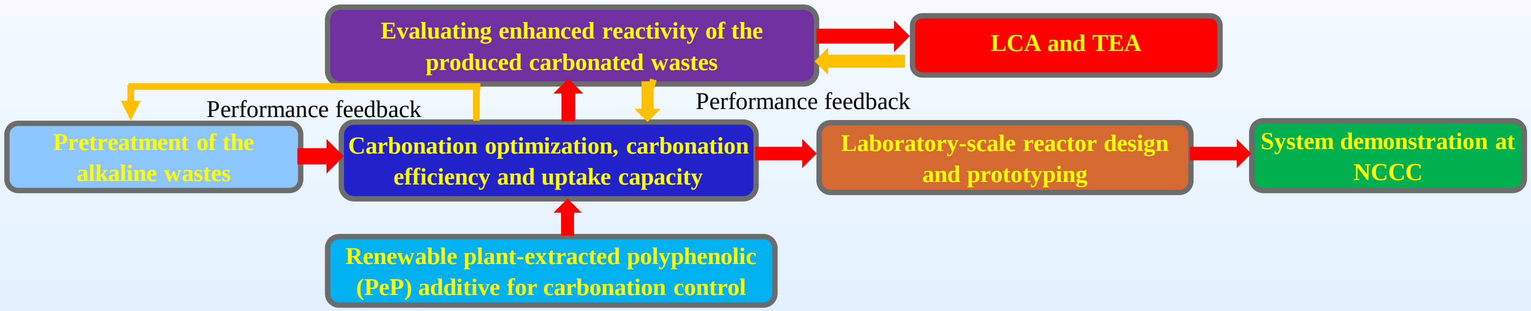
- More CO_2 uptake
- Higher reactivity of the resulting SCM
- Minimum processing energy.

Challenges:

- Variation of feedstock
- Cost of biomolecules
- Loss of workability

Project Scope

- Work plan



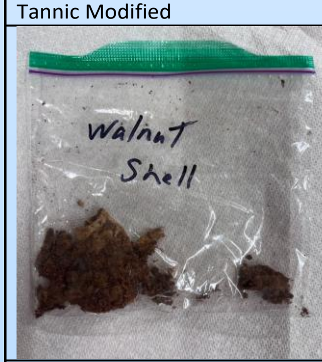
- Schedule

Key milestone	Criterion	Due Date
2	Extract of polyphenols at cost $0.25/lb	18 months after award
4.1	Identify optimal carbonation conditions	12 months after award
4.2	Carbonation efficiency reaches 80% and 50% for RCF and high calcium fly ash.	12 months after award
5.2	Complete reactor prototyping > 10kg/day CO_2 uptake	18 months after award
7	30% less cement can be saved without strength reduction; Carbon-negative concrete > 2500psi.	21 months after award
9	Site demonstration, reaching TRL5.	24 months after the award
Success metric	1) CO_2 fixation >10kg/day; 2) Reducing CO_2 emission of concrete over 200 CO_2 lb/yd^3; 3) Achieve TRL 4-5	24 months after the award

Plant-extracted Polyphenolic (PeP) Additive

a. Table 1 Pep additive produced by Sutterlin Research LLC

Sample	Quantity (grams)	Form
Pecan Hull	3.85	dried
Pecan Leaf	5.42	dried
Pine Bark	4.2	dried
Pecan Bark	5.07	dried
Pecan Shell	13.37	dried
Pine Straw	5.98	dried
Oak Gall	51.15	dried
Oak Bark	4.3	dried
Walnut Shell	4.1	In Process
Coffee	21.92	In Process
Coffee Grounds	14.05	In Process
Gall Powder	15.55	dried
Tea	17.3	In process
Tea Grounds	15.55	In process
Tannic 1	17.35	dried
Tannic 1 modified	15g in 37 ml	wet
Tannic 2	17.28	dried
Tannic Modified	15g in 37 ml	wet



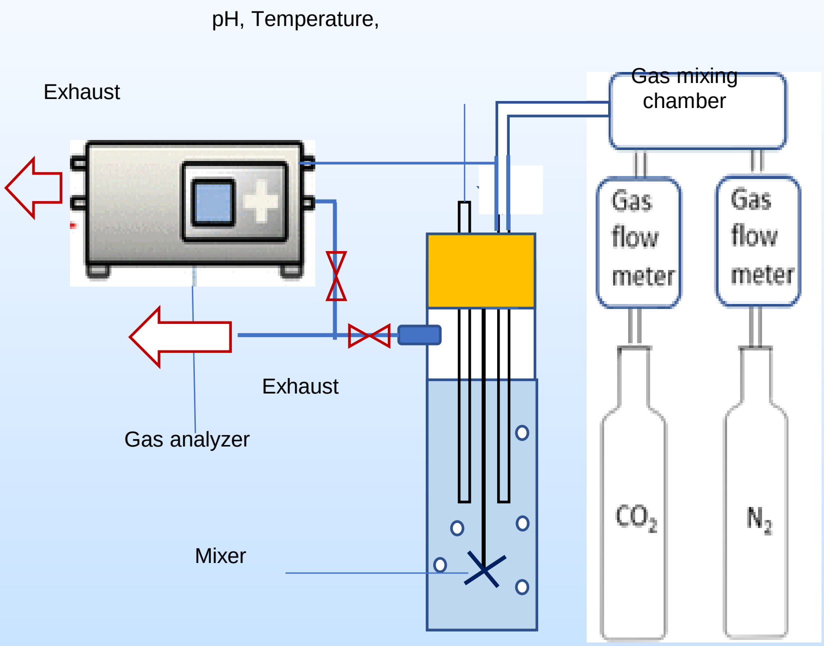
Extract of Walnut Shells

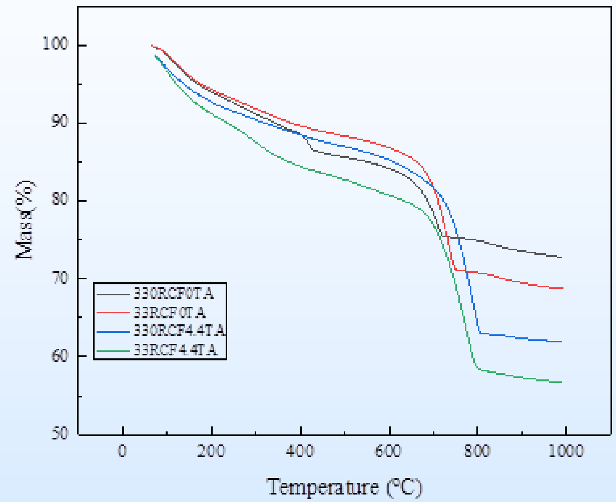
Extract of Spent Tea Leaves

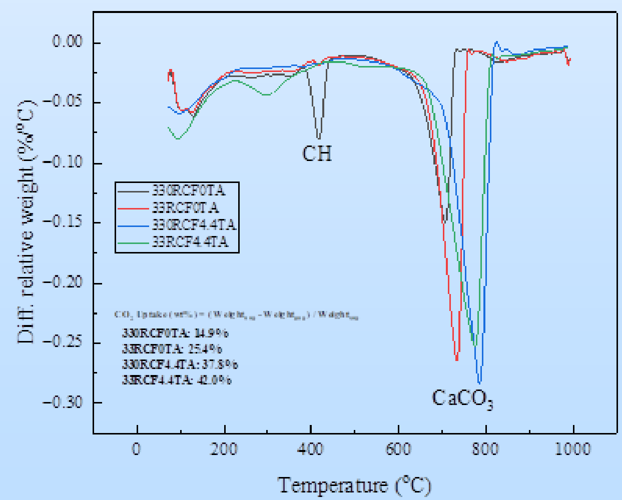
Extract of Spent Coffee

Figure 1. Extractives of three biomass waste materials.

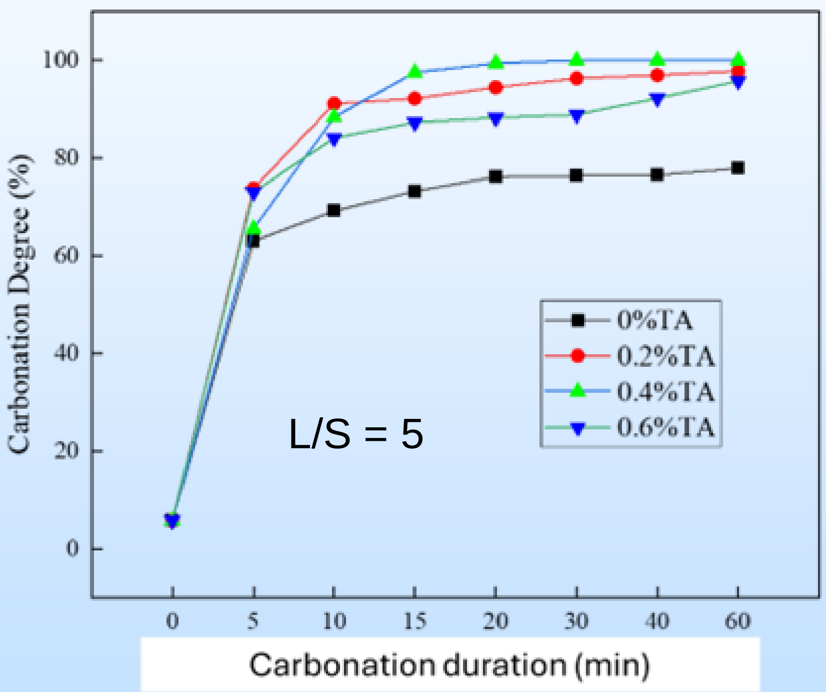
Carbonation Testing





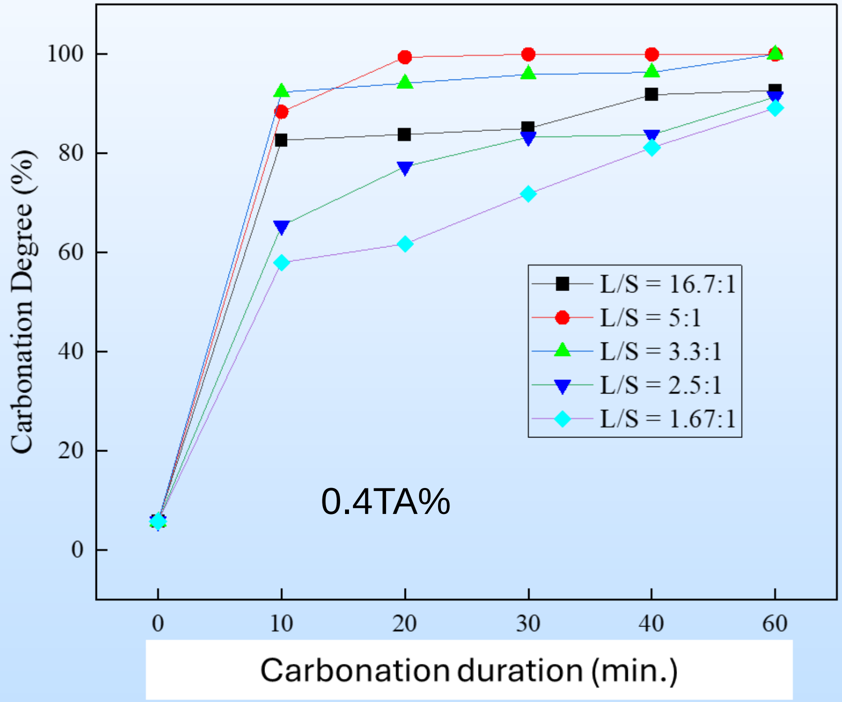


Effect of Additive Dose



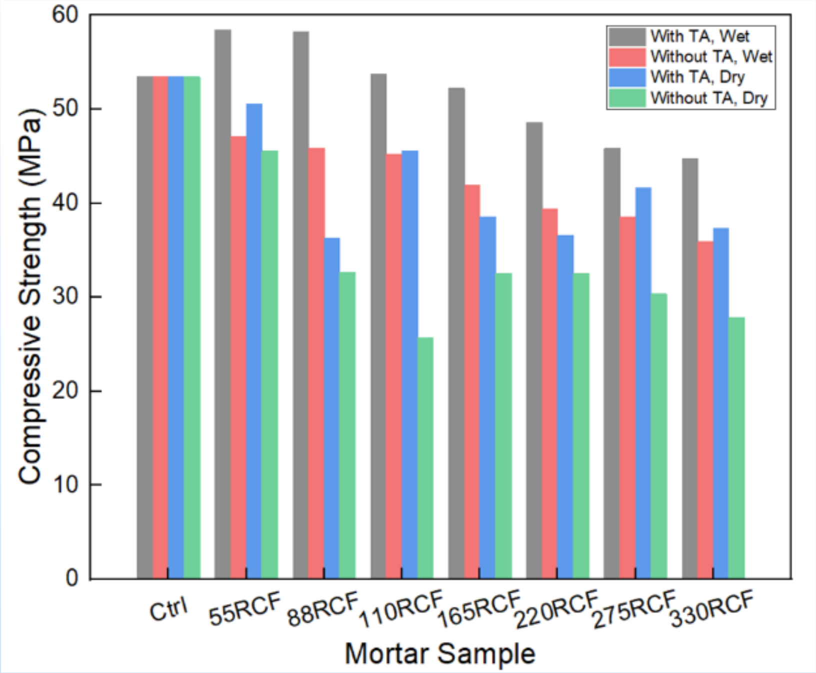
i. Most carbonation can be completed at the first 15min.
ii. TA can slightly slow down the carbonation.
iii. Without TA, more than 20% of RCF can't be carbonated.
iv. Higher dose of TA can increase the carbonation degree of the RCF.
v. However, too much TA may inversely affect the carbonation.
vi. With TA, RCF can be almost completely carbonated.

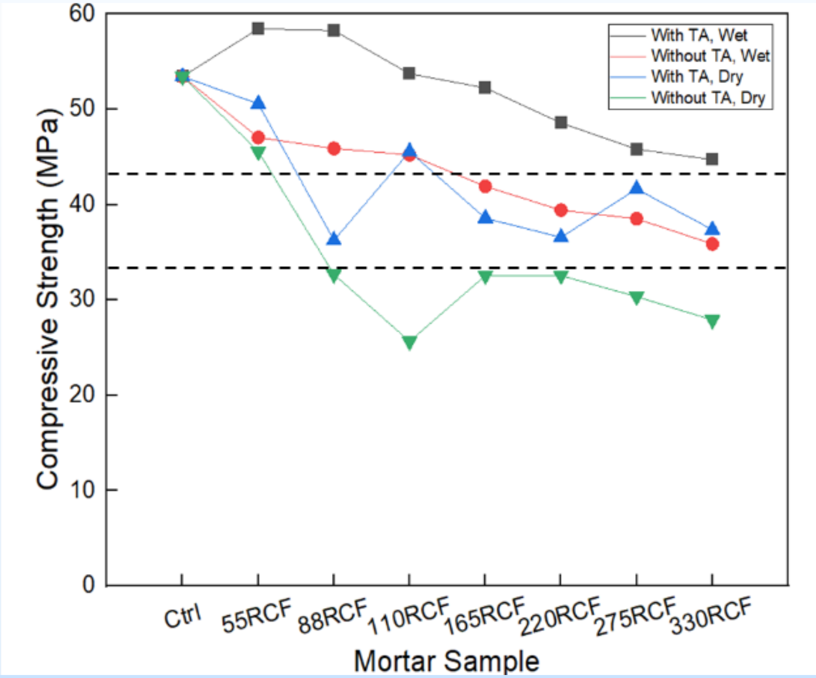
Effect of L/S ratio



i. Less time is needed to complete carbonation with higher L/S ratio.
ii. TA affects the maximum carbonation degree of the RCF.
iii. An optimal dose of TA exists for each different L/S ratio. Under such ratio, maximum carbonation degree can be obtained at a short period of carbonation.

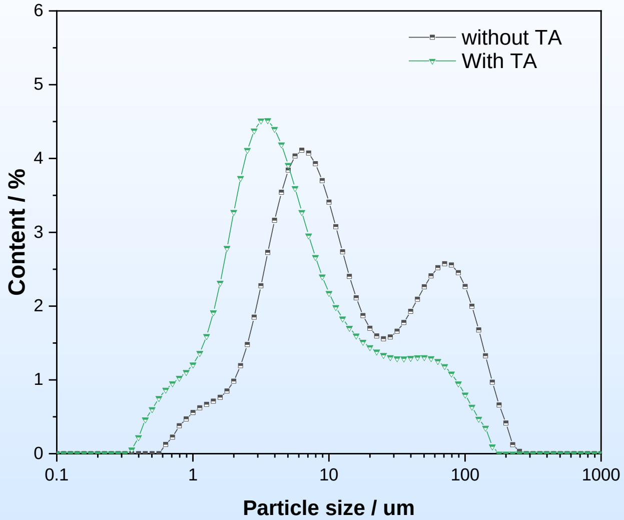
Why Carbonated Slurry?

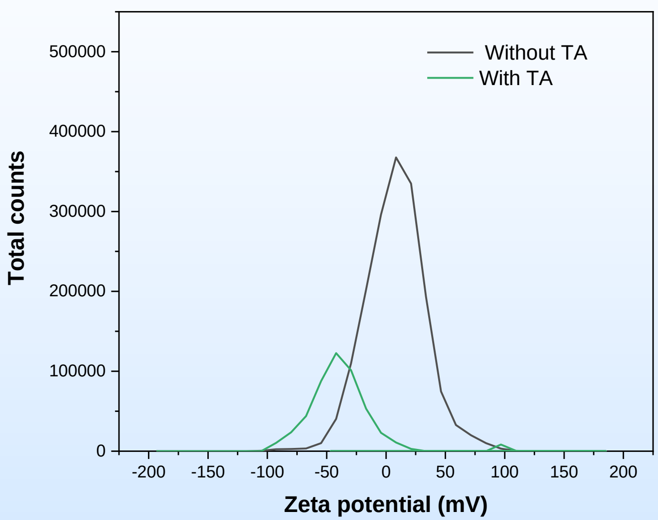


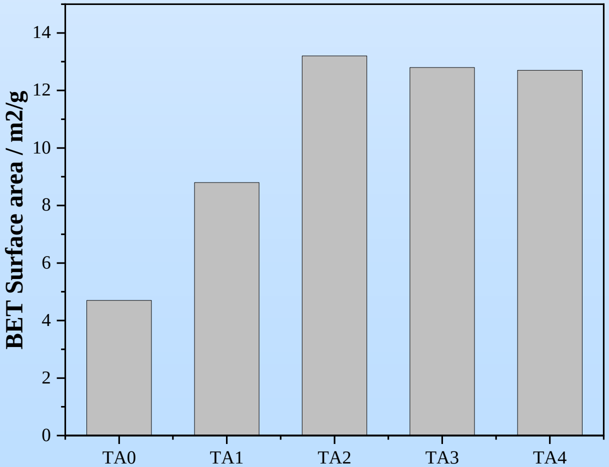


- The presence of TA during carbonation significantly enhances the compressive strengths of cement mortars.
- Directly adding the carbonated RCF slurry without drying drastically enhances the compressive strength of the cement mortar.

Dispersion of $CaCO_3$

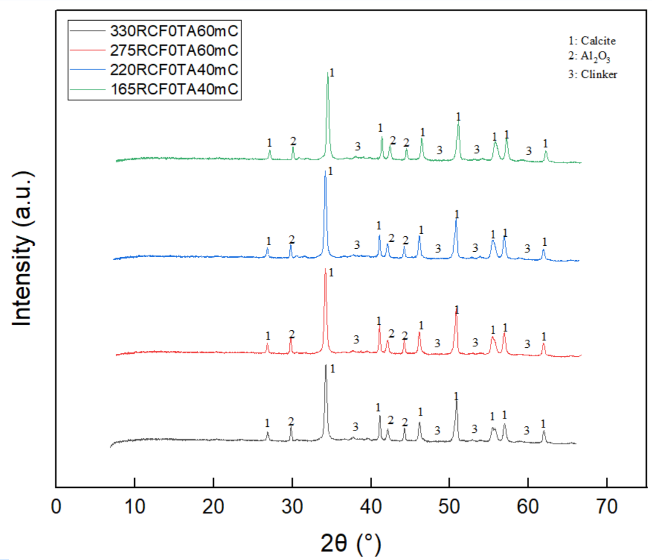


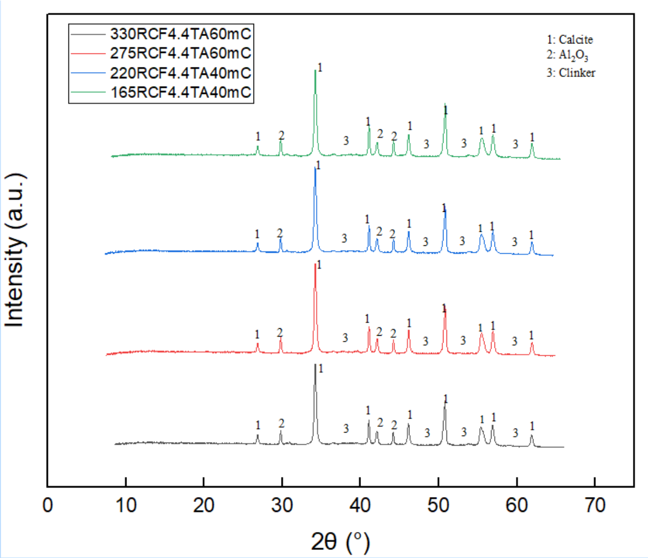


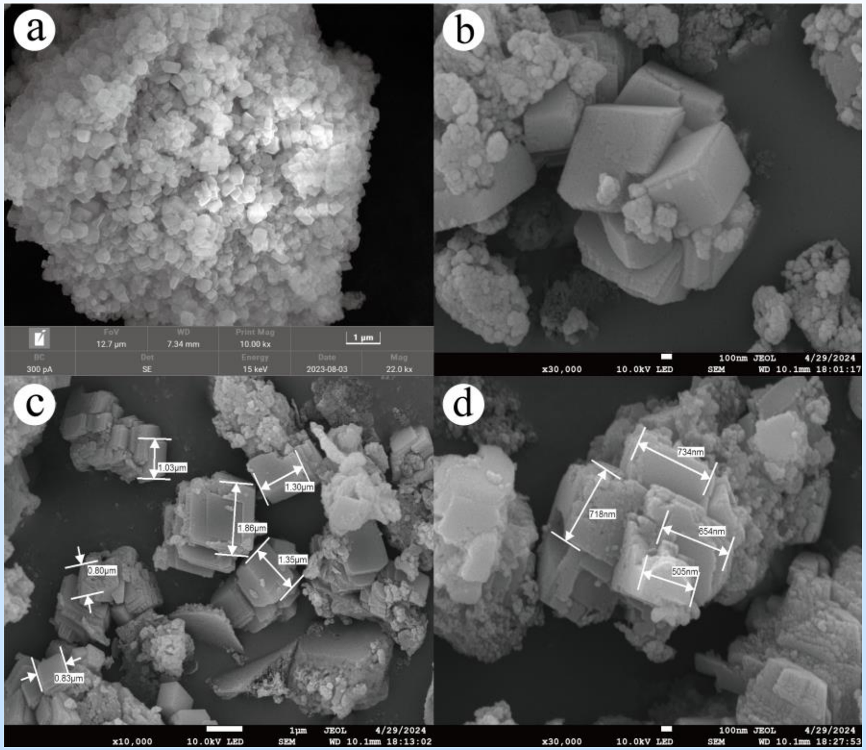


Dispersion of produced $CaCO_3$ particles leads to denser microstructure.

Carbonation Products







Lessons Learned

- Workability of concrete can be reduced by the carbonation. This can be mitigated by using water reducer.
- Over carbonation may reduce the performance of the produced concrete.
- A low L/S ratio reduces water demand; however, a longer carbonation duration is needed.

Plans for future testing/commercialization

a. Testing the efficacy of PeP
b. Co-Carbonation of class C fly ash and cement kiln dust
c. Scale up to absorb at 10kg CO_2/day
d. 30% replacement of OPC with the SCM without loss of strength.
e. Demonstration at NCCC.
f. Commercialization is undergoing supported by an NSF ART project.

Summary

- Biomolecules can promote CO_2 uptake of calcium-rich industrial wastes.
- Recycled concrete fines can be completely carbonated by the BioCarb process.
- Most carbonation can be completed within 30 min in aqueous condition.
- Higher strength of cement mortars is reached by wet carbonated slurry.
- Recycled concrete fine provides sustainable supply of Ca for CO_2 mineralization.