



Converting CO₂ and Alkaline Solid Wastes into Carbon-Negative Supplementary Cementitious Materials for Co-decarbonization of Multiple Sectors

(DE-FE0032395)

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2024 FECM/NETL Carbon Management Research Project Review Meeting
August 5 – 9, 2024



Outline

- Project Overview**
- Technology Background
- Project Scope
- Progress and Current Status
- Community Benefits
- Lessons Learned
- Plan for Future Development and Commercialization
- Summary



Project Overview

Overview

Title: Converting CO₂ and Alkaline Solid Wastes into **Carbon-Negative Supplementary Cementitious Materials (SCMs)** for Co-decarbonization of Multiple Sectors

Performance Period: 10/1/2023 – 9/30/2025

Total Funding:

DOE: \$2,000,000

Cost share: \$500,000



Project Overview

Team: Missouri S&T

- **STEM-intensive public research university**, founded in 1870, located in Rolla, MO
- **Campus and colleges**: 3 colleges; known primarily for the engineering programs
- **Fast developing innovation/education ecosystem**: Protoplex, Innovation Lab, STEM Center, BioX, etc., enabled by a \$300M gift and state investment
- **Strong support for research facilities**:
 - Materials Research Center
 - Center for Research in Energy and Environment
 - Center for Infrastructure Engineering Studies
 - *Advanced Materials for Sustainable Infrastructure* as a signature research area



Hongyan Ma

Associate Professor of Civil/Materials Engineering
Director, Laboratory of Future Cements and Carbon-Negative Initiatives (**FuCCI**)

Lab Space: 2,000 ft²

Expertise: Carbon-efficient and carbon-negative cement and concrete; functional construction materials; multi-scale modeling; energy storage...



Aditya Kumar

Associate Prof. of Materials Science and Engineering

Expertise: Sustainable cements; thermodynamics and kinetic modeling; machine learning



Mahelet Fikru

Associate Prof. of Economics

Expertise: Techno-economic analysis; economics modeling; optimization



Wenyu Liao

Assistant Research Professor of Civil Engineering
Manager, **FuCCI**

Expertise: Characterization of cement and solid wastes; concrete testing; carbon-negative innovation

Project Overview

Team: Lawrence Livermore National Lab

- **DOE/NNSA lab**,
founded in 1952, located in Bay Area, CA
- **Campus**: 2 sites; known for energy science & technology; open campus to academia & industry
- **Large innovation ecosystem**: Open campus; in vicinity to Berkeley, Stanford, MSIs; research programs with UC (e.g., Merced) & Livermore Lab Foundation
- **Strong supports on research & development**:
 - Advanced Manufacturing Lab
 - *Carbon Initiative*
 - High performance computing
 - *Getting to Neutral Report, etc.*



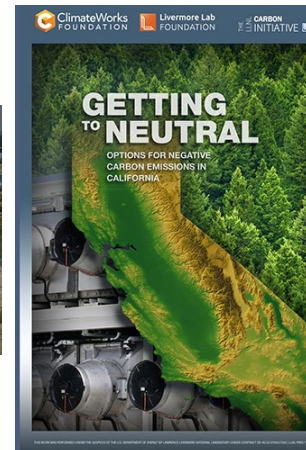
Jiaqi Li

Research Scientist

Porous media group: OpenLCA, SimaPro, GreenConcrete LCA; carbon analyzer, TGA-MS, etc.

Expertise: Carbon-efficient cement and concrete; advanced material characterization; life-cycle assessment

 Lawrence Livermore
National Laboratory



Project Overview

Overall project objectives

To develop a transformational process to convert CO₂ and alkaline solid wastes into carbon-negative blended SCMs to meet the demand of cement/concrete market, as well as co-decarbonize and economically benefit multiple industrial sectors

- CO₂/flue gases
- Alkaline solid wastes (steel slags, off-spec coal ashes, MSWI ashes, ...)
- Carbon-negative blended supplementary cementitious materials (SCMs)
- SCMs: substitution of portland cement (>50%)
- Multiple industrial sectors (e.g., cement manufacturing, power generation and steel making, which generate both CO₂ and solid wastes)



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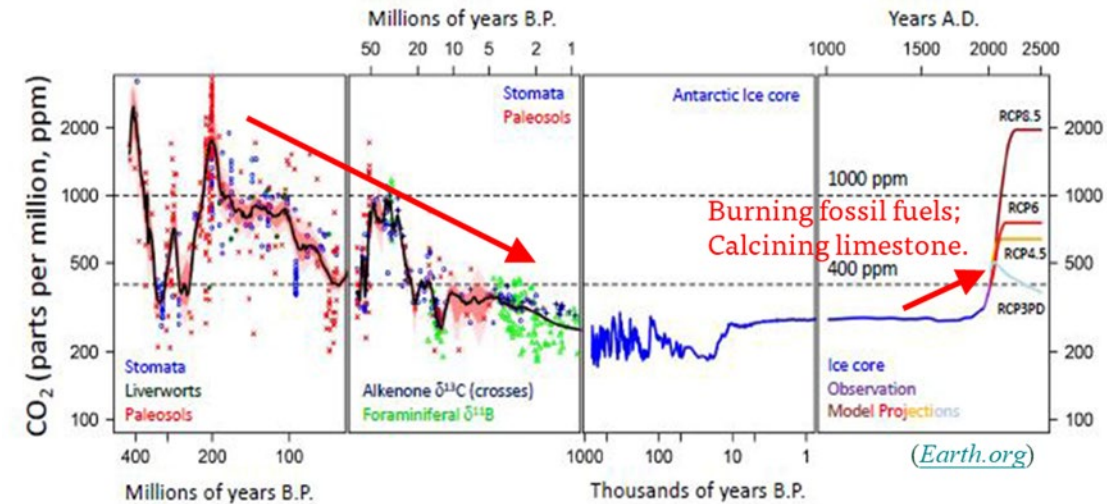


Technology Background

Why carbon-negative?

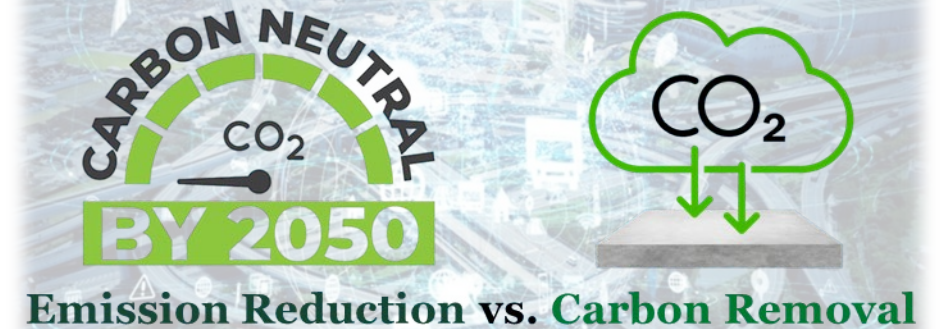
Driving Force of Climate Change:

- **422 ppm** in 2023 → **2-3°C** higher temperature and **> 6m** higher sea levels (90% reliability)
(Foster & Rohling, 2013)
- Warming effects may emerge after a **10-to-30** years lag
(Ricke & Caldeira, 2014)



Immediate Actions?

- Emission Reduction and Carbon Neutrality: necessary but not enough
 - **Carbon Negativity**: reduce CO₂ concentration (ppm)
- i.e.: utilize and/or permanently store captured CO₂**



Technology Background

Why SCMs?

Decarbonization of cement/concrete

Need?

- Annual production: 4.5 Gt cement; >30 Gt concrete
- ~10% of anthropogenic CO₂ emission

Challenges

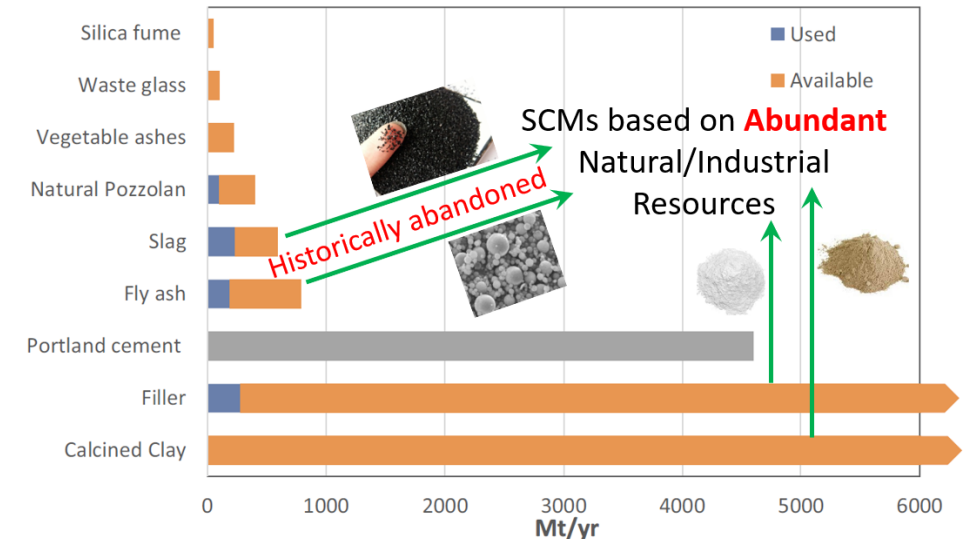
- Reduce CO₂ emission by 24% by 2050 (IEA 2018)
- Achieve net zero by 2050 (PCA/GCCA, 2021/2022)

Possibilities

- Increasing use of carbon-efficient **SCMs**
- Improving **efficiency of cement** production/utilization
- Developing sustainable **alternative cements**

Limitation of resources

- **Class C/F fly ash?** GGBS? Calcined clay?
- **How about off-specification industrial wastes?**



[UN Environment, Scrivener et al., 2018, Cement and Concrete Research]



[www.lc3.ch]



Technology Background

Why (off-specification) alkaline solid wastes?



Off-spec **coal combustion** residues (2.5 Gt + 20 Mt/year)



Slags (steel, copper, lead, etc.; >15 Mt/year)



Waste-to-Energy (**WtE**) residues (>10 Mt/year)



Recycled concrete



Cement kiln dust



Mine tailings/waste rock



Waste glass



Sludge and Dirt



C&D wastes

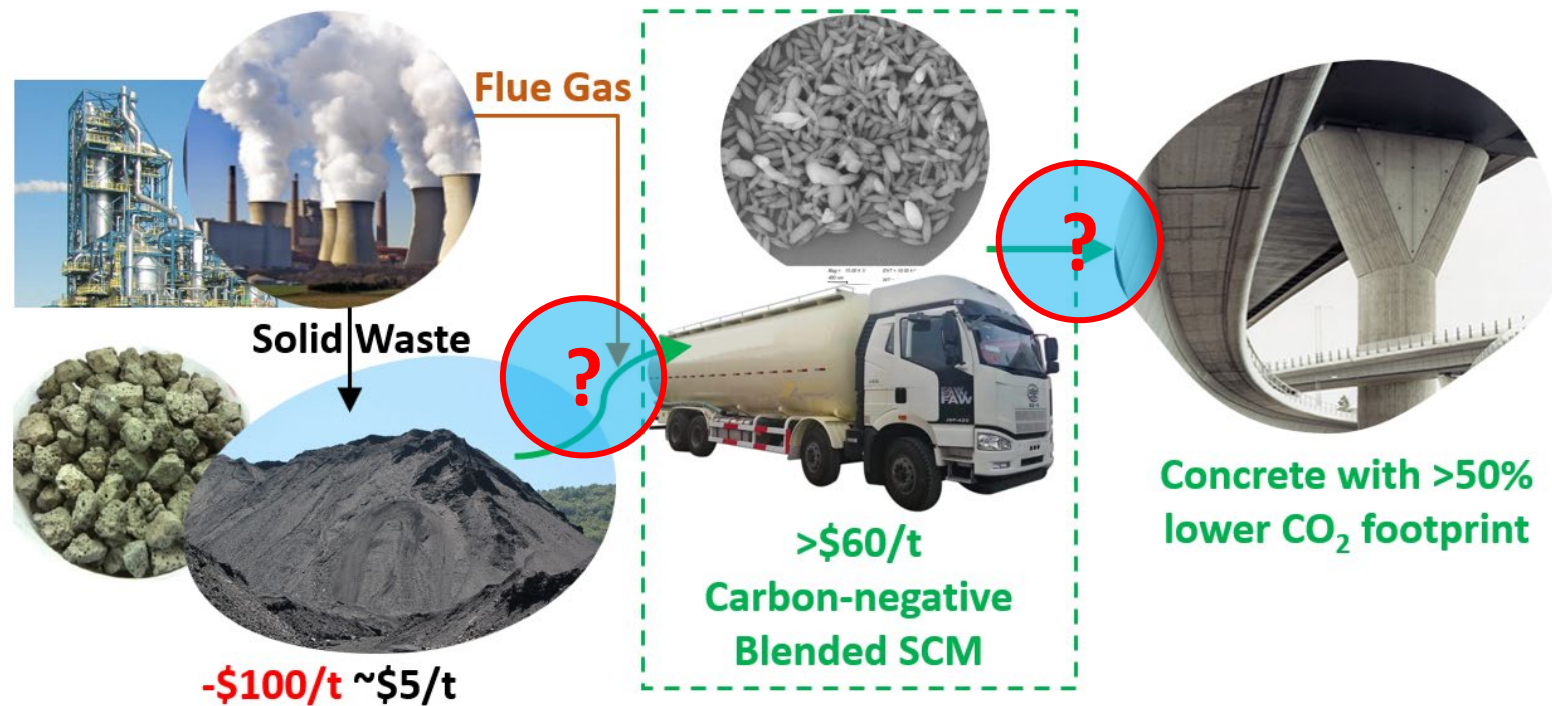


Technology Background

The Technology

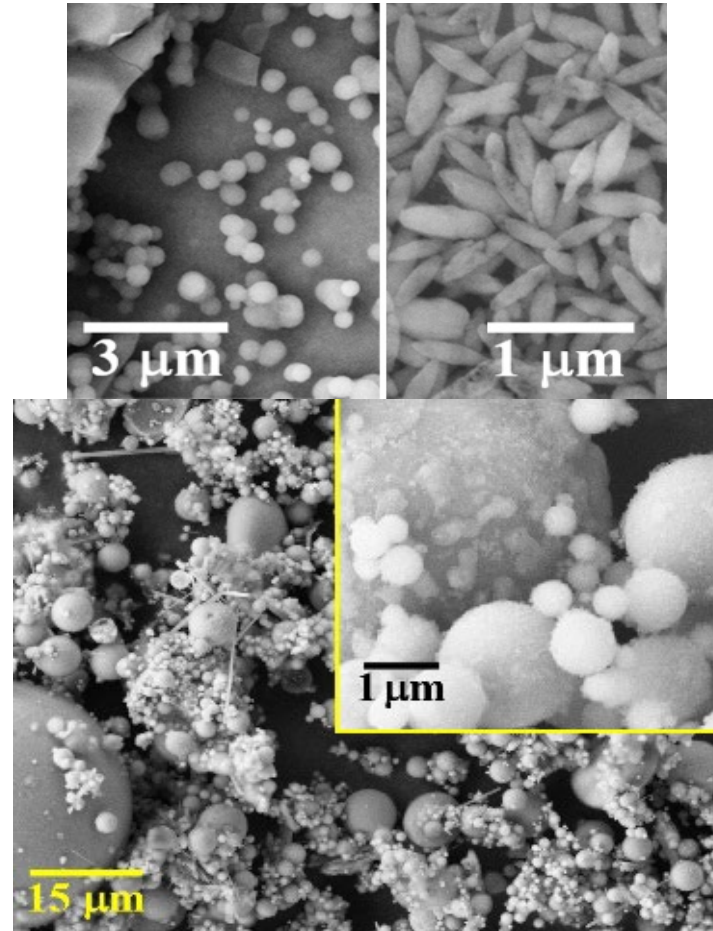
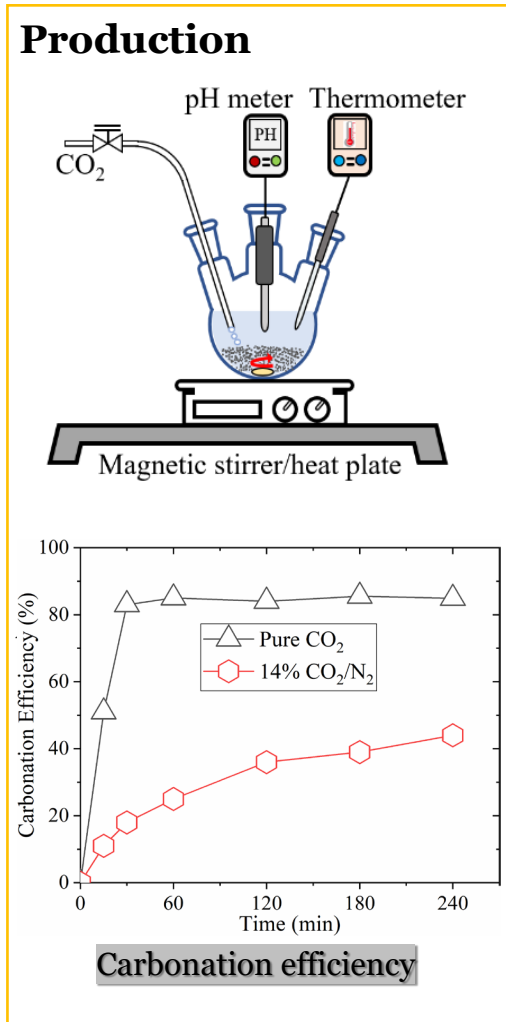
Use **CO₂** to **upcycle** alkali aluminosilicate **solid wastes** into **carbon-negative blended SCMs**, which can substitute **part** of cement without compromising the performance of concrete

The product: nano-/micro-carbonates-aluminossilicate (**nCAS**)



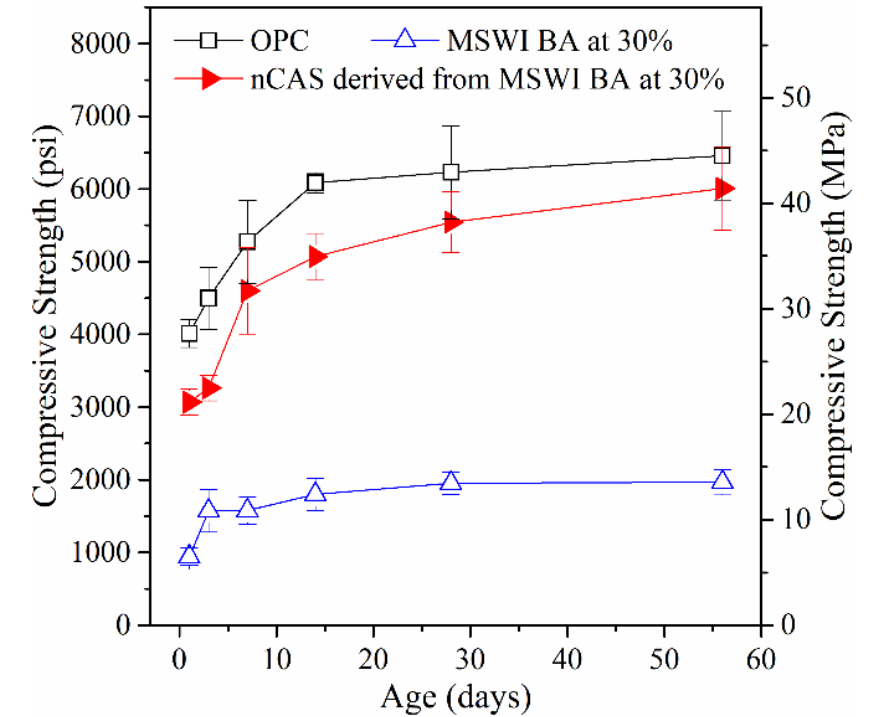
Background and Project Scope

The Technology: carbon-negative process



Carbonate nano-/submicron-particles formed following carbonation

Adaptive to low-quality feedstock?



MSWI bottom ash (BA):

Low reactivity?

Metallic aluminum?

Other detrimental substances?

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Project Scope

Steps and work plan

BP1

Task 1: Project Management and Planning

Task 2: Process model development and initial TEA and LCA

Task 3: Solid wastes sample collection and detailed characterization

Task 4: Optimization of carbonation route 1

Task 5: Optimization of carbonation route 2

Task 6: Tests of nCAS and nCAS-incorporated cement pastes

Task 7: Approach down-selection and final optimization

BP2

Task 8: Scale up production to achieve no less than 10 Kg CO₂ converted/day

Task 9: Standard performance tests of nCAS-incorporated cement-based materials

Task 10: Final TEA and LCA



Project Scope

Milestones/successful criteria

BP	Date	Success Criteria
BP1	9/30/2024	<ol style="list-style-type: none"> 1) Comprehensive database of alkaline solid wastes encompassing up to 200 samples from diverse sources; 2) Achieving CaO/MgO-to-carbonate conversion rate >65%; 3) Machine learning model trained and validated, capable of predicting carbonation efficiency with >90% confidence; 4) Cement paste (water-to-binder ratio=0.4) with ≥50% nCAS (containing ≥15% CO₂) substitution achieving 28-day compressive strength >40 MPa; 5) Supporting graduate students and postdoctoral researchers from underrepresented groups.



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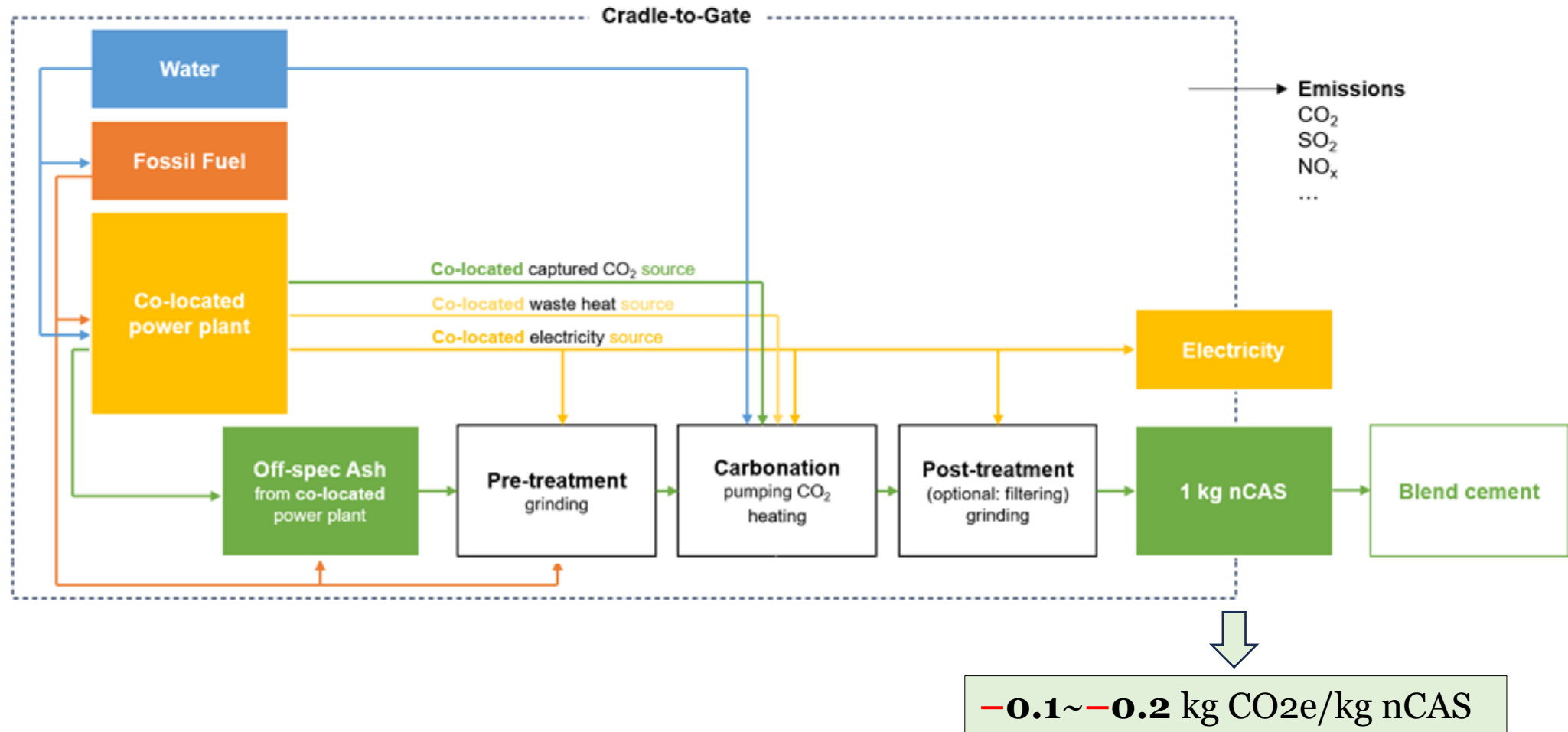


Progress and Current Status

Process model and preliminary LCA



□ Base scenario

Off-spec coal ash | co-located coal power plant (heat, electricity & CO₂ source) for carbonation



Progress and Current Status

Solid waste sampling and characterization

MSWI ashes						
	(a) Filter cake	(b) Small Aggregate	(c) Sand	(d) Filter cake	(e) Bottom Ash	(f) Fly ash
Coal ash						
	(g) C-Fly ash	(h) F-Fly ash	(i) Boral ash	(j) Bowen ash	(k) Harvested ash	
Steel slags & mine waste						
	(l) GGBFS	(m) LMF	(n) EAF	(o) RM	(p) Pb-Zn Tailing	

Municipal solid waste incineration (MSWI) ashes

Coal ashes

Steel slag

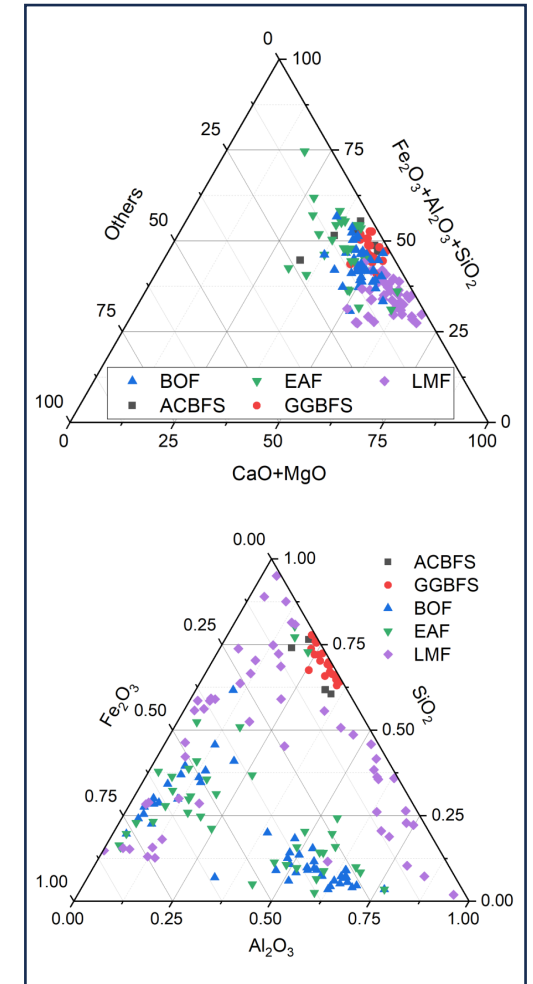
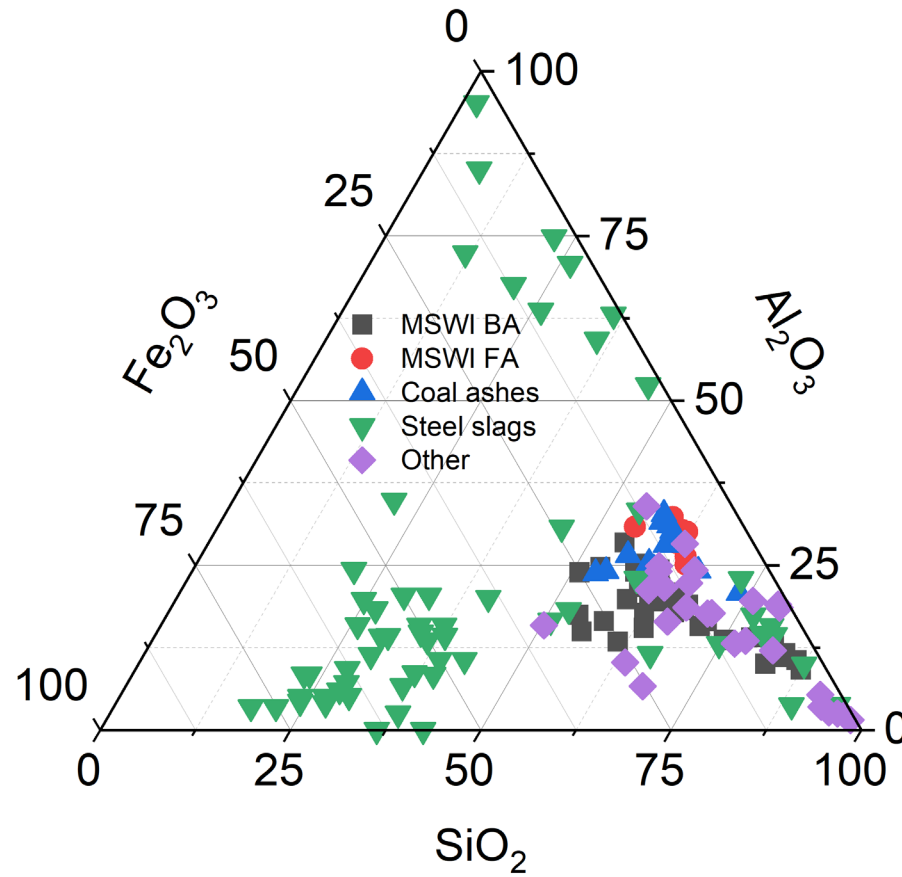
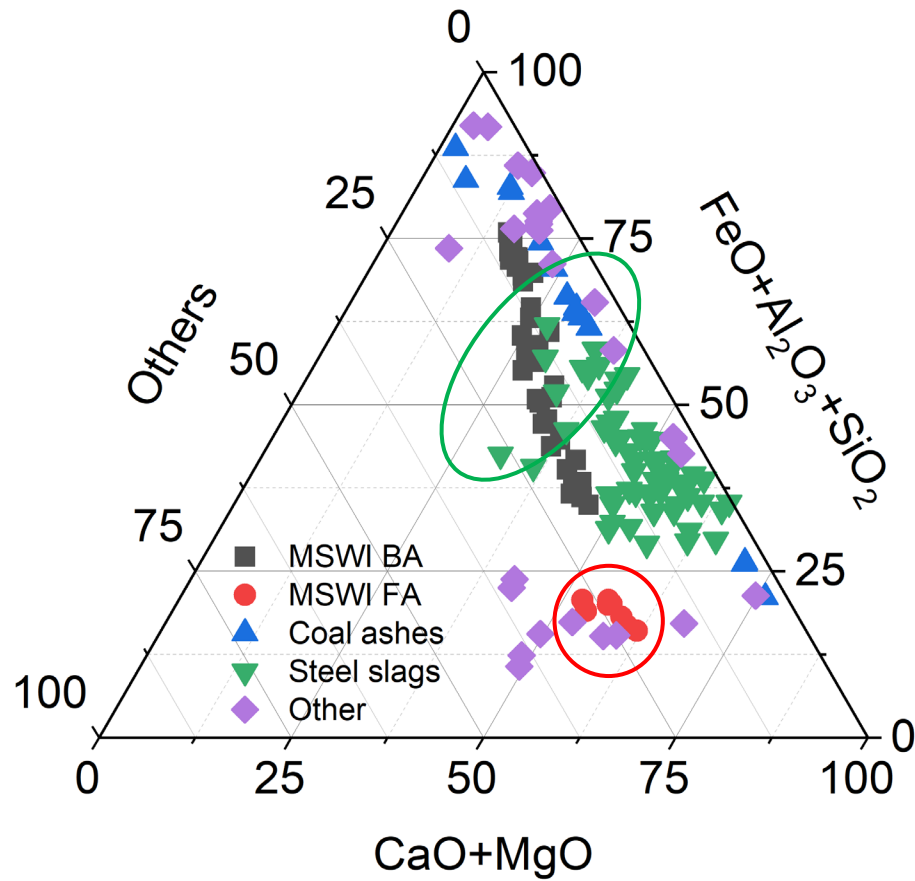
General

- York County Solid Waste & Refuse Authority
- Covanta/**Reworld**
- ...
- Electric Power Research Institute
- Local concrete producers
- ...
- SSAB
- Cleveland-Cliffs
- Harsco
- Reserve Management Group
- ...
- Continental Cement
- Ash Grove
- Quapaw Tribe, OK
- ...



Progress and Current Status

Solid waste sampling and characterization: Compositional variation

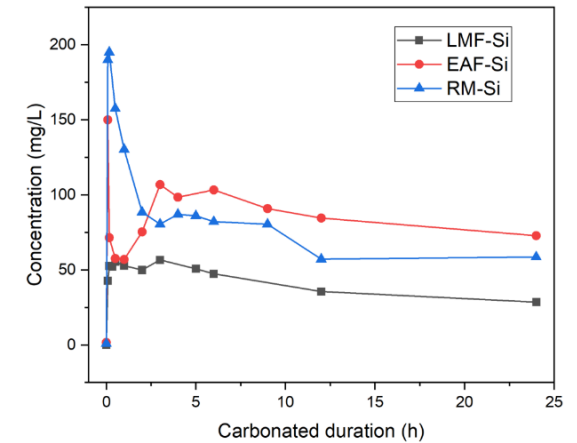
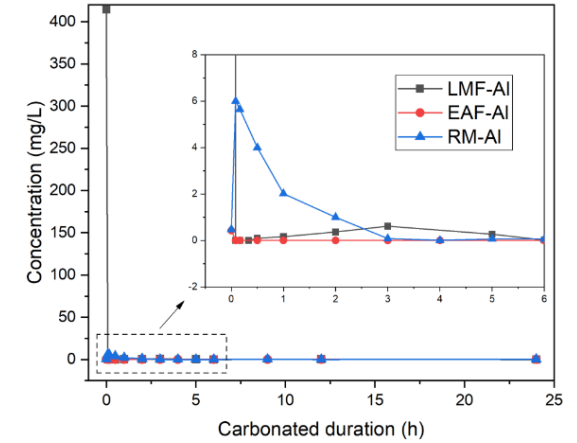
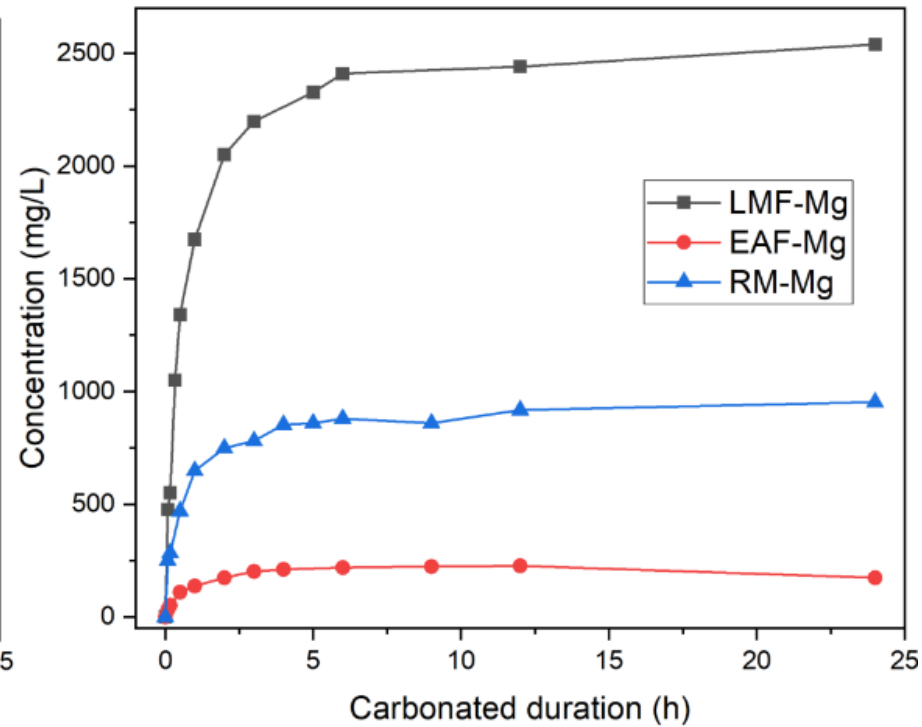
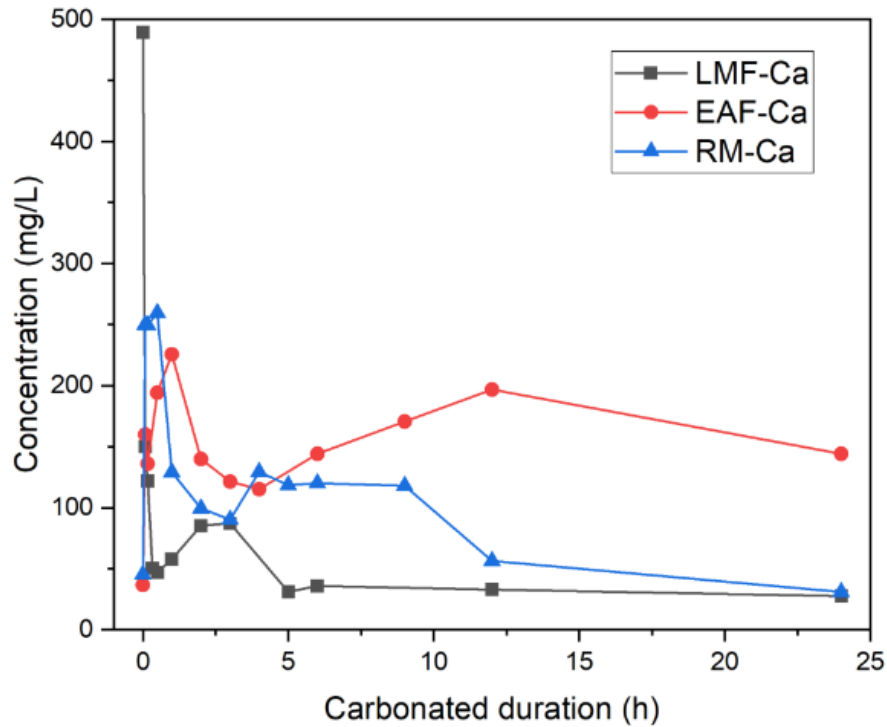


Steel slag



Progress and Current Status

Mechanism study: route 1, steel slag

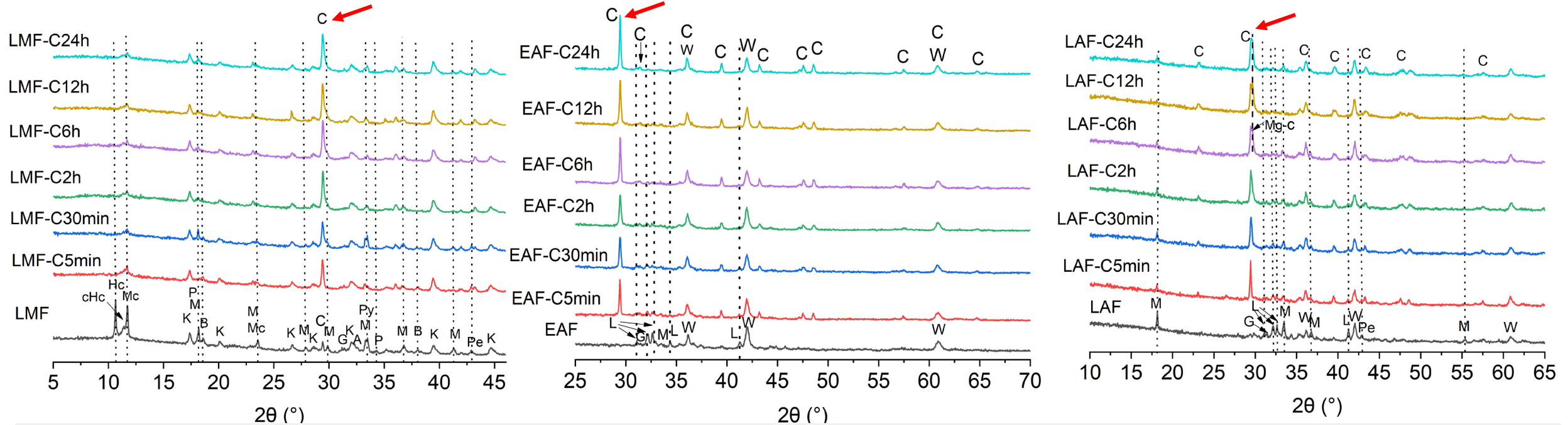


- Dissolution of Ca^{2+} and Mg^{2+} ions in water
- Carbonation: $\text{Ca}^{2+}/\text{Mg}^{2+} + \text{CO}_3^{2-} \rightarrow \text{CaCO}_3 / \text{MgCO}_3$



Progress and Current Status

Mechanism study: route 1, steel slag



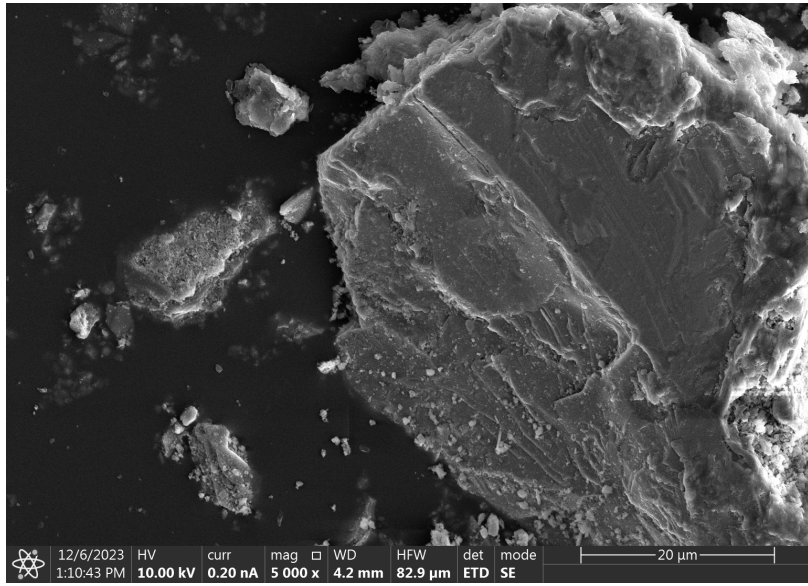
M: Mayenite, $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$; **C:** Calcite, CaCO_3 ; **L:** Larnite, Ca_2SiO_4 , **Am:** Amorphous; **Mc:** Calcium monocarboaluminate, $\text{Ca}_4\text{Al}_2(\text{OH})_{12}(\text{CO}_3)(\text{H}_2\text{O})_5$; **Hc:** Calcium hemi-carboaluminate, $\text{Ca}_4\text{Al}_2(\text{OH})_{13}(\text{CO}_3)_{0.5}(\text{H}_2\text{O})_4$; **cHc:** carbonated calcium hemi-carboaluminate, $\text{Ca}_4\text{Al}_2(\text{OH})_{12.4}(\text{CO}_3)_{0.8}(\text{H}_2\text{O})_4$

- The main carbonation **reactive minerals** in the slags are **larnite**, **mayenite**, and **AFm** phases (Mc, Hc, and cHc)
- The main carbonation **products** are **calcite** and **amorphous phases**, such as Al and Si gel

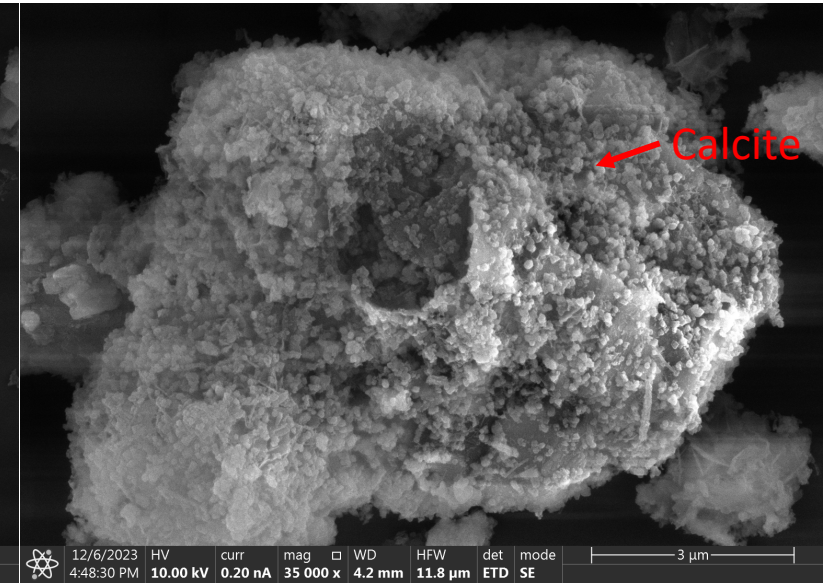
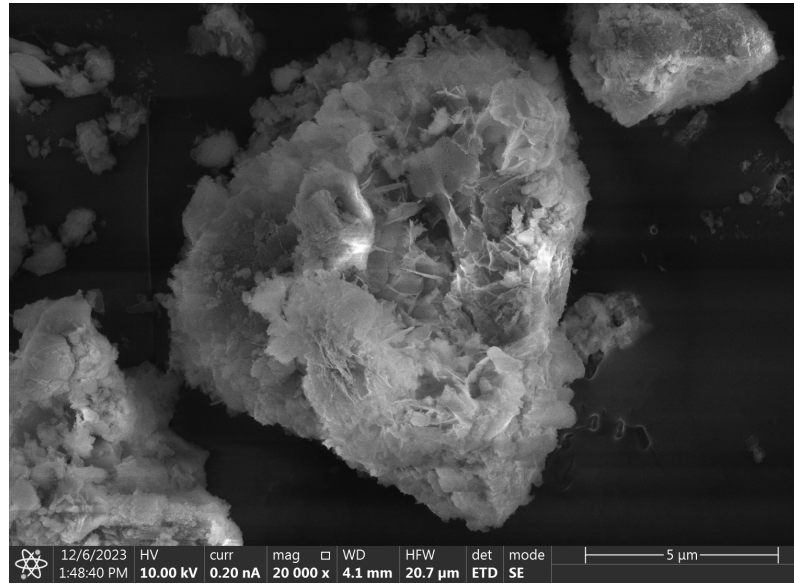


Progress and Current Status

Mechanism study: route 1, steel slag



Uncarbonated steel slag



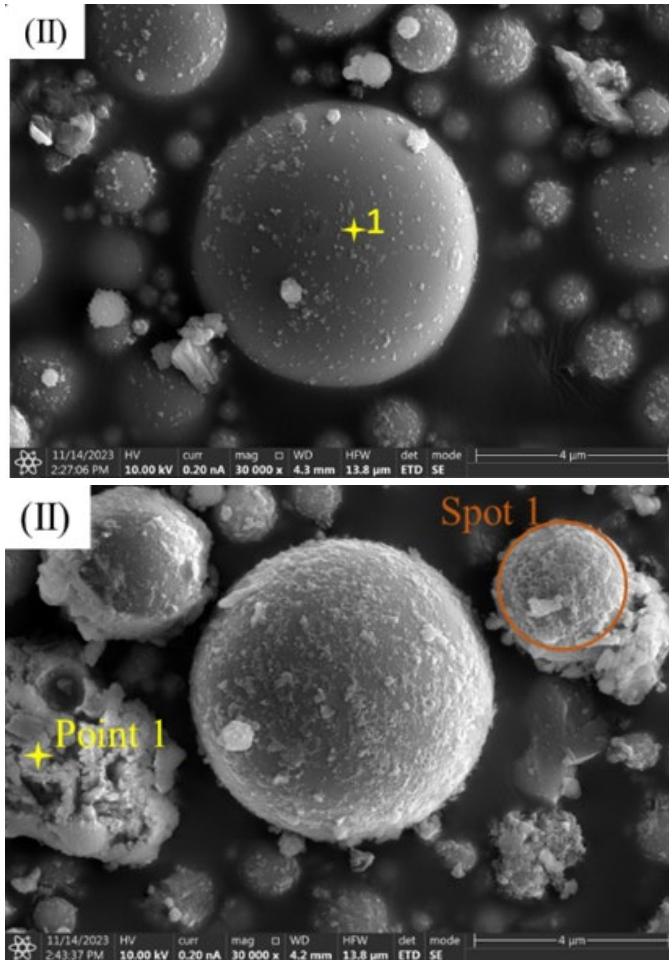
Carbonated steel slag

- The main carbonation **reactive minerals** in the slags are **larnite**, **mayenite**, and **AFm phases** (Mc, Hc, and cHc)
- The main carbonation **products** are **calcite** and **amorphous phases**, such as Al and Si gel



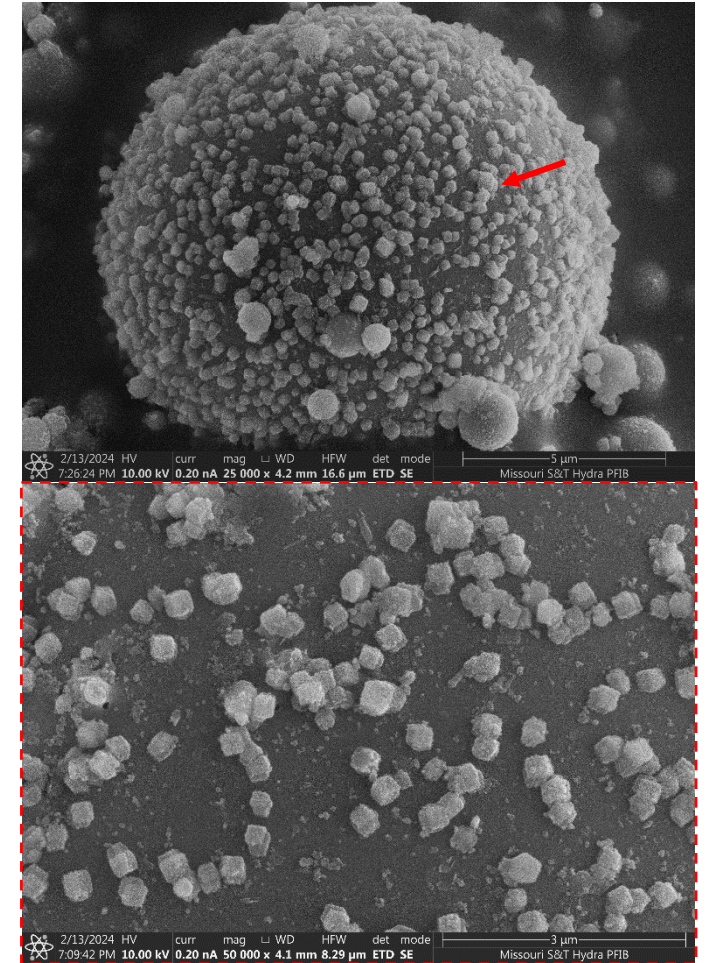
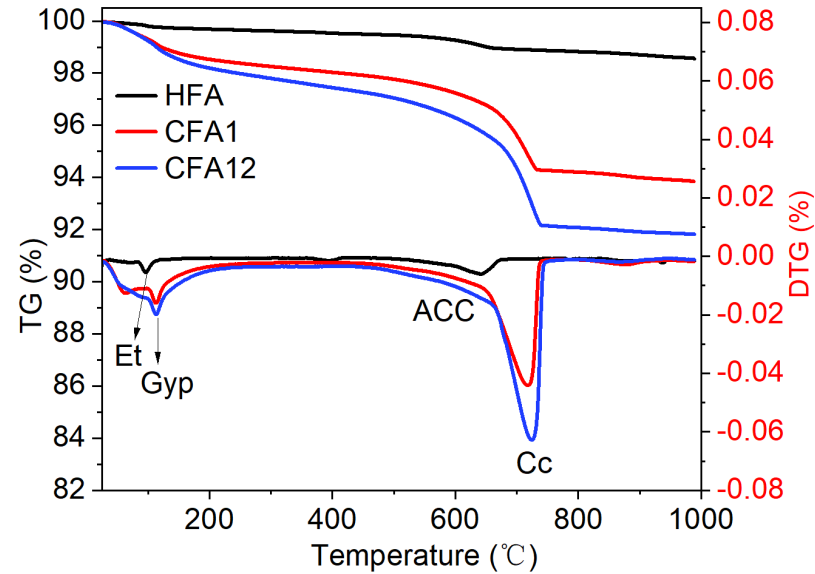
Progress and Current Status

Mechanism study: route 1 and route 2, fly ash



Less water: route 2

Carbonation of fly ash

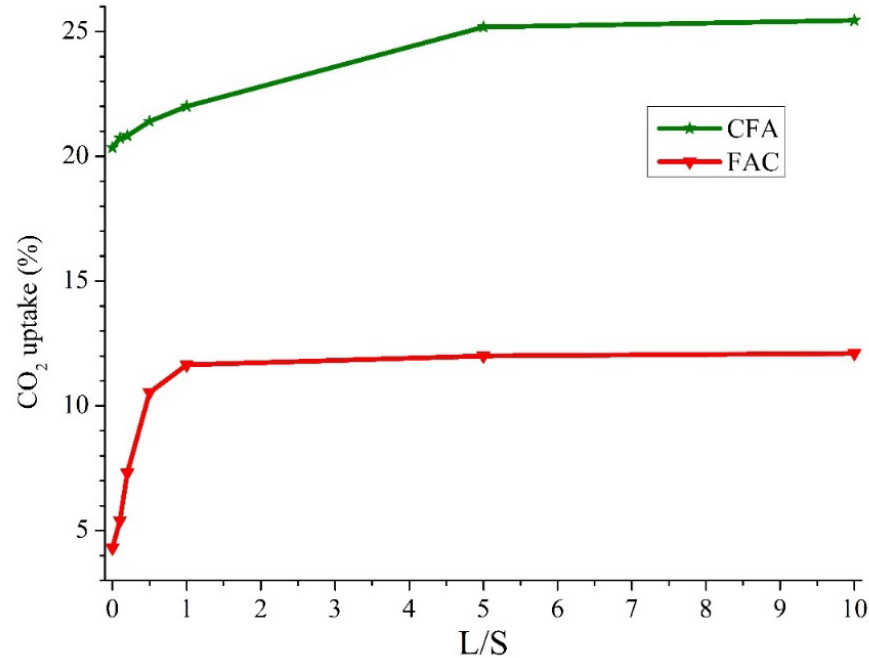


More water: route 1

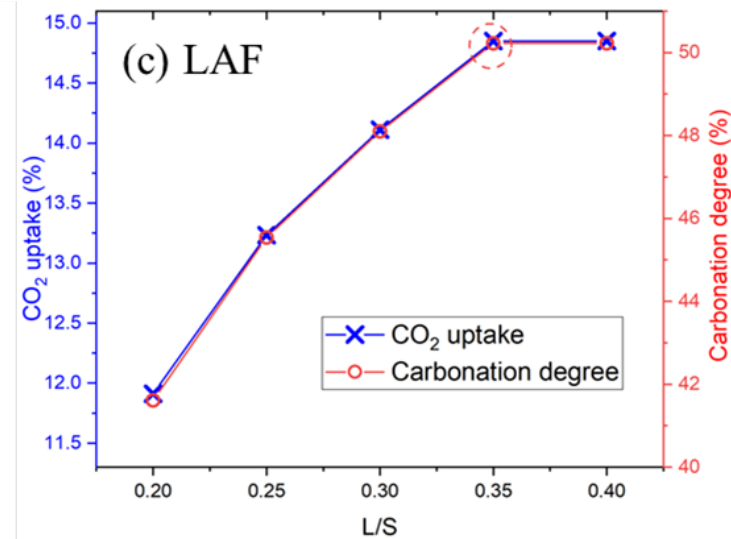
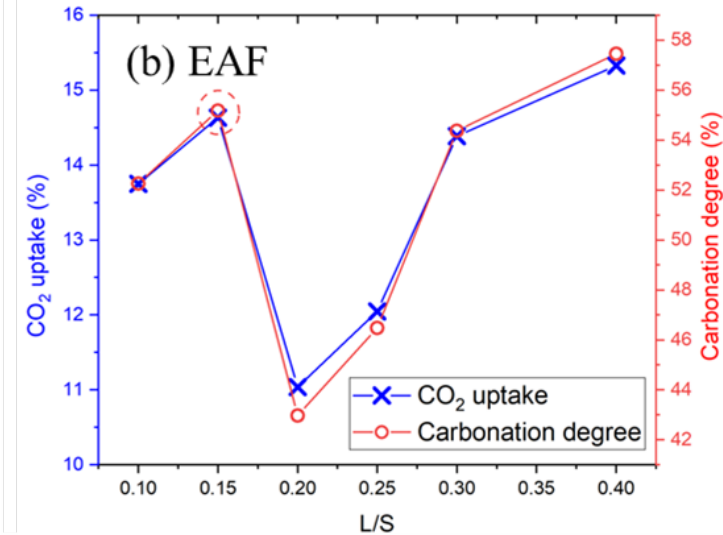
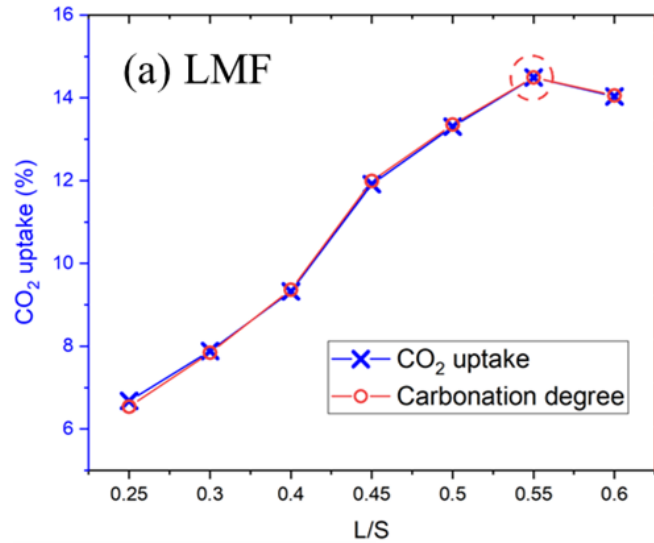


Progress and Current Status

Process optimization: effects of L/S, T, P, C_{CO_2} , ...



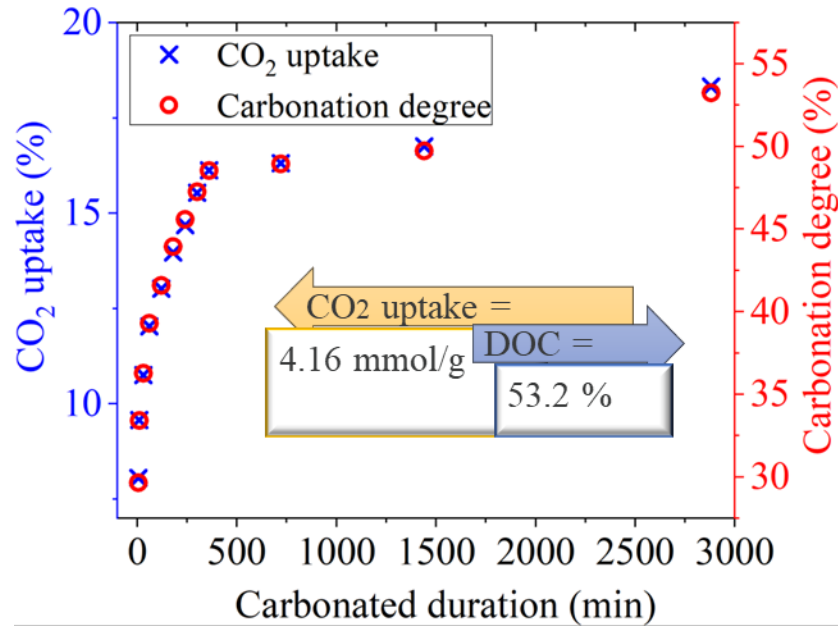
Fly ash (route 1 and route 2)



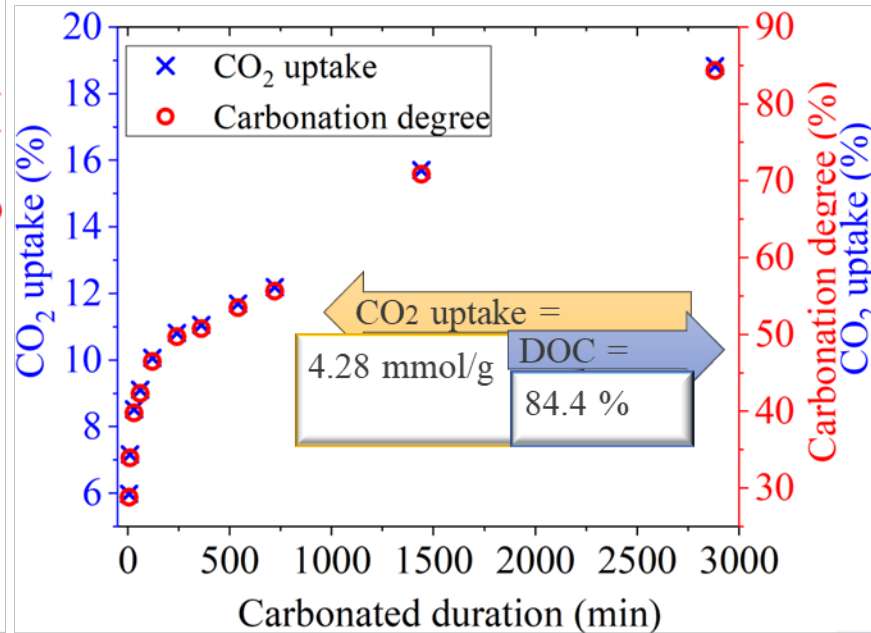
Steel slag (route 2)

Progress and Current Status

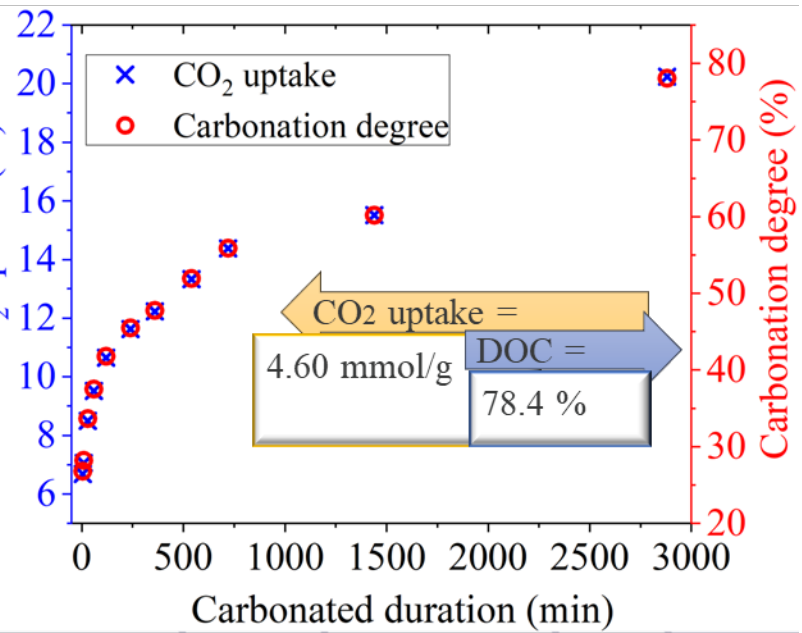
How much CO₂ uptake?



LMF slag



EAF slag



Blended slag

- Steel slags: 15%-20%; >4 mmol/g
- Ashes: 5%-25%; up to 5.7 mmol/g
- MSWI FA? 10 mmol/g?

Progress and Current Status

Rate and kinetics of conversion: route 1

CaO/MgO-to-carbonate conversion rate

Sample	Type	Conversion rate @ 30 min	Time when conversion rate >65%	Maximum conversion rate
Steel slag	LMF	36.3%	/	53.8%
Steel slag	EAF	39.8%, ambient 65.5%, @60°C	24h	84.4%
Steel slag	LAF	33.6%	48h	100%
Coal fly ash	FAC	70%	10 minutes	81%
MSWI fly ash	CFA-B8	69%	10 minutes	75%
MSWI fly ash	CFA-B7	79%	10 minutes	82%
MSWI filter cake	CFA-B8	46%	/	60%

Similar total CO₂ uptake



Progress and Current Status

Rate and kinetics of conversion: route 2

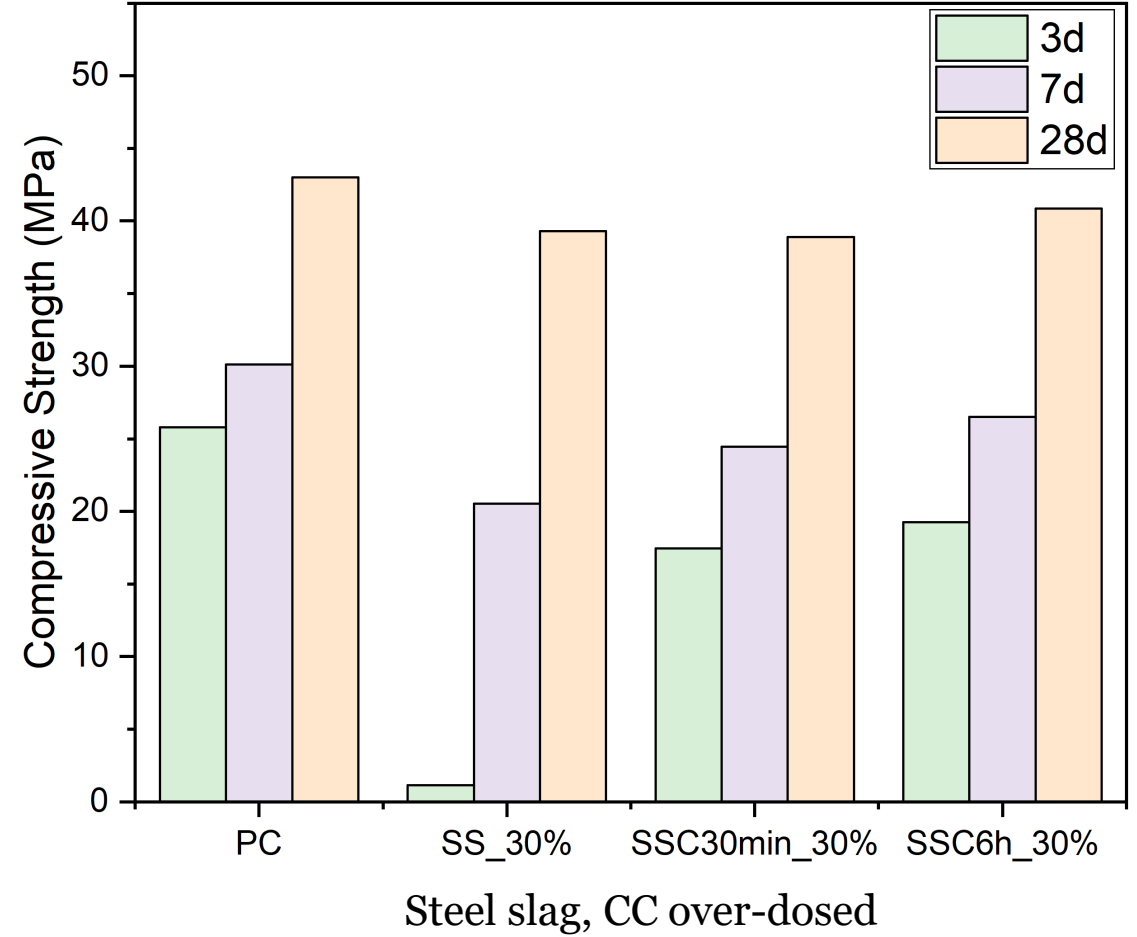
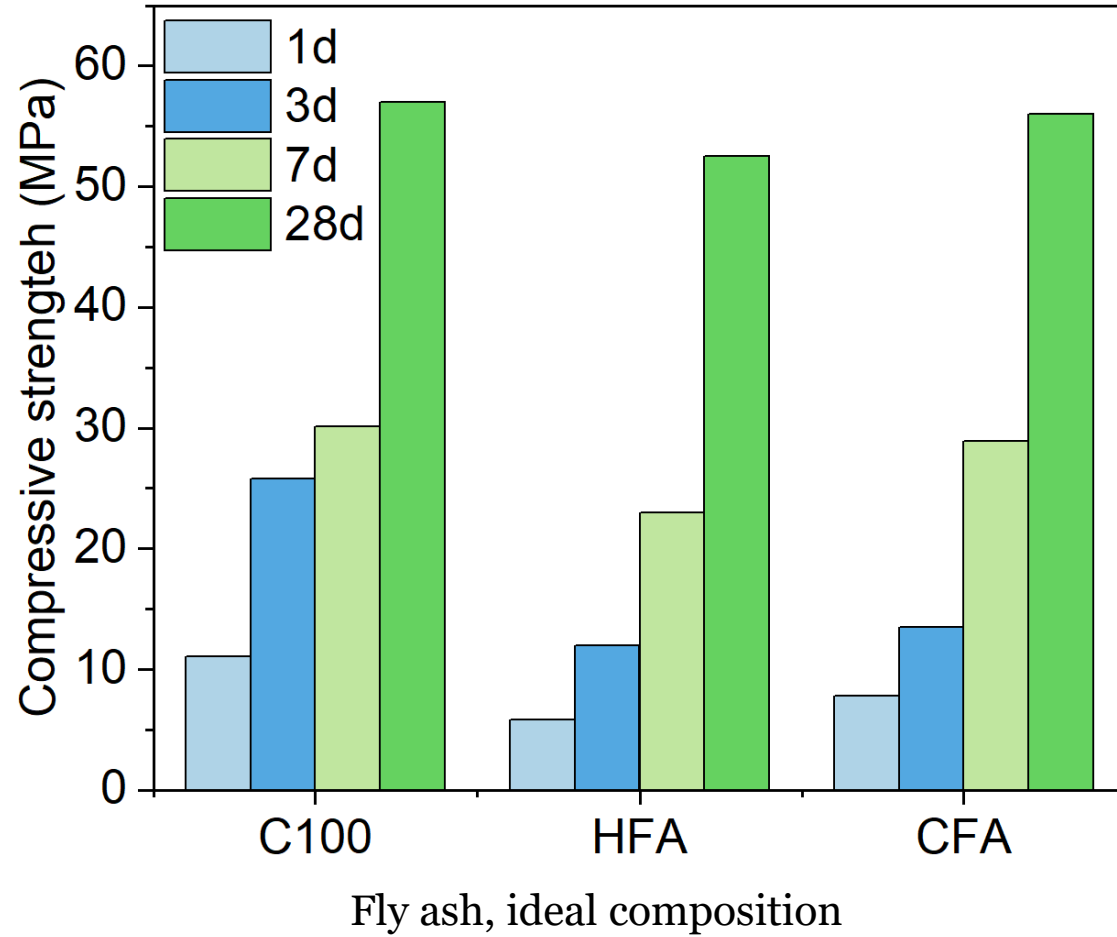
CaO/MgO-to-carbonate conversion rate

Sample	Type	Conversion rate @ 30 min	Conversion rate @ 120 min	Maximum conversion rate
Steel slag	LMF	32.3%	42.5%	44.9%
Steel slag	EAF	29.2%	41%	55.2%
Steel slag	LAF	20.9%	31%	53.1%



Progress and Current Status

nCAS-incorporated cement pastes



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Community Benefits

DEIA and workshop

- ❑ Hired one graduate student and one postdoctoral researcher from underrepresented groups
- ❑ Organized a high-school student workshop

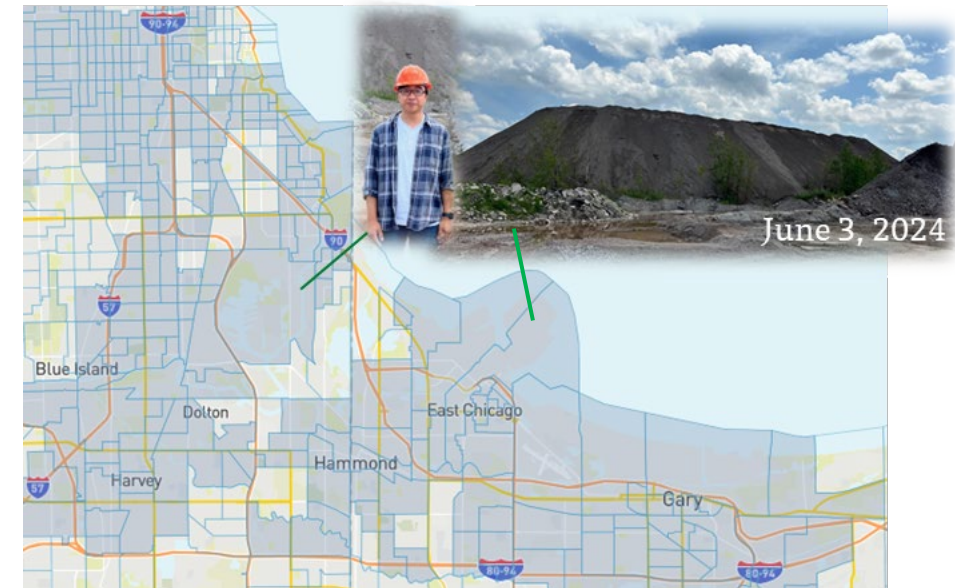
7/26/2024, half-day workshop →
“Storing CO₂ in Infrastructure Materials to Combat Climate Change”

As part of the
2024 **National Summer Transportation Institute**
Co-sponsored by
USDOT
MoDOT
Missouri S&T
The National Museum of Transportation
Center for Infrastructure Engineering Studies
Center for Intelligent Infrastructure
[Lab of Future Cements and Carbon-Negative Initiatives](#)
...



Community Benefits Justice40

- ❑ Prioritize processing solid wastes located in underserved communities
 - Sampling
 - Upscaling
 - Commercialization



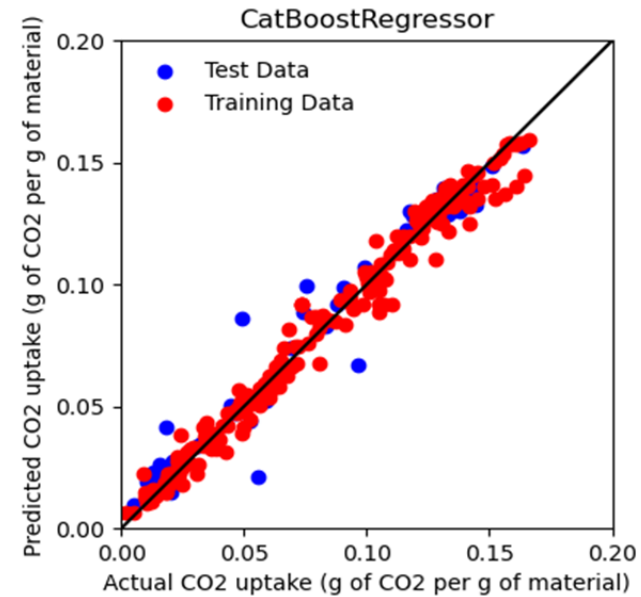
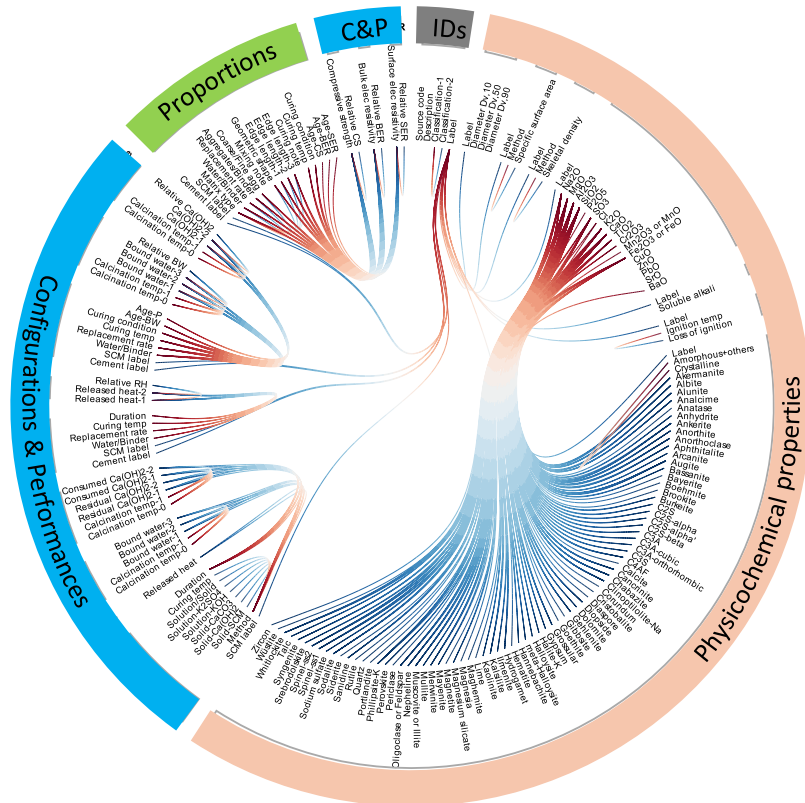
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Lessons Learned

- Not all solid wastes are available for upcycling (willingness of owners)
- Some solid wastes may not be “reliable” feedstock (viability)
- Some solid wastes may not be available anymore (changes in practice)



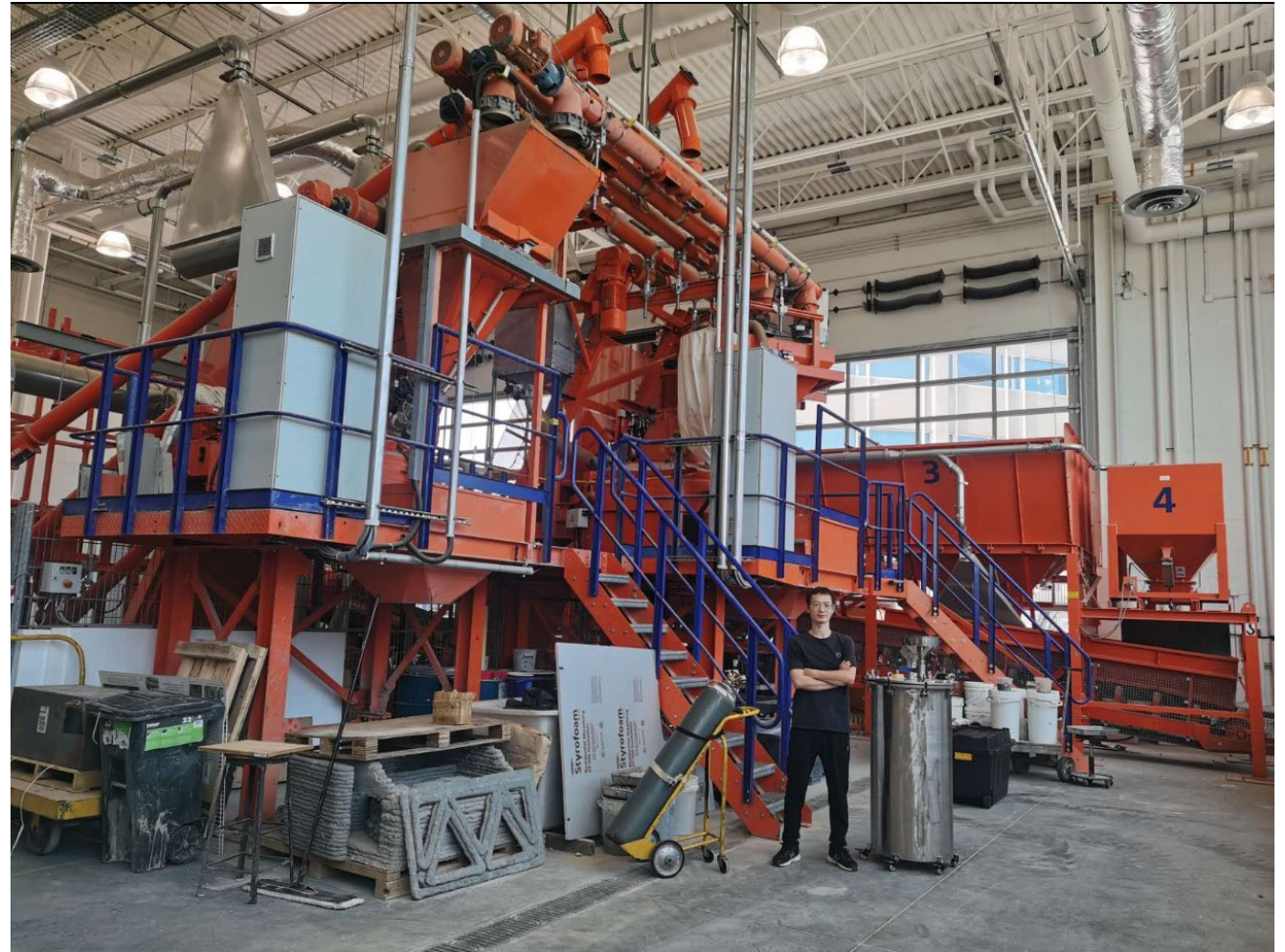
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Plan for Future Development and Commercialization

- Meet technical milestones
- Scale up the reactors
- Pilot trial at MS&T
- Demo project with an innovation ecosystem
- Launching a startup
- Licensing
- Two business models
- ...



The 0.2 m³ reactor and 1 m³ concrete batching plant at MS&T



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Summary

- 150 samples
- Carbonation protocols have been optimized
- Carbonation mechanisms have been studied
- >65% degree of carbonation conversion is feasible
- When used in cement paste to substitute cement, nCAS typically shows a filler effect in early age, and enables secondary reactions to boost late-age development
- Database and AI are under development



Open to more solid wastes, academic collaborators, and commercialization partners

Contact

Hongyan Ma

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