



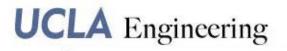
# Upcycled 'CO<sub>2</sub>-negative' concrete for construction functions

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**Developed for:** NETL CO<sub>2</sub> Capture Technology Project Review Meeting, August 13-17,2018, Pittsburgh, Pennsylvania

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#### **Presentation outline**

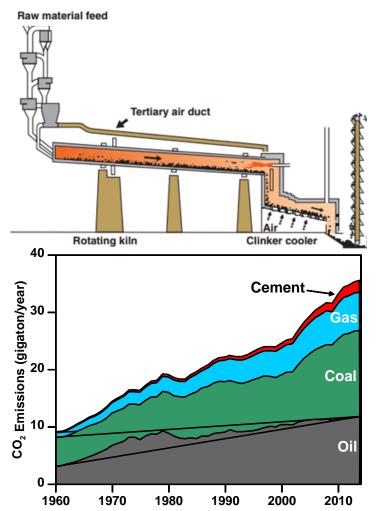
- Project Overview: Background, overall project objectives and timeline, funding, participants
- Technology Background: Upcycled concrete production process, advantages and challenges
- Technical Approach/Project Scope: Experimental design and work plan, key milestones, success criteria
- Progress and Current Status of Project
- Summary and future work

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# **Upcycled 'CO<sub>2</sub>-negative' concrete**

- 33 billion tons of concrete, 4.5 billion tons of portland cement (OPC) produced annually
- 0.9 ton CO<sub>2</sub> emitted per ton OPC produced—from energy input for processing at high T (~1600 °C) and CO<sub>2</sub> emitted during calcination
- Identify routes for large-scale utilization of CO<sub>2</sub> as a precursor in beneficial products and processes, by mineralization as stable carbonate compounds with cementitious properties



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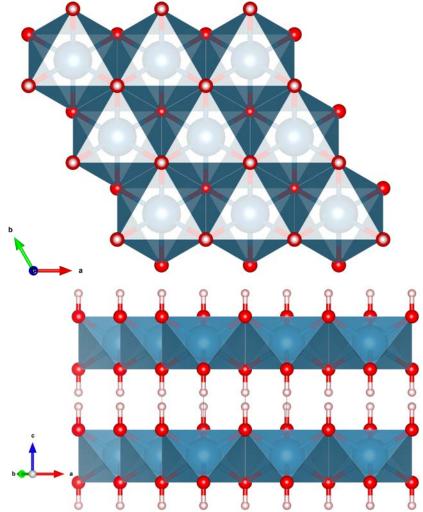


#### Low-temperature synthesis of portlandite

 Hydrated lime is an efficient material for CO<sub>2</sub> uptake (max. CO<sub>2</sub> uptake = <u>59%</u> by mass)

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

- Industrial methods of portlandite production require energy- and CO<sub>2</sub>-intensive calcination of limestone
- Low-temperature synthesis of portlandite using industrial byproducts and waste heat



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## **Objectives of the upcycled concrete technology**

#### Upcycling industrial wastes and CO<sub>2</sub>

 Utilize coal combustion and metal processing wastes as precursors for scalable CO<sub>2</sub> mineralization

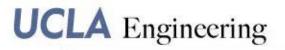
#### **Process design**

 Develop an integrated, 'bolt-on' technology solution for upcycled concrete production incorporating aspects of Ca-leaching, Ca(OH)<sub>2</sub> precipitation, mixture formulation, and structural shape-stabilization, while maximizing CO<sub>2</sub> uptake

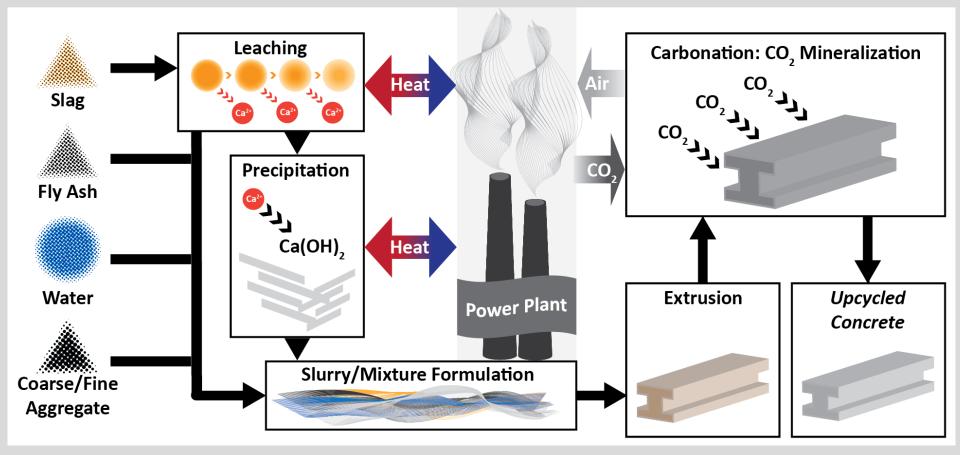
#### **OPC concrete replacement**

 Develop a novel CO<sub>2</sub>-negative upcycled concrete that is performanceequivalent or superior to OPC-based concrete

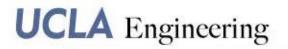
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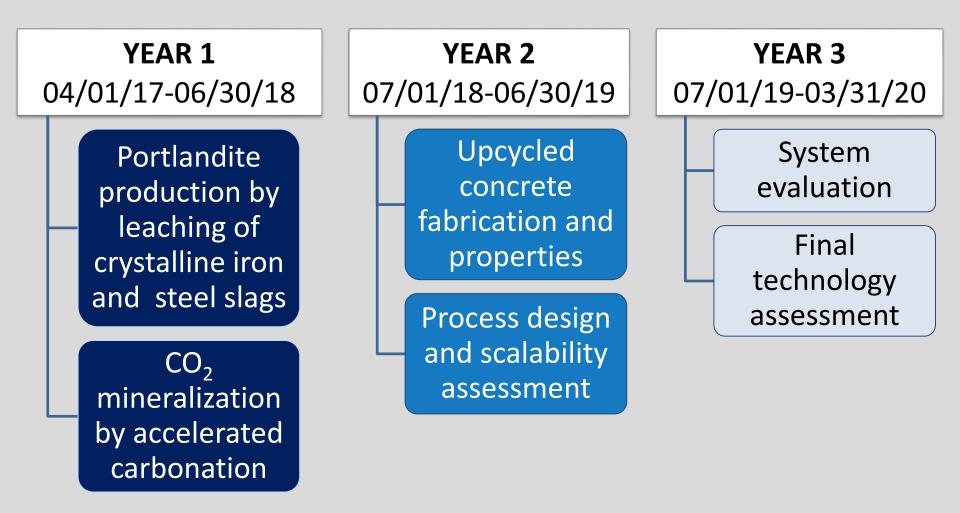
# **Overview of CO<sub>2</sub>-negative upcycled concrete** production process

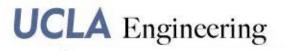


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#### **Project scope and current status**





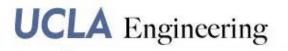


# **Project funding profile**

	Budget F	Period 1	Budget	Period 2	Budget P	Period 3	Total Droject			
	04/01/17-	06/30/18	07/01/18-06/30/19		07/01/19-0	03/31/20	Total Project			
	Gov't Share	Cost Share	Gov't	Cost Share	Gov't	Cost	Gov't	Cost Share		
	Gov t Share	COSt Share	Share	COSt Share	Share	Share	Share			
UCLA	\$344,436	\$155,533	\$274,142	\$119,467	\$181,421	\$25,000	\$799,999	\$300,000		
ASU	\$75,155	\$18,480	\$66,541	\$15,583	\$58,304	\$15,937	\$200,000	\$50,000		
Total	\$419,591	\$174,013	\$340,683	\$135,050	\$239,725	\$40,937	\$999,999	\$350,000		
Cost										
Share	71%	29%	72%	28%	85%	15%	74%	26%		

#### **Project participants:**

- University of California, Los Angeles (UCLA)
- Arizona State University (ASU)
- Boral North America





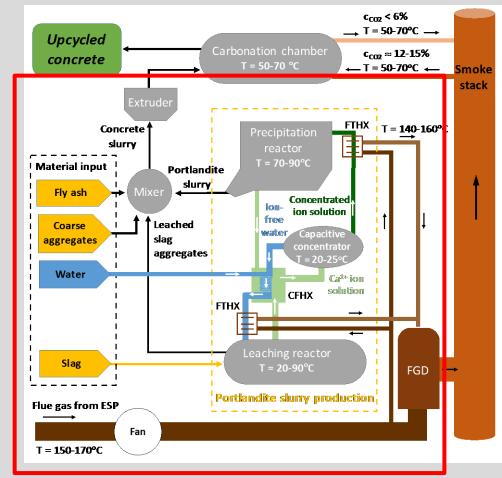
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# Process diagram for integrated production of upcycled concrete

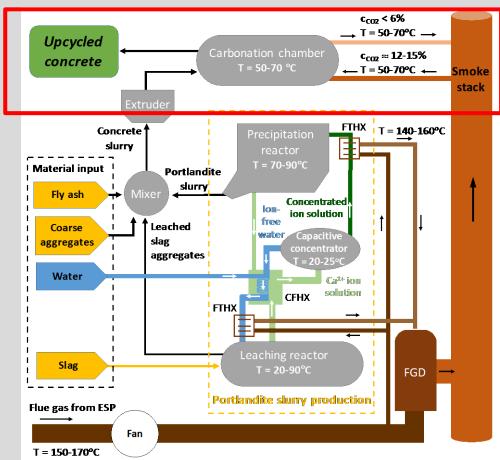
- Securing reclaimed solid reactants
- Ca extraction (leaching) within the leaching reactor
- Concentration of leaching solution in Ca, followed by Ca(OH)<sub>2</sub> precipitation
- Formulation of a rheologyoptimized slurry



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# Process diagram for integrated production of upcycled concrete

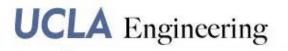
- Shape-stabilization of slurry into the form of a structural section (beam, column, etc.)
- Contacting structural section with flue-gas borne CO<sub>2</sub> within a carbonation chamber – *"upcycled concrete"* section
- Low-grade heat sourced from flue gas prior to, and following, desulfurization to optimize kinetics



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# Advantages of *upcycled concrete* technology and practical considerations

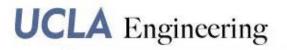
- Reduce construction period with precast/prefabricated components, compared to traditional "cast-in-place" construction, while ensuring repeatability and high quality
- Utilize CO<sub>2</sub> and waste heat carried by the flue gas in a typical coalfired power plant, and reject waste streams (e.g., crystalline slags, non-compliant fly ash in landfills and ash ponds)
- Path to carbon neutral/negative cementation through the production of hydrated lime, Ca(OH)<sub>2</sub>
- Considerations on (1) compositional heterogeneity (leaching/carbonation potential) of fly ash and slag, (2) carbonation kinetics, (2) concrete workability and (3) mechanical properties





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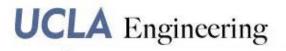




## Experimental design and work plan

- 1. Portlandite production by leaching of crystalline slags
- 2. CO<sub>2</sub> mineralization by accelerated carbonation
- 3. Upcycled concrete fabrication and properties
- 4. Process design and scalability assessment
- 5. System evaluation
  - System procurement and construction
  - Integrated laboratory-scale testing using simulated flue gas
- 6. Final technology assessment
  - Scalability assessment and economic feasibility study
  - Lifecycle and technology gap analyses

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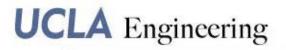
#### **Key milestones**

					Budget Period 2			_				
		1/17	7 - 6	/30	/18	7/1/	/18 -	6/30	/19	7/1/1	.9 - 3/	31/20
	Q1	Q2	Q3	Q4	Q5	<b>Q6</b>	Q7	<b>Q8</b>	<b>Q</b> 9	Q10	Q11	Q12
Leaching rate and extent for 3 slag types												
3 different CO <sub>2</sub> uptake levels (0.06-0.12 g												
$CO_2/g$ solid) with blended fly ash and $Ca(OH)_2$												
Rheology characteristics for upcycled												
concrete (UC) with 3 fly ash-Ca(OH) <sub>2</sub> blends												
Shape-stable upcycled concrete having												
compressive strength $\geq$ 15 MPa												
Process design for lab-scale test unit with												
production throughput of 10-100 kg/day UC												
Construction of lab-scale test unit above												
Production throughput of 10-100 kg/d UC,												
with CO <sub>2</sub> uptake of 0.06-0.12 g CO <sub>2</sub> /g solid												
Scalability, lifecycle CO <sub>2</sub> footprint and techno-												
economic feasibility												
Technology Gap Analysis												

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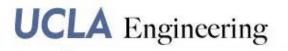
#### **Success criteria and decision points**

<ul> <li>Completion of BP 2</li> <li>Compressive strength of 15 MPa</li> <li>Lifecycle footprint that is &gt;75% smaller than OPC-concrete of equivalent performance grade (preliminary assessment)</li> <li>Design of laboratory-scale, integrated concrete production system with production throughput of 10-100 kg/day of upcycled concrete</li> <li>Real-time CO<sub>2</sub> uptake of the lab-scale test unit within 20% of the estimated "carbonation potential"</li> <li>Lifecycle footprint that is &gt;75% smaller than OPC-concrete of equivalent performance grade (final assessment)</li> <li>Conceptual scaled-up process design and completion of technical and economic feasibility study, market assessment, lifecycle analysis, and technology gap analysis</li> </ul>	Completion of BP 1	<ul> <li>Carbonation characteristics of fly ash and leached slag, and the process conditions for carbonation of upcycled concrete mortar</li> <li>The critical steps (leaching, portlandite production, and carbonation) can be carried out in 24-to-168 hours or less</li> </ul>
<ul> <li>Completion of BP 2</li> <li>Lifecycle footprint that is &gt;75% smaller than OPC-concrete of equivalent performance grade (preliminary assessment)</li> <li>Design of laboratory-scale, integrated concrete production system with production throughput of 10-100 kg/day of upcycled concrete</li> <li>Real-time CO<sub>2</sub> uptake of the lab-scale test unit within 20% of the estimated "carbonation potential"</li> <li>Lifecycle footprint that is &gt;75% smaller than OPC-concrete of equivalent performance grade (final assessment)</li> <li>Conceptual scaled-up process design and completion of technical and economic feasibility study, market assessment, lifecycle analysis, and</li> </ul>		
<ul> <li>Completion of BP 3</li> <li>Conceptual scaled-up process design and completion of technical and economic feasibility study, market assessment, lifecycle analysis, and</li> </ul>		<ul> <li>Lifecycle footprint that is &gt;75% smaller than OPC-concrete of equivalent performance grade (preliminary assessment)</li> <li>Design of laboratory-scale, integrated concrete production system with</li> </ul>
<ul> <li>Completion of BP 3</li> <li>Conceptual scaled-up process design and completion of technical and economic feasibility study, market assessment, lifecycle analysis, and</li> </ul>		
		<ul> <li>estimated "carbonation potential"</li> <li>Lifecycle footprint that is &gt;75% smaller than OPC-concrete of equivalent performance grade (final assessment)</li> <li>Conceptual scaled-up process design and completion of technical and economic feasibility study, market assessment, lifecycle analysis, and</li> </ul>

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#### **Presentation outline**

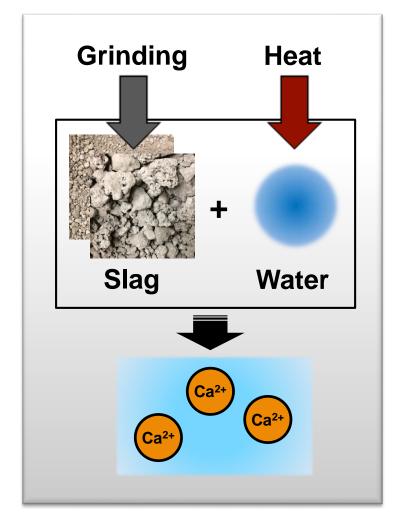
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# Sourcing of Ca from slags

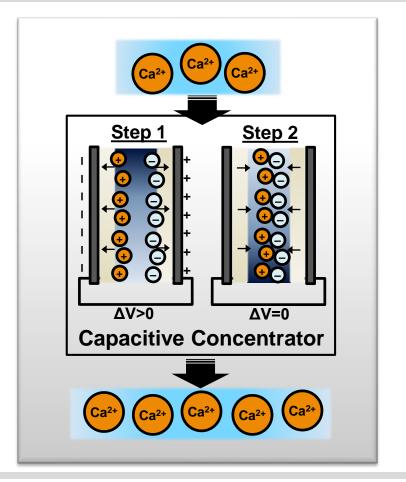
- Slags contain about 30-50% CaO by mass – Ca leaching potential of ~0.2–0.3 g per g slag
- Crystalline slags, which are used as low-value aggregates, are the focus of our process
- Up to 10 mM (400 ppm) Ca leached in water after 24 hours
- Rapidly evolves to a highly alkaline solution amenable to portlandite precipitation (final step)



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# Enhancing Ca concentrations to reach Ca(OH)<sub>2</sub> (portlandite) saturation

- Next step involves increasing Ca concentrations in the leachate to reach Ca(OH)<sub>2</sub> saturation
- Capacitive concentration cell with activated carbon or stainless steel electrodes
- As voltage is applied, ions migrate and adsorb on electrode surfaces; when voltage is reversed (or changed to zero), ions desorb, concentrating the flowing solution

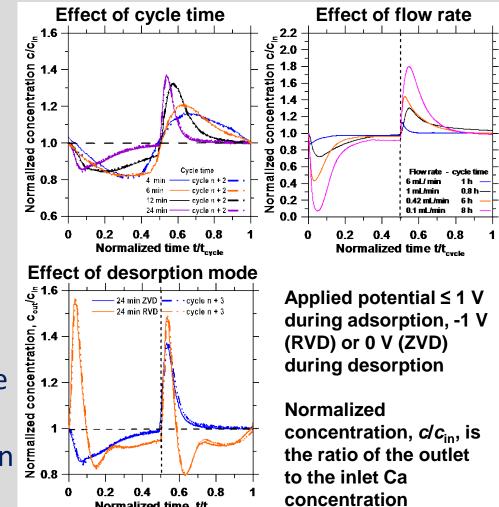


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#### **Capacitive concentration of CaCl**<sub>2</sub> solutions

- Initial experiments used 10 mM CaCl<sub>2</sub> as inlet solution – easily handled, high solubility of CaCl<sub>2</sub> in water, and represents a solution rich in Ca<sup>2+</sup> ions
- **Concentration factor increases** with decreasing flow rate and increasing cycle time, up to a value of 1.8x
- Slag leachates are highly alkaline – substantial decrease in extent of desorption (and concentration factor) for Ca(OH)<sub>2</sub> solutions



Normalized time, t/t

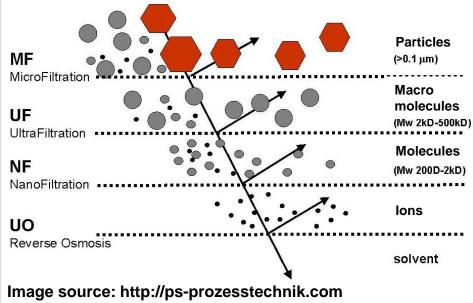
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# Alternative routes to concentration of alkaline Ca-containing solutions

- Capacitive concentration is not appropriate for alkaline solutions containing high [Ca]
- Stainless steel and nickel are electrochemically more inert than carbon under the relevant solution conditions, but their performance is limited by low SA
- Activated carbon electrodes have superior SA, but unstable in the presence of Ca

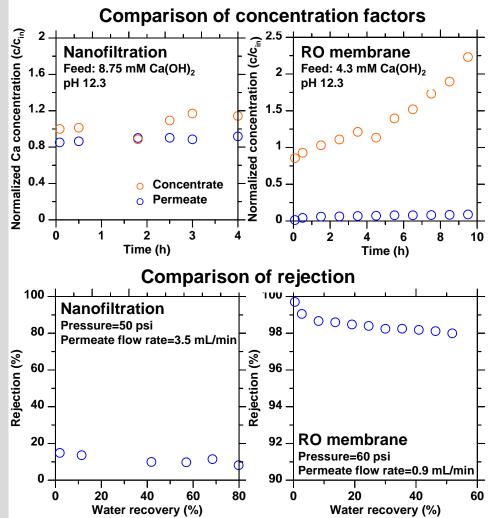
Membrane filtration which operates based on size exclusion and/or electrostatic repulsion is an alternative process to concentrate Ca<sup>2+</sup> ions



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# **Concentration of Ca(OH)**<sub>2</sub> solutions

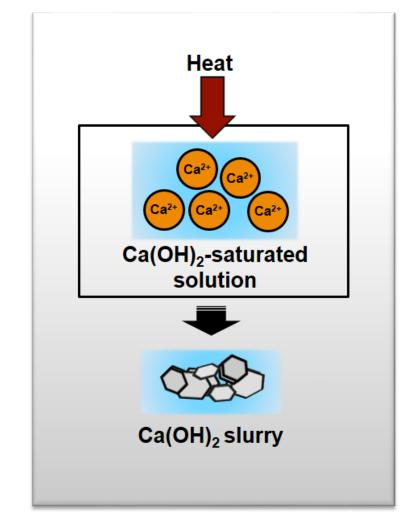
- Ca concentration factor for Ca(OH)<sub>2</sub> solutions reached up to >2x using reverse osmosis membrane (vs. <1.2x using nanofiltration)
- RO membrane showed greater Ca rejection (>98%) than nanofiltration (<20%)</li>
- RO membrane filtration suitable for concentration of alkaline Ca-rich solutions



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#### **Temperature ramping to induce precipitation**

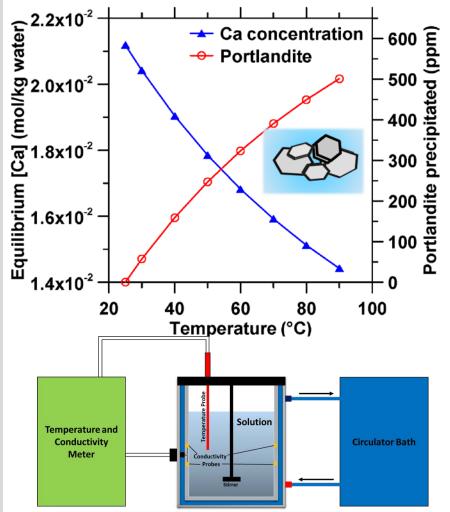
- Portlandite solubility decreases with increasing temperature
- Up to 500 ppm of portlandite can be precipitated from a saturated solution by temperature ramping
- pH adjustment is not necessary because of the alkaline nature of the slag leachate
- Precipitated portlandite is added to upcycled concrete formulation for subsequent carbonation

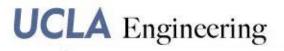


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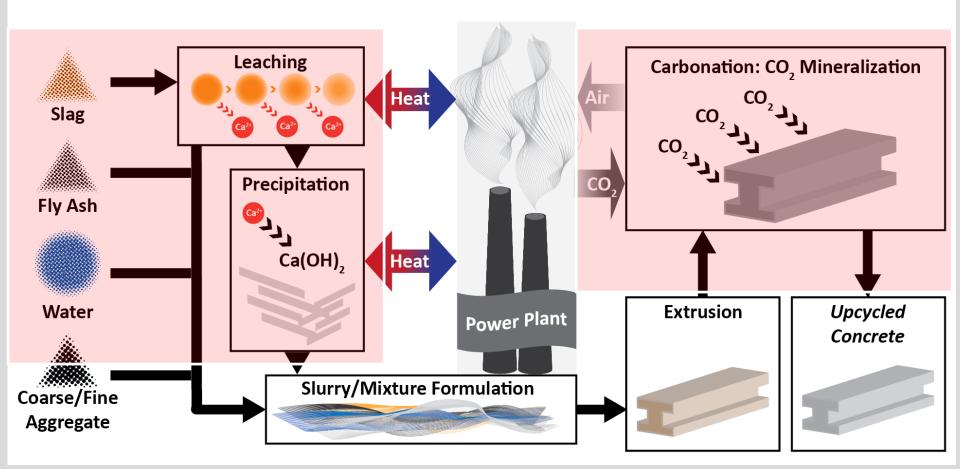
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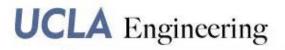
#### **Carbonation of** *upcycled concrete* mortars



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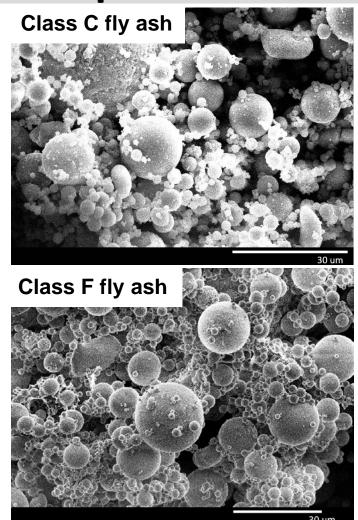
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# **Carbonation of fly ash and portlandite**

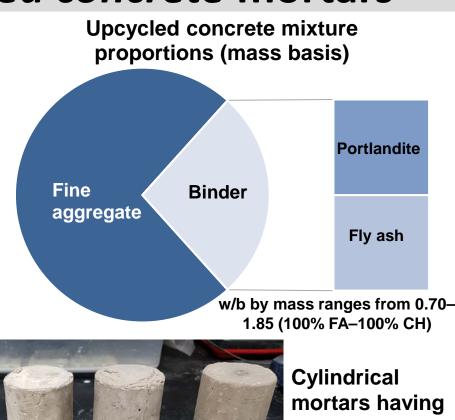
- Fly ash is a coal combustion byproduct which has cementitious properties and potential for CO<sub>2</sub> uptake
- Depending on type, can sequester up to 0.05–0.3 g CO<sub>2</sub>/g (0.59 for portlandite, CH)
- Carbonation of *suspensions* of CH–FA blends showed linear scaling of CO<sub>2</sub> uptake with CH
- Mortar samples prepared to represent *upcycled concrete*



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## **Carbonation of** *upcycled concrete* mortars

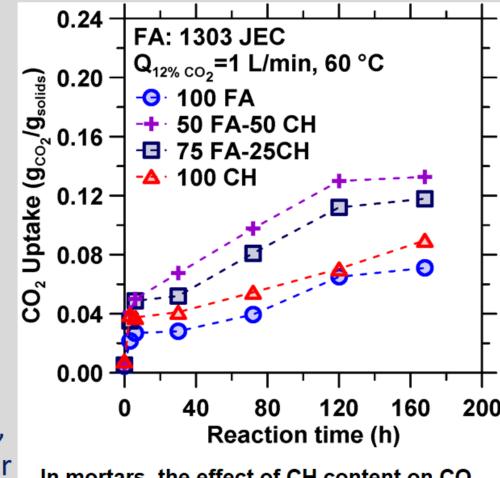
- Mortars cured at 45 °C for 5 h, then removed from molds prior to carbonation
- Carbonation carried out in reactors by flowing a gas mixture containing 12% CO<sub>2</sub> (v/v) at a rate of 1 slpm
- TGA carried out on powder samples (extracted using a drill) to evaluate temporally evolving CO<sub>2</sub> uptake for the different mortar compositions



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# Carbonation of *upcycled concrete* mortars

- CO<sub>2</sub> uptake increased with CH content, up to a point–not a simple linear scaling
- Optimum carbonation levels obtained for moderate dosages of CH (~50% by mass)
- Microstructure effects revealed and can be explained by (1) higher water content in CH-rich mixtures and/or (2) rapid carbonation of CH-rich mixtures, forming an outer carbonate layer having low porosity



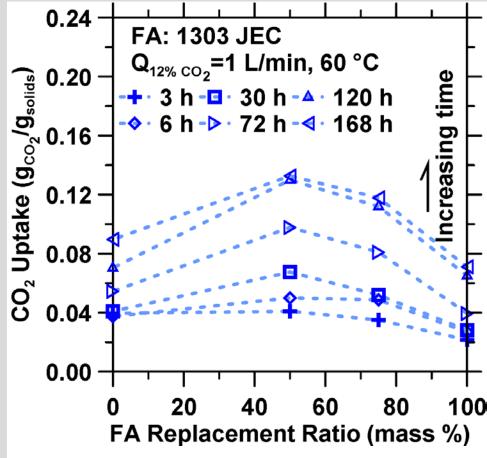
In mortars, the effect of CH content on CO<sub>2</sub> uptake is not a simple linear scaling

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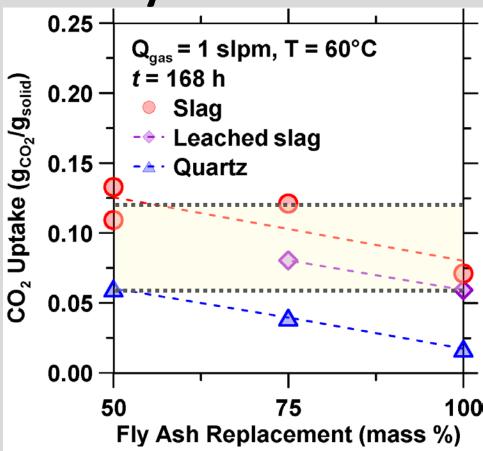
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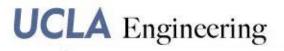
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## CO<sub>2</sub> uptake in portlandite-fly ash mortars

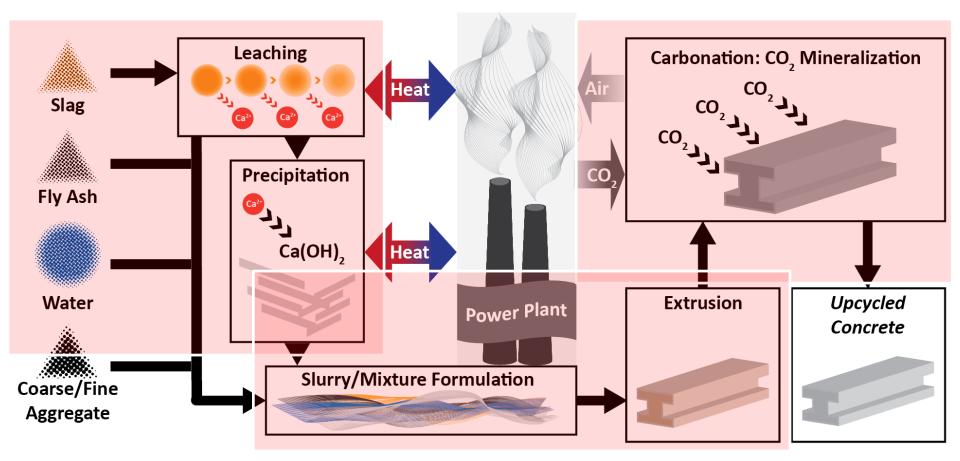
- Type of aggregate influenced CO<sub>2</sub> uptake – slag contributed significantly to carbonation
- Target CO<sub>2</sub> uptake of 6–12% achieved using ground asreceived and leached slag
- Carbonation rates decreased over time – significant uptake at t ≤24 h
- Weak dependence on temperature over the range 45– 85 °C



Target  $CO_2$  uptake of 6–12% reached at 168 h for slag-containing mixtures having  $\leq$ 50% CH



# Fabrication of *upcycled concrete* mortars for carbonation



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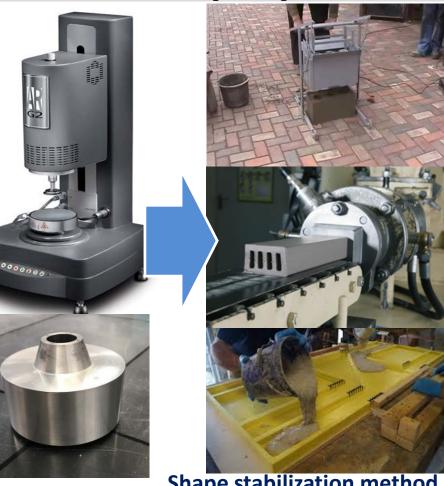
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## **Upcycled concrete fabrication and properties**

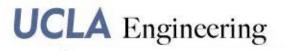
- Determine mixture proportions to optimize rheology (yield stress, plastic viscosity, suspension stability), workability
- Mixing ratios between the fly ash-portlandite-slag particulate blends, fine aggregates, water and chemical admixtures
- Select suitable shape stabilization process
- Optimize mechanical properties (compressive, flexural strength, fracture properties, etc.)



Laboratory tests

Shape stabilization method based on application

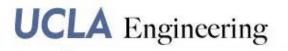
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#### Process design and system evaluation

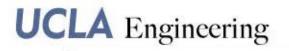
- Establish process design for laboratory-scale demonstration of integrated *upcycled concrete* production system
  - Component selection and design
  - System design and process optimization
  - Operating procedures and test plan
- Perform test runs using simulated coal-fired power plant flue gas
- Produce upcycled concrete with different CO<sub>2</sub> uptake levels
- Performance data from experimental test runs: CO<sub>2</sub> uptake, mass flow rate, production throughput, energy consumption, etc.





#### Summary

- Upcycled 'CO<sub>2</sub>-negative' concrete project utilizes coal combustion and metal processing wastes to develop an integrated technology solution for the production of an OPC concrete replacement, while maximizing CO<sub>2</sub> uptake
- In the first year, we have demonstrated portlandite synthesis from crystalline slags and CO<sub>2</sub> uptake levels of 6– 12% by mass of solid reactants
- Ongoing work includes the rheological characterization of upcycled concrete formulations and process design, including component selection for the laboratory scale upcycled concrete reactor





#### Acknowledgments

- DOE-NETL Project DE-FE0029825
- Project Manager: Andrew Jones

