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Application of Cellulose Nanomaterials in 3D Printed Sustainable Building Composites

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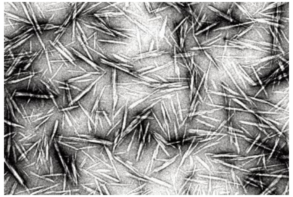
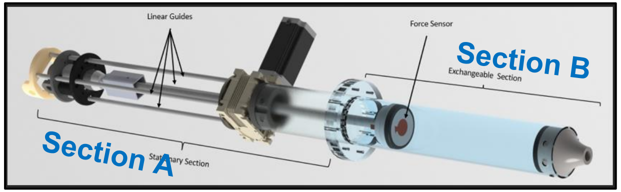
Motivation & Innovation

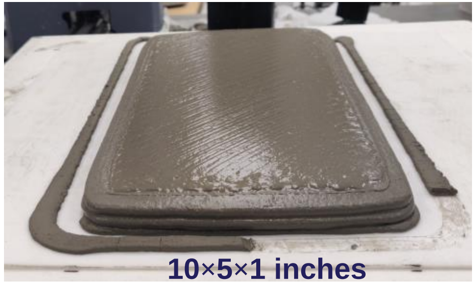
- 3D printed cementitious materials has been receiving increasing attention
- Challenges exist to deliver cost-effective and sustainable 3D printed cementitious mixtures
- Cellulose Nanomaterials are "green" nanoparticles that can improve the mechanical properties of cement-based materials
- **Application of CN-materials in alkali-activated composites**



Project Objectives

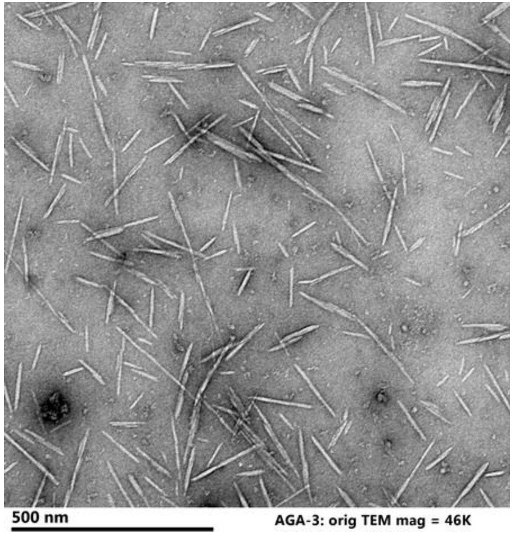
- Develop high-performance 3D printed sustainable cementitious composites using CN-materials for architectural applications (Impact on Printability and hardened properties)
- Provide input data for the techno-economic feasibility of the application of 3D printed CNC/CNF composites in architectural members





Background

- Cellulose nanocrystals (CNC) are the crystalline part of these polymers usually extracted from trees and plants.
- CNCs are typically 0.05 – 0.5 µm long and have a width of 3 – 5 nm.
- CNCs are renewable, biodegradable, sustainable, and present in high abundance in nature.



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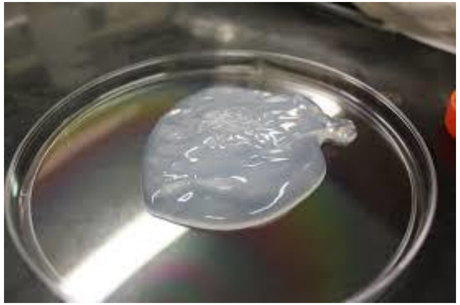
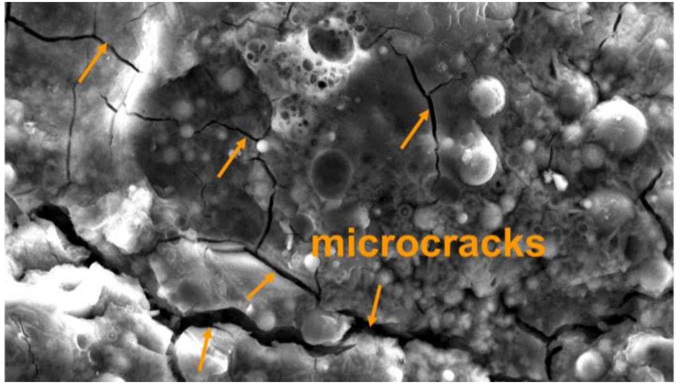


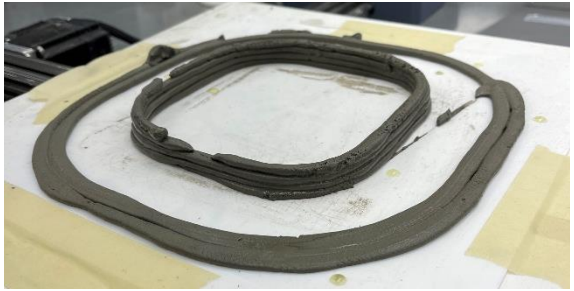
Research Approach

- Developing Printable Mixtures With CN-materials
- Impact of CN-materials on fresh properties
- Impact of CN-materials on performance and microstructure

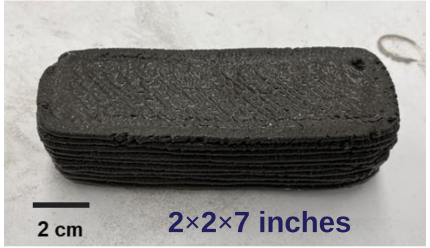


Experimental Program

- **Extrudability and Buildability of Mixtures**
  - The consistency of the extruded filaments (extrusion force)
  - No splitting or tearing of the filament
  - The number of successfully stacked layers before failure (e.g., 11 layers)
  - Excessive water leak (i.e., open print time)



Experimental Program



- **Printable Mixtures with/without CNC**
  - AA precursor consists of 70% Class F FA and 30% GGBFS by mass
  - The CNCs were in aqueous suspension (10.6% solids)

OPC Systems

Mixture ID	Cement (g)	w/c	Water (g)	CNCs slurry (g)	VMA (g)	HRWR (g)	CNCs/cement (vol%)
OPC–Control	1200	0.26	312.00	0	21.60	9.84	0.00
OPC–0.25% CNC	1200	0.26	299.95	13.48	21.60	9.84	0.25
OPC–1.00% CNC	1200	0.26	263.81	53.91	14.40	12.00	1.00
OPC–1.75% CNC	1200	0.315	293.66	94.34	38.40	11.40	1.75

AA Systems

Mixture ID	FA (g)	GGBFS (g)	Liquid /binder	NaOH (g)	Na_2SiO_3 (g)	CNCs slurry (g)	CMC (g)	CNCs/binder (vol%)
AA–Control	840	360	0.32	326.40	57.60	0.00	6.00	0.00
AA–0.30% CNC	840	360	0.32	311.22	54.92	19.98	0	0.30
AA–1.00% CNC	840	360	0.335	291.10	51.37	66.59	0	1.00
AA–1.50% CNC	840	360	0.345	275.99	48.70	99.89	0	1.50

Experimental Program

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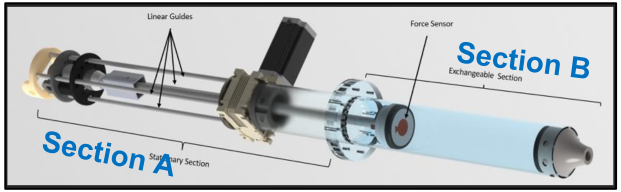
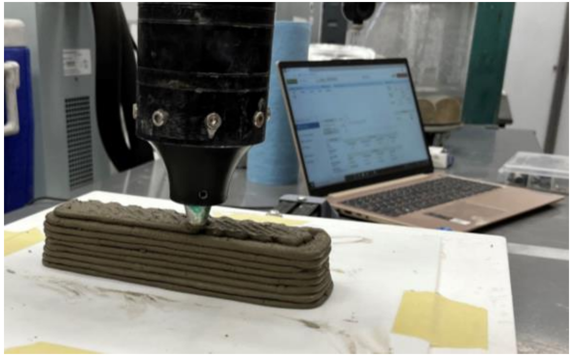
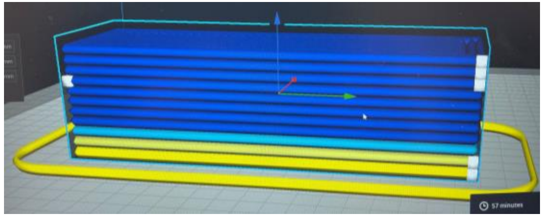
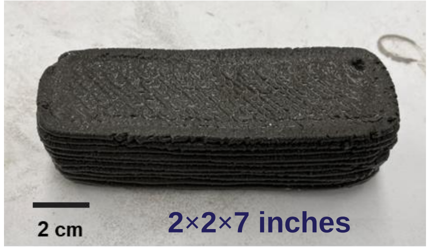
- **Printing Setup**
  - The cartridge assembly was separated into two pieces (material can be easily loaded onto "Section B" and consolidated)
  - The perimeter filament was printed in a straight-line pattern (5 mm/s) and the infill filaments were printed in a zigzag pattern (10 mm/s)



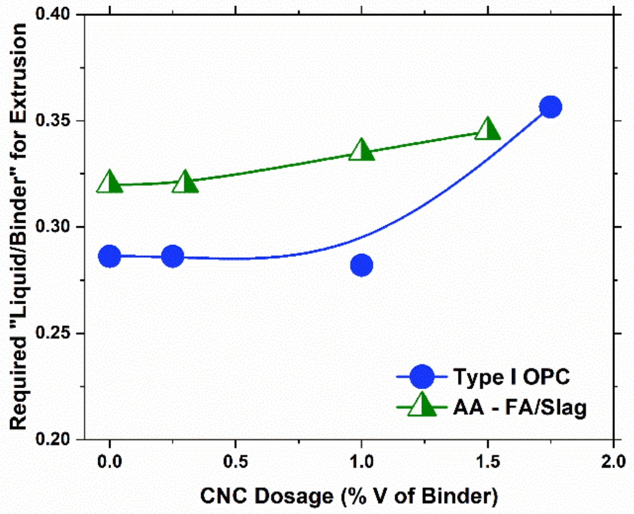






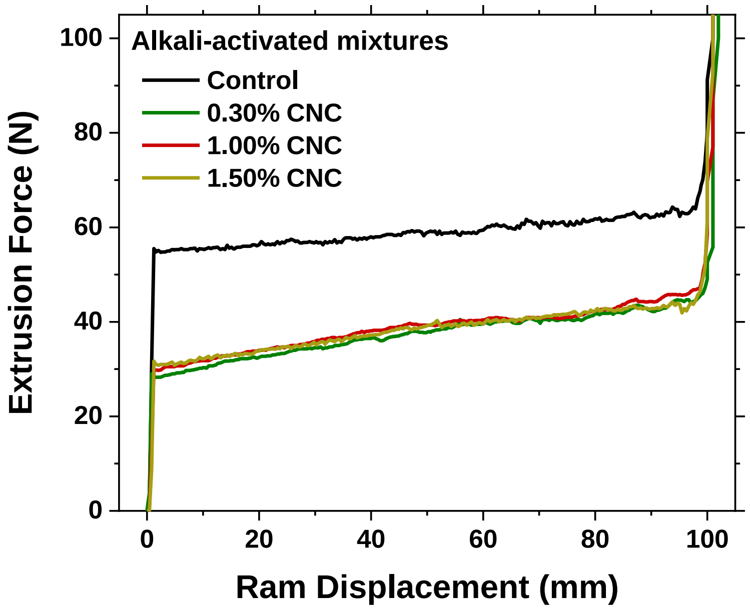
Extrudability and Buildability of Mixtures

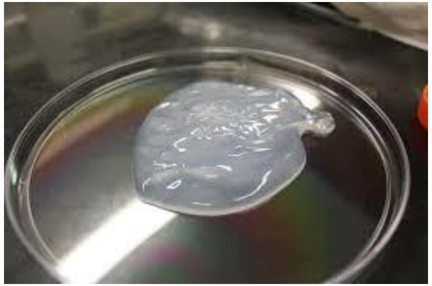
- The required "liquid/binder" was higher in AA mixtures compared to OPC mixtures
- However, OPC mixtures required a more dramatic increase of "liquid/binder" above 1.00% CNC concentration
- This can be attributed to the dispersion quality of CNC in different systems.

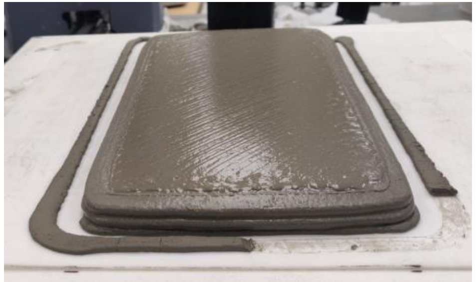


Extrudability and Buildability of Mixtures

- The addition of CNC in AA mixtures reduces the extrusion pressure (i.e., CNC performs as a VMA in AA mixtures).







Extrudability and Buildability of Mixtures

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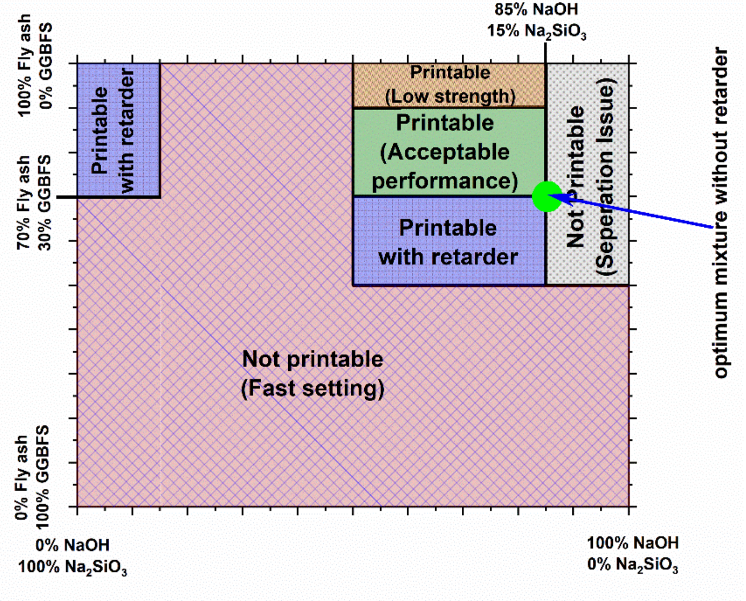
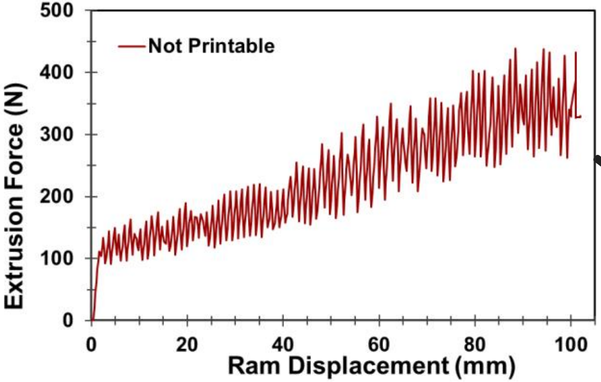
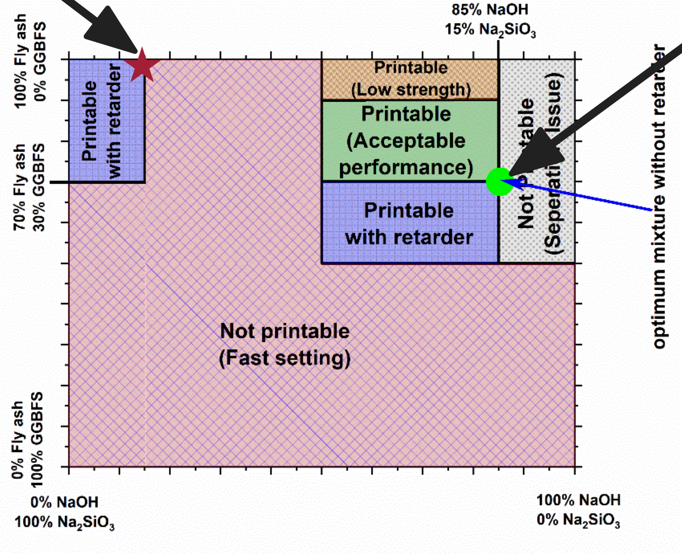
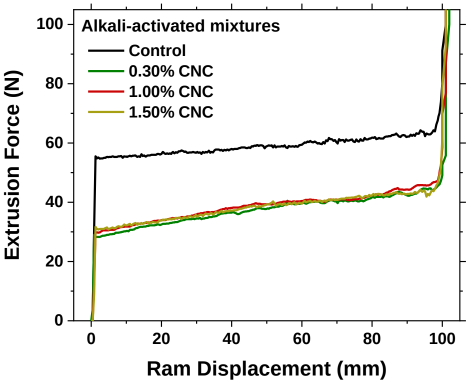
- The AA mixture without the addition of either CMC or CNC was not buildable. It seems that the CNC performs as a VMA in AA mixtures.
- The AA mixture with a "$NaOH/Na_2SiO_3$" mass ratio of 85:15 and a "FA/GGBFS" mass ratio of 70:30 was selected as the optimum AA mixture for printing.



Extrudability and Buildability of Mixtures

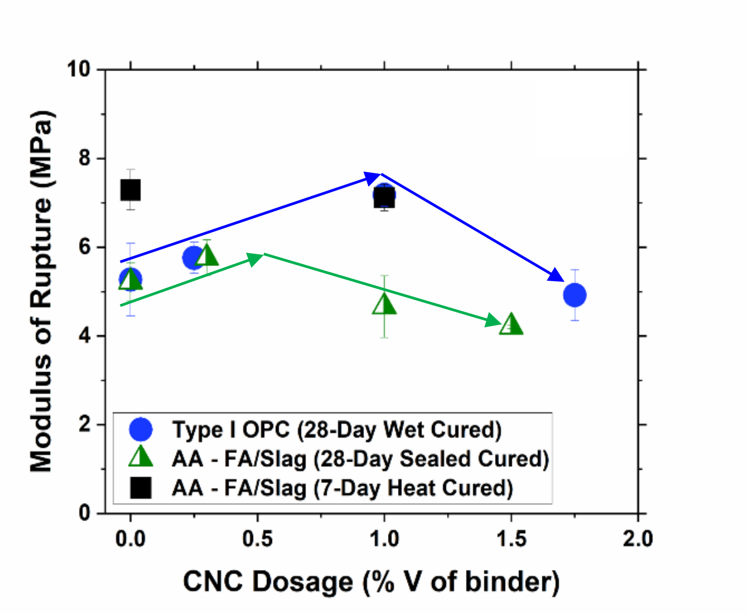


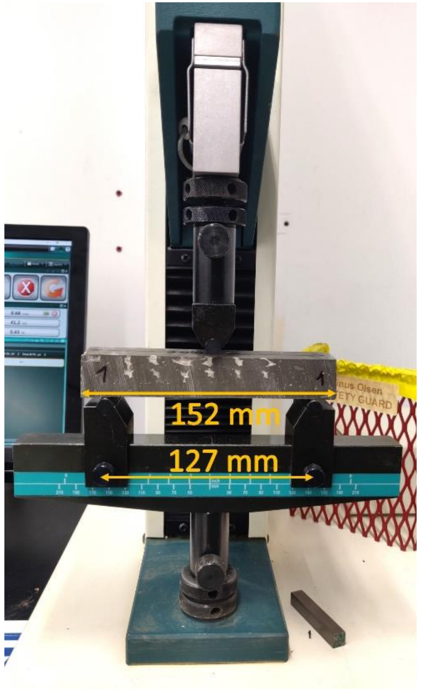




Mechanical Performance

- The inclusion of CNCs up to 1.00% (by volume of the binder) improves the overall mechanical performance





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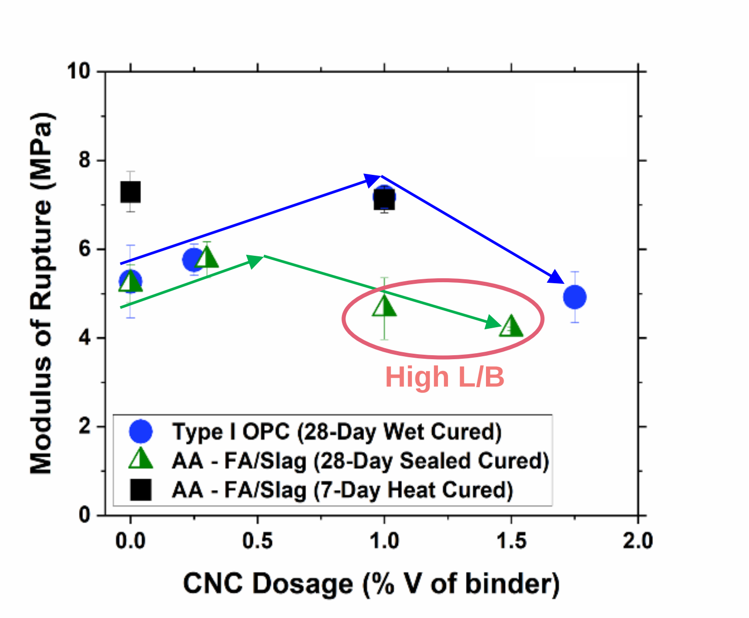

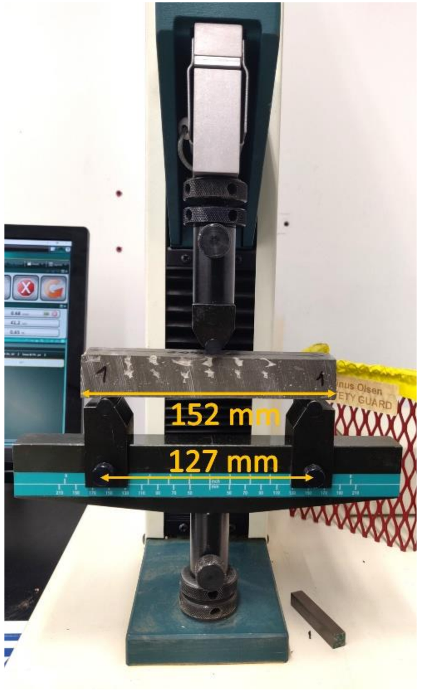

Mechanical Performance

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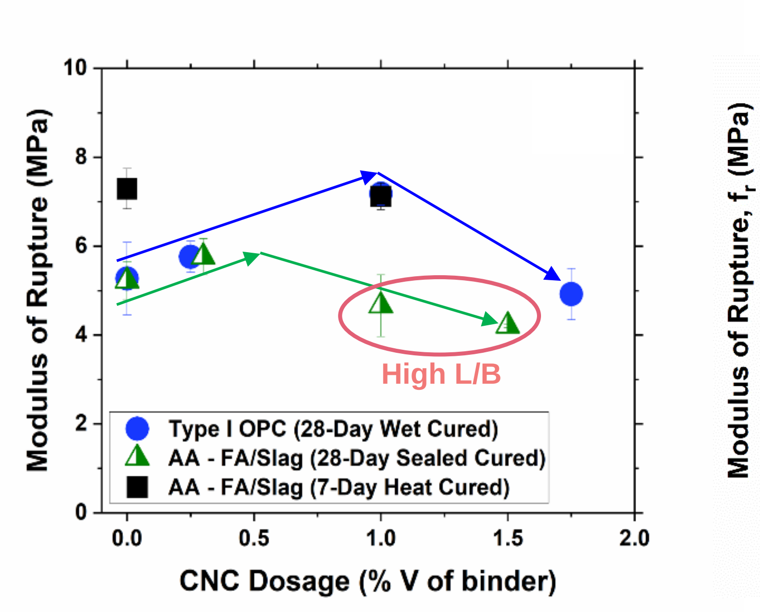

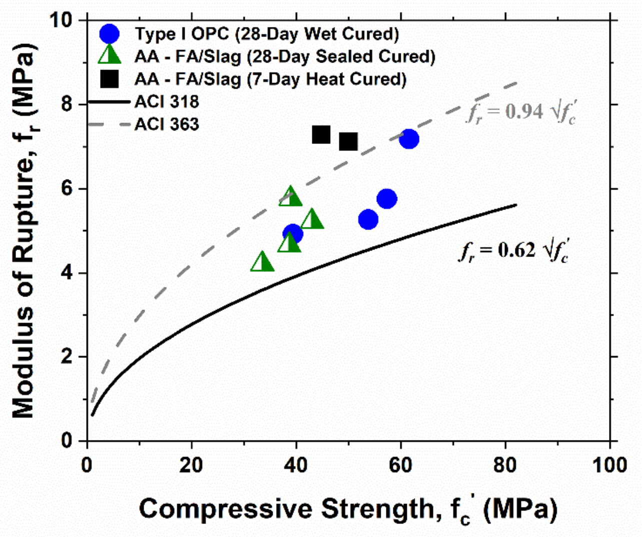

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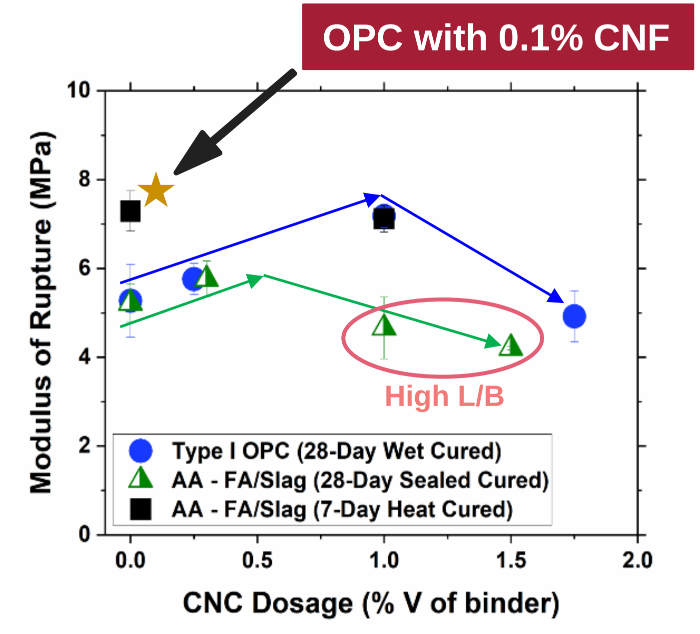

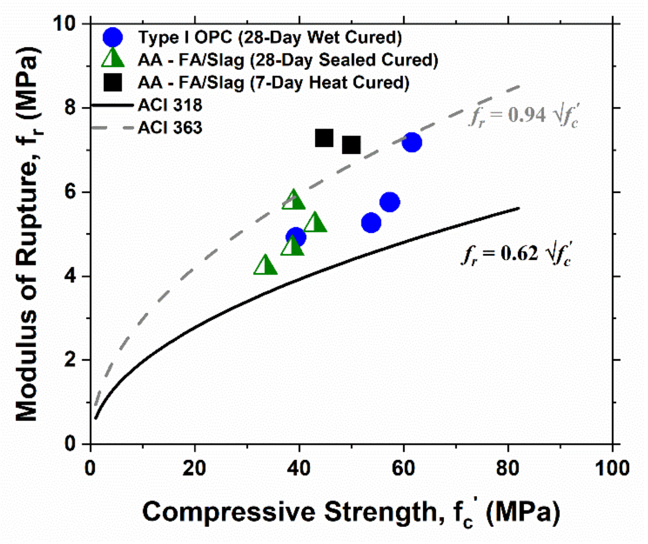

Porosity and Electrical Resistivity

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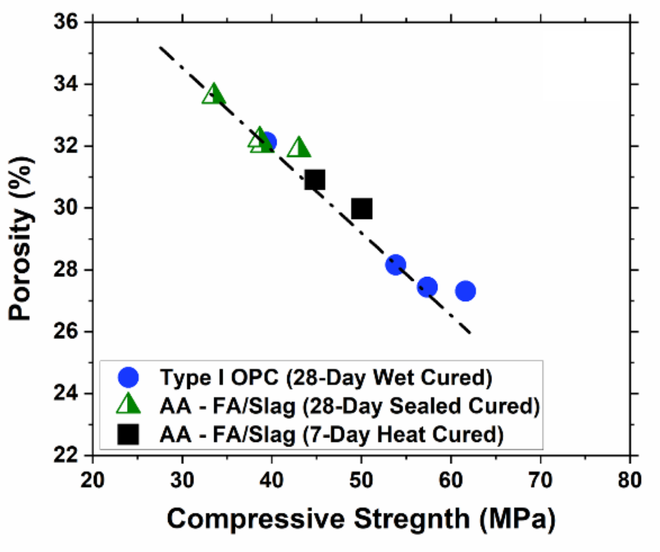
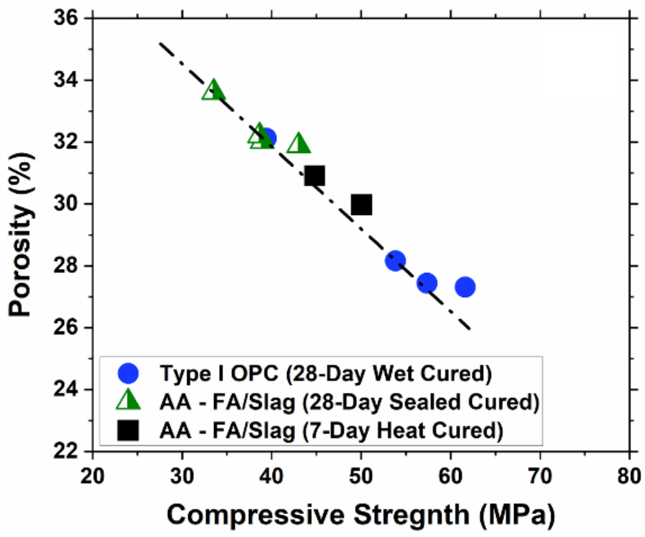
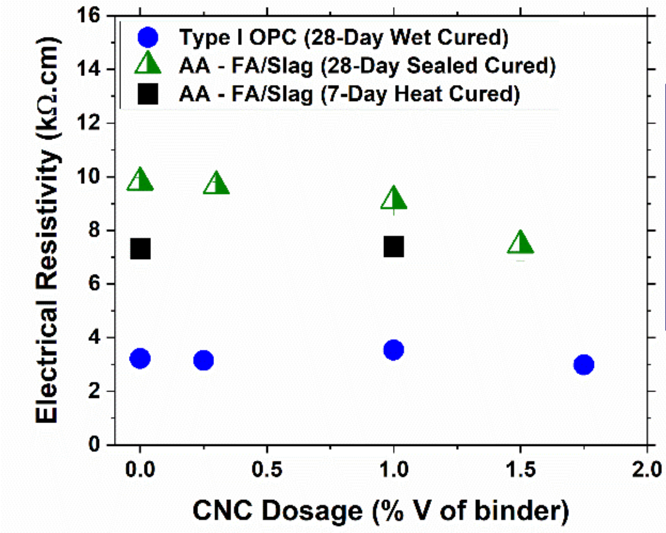
- There is a strong linear correlation between the porosity and compressive strength results



Porosity and Electrical Resistivity

- There is no significant change in the electrical resistivity of OPC samples with the addition of the different dosages of CNC.

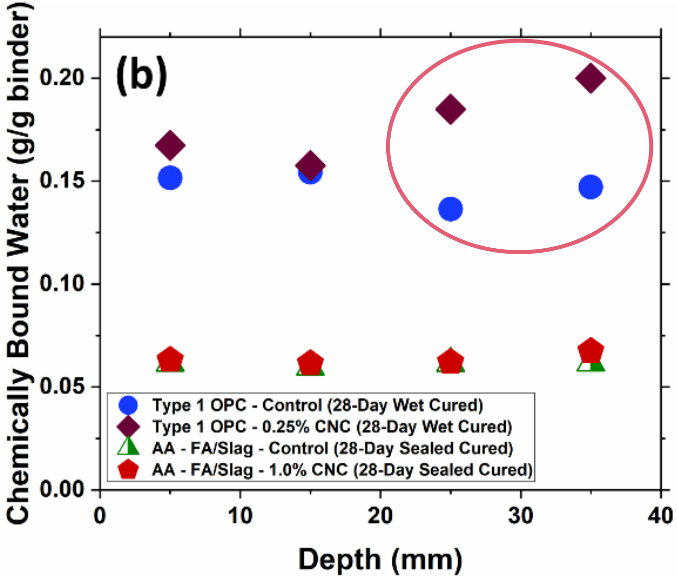




The AA samples have significantly higher electrical resistivity compared to OPC samples.

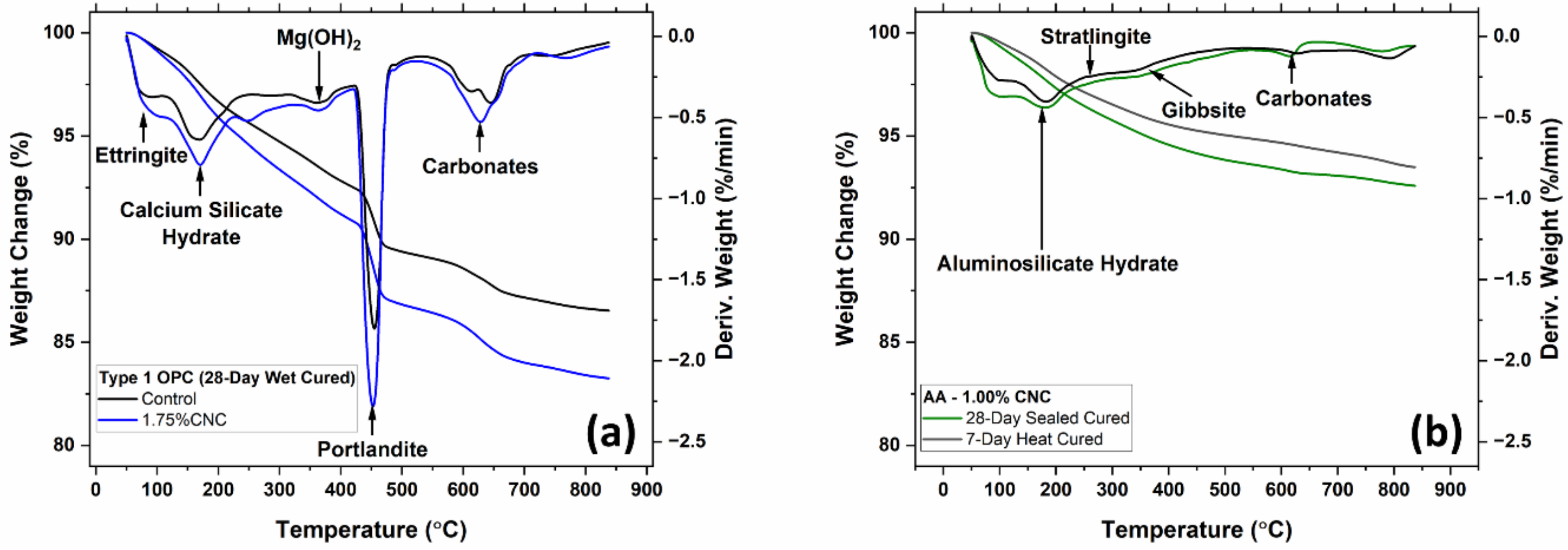
Chemically Bound Water/Hydroxyls from TGA

- OPC with 0.25% CNC approximately show a 25% increase in DOH.
- This is beneficial in low w/c systems such as 3D printed elements where the permeability of the matrix is low



Chemically Bound Water/Hydroxyls from TGA

- The OPC–CNC samples have higher chemically bound water content, suggesting that the addition of CNC improves the microstructure.



Microstructure Analysis

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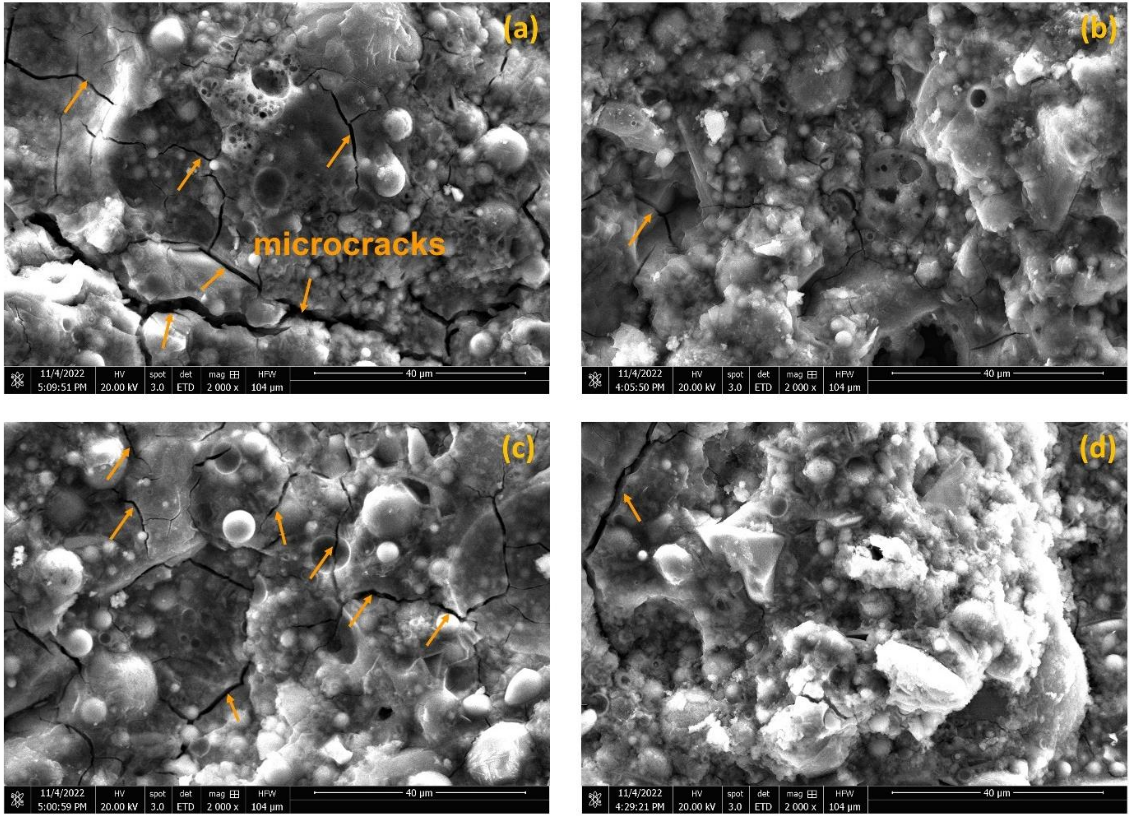
- The sealed-cured samples have a significantly higher amount of microcracks due to flexural stresses
- In heat-cured samples, the FA particles are well embedded and connected to the matrix.

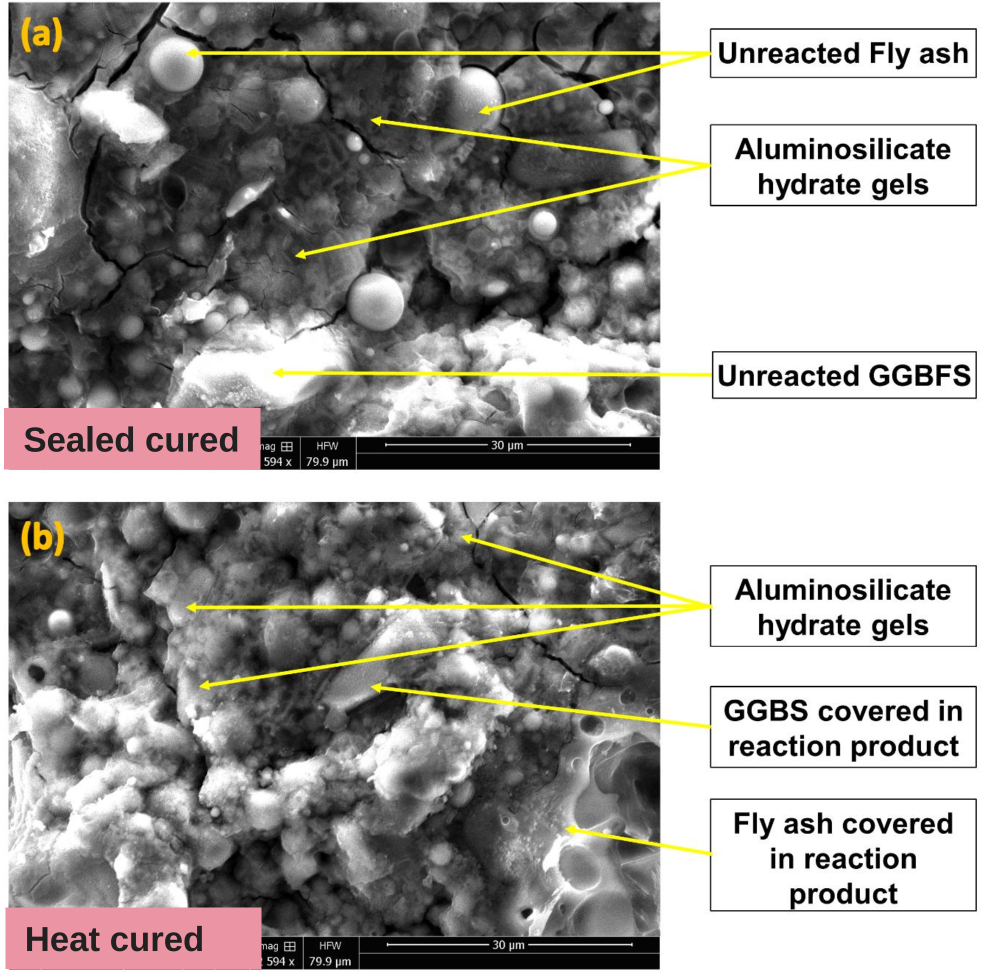
AA
Sealed cured

AA
Heat cured



Microstructure Analysis

- 28-day sealed cured samples show a higher amount of unreacted fly ash spheres
- Non-crosslinked N-A-S-H and C-A-S-H or crosslinked C-N-A-S-H are the main reaction product



Techno-Economic Analysis Inputs

- The potential to eliminate or reduce the need for chemical admixtures (e.g., viscosity modifiers)
- Impact of CN-materials on performance of 3D printed elements
- Replacing the ordinary portland cement with waste materials (fly ash and slag)
- Reductions in both the capital and operating costs potentially result from the application of the 3D printing process.

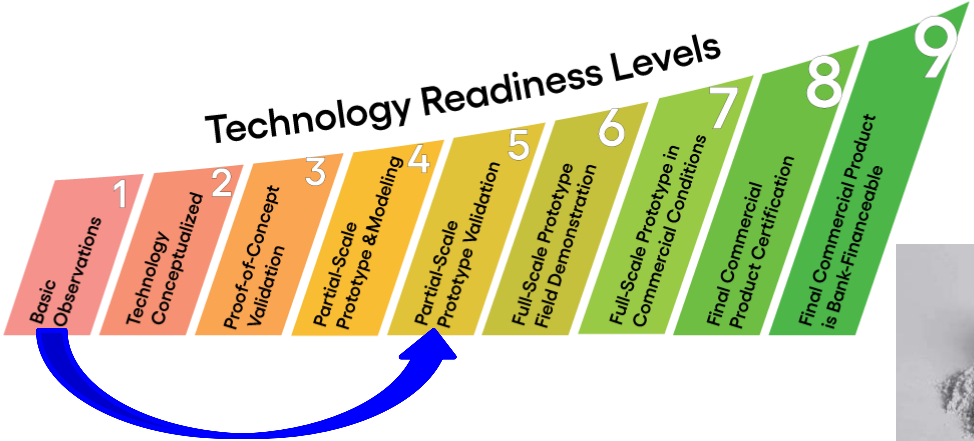
Conclusions

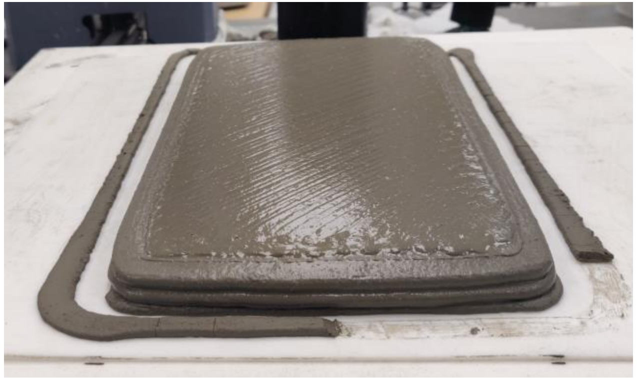
- The buildability of the AA mixtures was improved by increasing the dosage of CNC, suggesting that the CNC performs as a viscosity-modifying agent in AA mixtures.
- The inclusion of CNCs up to 1.00% (by volume of the binder) improves the overall mechanical performance and reduces the porosity of 3D-printed OPC and heat-cured AAM samples.
- The inclusion of CNC showed greater chemically bound water content for OPC samples, suggesting that the addition of CNC (below critical concentration) improves the microstructure.

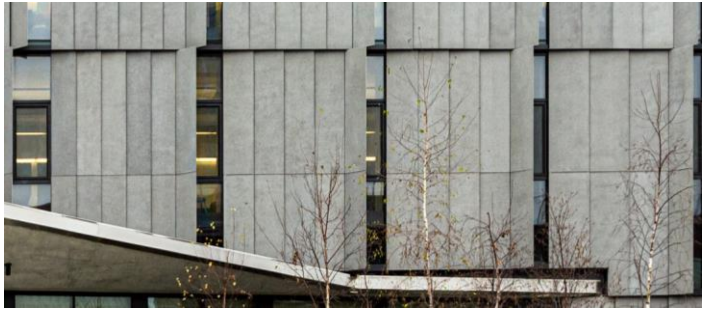
Conclusions

- The addition of CNC in OPC significantly increased the chemically bound water in the inner depths of the printed samples. This is beneficial in low water-to-binder systems such as 3D-printed elements where the permeability of the matrix is low.
- The developed printable “alkali-activated-CNC” composites can provide an overall reduction in the environmental impacts of the 3D-printed cementitious composites by eliminating/reducing the need for different chemical admixtures (e.g., viscosity modifying agents) to improve 3D-printed material consistency and stability, and replacing 100% of portland cement with fly ash and slag.

Ongoing/Future Work







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