

Hybrid Polylactic Acid (PLA) Composites: Impact of Varying the Cellulose Material (CM) Source

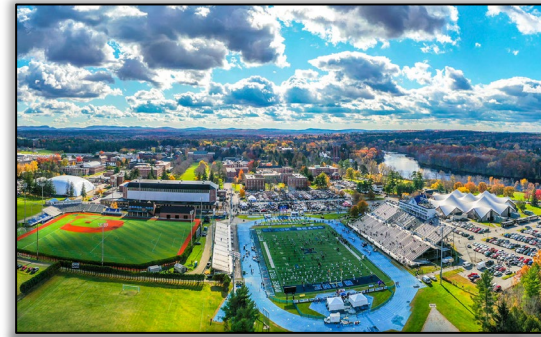
Amber M Hubbard, Katie Copenhaver, Caitlyn Clarkson,
Meghan Lamm, and Soydan Ozcan

Oak Ridge National Laboratory
Manufacturing Sciences Division
Sustainable Manufacturing Technologies Group



Hub & Spoke Program

Building a Sustainable Future



Bio-based Composites



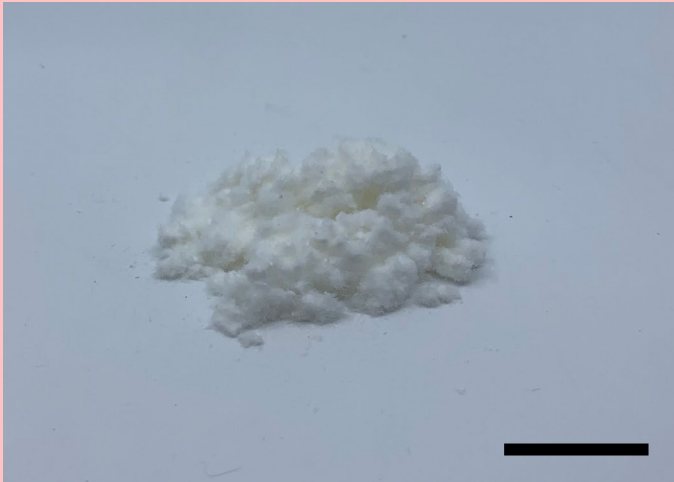


BioHome3D

- Printed at the Advanced Structures and Composites Center (ASCC) at the University of Maine
 - Revealed November 2022
- Natural Fiber Composites for Additive Manufacturing
 - Reduced Coefficient of Thermal Expansion (CTE)
 - Increased Heat Deflection Temperature (HDT)



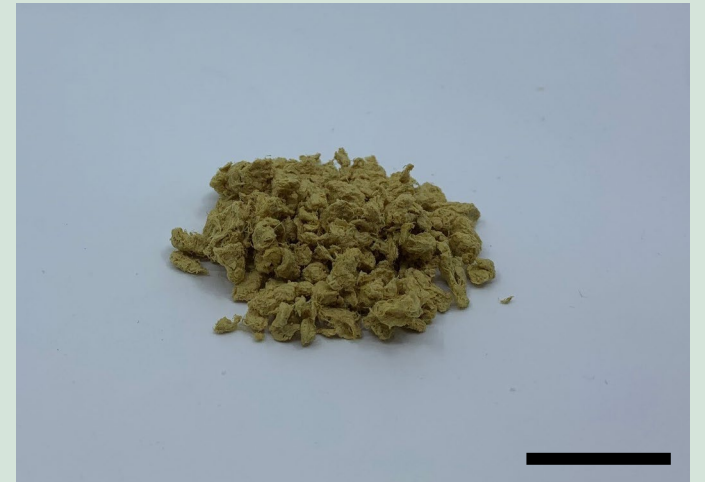
Creafill



Flax

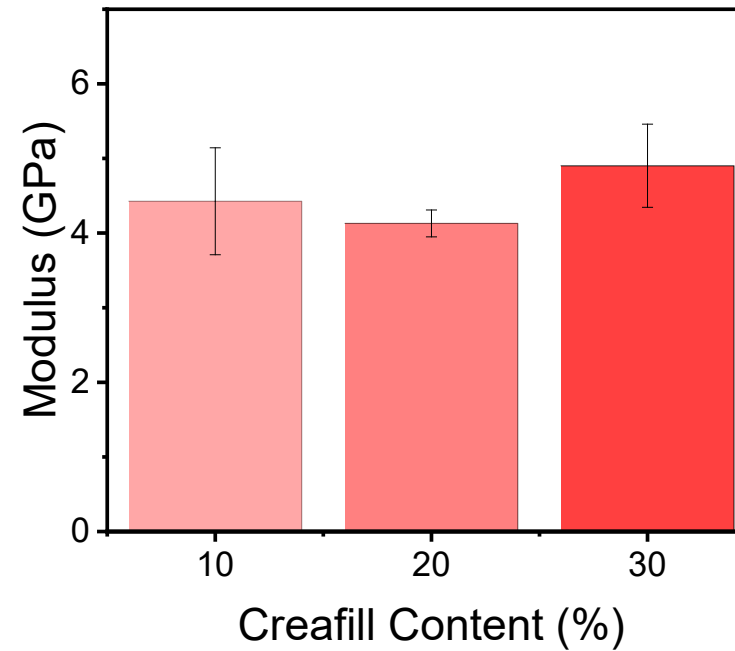
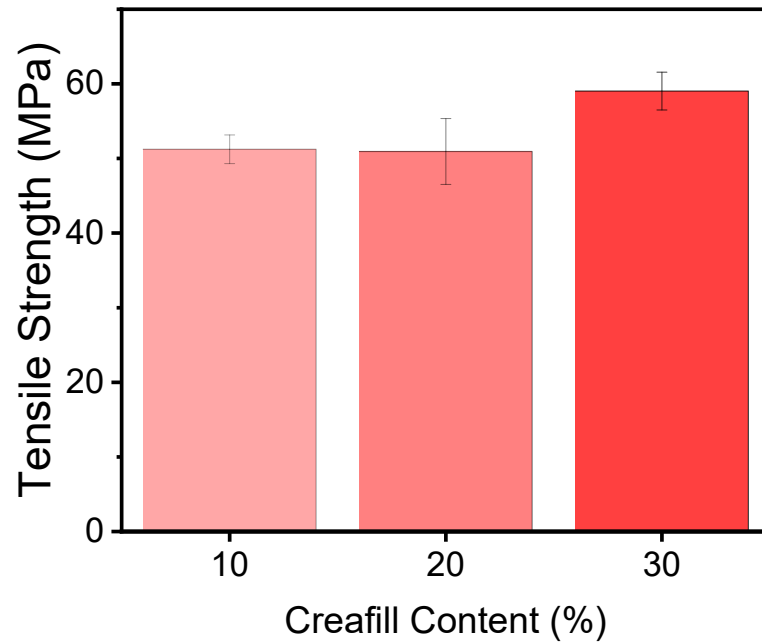


Wheat Straw



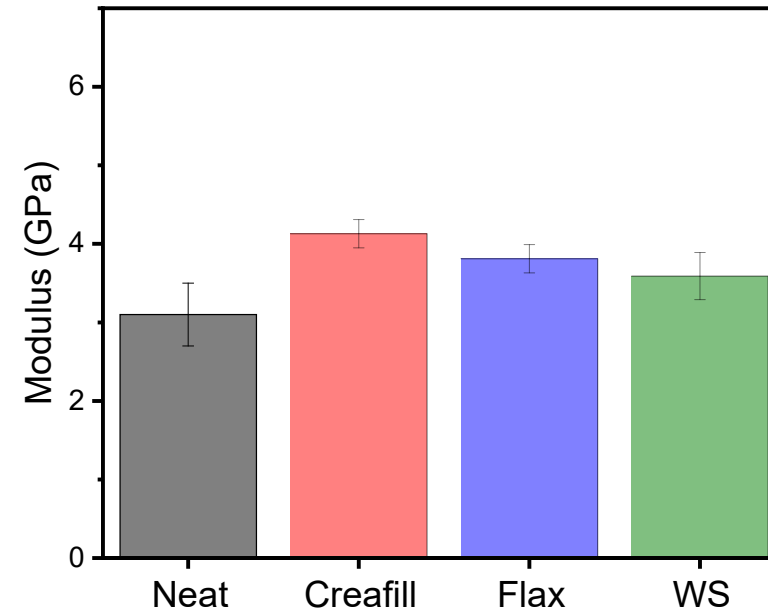
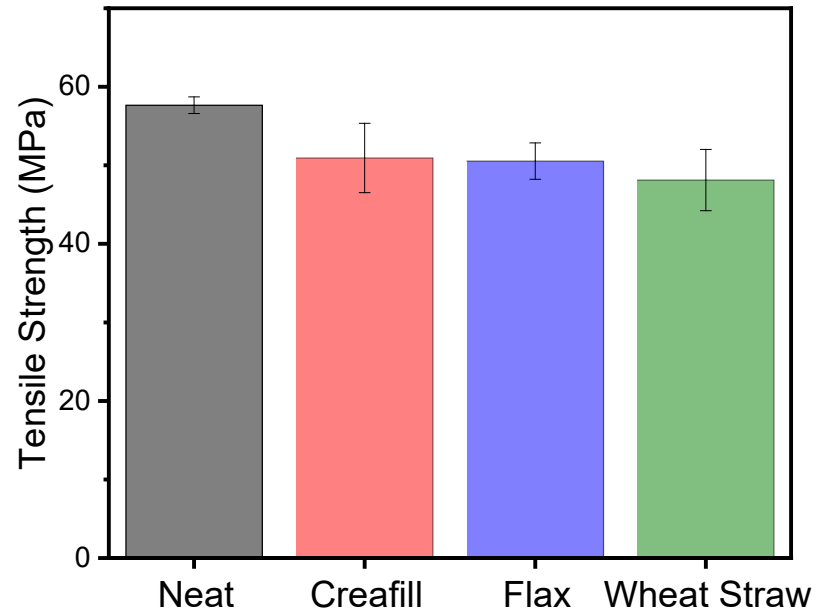
Macrofibers

Varying Filler Concentration

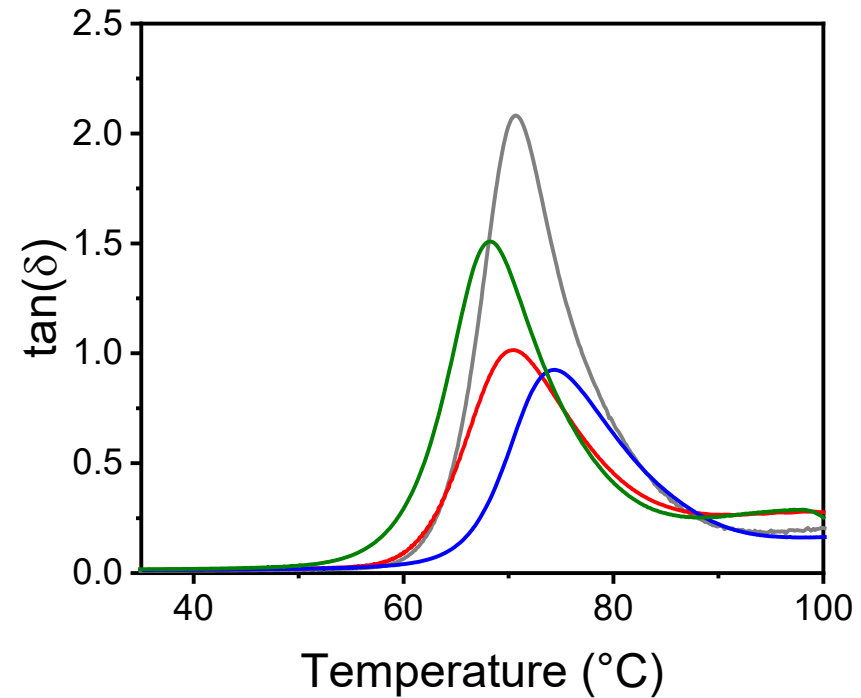
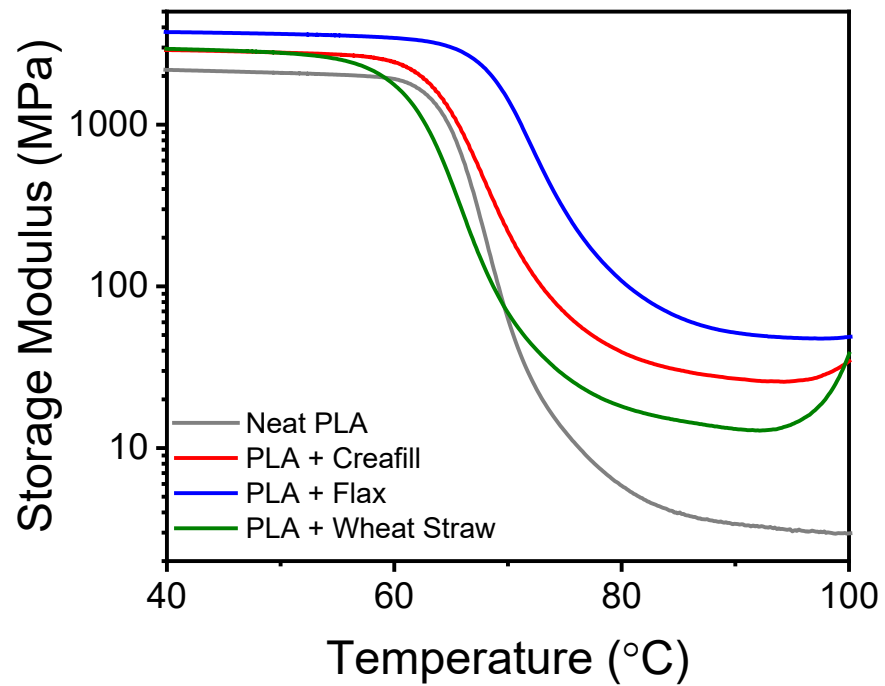


Takeaway: There is an optimum amount of filler to maximize both strength and modulus.

20 wt% filler content (historically used) chosen as optimum

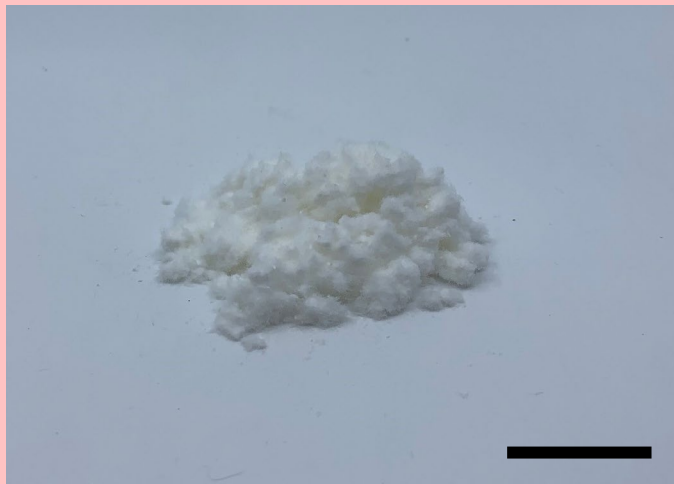


Takeaway: Natural fiber addition (20wt% fiber) into PLA results in increased modulus and slightly decreased strength.

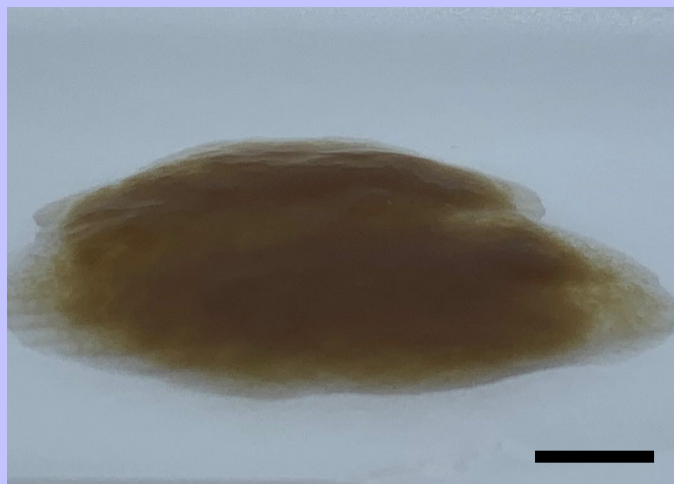


Takeaway: Dynamic mechanical analysis confirms an increase in modulus with filler, regardless of temperature.

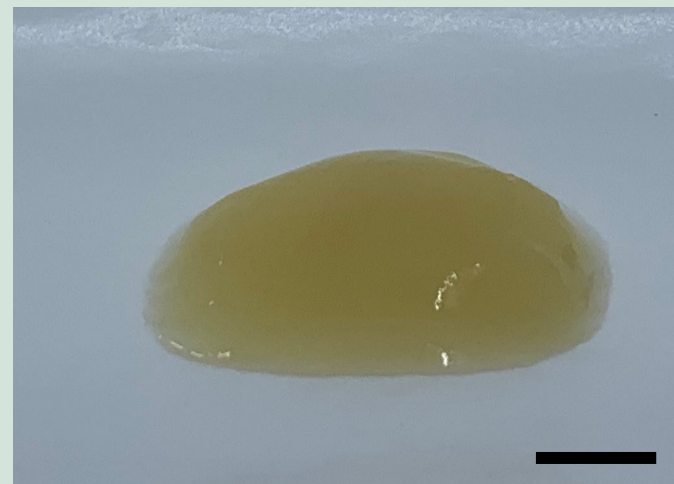
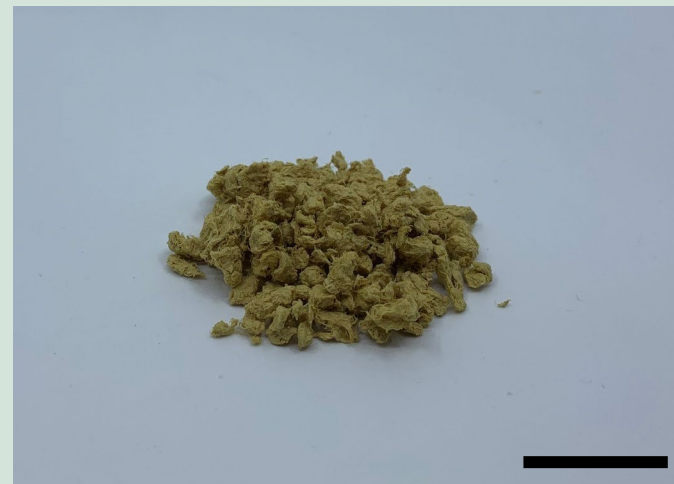
Creatin



Flax



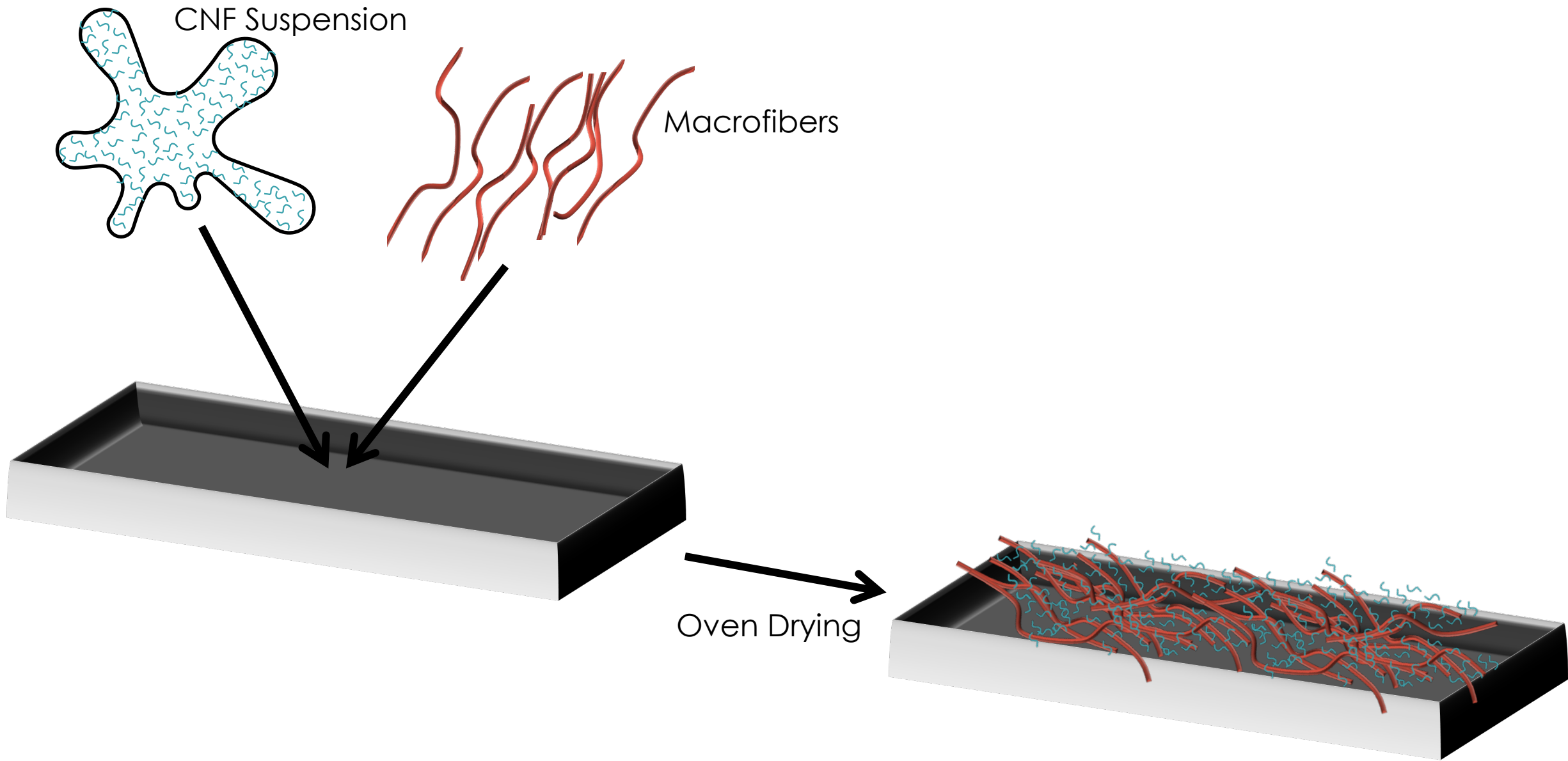
Wheat Straw



Macrofibers

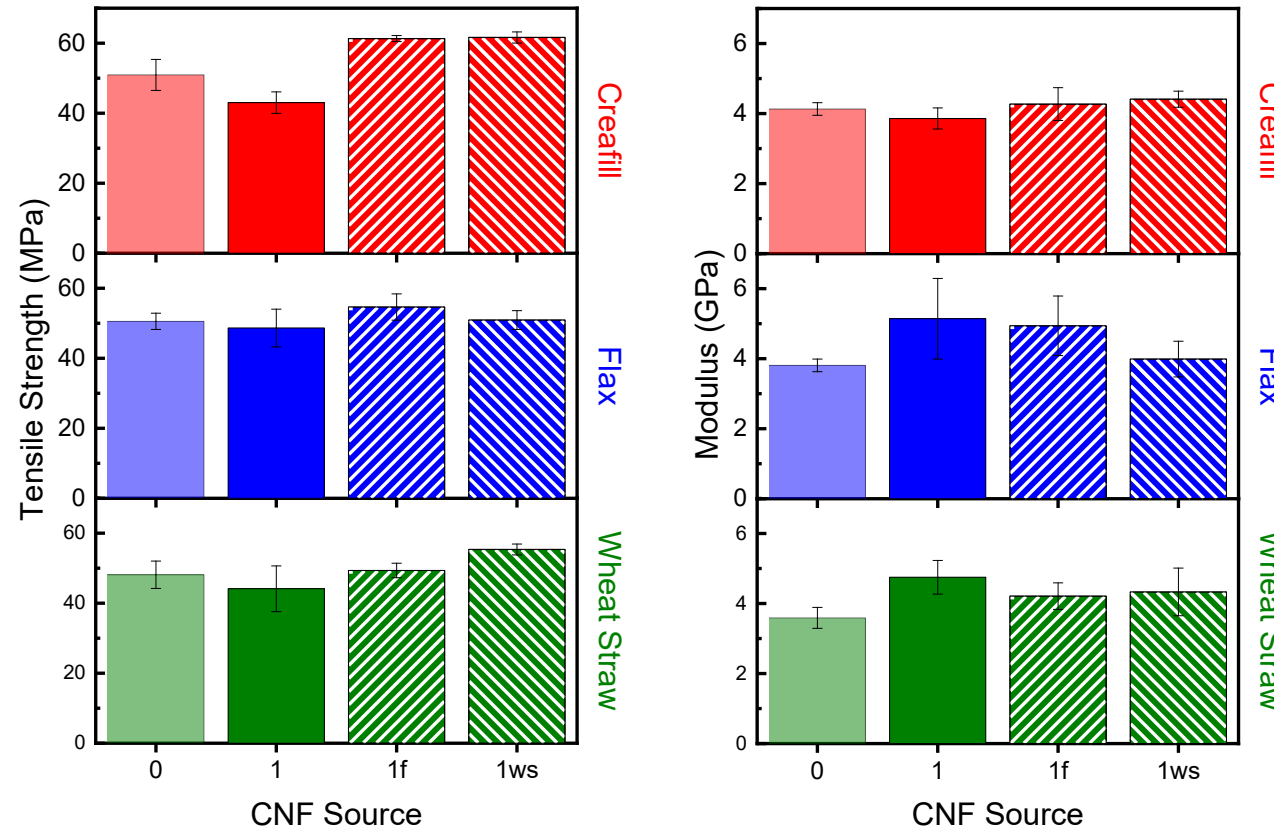
CNF Suspension

Scale bars: 1 cm



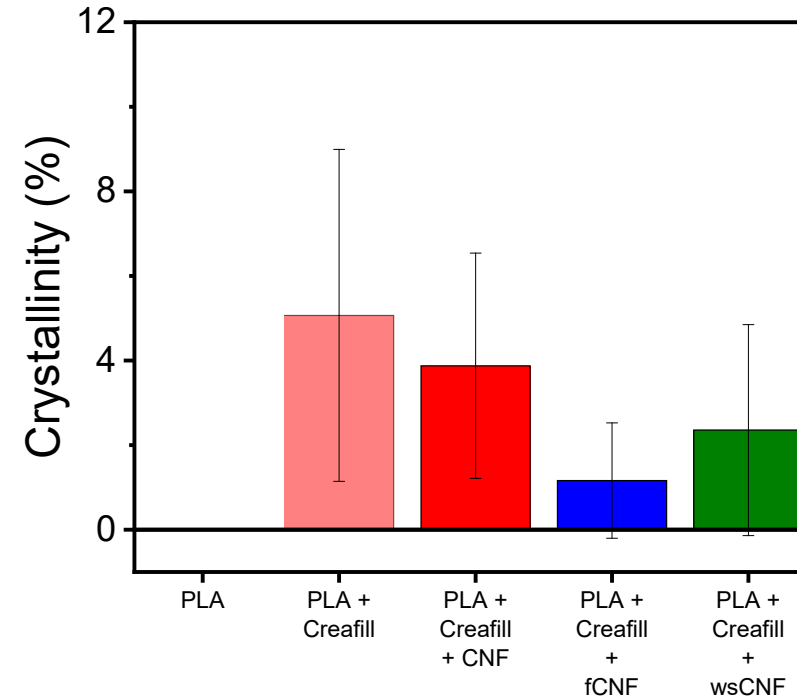
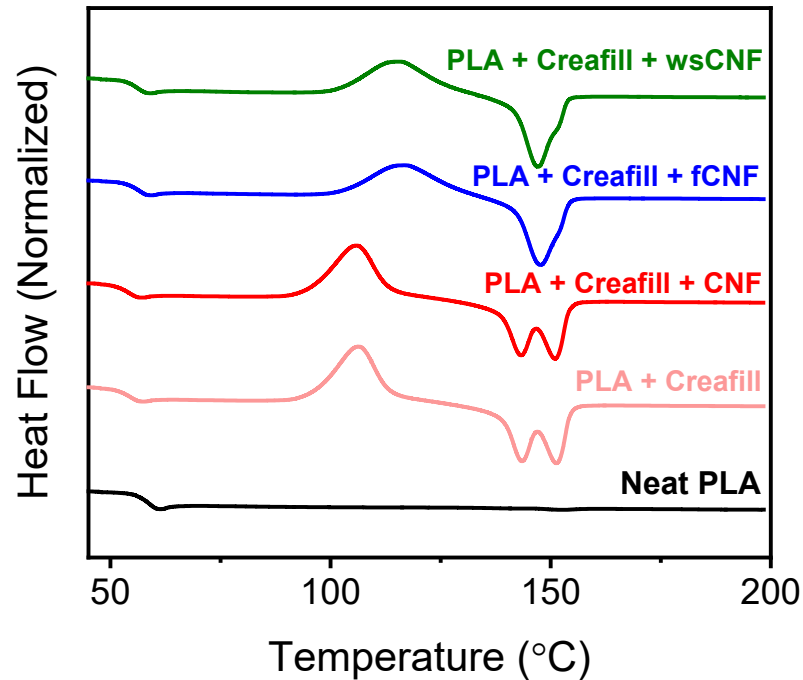
Hybrid Fibers = Macrofiber + CNF





Takeaway: Hybridization of fibers increases both tensile strength and modulus.

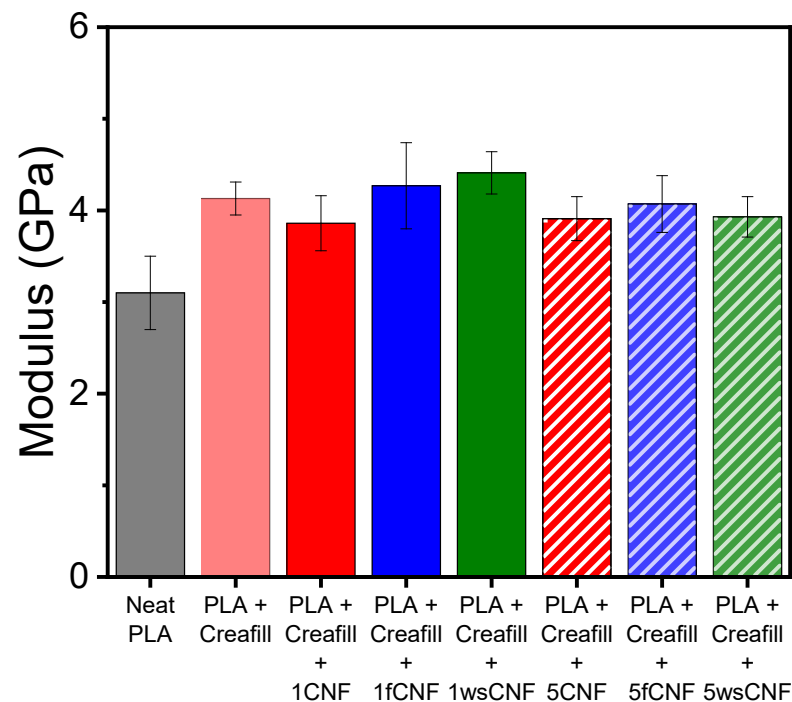
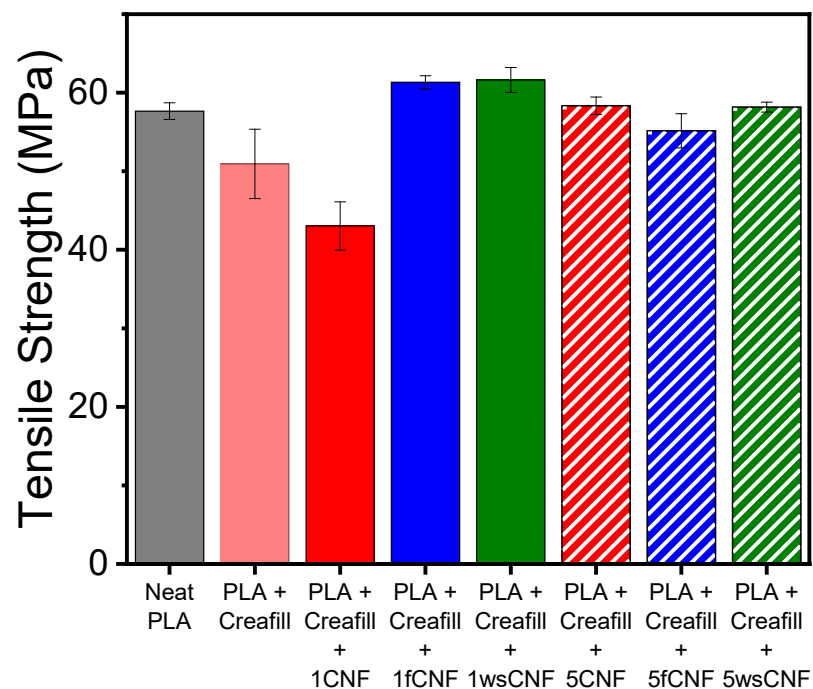
Hypothesis: Both topology and surface chemistry impact the adhesion between the filler and matrix.



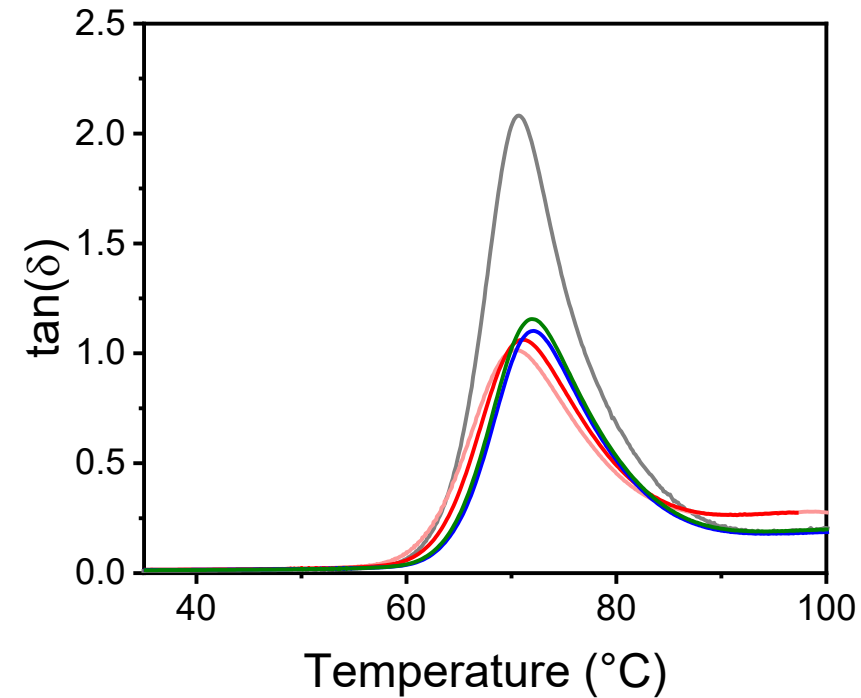
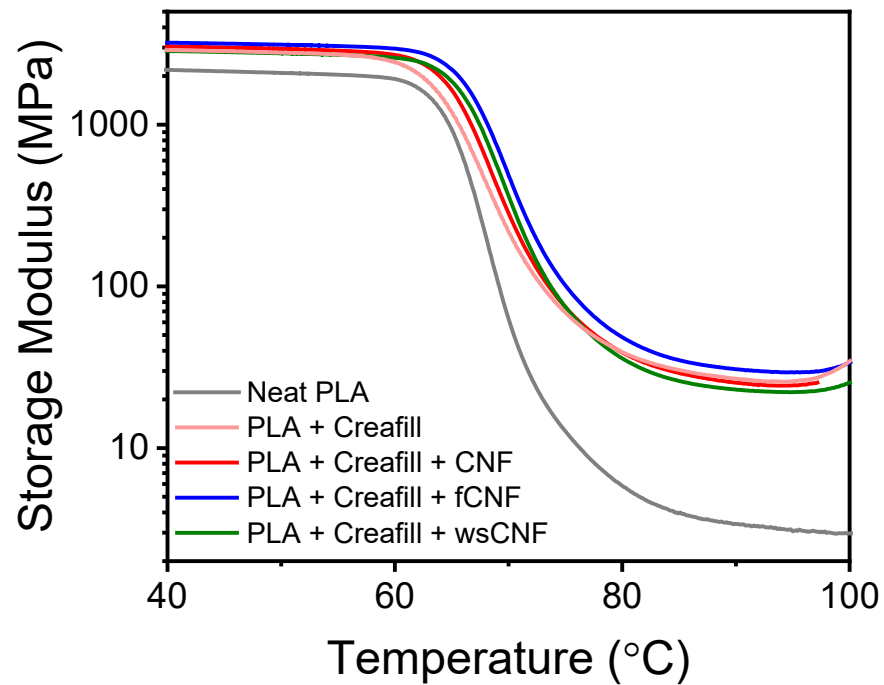
Takeaways: The ideal hybrid filler composition is **Creafill + fCNF** or **Creafill + wsCNF** based on minimized crystallization and increased strength/modulus.

There is a shift to higher T_{cc} and a suppression in the melting peak for these compositions.

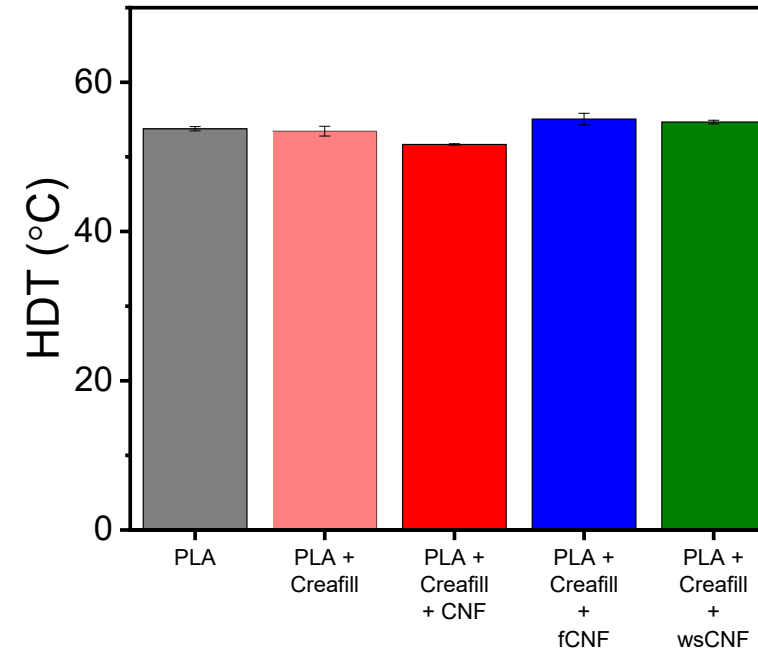
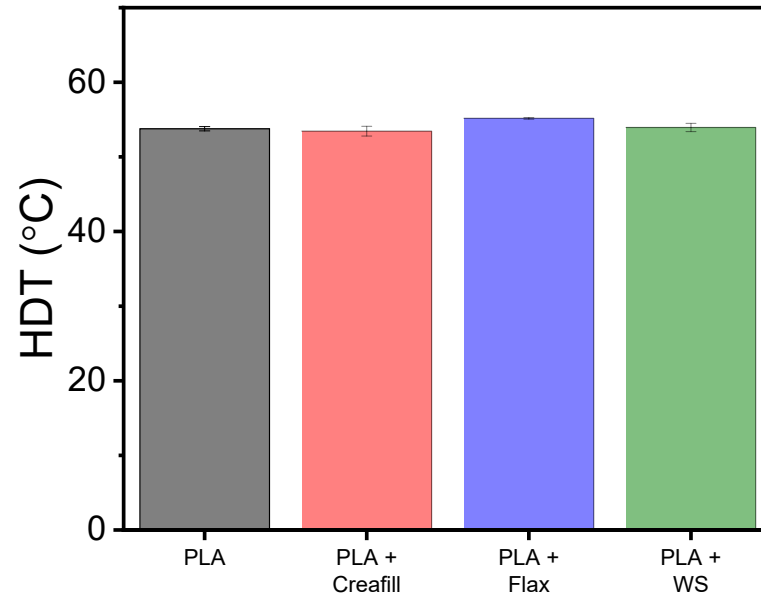
Varying CNF Source and Concentration



Takeaway: There is an optimum amount of CNF addition (1%) for fiber hybridization to maximize both strength and modulus.

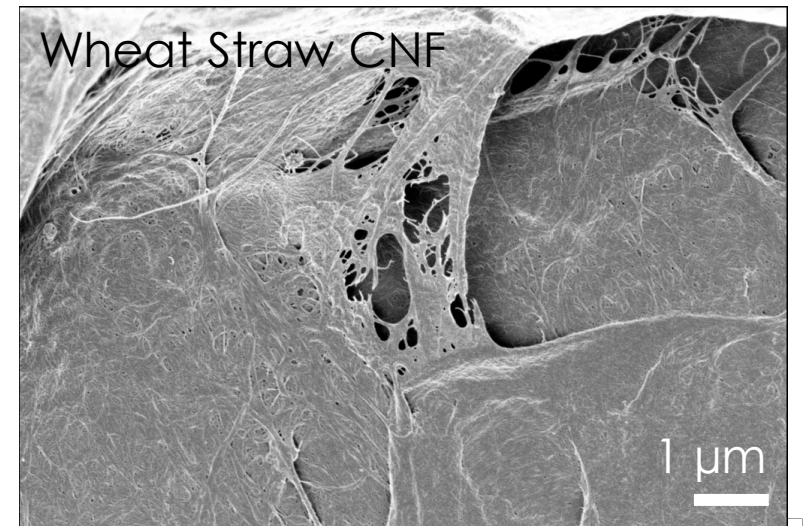
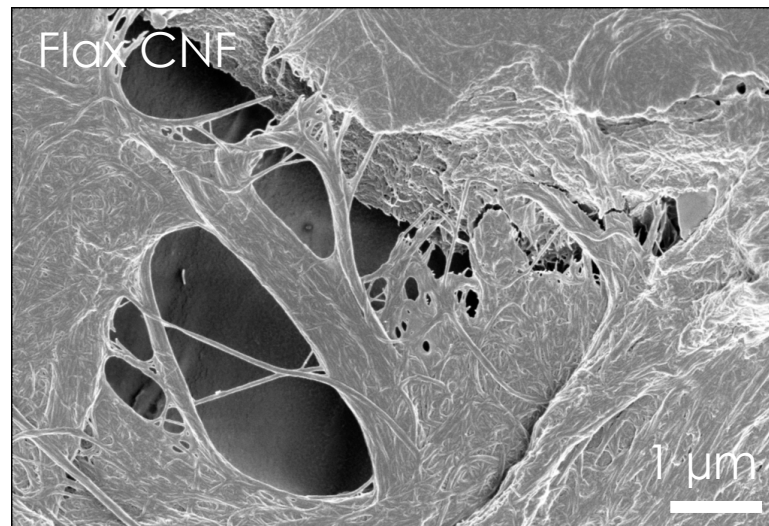
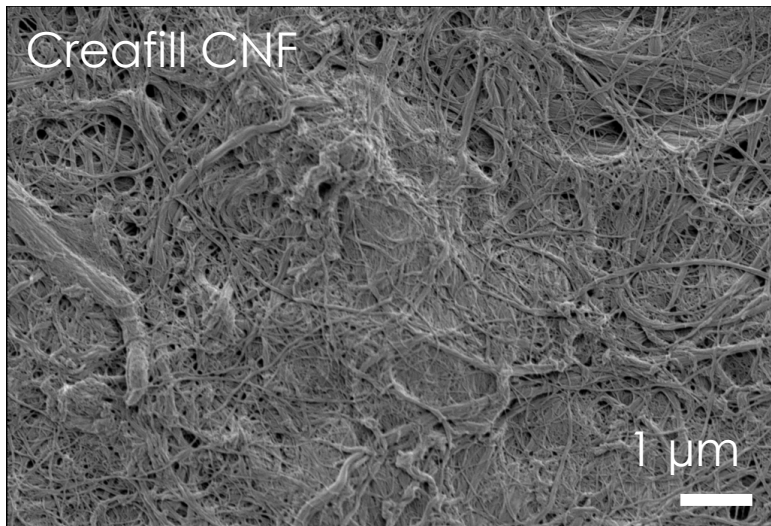
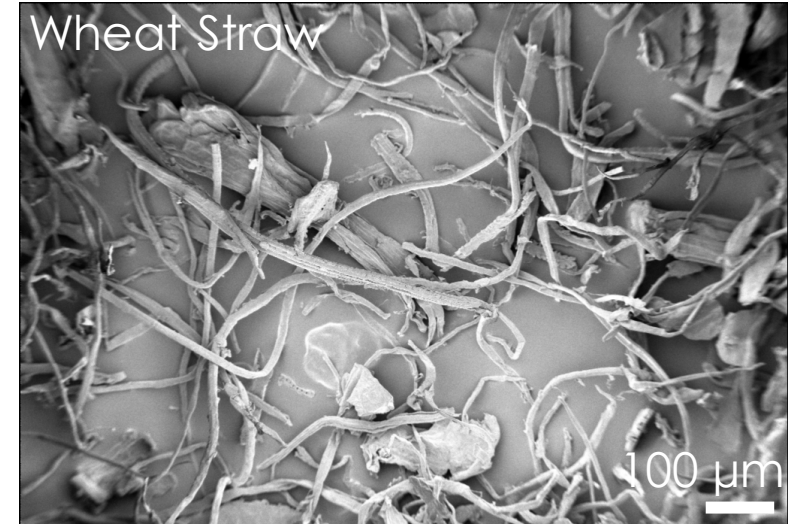
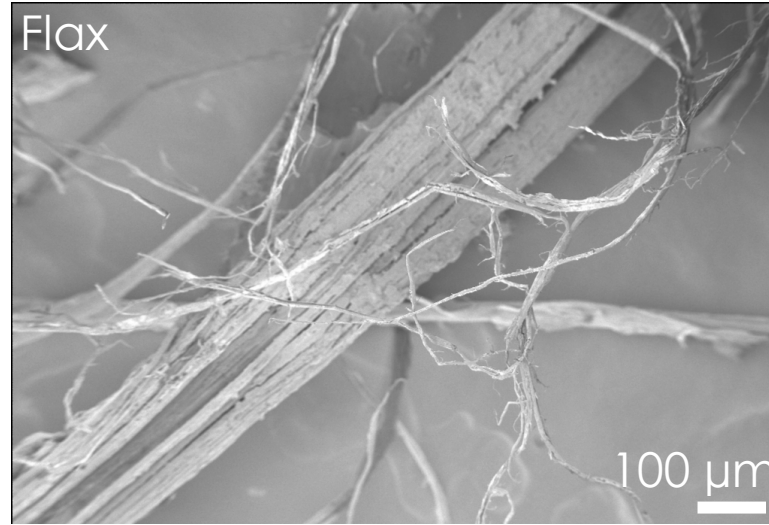
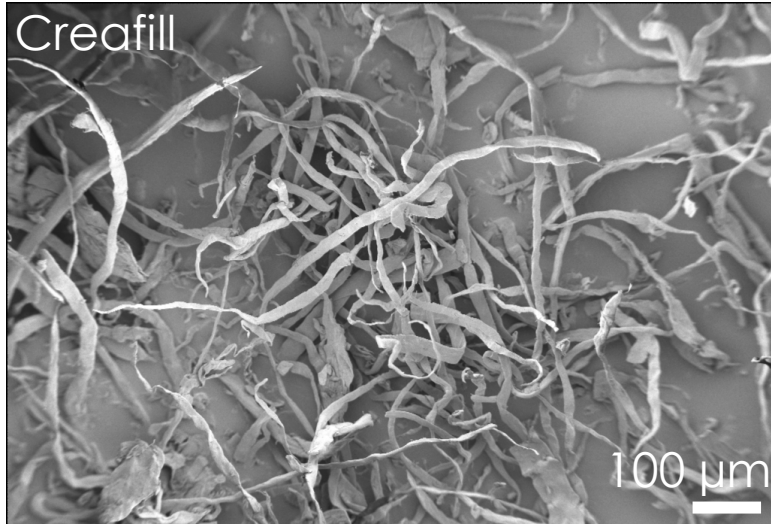


Takeaway: Dynamic mechanical analysis confirms no appreciable difference in modulus for all Creafill hybridized composites.

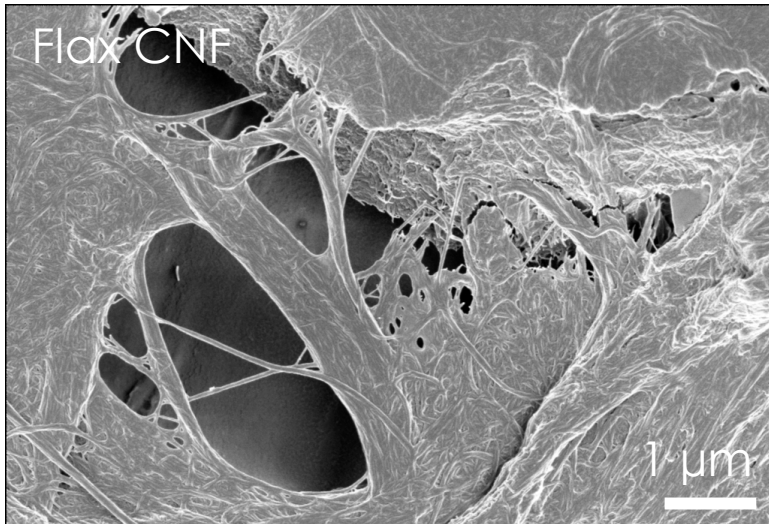
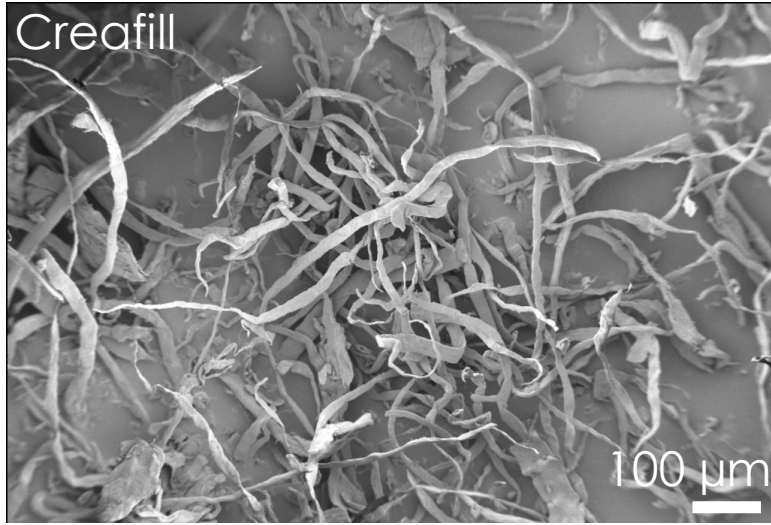


Takeaway: There is minimal variability in heat deflection temperature (HDT) regardless of filler type.

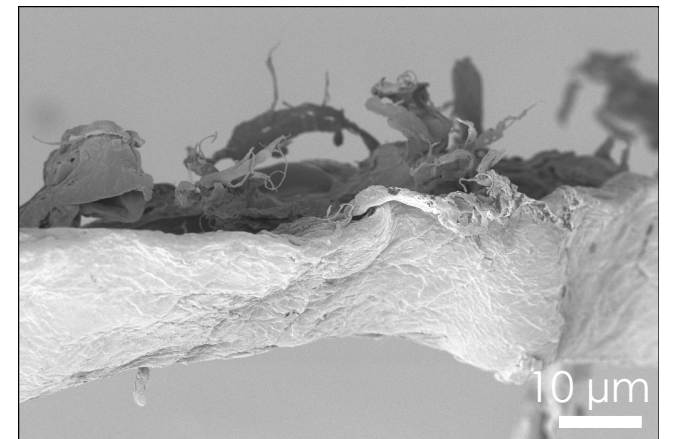
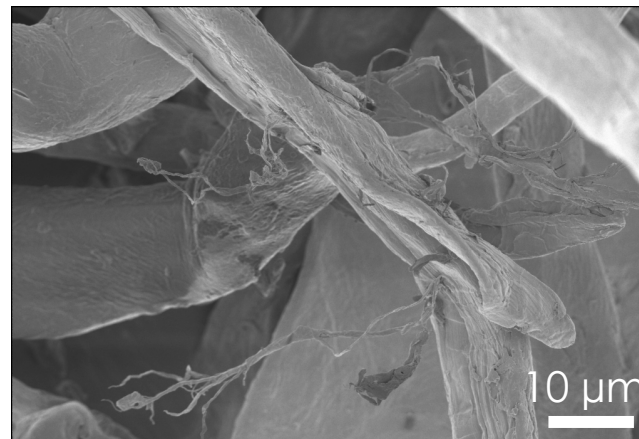
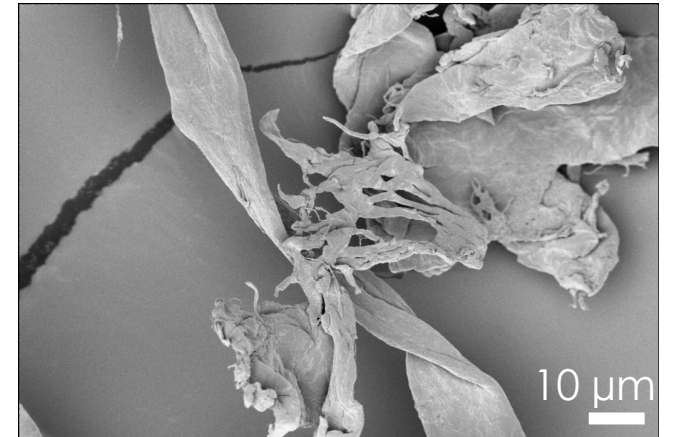
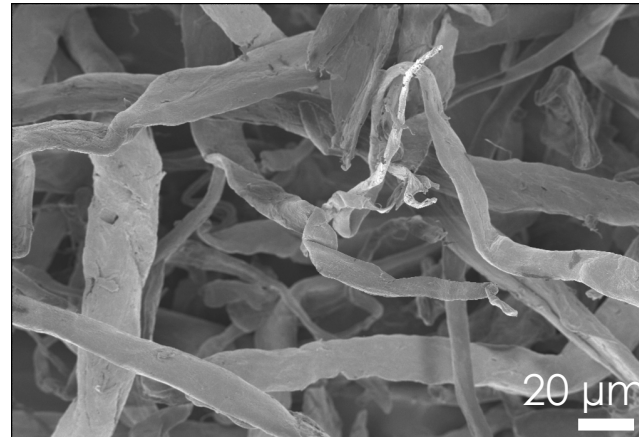
Hypothesis: Both topology and surface chemistry impact the adhesion between the filler and matrix.

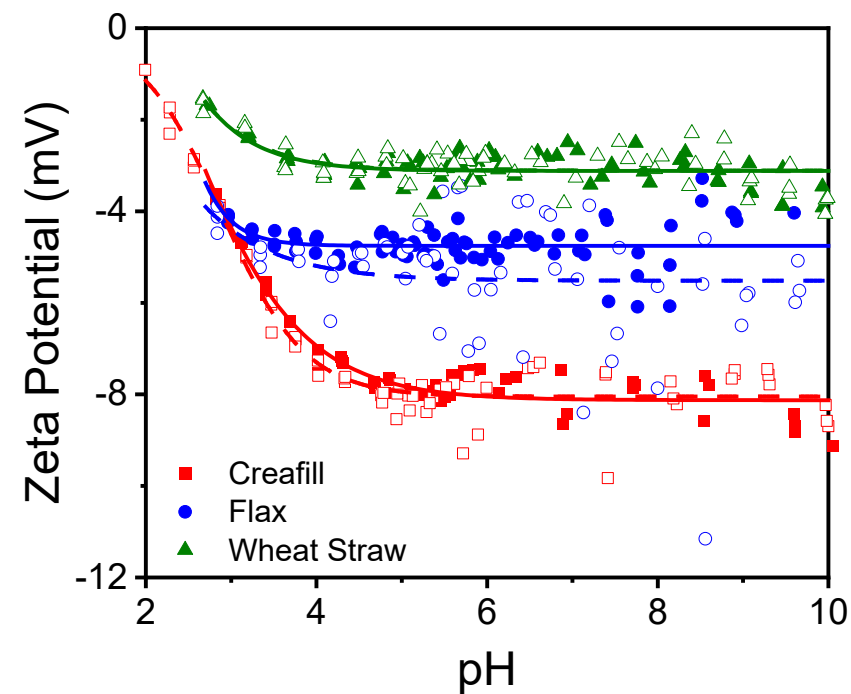


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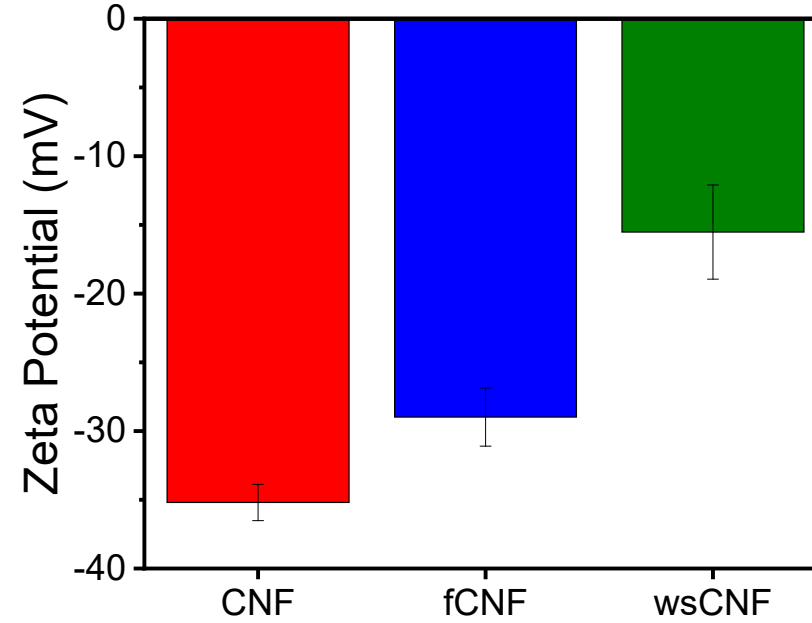


Hybridization





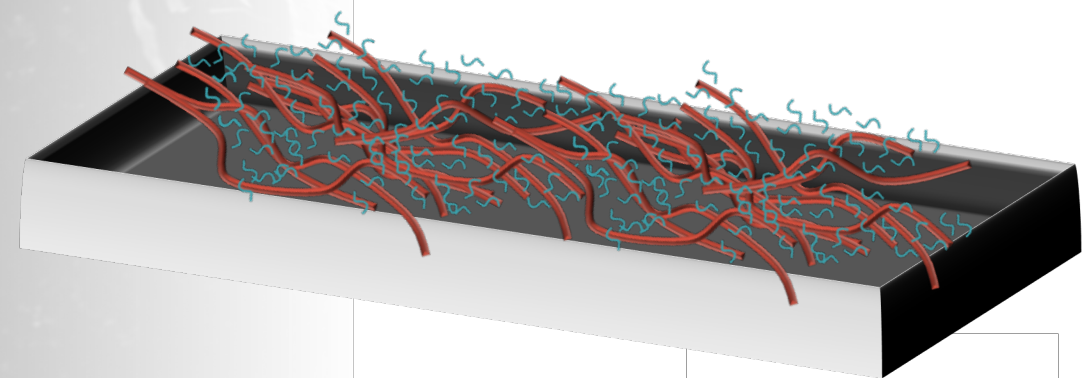
Takeaway: Most Hydrophilic → Least Hydrophilic
Wheat Straw → Flax → Creafill



Takeaway: Most Hydrophilic → Least Hydrophilic
Creafill → Flax → Wheat Straw

Conclusions

- Hybrid fillers increase the strength and modulus of PLA composites.
- CNF sourced from flax (fCNF) and wheat straw (wsCNF) outperforms CNF sourced from bleached pulp (CNF) as a composite reinforcement.



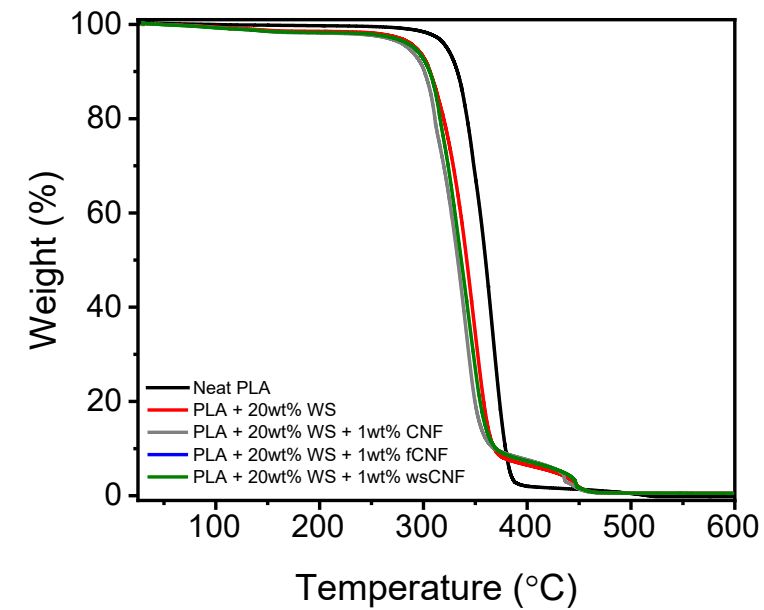
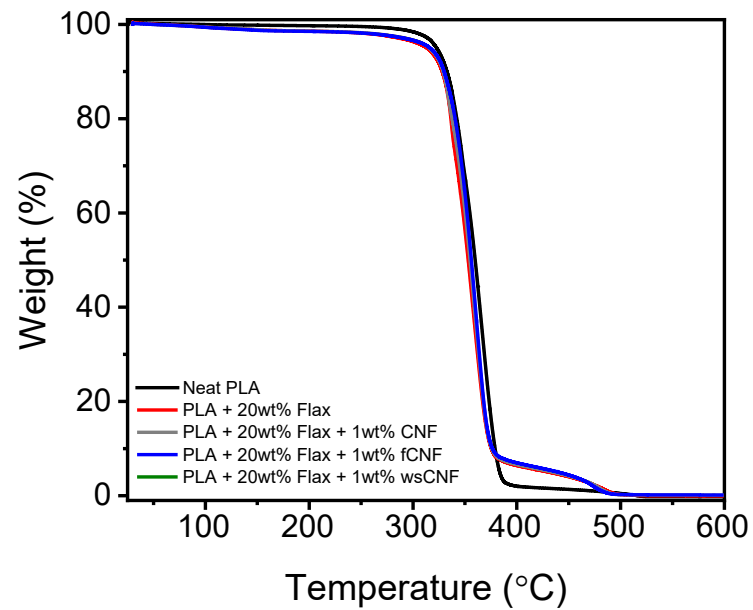
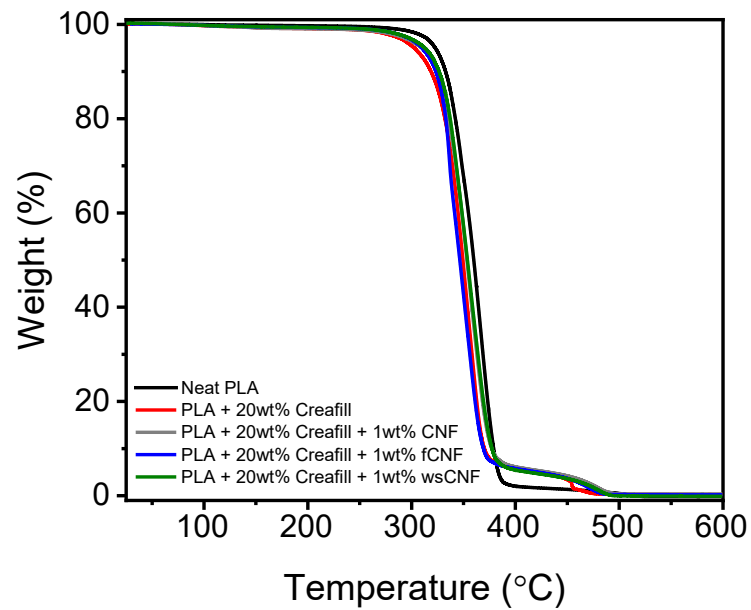
A scanning electron micrograph (SEM) showing a cross-section of a plant stem. The image reveals a complex, fibrous internal structure with various layers and textures, including what appears to be a vascular bundle. The stem is light-colored against a dark background.

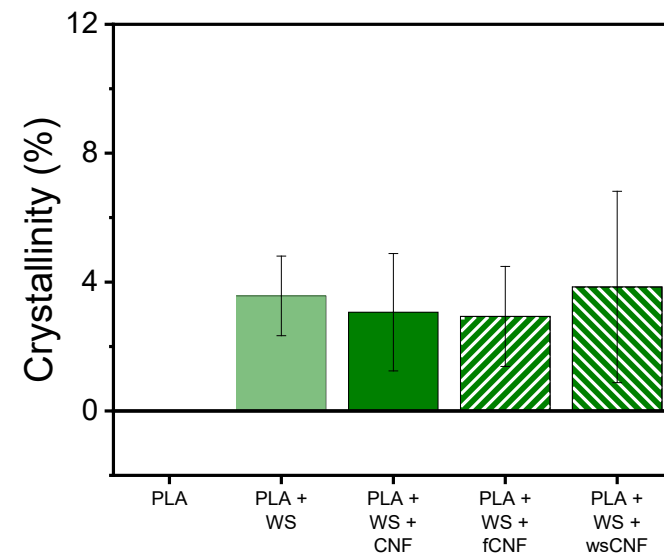
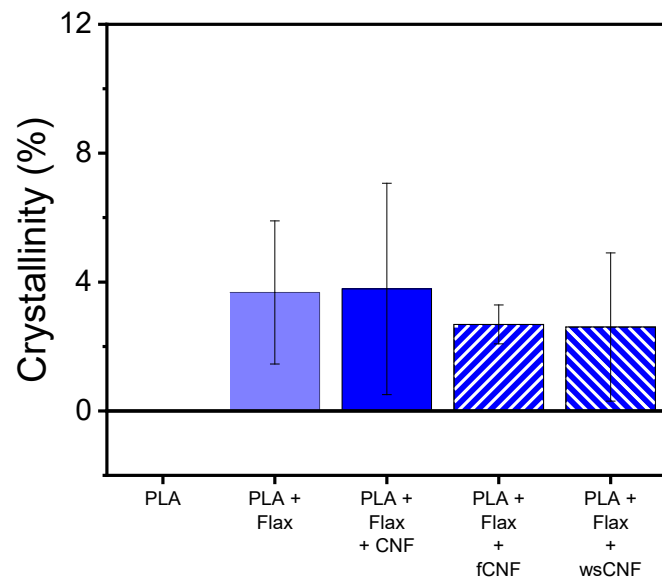
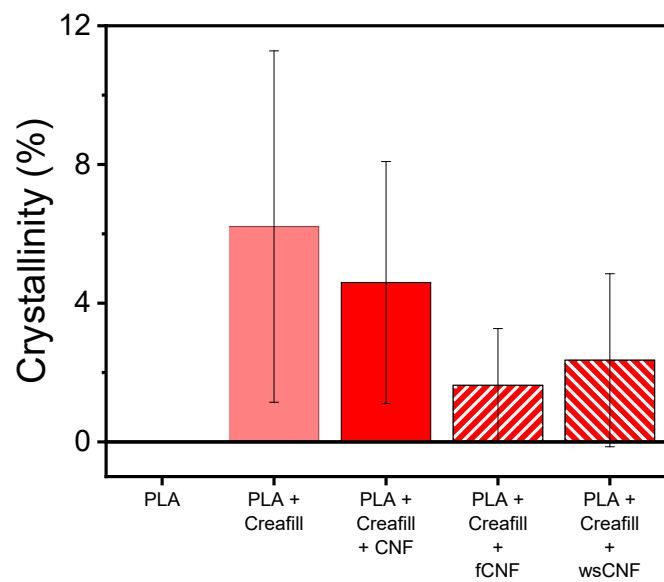
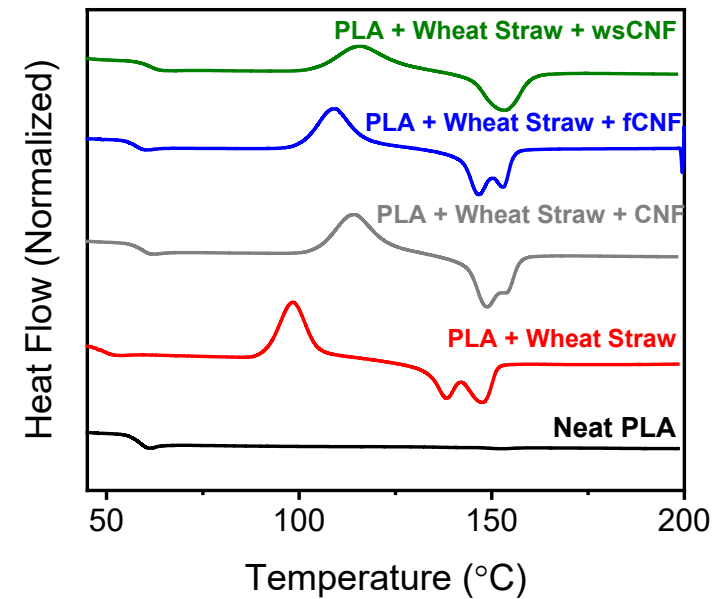
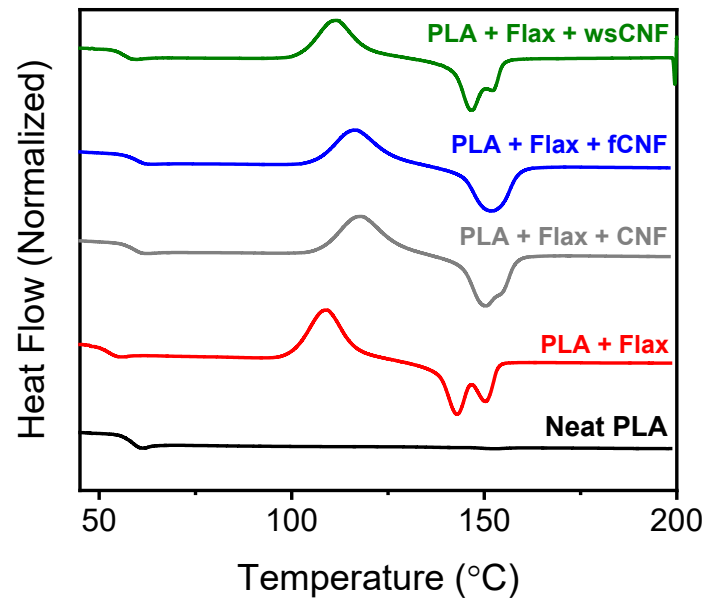
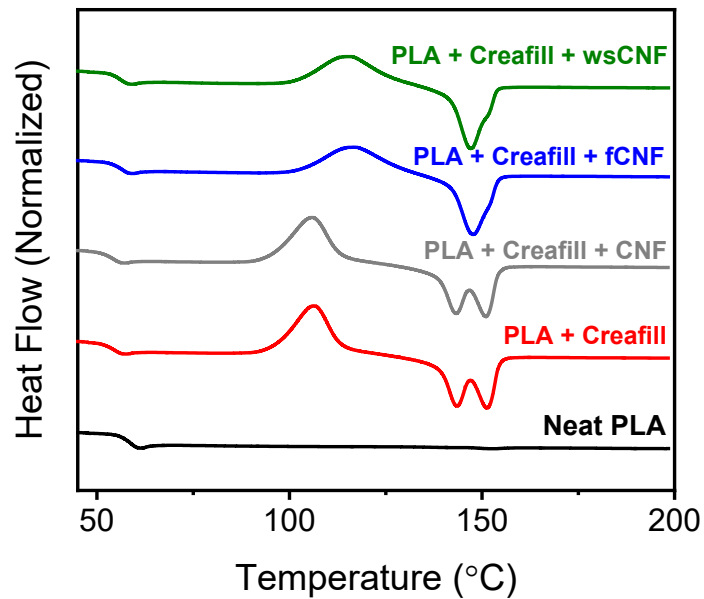
Questions?

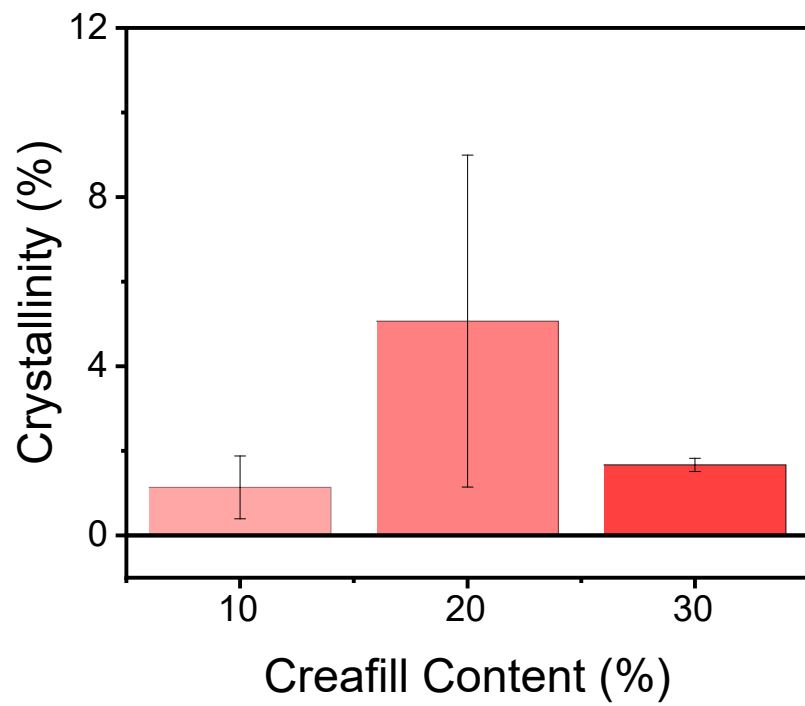
Amber M Hubbard
hubbardam@ornl.gov

Abstract

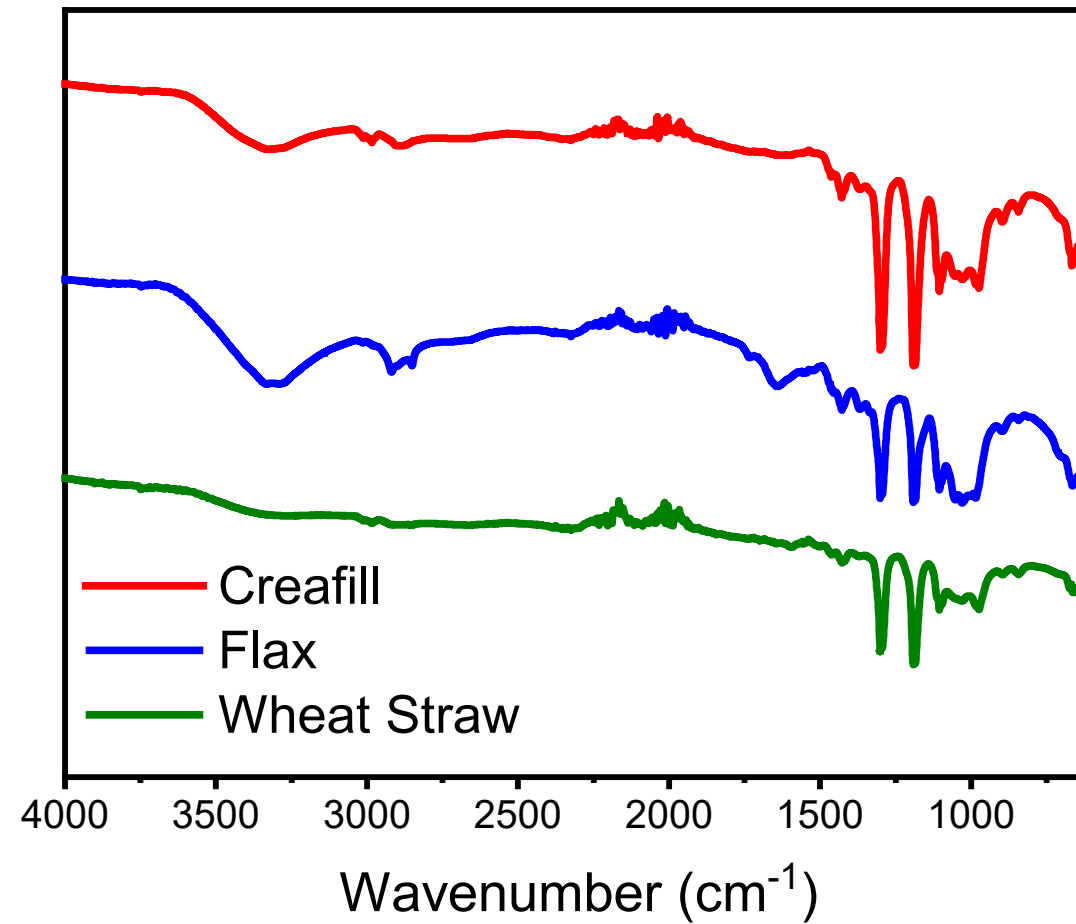
Thermoplastic composites are of significant interest in a variety of industries ranging from automotive to construction as they present a recyclable, low cost, and lightweight alternative material without sacrificing performance. Additionally, hybrid composites which contain more than one type of reinforcement (*e.g.*, natural fibers and cellulose materials (CM)) have gained increasing attention where target properties are enhanced via composite formulations. Herein, three different fibers (*i.e.*, creafill, flax, and wheat straw) are mixed with three different CM and incorporated into polylactic acid (PLA) via melt compounding. PLA was chosen as the matrix material for all hybrid composites due to its industrial compostability and its demonstrated applicability in 3D printing. The mechanical and thermomechanical properties of the resulting thermoplastic, hybrid composites are reported where the composite strength and modulus exhibit marked increases compared to the non-hybrid composites. In addition, by matching the fiber and CM, the material demonstrates further improvements; the mechanism for property enhancement is explored as a function of CM. Finally, these hybrid composites are formulated and targeted for automotive and tooling applications.



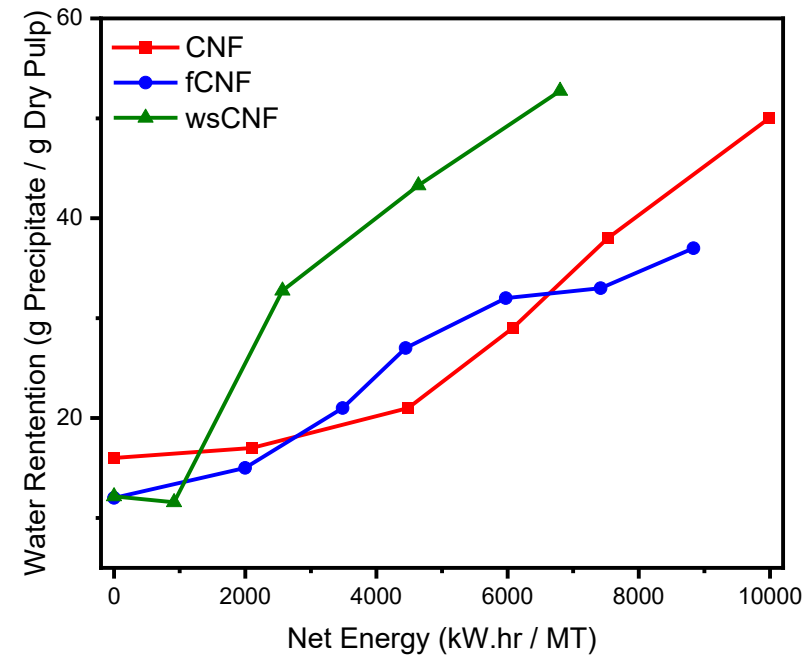
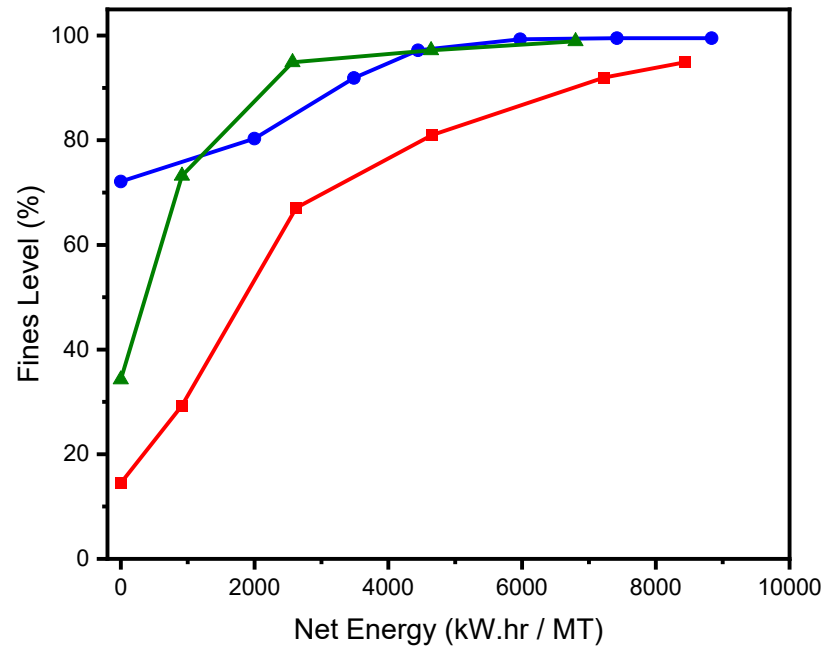




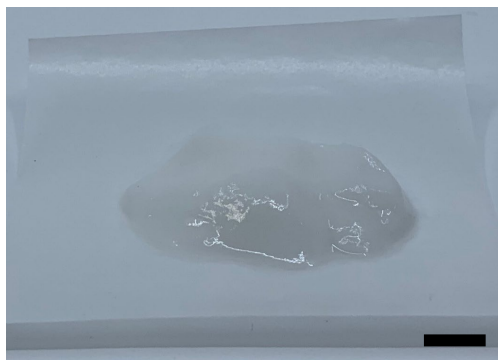
FTIR of Neat Macrofibers



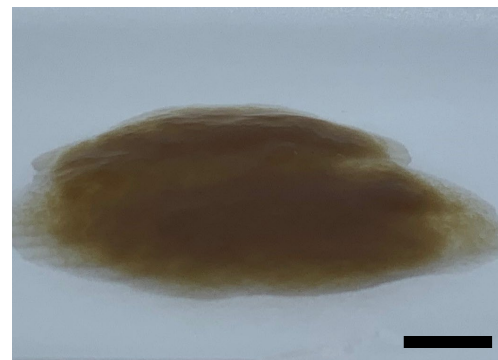
CNF Characterization



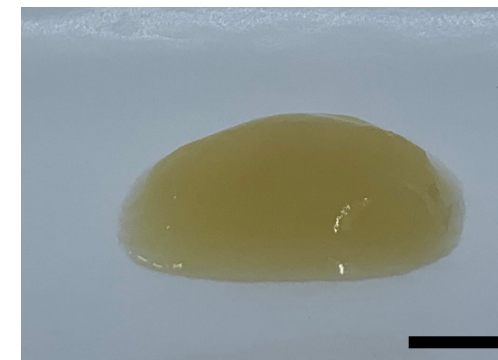
Creatill CNF



Flax CNF



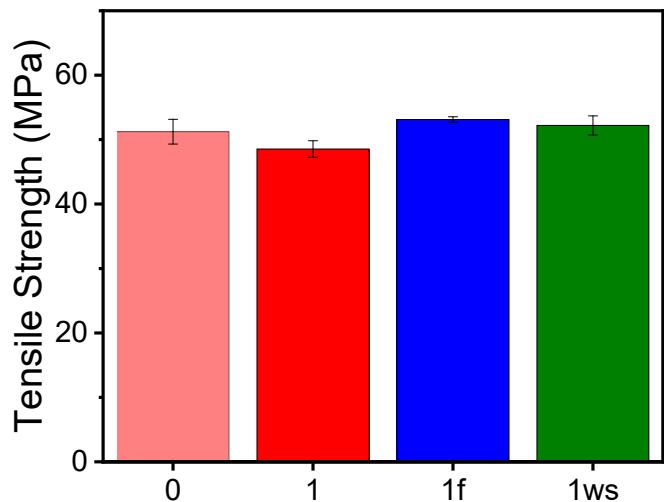
Wheat Straw CNF



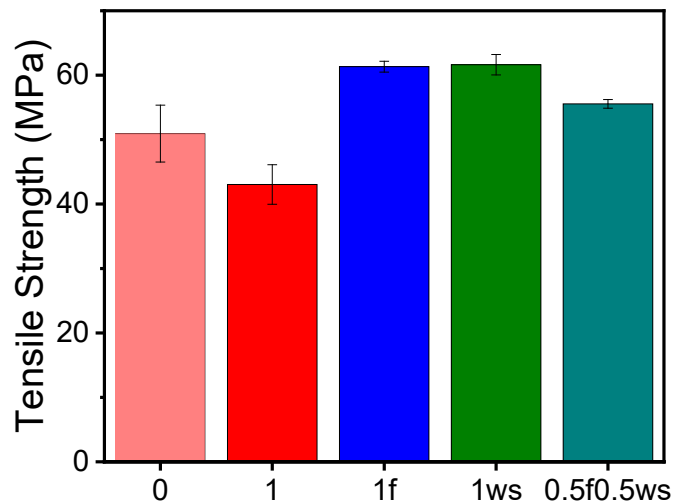
Scale Bar: 1 cm

Varying Hybrid Filler Concentration

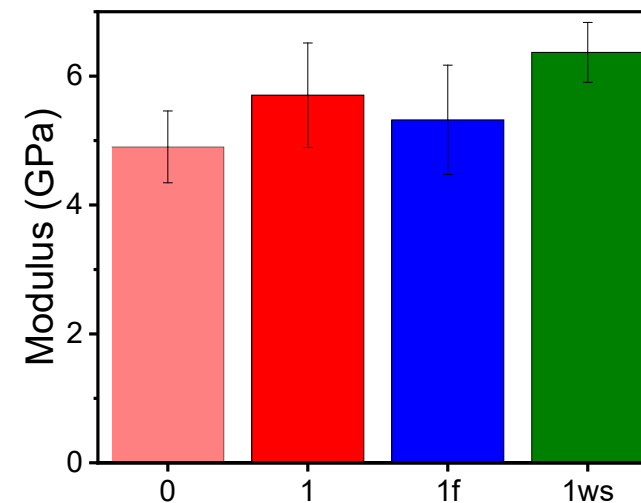
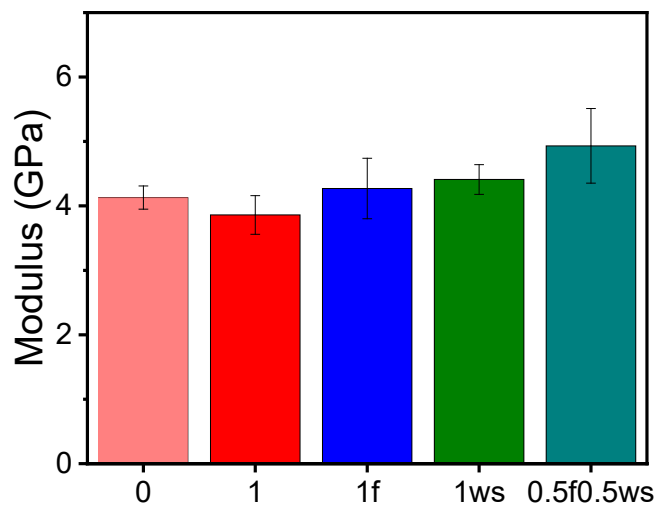
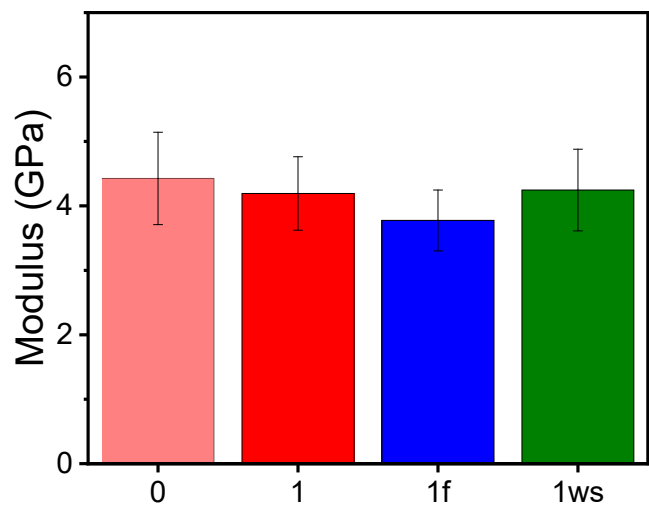
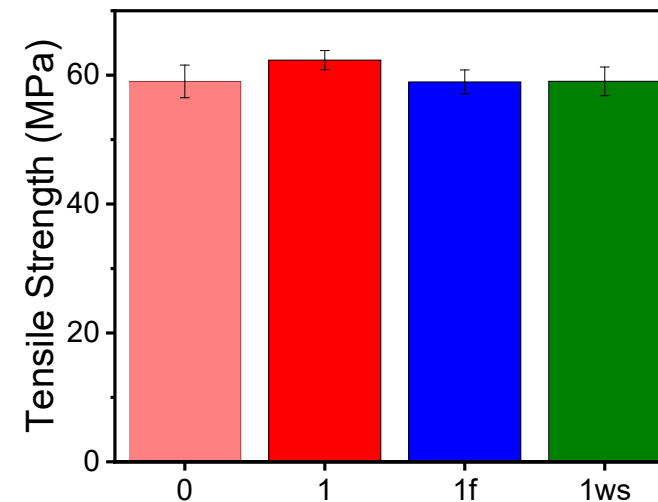
10% Filler Content

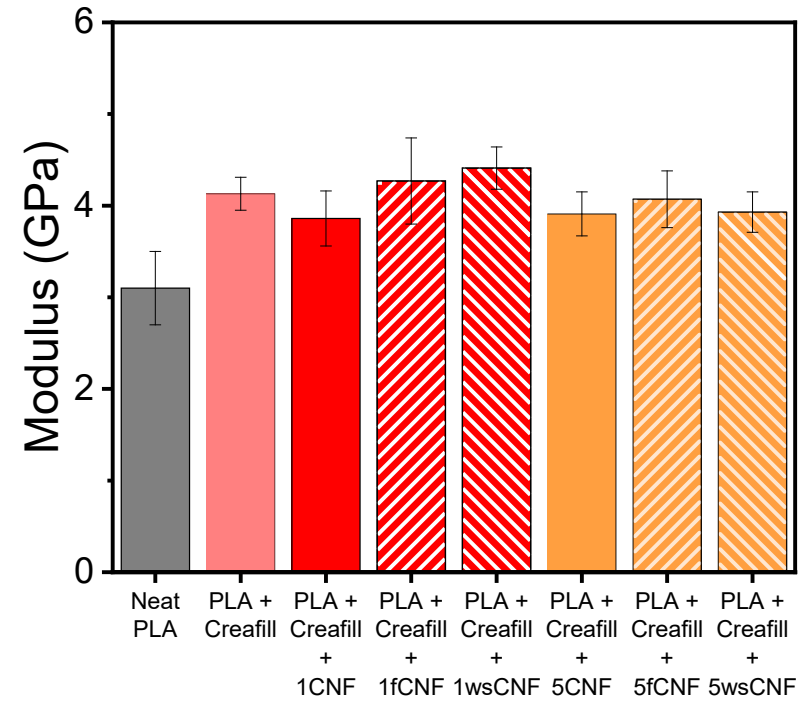
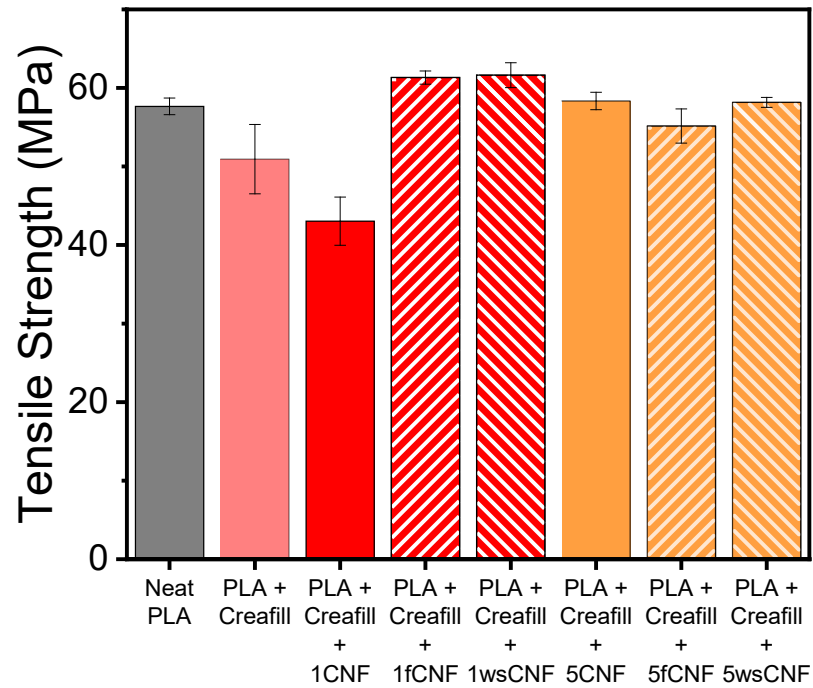


20% Filler Content

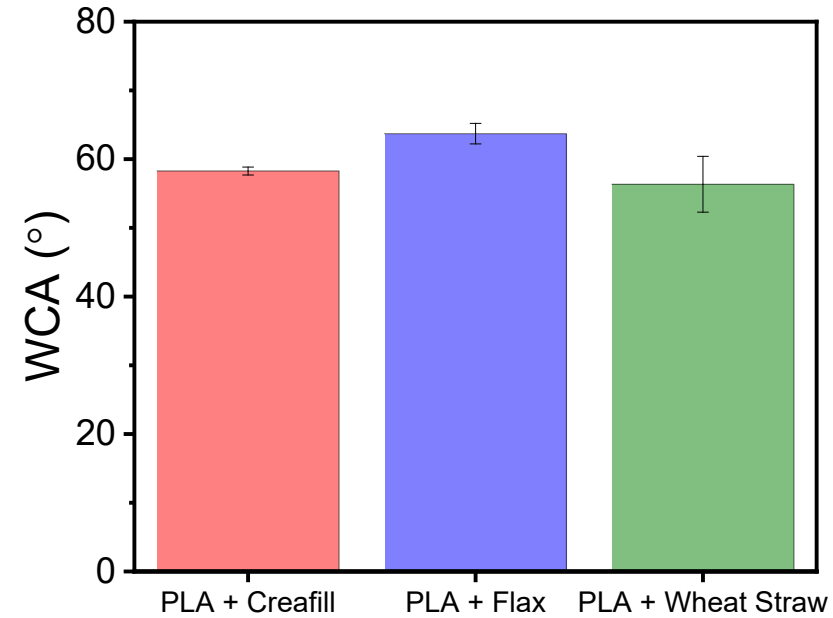


30% Filler Content

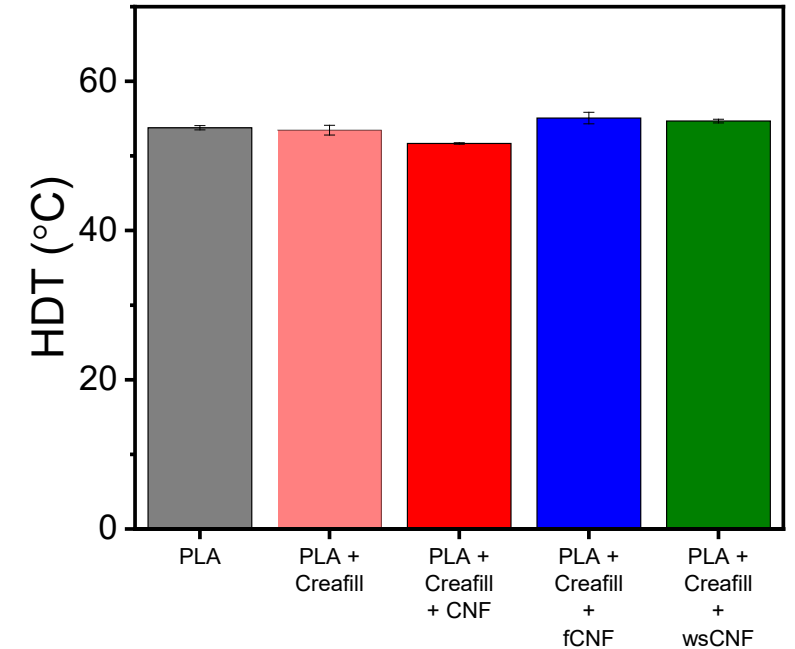
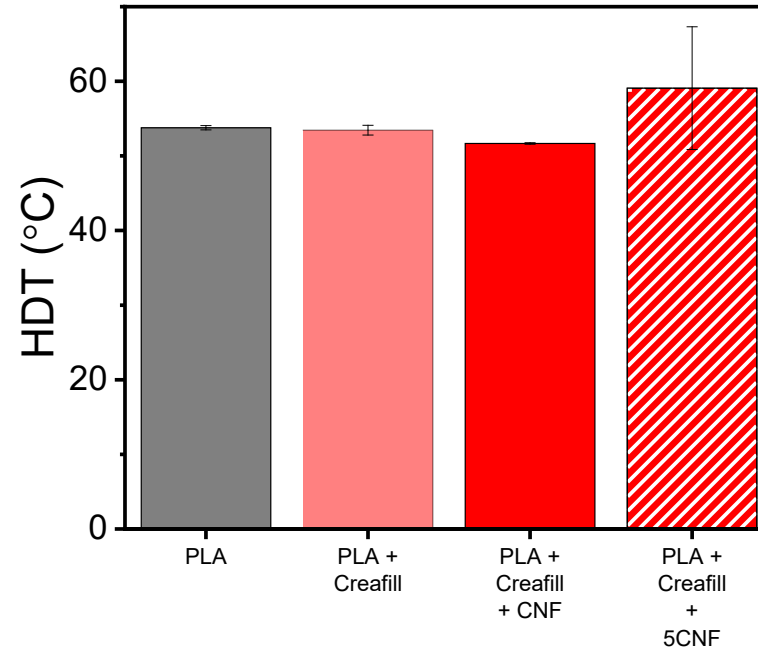
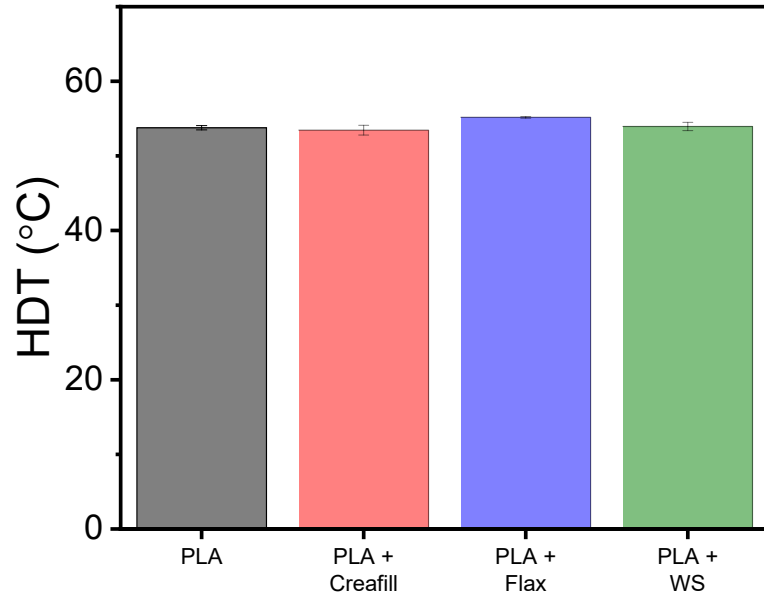




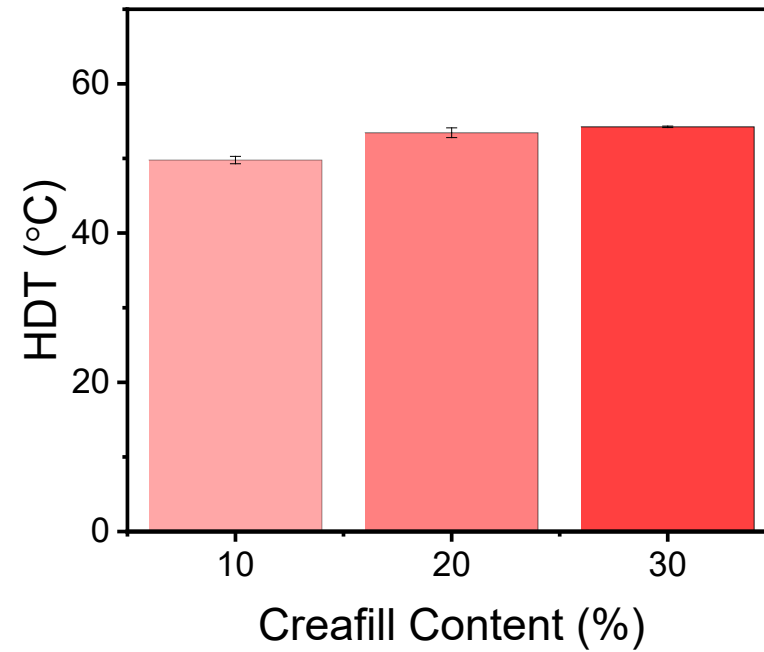
Takeaway: There is an optimum amount of CNF addition for fiber hybridization to maximize both strength and modulus.



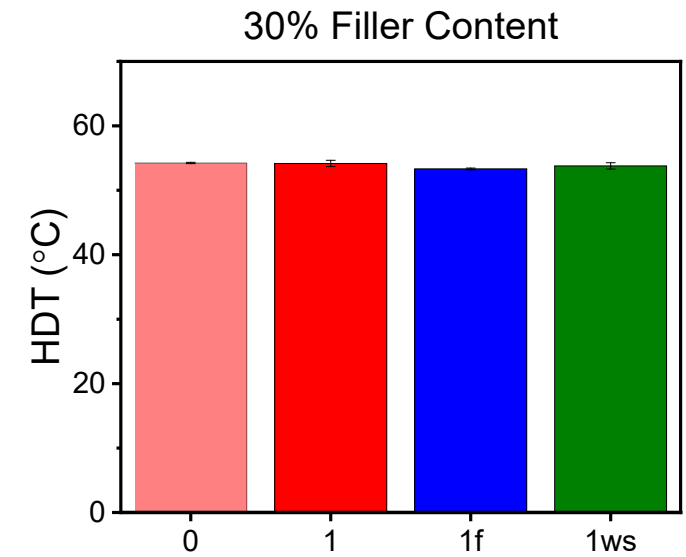
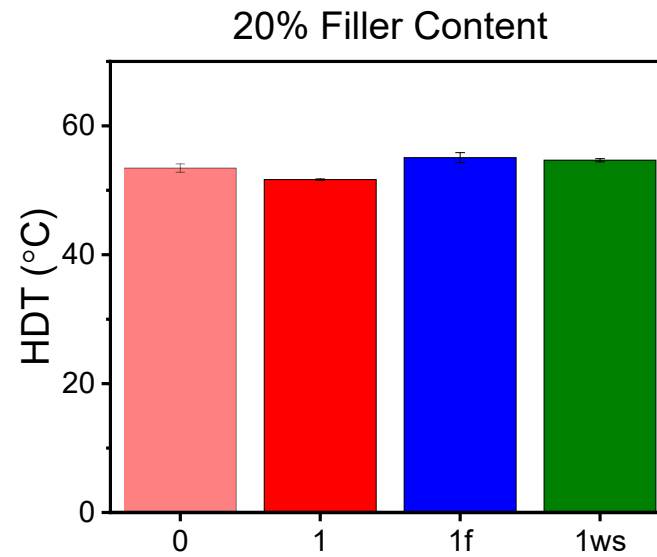
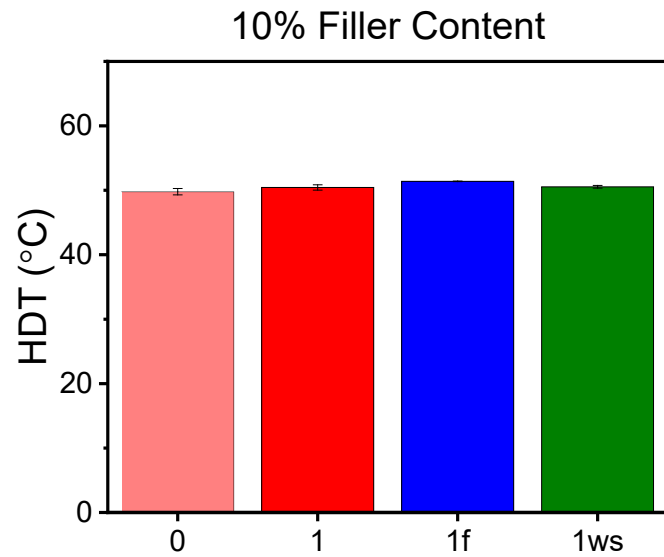
Takeaway: No appreciable difference in hydrophilicity of the composites.



Takeaway: There is minimal variability in heat deflection temperature (HDT) regardless of filler type.



Takeaway: The heat deflection temperature (HDT) is optimized at 20wt% filler concentration.



Takeaway: The heat deflection temperature (HDT) is optimized at 20wt% filler concentration; minimal changes seen throughout.

Biomass Composition

Filler Type	Glucan (%)	Xylan (%)	Lignin (%)
Creafill	79.33 ± 1.65	18.4 ± 0.67	-
Flax	73.46 ± 0.71	6.97 ± 1.73	15.34 ± 0.26
Wheat Straw	49.92 ± 0.92	21.8 ± 0.47	24.82 ± 2.17

Hypothesis: Both topology and surface chemistry impact the adhesion between the filler and matrix.

