Development and Validation of a Direct Ink Writing Platform for Lignin-Based Materials

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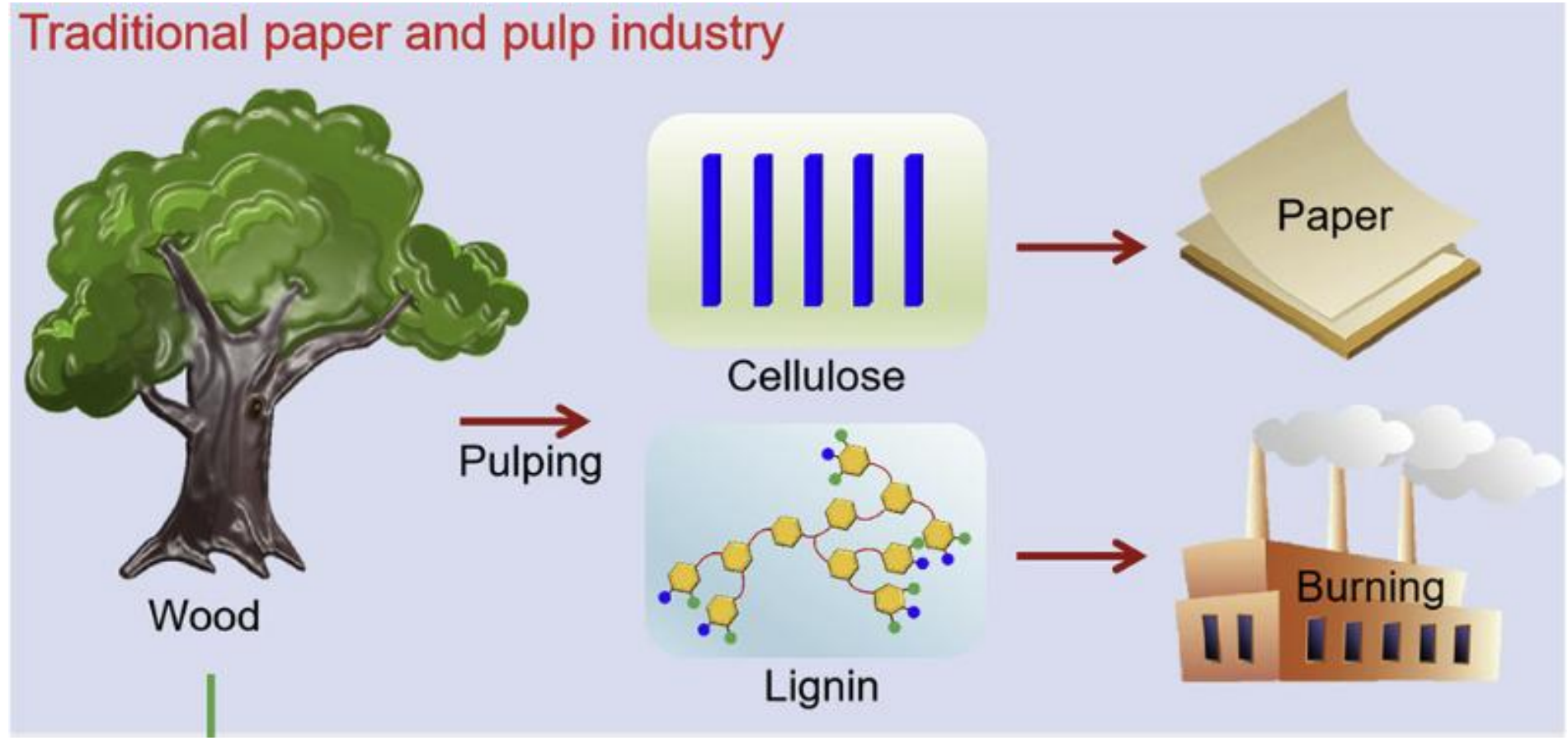
1 Background & Motivation

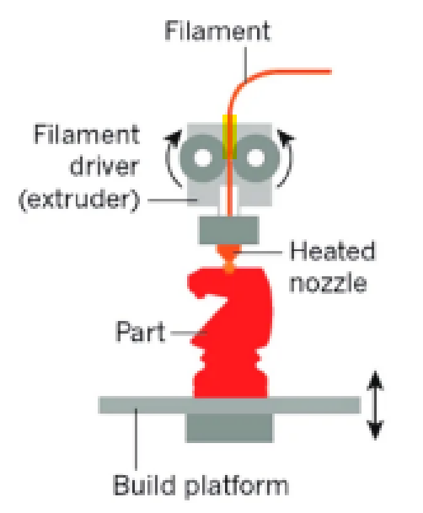
- Lignin is the second most abundant renewable polymer on Earth after cellulose, yet much of it is still burned as a low-value fuel.
- Direct ink writing (DIW) can process viscous pastes and gels under mild conditions.
- This makes DIW a promising route for sustainable, high-lignin-content feedstock.

Traditional paper and pulp industry



DIW vs Conventional Printing

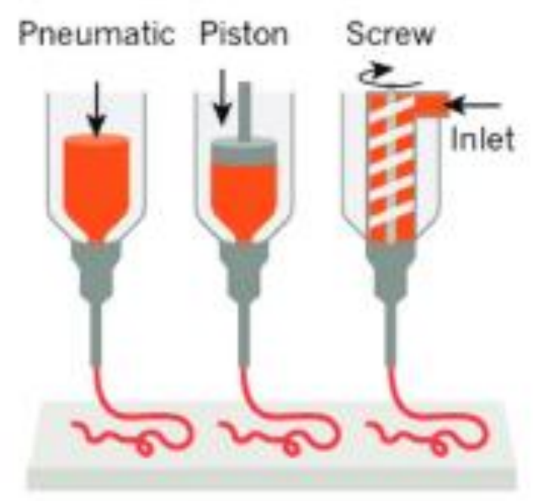
Conventional



- Lower-viscosity materials
- Melting or UV curing
- Limited to thermoplastics or resins

VS

DIW (This Work)

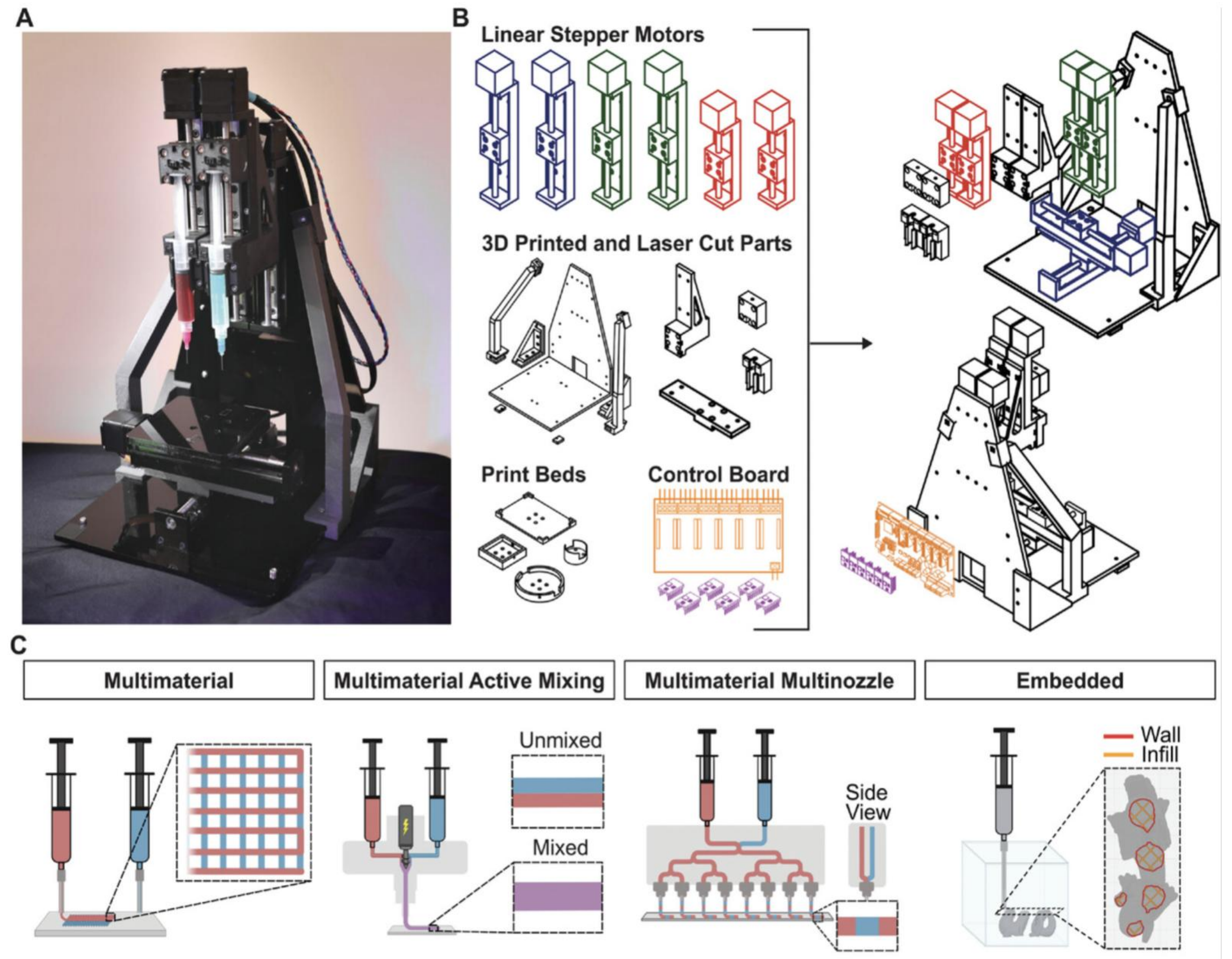


- High-viscosity pastes and gels
- Mild temperatures (room temperature)
- Compatible with lignin-rich feedstocks

3 Objectives

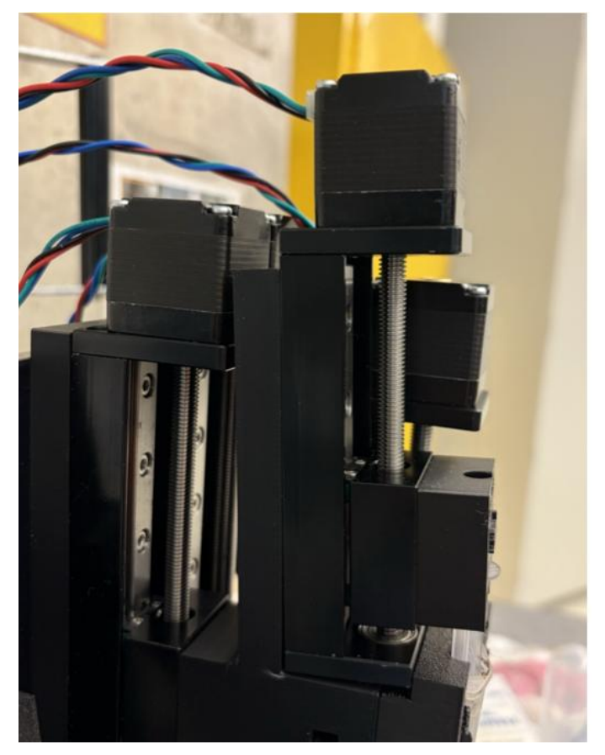
- Assemble and validate a low-cost DIW printer based on the Printess architecture.
- Establish a repeatable CAD-to-print workflow for syringe-based extrusion.
- Screen lignin-based DIW ink systems and select a practical starting formulation.
- Develop and refine a workable lignin-rich ink through screening of pH, lignin preparation, sonication, and air-drying versus freeze-drying routes.
- Demonstrate preliminary printing, post-processing, and basic performance testing of printed structures.

2 Platform Development

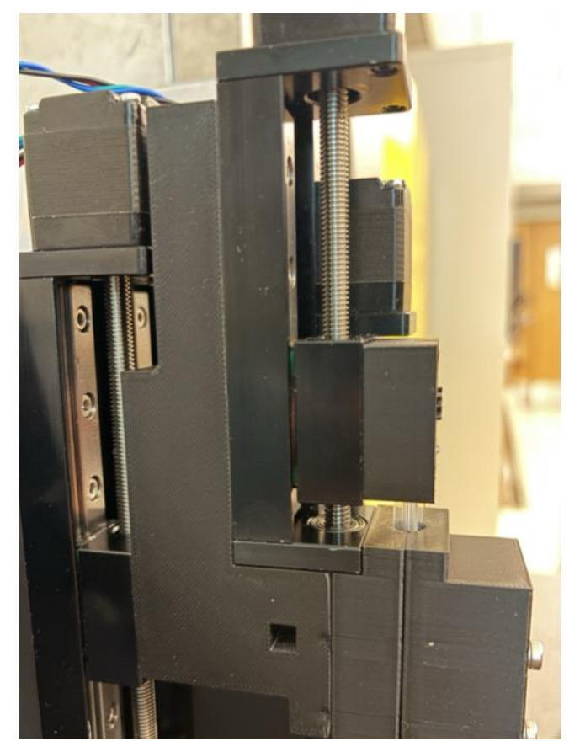


Hardware Upgrades for High-Viscosity Extrusion

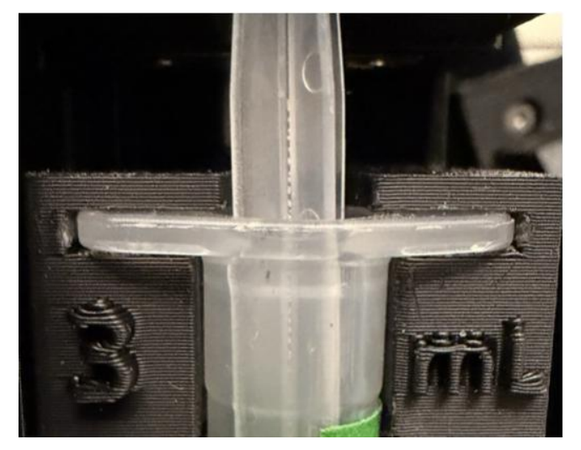
- Custom supports reduced deflection and improved force transfer during extrusion.
- Added syringe-body support improved alignment and extrusion consistency.



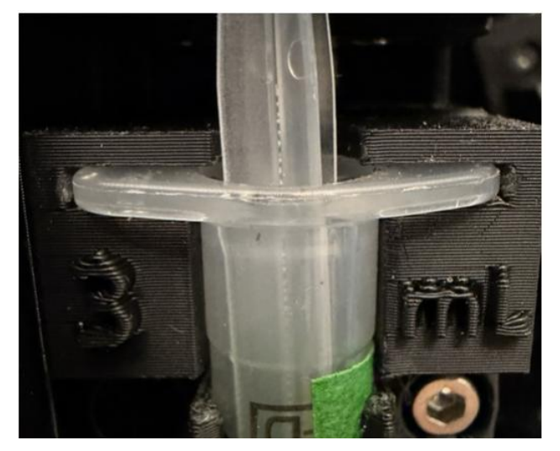
Original support under load



Upgraded support under load



Original syringe support under no-load



Original syringe support under load

Platform Key Points

- Low-cost, open-source Printess-based DIW platform
- Controlled using Marlin firmware and Pronterface
- Repeatable workflow: CAD → STL → Cura → G-code → Python conversion → Print
- Validated using toothpaste as a paste-like surrogate material

Total Cost: CAD $584.64

3 Materials & Ink Development

- Selected route: lignin–Pluronic F127
- Why selected: high lignin loading, simple room-temperature preparation, and demonstrated printability.
- Two alkali lignin routes were compared: pH 10.5 and pH 6.5
- Final practical formulation: 1:1.2 lignin:(F127 + water), prepared using pH 5 water–F127.

pH 10.5 lignin route (not selected)

Water-soluble, but putty-like and showed water separation under pressure.

pH 6.5 lignin route (selected)

More paste-like and workable after preparation.

Lignin Preparation Workflow

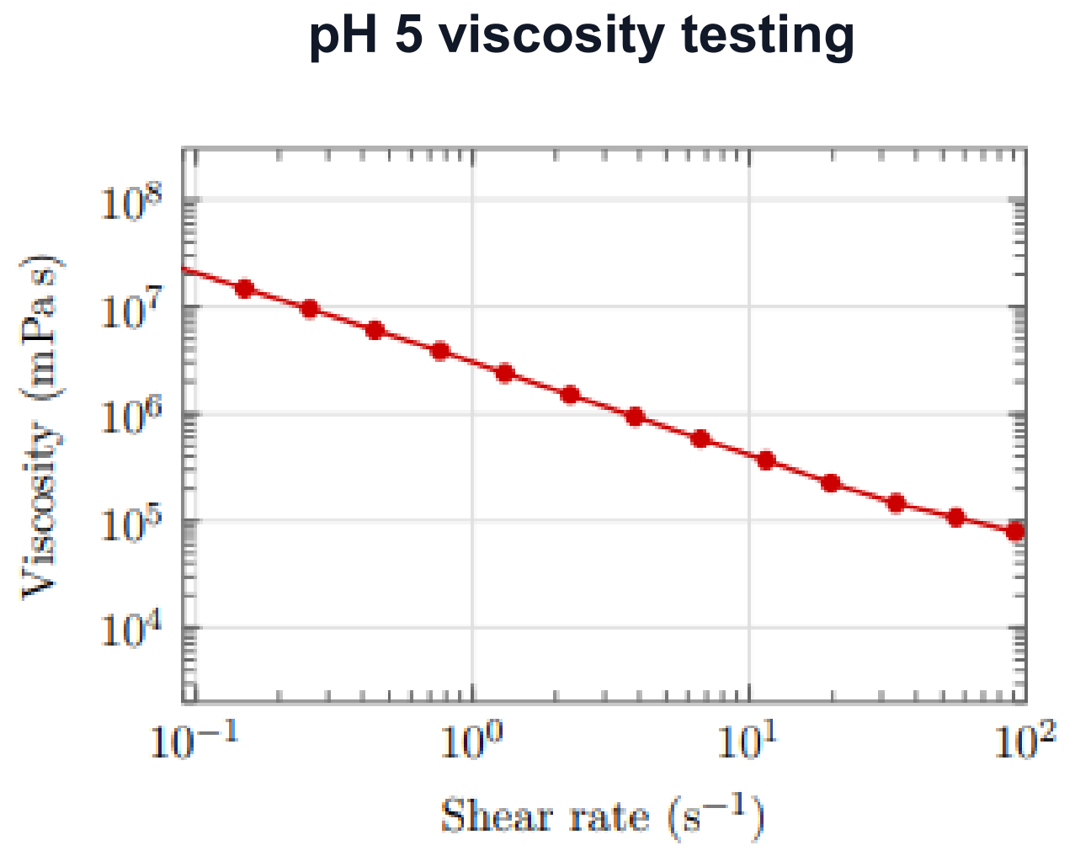
1. Tip sonication — reduce particle size and break up lignin agglomerates
2. Centrifugation — separate larger particles and remove supernatant
3. Drying method — compare air drying and freeze-drying of prepared lignin
4. DLS evaluation — use DLS to compare apparent particle size after each drying route

DLS Results

- Freeze-dried: Z-average particle size = 1027 nm; PDI = 0.7408
- Air-dried: Z-average particle size = 2217 nm; PDI = 0.4487
- Freeze drying gave smaller apparent particles, but print performance was similar.

Effect of Water–F127 pH when Mixed with Lignin

- pH 5: lower viscosity and better flow; enabled higher lignin loading and the best balance of extrusion and shape retention.
- pH 7: intermediate behavior and a neutral baseline.
- pH 9: higher viscosity, crumblier, and reduced printability.



4 Printing & Post-Processing

Print Process

Print → Freeze-dry → Thermal stabilize at 100 °C for 2 h

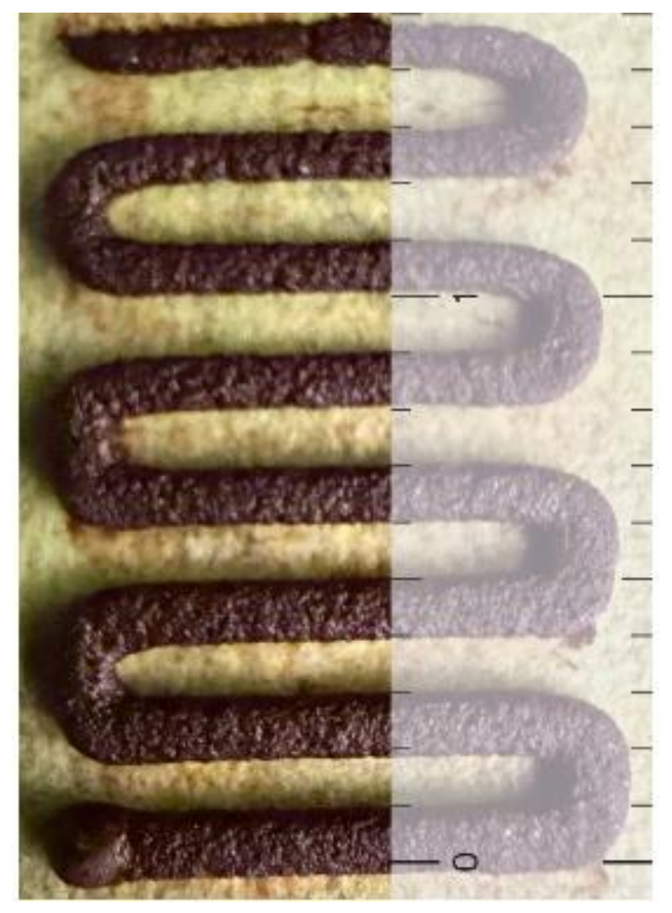
Printing Trials & Stabilization

- Printed lignin–F127 structures through a 0.965 mm nozzle to evaluate deposition quality.
- Line-spacing trials compared filament overlap, spreading, and definition.
- Print quality depended on bed leveling, nozzle height, and formulation consistency.
- Freeze-drying improved handling, but thermal stabilization at 100 °C for 2 h was required for water and oil stability.

Line-Spacing Trials

0.25 mm spacing

0.50 mm spacing



1.00 mm spacing

Printed Rings & Post-Processing

After printing (green state)

Stable after freeze-drying and thermal stabilization

Compression Results

- Ring specimens had an outer diameter of approximately 10 mm and a wall thickness of about 2.0 mm.
- Freeze-dried and thermally stabilized samples withstood about 15 lb (~66.7 N) before failure.
- The corresponding approximate compressive stress was about 1.3 MPa.
- This was a rough weight-based estimate rather than a standardized mechanical test.

References

- Sarge et al., Small, 2020
- Du et al., ACS Sustainable Chemistry & Engineering, 2020
- Decker et al., Green Materials, 2020
- Verma et al., Advanced Materials, 2023

Conclusions

- A low-cost, open-source DIW platform was successfully built and validated.
- Lignin preparation, formulation pH, and post-processing strongly influenced printability and material behavior.
- This work provides a promising foundation for sustainable, high-lignin-content printed structures.

Limitations

- Bed leveling and frame stiffness affected print repeatability.
- Ink viscosity changed quickly after mixing and air exposure.
- Mechanical testing was preliminary.
- Samples were brittle in bending.

Future Work

- Improve frame stiffness, bed leveling, and motor cooling.
- Test conical nozzle tips to improve flow.
- Perform controlled rheology and mechanical testing.
- Optimize formulation for toughness and stability.