Fused Deposition Modeling Additive Manufacturing of Carbonized Structures via Waste-Enhanced Filaments

DE-FE0032143

Yahya Al-Majali, Ph.D.

Ohio University

U.S. Department of Energy

National Energy Technology Laboratory
Resource Sustainability Project Review Meeting
April 2-4, 2024

Disclaimer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Project Overview

Project Specifics

- DOE/NETL Cooperative Agreement No. DE-FE-0032143
- DOE Project Manager: Mark Render
- Principal Investigator (PI): Yahya Al-Majali
- Participants: CONSOL Innovations, JuggerBot 3D, IC3D, CONSOL Energy, and Clear Skies Consulting

Project Budget

- Federal: $1,000,000
- Non-Federal: $250,000

Project Duration

- April 15, 2022 – April 14, 2025

DOE-NETL Carbon Ore Processing Program

Project Objectives

Overall: Develop carbonizable coal-enhanced polymer filaments for use in commercially available 3D printers to manufacture structures for building/construction and tooling applications.

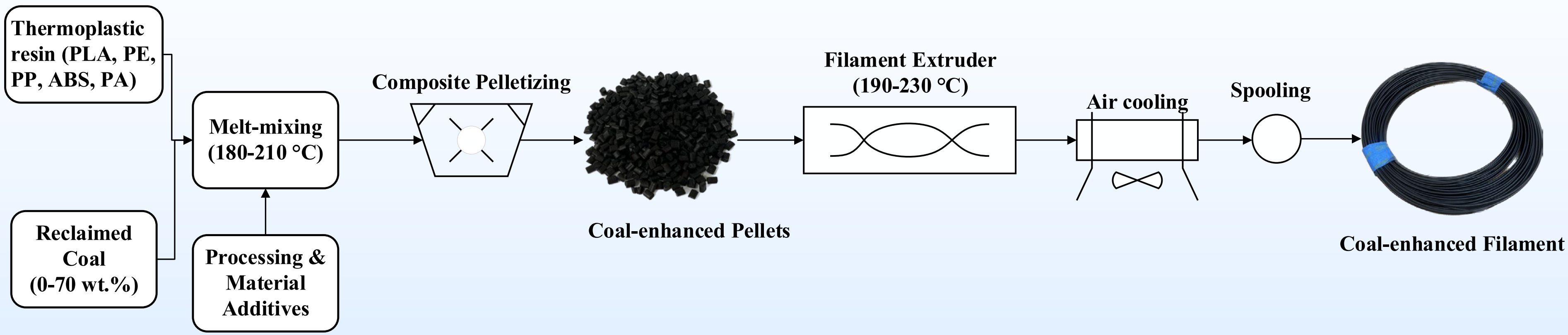
Budget Period 1 objectives

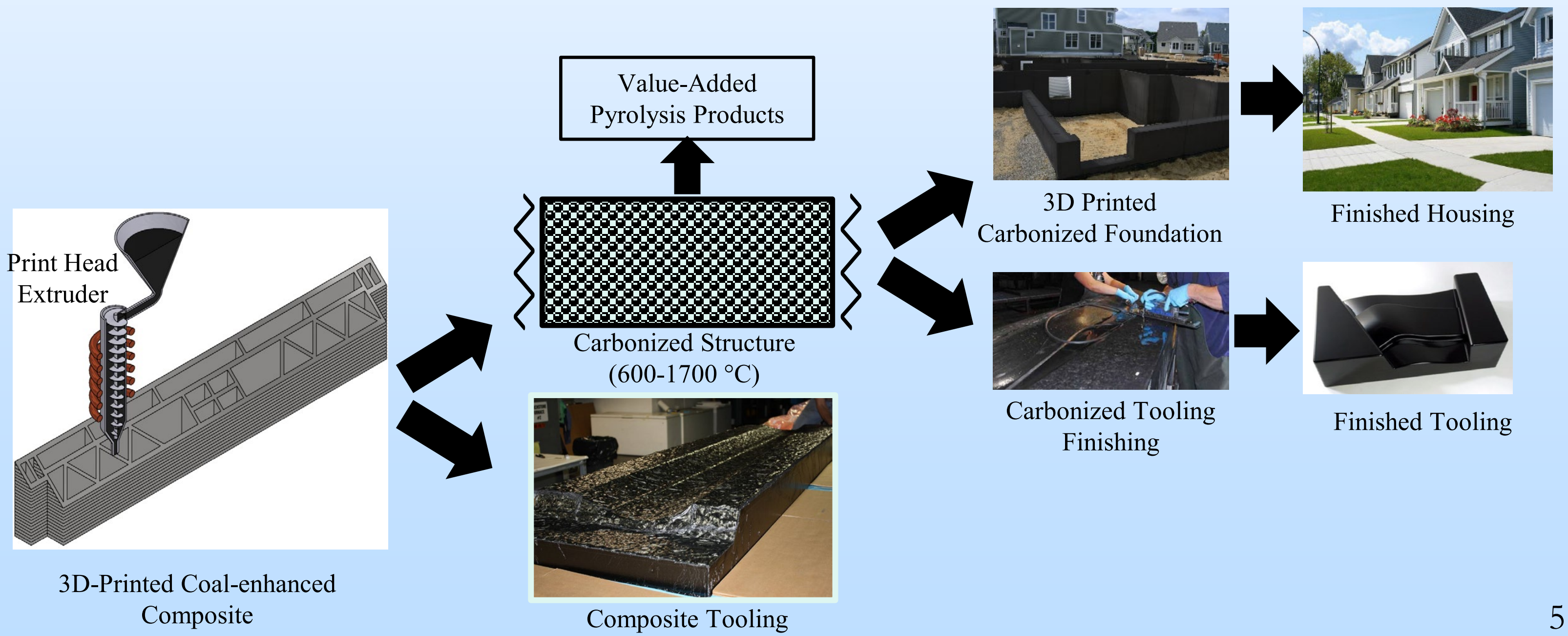
– Develop coal-enhanced filament (CEF) formulations that maximize the coal content
– Develop and refine CEF extrusion parameters
– Assessing CEF mechanical and physical properties
– Assess microwave processing to improve welding between printed CEFs

Budget Period 2 objectives

– Develop CEF printing parameters for commercial 3D printers
– Print tooling articles with commercial 3D printers
– Assess the performance of non-carbonized and carbonized 3D-printed structures
– Develop detailed techno-economic analyses
– Identify additional commercial applications for the CEF materials
– Develop Environmental, Safety, and Health (ES&H) analysis

Technology Background





Technology Background

Technical, Economic, and Environmental Advantages

- Higher thermal stability compared to unfilled plastics
- Reduced warpage/shrinkage
- Equivalent/superior performance compared to some commercial AM composite materials
- Integrate with several types of commercially available 3D printers
- Require less energy to manufacture and generate less emissions
- Preliminary techno-economics
  - Commercial filament costs: $10-30 /kg
  - CPC filament production costs: $4.28-4.50 /kg

Technical Challenges

- Processing parameters scale nonlinearly
- Warpage/delamination during printing of PE-based formulations

Technical Approach/Project Scope

Milestones

Task	Description	Planned Completion Date	Actual Completion Date	Verification Method
1.0	Updated PMP submitted	May 20, 2022	May 19, 2022	Submission of updated PMP to NETL FPM
1.1	Project Kick-Off meeting held	May 23, 2022	May 23, 2022	Presentation file
1.2	Technology Maturation Plan (TMP)	July 14, 2022	July 15, 2022	Report file
1.5	Environmental, Safety, and Health (ES&H) Analysis	July 14, 2022	July 15, 2022	Report file
2.1	Coal-Enhanced Filament Formulation Report	January 14, 2023	January 14, 2023	Quarterly Report
2.2	Coal-Enhanced Filament Properties Report	April 14, 2023	April 14, 2023	Quarterly Report
4.0	Coal-Enhanced Filament Printability Analyses	October14, 2024		Quarterly Report
5.0	Techno-economic and Market Analyses	January 14, 2025		Quarterly Report

Technical Approach/Project Scope

Project Success Criteria

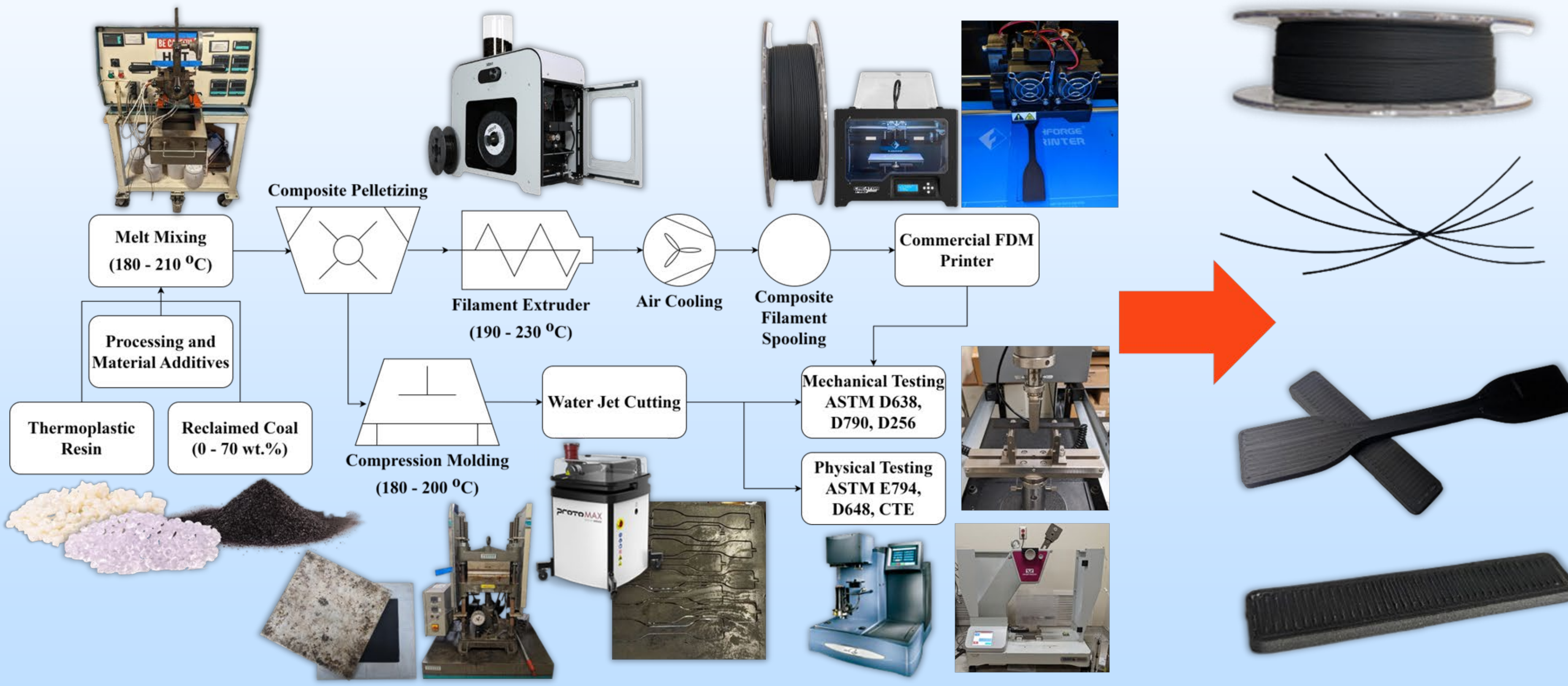
- Develop a coal-enhanced filament containing >50 wt.% coal content which can be utilized in a commercial FDM printer
- Integrate carbonizable coal-enhanced filament with lab- and large-scale commercially available FDM printers and develop associated 3D printing parameters
- Demonstrate utilization of carbonizable coal-enhanced filament in printing of representable building/construction and tooling parts and quantification of application-specific properties

Technical Approach/Project Scope

Risks and Mitigation Strategies

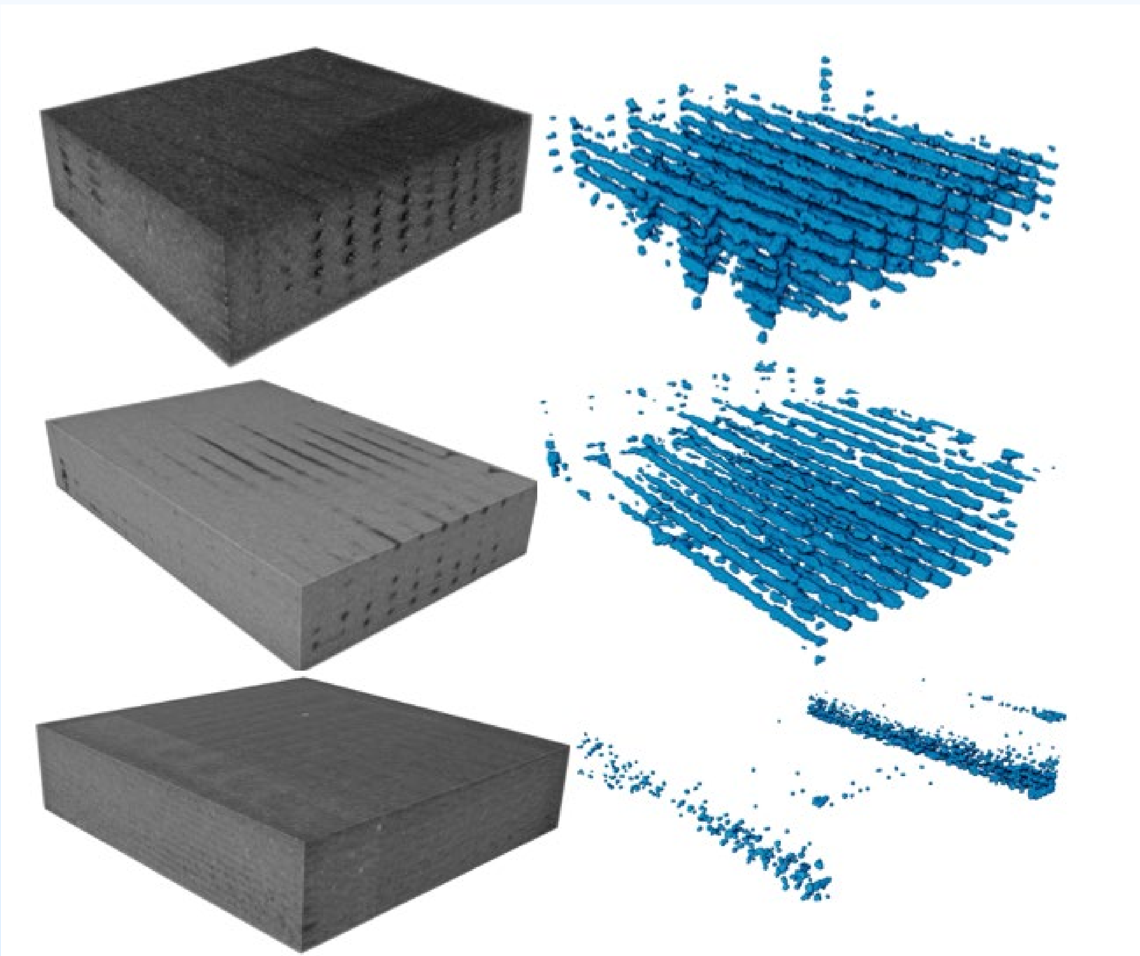
Perceived Risk	Risk Rating (Low, Med, High) Probability	Impact	Overall	Mitigation/Response Strategy
Feedstock Availability	Low	Medium	Low	The project has commitments from industry to supply coal-derived resources to support filament formulation development. The project team plans to test a host of coal, reclaimed coals, and thermoplastics to develop coal-enhanced filaments for a wide range of commercial applications.
Printer Integration	Low	Medium	Medium	The ease of utilizing materials with commercial FDM printers is an important factor for commercial adoption of coal-enhanced filaments. A range of filament diameters and formulations will be evaluated with commercial FDM printers throughout the project to demonstrate integration of coal-enhanced filaments with commercial FDM printer technology.
Equipment Wear	Medium	Medium	Low	Processing of coal in extrusion equipment could increase wear of extruder parts. Bench-scale compounding, extrusion, and spooling trials will allow wearing on filament production equipment to be assessed and determine materials best suited for the process.
Market Acceptance	Medium	High	High	Failure for the coal-enhanced filaments to be accepted by the market would prove commercialization difficult. OHIO is working with project commercial partners to assess carbonizable coal-enhanced filaments for industry applications to print and quantify properties of 3D printed articles.
Process Economics	Medium	Medium	Medium	To minimize coal-enhanced filament manufacturing costs, multi-variable techno-economic studies will be completed including but not limited to feedstock pricing, financing, product sales price, location, and capacity.

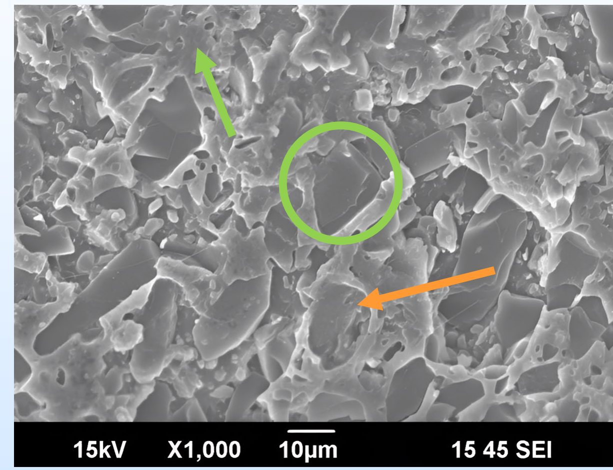
Progress and Current Status of Project

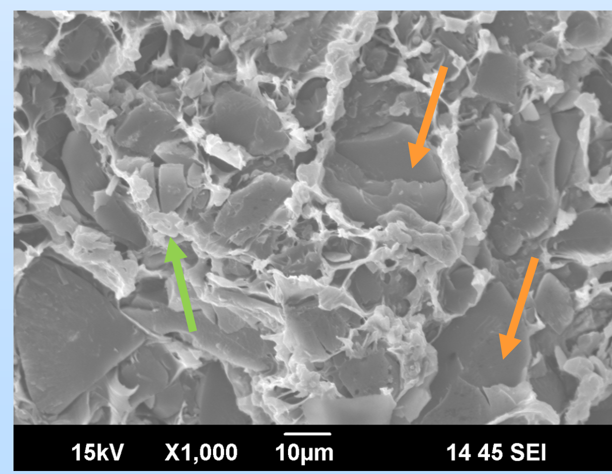


Progress and Current Status of Project

Composite Microstructure

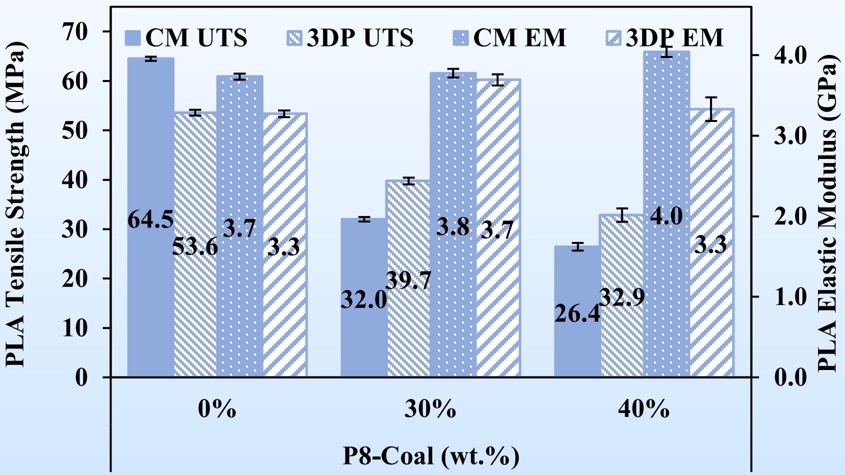


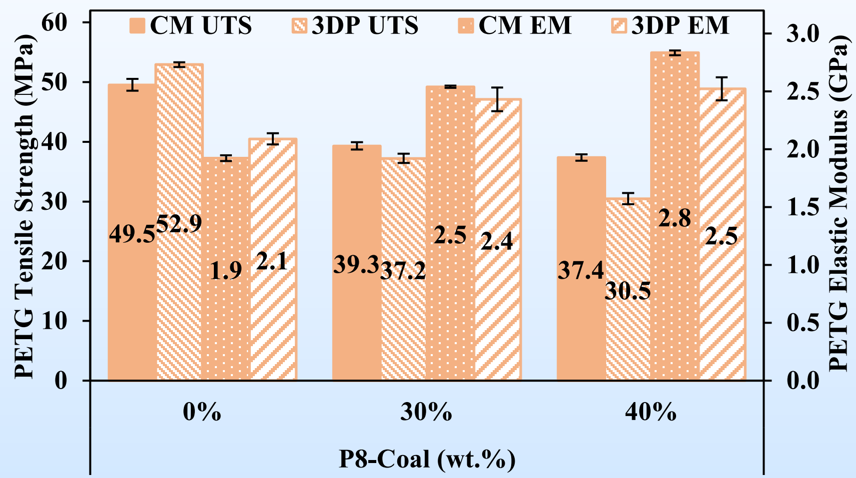


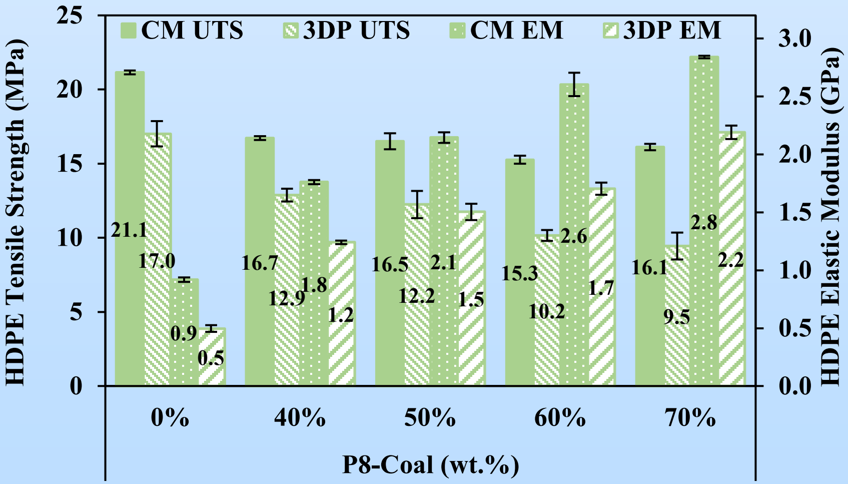


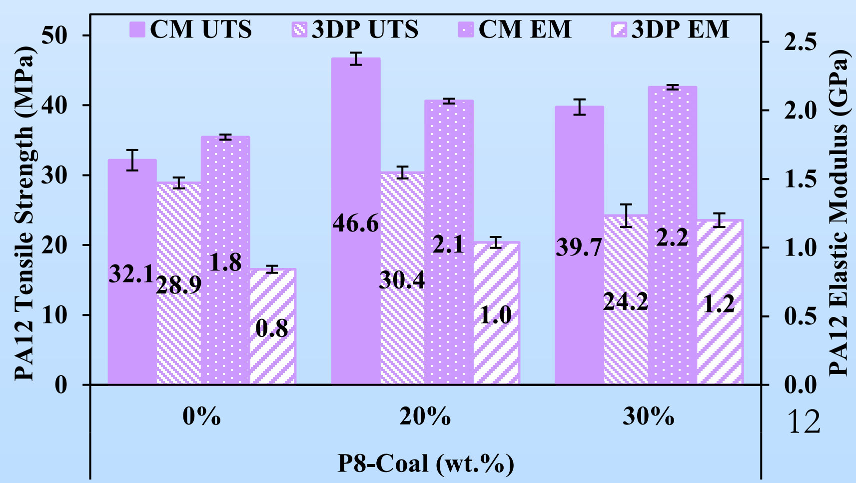
Progress and Current Status of Project

Tensile Properties



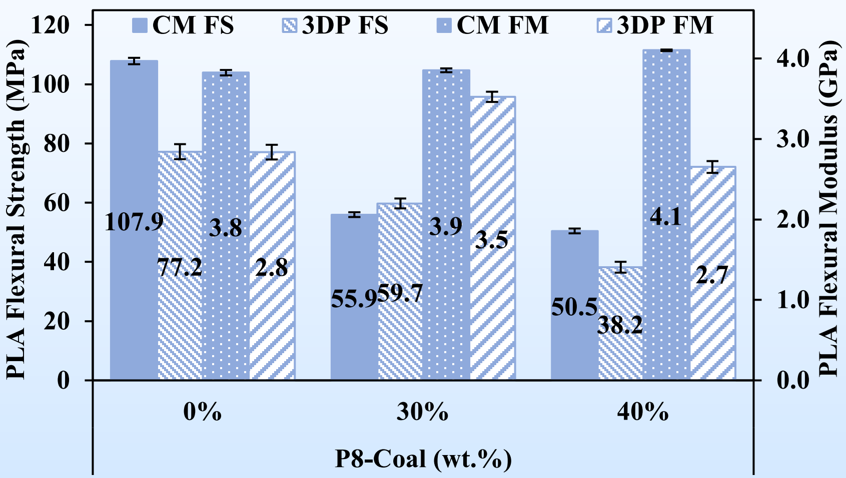


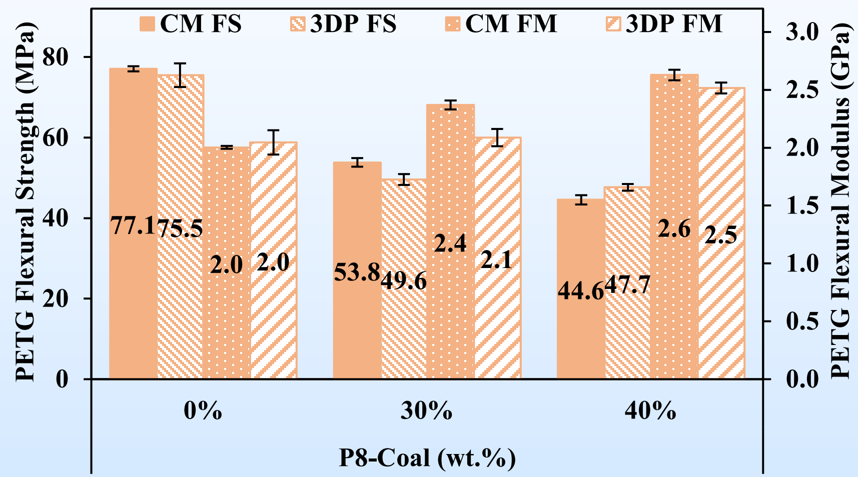


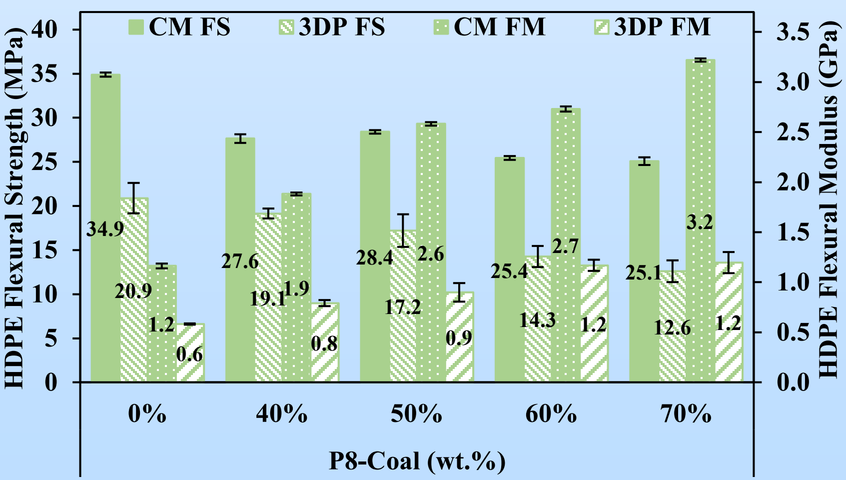


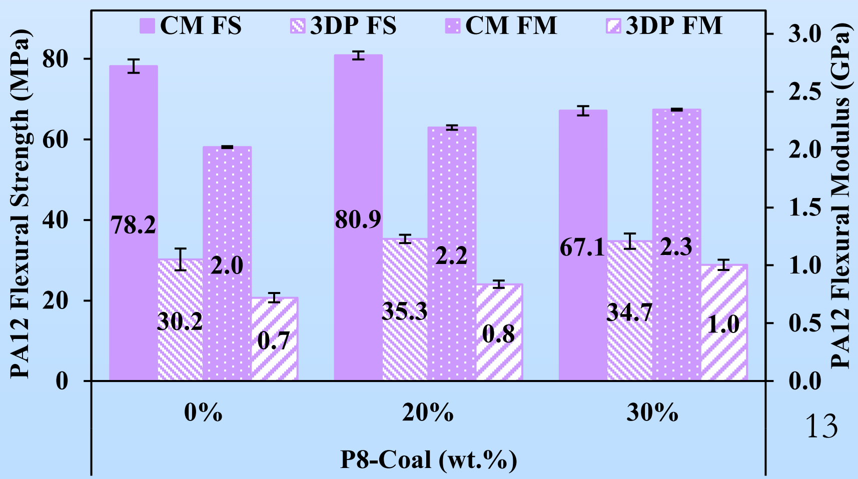
Progress and Current Status of Project

Flexural Properties



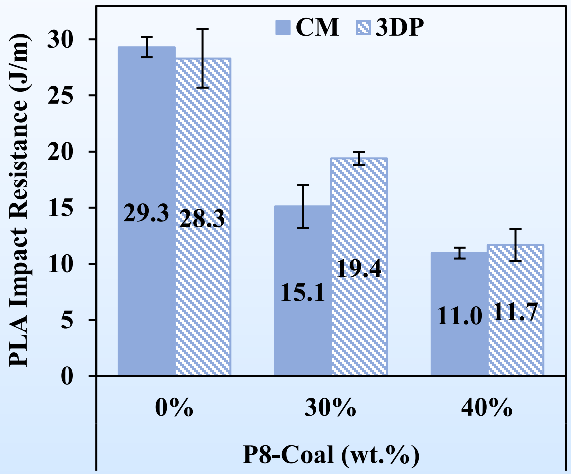


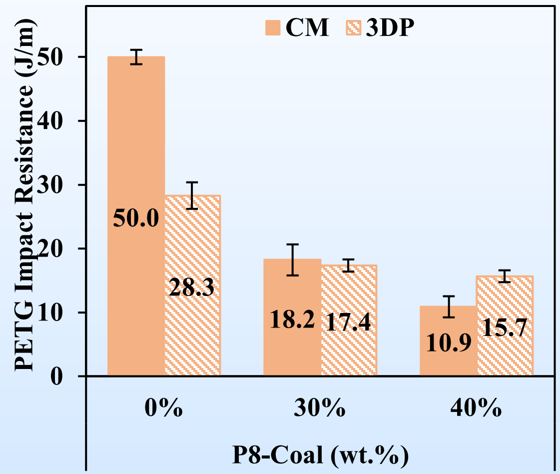


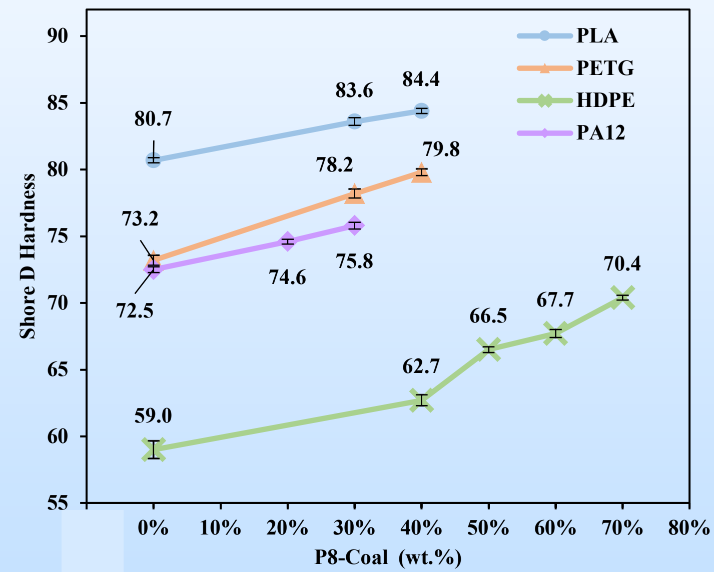


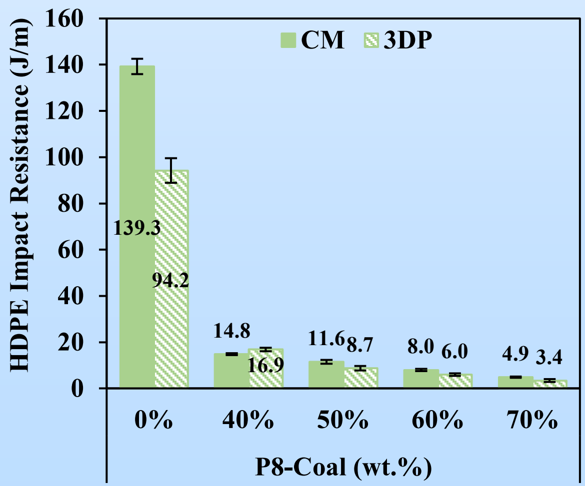
Progress and Current Status of Project

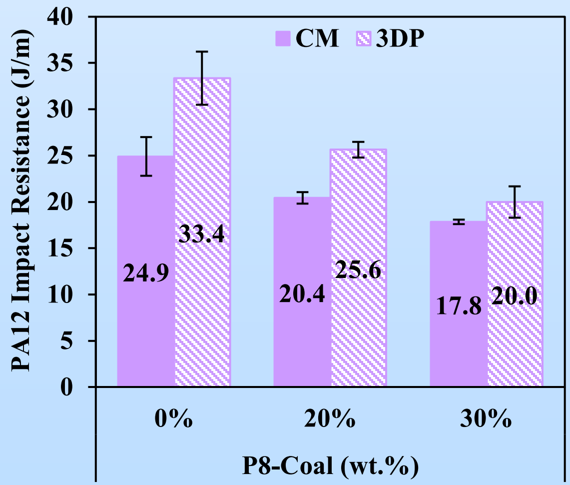
Impact Resistance and Hardness





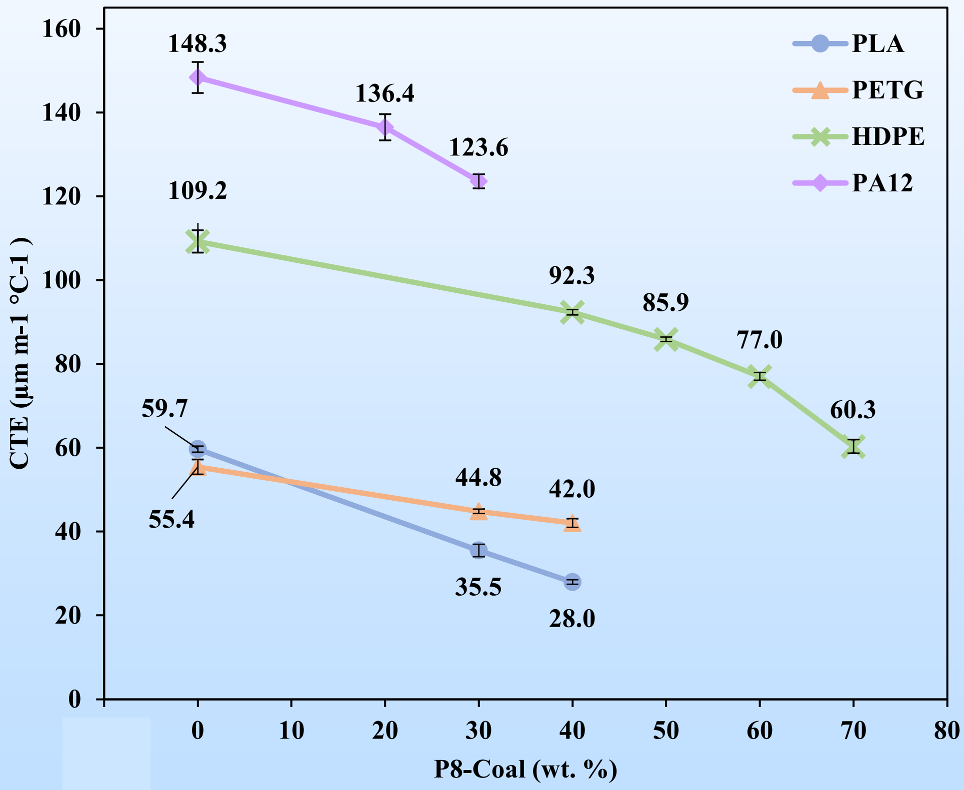


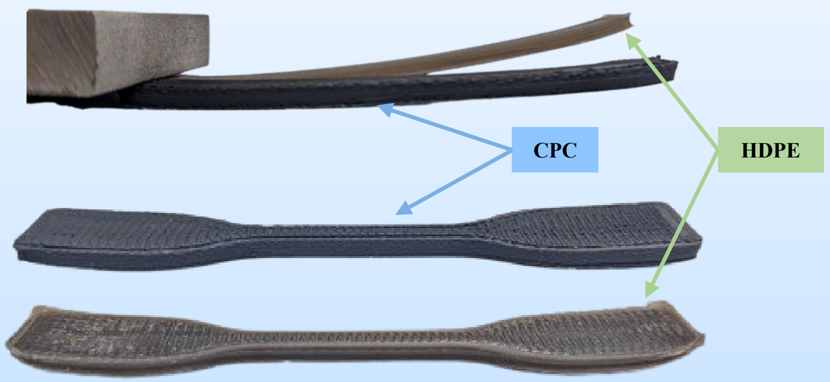




Progress and Current Status of Project

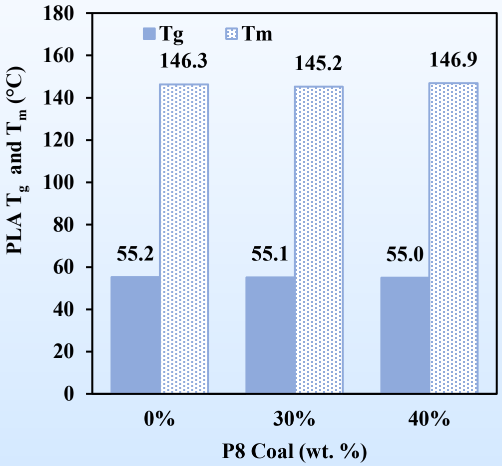
Coefficient of Thermal Expansion

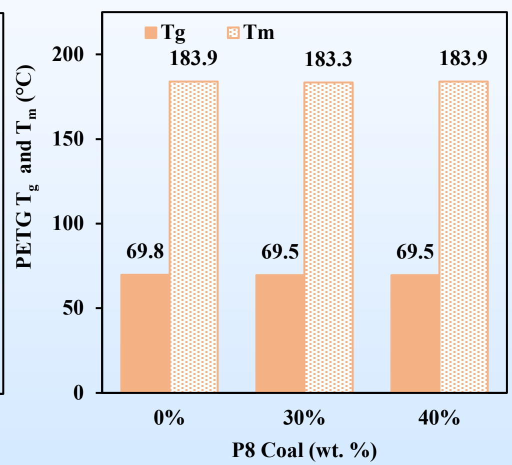


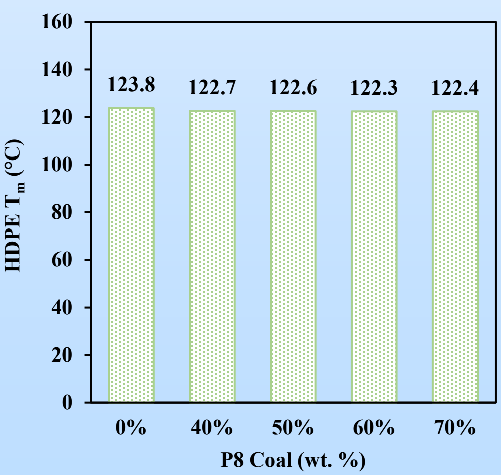


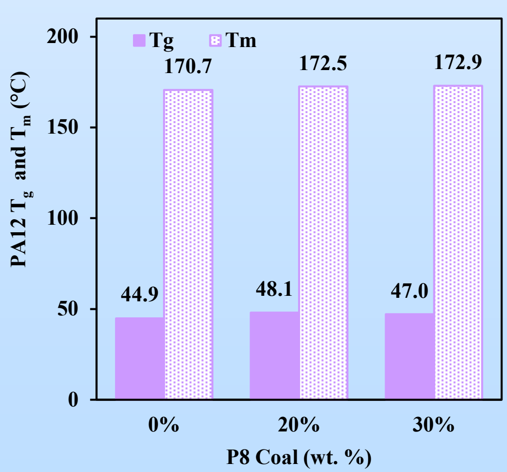
Progress and Current Status of Project

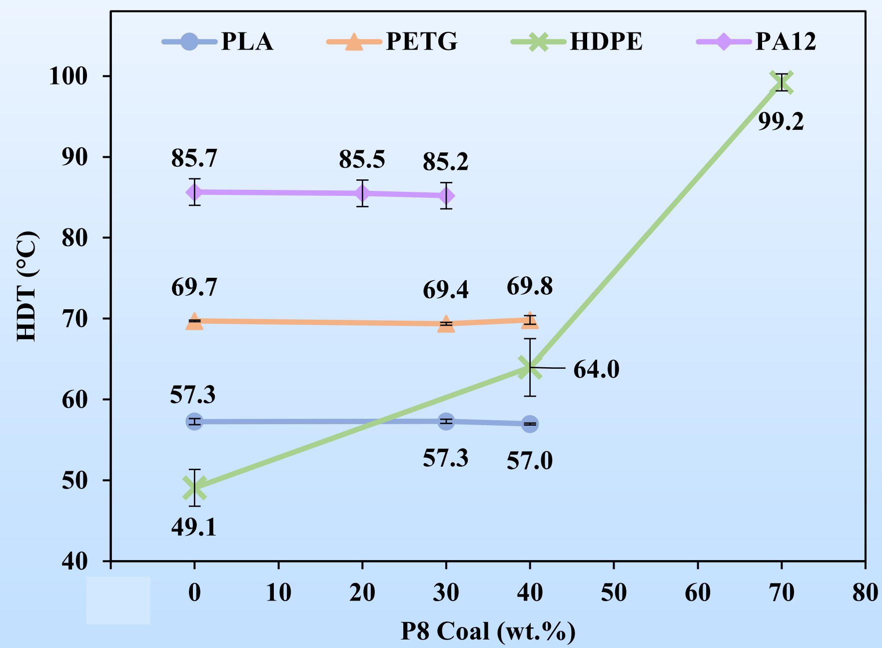
Melting Temperature, Glass Transition Temperature, and Heat Deflection Temperature





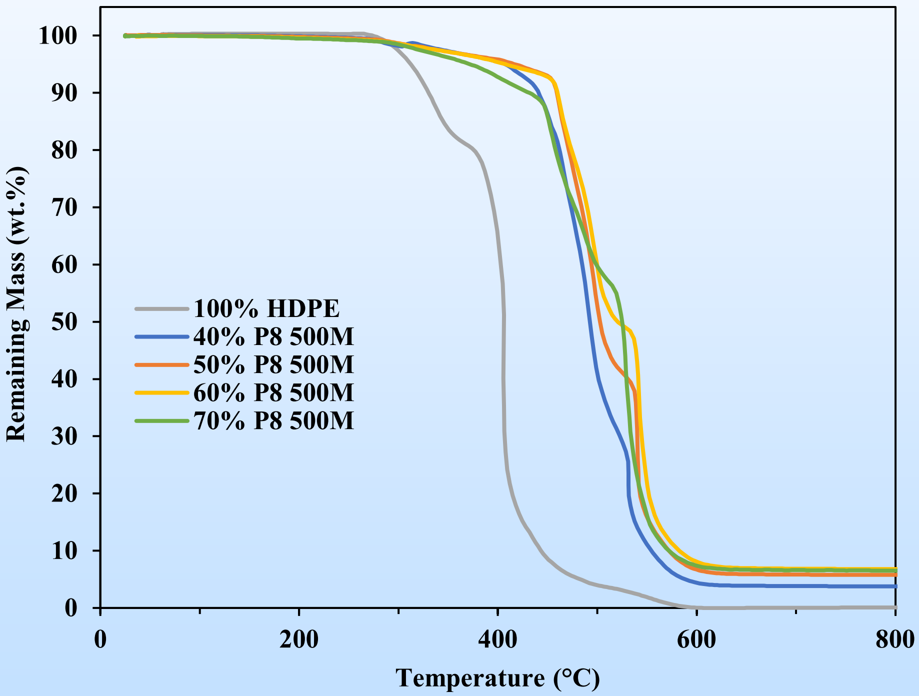


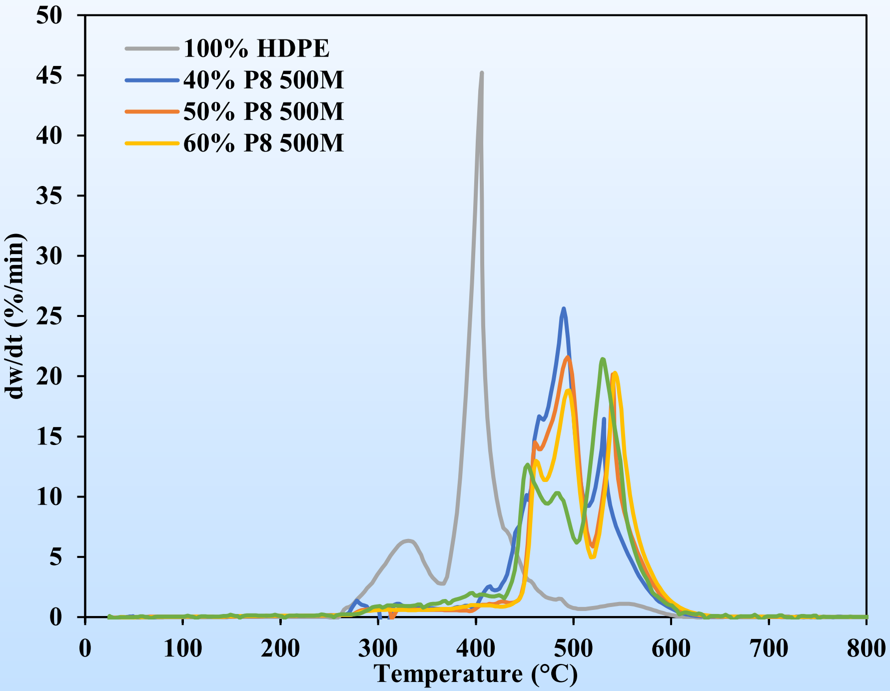




Progress and Current Status of Project

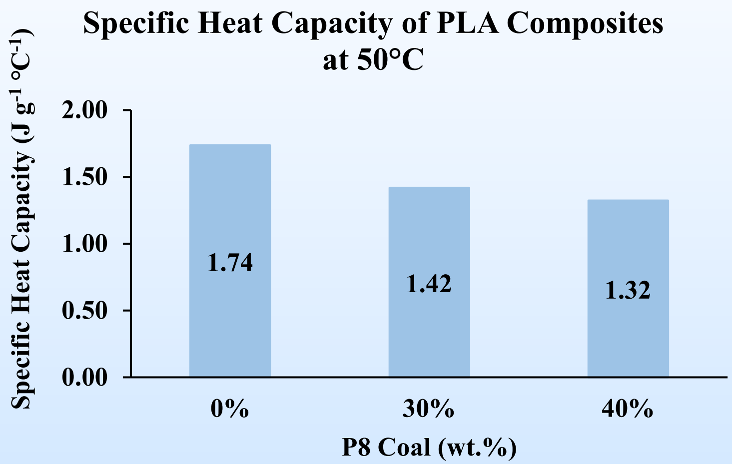
Thermal Stability

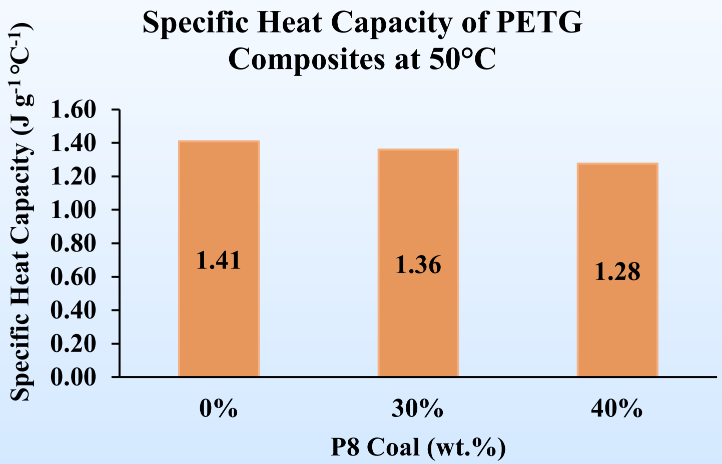


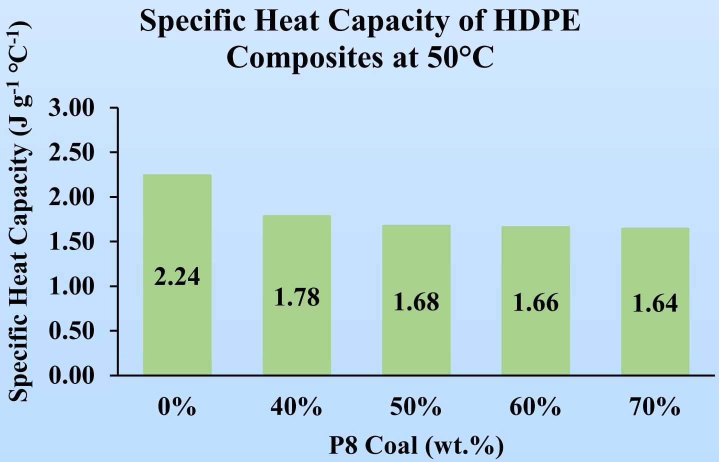


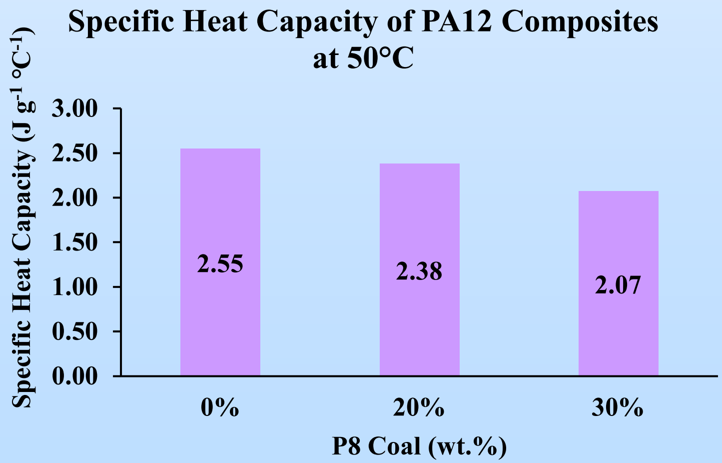
Progress and Current Status of Project

Specific Heat Capacity



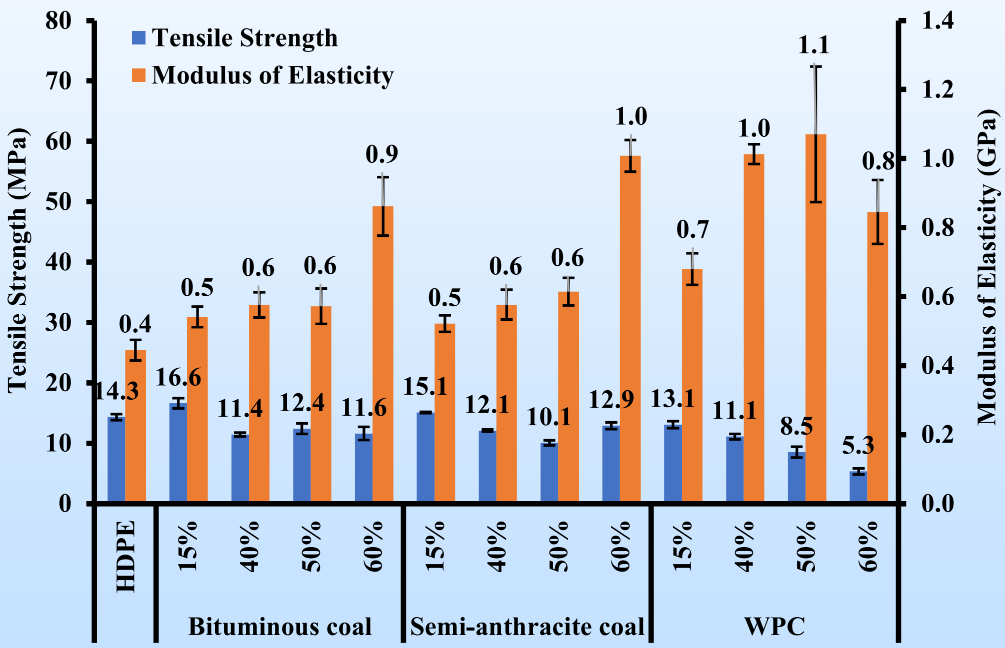


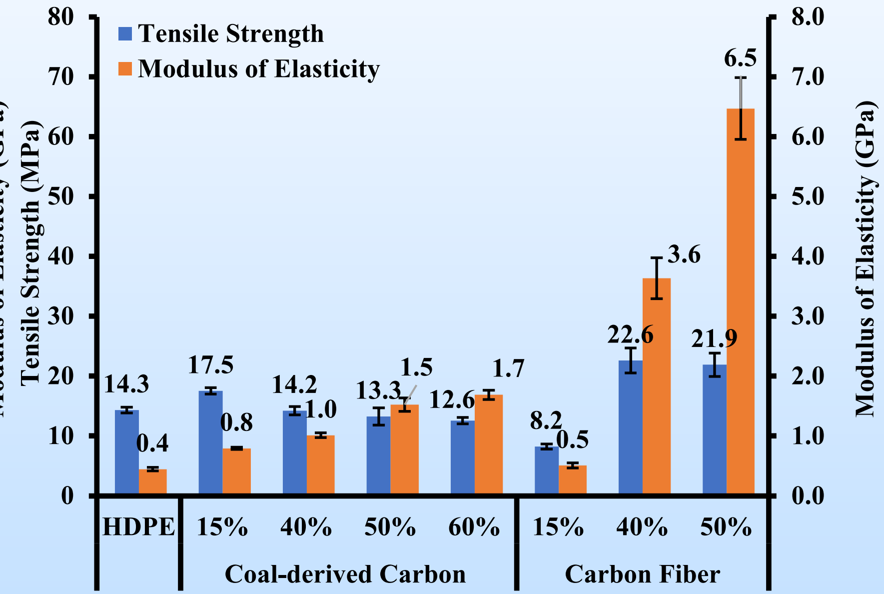




Progress and Current Status of Project

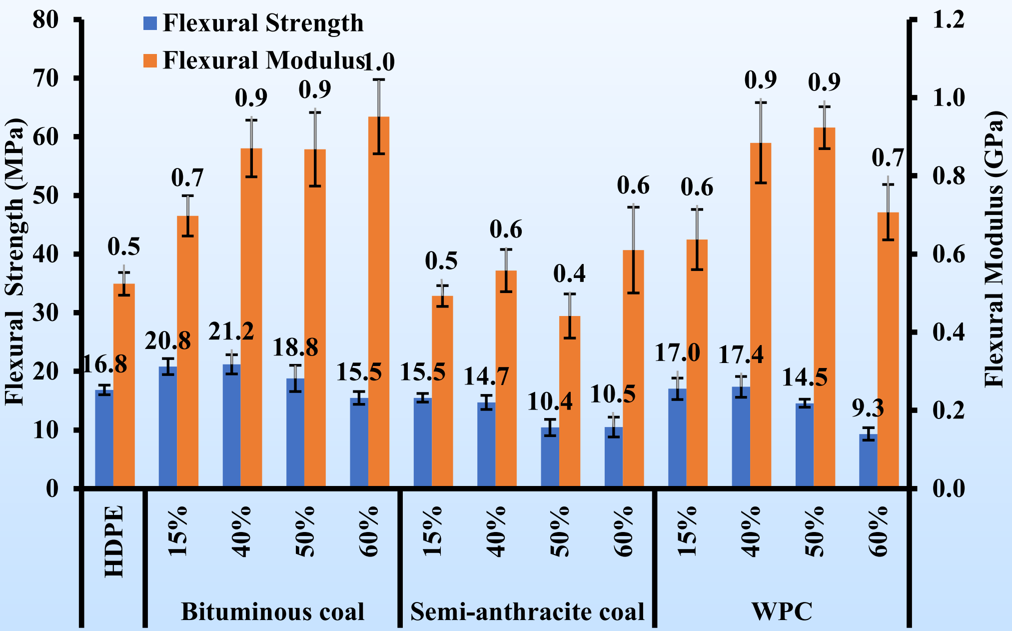
Performance Compared to Commercial Materials

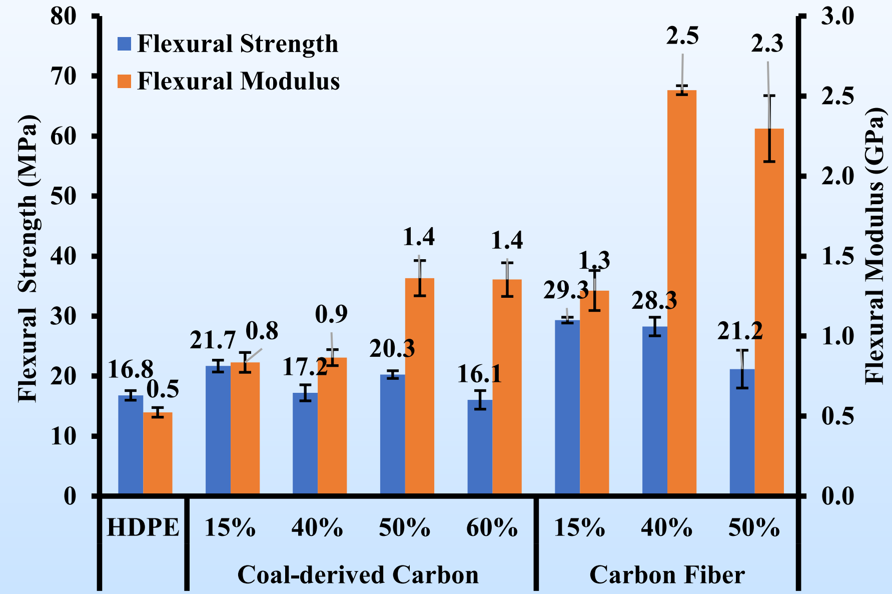




Progress and Current Status of Project

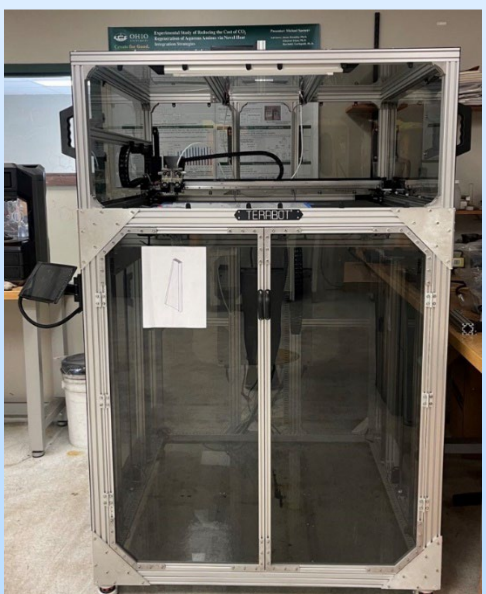
Performance Compared to Commercial Materials



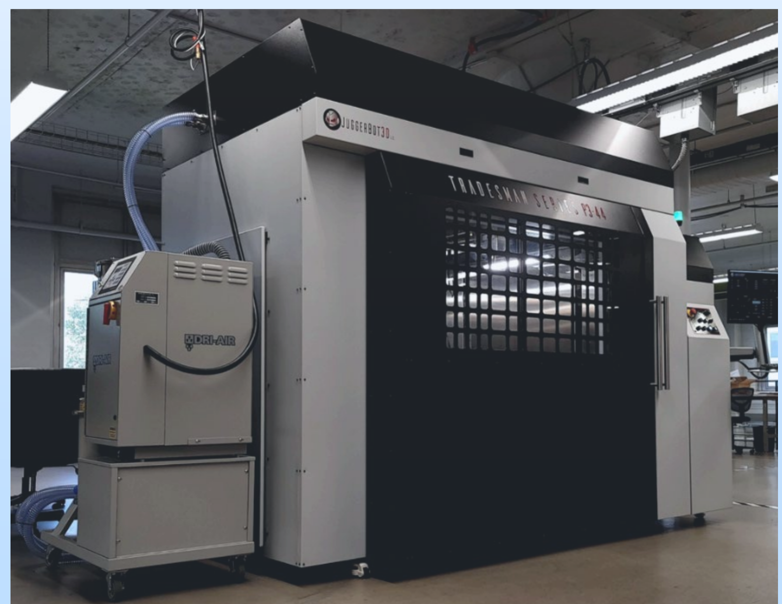


Progress and Current Status of Project

Scaled 3D Printing



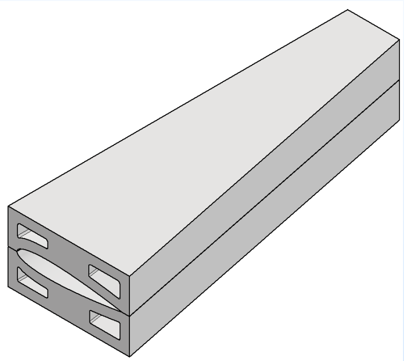
re3D Terabot 4 3D Printer

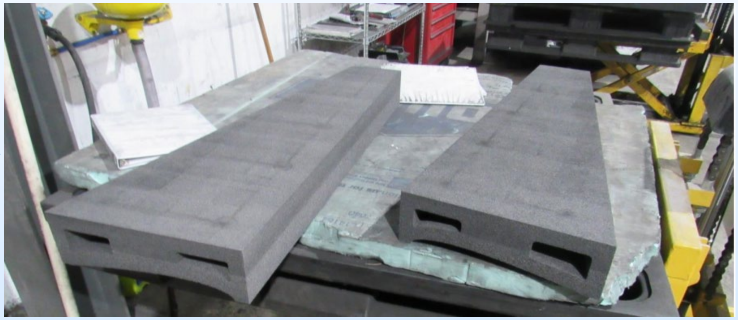


JuggerBot 3D Tradesman Series™ P3-44 additive system

Progress and Current Status of Project

Wind Turbine Blade Tooling-Traditional Method



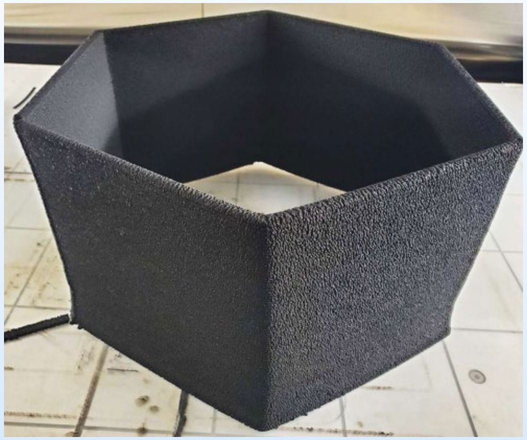


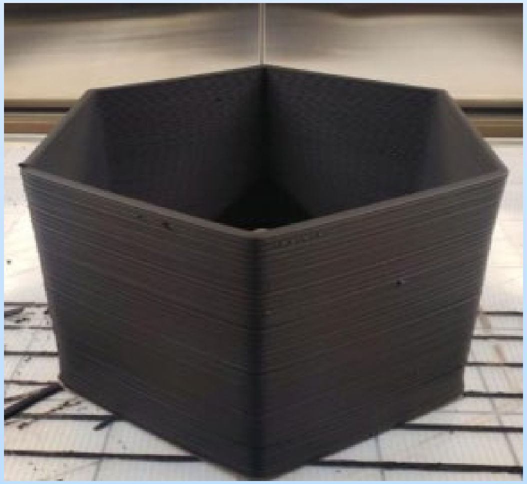
Material: Coal-derived carbon foam with polymer coating
Material Cost: $4,765
Labor Hours:106.6 hr
Labor cost:$12,635
Total Cost:$17,400

Progress and Current Status of Project

Material Testing & Assessment (MT&A)

Nozzle Diameter (mm)	Extrusion Speed (RPM)	Bead Width (mm)	Layer Height (mm)	Estimated Gantry Speed (mm/min)	8" Hexagon Layer Time (sec)
2.0	5	3.00	0.75	852	84.4
	10	3.00	0.75	1733	41.5
	20	3.00	0.75	3594	20.0
	30	3.00	0.75	5543	13.0
	50	3.00	0.75	9503	7.6
	75	3.00	0.75	14379	5.0
	100	3.00	0.75	19213	3.7
	130	3.00	0.75	24787	2.9
	165	3.00	0.75	29894	2.4
4.0	10	6.0	1.5	462	155.7
	25	6.0	1.5	1165	61.7
	50	6.0	1.5	2298	31.3
	75	6.0	1.5	3385	21.2
	100	6.0	1.5	4435	16.2
	150	6.0	1.5	6474	11.1
	200	6.0	1.5	8494	8.5
	250	6.0	1.5	10506	6.8
	300	6.0	1.5	12391	5.8
6.0	15	9.00	2.25	307	320.0
	25	9.00	2.25	684	133.1
	50	9.00	2.25	1292	69.5
	75	9.00	2.25	1868	50.0
	100	9.00	2.25	2418	39.6
	150	9.00	2.25	3457	27.8
	200	9.00	2.25	4457	21.1
	250	9.00	2.25	5468	16.8
	300	9.00	2.25	6547	14.0

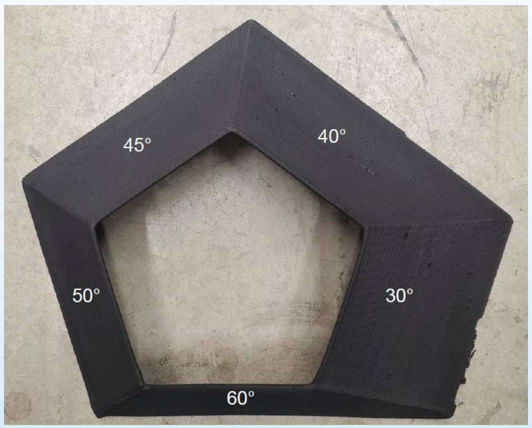




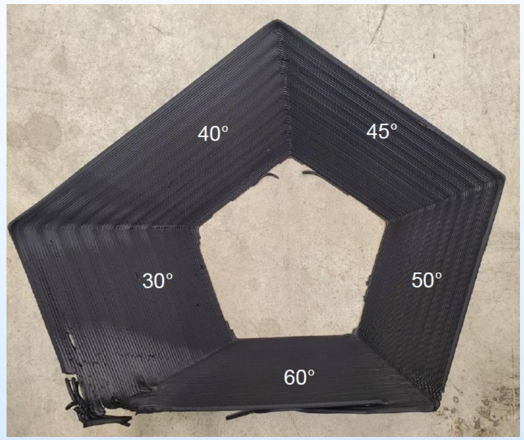
Hexagonal test prints

Progress and Current Status of Project

Material Testing & Assessment (MT&A)



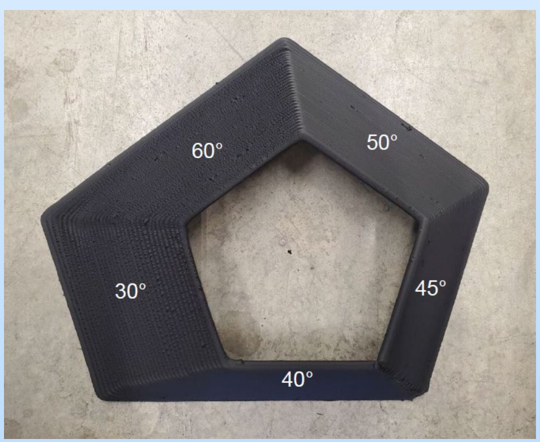
Negative overhang test (2 mm nozzle)



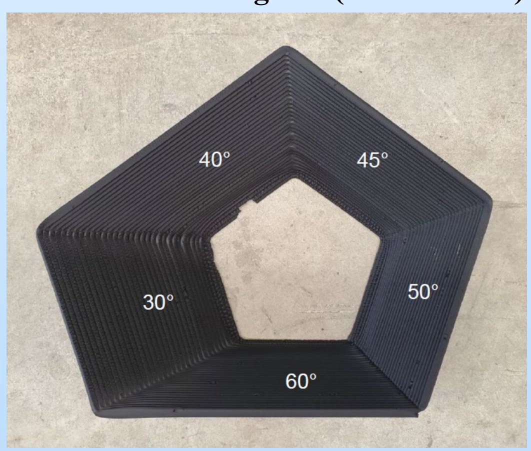
Positive overhang test (2 mm nozzle)


Bridge test (2 mm nozzle)

Bridged Gap (mm)	*Sag (mm)*
2.32	2.29
4.17	1.84
4.62	2.06
5.54	2.02
8.26	2.94
9.52	1.13
14.00	1.85
19.37	9.33



Negative overhang test (4 mm nozzle)



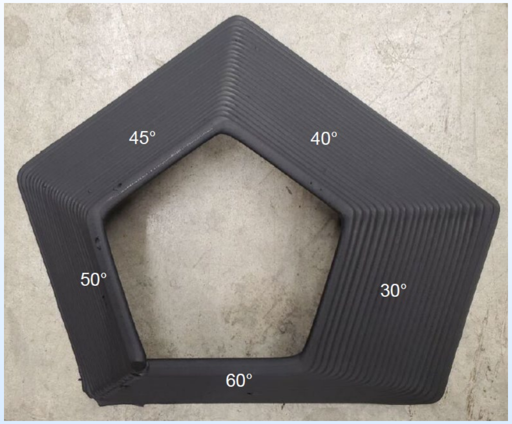
Positive overhang test (4 mm nozzle)


Bridge test (4 mm nozzle)

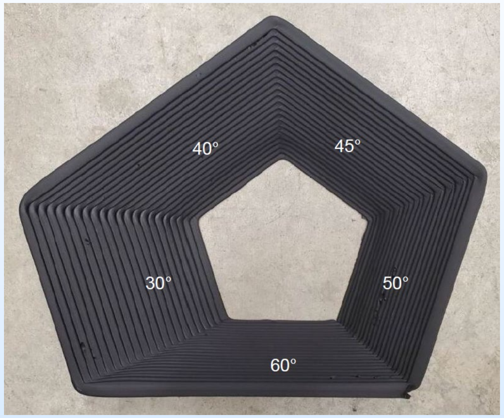
Bridged Gap (mm)	*Sag (mm)*
3.98	3.50
5.90	2.59
5.42	2.17
7.85	3.93
9.95	3.88
10.94	2.84
15.51	3.89
22.79	4.85

Progress and Current Status of Project

Material Testing & Assessment (MT&A)



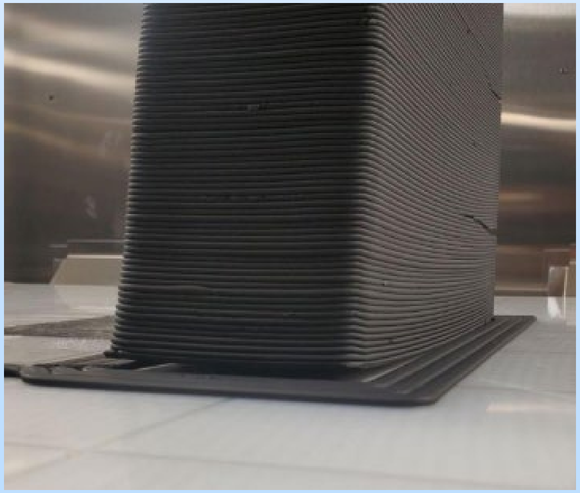
Negative overhang test (6 mm nozzle)



Positive overhang test (6 mm nozzle)

Bridge test (6 mm nozzle)

Bridged Gap (mm)	*Sag (mm)*
4.79	5.35
5.75	4.77
5.64	4.91
6.78	5.09
10.16	5.85
10.65	5.23
16.61	5.37
21.30	5.57



Wind Turbine Blade Tooling Printing Trial

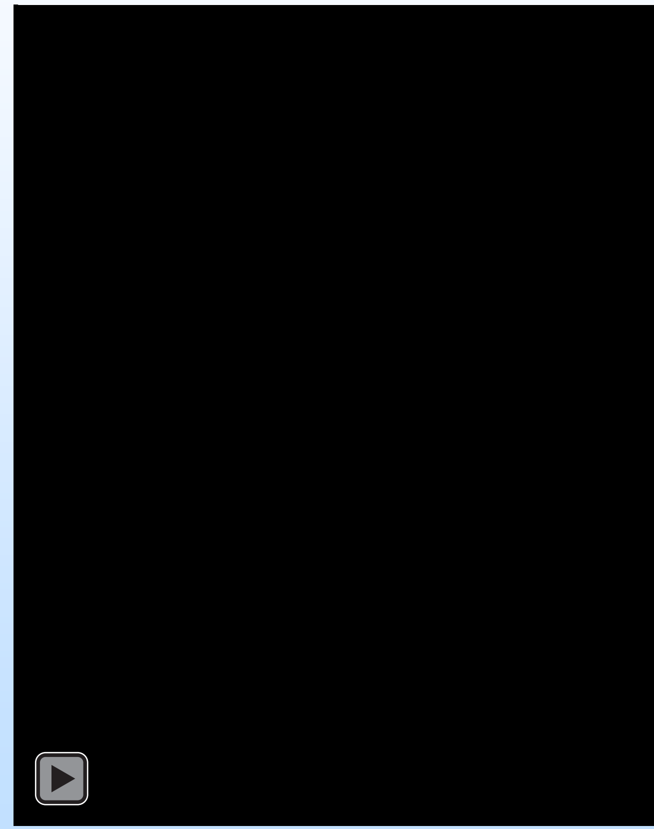
Plans for future testing/development/ commercialization

Current Project

- Conduct MT&A on additional composite formulations
- Continue assessment of large-scale printing for building/construction and tooling applications
- Assess the impact of porosity on performance using FEA
- Finalize the TEA and Market Analyses

Next Project:

- Pilot scale production
- ASTM and building codes compliance testing
- Target additional applications



Outreach and Workforce Development Efforts or Achievements

Outreach/Dissemination

- Technical Presentations: TMS (2023), SME (2024), SAMPE (2024)

ACS APPLIED POLYMER MATERIALS

pubs.acs.org/acsapm Article

3D Printing of Sustainable Coal Polymer Composites: Study of Processing, Mechanical Performance, and Atomistic Matrix–Filler Interaction

Logan E. Veley,* Chinonso Ugwumadu,

Cite This: https://doi.org/10.1021/acsapm.3c01784

ACCESS | Metrics & More

ABSTRACT: Bituminous coal was utilized as a polymer-based composites to fabricate standard 1. composite filaments for material extrusion 3D printi were formulated by incorporating Pittsburgh No. 8 acid, polyethylene terephthalate glycol, high-densit polyamide-12 resins with loadings ranging from 20 plastic composite filaments were extruded and printe processing parameters as the respective neat plastics. coal ameliorated the warping problem of 3D

Materials Today Communications 37 (2023) 106989

Contents lists available at ScienceDirect

Materials Today Communications

journal homepage: www.elsevier.com/locate/mtcomm

ELSEVIER

3D Printing of sustainable coal polymer composites: Thermophysical characteristics

L. Veley [a,b,*], J. Trembly [a,b], Y. Al-Majali [a,b,*]

[a] Institute for Sustainable Energy and the Environment (ISEE), Department of Chemical and Biomolecular Engineering, Ohio University, At
[b] Department of Mechanical Engineering, Ohio University, Athens, OH 45701, USA

ARTICLE INFO

Keywords:
Coal Plastic Composite
Additive Manufacturing
Fused Deposition Modeling
High-density Polyethylene
Sustainable Composite Materials

ABSTRACT

Recently, there has been a growing interest in enlisting polymer- structural applications such as wind turbine blade tooling and availability of sustainable and environmentally friendly 3D printal precluding the technology expansion to such applications. This re and environmentally friendly coal-plastic composites for use i polyethylene terephthalate glycol, high-density polyethylene, an

Carbon xxx (xxxx) 119086

Contents lists available at ScienceDirect

Carbon

journal homepage: www.elsevier.com/locate/carbon

ELSEVIER

The structure of appalachian coal: Experiments and *Ab initio* modeling

Chinonso Ugwumadu [a,*], David A. Drabold [a,**], Natasha L. Smith [b], Jason Trembly [c], Rudolph Olson III [b], Eric Shereda [d], Yahya T. Al-Majali [c,***]

[a] Department of Physics and Astronomy, Nanoscale and Quantum Phenomena Institute (NQPI), Ohio University, Athens, OH, 45701, USA
[b] CONSOL Innovations, Triadelphia, WV, 26059, USA
[c] Department of Mechanical Engineering, Institute for Sustainable Energy, and the Environment (ISEE), Ohio University, Athens, OH, 45701, USA
[d] CONSOL Energy, Canonsburg, PA, 15317, USA

ARTICLE INFO

Keywords:
Coal
NMR
FTIR
XPS
ReaxFF
DFT

ABSTRACT

This study focuses on the construction of small-scale atomistic representations of three Appalachian coals of different ranks– high-volatile bituminous, low volatile bituminous, and semi-anthracite. Ultimate, proximate, ^{13}C NMR, FTIR, and XPS data were used to infer 2D and construct multiple 3D molecular models for each coal. A new simulation strategy for structure optimization, termed "Sectioned Optimization", is introduced to ensure energetically stable configurations of the 3D coal models. Density functional methods are applied to the models, improving their quality, and giving insight into the limitations of empirical simulations in this complex system. The structural models were validated *ex post facto* using a density functional code, VASP. We describe the distribution

Outreach and Workforce Development Efforts or Achievements

Training and Professional Development

- Graduate Students: Trained three students on additive manufacturing, composite materials characterization techniques, and project reporting/presentations. One student successfully completed their program.
- Undergraduate Students: Provided hands-on experience for more than 15 students and up to seven students from different engineering disciplines will be trained this summer.

Summary

- High coal content composites were successfully extruded and 3D printed using lab- and large-scale commercially available 3D printers
- With increasing filler contents:
  - Generally, decreases in tensile strength, flexural strength, and impact resistance
  - Increases in tensile and flexural moduli
  - Increases in hardness
  - Decreases in CTE and warping
- Pellet-based AM will be more viable for large-scale applications
- Costs for AM of composite tooling are expected to be significantly lower than traditional methods

Acknowledgements

DOE/NETL

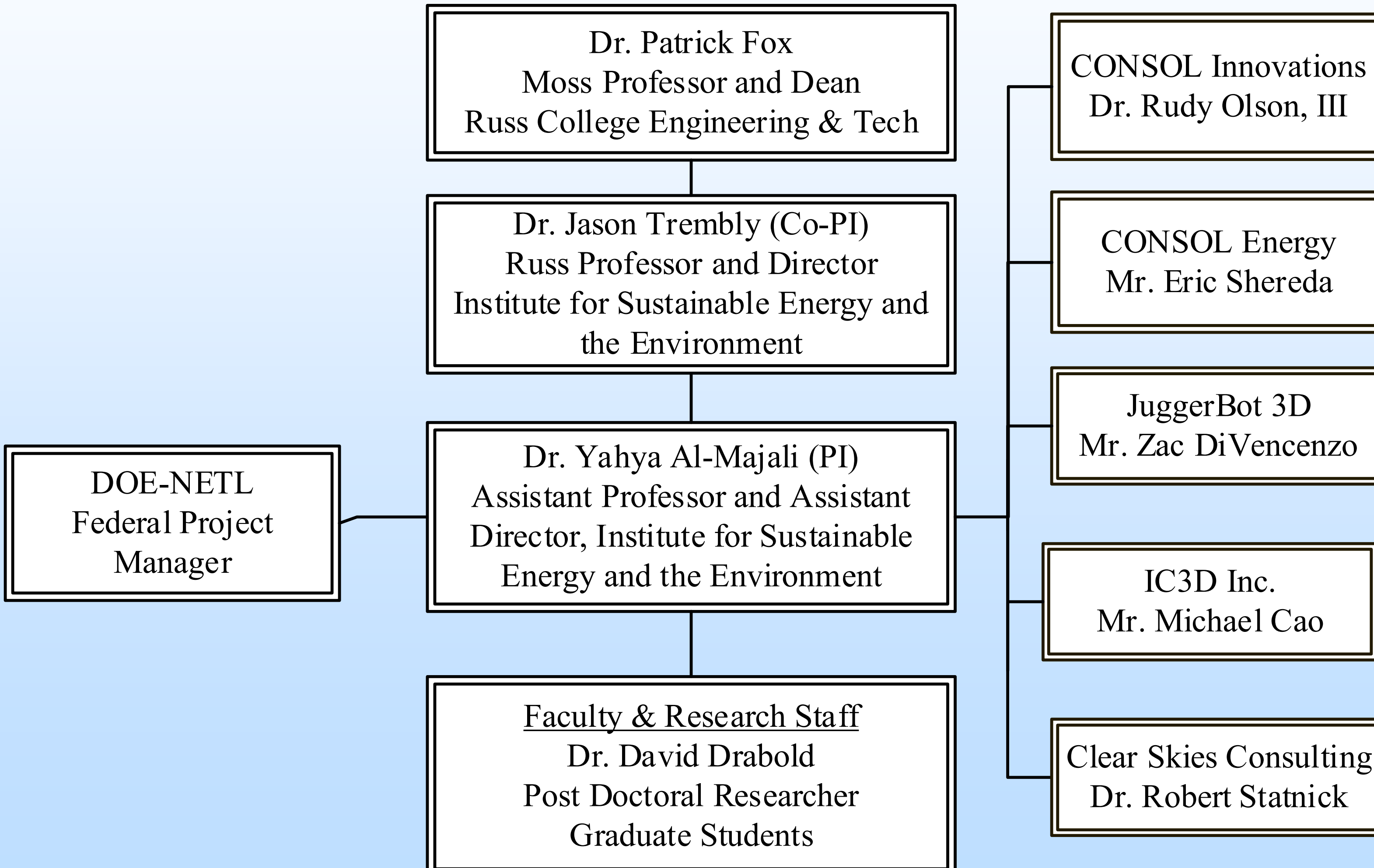
- Project Manager: Mark Render
- Technology Manager: Dr. Joseph Stoffa

Project Team

- OHIO: Grace Baranack, Logan Veley, Chinonso Ugwumadu, Dr. Jason Trembly, Dr. David Drabold
- CONSOL Innovations: Dr. Rudolph Olson
- CONSOL Energy: Eric Shereda
- JuggerBot 3D: Zac DiVencenzo, Daniel Fernback
- IC3D: Michael Cao
- Clear Skies Consulting: Dr. Robert Statnick

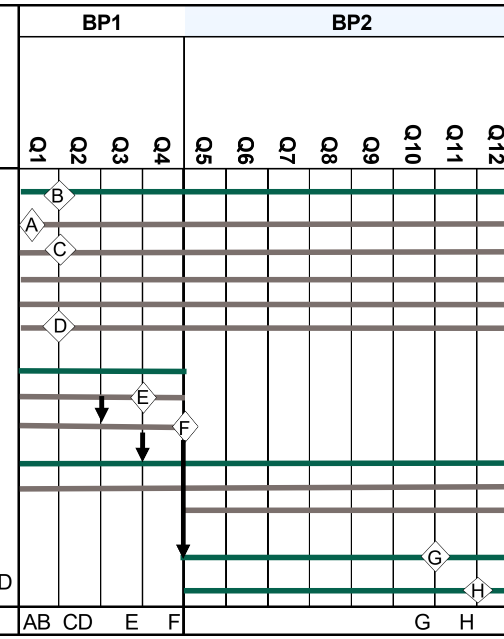
Appendix

Organization Chart



Gantt Chart

Task	Responsible Organizations
Task 1.0 - Project Management and Planning	OHIO
Sub-task 1.1 - Project Management Plan	OHIO
Sub-task 1.2 - Technology Maturation Plan	OHIO
Sub-task 1.3 - Summary of Environmental Justice Considerations	OHIO
Sub-task 1.4 - Summary of Economic Revitalization and Job Creation Opportunities	OHIO
Sub-task 1.5-Environmental, Safety, and Health (ES&H) Analysis	OHIO
Task 2.0 - Coal-Enhanced Filament R&D	
Sub-task 2.1 - Coal-Enhanced Filament Formulation Development	OHIO, CE
Sub-task 2.2 - Coal-Enhanced Filament Performance	OHIO, CFOAM
Task 3.0 - Multi-scale Computational Material Simulations	OHIO
Sub-task 3.1 - Molecular Dynamic Simulation of Carbonized 3D Printed Coal-enhanced Filaments	OHIO
Subtask 3.2 –Finite Element Analysis of 3D Printed Structures	OHIO
Task 4.0 – Assessment of Coal-Enhanced Filament Printability	OHIO, CFOAM, JuggerBot, IC3D
Task 5.0 –Techno-economic Studies and Market Analyses	OHIO, CFOAM, CE, JuggerBot, IC3D
Milestone Log	



Ohio University (OHIO), CONSOL Energy (CE), CFOAM LLC. (CFOAM), JuggerBot 3D (JuggerBot), IC3D Inc. (IC3D)

Milestones: A: Updated Project Management Plan; B: Project Kickoff Meeting; C: Preliminary Technology Maturation Plan; D: Environmental, Safety, and Health (ES&H) Analysis; E:Coal-Enhanced Filament Formulation Report; F:Coal-Enhanced Filament Properties Report; G: Coal-Enhanced Filament Printability Analyses; H:Techno-economic and Market Analyses