3D Printed Hybrid Composite Materials with Sensing Capability for Advanced Vehicles

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Project ID: 36928.

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Overview

Timeline

- Start: October 2020
- End: October 2023
- 22% Complete

Budget

- Total Project Funding
- $ 1.5 Million in 3 Years
- $ 500/year in 3 Labs

Barriers

- Continuous fiber extrusion and 3D printing machine needs accelerated development for thermoset type of materials.
- Synthesis of new epoxy and thermoset materials blends that have the right viscosity and curing behavior for continuous fiber printing.
- Initial demonstration of a sandwiched sensor device and a fabricated composite layer

Partners

- ORNL: CNMS
- ORNL: CSD
- University of North Texas

Relevance

- Additive manufacturing (AM) enables rapid development of prototype parts and digital manufacturing yet there is an unmet need on high performance parts from continuous fiber composites.
- Materials development and simulation simultaneous with new 3D printing process development is key to improving advanced vehicle manufacturing throughput.
- *If successful,* new vehicle parts development methods can take advantage of AM early with integration of parts design simplification and provision for device health monitoring.

OBJECTIVES

- *1) enhanced organic-inorganic interface for long-term performance via covalent bonding between CF and polymer matrix and,*
- *2) real time evaluation of material properties with embedded sensors.*

… will be addressed in 4 Main Tasks:

Mission statement: *We will pursue research and development of advanced high-performance carbon fiber composites and structures through interfacial chemical optimization in demonstrated additive manufacturing (3D Printing) and structural monitoring through embedded sensors.*

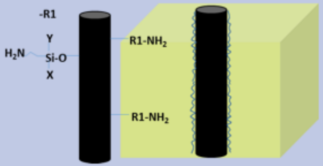
Milestones

Milestone Description		STATUS	FY	Qtr
1. Precise chemical reaction control in composite materials				
1.1	Study on resin development and chopped carbon fiber composite reactivity	Complete	21	Q1
1.2	Surface modification of low-cost carbon fiber with reactive group	On Schedule	21	Q3
1.3	Demonstration of chemical reaction of carbon fiber and polymer matrix	On Schedule	21	Q4
2. Computational studies of interfacial failure and mechanical properties				
2.1	Simulation of Matrix and mechanical properties of a composite*	Complete	21	Q1
2.2	Develop cohesive models to simulate interfacial interactions	On Schedule	21	Q3
3. 3D printing of continuous fiber/composite system with enhanced interface				
3.1	3D Printing of resin epoxy composite with initial tensile testing measurements	On Schedule	21	Q4
3.2	Optimization of 3D printing process for continuous carbon fiber/polymer composite with ultimate tensile strength > 200 MPa and Young's modulus > 10GPa	On Schedule	21	Q4
4. Sensor-embedded fiber/polymer composite with enhanced interface				
4.1	Sensor material development and compatibilization	Complete	21	Q1
4.2	Identify most suitable printable solution for 2D materials printing	On Schedule	21	Q4

* Paper submitted: "Characterize traction-separation relation and interfacial imperfections by data-driven machine learning models", which was submitted to *Scientific Reports* .

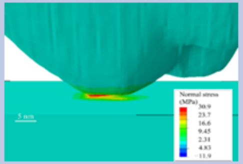
Approach

Precise Chemical Reaction Control



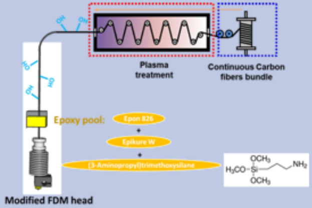
CF/polymer composites with enhanced interface via covalent reactions

Computation Interfacial Interaction



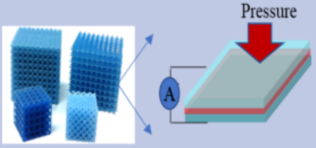
Failure mechanisms of the printed structure & stability by finite element analysis

3D Printing CF /Polymer Composites



Epoxy/continuous CF composites with controlled distribution & adhesion

Embedded Sensor Composite Printing



Embedded mechano-electric sensor synchronously with 3D printed CF composite

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Data for Interface Bonding & Composition

Model for Interface Failure Mechanism & Stability

Novel Printed Structure & IP, Publication

Novel Embedded Sensor & Publication

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The focus is on enhancing interfacial interaction between CF/polymer and monitoring material performance, leading to greater *isotropic* enhanced mechanical properties (Young's modulus >15 GPa and tensile strength> 250 MPa)[1] in technical readiness level (TRL 3) metrics and future TRL 4 studies.

Tasks Summary:

Task 1. Precise chemical reaction control in resin materials

Principal Investigator: Pengfei Cao of ORNL

Objective: Develop CF/polymer with enhanced inorganic-organic interface covalent interaction

End-of-project Goal: Composites with optimum chemistry, processability, and high performance.

Collaborators: Rigoberto C Advincula and UNT Group

Task 2. Computational studies of interfacial interaction between polymer matrix and CF

Principal Investigator: Wonbong Choi and Yijie Jiang of UNT

Objective: Simulation of interfacial properties and optimum reactive chemical species

End-of-project Goal: Achieve high correlation of printed components with predictive tools.

Collaborators: Pengfei Cao and Rigoberto Advincula

Task 3: 3D printing of continuous CF/epoxy composite with enhanced fiber-polymer adhesion

Principal Investigator: Rigoberto Advincula of ORNL

Objective: 3D printing continuous CF-epoxy matrix with optimal fabrication parameters.

End-of-project Goal: Achieve AM process and materials combination for high performance.

Collaborators: Pengfei Cao, Yijie Jiang of UNT and Hyrel 3D and TCPoly.

Tasks 4. Continuous sensor-embedded polymer/carbon fiber composite 3D printing

Principal Investigator: Rigoberto Advincula and Wonbong Choi of ORNL and UNT

Objective: 3D print continuous CF/polymers with embedded sensor geometries and testing.

End-of-project Goal: Achieve sensing capability in continuous CF/epoxy 3D printed parts

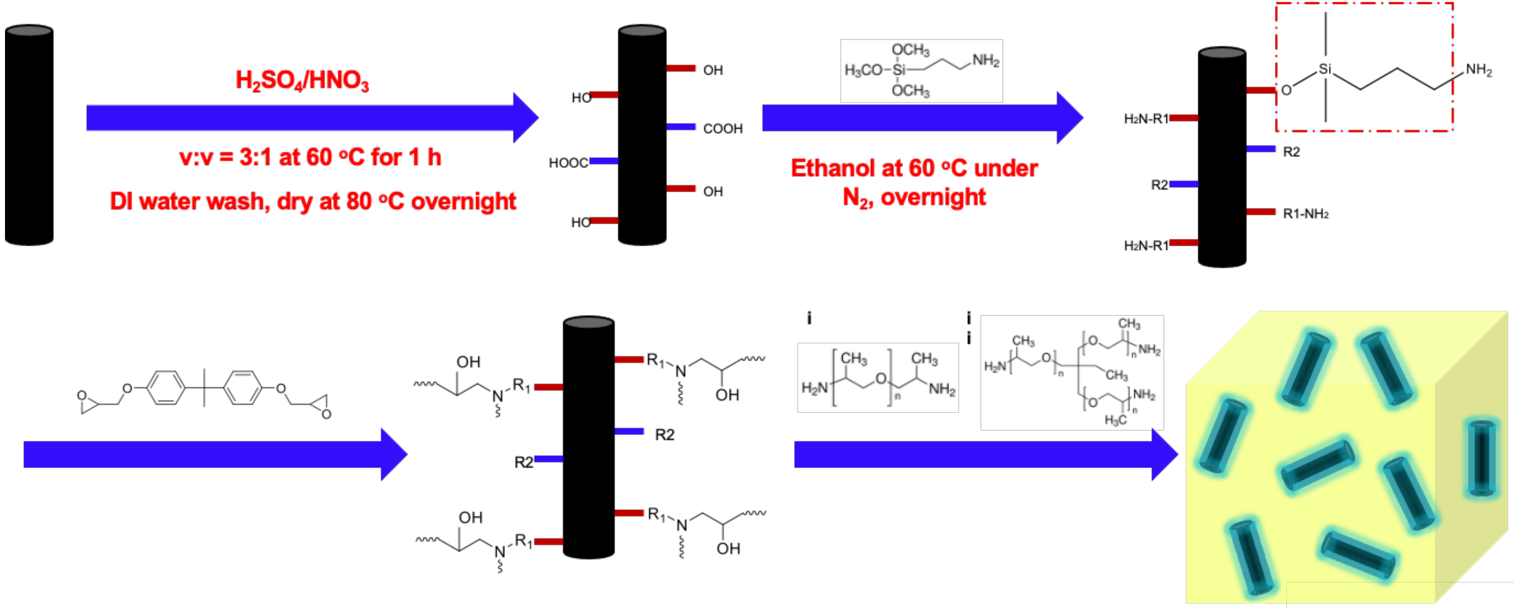
Collaborators: Pengfei Cao, Yijie Jiang and Hyrel 3D

Accomplishments:

R1

Task 1: Precise chemical reaction control in resin materials

- A comprehensive study of epoxy/chopped CFs has been performed; the sample with 50 μm CFs shows promising results, which is good for the fundamental study.
- Model epoxy resins were investigated along with optimized protocols for grafting of surface function al groups in CFs.
- improved the performance of composites allowing the desired 200 MPa of strength to be reached even with chopped CFs. Initial target: **10 GPa**, tensile strength of **200 Mpa.**



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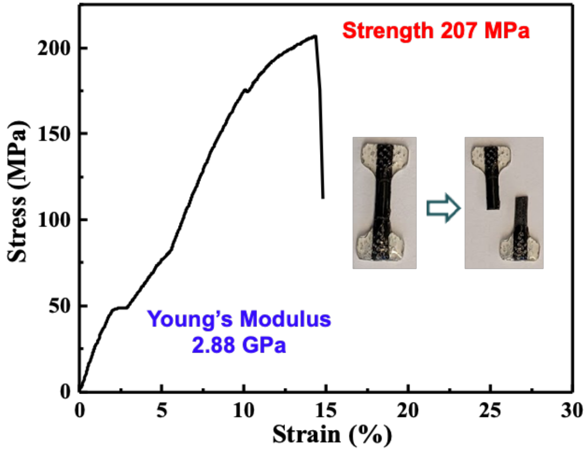
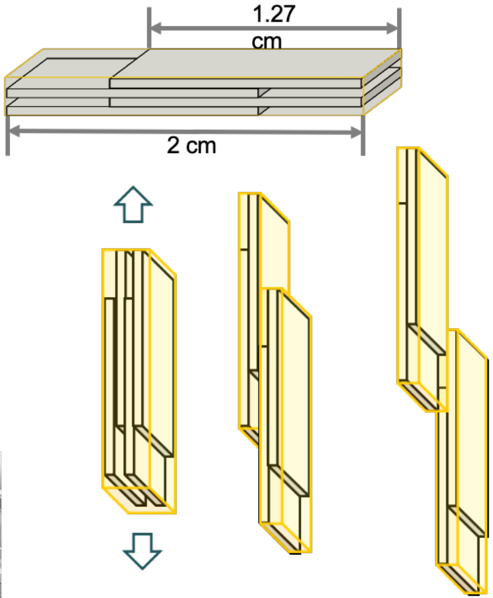
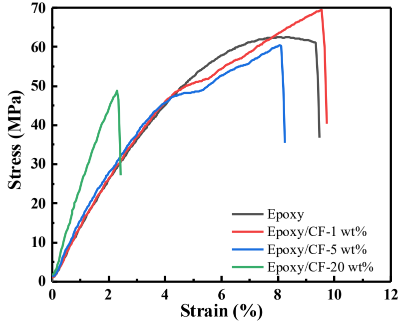
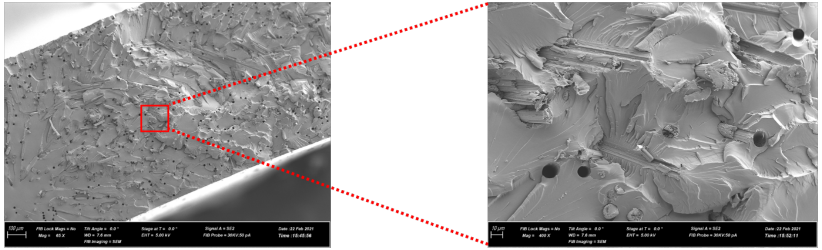




SEM images of Epoxy with 10 wt.% silane-treat CFs





➢ No obvious phase separation observation, homogenous distribution of CFs

- The mechanical strength of the sample with 1 wt.% CF is comparable to that of pristine polymer, while decreases with further increment of CF loading (i.e., 5 wt.%, 20 wt.%)
- The phase separation could be observed for the samples with 1 and 5 wt.% CFs
- The uniform distribution of CFs into the system during the whole process is critical

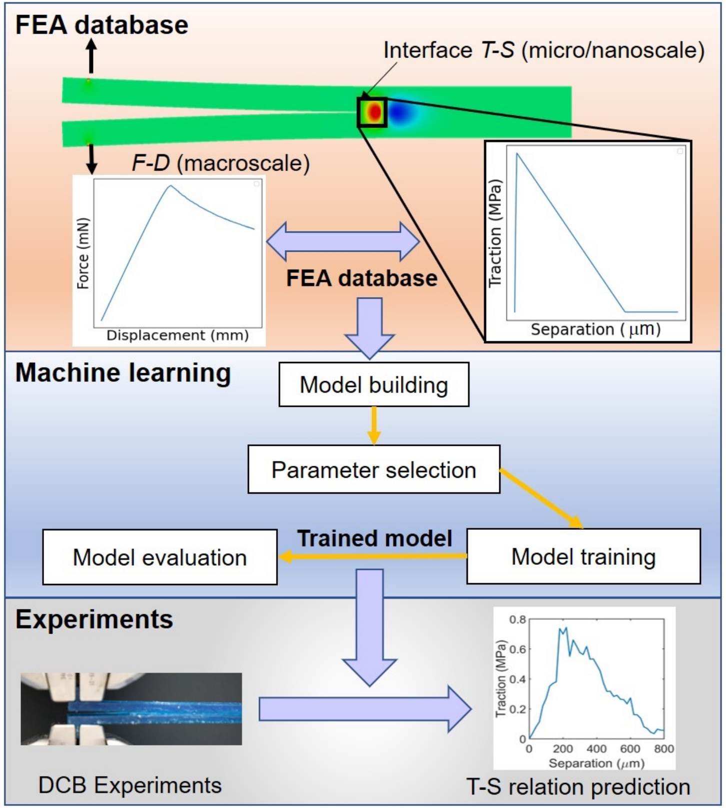
Accomplishments:

Task 2: Computational studies of interfacial interaction between polymer matrix and CF.

- Established data-driven ML models to precisely predict the interface adhesion properties and identify imperfections along interfaces from finite element analysis (FEA) and standard experiments

- Preliminary FEA simulations on fiber reinforced composites with multiscale fibers are performed

- Developed algorithms for material programming of crack design and high tough and high strength composites are on-going. Establishment of genetic algorithm and FEA iterations: crack propagation simulations with designed composites



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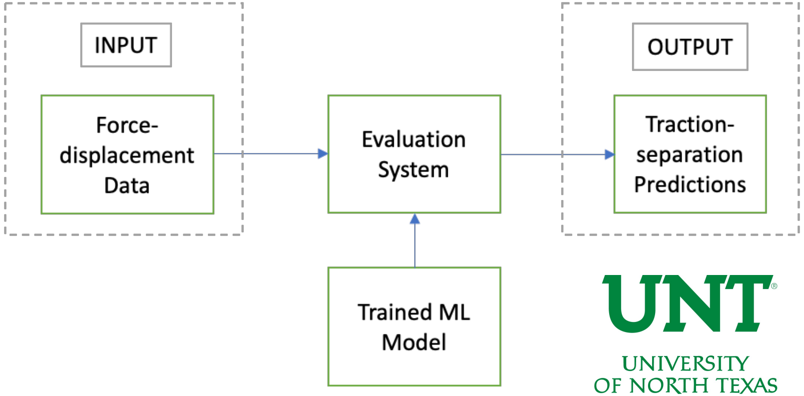

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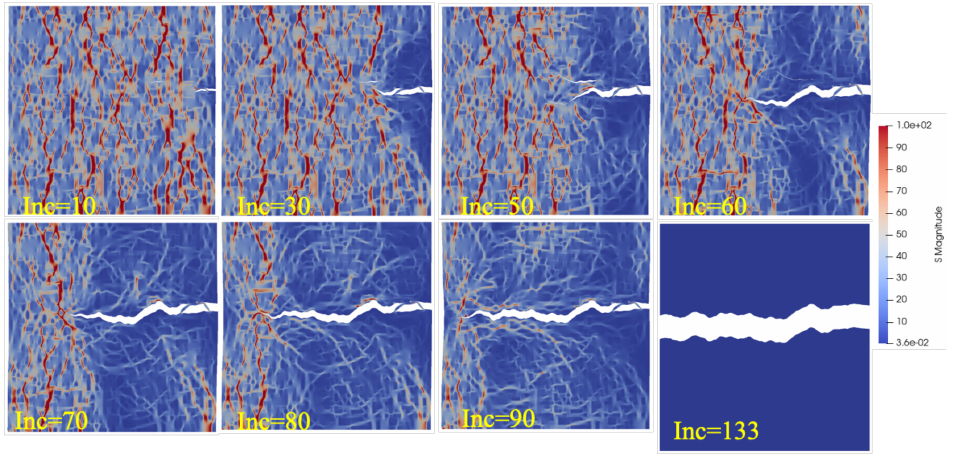
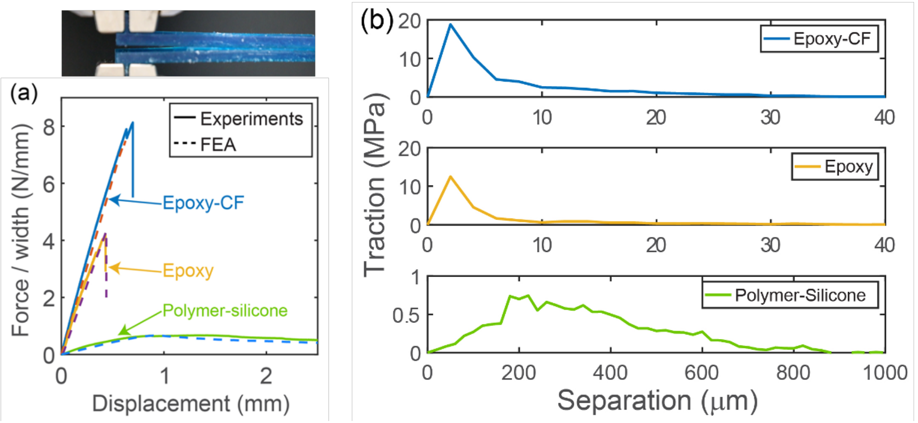




- We experimentally characterized the interface between epoxy, epoxy-carbon fiber, and polymer-silicone interfaces and obtain force-displacement data.
- Using the experimental force-displacement data, T-S laws at interfaces are predicted and evaluated using FEA to compare with experiments.
- Crack propagation model: Initial tests using an A-FEM method allow failure across the composite plate

* Paper submitted: "Characterize traction-separation relation and interfacial imperfections by data-driven machine learning models", which was submitted to *Scientific Reports* .

Accomplishments:

Task 3: 3D printing of continuous CF/epoxy composite with enhanced fiber-polymer adhesion

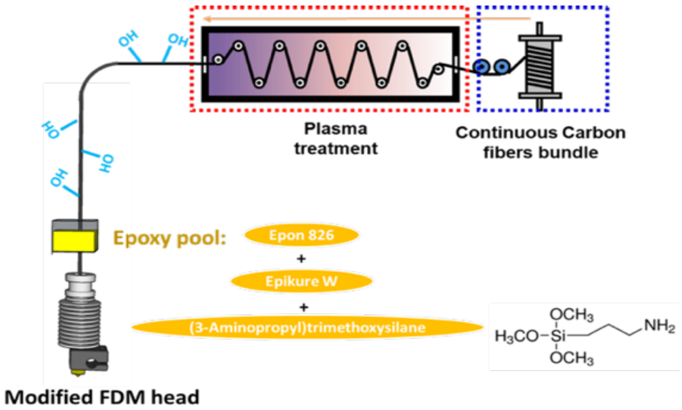
- 3D Printing of epoxy-CF composite was achieved using a viscous solution printing (VSP) based method. The wt% composition was studied up to 40%

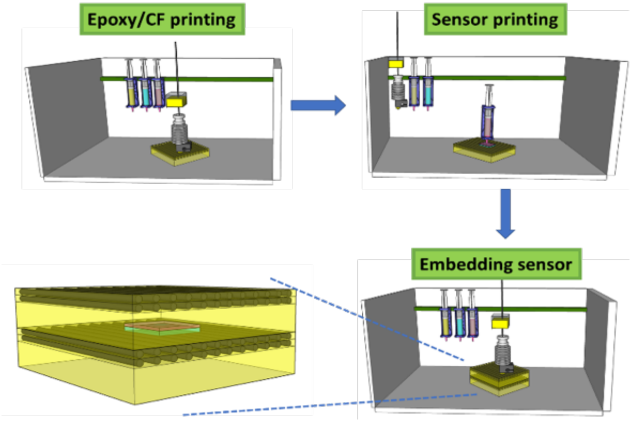
- Rheology and mixing of commercial based epoxy and chopped CF was used to demonstrate upscalability of the process: Epon, Epikure, silica nanoparticles added.

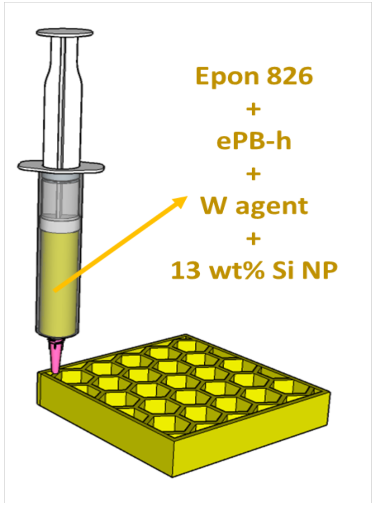
- Loading of CF at 40 wt%. Tensile strength: 73 MPa. Modulus at 2.8 Gpa but Elongation decreased to 3%. The toughness is maximized at 20 wt%

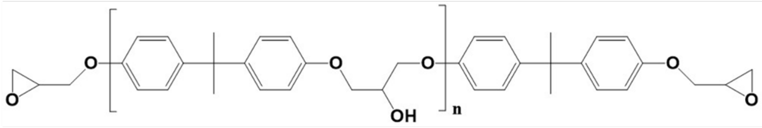
- Investigate early the multimaterial 3D printing with CF mat pre-preg.

Design for Continuous CF 3D Printing

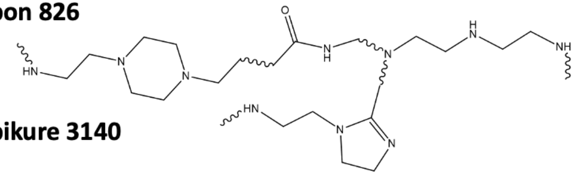




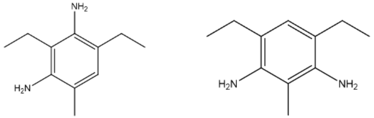




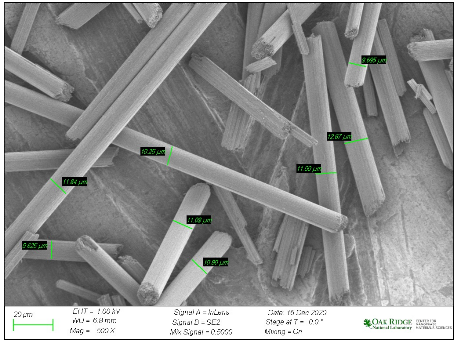
Epon 826

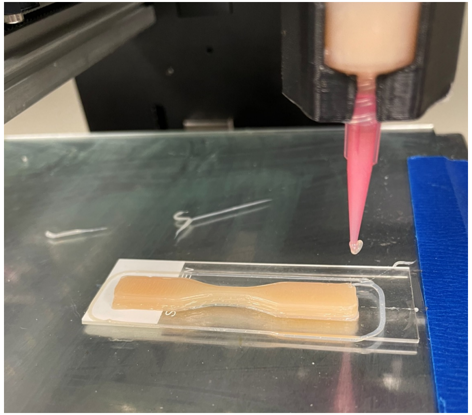


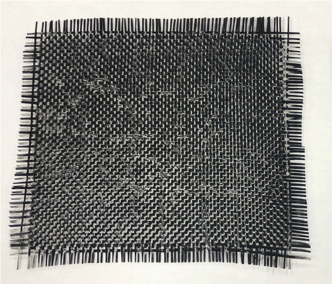
Epikure 3140

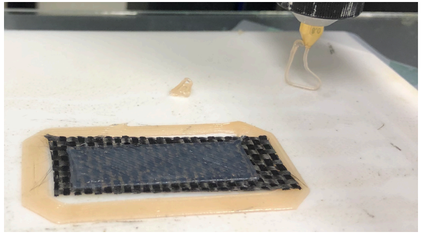


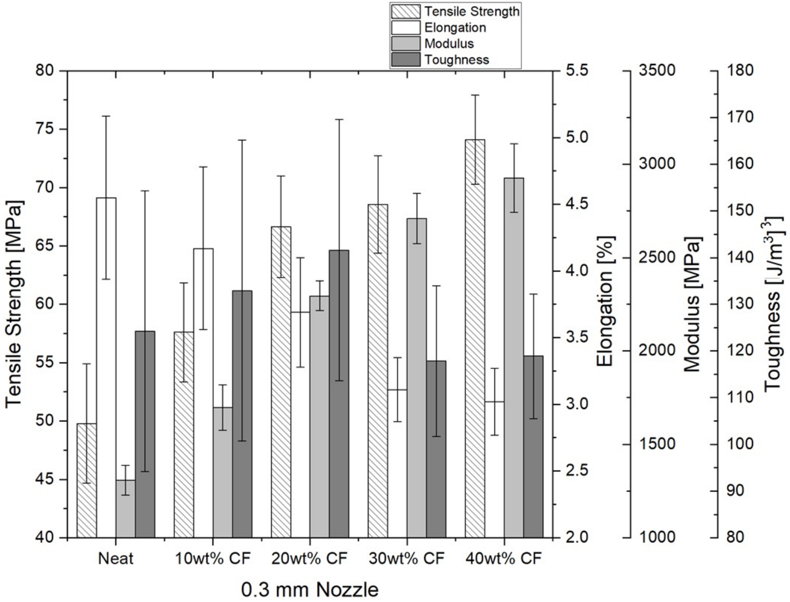
Epikure W







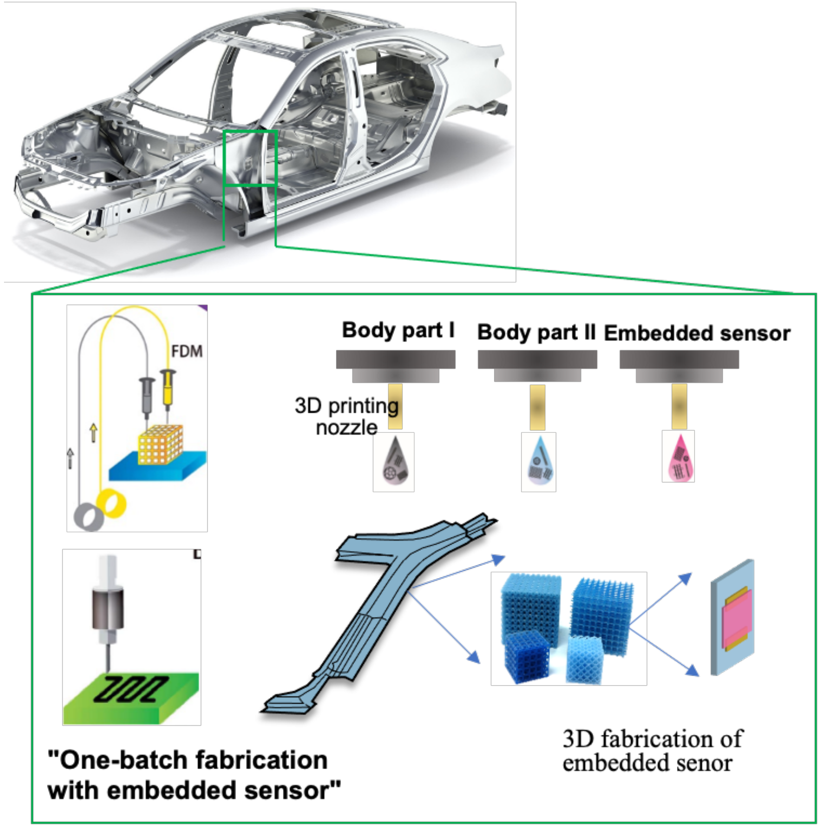




Accomplishments:

Task 4: Continuous sensor-embedded polymer/carbon fiber composite 3D printing

- The end goal is to design and demonstrate multi-material 3D printing to incorporate in-situ and embedded sensors with the concept of printing vehicle components that can be monitored with stress and time.
- We have developed protocol and 3D printing inks for embedded sensors and Zn-ion battery.
- Designed and fabricated porous structures of Zn-anode having high surface area for high efficiency Zn-anode.
- Successfully fabricated flexible Zn-anode and presented high specific capacity of 650mAh/g.



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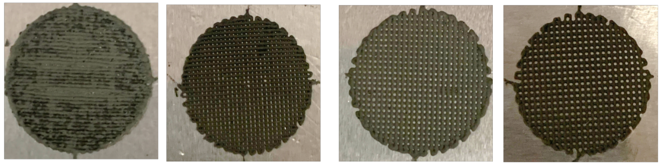
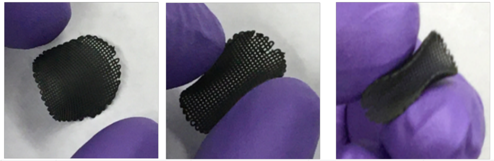
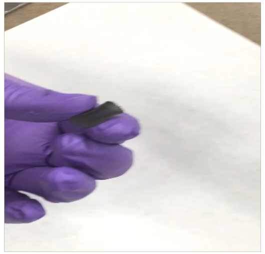
S0 S1 S2 S3

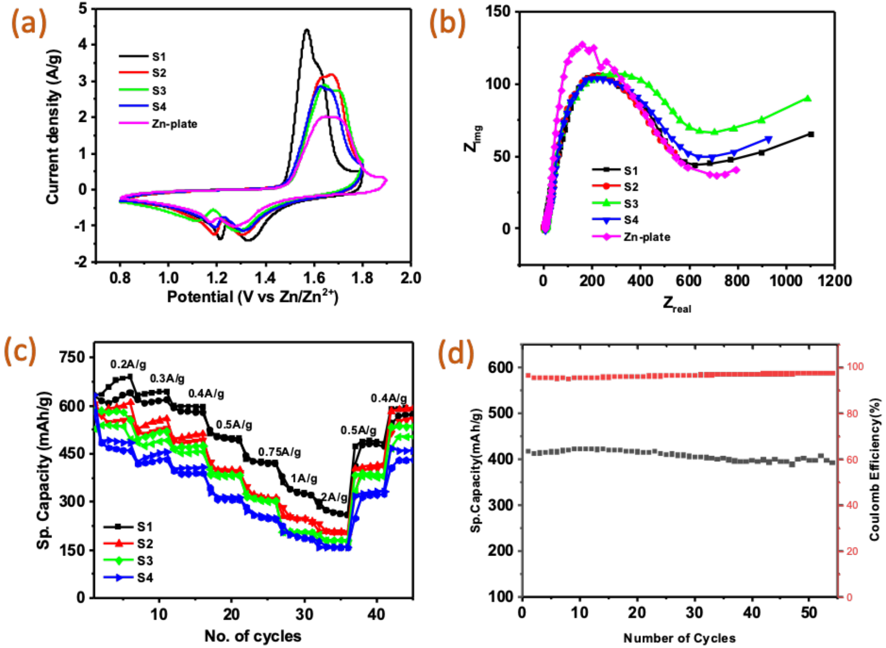


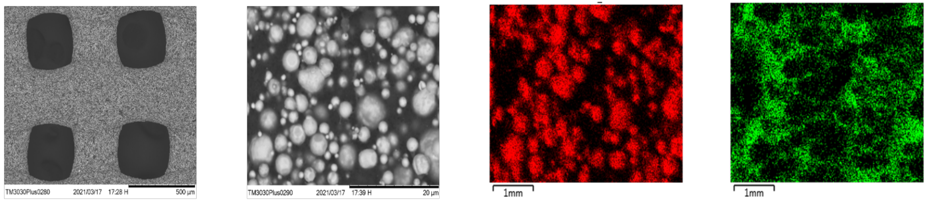




Samples	Filament Distance(cm)	Surface area (Cm2)	Geometry Porosity	S-V ratio (1/cm)
S0	0	2.7	0.5	18.9
S1	0.062	6.1	0.71	65.2
S2	0.079	5.5	0.78	63.6
S3	0.089	5.1	0.79	60.4

3D Printed Zn Anode:
Macropore structures in 3D printed anodes show homogeneous distribution of ink materials with no agglomeration in printed structure Carbon particles act as bridges between zinc particles improving conductivity.





Full Cell Characterization: (a) High reaction kinetics with increasing surface area. (b) Series resistance does not differ significantly; Zn-plate has high charge transfer resistance; (c) High specific capacity of 650mAh/g at 0.2A/g C-rate; (d) 3D anode maintained stable and high capacity for long term stability test

Response to Previous Year Reviewers' Comments:

This is the first year that the project has been reviewed.

Collaboration and Coordination:

- *Since the beginning, the Task leaders and the members of the groups meet continuously every 2 weeks to discuss results and coordinate future work.*

- We have divided the effort into corresponding Tasks in order to have a parallel approach that allow comparison of data and plan for future work more efficiently.

- The coordination between ORNL and UNT is seamless and beyond the regular meeting, a lot of discussions and joint experiments are done in-between group members- email and calls (zoom).

- Successful outcomes on milestones and a first publication has been achieved.

Task 1. Precise chemical reaction control in resin materials: *Pengfei Cao of ORNL*

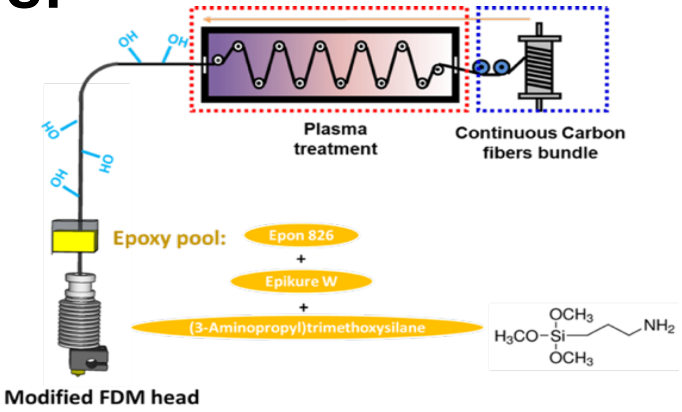
Task 2. Computational studies of interfacial interaction between polymer matrix and CF: *Wonbong Choi and Yijie Jiang of UNT*

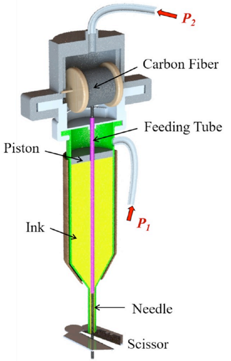
Task 3: 3D printing of continuous CF/epoxy composite with enhanced fiber-polymer adhesion: *Rigoberto Advincula of ORNL*

Tasks 4. Continuous sensor-embedded polymer/carbon fiber composite 3D printing: *Rigoberto Advincula and Wonbong Choi of ORNL and UNT*

Remaining Challenges and Barriers:

- Continuous fiber extrusion and 3D printing machine needs accelerated development for thermoset type of materials. Discussions with Hyrel is underway but also in-house development – Up to 21/Q4
- Synthesis of new epoxy and thermoset materials blends that have the right viscosity and curing behavior for continuous fiber printing. Two sets of epoxies were used initially but we will need to have a unified formulation approach.
- Initial demonstration of a sandwiched sensor device and a fabricated composite layer. The two tasks will eventually be using the same printer platform – Up to 21/Q4.
- Long-Term: demonstrated AM (3D Printing) and structural monitoring through embedded sensors.





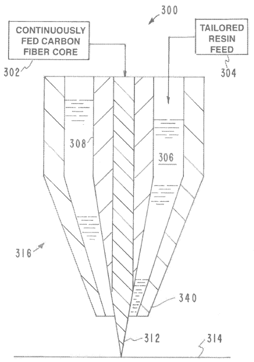


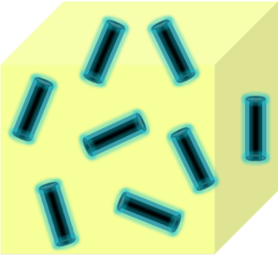
FIG. 3

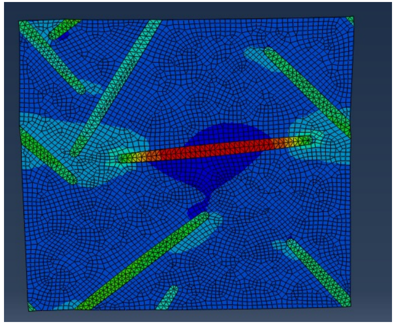
Additive Manufacturing 40 (2021) 101921

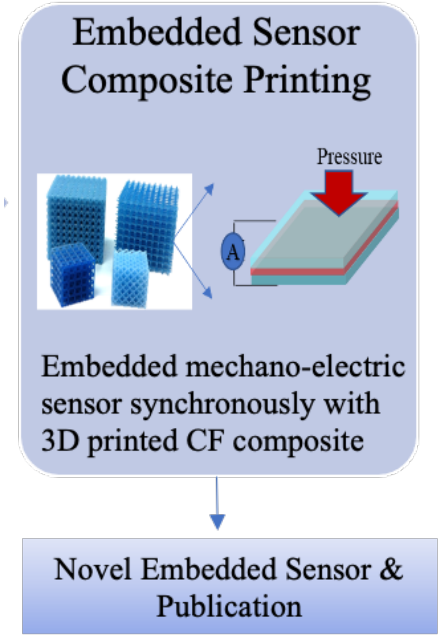
Lewicki, J., Lawrence Livermore National Security LLC, 2018. *High performance, rapid thermal/UV curing epoxy resin for additive manufacturing of short and continuous carbon fiber epoxy composites*. U.S. Patent 9,944,016.

Proposed Future Research: FY 2021-2022

- Test composites of different concentrations and T and do the corresponding characterization (i.e., SEM, TEM, XPS, etc.)
- Correlate experimental study with simulation to unveil the specific mechanism: examine its 3D printing performance and investigate long-term thermo-mechanical properties of CF/polymer composites.
- Establishment of genetic algorithm and FEA iterations for the rest of the project including specific crack propagation simulations with future designed composites
- Higher wt% CF/epoxy composites and work with Hyrel to fabricate the continuous carbon fiber/epoxy 3D printing system.
- Develop 2D printable inks for sensor and printing: collect and analyze 3D printed sensors and Zn-ion battery data and develop a high-resolution sensor 3D fabrication with the epoxy/CF composite.







Summary

TECHNICAL ACCOMPLISHMENTS:

- A comprehensive study of epoxy/chopped CFs has been performed; Model epoxy resins were investigated along with optimized protocols for grafting of surface function al groups in CFs.
- Established data-driven ML models to precisely predict the interface adhesion properties and identified imperfections from FEA and standard experiments. Developed algorithms for material programming of crack design and high tough and high strength composites.
- 3D Printing of epoxy-CF composite was achieved using a viscous solution printing (VSP) based method and the development of a continuous CF printer is ongoing
- Developed a protocol and 3D printing inks for embedded sensors and flexible Zn-ion anode and battery to enable high-resolution sensor 3D fabrication with the epoxy/CF composite.

OPERATIONAL HIGHLIGHTS

- Initiated the project in 2020 on four Tasks on schedule with short-term (FY-21) and path for long-term (FY22)
- Cohesive team between ORNL and UNT that meets regularly: every two weeks.
- Submitted the first paper.