Integrated Computational Materials Engineering (ICME)

Predictive Tools Development for Low Cost Carbon Fiber for Lightweight Vehicles

- 2021 Annual Merit Review -

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June 22, 2021

Project ID: MAT124

University of Virginia Engineering

Timeline & Budget

Timeline

- Start Date: October 1, 2017
- End Date: March 31, 2021 (with 6 month no-cost extension)
- This period was broken into three annual phases of work
- Percent Complete: 100%

Budget

- *Approved* Project Funding: $4,408,032
  - $ 3,000,000 Federal
  - $ 418,032 Cost Share
  - $ 990,000 LightMat Consortium (Leveraged)
- *Actual* Project Funding: $4,408,629
  - $ 3,000,000 Federal (UVA, PSU, ORNL)
  - $ 418,629 Cost Share (Contributions by Solvay *exceed* required cost share)
  - $ 990,000 LightMat Consortium (Leveraged by ORNL & Co-PIs Klett and Vautard)

Project ended on March 31, 2021

Timeline & Budget

Budget

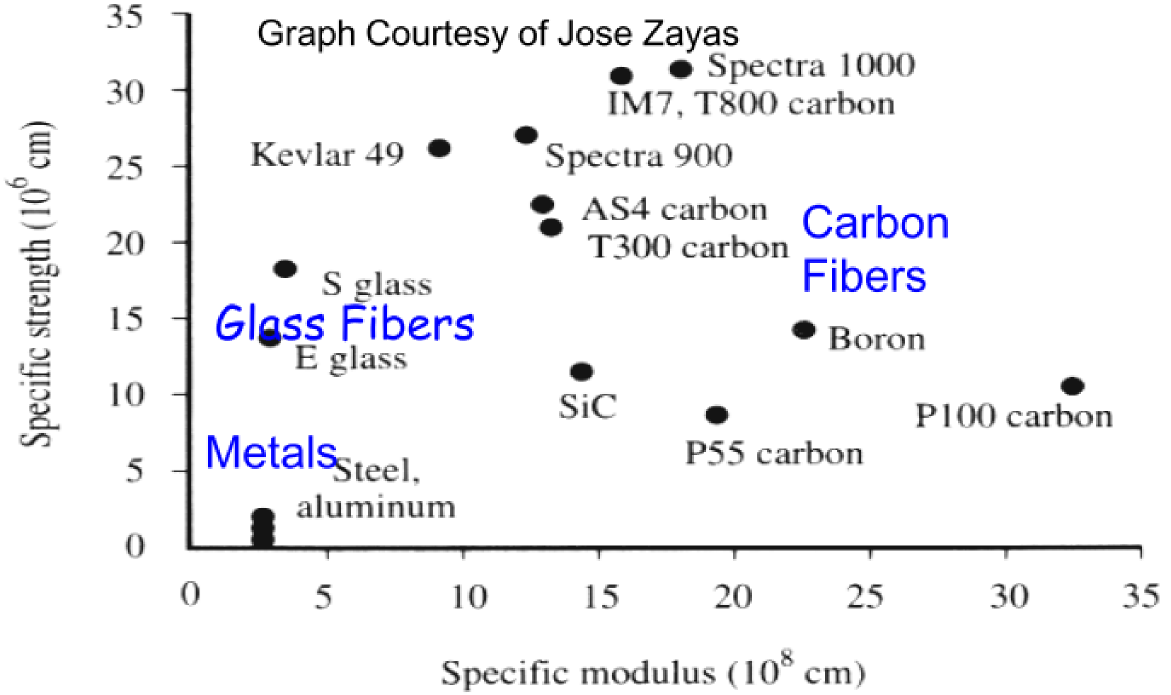
- Below is a breakdown of the actual costs incurred
- UVA, PSU, and ORNL used all available funding
- Solvay contributed more than the required cost share, exceeding our 20% target

	Approved Budget			Actual Costs Incurred		
	EERE	**FFRDC**	**Cost Share**	**EERE**	**FFRDC**	**Cost Share**
UVa	$2,175,000	$0	$0	$2,175,000	$0	$0
PSU	$525,000	$0	$0	$525,000	$0	$0
ORNL	$0	$300,000	$0	$0	$300,000	$0
Solvay	$0	$0	$418,032	$0	$0	$418,629
Total ($)	**$2,700,000**	**$300,000**	**$418,032**	**$2,700,000**	**$300,000**	**$418,629**

Project ended on March 31, 2021

Relevance

- Reduction of vehicle weight necessitates lower-density materials with suitable mechanical properties, low-cost carbon fiber
  - Source: 2017 U.S. DRIVE MTT Roadmap Report, Section 3

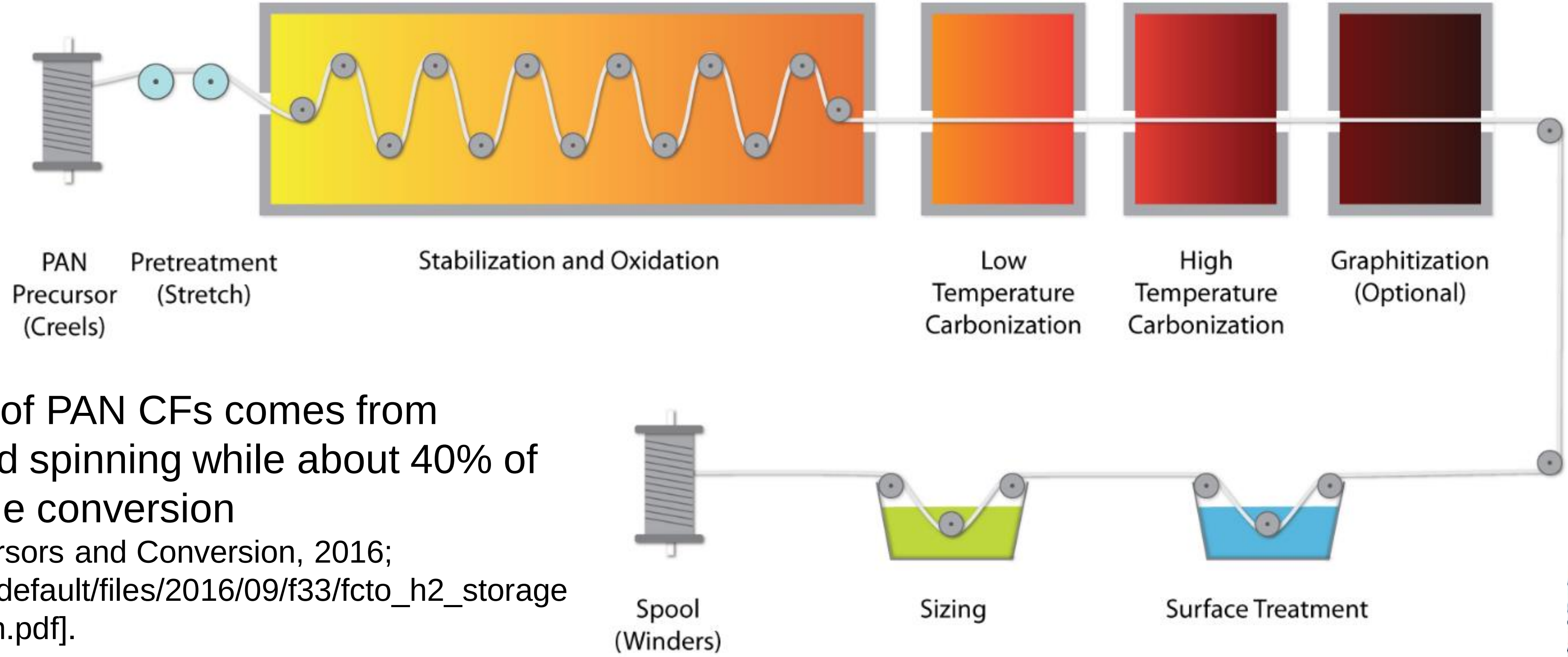


Graphical comparison of strength and modulus properties of structural metals, glass fibers, and carbon fibers

[Warren, Carbon Fiber Precursors and Conversion, 2016; https://www.energy.gov/sites/default/files/2016/09/f33/fcto_h2_storage_700bar_workshop_3_warren.pdf].

Relevance

- PAN-derived carbon fibers are the current state-of-the-art
  - Possess good properties (moderate modulus ~230 GPa, high strength >3500 MPa, moderate ductility >1.5 %)
  - Expensive to produce (10 to 13 $/lb)



About 50% of the cost of PAN CFs comes from precursor materials and spinning while about 40% of the cost comes from the conversion
[Warren, Carbon Fiber Precursors and Conversion, 2016; https://www.energy.gov/sites/default/files/2016/09/f33/fcto_h2_storage_700bar_workshop_3_warren.pdf].

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Project ended on March 31, 2021

Relevance

Objectives

- A low-cost, high strength alternative to PAN is needed for lightweight vehicles
- We sought to develop a calibrated ICME predictive tool that can identify & optimize fiber processing parameters
- Exploit this ICME framework to identify alternative precursors for carbon fiber with the following properties
  - Cost < $5/pound
  - Strength ≥ 250 Ksi (1725 MPa)
  - Modulus ≥ 25 Msi (172.5 GPa)
  - Strain ≥ 1%

Project ended on March 31, 2021

Phase 1 Tasks & Milestones

- Each task had a corresponding milestone

Date	Milestones	Status
September 30, 2018	**M1:** Statistical analysis of PAN precursor oxidation, baseline reference for comparison with statistical results of thermomechanical testing laboratory synthesized fibers.	**Met**
September 30, 2018	**M2:** Chemical conversion of PAN-based carbon fibers and verification through direct comparison with M1. Milestone will be met if simulations correctly identify resultant properties within 15% error margin.	**Met**
September 30, 2018	**M3:** Fiber and fiber/matrix mechanics correctly predicted by ICME simulations as compared to resultant properties and characteristics of synthesized fibers.	**Met**
September 30, 2018	**M4:** Wet/melt spinning optimization for preliminary production-scale synthesis able to reproduce laboratory-scale fiber properties at-scale.	**Met**
September 30, 2018	**Go/No Go M5:** Accurate prediction of PAN-fiber properties. Simulations will estimate PAN-based fiber properties (strength, modulus, strain) within a 15% error margin. Results will be verified with mechanical testing of the fibers to confirm a go-decision.	**Met**

Phase 2 Tasks & Milestones

- Each task had a corresponding milestone

Date	Milestones	Status
September 30, 2019	**M1:** Statistical analysis of alternative precursor conversion, baseline reference for validation of simulation predictions, and exploratory investigation and mechanical testing of low-cost alternative fibers.	**Met**
September 30, 2019	**M2:** Chemical conversion of alternative carbon fibers and verification through direct comparison with M1. Milestone will be met if simulations identify resultant properties within 15% error margin.	**Met**
September 30, 2019	**M3:** Large-scale simulations of fiber mechanics to predict resultant properties with verification through comparison with M4. Milestone will be met if simulations identify properties within 15% error margin.	**Met**
September 30, 2019	**M4:** Synthesis and evaluation of prototype carbon fibers produced via alternative precursors. Characterization and mechanical testing of alternative fibers to track progress toward DOE targets.	**Met**
September 30, 2019	**FY 19 Go/No Go:** The ICME framework shall identify at least one potential alternative precursor yielding a carbon fiber that is projected to meet cost, strength, modulus, and strain requirements.	**Met**

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Project ended on March 31, 2021

Phase 3 Tasks & Milestones

- Each task had a corresponding milestone

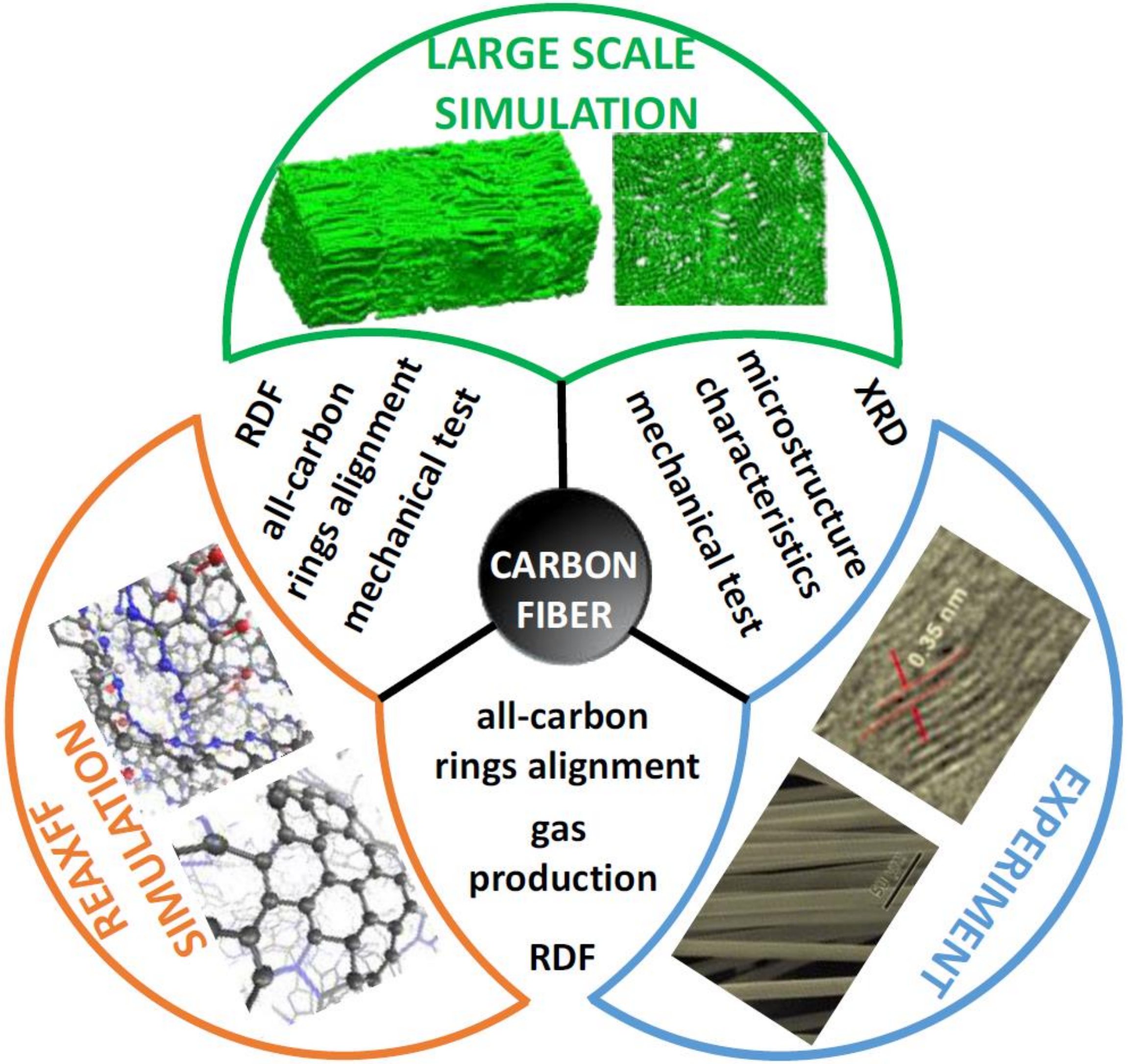
Date	Milestones	Status
March 31, 2021	**M1:** Scalability study of carbon fiber production via optimization of pilot-scale alternative precursor-derived carbon fibers	**Met**
March 31, 2021	**M2:** Experimental validation of carbon fiber properties to quantify the effect of fiber conversion parameters	**Met**
March 31, 2021	**M3:** Strategic framework for industrial production by polishing the ICME framework with ReaxFF simulations of the pretreatment, with targeted design of future precursors for fast conversion, and with large scale MD and continuum modelling of structure/property relationships	**Met**
March 31, 2021	**FY21 Go/No-Go:** Validation of ICME framework to predict the properties of carbon fiber produced via pilot production based on realistic fiber microstructure (porosity, diameter, core/shell, *etc*.)	**Met**

- Project ended on March 31, 2021 with 6-month no-cost extension

Project ended on March 31, 2021

Approach

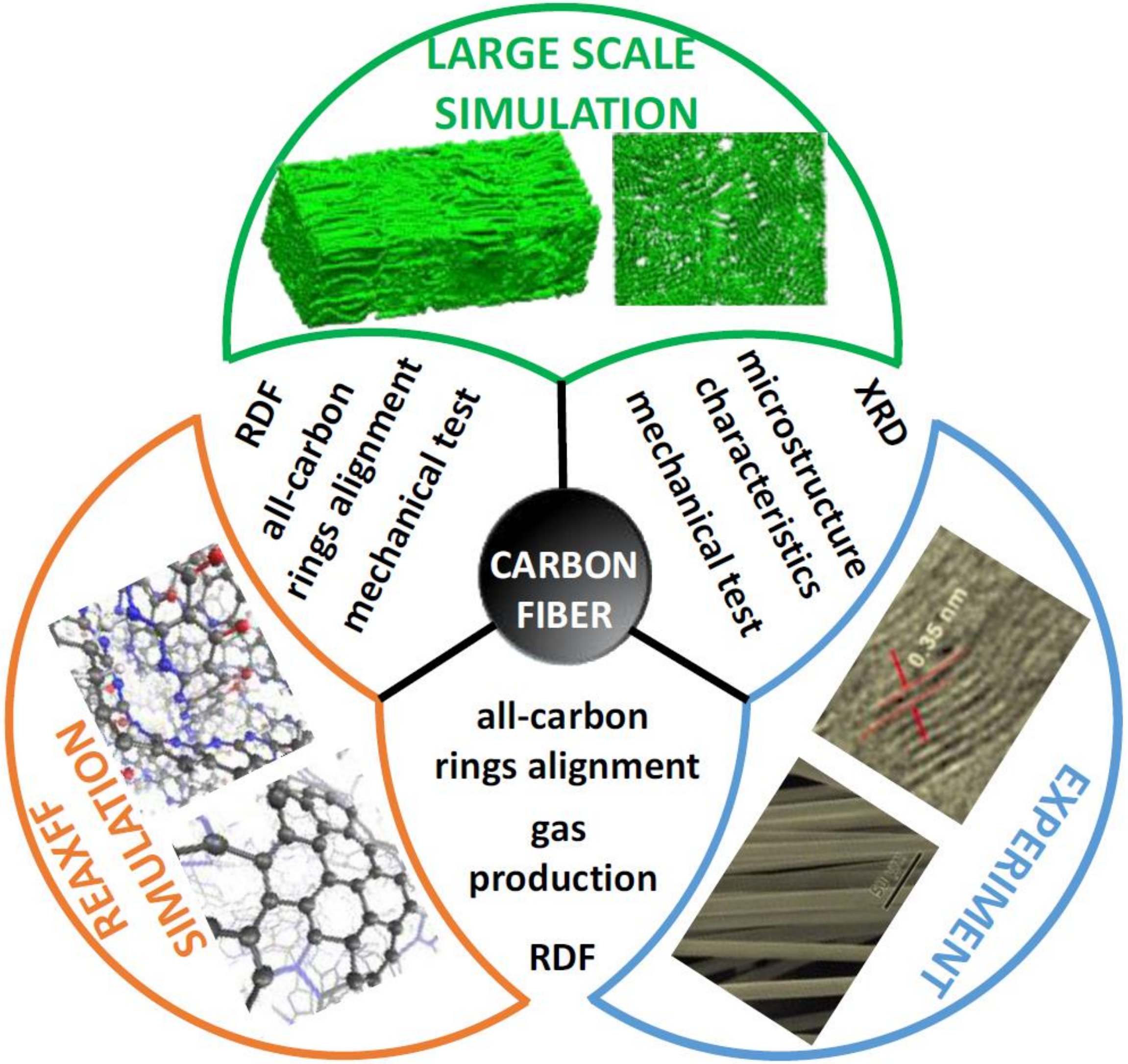
- **Closed loop validation** of modeling framework with experimentation
  - **ReaxFF simulations** probe the effect of conversion parameters on resultant **fiber chemical structures**
  - Resulting structure provides input to MD simulations of fiber mechanics
  - **MD and continuum FE simulations** translate chemical structure to realistic fiber microstructure and properties, validated against experimental results



Project ended on March 31, 2021

Approach

- **Experimentation** will provide input conditions and validation for modeling
  - In turn, the models help guide the experiments to explore new precursors and conversion parameters
- **Conduct pilot-production** to evaluate feasibility of scaling fiber production up from lab-scale results
  - Lab conversion procedures need to be optimized at different production scales



Project ended on March 31, 2021

Task 1: Scalability Study of Alternative Precursor CF - MET

- Task 1 aimed to determine the effects of production scale conversion and to optimize conversion at larger scales
- New larger-scale production tools were developed, including a continuous bath treatment line
- By using an initial alternative heat treatment, we reduced conversion time yet maintain our target properties
(the cost of conversion based on the CF cost breakdown approach presented by Felix Paulauskas (ORNL))

Metrics	DOE Target	Alternative Precursor* (Short conversion 1)	Alternative Precursor* (Short conversion 2)
Strength (GPa)	1.73	2.49	1.78
Modulus (GPa)	173	173	169
Strain (%)	1	1.44	1.05
Cost ($/lb)	5	4.1	3.58

*These properties reflect pilot-scale precursor with lab-scale conversion optimization.
*Reported by F. Paulauskas at VTO AMR (6/3/2020) and supported by literature: CD Warren/ORNL; Gill et al., (2016); Nunna et al., (2019); Choi et al., (2019)

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Project ended on March 31, 2021

Task 2: Experimental Validation - MET

- In Task 2, the simulations were refined to continue to support the experimental characterization and optimization of CFs
- As presented in Task 1, alternative CFs have been demonstrated to achieve all target properties, thus satisfying this milestone
- ORNL characterized fiber microstructure and correlated structure with processing parameters
- We investigated the fiber microstructure-property relationships via experimental and modeling methods
- We also developed a continuum model to quantify the effect of pore size across different scales

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Task 3: Strategic Frame for CF Production - MET

- We have integrated CF conversion insights gained from simulations and experiments into a testbed system to facilitate rapid technology transfer
- We have developed/performed a machine learning ICME testbed, which can predict optimized conversion parameters
- In tandem with the completion of the machine learning ICME testbed, we have performed a parametric study of alternative precursor CF recipes to test the testbed accuracy
- Tensile properties from up to 20 different alternative precursor CF samples with randomly chosen processing variables were used for training and accuracy testing

Project ended on March 31, 2021

Success Criteria: 15% ICME Accuracy - MET

- Our Phase 3 success criteria is to demonstrate the ICME framework can predict fiber properties with better than 15% accuracy
- The machine learning alternative precursor CF testbed parametric study has demonstrated accuracy of this testbed within 15% accuracy
- We have demonstrated the continuum FE simulations can accurately predict CF properties

Project ended on March 31, 2021

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Responses to Previous Year Comments

- Previous year comments are all very positive! Below are two minor comments.
- The reviewer expressed interest in seeing a table of how often there was contact, meetings, or reviews between and among the five entities.
  - Below is the table showing the communications between and among the five entities.

	UVA	PSU	ORNL	Solvay	Oshkosh
Ad Hoc Contact	Daily-Weekly	Daily-Weekly	Weekly--Monthly	Monthly	Monthly
Formal Meetings	Monthly	Monthly	Monthly	Monthly	Monthly
Reviews	Monthly, Quarterly, Annual	Monthly, Quarterly, Annual	Monthly, Quarterly, Annual	Monthly, Quarterly, Annual	Monthly, Quarterly, Annual

- The reviewer would also like to see further development of ICME tools, especially the integration of more detailed process analysis into optimization when considering scalability. Continuum scale simulations to reveal the effect of multiscale defects on CF properties.
  - We established/performed a machine learning ICME testbed which can predict optimized conversion parameters. We also developed a continuum model to quantify the effect of pore size across different scales.

Project ended on March 31, 2021

Collaboration

- **University of Virginia**, PI Li & Co-PI Zhigilei
  - Experimental and statistical analysis of carbon fiber conversion supported by MD and continuum FE simulations
- **Pennsylvania State University**, Co-PI van Duin – Subcontractor
  - Simulations of chemical conversion of precursors into carbon fiber
- **Oak Ridge National Laboratory**, Co-PIs Klett & Vautard – Subcontractor
  - Experimental testing and characterization of alternative precursors
- **Solvay S.A.**, Co-PI Des Cook – Subcontractor
  - Industry guidance on fiber characterization and operation of pilot production run of carbon fiber from alternative precursors
- **Oshkosh Corporation**, Co-PI Robert Hathaway – Subcontractor
  - Industry insight on constraints and priorities for technology transfer from research laboratories to industrial production

Project ended on March 31, 2021

Summary

- We have **established an ICME framework** to predict fiber properties
  - **ReaxFF simulations** target the fundament chemical evolution from precursor to CF
  - **Atomistic MD simulations** translate molecular structure to fiber microstructures, and **continuum models** translate mechanical properties to relevant CF length scales
  - **Experimental testing** validates these conversion mechanisms and unveils new pathways (alternative treatments) to low-cost CF conversion
- We have demonstrated low-cost, high-performance CF from alternative precursors
  - **One alternative precursor** has been shown to convert to CF meeting all target metrics

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Project ended on March 31, 2021

Technical Backup Slides

Project ended on March 31, 2021

Technical Backup Slides – Alternative Treatments

- Alternative conversion and optimal conversion parameters were explored
  - TGA analysis shows that plasma conversion is promising for producing CF from alternative precursor
- DSC (performed by Solvay) of alternative precursor fibers shows promising
  - This result provided optimal conversion parameters for this material
- Using experiments and ReaxFF simulations, we investigated alternative bath treatments; a promising treatment was determined to be the best stabilizing alternative precursor conversion

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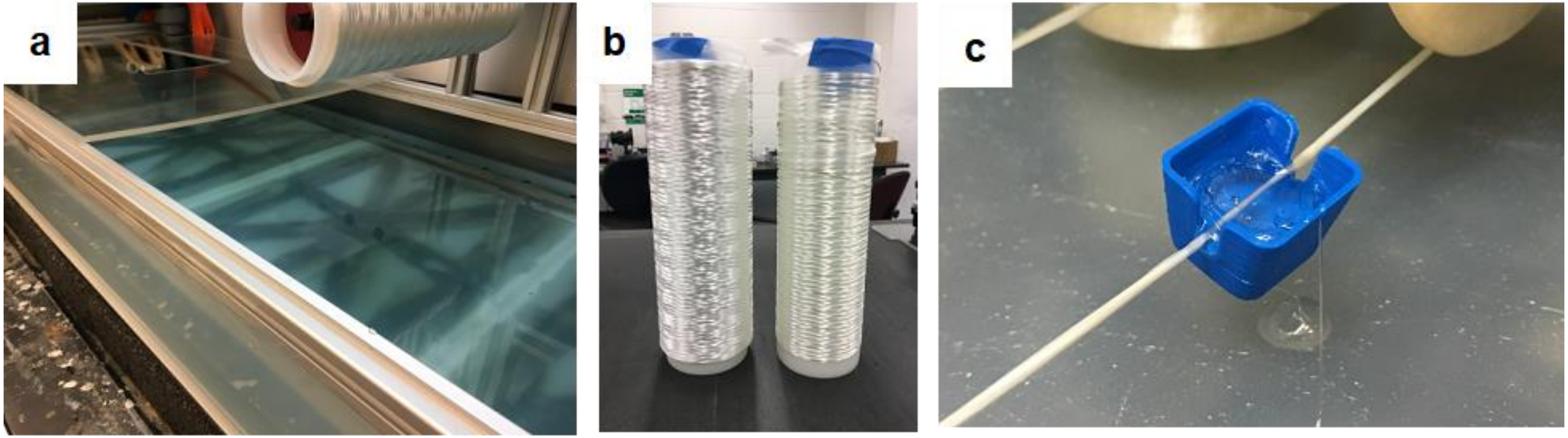
Technical Backup Slides – Continuous Treatment Line

A continuous bath line was developed for scale-up treatments



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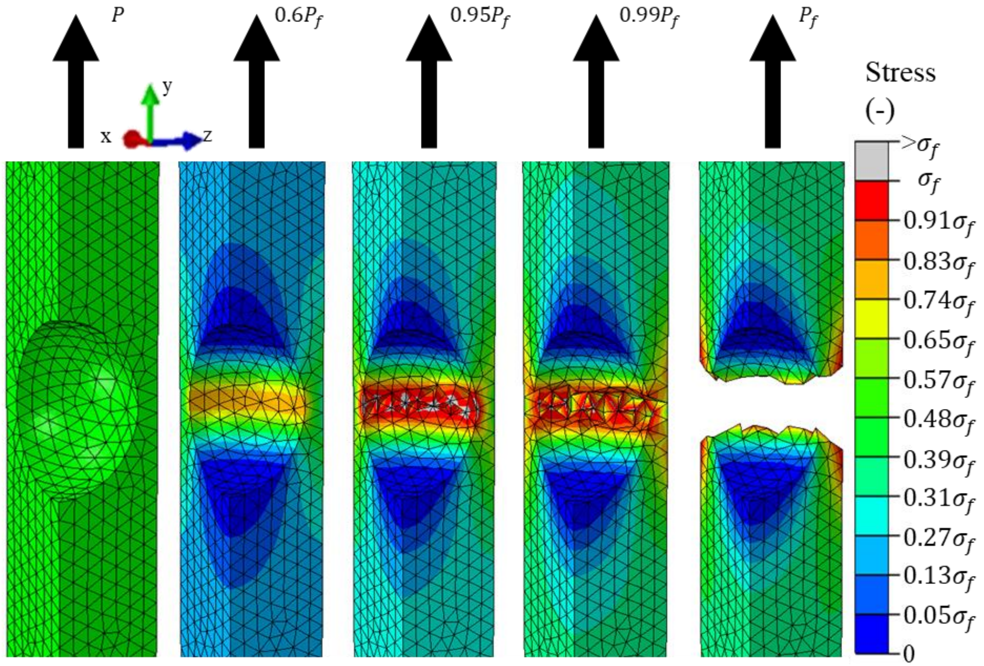
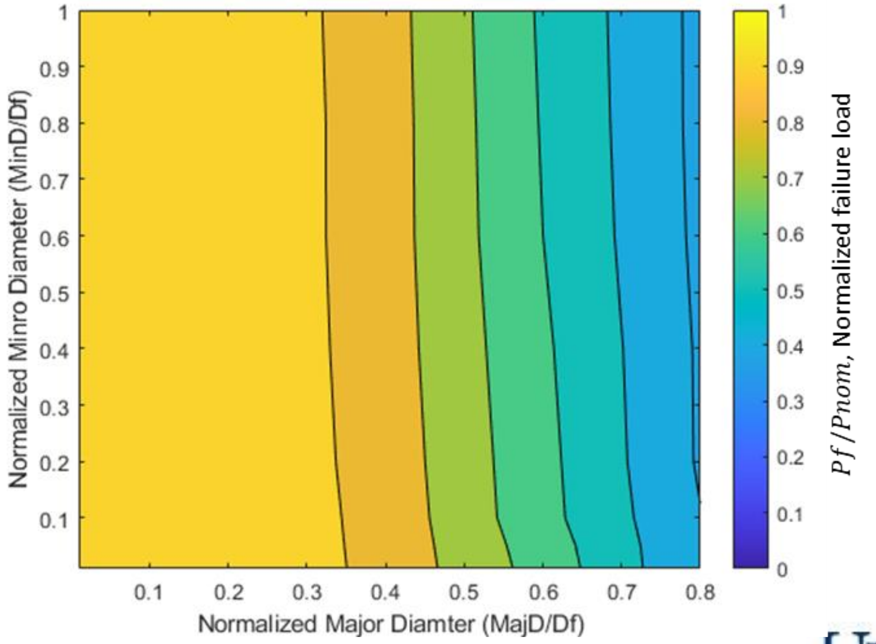
Technical Backup Slides – Continuum Finite Element Model

- A continuum finite element model was developed to explore the effect of defects on the stiffness and failure strength of the fiber
- The finite element model predicted the material behavior using a dynamic explicit analysis in conjunction with an element deletion technique





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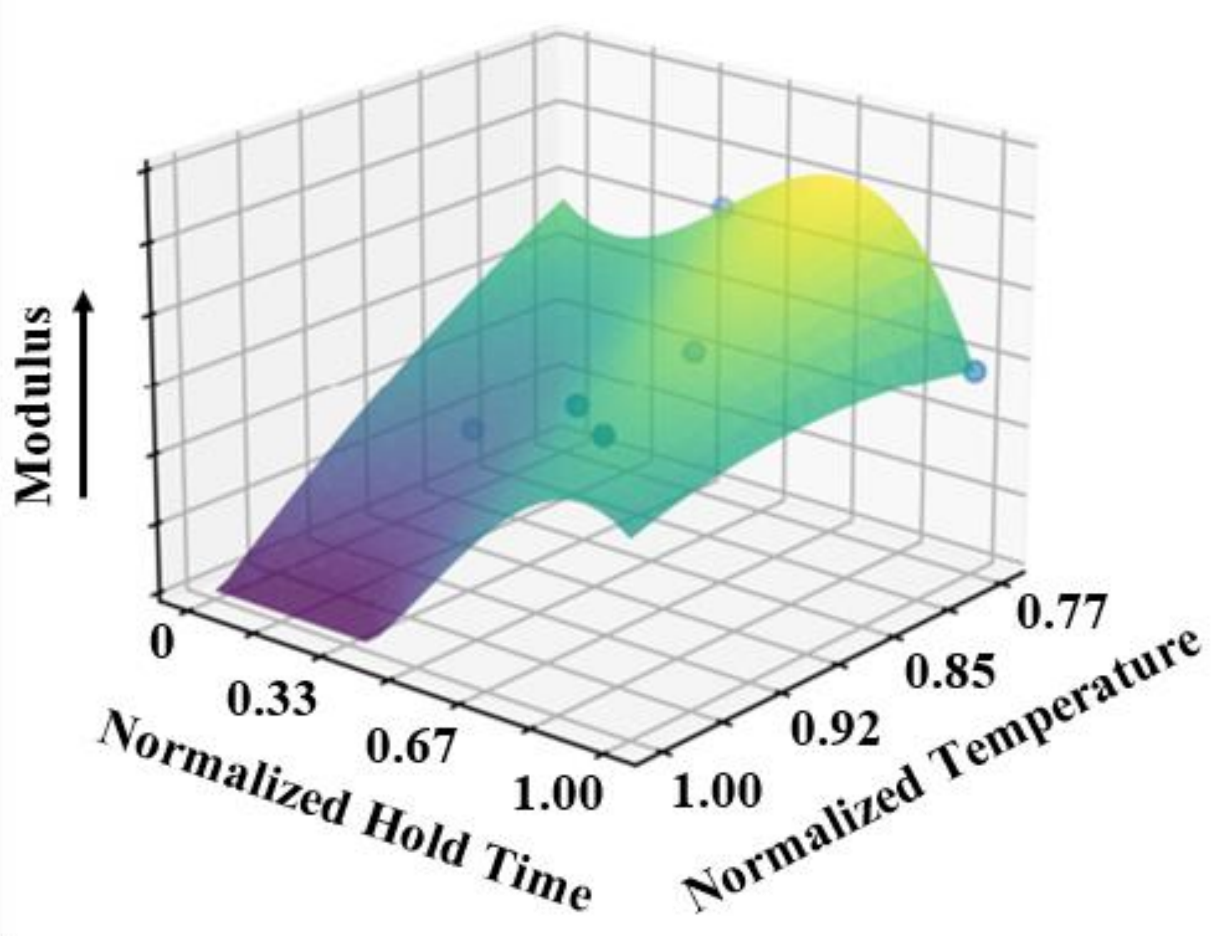
Technical Backup Slides – Machine Learning Algorithm

The machine learning algorithm is capable of predicting the properties (shown here for a modulus prediction) based on conversion processing parameters



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