Center for Composite Materials

Closed Loop Recycling of Composite Waste Streams into High Performance Aligned Short Fiber Composites

WP20-1483 Project Outbrief

Dirk Heider | July 31 2023

University of Delaware

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14. ABSTRACT
The technical objectives of this project are listed below:
- Global market for carbon fiber reinforced plastics (CFRP) was less than 20,000 tons annually in 2020 and grew to 140,000 tons in the early 2020s.
- Reducing CF waste with its high embodied energy through recycling is energy-efficient.
- Our approach demonstrates closed loop recycling of DoD relevant composite materials, scrap reduction and fabrication of complex part geometries with the TuFF technology.
- This reduces waste and lifecycle costs associated with the manufacturing of DoD systems.

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composites, corrosion, emissions and waste reduction, manufacturing, Closed Loop Recycling

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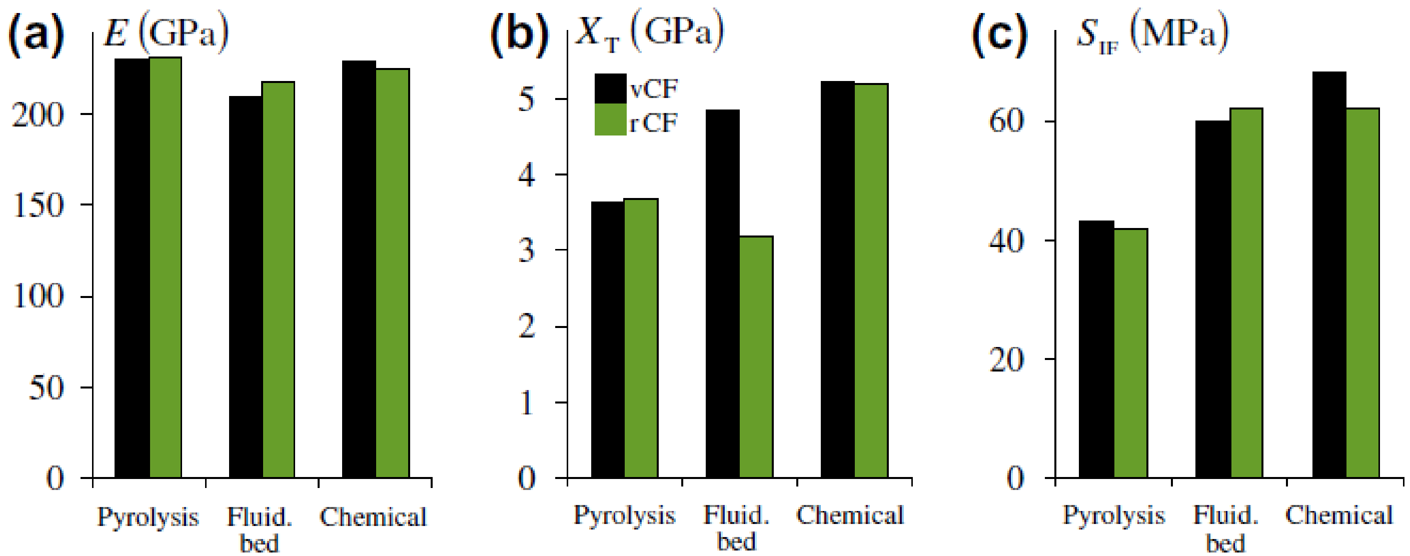
Background & Motivation

- Global market for carbon fiber reinforced plastics (CFRP) was less than 20,000 tons annually in 2020 and grew to 140,000 tons in the early 2020s.
- Reducing CF waste with its high embodied energy through recycling is energy-efficient.
- Our approach demonstrates closed loop recycling of DoD relevant composite materials, scrap reduction and fabrication of complex part geometries with the TuFF technology.
- This reduces waste and lifecycle costs associated with the manufacturing of DoD systems.

Recycled Fiber Property Retention Has Been Demonstrated in Literature

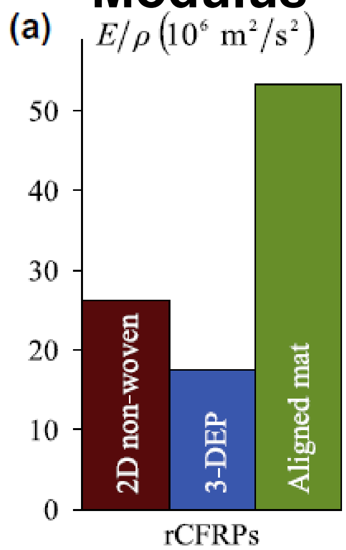
- In general fiber (not recycled CFRP) properties after recycling show no or small amount of degradation
  a) Modulus
  b) Strength
  c) Interfacial Shear Strength with Epoxy resin



S. Pimenta, S.T. Pinho," Recycling carbon fibre reinforced polymers for structural applications: Technology review and market outlook"< Waste Management 31 (2011) 378–392

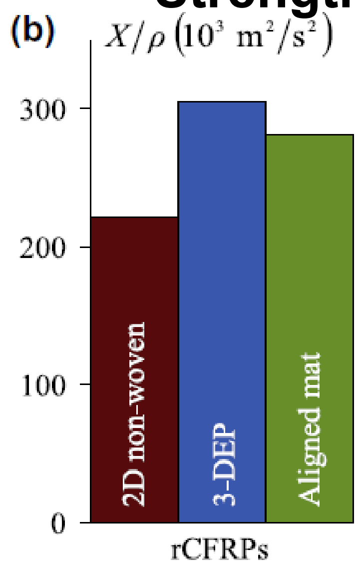
Recycling Challenge is Creation of High-Performance Composite Parts

Modulus



$E_{recycled}$=25-80 Gpa
$E_{virgin,\ IM7\text{-}8552\ @57\%}$=150 GPa

Strength



$X_{recycled}$=300-420 MPa
$X_{virgin,\ IM7\text{-}8552\ @57\%}$=2600 MPa

Recycled Data from S. Pimenta, S.T. Pinho," Recycling carbon fibre reinforced polymers for structural applications: Technology review and market outlook" Waste Management 31 (2011) 378–392

A high-performance recycled part exhibits following properties:

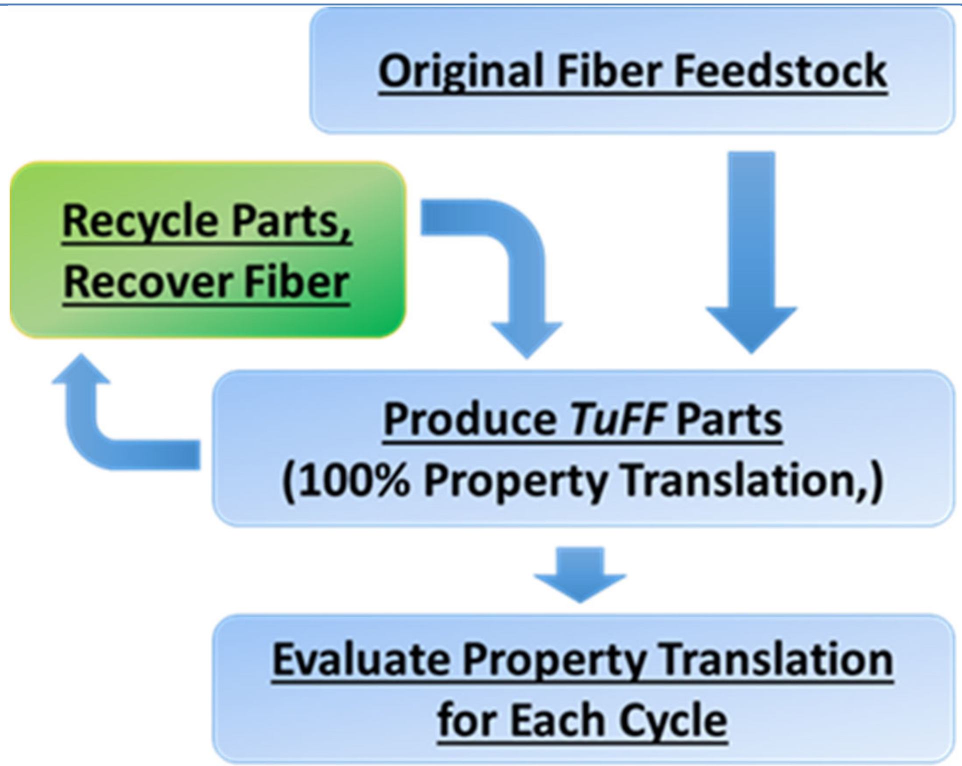
- **Good fiber strength retention after recycling**
- **Good interfacial shear strength of recycled fiber to polymer**
- **High fiber volume fraction part**
  - **High degree of fiber alignment**
- **Control of fiber aspect ratio of >600 (length over diameter)**

➔ Historical data shows better stiffness than strength translation

TUFF Approach to Closed-Loop Recycling

1. *TuFF* is utilizing short fibers as original feedstock into process
2. A part can be produced with conventional processing approaches
3. The *TuFF* part is pyrolyzed and the fiber content is recovered
4. Fiber surface may have to be treated to ensure good bonding of polymer to fiber
5. The short fiber are reinserted into *TuFF* process and the process can be restarted

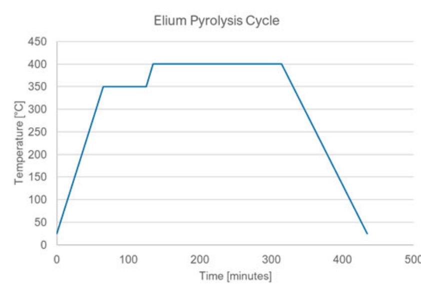
➔ Composite properties are evaluated after each recycling step and compared to original panel performance



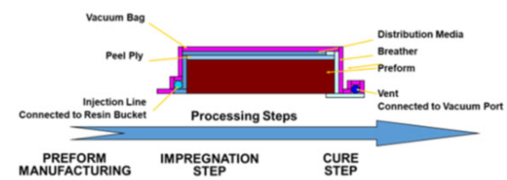
Process Flow for SERDP Project

1. Fibers are recovered from aerospace-grade prepreg (IM7 fiber with 977-3 resin) using pyrolysis
2. The TuFF process is used to align fibers
3. The aligned sheet material is infused with 977-3 resin film
4. The composite part tension properties were tested
5. Cycle can be repeated if desired

TuFF Process



Pyrolysis



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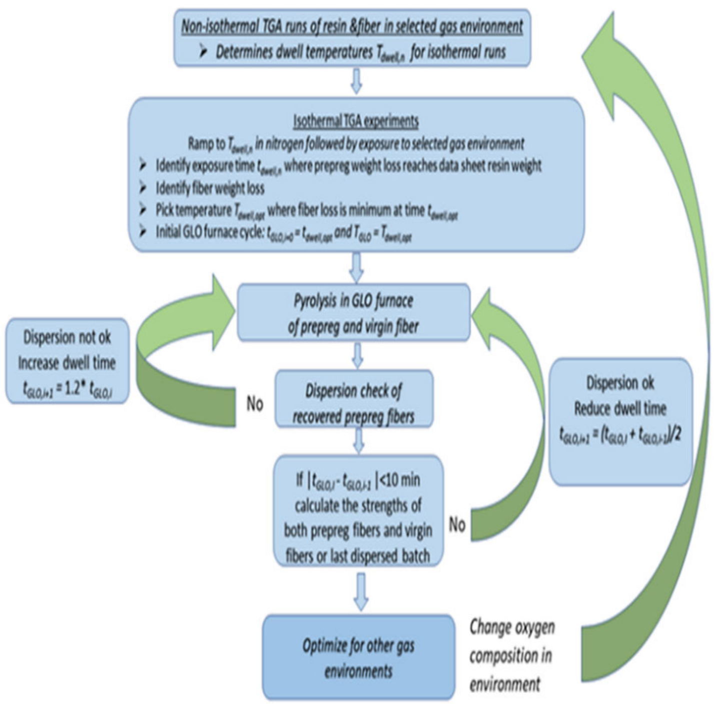
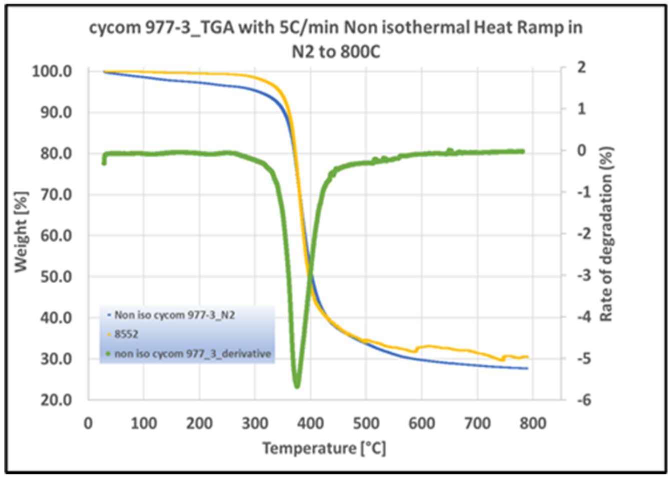
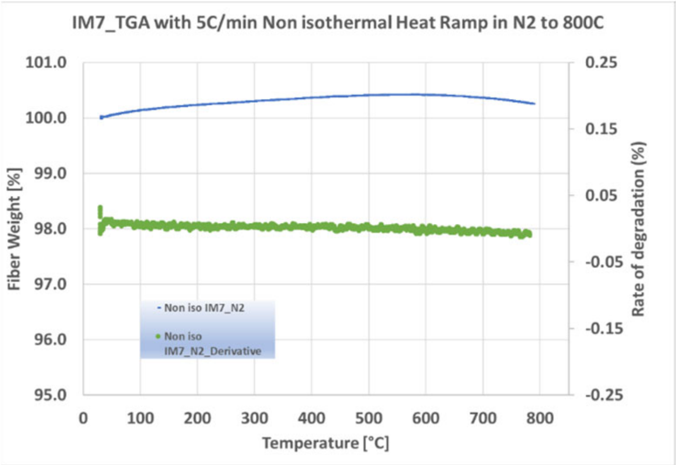
Optimization of Pyrolysis Cycle



- Utilizes TGA data to understand resin and fiber degradation process
- Goal is to obtain fiber content only with minimum reduction of fiber properties
- 977-3 epoxy resin degrades at lower temperatures compared to carbon fibers

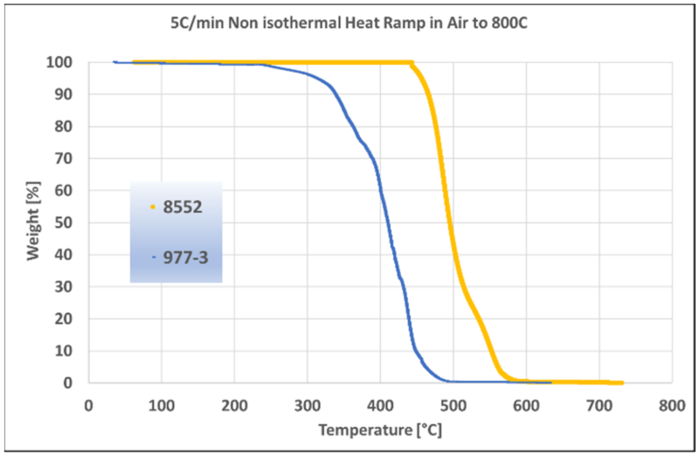
TGA Results under Inert Gas Environment

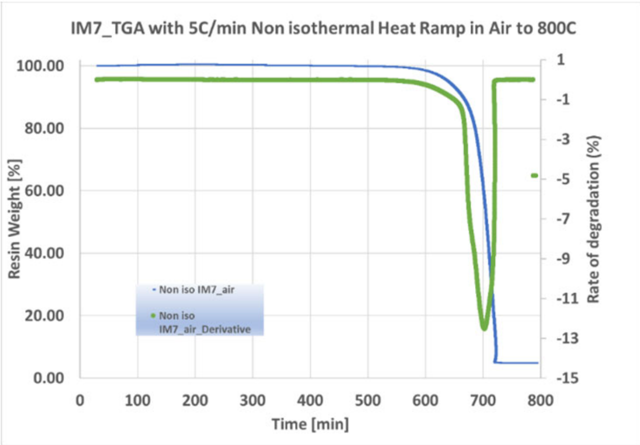




- Under nitrogen environment, the carbon fiber does not degrade until 600C
- The epoxy resin material shows ~70% weight loss at 450C without any further weight loss at higher temperatures

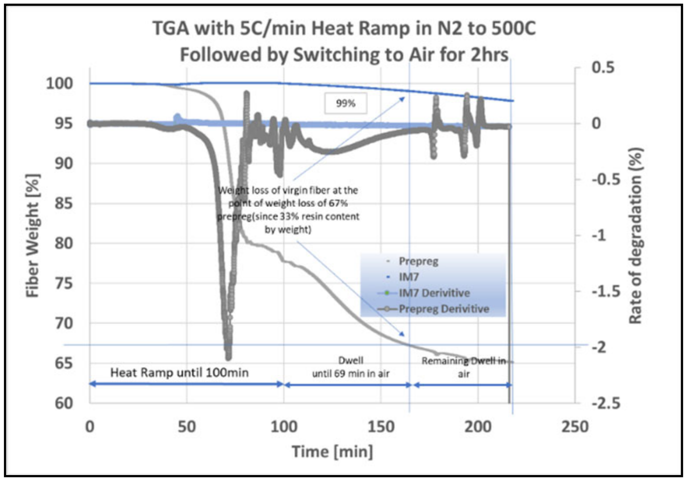
TGA Results under Oxygen Environment

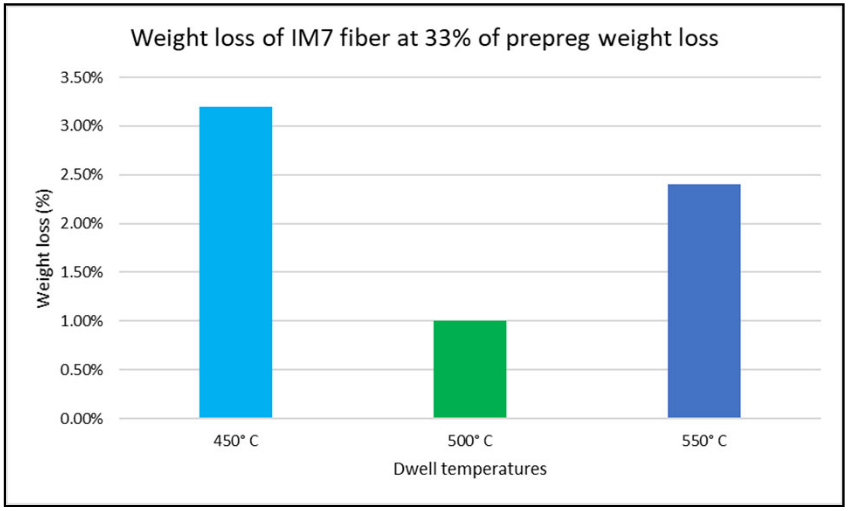




- Under air, weight loss is significantly higher for both materials
- IM7 degradation starts above 500C with peak loss at ~700C, while the neat resin weight loss shows start of degradation at 300C

TGA Result of Optimized Pyrolysis Process





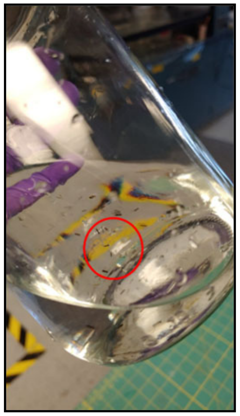
- Optimized heat ramp is in nitrogen and then switched to air during dwell. This minimizes fiber degradation while some resin loss is expected.
- The prepreg material has nominal 33.5% resin by weight. At 500C, 33.5% weight loss is achieved ~70min into the dwell cycle.
- Increasing dwell temperature while maintaining resin loss goal of 33.5% increases degradation of carbon fibers. Minimum weight loss of approximately 1% at 500C.

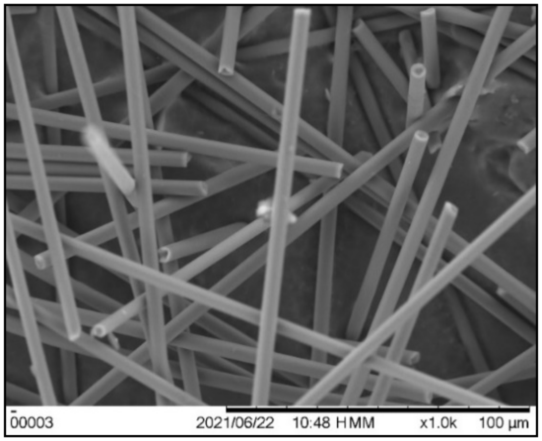
Pyrolysis of Composite Samples

$$Q_{GLO} = \frac{A_{GLO}}{A_{TGA}} * Q_{TGA} = \frac{706.5\ cm^2}{5.06\ cm^2} * 120\ {}^{ml}/_{min} = 1000\ {}^{l}/_{hr}$$

- A GLO 40/11-2G semi-automatic furnace allows control of the gas environment and heating cycle.
- A sample holder allowed material stacking while keeping material physically apart.
- Flow rate was calculated using chamber size and TGA sample holder volume.

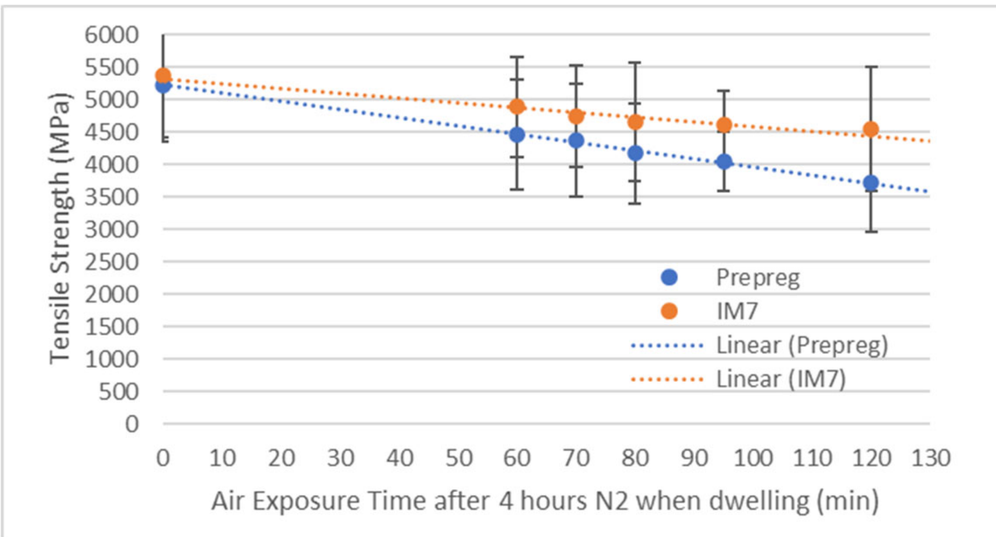
Dispersion of Recycled Fiber Material





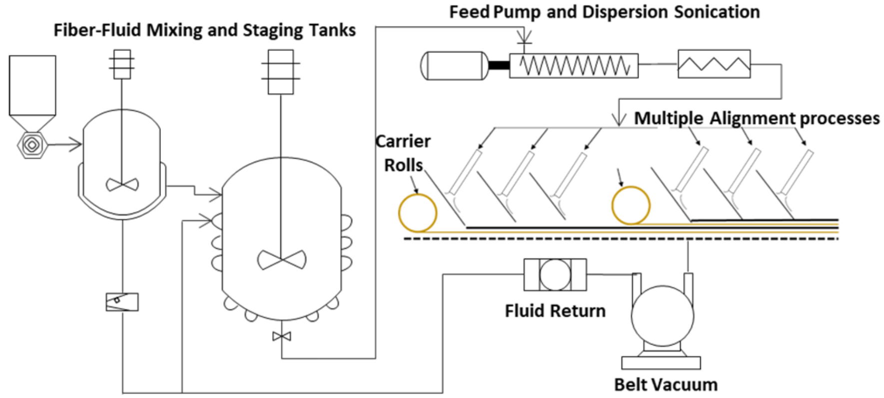
- Filament level dispersion after pyrolysis is needed for TuFF alignment process
  - Visual observation after mixing and SEM are used to evaluate fiber separation
- TGA optimized cycle is starting point for dispersion optimization
  - Dwell of 95 min allowed good dispersion of fibers

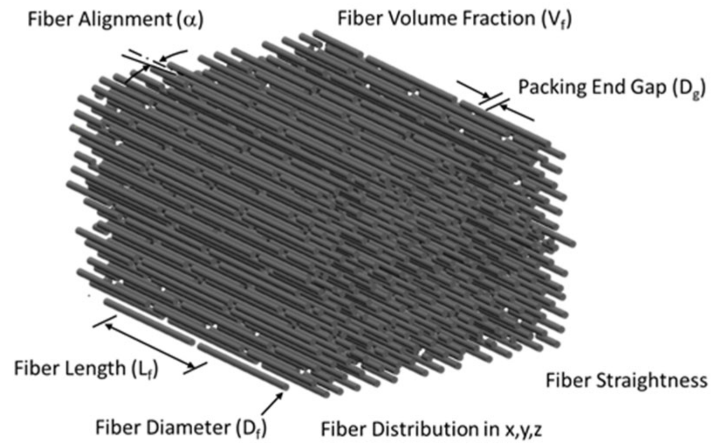
Strength Evaluation as a Function of Dwell Cyle



- Long fibers recovered from the prepreg and are tested in Dia-stron (Single Fiber Tensile Test) machine to evaluate fiber strength.
- Virgin fiber strength (5450 MPa ± 815 MPa) was within one standard deviation compared to the Hexcel datasheet numbers of IM7 at 5516 MPa..
- The fibers didn't degrade after the nitrogen step but air exposure reduced strength significantly.
- At the air exposure of 95 min, fiber strength of the recovered virgin and prepreg fiber material has been reduced to 4608 MPa ± 531 MPa and 4055 MPa ± 452 MPa, respectively.
  - This is a 16% and a 26% reduction in fiber strength compared to the virgin material.
- It is interesting to observe that the virgin material has higher strength numbers compared to the fiber material recovered from the prepreg. This might be due a loss in fiber strength during prepregging, , where the fiber goes through various material handling processes.

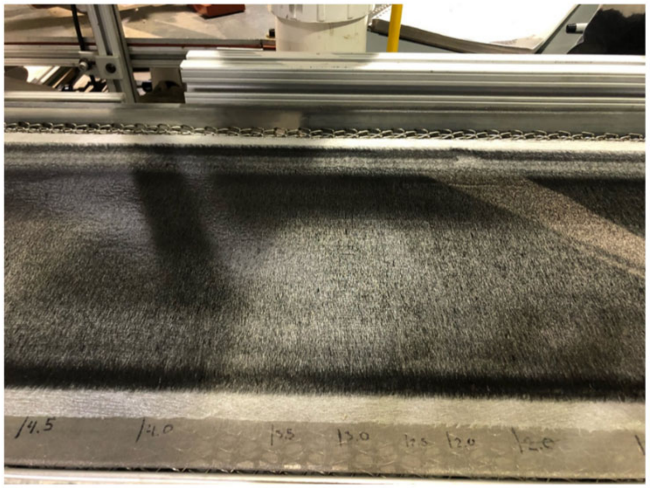
TuFF Alignment Process





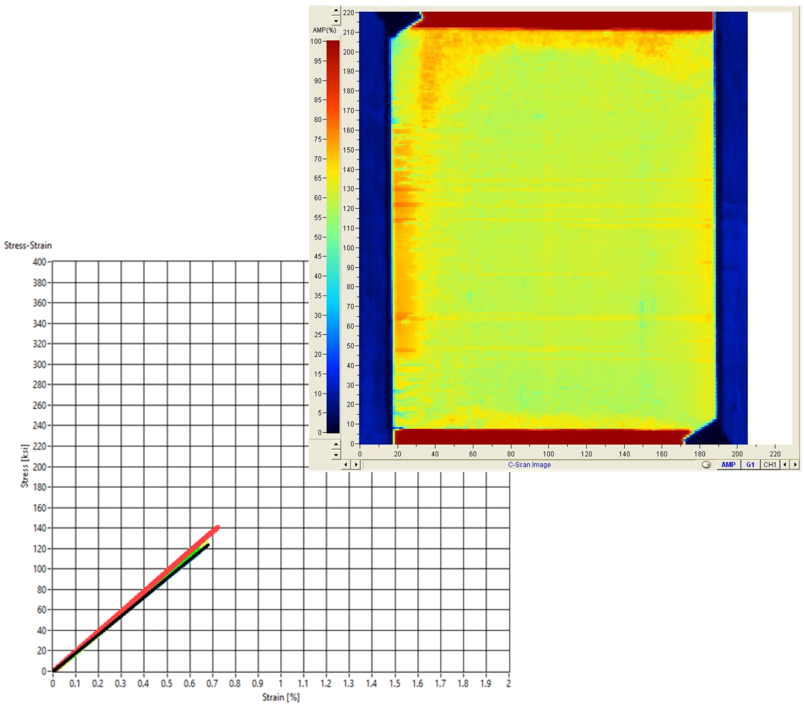
- Wetlaid process which disperses fibers in fluid, aligns fibers in patented headbox followed by water-fiber separation
- Filament level alignment control: > 95% (± 5◦) has been demonstrated with virgin fiber
- High fiber volume fraction >57%

TuFF Sheet Production



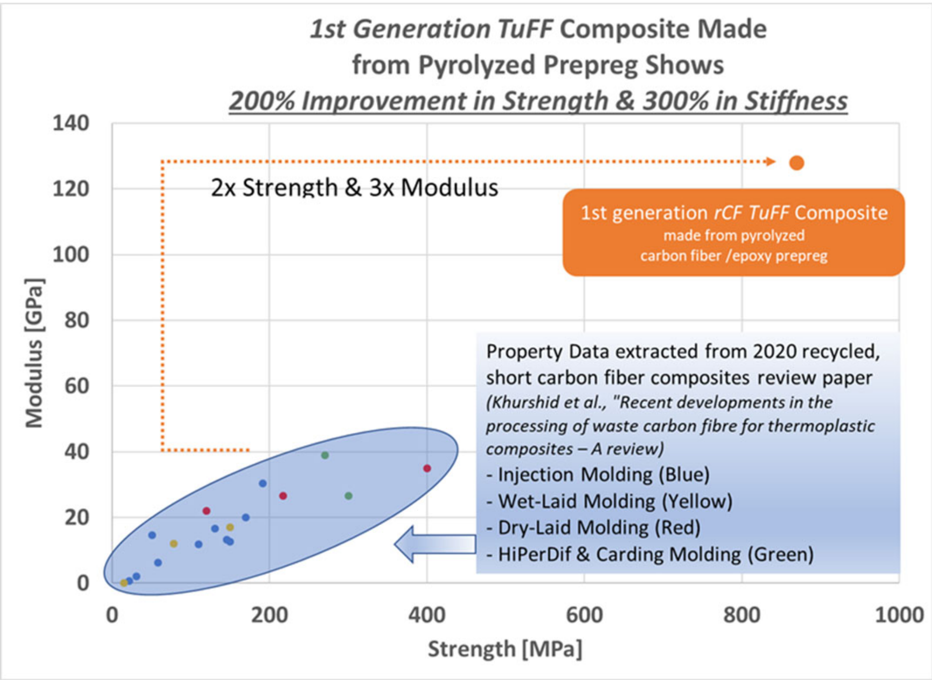
- 100 grams of short fiber prepreg material was pyrolyzed with the 95 minutes dwell cycle
- Fibers were processed into a panel using the TuFF process.
- Alignment quality of the produced were good indicating good dispersion of the fibers in the mix tank.
- Fiber length was also measured and showed that most of the fiber length are centered around the desired 3mm cut length.

QA and Mechanical Test Results



- TuFF sheets were stacked with resin film and autoclaved to produce flat coupon panel
- Ultrasonic C-scan showed uniform panel quality
- Tensile coupon were cut and tested
- A tensile strength of 869 MPa (126 ksi) ± 66 MPa (9.6 ksi) and tensile modulus of 128 GPa (18.6 msi) ± 4.84 GPa (0.702 msi) was measured.
- 100% stiffness translation compared to virgin continuous fiber prepreg material @ 45% FVF
- Strength translation is lower at 44.2% compared to the prepreg

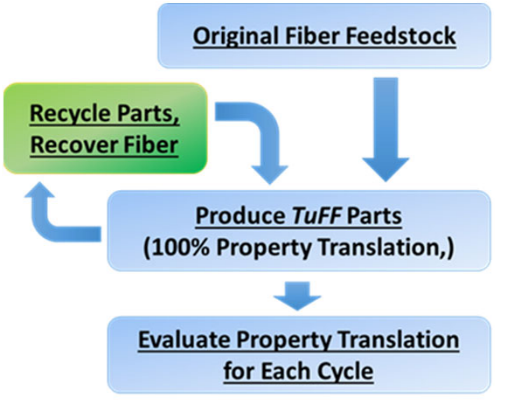
Summary and Accomplishments



- Composite properties of the recycled fiber material is higher than any other reported properties.
- Future work will evaluate different process conditions in the GLO oven to improve fiber strength while maintaining the ability to disperse fibers.
- We have not evaluated new surface treatments of the recovered fiber material

➔ Overall, a methodology has been developed to recover carbon fiber from uncured prepreg and reuse them in our *TuFF* process to demonstrate true recycling of high-performance composites.

WP20-1483: Closed Loop Recycling of Composite Waste Streams into High Performance Aligned Short Fiber Composites



Performers: University of Delaware

Technology Focus

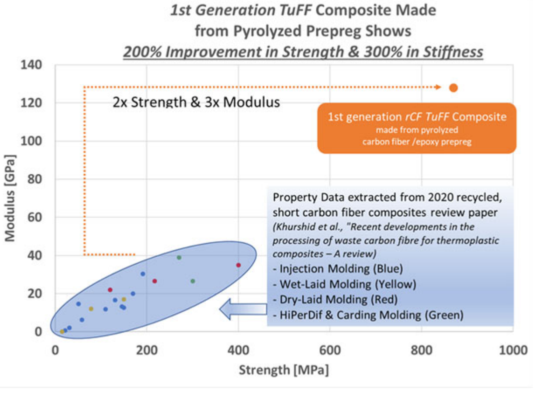
- *Reduce waste and lifecycle costs associated with the manufacturing of DoD composite material systems.*

Research Objectives

- *Demonstrate closed loop recycling of DoD relevant composite materials, scrap reduction and fabrication of complex part geometries with the TuFF technology.*

Project Progress and Results

- *A methodology has been developed to recover carbon fiber from uncured prepreg*
- *Reuse of rCFs with the TuFF process to create recycled composite parts with true recycling of high-performance composites has been demonstrated for the first time.*
- *Property translation exceeds any other reported short, recycled fiber composite by 2-3 times.*



Technology Transition

- *Targeting automotive, sporting good companies to replace virgin structures with our sustainable, low-cost material solution.*