Continuous-Fiber, Malleable Thermoset Composites with Sub-1-Minute Dwell Times: Validation of Impact Performance and Evaluation of the Efficacy of the Compression Forming Process

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Project ID: mat147

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Bo Song — Sandia National Laboratories

Michael Larche/ Leo Fifield — Pacific Northwest NATIONAL LABORATORY

Robert Norris — OAK RIDGE National Laboratory

Overview

Timeline

- Project Start: 9/2018
- Project End: 6/2021
- Percentage complete: 95%

Budget

- Total project funding: $ 1 M
  - DOE share: $ 500k
  - Contractor share: $ 500k

- FY 2018: $325k
- FY 2019: $575k
- FY 2020: $100k
- FY 2021: No Cost Extension

Barriers Addressed[1]

- Low cost, high volume manufacturing of carbon fiber composites, with cycle time < 3 minutes
- Non-destructive evaluation (NDE) of malleable thermoset composites- specifically acoustic approach to QA/QC
- Enhancing crash energy management

[1]*2017 U.S. DRIVE MTT Roadmap Report, sections 4 and 5*

Partners

- Mallinda (lead): Philip Taynton
- PNNL: Michael Larche/ Leo Fifield $225k 49% spent
- SNL: Bo Song $175k 96% spent
- ORNL: Robert Norris $100k 93% spent

Any proposed future work is subject to change based on funding levels

Relevance

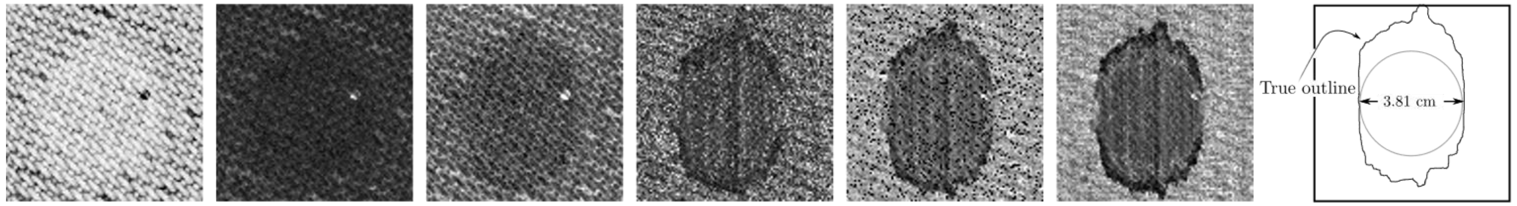
IMPACT	APPROACH
Under 1-minute dwell times	Precured malleable thermoset prepregs
Non-destructive evaluation	Acoustic microscopy towards QA/QC
Crash energy management	Material development informed by split Hopkinson high speed impact testing, & moderate strain rate tensile & flexural testing

OBJECTIVES

- Develop malleable thermoset resin/fiber combinations for improved crash energy management
- Understand relationship b/n compression forming conditions, acoustic response, and defects & voids in the composite
- Determine high speed split-Hopkinson impact characteristics of candidate composite materials
- Demonstrate feasible, >20% lightweighting of CEM structures vs. glass mat thermoplastic via moderate strain-rate testing

Approach - Acoustic analysis

Technique: Joint Entropy Analysis of C-Scan Data



Comparison of imaging techniques for a difficult to detect resin-rich region. Peak-to-peak and $log[E_f]$ (log sum of squares) images are shown in the first two panels. They show little evidence of the resin rich region and have very different appearance from all of the entropy images: H_f (differential entropy), $I_{f,\infty}$ (Renyi entropy) and Joint entropies $H_{f,g}$ and $H_{f,gO}$.

Optimize composite stamping process & establish Acoustic response

- ☑ Study the impact of temperature, time and pressure on interlaminar shear strength in consolidation of pre-cured vitrimer composite plies.
- ☑ Validate compression forming results in 3D part consolidation
- ☑ Use selected range of sub-1-minute stamping conditions for further study of ultrasonic response
  - Investigate correlation of bond strength with C-scan results by mapping samples, conducting short beam shear analysis in areas of interest
  - Microscopic crosscut analysis to characterize defects

- **Reporting on Ultrasonic analysis**
  - Report analysis of material signatures and defects
  - Discuss limitations, potential and appropriateness of techniques
  - Report recommendations for technique refinement and future work

References:

[1] Larche, et. Al. Proceedings Volume 10169, Nondestructive Characterization and Monitoring of Advanced Materials, Aerospace, and Civil Infrastructure; 101690T (2017).

[2] Luzi, et. Al. Journal of the Acoustical Society of America **148**(1) (2020), pp. 292-301.

[3] Hughes, et. Al. (L.J. Bond, and D.E. Chimenti Eds.), Vol. 42, A.I.P. Press, New York, 2015, pp 1168-1174.

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Approach - Impact testing

Impact & strain rate response

1. Interaction with OEM partner to guide targets & provide design & modeling baseline

    - redesign component for simplicity of production, lightweight, and simulated crash performance
    - produce tooling, validate compression forming process in a full-scale component mold.

2. Strain rate studies using Split Hopkinson Bar to understand material response vs. quasi-static.

    - re-run part-specific crash simulations using strain rate response data.

3. Scaled-up testing of Flexural & tensile test samples across strain rates.

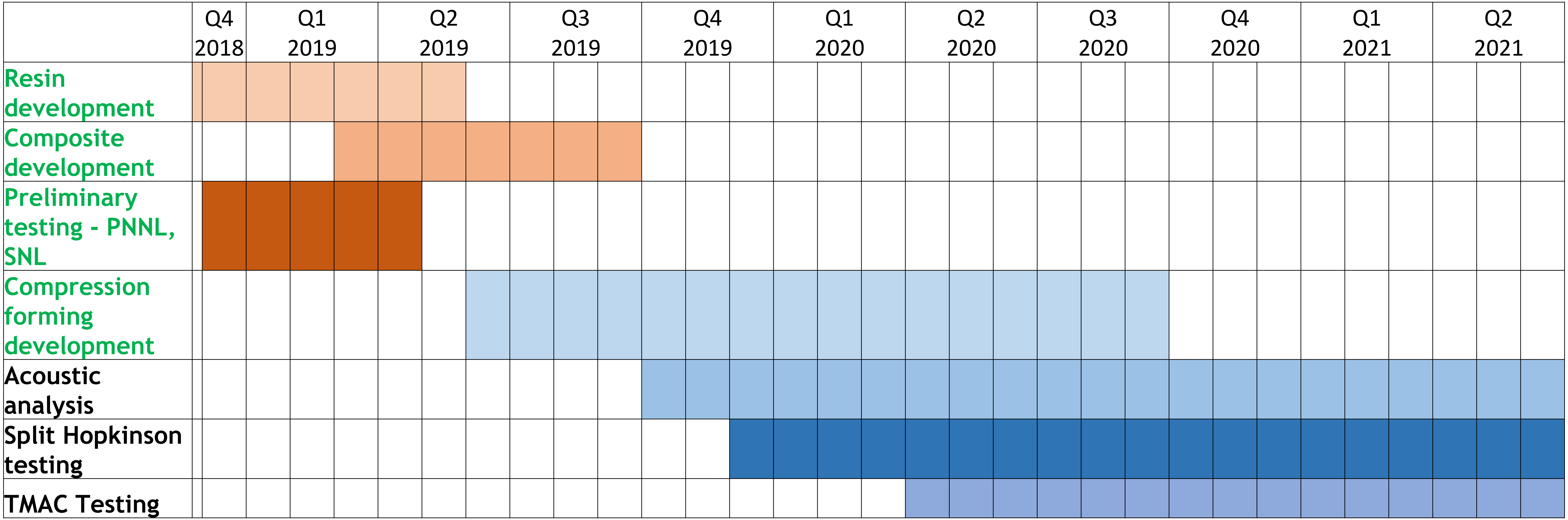
    - re-run part-specific crash simulations using strain rate response data.

TMAC Machine

Sandia National Laboratories

Oak Ridge National Laboratory

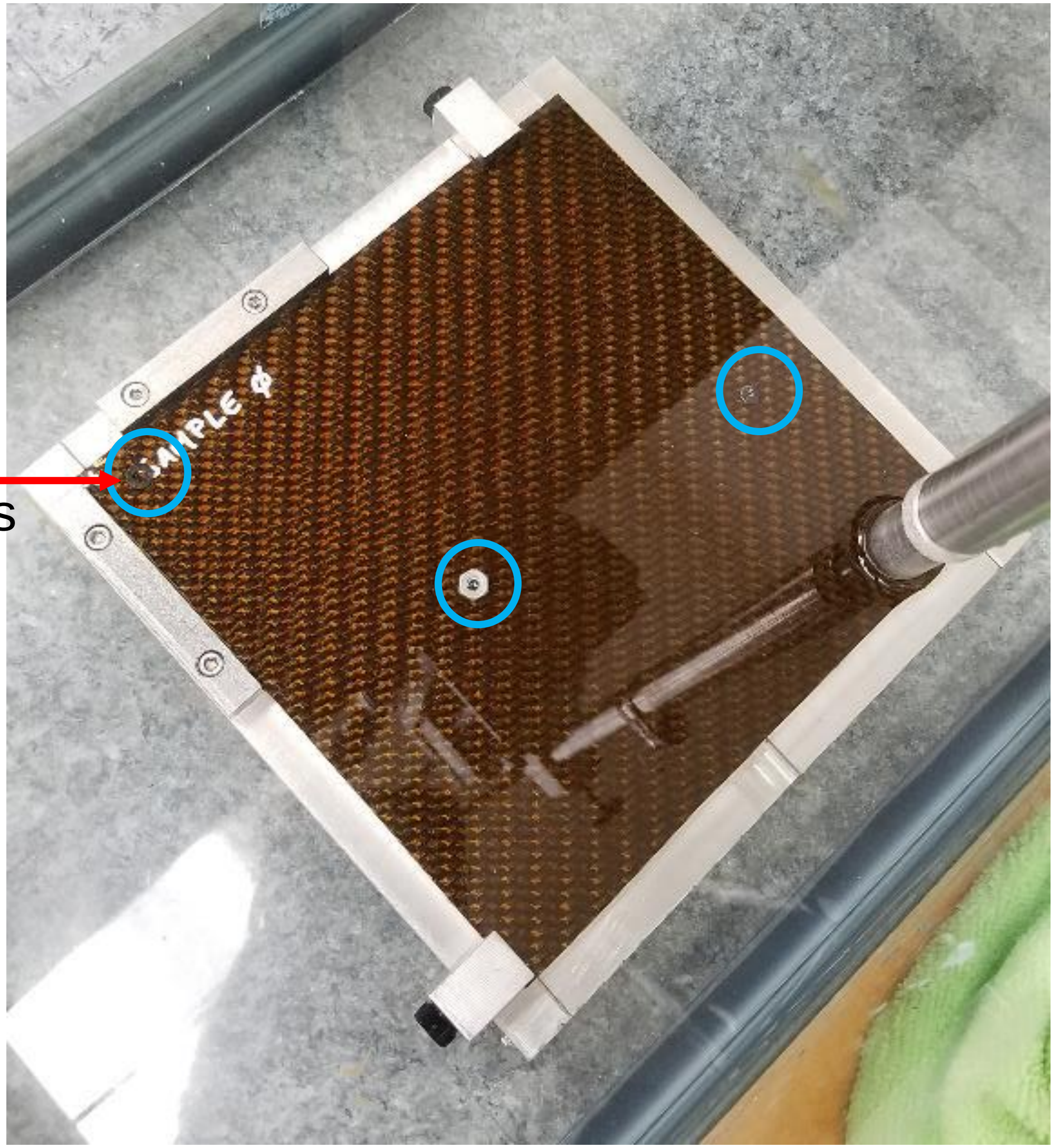
Approach - Milestones



Exam Setup

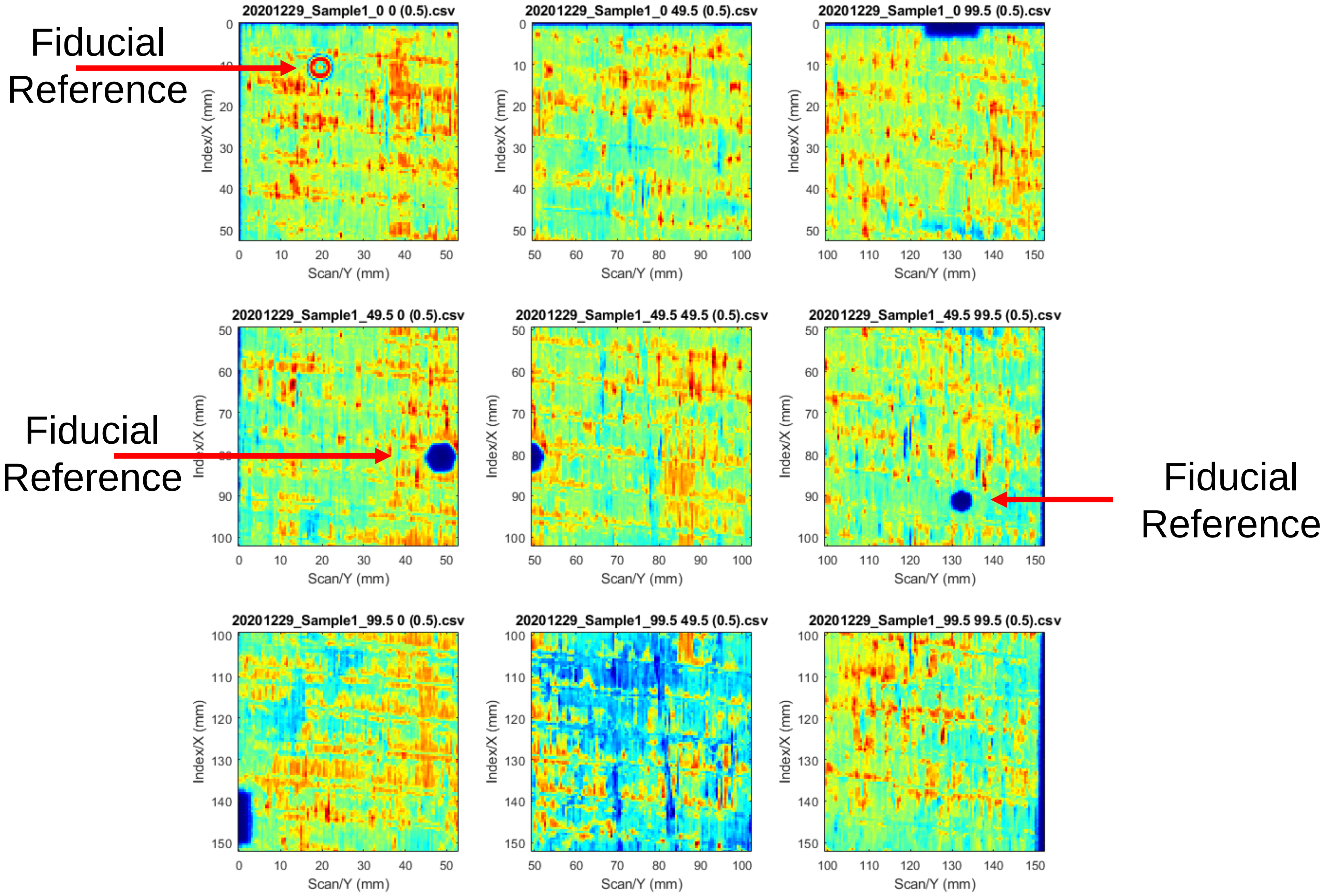
- Plaques were ultrasonically examined in immersion tank in 9 sections, results were stitched into a mosaic
- Overlapping 53x53 mm area sections were examined in 0.5 mm steps using a 10 MHz, 0.5” diameter, 1” focused probe
- Samples were soaked in water for 24 hours prior to testing. Samples were soaked for an additional 7 hours during testing, removed promptly after testing and dried

Fiducial References



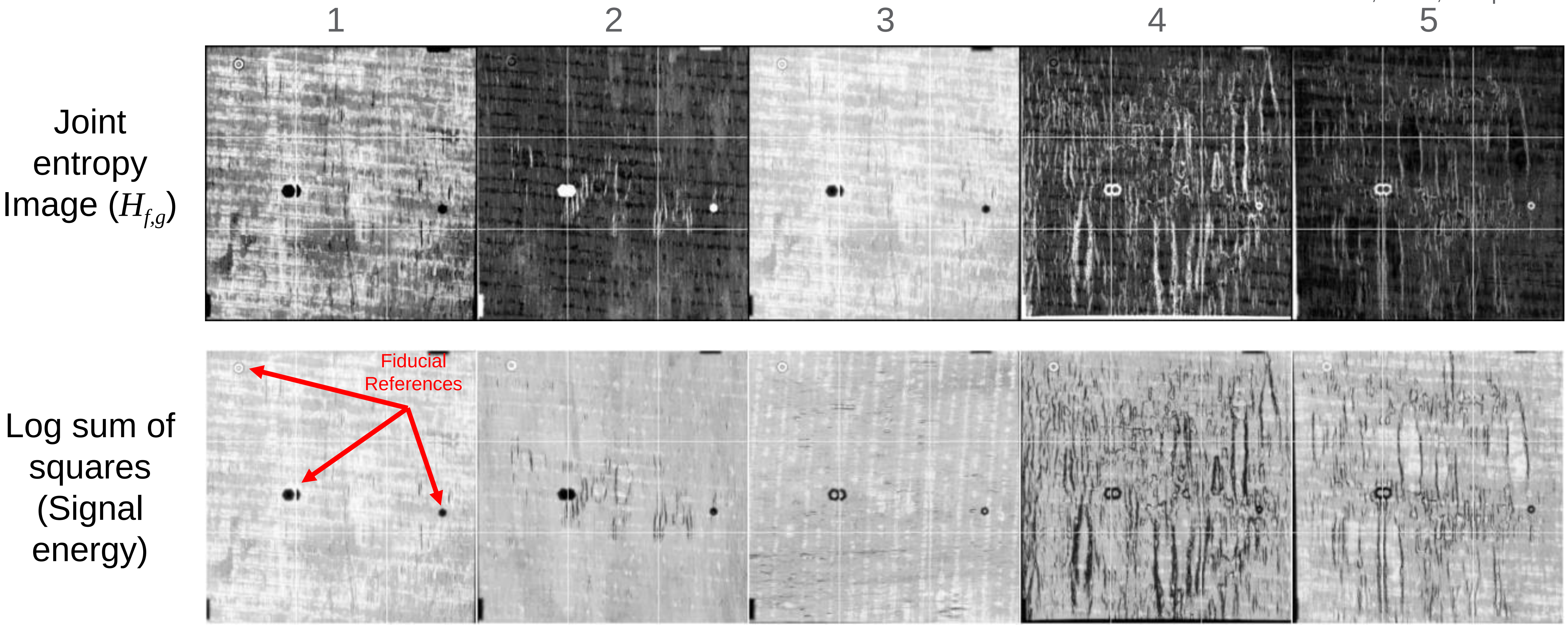
Sample 1 regions scanned and stitched together

- Plaques were examined in 9 sections as independent data sets.
- Fiducials were placed on top of the samples is documented positions and used for image registration for stitching sections into a mosaic of each sample



Ultrasonic Analysis

CONSOLIDATION CONDITIONS:
- 1. 600 sec, 180C, 400 psi
- 2. 300 sec, 200C, 200 psi
- 3. 30 sec, 180C, 400 psi
- 4. 60 sec, 200C, 100 psi
- 5. 30 sec, 160C, 100 psi



The entropy analysis of the ultrasonic data shown in the top row is more sensitive to small defects than typical Log sum of squares c-scan image. The similarities between the imaging techniques indicate gross defects across samples.

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Next steps

Current scope

- Receive updated samples with sealant on edges to prevent water ingress
- Ultrasonically examine sealed samples
- Perform destructive analysis of identified regions of interest
- Correlate defect nature with mechanical response and NDE signals

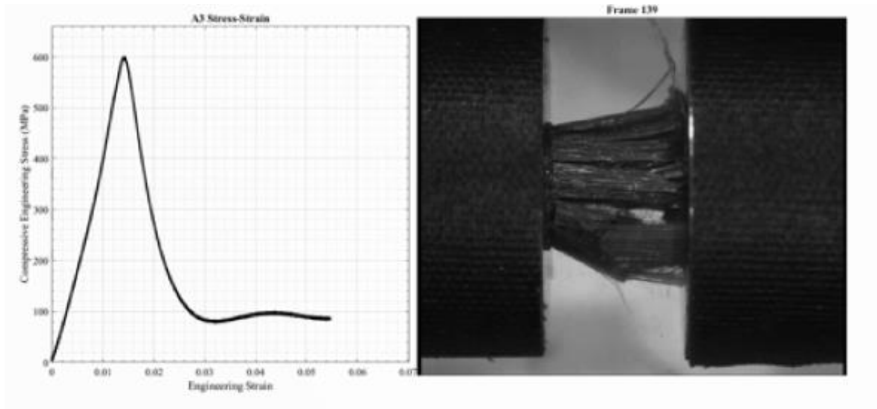
Potential future work

- Continue validation of ultrasonic inspection approach with additional samples and iteration with destructive analysis to develop statistical relationships
- Apply inspection technique to representative sample geometries with complex shapes including curvature
- NDE of samples prior to impact and moderate strain-rate testing at other national labs

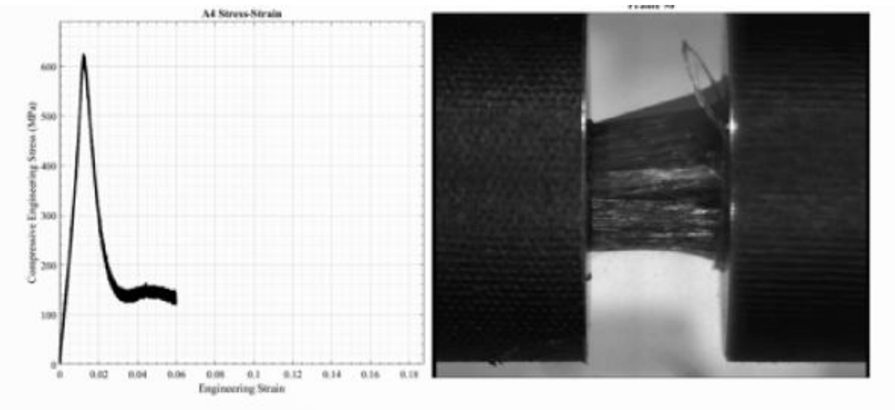
Any proposed future work is subject to change based on funding levels.

SNL: Split Hopkinson Analysis

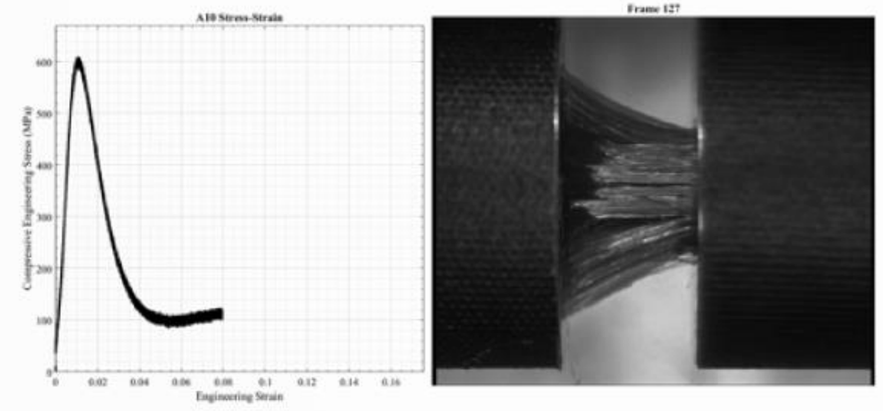
Dynamic compression failure along fiber direction



50 1/s



100 1/s



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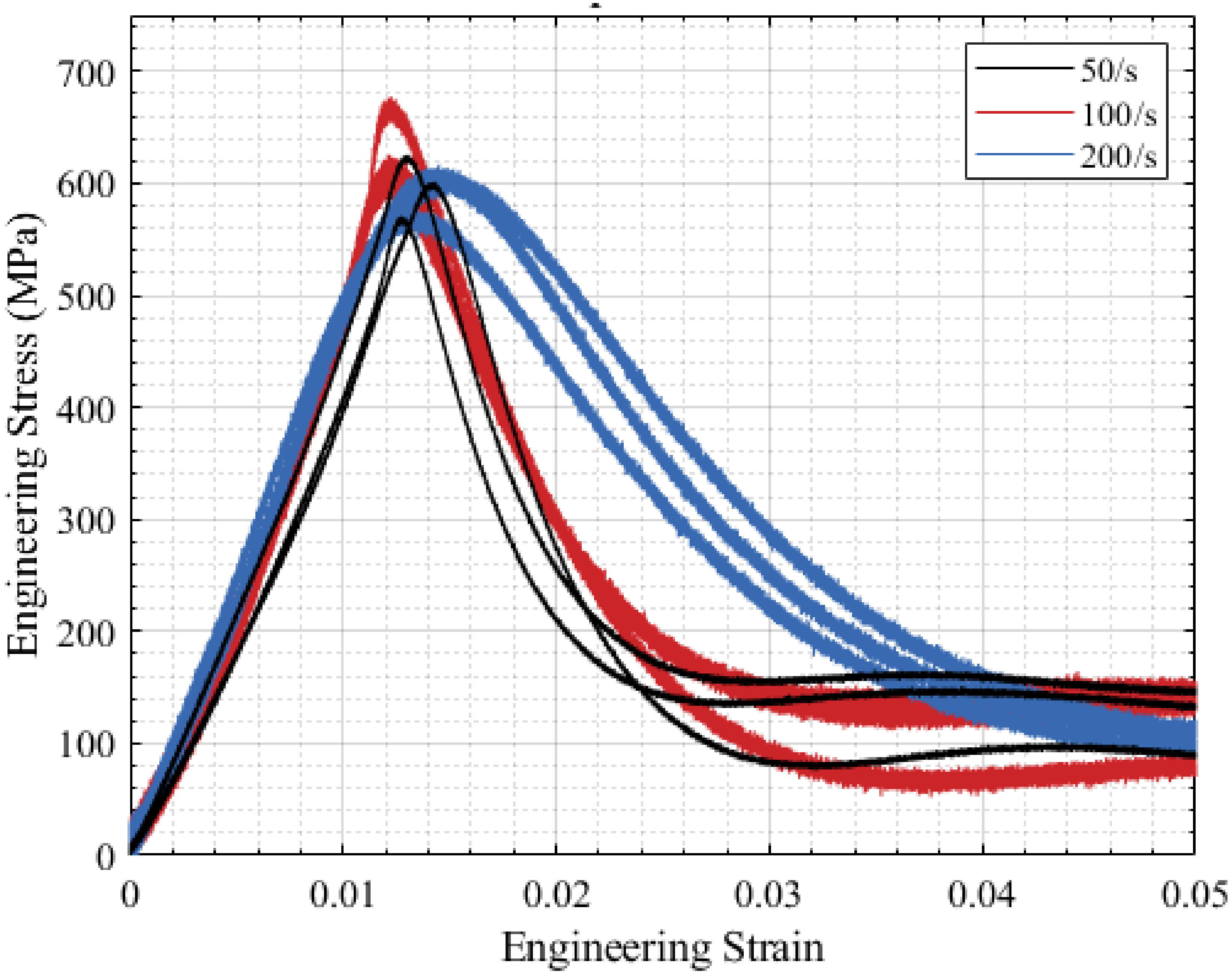
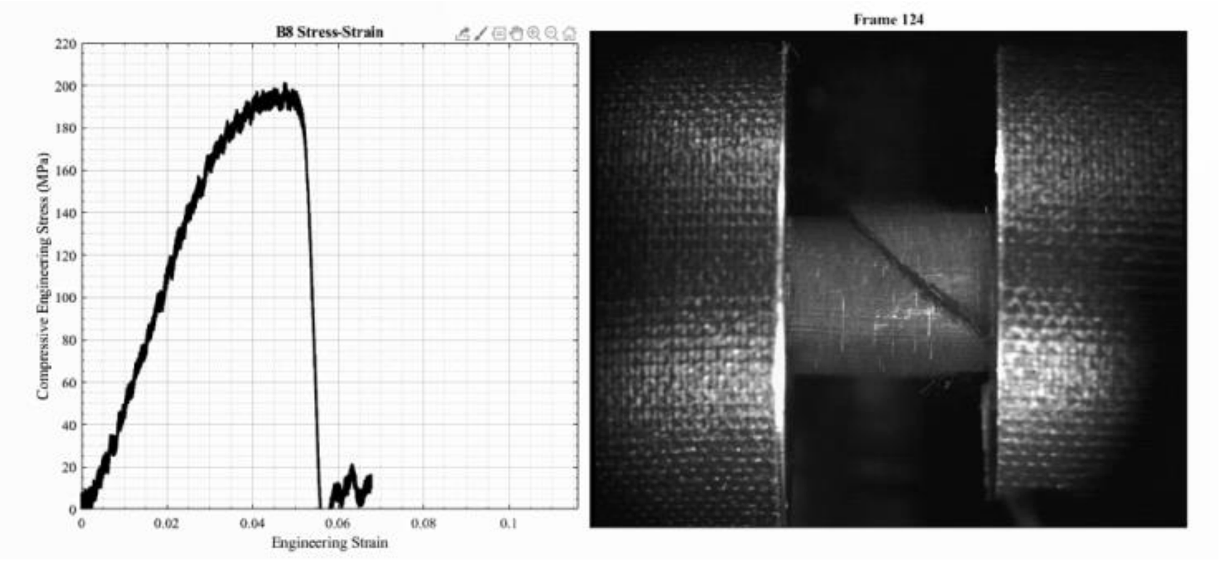


Dynamic compression failure perpendicular to fiber direction



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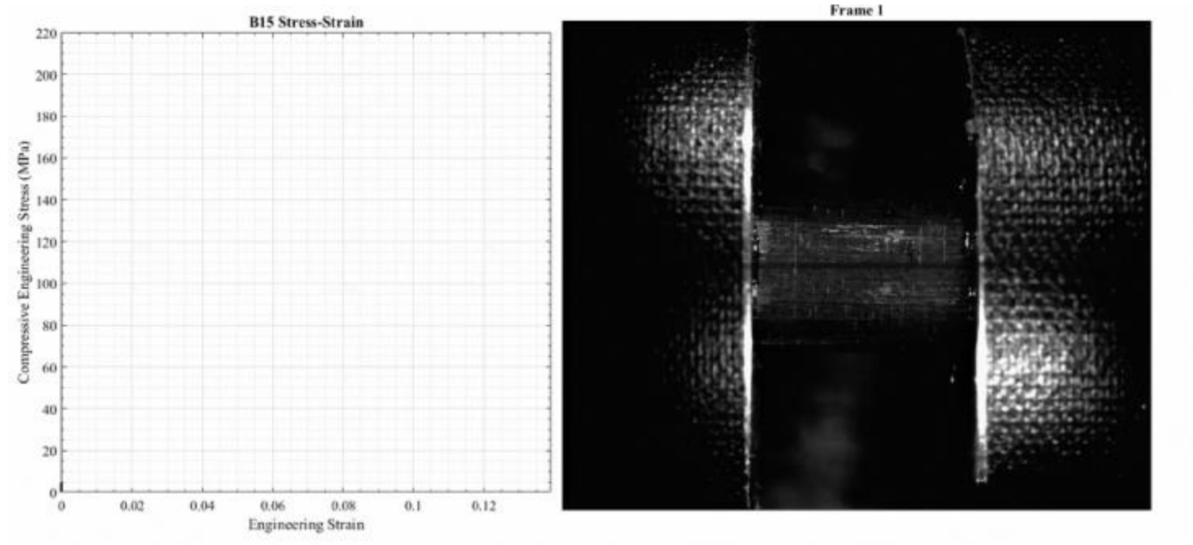
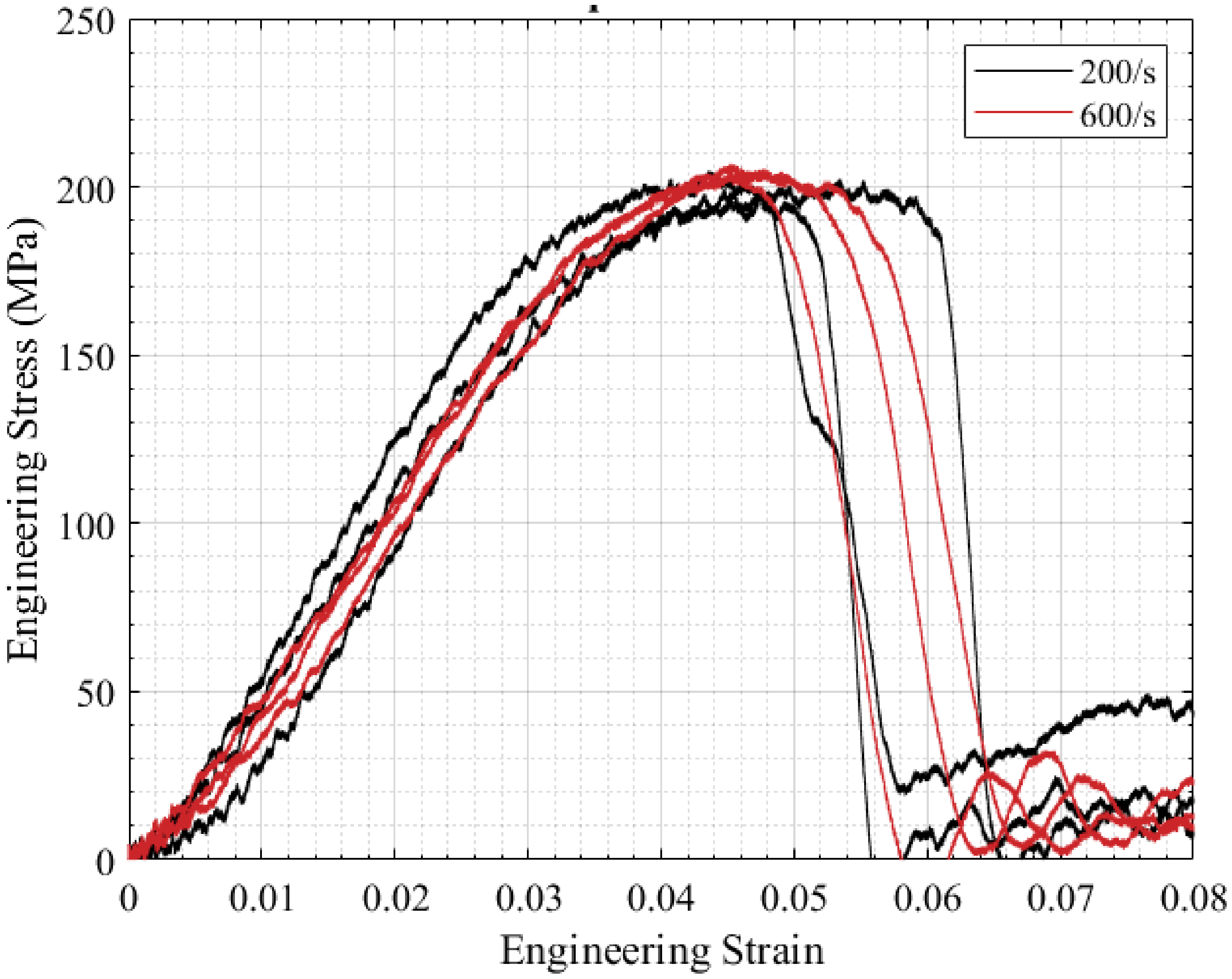
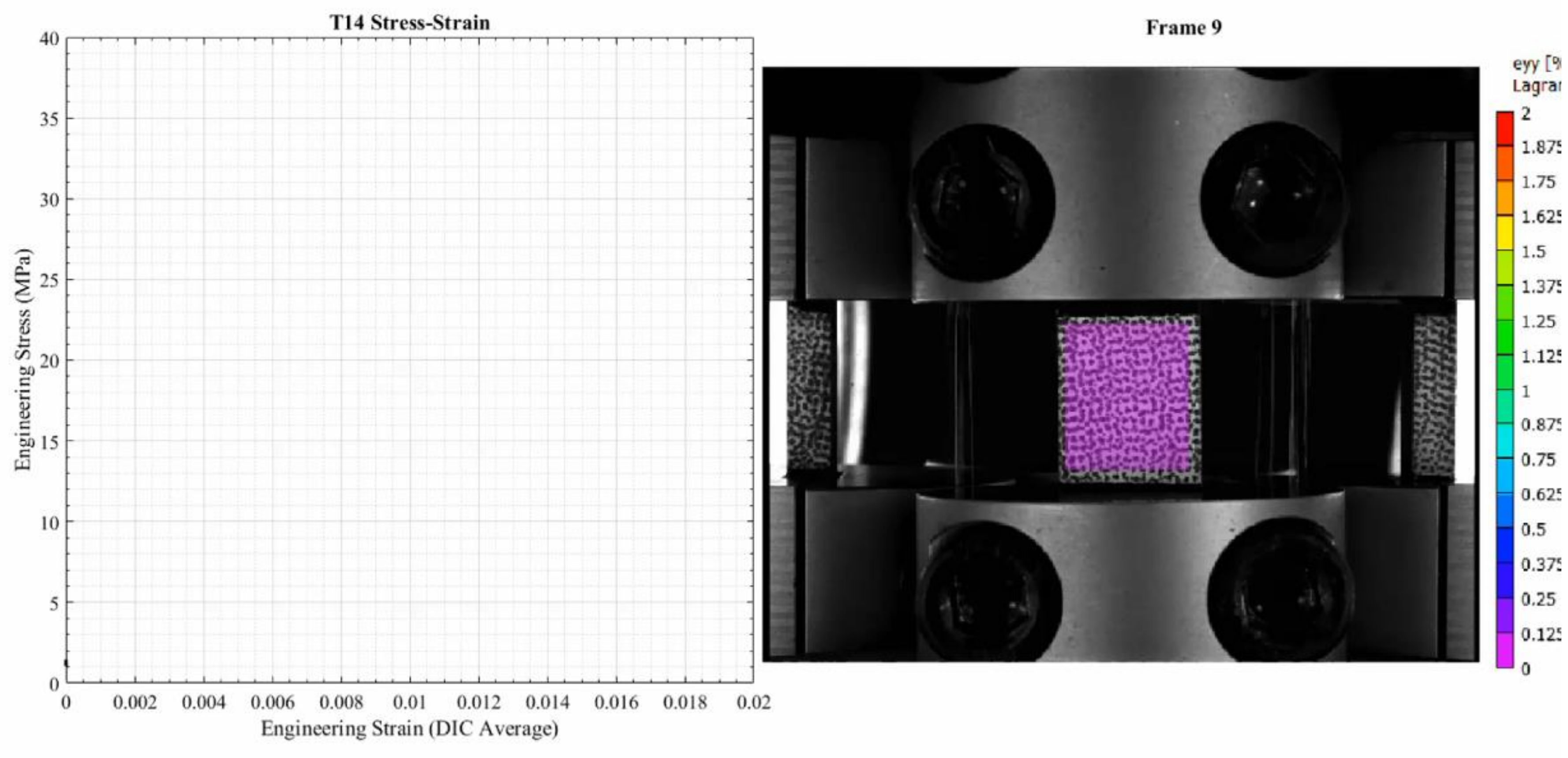
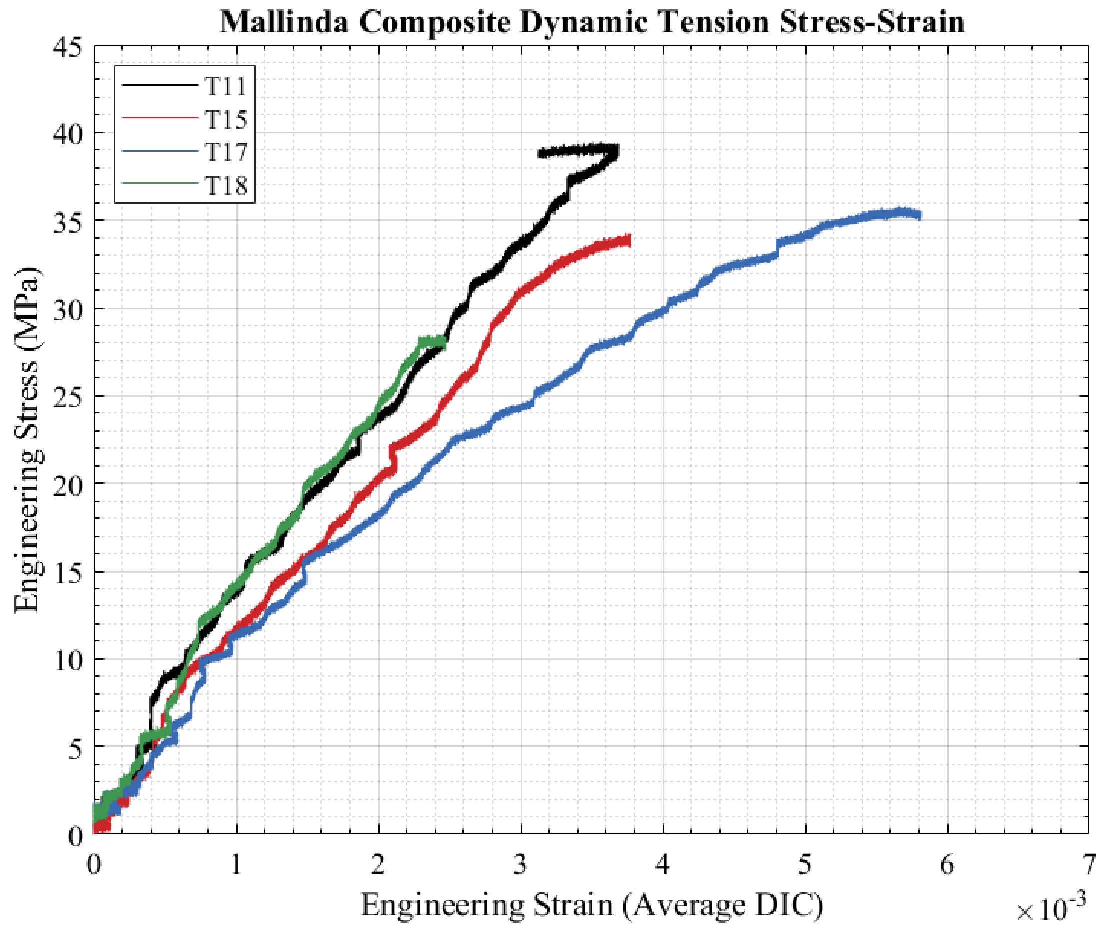


600 1/s



Dynamic tensile failure





Impact, Current Status, Challenges, and Path Forward

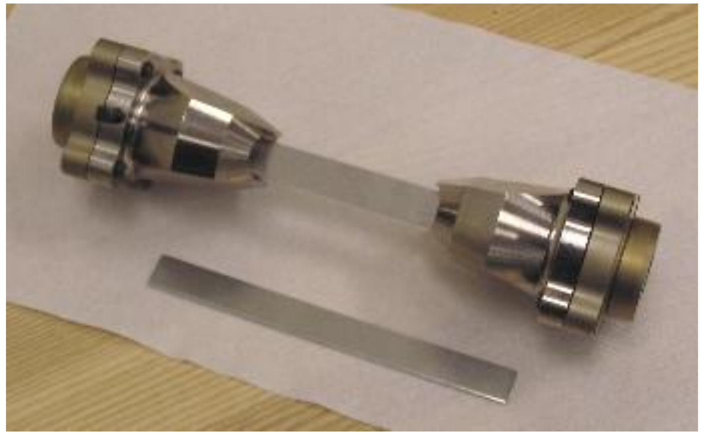
- Impact
  - Mallinda's unidirectional carbon fiber composite was dynamically characterized in compression and tension at various strain rates
  - The material exhibited minimal strain rate effect, but the material response significantly depends on orientation
- Current status
  - Mallinda is sending one more composite panel for more tensile specimens (align with fiber direction)
- Challenges
  - Technical challenge: DIC resolution for dynamic tensile characterization due to brittleness of the composite
  - Financial/time challenge: we have very limited time and funding left for the new panel that Mallinda is sending
- Path forward
  - Environmental temperature effect on dynamic response of the composite
    - Very important to automotive industries

Any proposed future work is subject to change based on funding levels.

High Speed Tensile Test Tests

Test	Sample Dimensions	Strain Rates	Number of Samples/Strain Rate	Total Samples	Material
(Dynamic)Tensile Tests	8”x TBD x 0.03”	125/s, 250/s, 500/s, 1000/s	Up to 5	Up to 20 per material	Mallinda CF Unis and GMT fabric

- Tensile tests to compare rate scaling against earlier tests and for comparing Mallinda/CF system vs GMT commercial system
- Downsized test coupons and strain rates due to failure loads in unidirectional loading



Low-mass grip used in high-rate test machine.

Typical coupon test set-up on high-rate tension machine.

High Speed Flex Testing

- Due to cost and availability of test specimens, TMAC testing of a single OEM part replaced by flex testing of coupons

Customized 3-Point Flex Fixture Utilized for Dynamic Testing of Coupons

Test	Sample Dimensions	Strain Rates	Number of Samples/ Strain Rate	Total Samples	Material
3-pt Flex Testing	10” x 1” x 0.2”	8.3/s, 4.2/s, 2.1/s	~4	Up to 15 per material	Mallinda CF Unis and GMT fabric

OAK RIDGE National Laboratory

Value of Dynamic Test Results

- These tests can be used to evaluate strain rate effects in materials systems from small coupons to components to in many cases actual structures

- Composite failures can be greatly affected by (among others):
  - Multiple constituents utilized (fibers, resins, fillers, etc.)
  - Levels of each of the constituents utilized
  - Arrangements/architecture of the constituents and interfaces
  - Design, shape, joints, etc., involved in making the final components
  - Quality of each manufacturing step
  - Damage present at each stage of life

- Dynamic testing as demonstrated in this work allows direct comparison of many potential approaches/combinations and provides data for design and modeling

Response to previous reviewers' comments

No Comments Received.

Collaboration & Coordination

Pacific Northwest National Lab:
Leading Acoustic method development
-Michael Larche/ Leo Fifield

Sandia National Lab:
Leading split Hopkinson bar high speed testing
-Bo Song

ORNL:
Leading Strain rate testing
-Robert Norris

Remaining Challenges & Barriers

- Completing the testing on-time is a challenge: sample prep and machine availability have been hurdles

- Per prior year reviewers' prior comments, and project team discussions, the scope of work has been modified to characterize a baseline vitrimer composite material rather than optimizing for impact performance. Even then, Covid-related shut-downs proved a challenge.

Recommended Follow-on Studies

- Analysis of recovered/recycled composite material- LCA.
- C-scan assessment of structural repair using Imine-exchange mechanism
- Further study of strain rate effect varying fiber type (including natural fiber) and varying resin : fiber ratio

Any proposed future work is subject to change based on funding levels

Summary

PREVIOUS KEY TAKE AWAYS

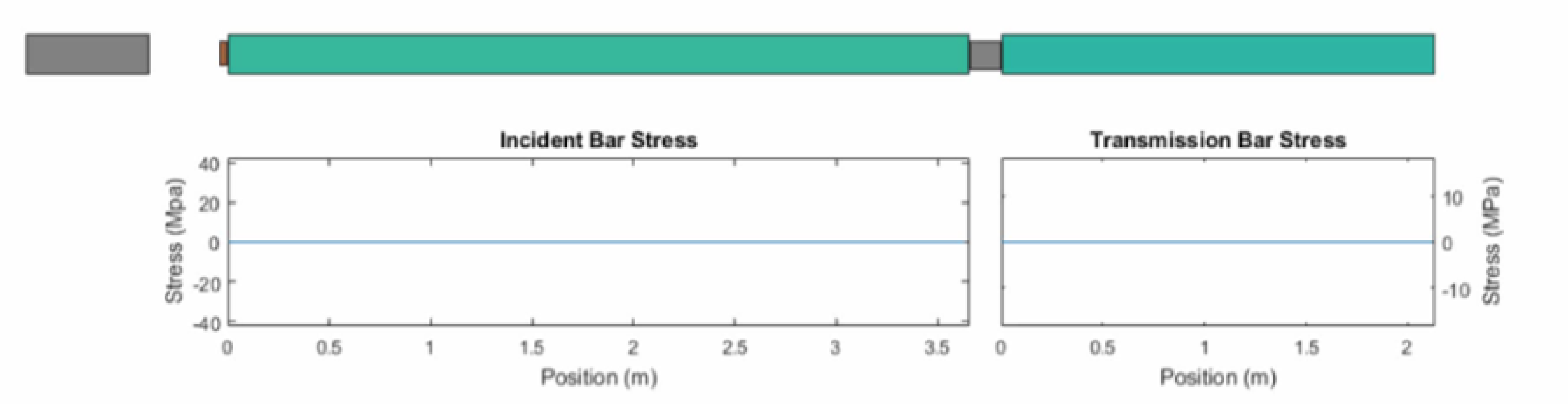
- Mallinda Inc. successfully developed a high Tg resin system for automotive application.
- Mallinda Inc. manufactured in-house scaled production of resin and tolled 200 linear yards of prepreg.
- The 3-minute in mold compression forming was performed to produce an OEM automotive part.
- Complete recycling of part into resin and fiber was successfully demonstrated.
- Initial intermediate strain dynamic tensile and compression tests revealed mild strain rate response.
- The Time-Temperature-Pressure parameters were systematically studied to determine impact of each variable on vitrimer weld-strength as determined by short beam shear testing. While both time and pressure have a positive impact on bond strength, temperature is more dynamic, and must be optimized.

KEY TAKE AWAYS

- Material has been delivered to ORNL, SNL, & PNNL- Final testing run is under way in each facility.
- Feedback from final round testing & resulting re-sampling has occurred for ORNL, SNL & PNNL
- Once complete, the first studies of vitrimer composite strain-rate response, and ultrasonic analysis of vitrimeric consolidation will have been enabled by this LightMAT project.
- Imine-linked Vitrimer composites competitive mechanical properties, rapid processability, and facile recyclability have been successfully demonstrated in this LightMAT project

Technical Back-up Slides

Kolsky bar technique for material property characterization at high strain rates



One-dimensional Elastic Stress Wave Propagation

Mass Conservation

$$V_1 = C_0(\varepsilon_i - \varepsilon_r)$$

$$V_2 = C_0\varepsilon_t$$

$$\dot{\varepsilon} = \frac{V_1 - V_2}{l_s} = \frac{C_0}{l_s}(\varepsilon_i - \varepsilon_r - \varepsilon_t)$$

$$\varepsilon = \int_0^t \dot{\varepsilon}(\tau)d\tau$$

Momentum Conservation

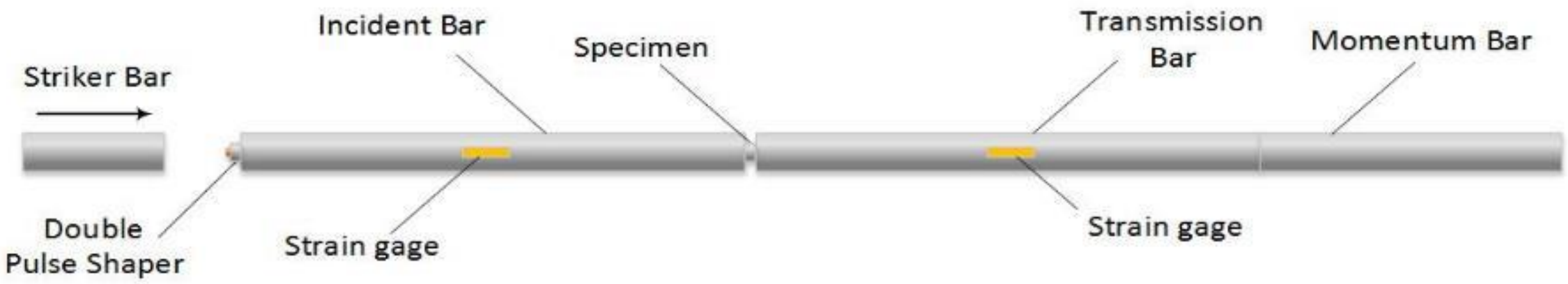
$$F_1 = E_0A_0(\varepsilon_i + \varepsilon_r)$$

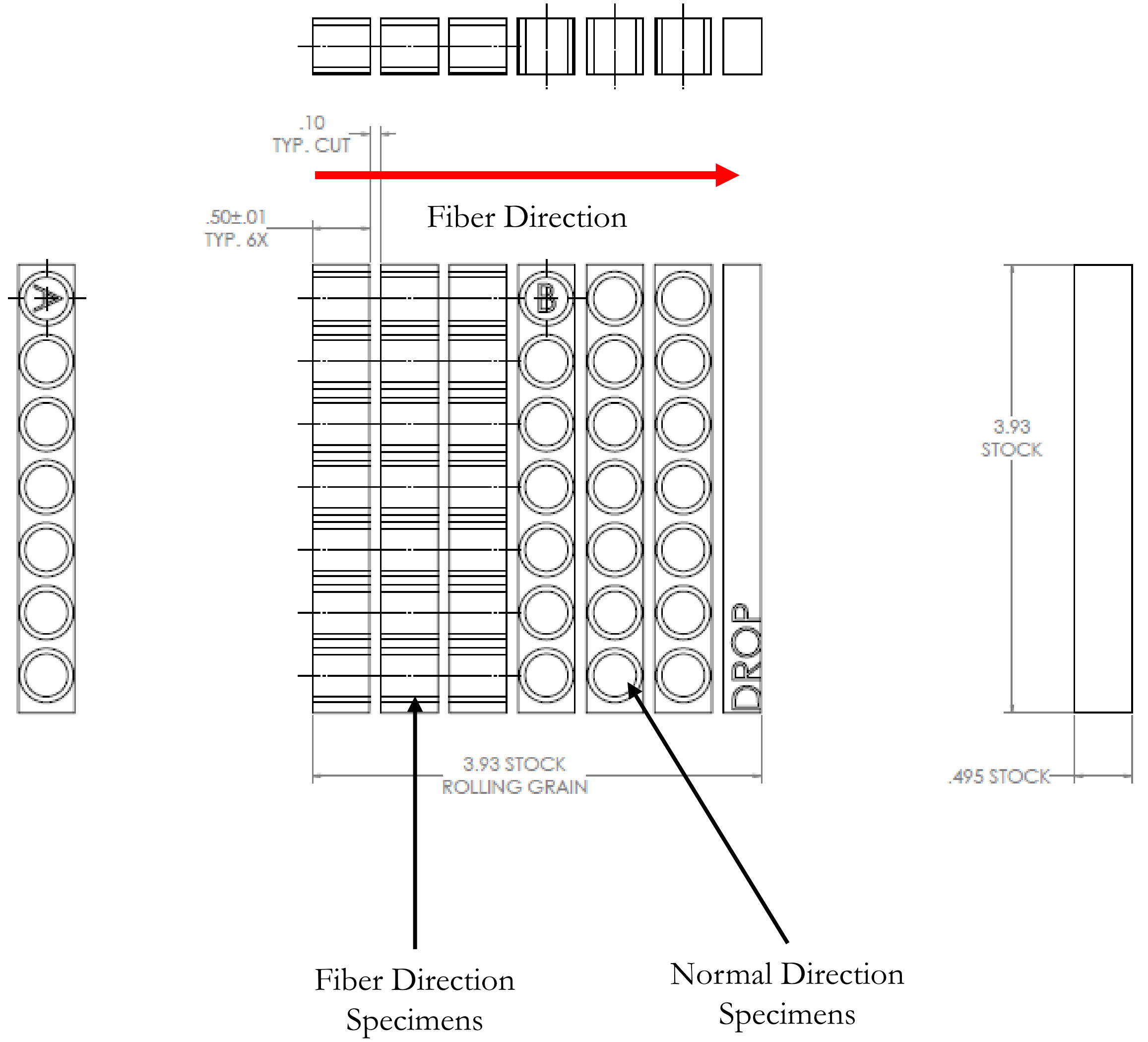
$$F_2 = E_0A_0\varepsilon_t$$

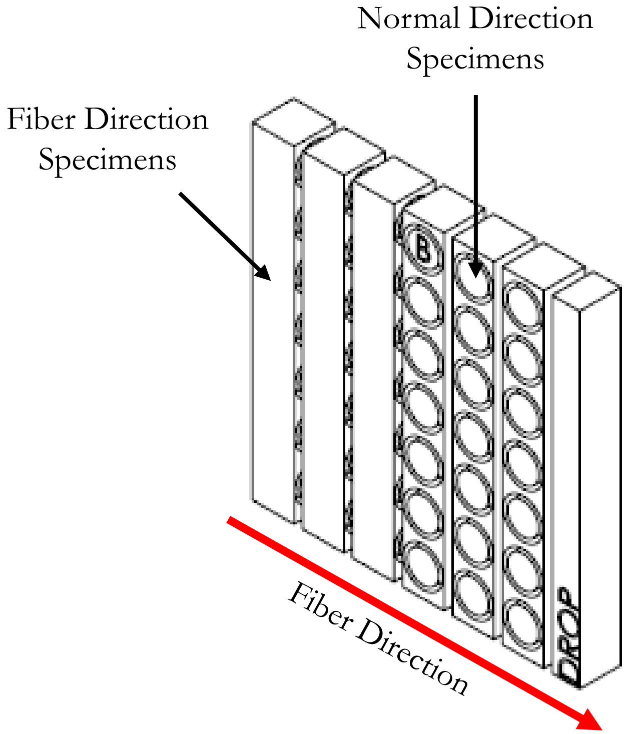
$$\sigma = \frac{F_1 + F_2}{2A_s} = \frac{E_0A_0}{2A_s}(\varepsilon_i + \varepsilon_r + \varepsilon_t)$$

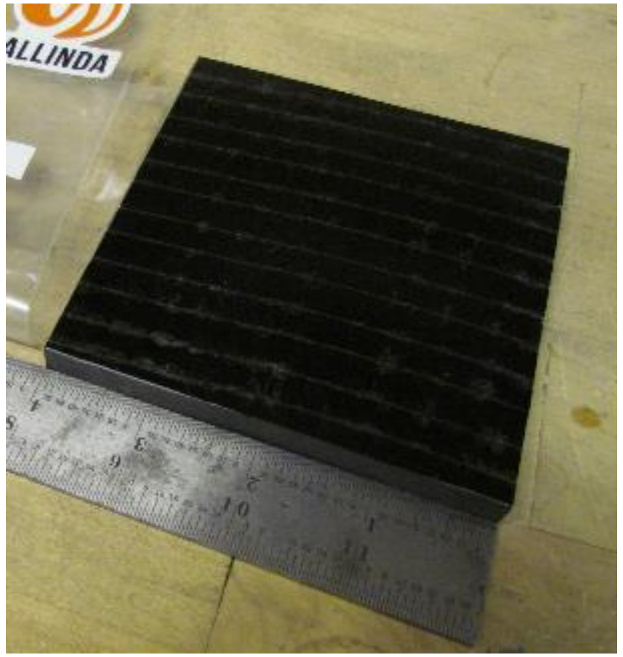
$$\sigma \sim \varepsilon$$

Kolsky compression bar for Mallinda unidirectional carbon fiber composite









Original Planned Dynamic Coupon/Component Tests

TMAC Machine Has Been Recently Restored/Upgraded

- **Test Machine for Automotive Crashworthiness**
  - Tube crushing or tension at 100's of kN
  - Constant velocity to 8 m/s (18 mph) thru 25 cm stroke
- **High Speed Coupon Tester**
  - Coupon tensile testing at loads to 22 kN
  - Constant velocity to 18 m/s (40 mph)
- **High Speed Video and Digital Image Correlation for Failure Analysis**
- **TMAC Specifications:**

Physical

1 m daylight

490 kN actuator cap. (static)

250 mm stroke

> 490 kN side-load capacity

Attachable 450 kg mass

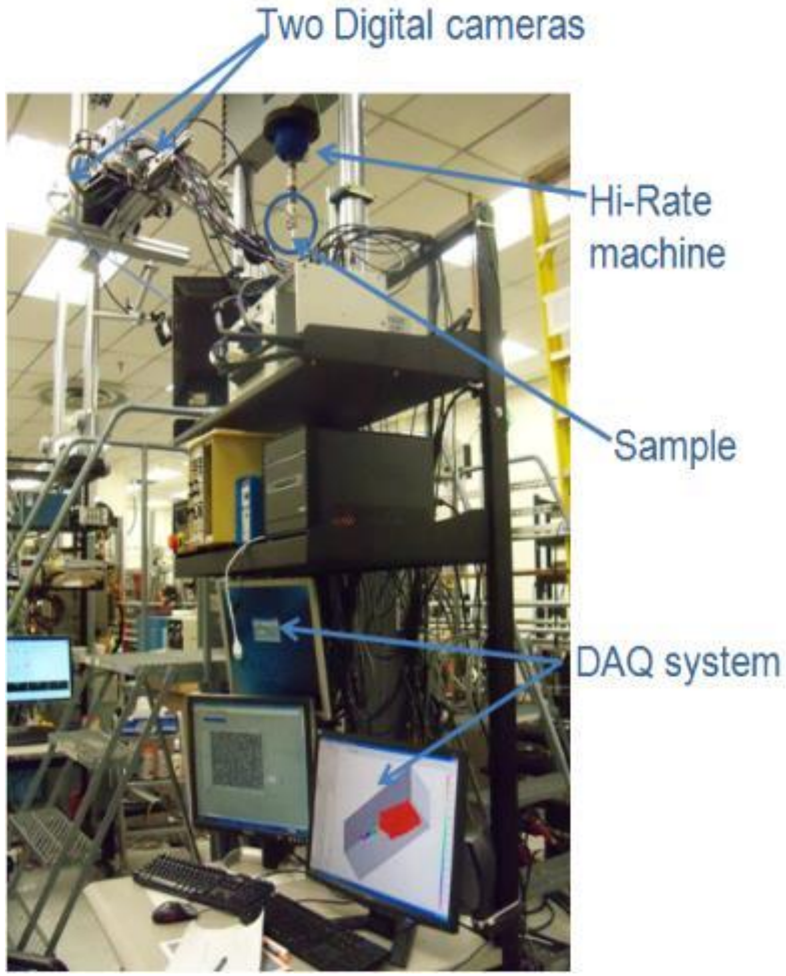
Operating

No load: 230 mm travel at 8 m/s

113 kN: 230 mm travel at > 6 m/s

267 kN: 115 mm travel at 6 m/s

Target velocity constant within ± 10% for 115 mm



High Speed Coupon Tester