OLD DOMINION UNIVERSITY
IDEA FUSION

Automotive Composites Conference and Exhibition, Novi, Michigan, Sept. 7-9, 2022

# Improving Mechanical Properties of 3D Printed Short Glass Fiber Nylon Composites using Isostatic Compaction

ODU MECHANICAL AND AEROSPACE ENGINEERING

COMPOSITES MODELING AND MANUFACTURING GROUP

Patricia Revolinsky, Pushpa Jain, John Barber, Siavash Sattar, Alex Kravchenko

Department of Mechanical and Aerospace Engineering, Old Dominion University, Norfolk, Virginia

Composites Modeling and Manufacturing Group

# Background

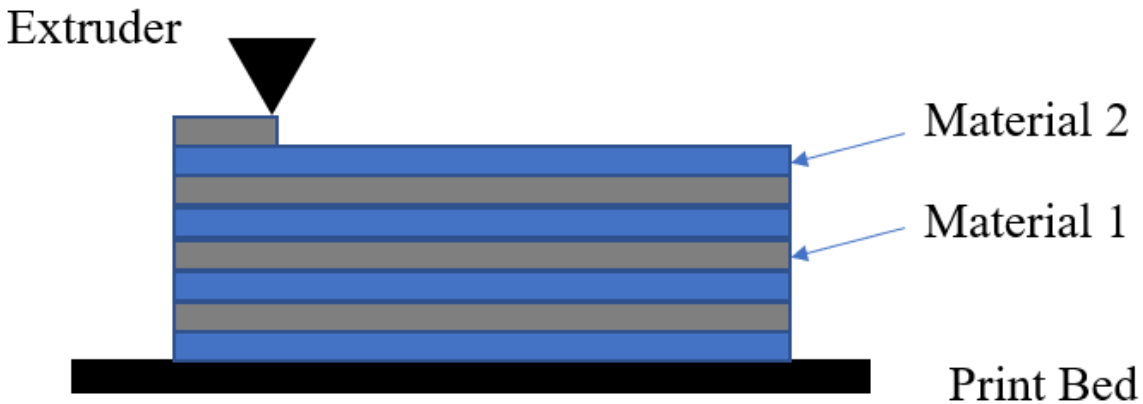
- Secondary structural components
  - Complex geometry
  - Composite
    - Multiple materials
  - Difficult to make using traditional manufacturing methods with fiber reinforced polymers
- Highest strength around mount
  - Control fiber direction using additive manufacturing (AM)

Image courtesy of Composites World

# Additive Manufacturing

COMPOSITES MODELING AND MANUFACTURING LAB

- Fused Deposition Modelling (AM)
  - Deposit molten material
  - Layer by layer
- Composites

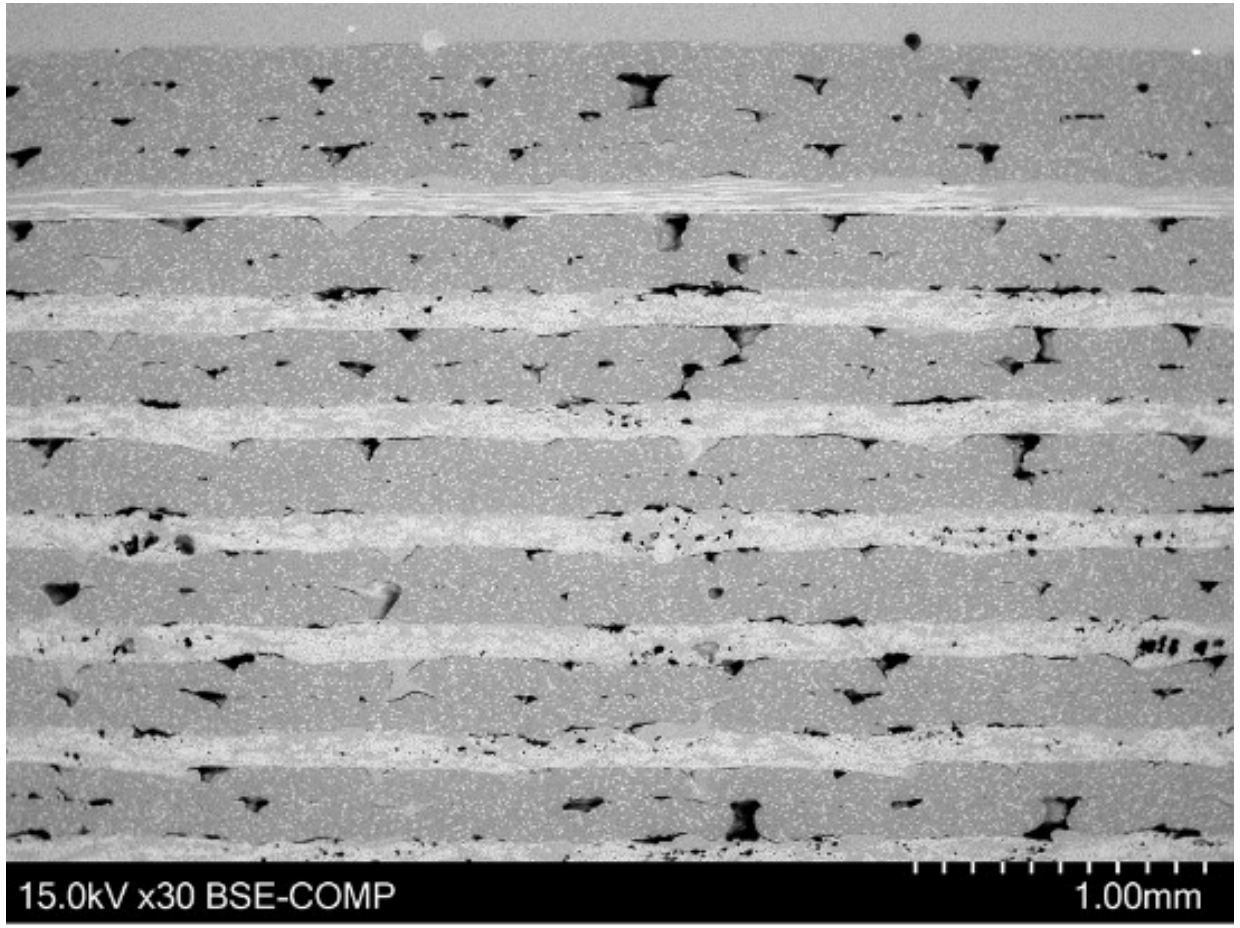



- Voids
- Compression Mold
  - Reduce voids
  - Connect pieces

9T Labs Red Platform Image courtesy of Composites World

# Voids

- Voids act as ***stress concentration*** points
- Inter-bead Voids
  - Between printer nozzle passes
- Deconsolidation
  - Carbon Fiber (CF) decompaction
    - Stress due to CF overcomes sizing material bonding

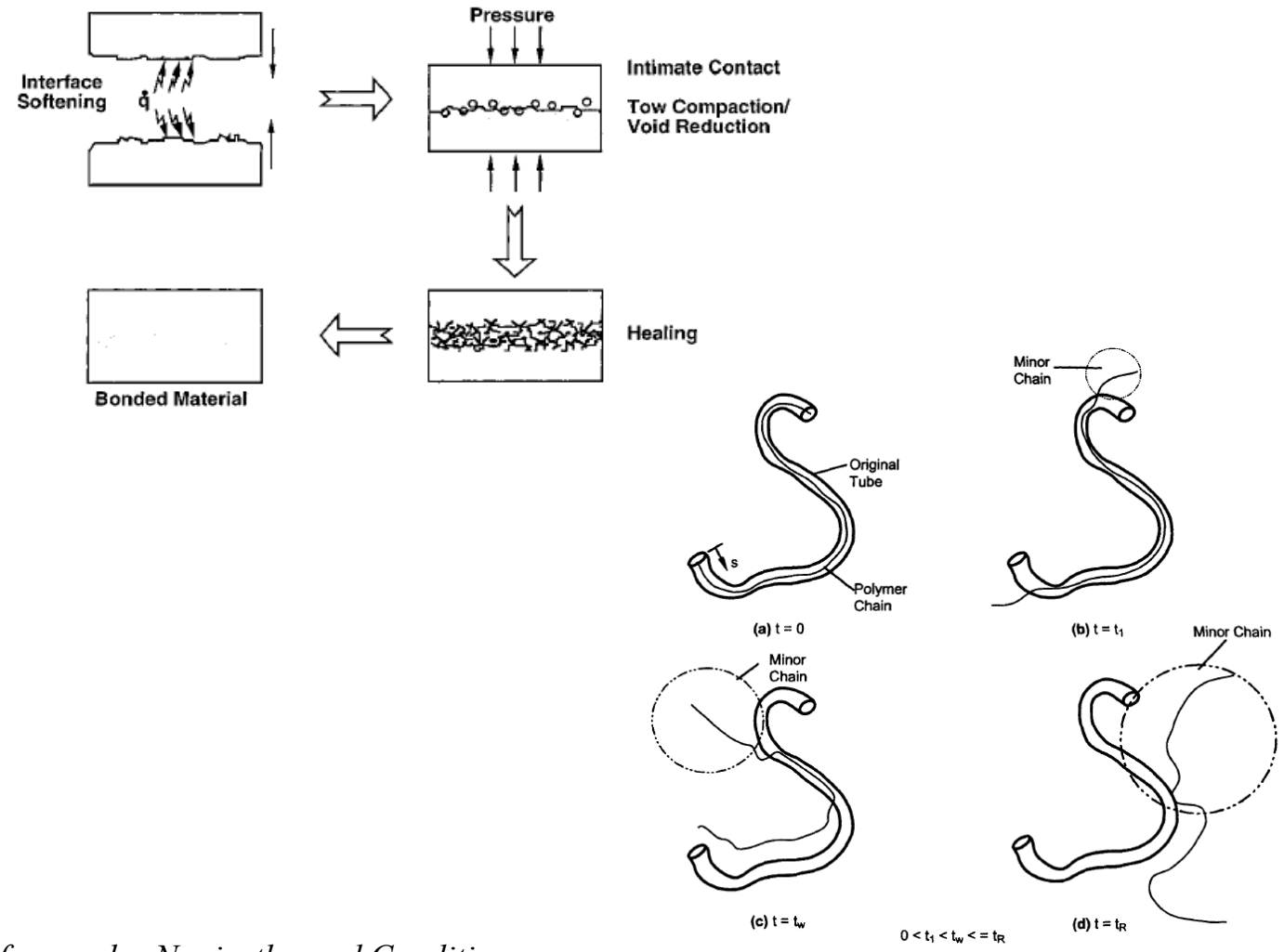



O-025-015

[1] Zhang et al. *Research on the mechanical properties prediction of carbon/epoxy composite laminates with different void content, 2016*
[2] De Almeida et al. *Effect of void content on the strength of composite laminates, 1994*
[3] Tekinalp et al. *Highly oriented carbon fiber-polymer composites via additive manufacturing*

# Reptation

- Non-isothermal problem
  - Uneven temperature
  - Layers cool during placement
- Fusion bonding
- Long polymer molecules
- Atomic vibration
- Welds surfaces
- Temperature and pressure dependent





[15] Yang et al *Healing of Thermoplastic Polymers at an Interface under Nonisothermal Conditions*

[16] Yang et al *Nonisothermal Healing and Interlaminar Bond Strength Evolution During Thermoplastic Matrix Composites Processing*

# Nylon 6 Phase Transformation

## Nylon 6 phases: amide molecule chains attached with hydrogen bonds

**Gamma**

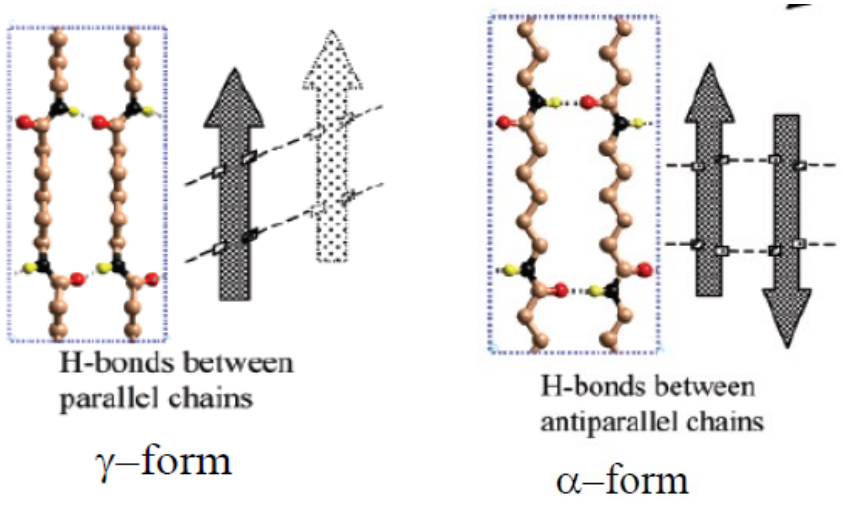
- Most ductile
- Least dense

Beta

- Transitional

**Alpha**

- ***Highest strength phase***
- Lowest ductility
- Most tightly packed
- ***150*** to ***200*** degrees C in nylon 6



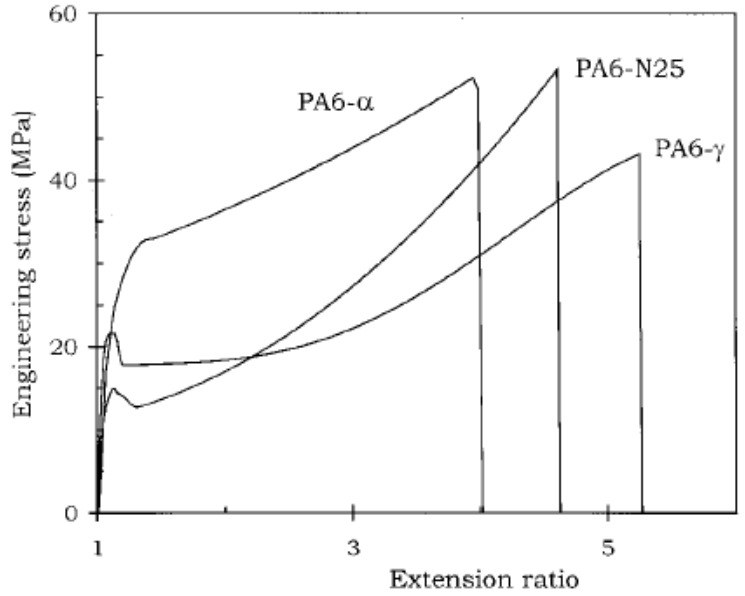




**Figure 3.** Engineering stress–strain curves of PA6-α, PA6-γ, and PA6-N25 at 120 °C at a crosshead speed of 50 mm/min.


[4] Ramesh et al. *High-Temperature X-ray Diffraction Studies on the Crystalline Transitions in the α- and γ-Forms of Nylon-6, 2001*
[5] Sang et al. *A comparative study of the crystalline structure and mechanical properties of carbon fiber/polyamide 6 composites enhanced with/without silane treatment, 2016*
[6] Cavallo, et al. *Effect of cooling rate on the crystal/mesophase polymorphism of polyamide 6, 2011*
[13] Penel-Pierronet al., "Structural and Mechanical Behavior of Nylon-6 Films. II. Uniaxial and Biaxial Drawing"

# Transcrystallinity

- ***Carbon fiber acts as nucleation site*** for nylon
- Increased alpha phase leads to increased nylon 6 strength
- Limitation
  - Upper threshold on transcrystalline layer thickness
- Pure annealing
  - gamma to alpha
- Pressure facilitates transcrystallinity

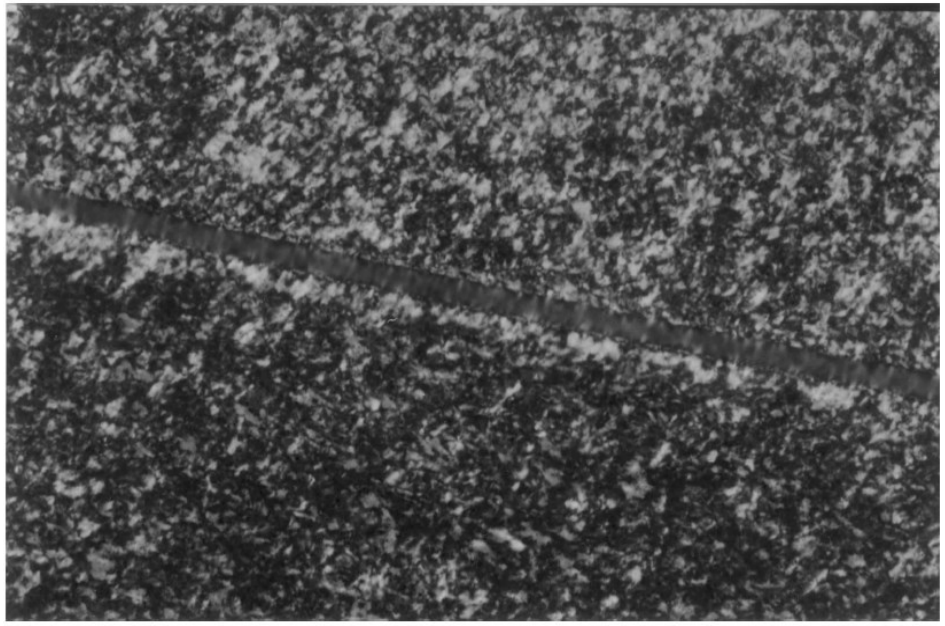


*Figure 13* Polarised optical image of GF/PA6 thin film cooled in hot-press (cooling rate:- −1 °C/min, holding pressure: 1.5 MPa, magnification: ×250).

Cartledge et al

[7] Wang et al. *A comparative study of the crystalline structure and mechanical properties of carbon fiber/polyamide 6 composites enhanced with/without silane treatment, 2016*
[8] Harel et al. *On crystalline interfaces in composite materials, 1998*
[14] CARTLEDGE et al., "Studies of microstructural and mechanical properties of nylon/glass composite"

# Goals and Objectives

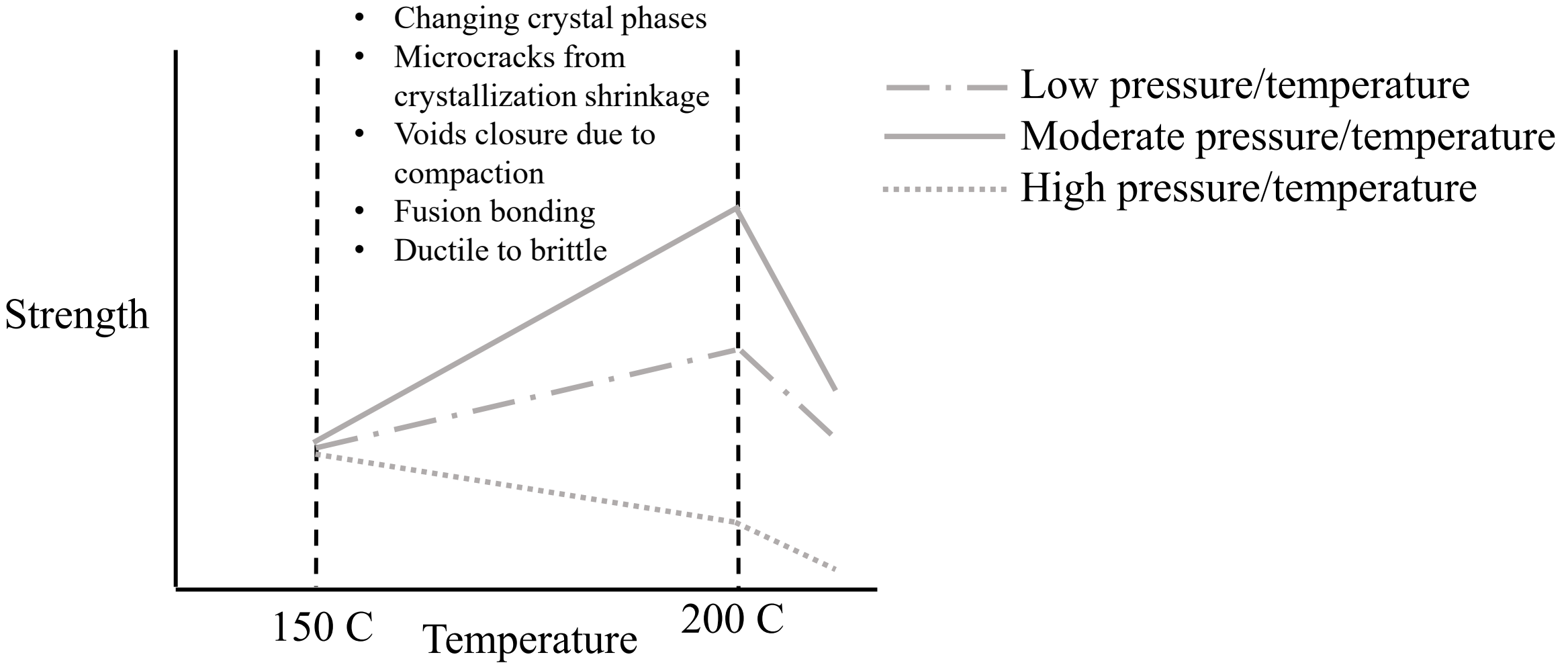
Objective:

- Produce post-processing method that reduces void content

Goals:

- Characterize AM CFRP performance for different post-processing treatments
- Identify contributors to different performance among treatments
- Quantify effect of parameters on performance

# Physics of Compaction with Annealing



# AM Part Manufacturing

COMPOSITES MODELING AND MANUFACTURING LAB

Need for Simulation Tools to Ensure Quality During Post-Processing

Acceptable Choice
of Post-Processing Parameters

Defective Choice
of Post-Processing Parameters

# Short Carbon Fiber Nylon: Post Treatment

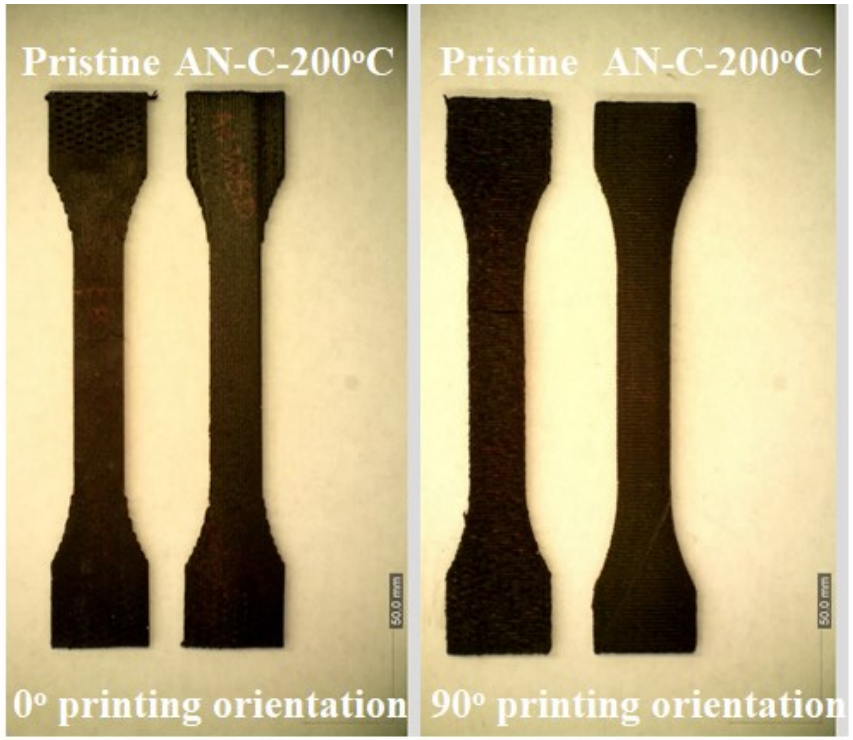



**Table**. Post-processing conditions for annealing and iso-static compaction

| Post fabrication treatment | Temperature |
|---|---|
| **Annealing (AN)** | 150 °C for 4hr |
| | 170 °C for 4hr |
| | 200C for 30min |
| **Annealing with Compaction (AN-C)** | 150 °C for 4hr at 0.55 MPa (80 psi) |
| | 170 °C for 4hr at 0.55 MPa (80 psi) |
| | Step 1: 200 °C for 30min at 0.55 MPa (80 psi)<br>Step 2: 150 °C for 2hr (without pressure) |

# Effect of Post-Treatment: Crystal Structure

**Figure**. Raman spectroscopy analysis of pristine 3D printed nylon treated at 150 °C, 170 °C and 200 °C.

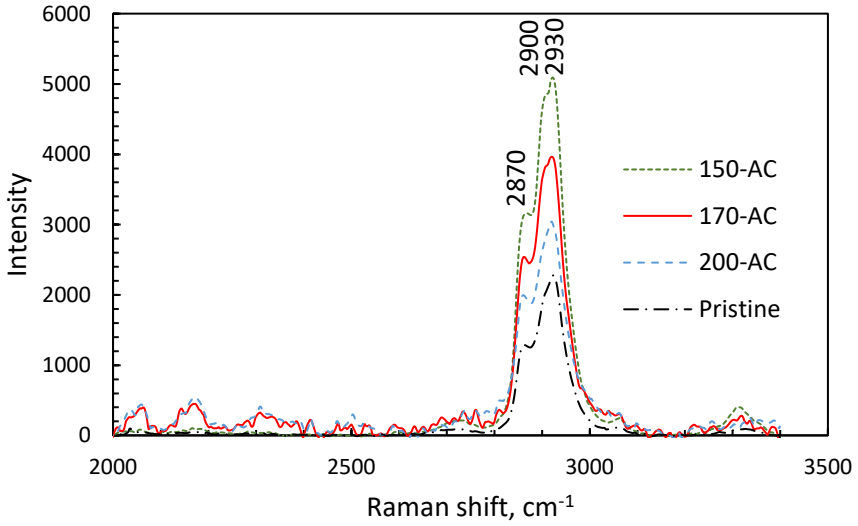
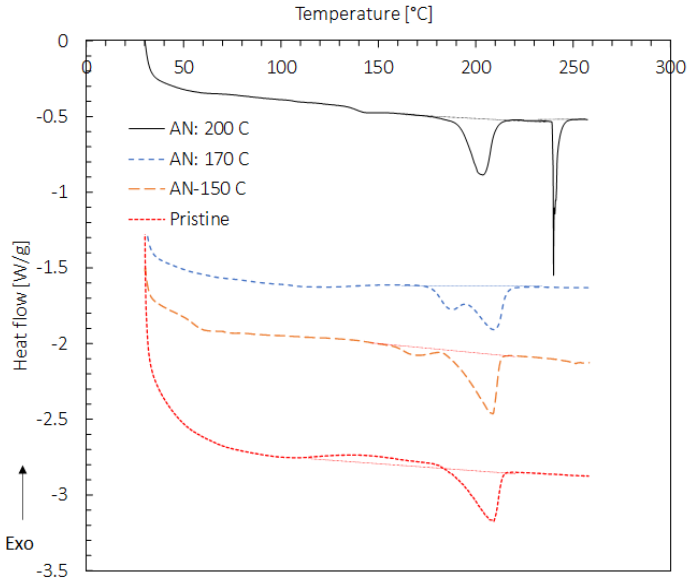



**Table**. Degree of crystallinity in pristine and treated PA6 glass fiber composites

| Treatment temperature | Degree of crystallinity |
|---|---|
| Pristine | 34.0% |
| 150 °C | 38.6% |
| 170 °C | 40.3% |
| 200 °C | 46.0% |



# Effect of Post-Treatment: Micro Structure

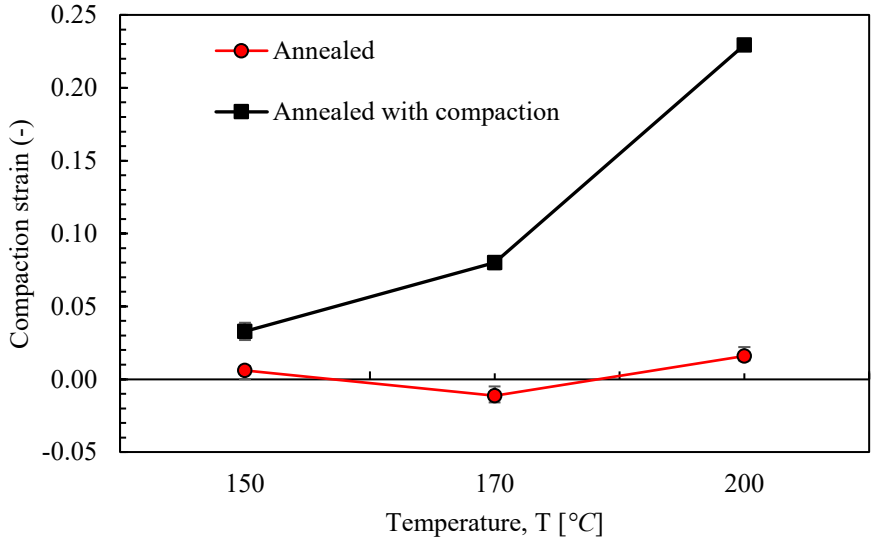
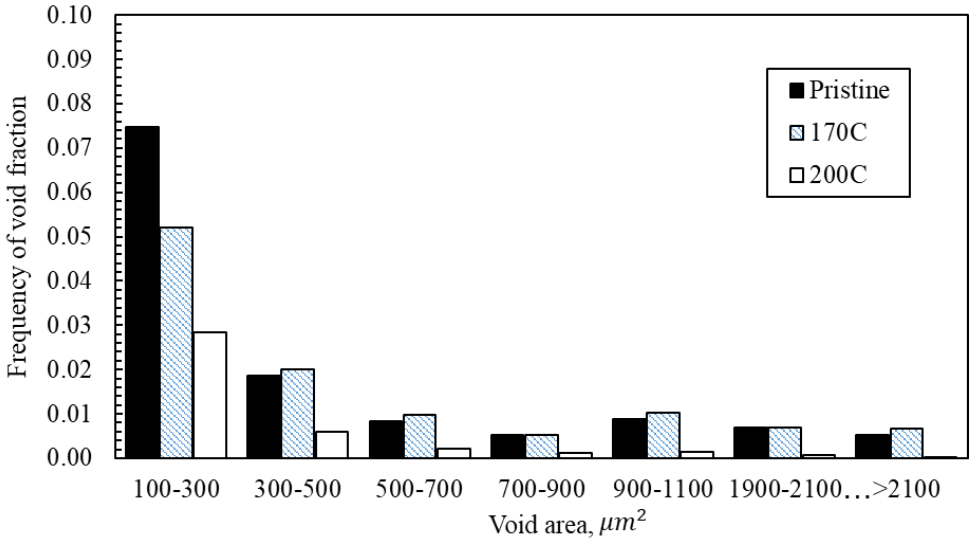
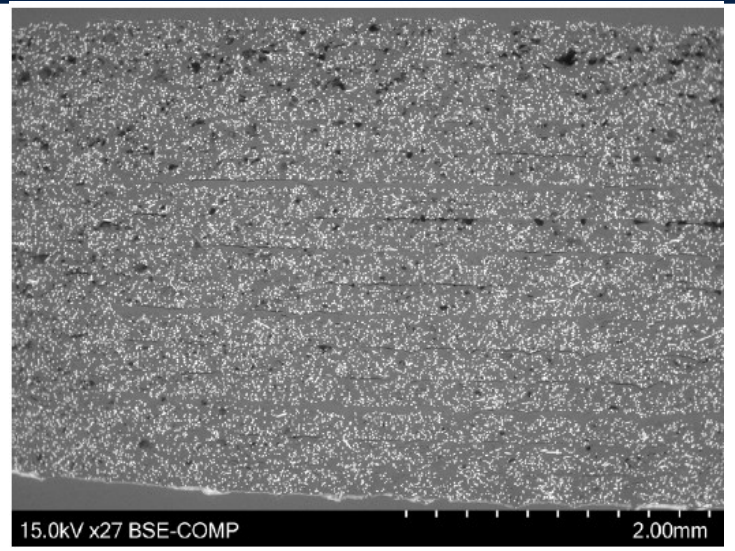



**Figure 9**. Histogram of void content in annealed and compacted samples




As-printed morphology of an AM composite




Morphology of an AM composite after compaction & annealing

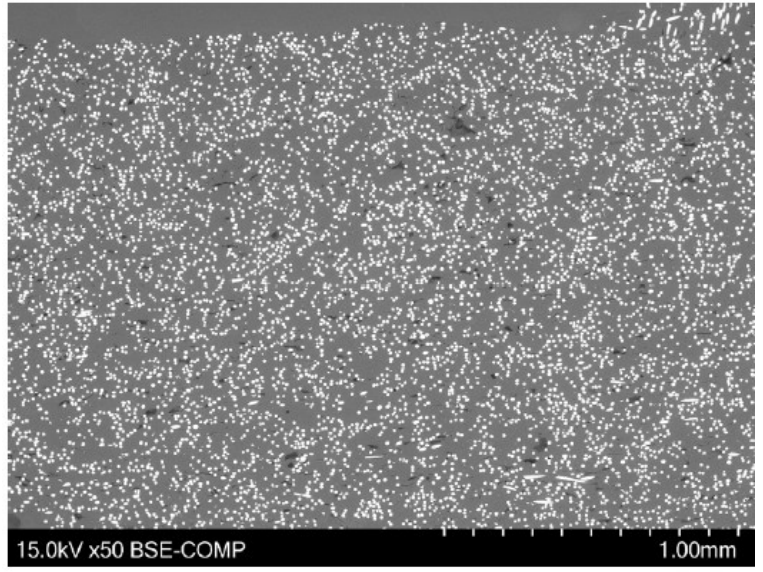
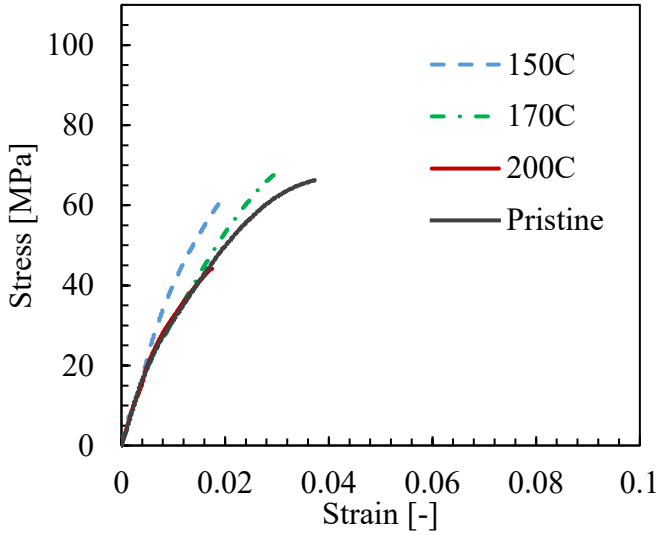


**Figure**. Representative stress-strain curves for annealed samples at different temperatures for 0° (a) and 90° (b) samples



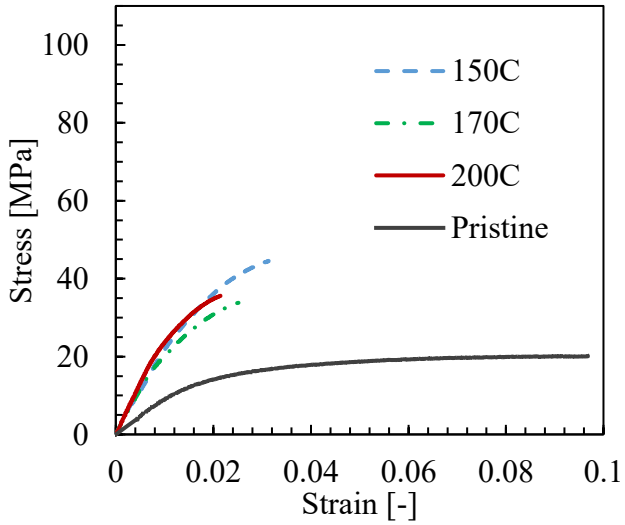
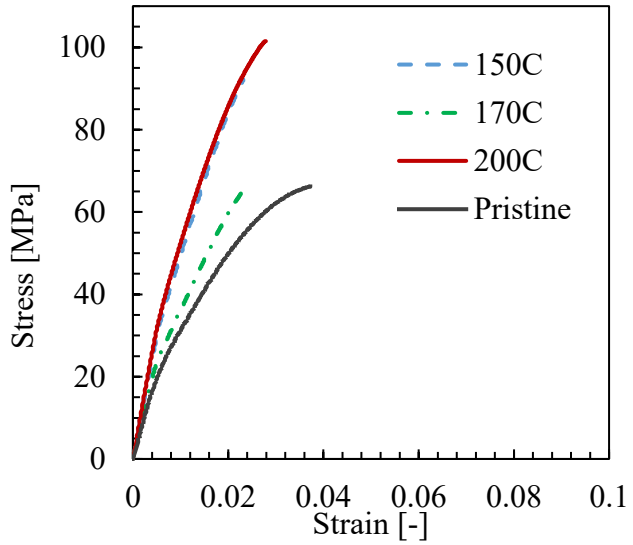
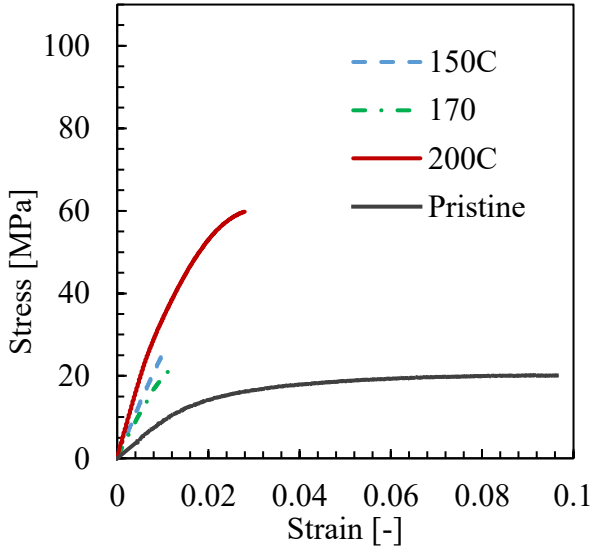
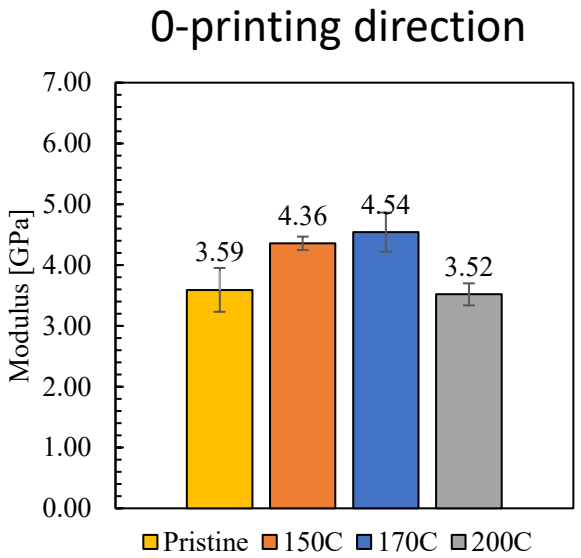




**Figure**. Representative stress-strain curves for compacted samples at different temperatures for 0° (a) and 90° (b) samples

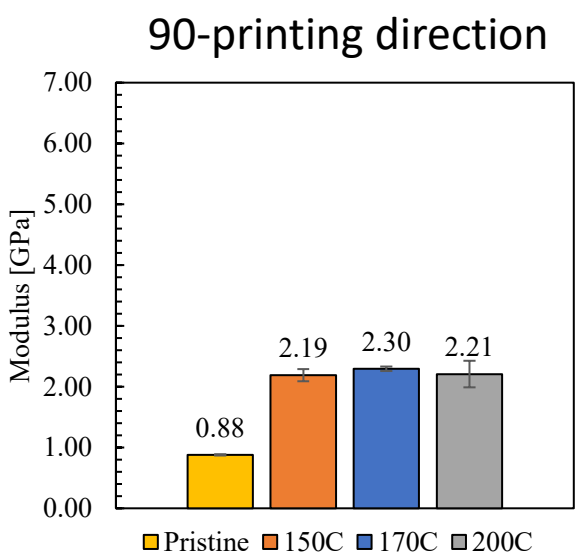


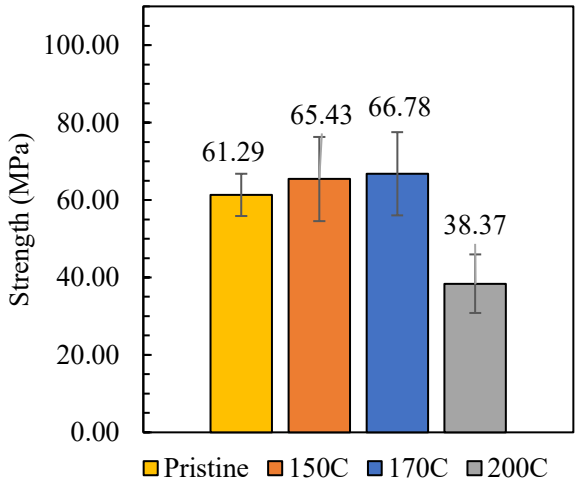
# Effect of Annealing: Mechanical Properties



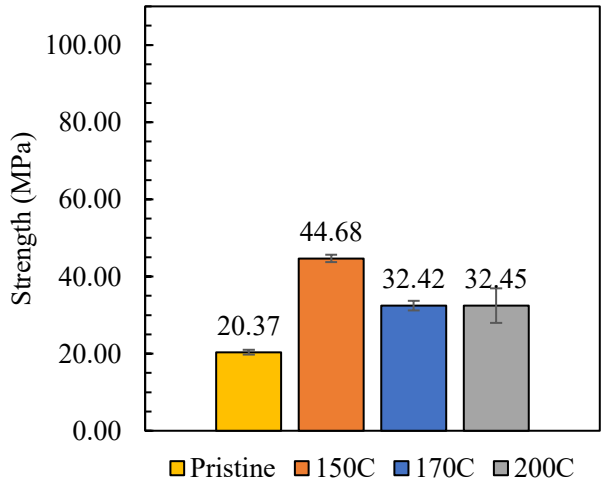

a.




b.



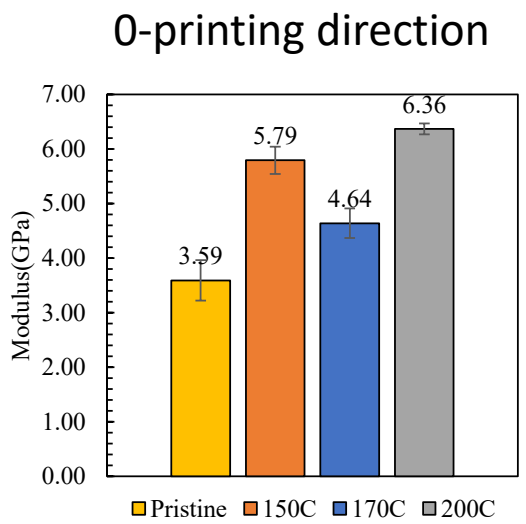

c.

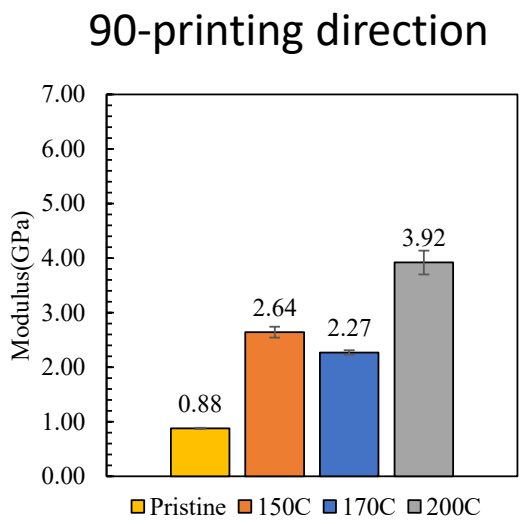


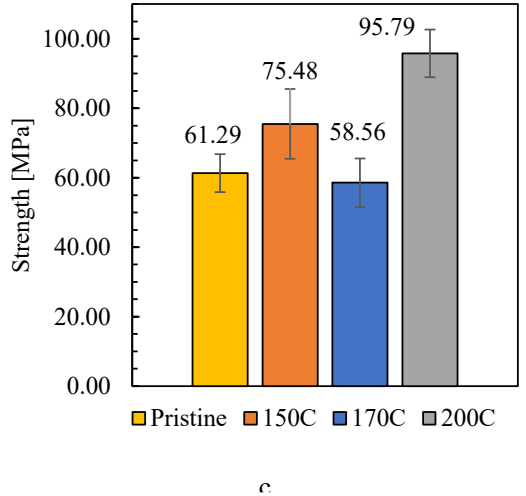

d.

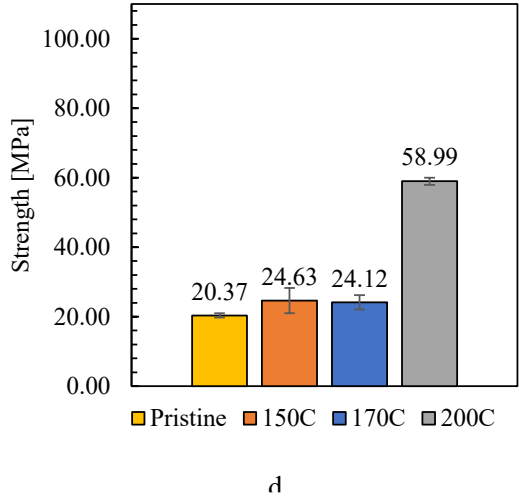
Role of annealing and isostatic compaction on mechanical properties of 3D printed short glass fiber nylon composites, PAK Jain et al. 2022

# Effect of Compaction: Mechanical Properties



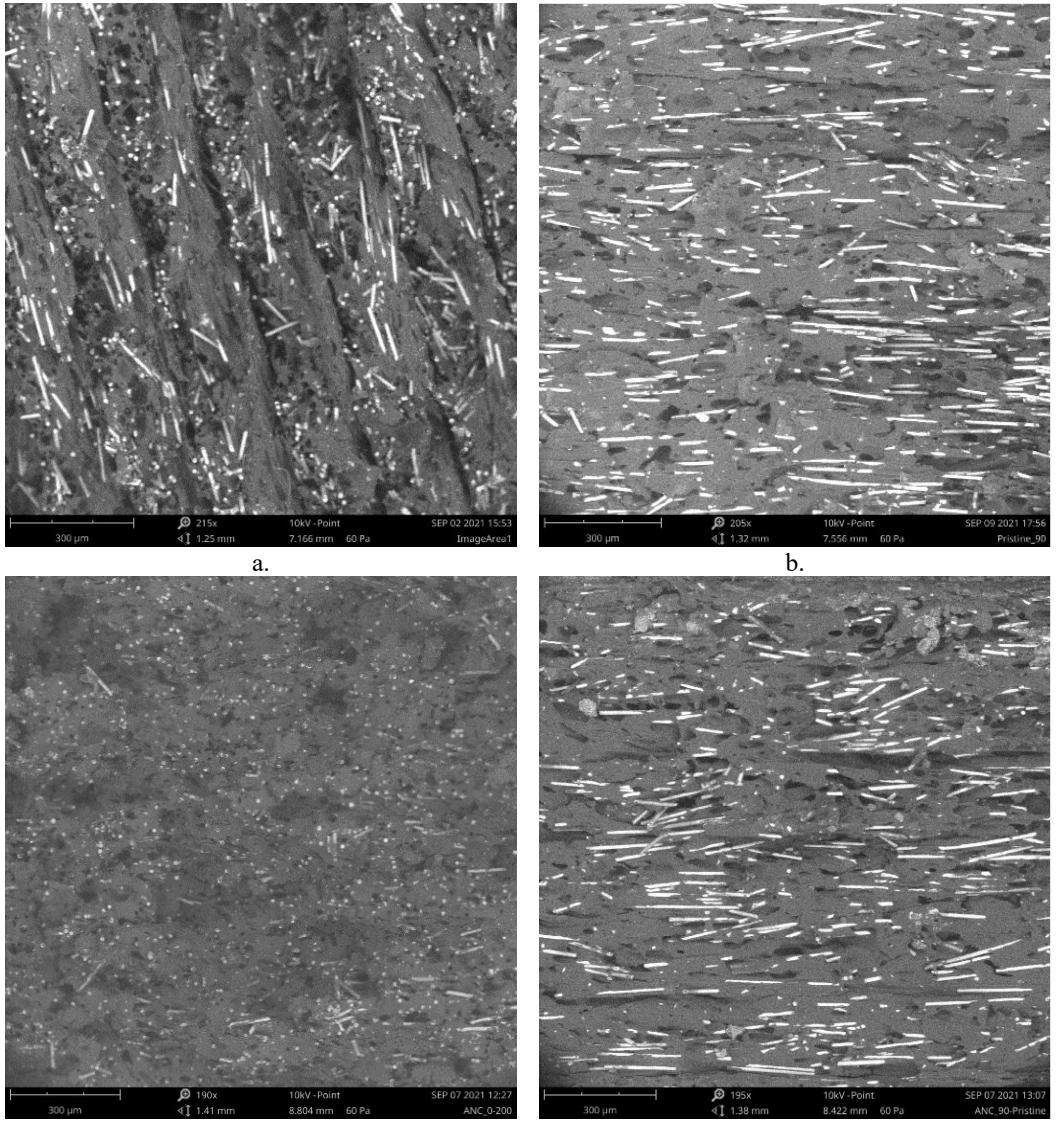



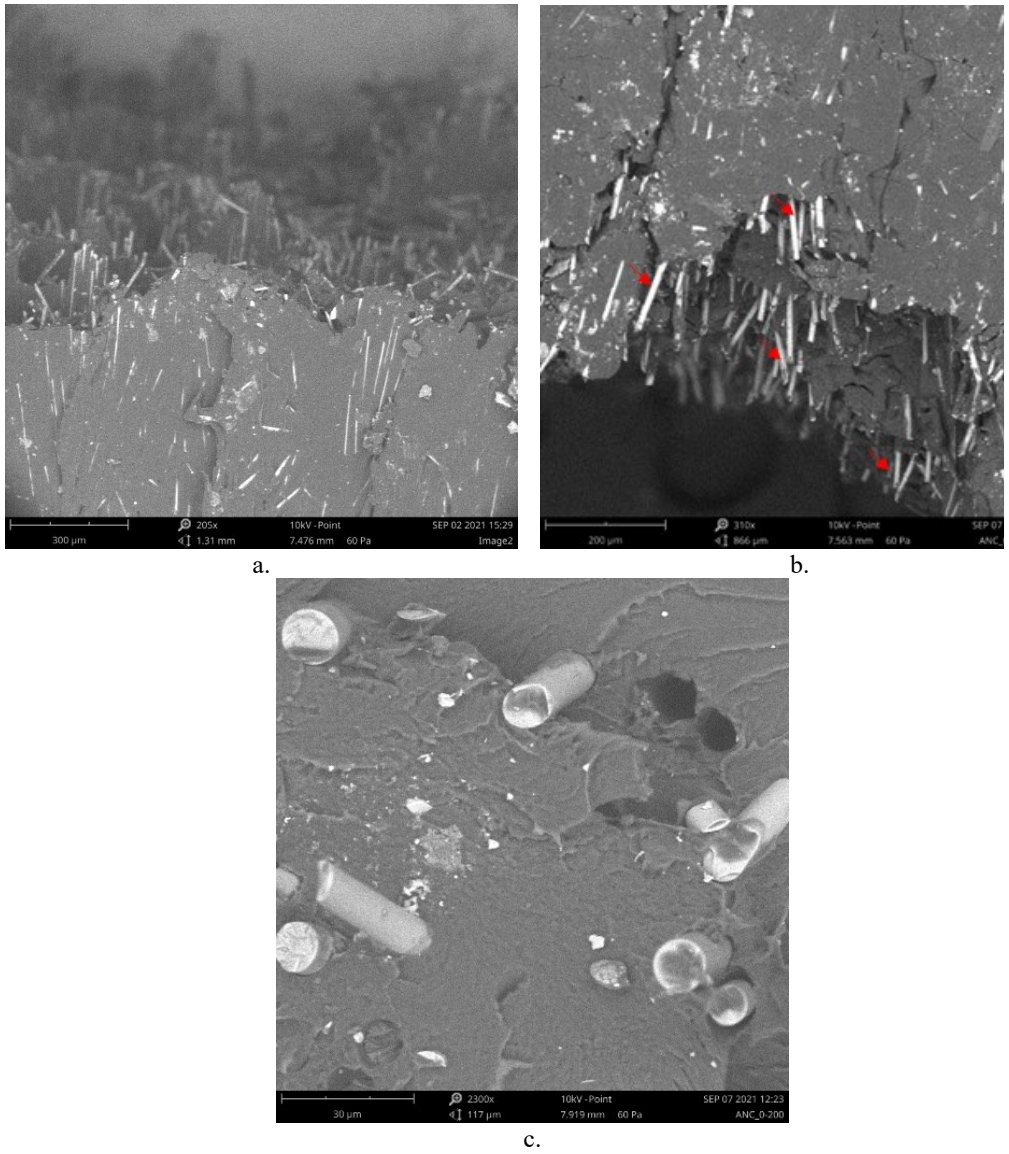

Role of annealing and isostatic compaction on mechanical properties of 3D printed short glass fiber nylon composites, PAK Jain et al. 2022

# Failure Surface



a. b. c. d.

**Figure 12**. Fracture surface of pristine 0º sample (a) and pristine 90º sample(b); Fracture surface of AN-C-200 ºC 0º sample (c) and AN-C-200 ºC 90º sample (d)



a. b. c.

**Figure 13**. Top view on the edge of the fracture surface for 0º pristine sample (a) and for 0º AN-C-200ºC sample (b-c). Arrows in (b) indicate fiber pullout

# Combining Long and Short Carbon Fiber using AM

COMPOSITES MODELING AND MANUFACTURING LAB

## Materials

- Polyamide 6 (PA6) matrix
- Short carbon fiber (Onyx)
  - PA6 with short carbon fiber
  - 15% CF by weight
- Long Carbon Fiber (CF)
  - Nylon 6 sizing

## Equipment

- Markforged Mark Two

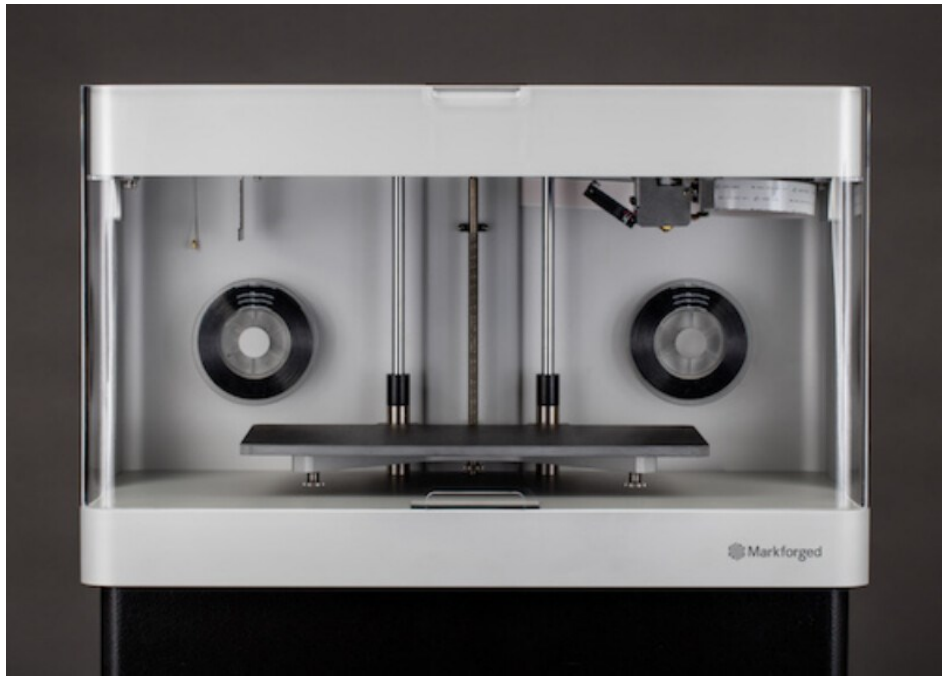
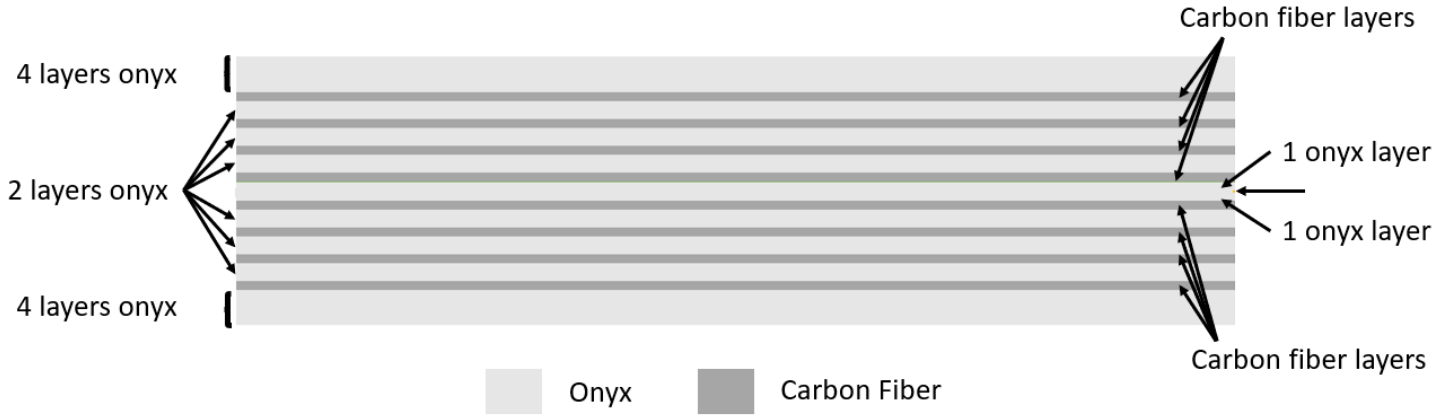



Image courtesy of Markforged

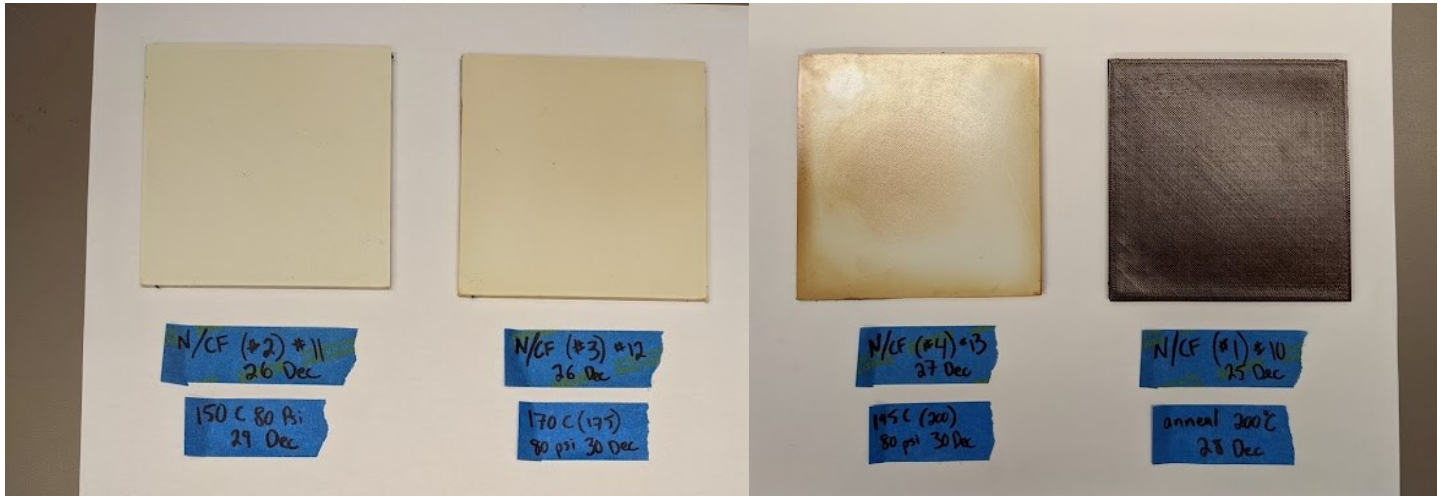
# Test Matrix




| Factor | Levels | | | | |
|---|---|---|---|---|---|
| Carbon Fiber Angle | Multidirectional | | Longitudinal (0°) | | Transverse (90°) |
| Treatment Pressure (psi) | 15 | | 80 | | 100 |
| Treatment Temperature (deg C) | 25 | 155 | 175 | 190 | 200 |

# Post Processing

- Annealing
- Pressure and temperature treatments
  - Preheat press to treatment temperature
  - Apply treatment pressure in 15 minutes increments
  - Cycle for a total of 4 hours
  - Remove from press and let cool

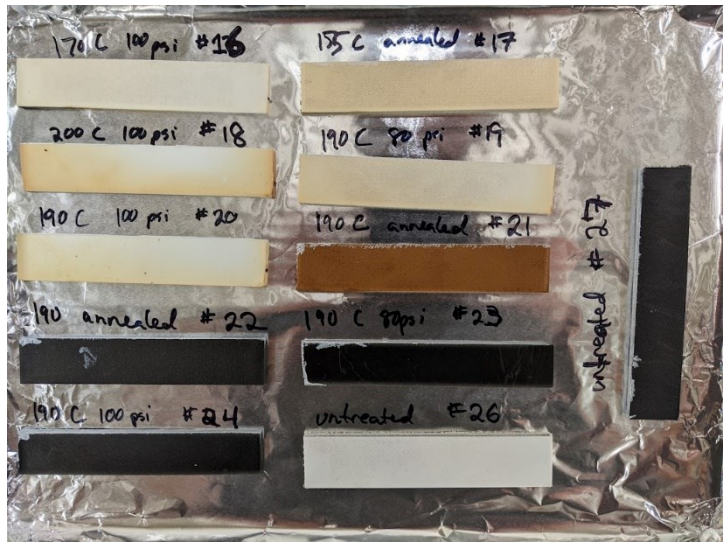


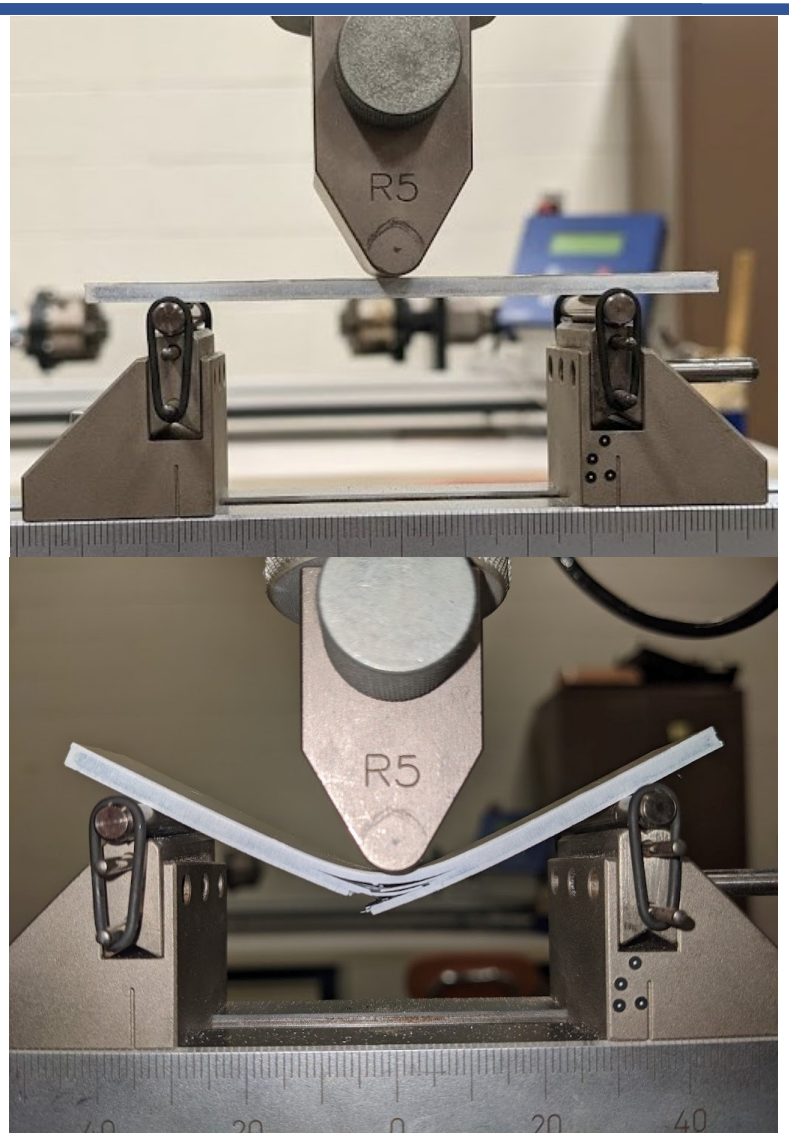
Treated nylon plaques

Hydraulic press

# 3 Point Bending

- Cut plaques into 5 samples each
- 5 Blocks, 1 block per replicate
- Ran a sensitivity study to determine test setup
  - 60 mm span
  - 4 mm/min rate
- Used Tinius Olsen S10 for 3 point bending



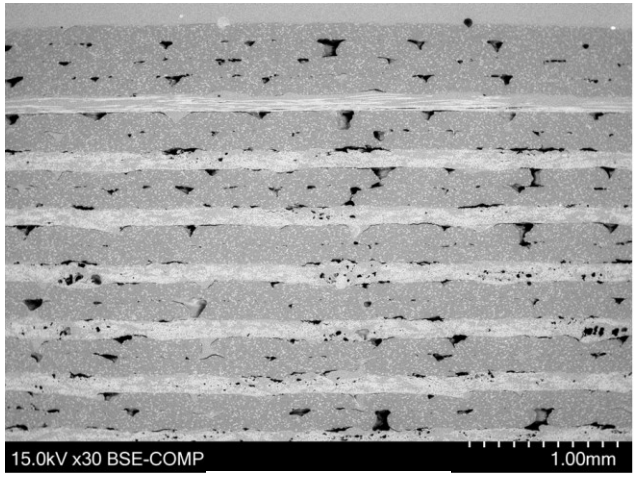



# Shrinkage: LCF single layer

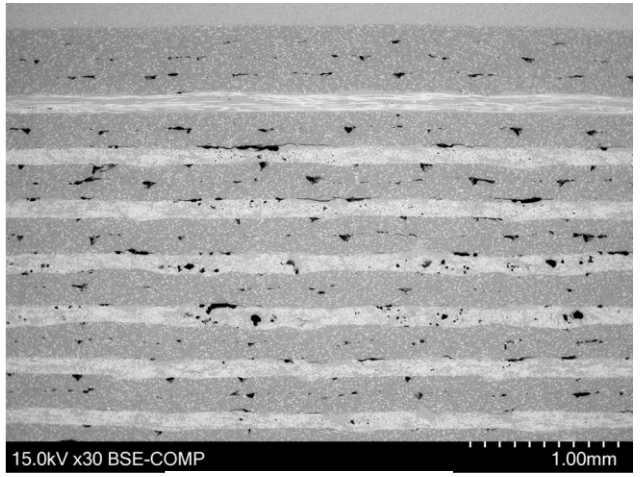
COMPOSITES MODELING AND MANUFACTURING LAB

| | 155 C | 175 C | 190 C |
|---|---|---|---|
| Pristine | | | |
| No pressure | | | |
| 80 psi | | | |

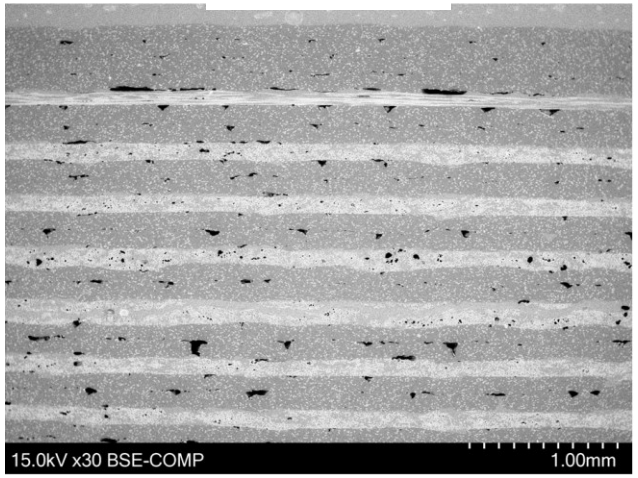
# Microstructure



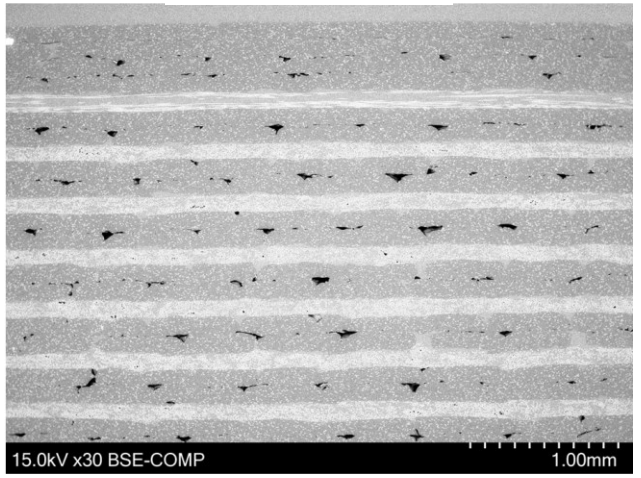

As-printed




155 C, 80 psi

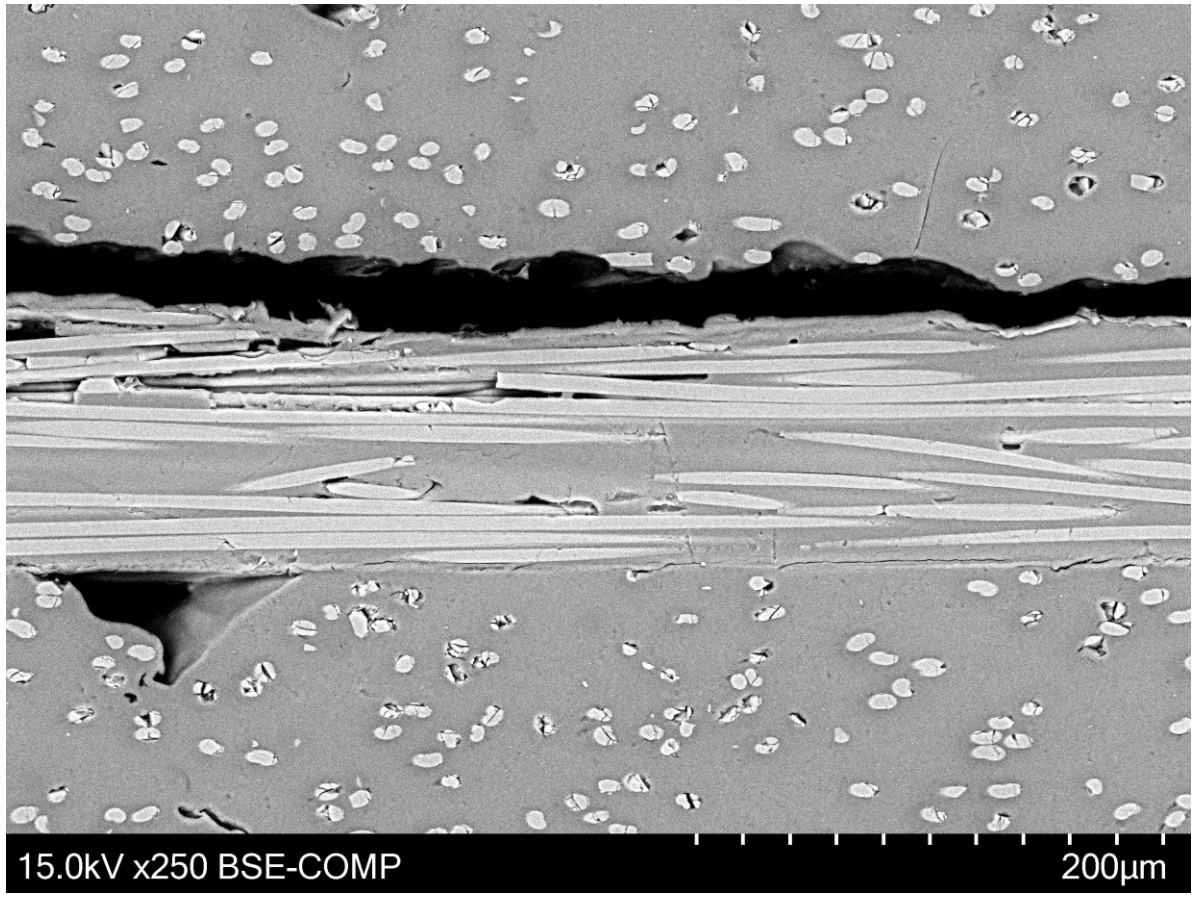



175 C, 80 psi

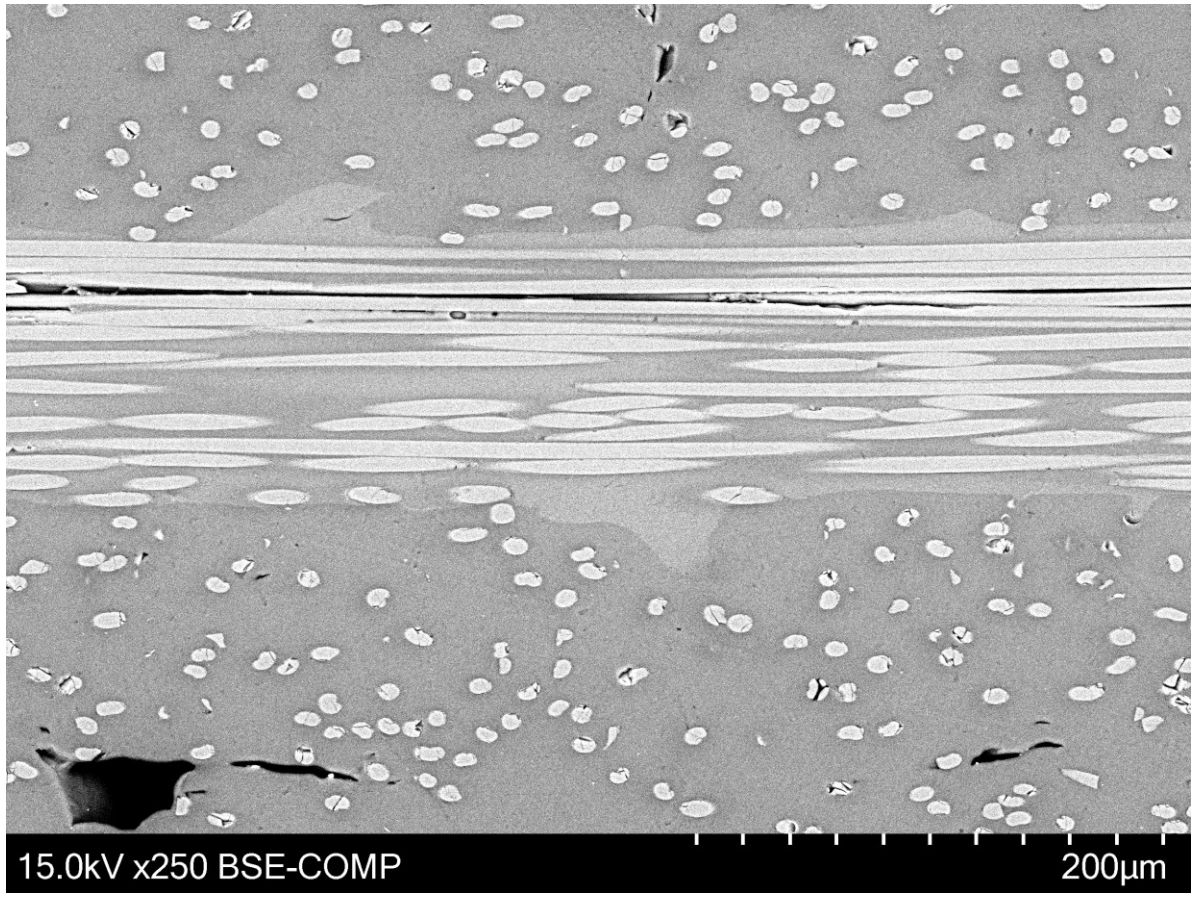



190 C, 80 psi

# Microstructure

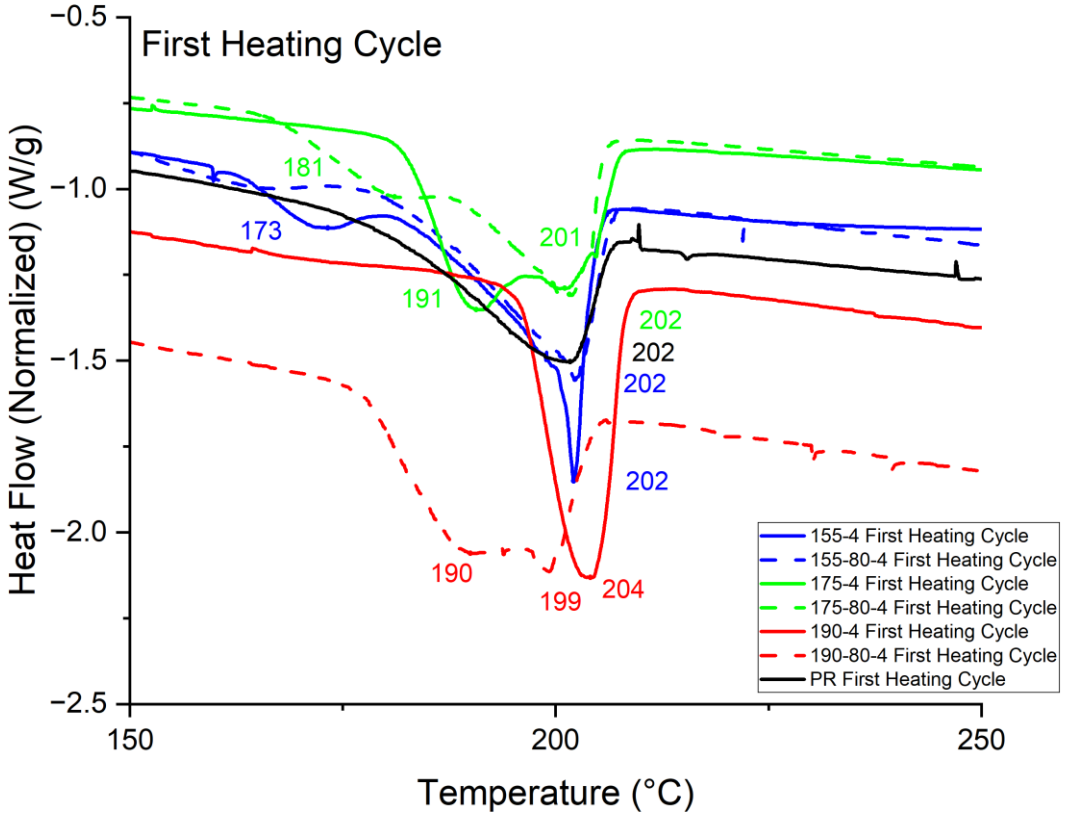



As printed



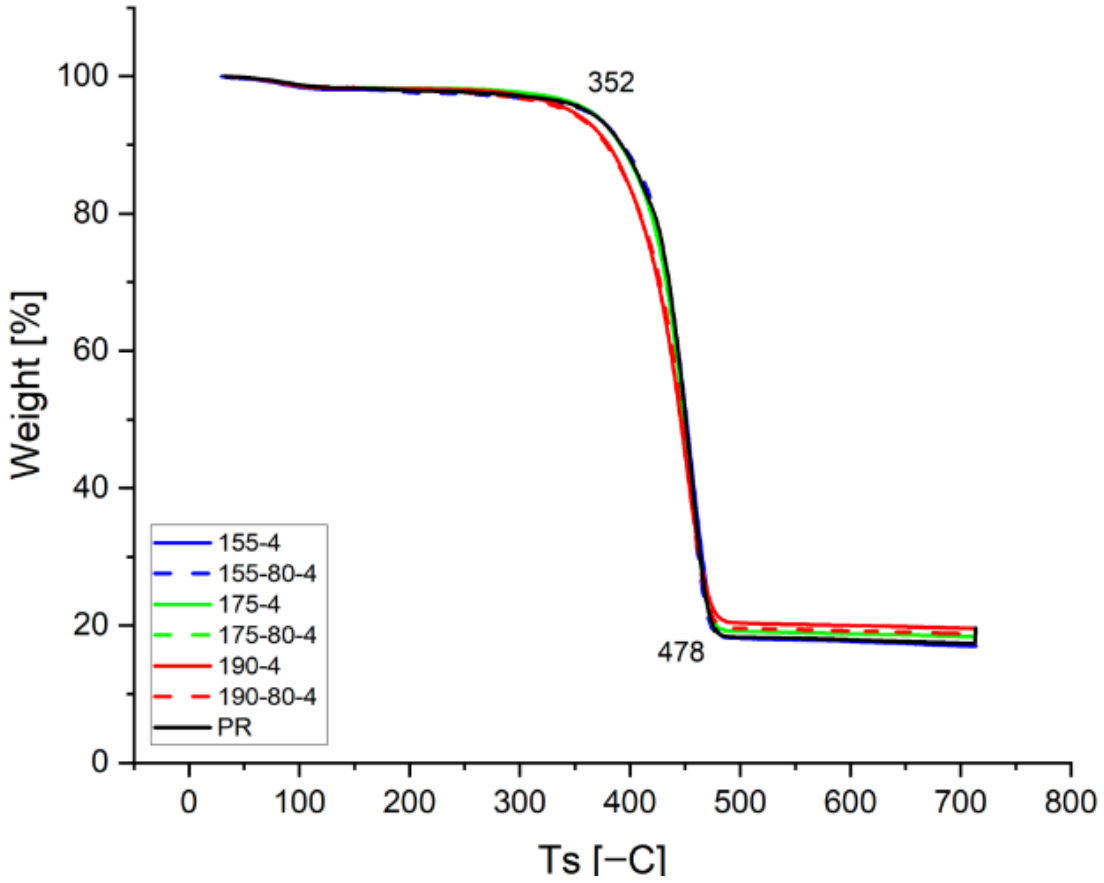

190 C, 80 psi

# DSC

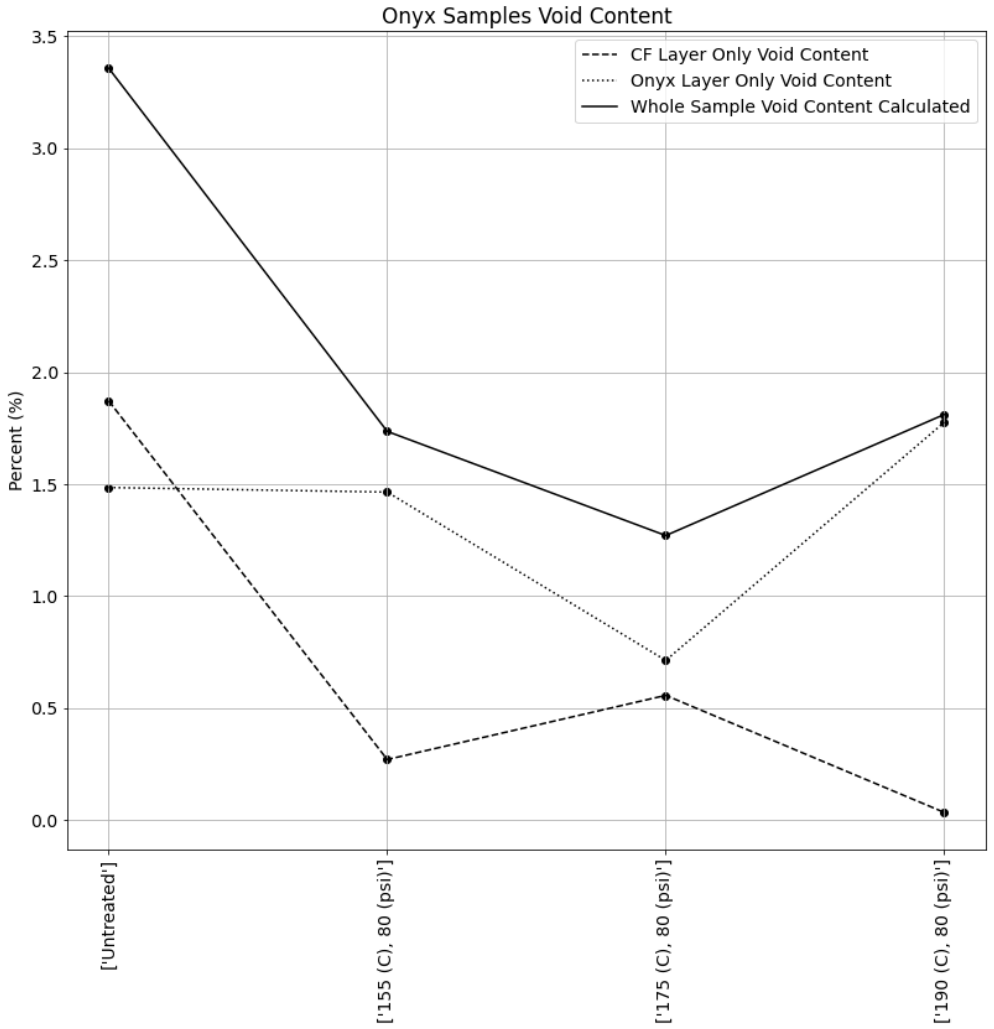



| | First Heating Ramp | |
|---|---|---|
| | Peak Melting Temperature (C°) | Peak Melting Temperature (C°) |
| PR | 202 | |
| 155/4 | 173 | 202 |
| 155/80/4 | | 202 |
| 175/4 | 191 | 201 |
| 175/80/4 | 181 | 202 |
| 190/4 | 204 | |
| 190/80/4 | 190 | 199 |

# TGA


100
80
60
40
20
0
Weight [%]
0
100
200
300
400
500
600
700
800
Ts [−C]
352
478
155-4
155-80-4
175-4
175-80-4
190-4
190-80-4
PR

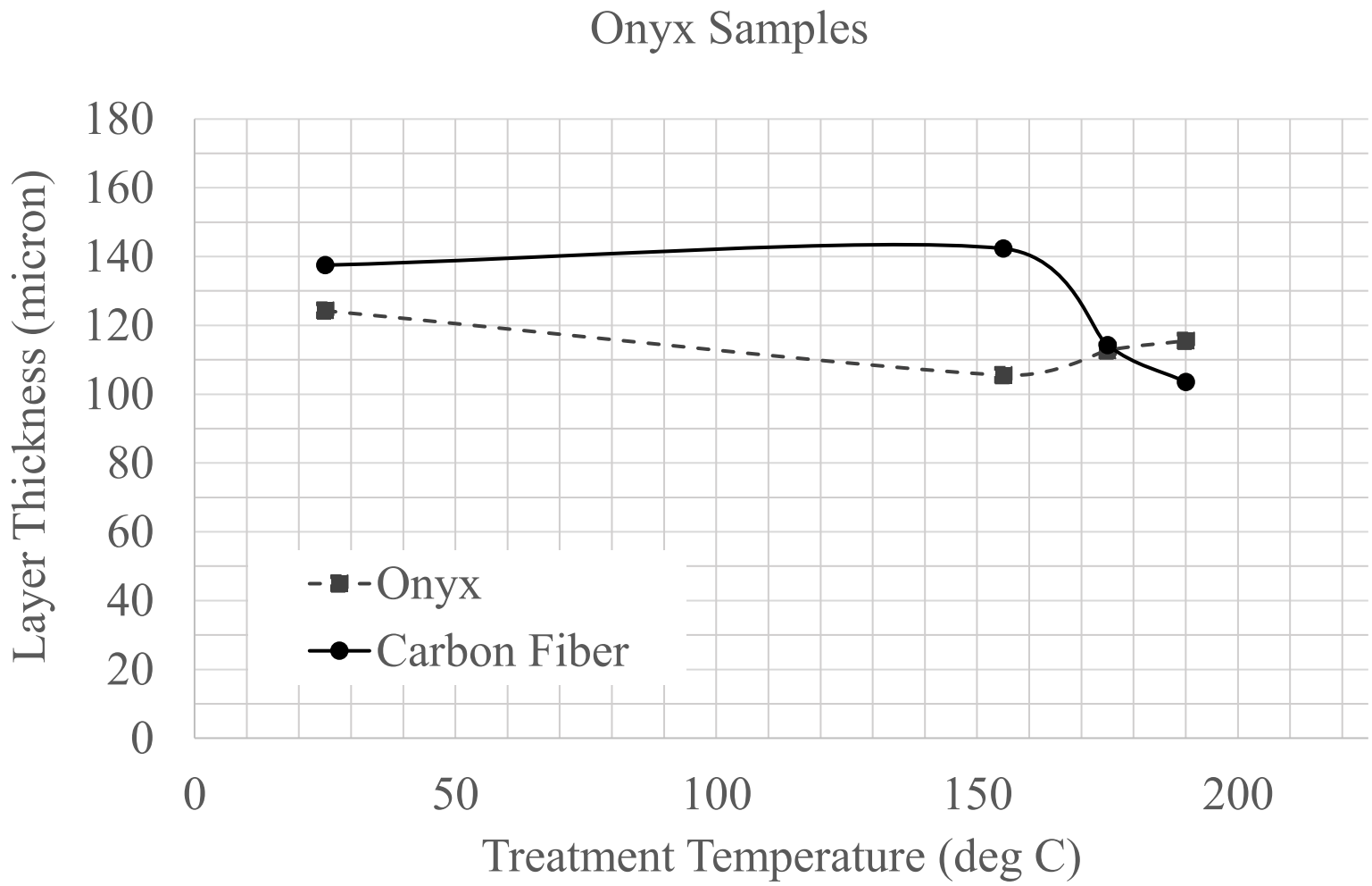
# SEM Void Content and Compaction Strain



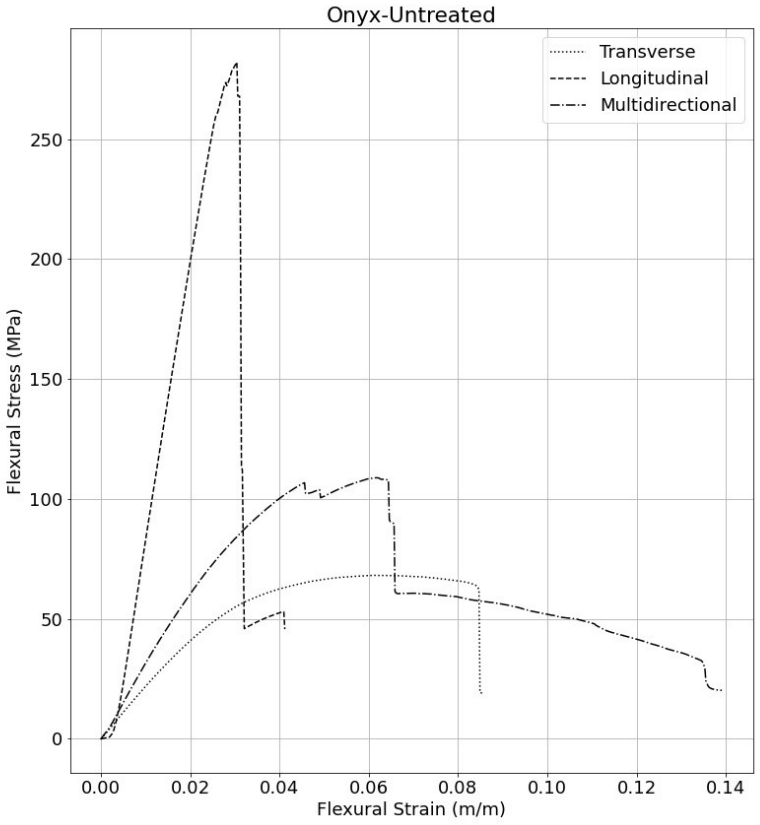

Multidirectional

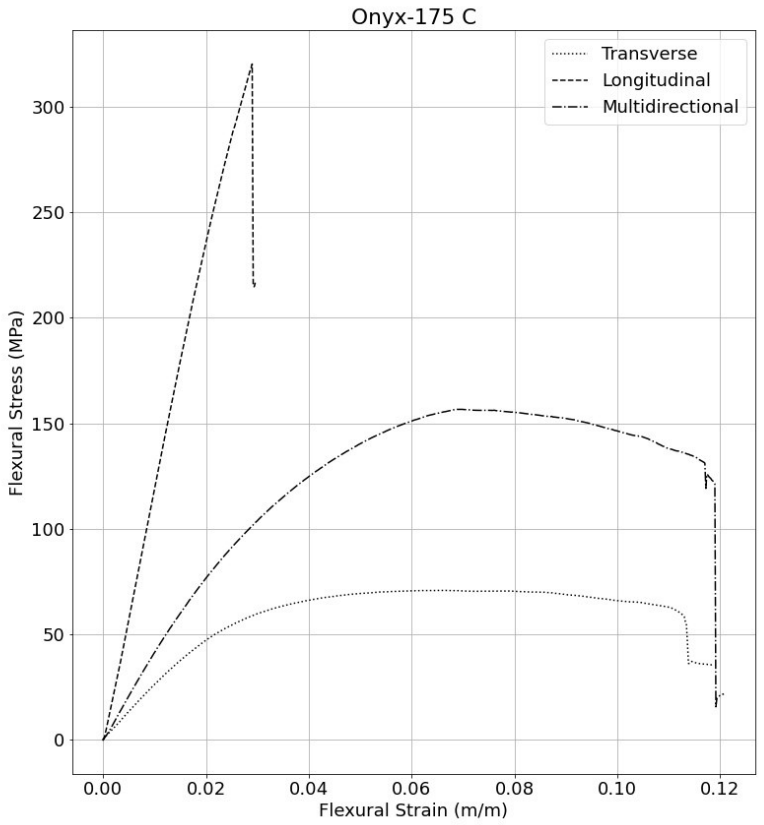
| | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Temp (deg C) | **155** | **155** | **155** | 175 | 175 | 175 | **190** | **190** | **190** | 200 | 200 | 200 | 25 |
| | Pressure (psi) | **15** | **80** | **100** | 15 | 80 | 100 | **15** | **80** | **100** | 15 | 80 | 100 | 15 |
| Comp. (%) | Onyx | **109** | **95** | **92** | 139* | 89 | 91 | **97** | **88** | **88** | 148* | 70* | 72* | 100 |

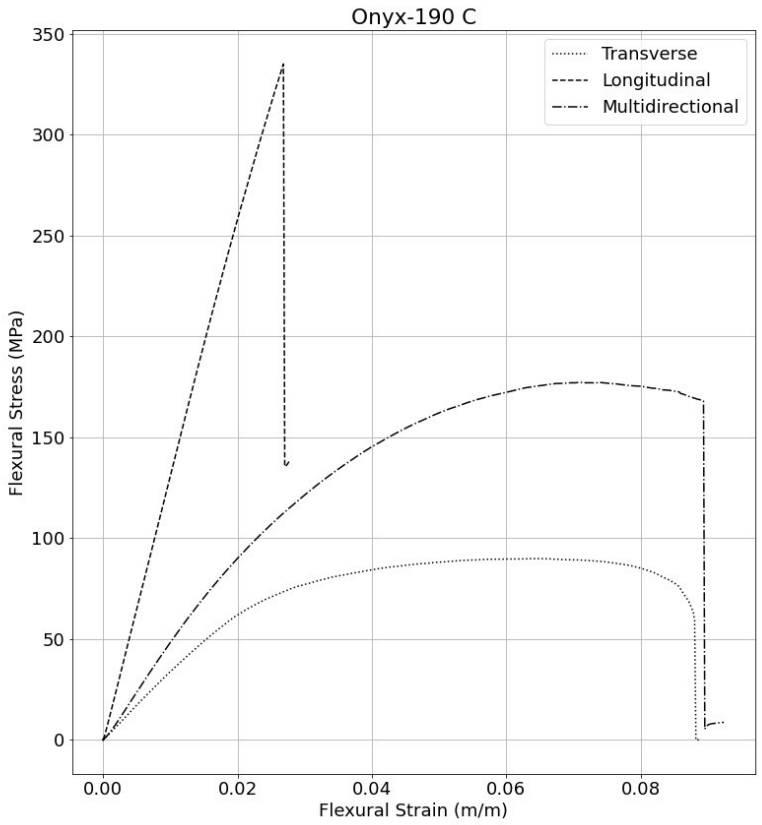
# SEM Layer Thickness - Onyx



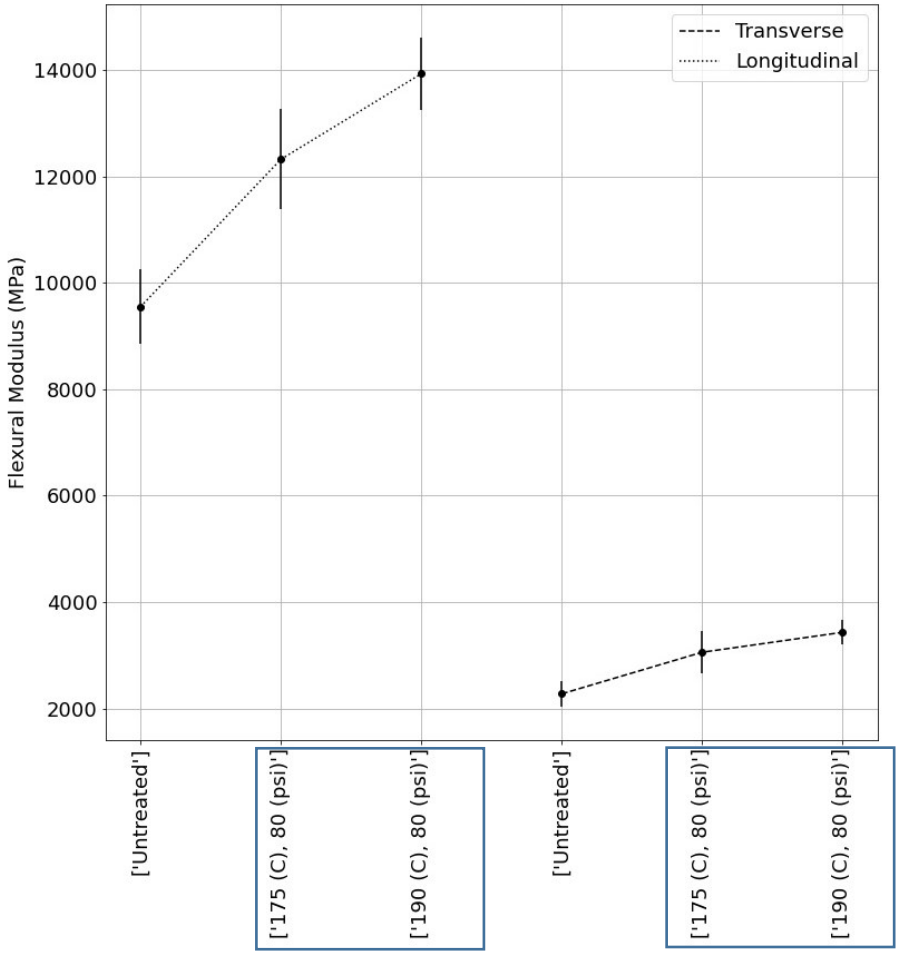
# Flexural Stress/Strain Data

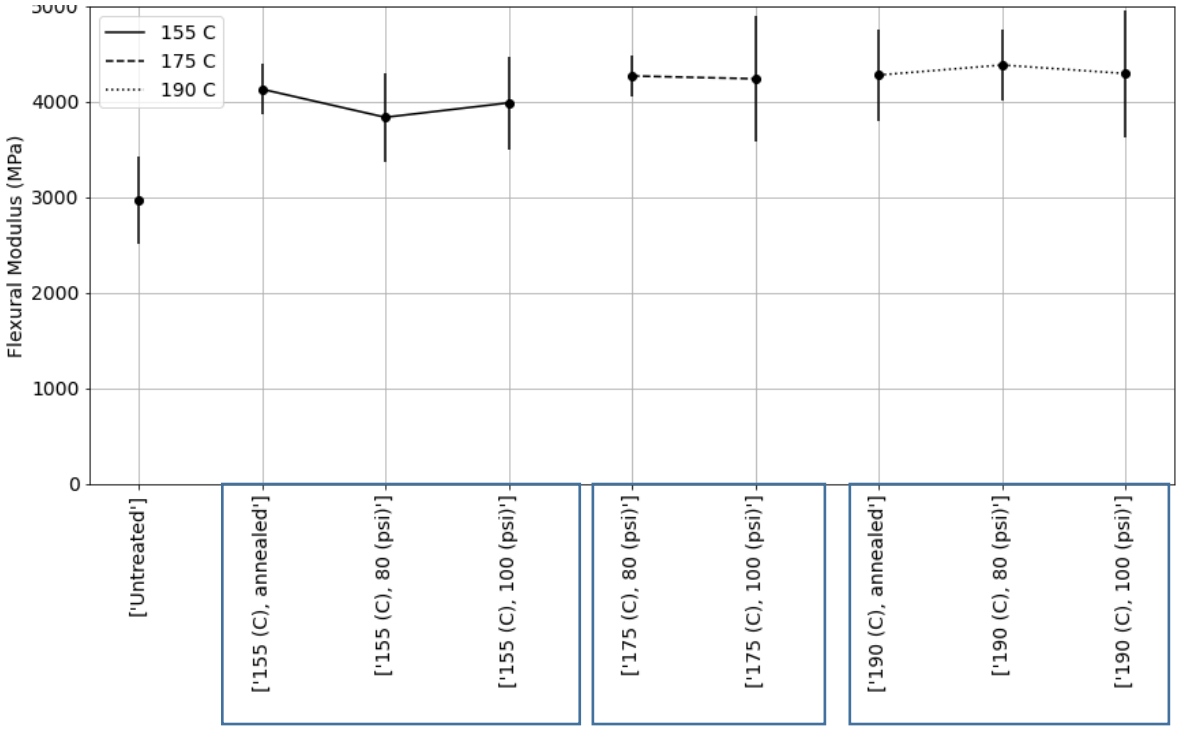


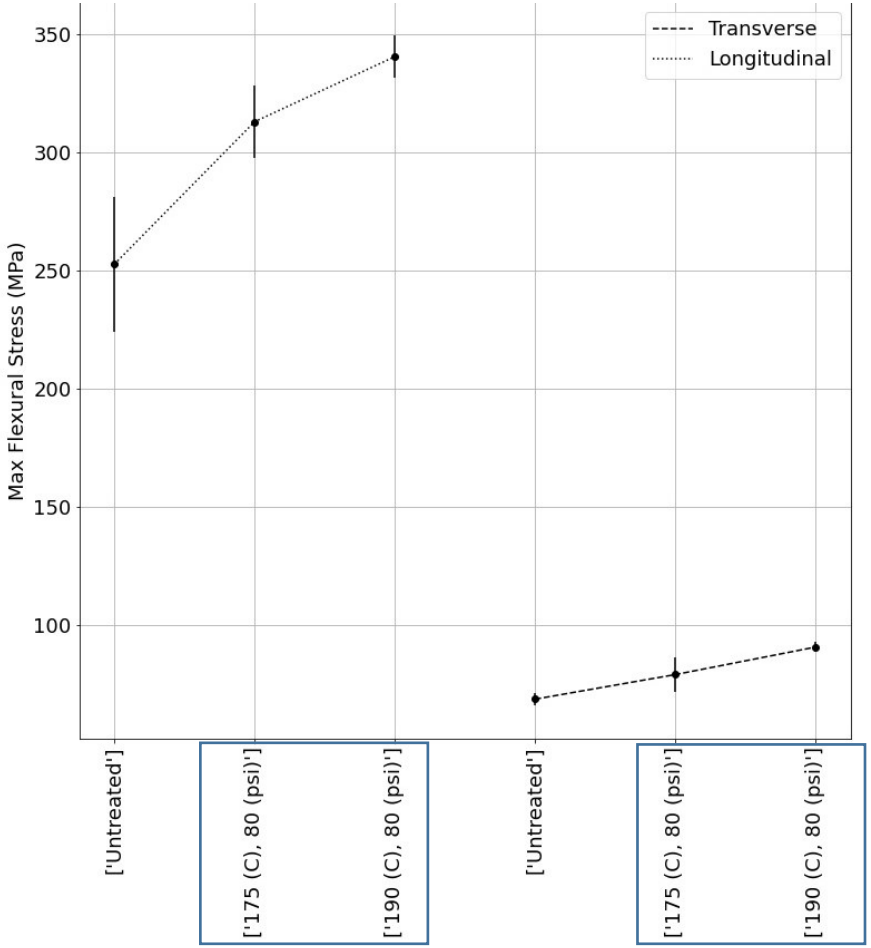



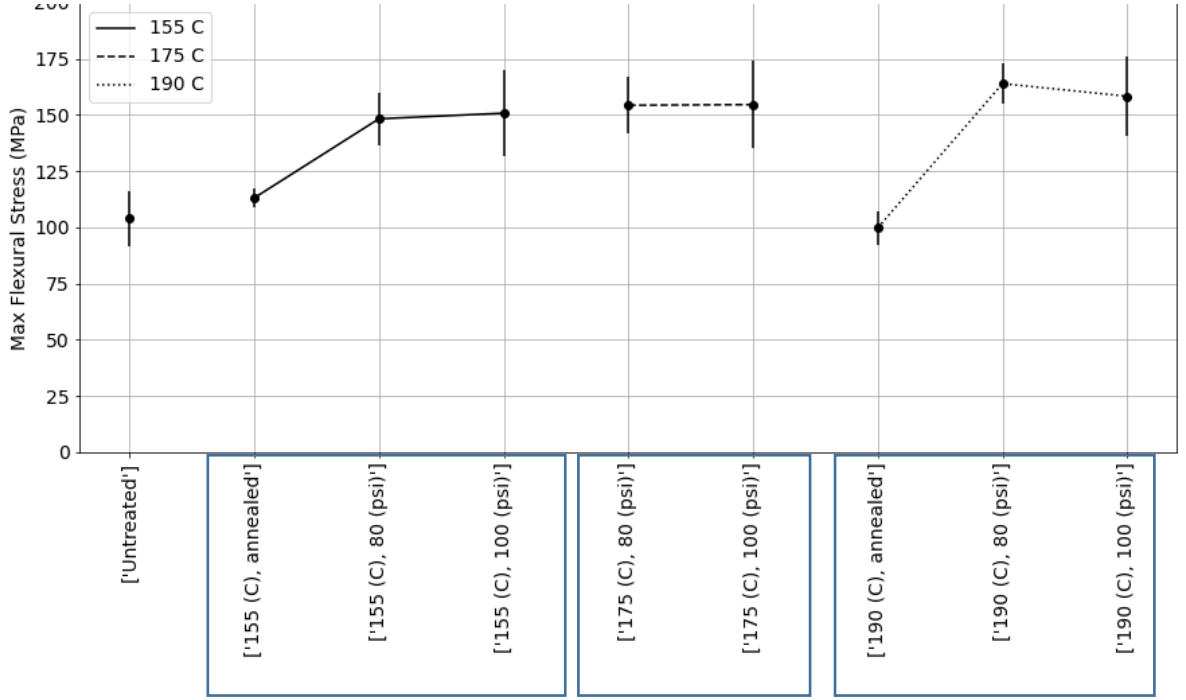
# Flexural Modulus



# Flexural Strength



# Conclusions

- Mechanical properties of nylon short and long glass fiber can be improved by as high as 50%
- Dimensional stability of iso-statically compacted part can be preserved when pressure and temperature conditions are balanced
- Changes occurring in the material have both micro- and molecular scale

**Acknowledgement**