

DIMENSIONAL STABILITY OF LOW-COST THERMOPLASTIC COMPOSITE MOLDS



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FINAL TECHNICAL REPORT

DIMENSIONAL STABILITY OF LOW-COST THERMOPLASTIC COMPOSITE MOLDS

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1. LISTS

1.1 List of Acronyms

AM	additive manufacturing
EDAM	extrusion deposition additive manufacturing
CF	carbon fibers
CLTE	coefficient of thermal expansion
FDM	fused deposition modeling
FOD	fiber orientation distribution
PEI	polyetherimide
ULTEM	brand name given to polyetherimide by SABIC company

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2. EXECUTIVE SUMMARY

This report addresses a key question for using additively manufactured tooling from carbon fiber reinforced thermoplastic composites. How does the distortion of the molded part change with the print orientation used in the manufacture of tooling? The objective of this research was to discuss the distortion of parts molded with such tooling, in relation to the deflections in the tooling during the compression molding process. This was achieved through both model simulations and experimental molding runs. A vertical orientation was used in printing the mold with a distinctly non-planar surface: this orientation allowed for higher performance along the press closing direction -- higher thermal conductivity, higher stiffness, and lower coefficient of thermal expansion. Epoxy-carbon fiber twill weave fabric prepreg was then compression molded in this mold. The thermomechanical properties of the tooling material and the molded part were measured and used in simulation of mold deformation as well as part distortion. The thermomechanical anisotropy of the mold is quite different from that of the molded part because in the mold, the stiffest direction is the z-axis, while in the molded thermoset part, the z-axis is the weakest direction. The connection between mold deformation during the compression molding process and the final part distortion can be seen from the simulation results. The simulation results for the case where the mold was four times as stiff as the part along the press closing direction compared well with experiment. When the mold stiffness was lowered in relation to the part stiffness, the mold deformation during the compression molding increased; but this led to a smaller extent of part distortion.

3. INTRODUCTION

Development of lightweight, lower-cost tooling that is additively manufactured from thermoplastic composites, to replace metal molds, has great potential in the automotive and aerospace industries [1, 2]. The use of chopped carbon fiber reinforced thermoplastics and the development of high speed extrusion deposition machines has made this more promising [3-5]. Still, the use of such tooling has some potential problems and unknowns: a key question is how the use of such tooling changes the distortion of molded parts. The objective of this research project was to investigate the distortion of parts molded with such tooling, in relation to the deflections in the tooling during the compression molding process. This was done by comparing results of model simulations and experimental molding runs.

4. BACKGROUND

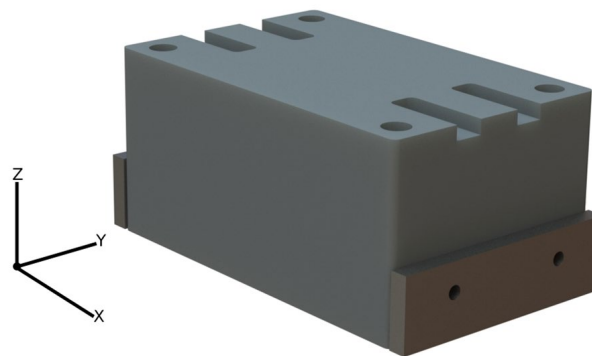
One drawback to use of tooling made from thermoplastic composites is the difficulty in uniform heating of the mold because of the highly anisotropic thermal conductivity of the mold material. Dinwiddie et al. [6] have reported that the thermal conductivity values of chopped carbon fiber reinforced thermoplastics are in the proportion 4:2:1 along the print direction, the cross or lateral direction, and the stacking direction. The difficulty with heating such molds was documented in a recent study reported by Bogdanor et al. [7]; in this work, they printed a mostly planar mold from polyether-sulfone reinforced with 25

wt% carbon fibers, using a print direction aligned with the mold surface. The thermal conductivity in this case was particularly poor along the press closing direction, making for low heat transfer rates from the press platens to the mold; hence cartridge heaters were inserted within the mold by Bogdanor et al. [7] to maintain the temperature in the mold. Another potential drawback of using such tooling is that the anisotropy of stiffness and strength of the mold material, arising from preferential fiber orientation along the print direction during the deposition process, may lead to excessive deflection of the mold while molding thermosetting composites. This will in turn affect the final distortion of the molded part. The print orientation was designed to be uniformly vertical, aligned with the press closing direction in both mold halves of the present study.

5. RESULTS AND DISCUSSION

3D-printing of tool

A block of polyetherimide reinforced with 20 wt% chopped carbon fibers was manufactured by extrusion deposition additive manufacturing (EDAM) with the fibers oriented uniformly along one direction. This was carried out with a BAAM machine using a bead width of 0.5 in. and a layer thickness of 0.15 in. Mating mold halves were then machined from this block so that fiber orientation throughout the mold was along the closing direction of a press; this allowed for higher performance along the press closing direction -- higher thermal conductivity, higher stiffness as well as lower coefficient of thermal expansion (CTE) to be obtained along the press direction. Cartridge heaters were found to be inconvenient in our configuration since the lower in-plane conductivity of the mold could fuse the thermoplastic locally around cartridge heaters. The use of blocks with uniform orientation was also found to be useful for consistent repair of the mold where required. In the event of local damage, the damaged area would be cut out and a printed block with the same orientation would be inserted and fused together.



All print paths go along the z-direction so that they align vertically in the press closing direction for both mold halves

Figure 1. The complete mold assembly machined from a 3-D printed block

Part design

The molded part shape was chosen to be distinctly non-planar with two inclined faces on either side of a flat as shown in Figure 2a; the designed angle of incline was 135 degrees on either side. This choice was made to heighten the sensitivity of molded part distortion to the anisotropic thermomechanical properties of the mold and the part, consistent with the objective of the paper. The molded part was 200 mm long and 90 mm wide without any bounding walls along the width or y-direction. The top half mold or punch is displayed in Figure 2b and the bottom half mold or die is displayed in Figure 2c. The mold halves are machined to allow a uniform part thickness of 4.1 mm and have matching post holes in the corners. The lower mold half was reinforced with metal sides on either side of the length (see Figure 1) before mounting in the press.

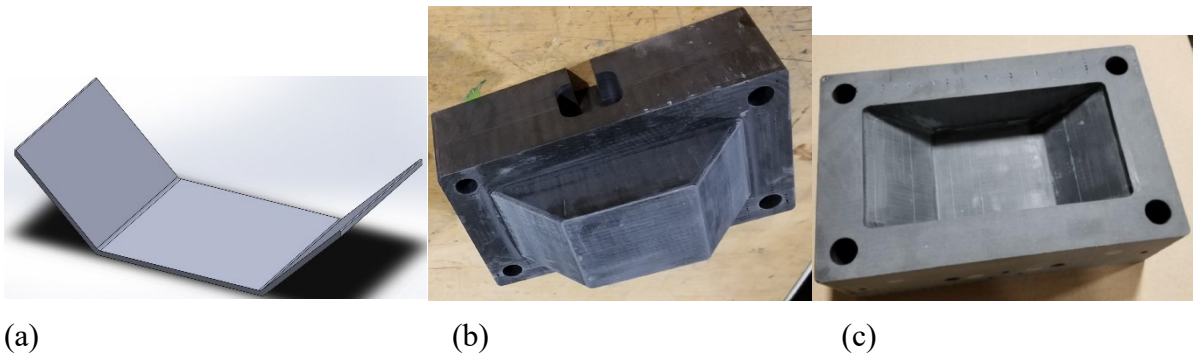


Figure 2. (a) the part geometry; (b) the male mold or punch and (c) the female mold or die

Prepreg layup and compression molding

The carbon fabric used here was a twill weave with a weight of 670 g/m² dry and 1124 g/m² as prepreg; the fiber volume fraction was 0.58. The average ply thickness of the prepreg was 0.8 mm and the gap thickness in the mold was 4.1 mm. Five plies of the twill weave carbon-fabric/epoxy prepreg were stacked in the following sequence with mold release films above the top and below the bottom as shown in Figure 3. This stacking will lead to the lowest modulus along the z-direction for the molded composite part.

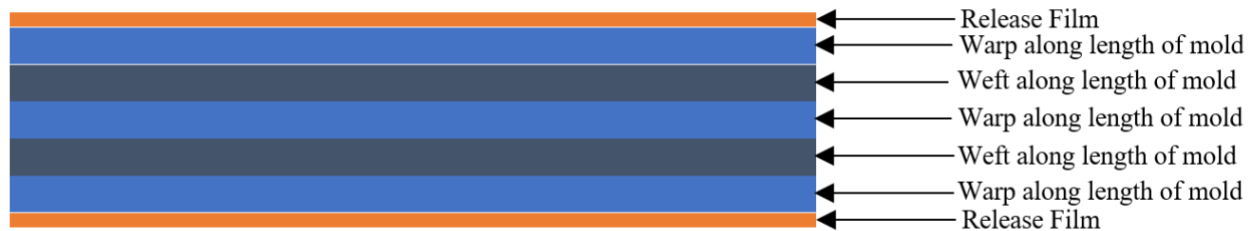


Figure 3. Prepreg layup of fabric

The thermoplastic composite tool was preheated to 121°C in a portable oven located next to the press before mounting in the press with heated platens—the top platen at 143°C and the

bottom platen at 149°C. The prepreg was compacted with a pressure of 0.27 MPa (40 psi); the fabric being inextensible. The press opened when the mold cooled to 104°C. The epoxy composite part stayed attached to the lower half mold until it cooled to room temperature, 23°C.

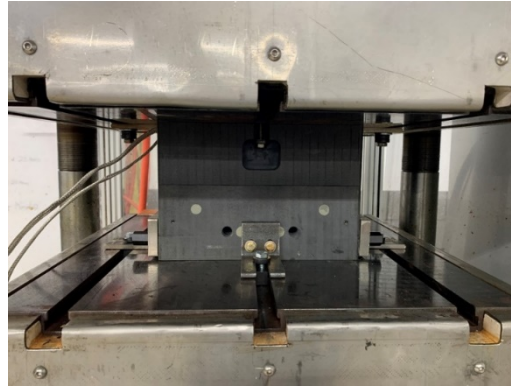


Figure 4. The smaller and lighter 3D-printed composite tool mounted in the press

Model Simulations

The molding process was simulated with a finite element software package to predict distortion at various stages of the process in the mold and in the molded part. The simulation was carried out in three distinct stages: the cure stage, where the part is in contact with both mold halves, followed by a cooling stage where the part is in contact only with the bottom mold half and finally the cooling stage where the part is fully demolded. Thermal gradients are generated in the composite mold as well as the curing part during the process. Anisotropic thermal conductivity values were specified for both the composite mold and the part being molded, along with convective heat transfer coefficients at interfaces: 20 W/m²K between the mold surface and the part and 10 W/m²K between the mold surface and air.

Molded Part Results

Distortion of molded part

The profile of the molded part is shown in Figure 5. The flat edge of the part was curved slightly inwards to a concave down profile.

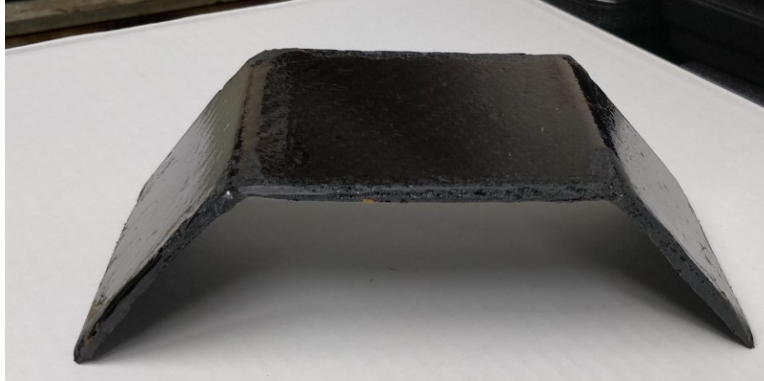


Figure 5: Profile of molded part

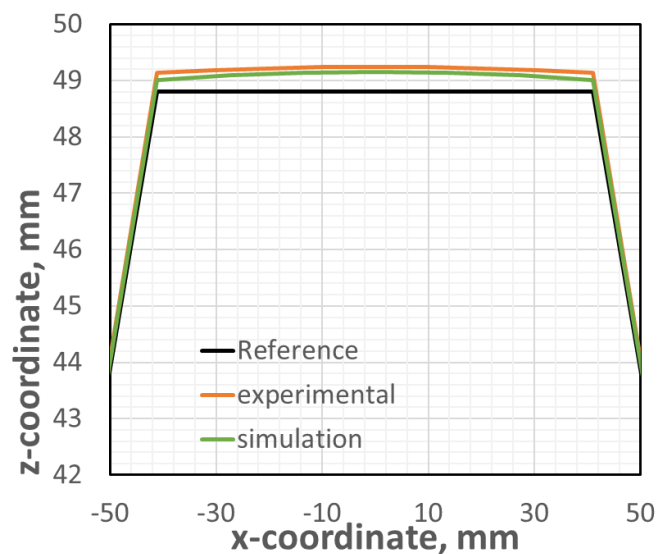


Figure 6: z-displacement profile along the length of the molded part from experiment

The displacements along the z-axis from the reference part design were recorded along the length of the part and have been presented in Figure 6. A slight curvature was noted: the maximum deviation was recorded at the center with a z-displacement value of 0.45 mm. The angle measured on the demolded part was pushed in by 2.5° to 132.5°.

Simulation results

Thermal gradients

The mold and the part are anisotropic and therefore experience uneven heating and cooling during the 3 stages of simulation. This results in the development of an evolving temperature profile within the composite part and mold. These thermal gradients lead to development of thermal stresses that ultimately lead to displacements and distortion of the part.

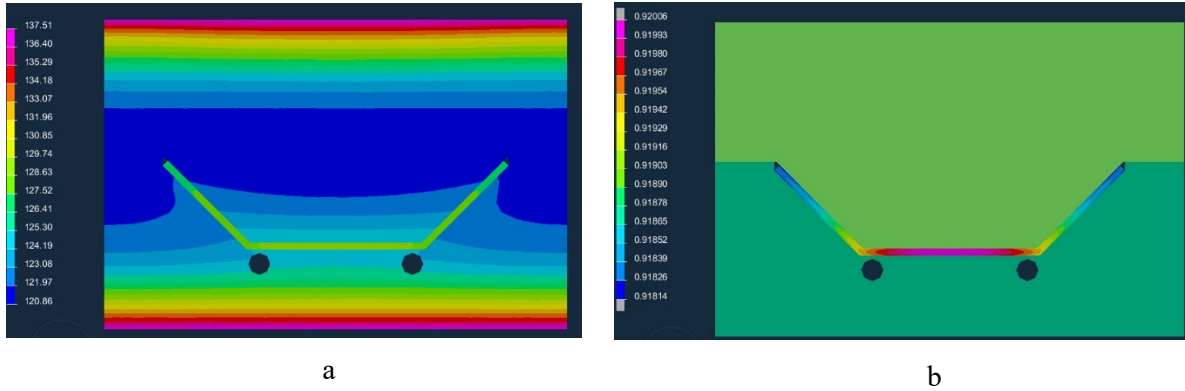


Figure 7. (a) Temperature profiles in mold and part and (b) cure profile in part, at end of cure.

Mold deformation

The thermomechanical anisotropy of the mold is quite different from that of the molded part because in the mold, the stiffest direction is the z-axis, while in the molded thermoset part, the z-axis is the weakest direction. This may be seen from the tensile moduli and the coefficients of thermal expansion presents in Table 1. The coefficients of thermal expansion were obtained from measurements carried out on samples cut from plaques of the polyimide/CF composite that were 3d printed under conditions similar to the mold.

Table 1. Thermomechanical properties for molded part and composite mold

Property	Part (glassy state)	Composite mold
Tensile Modulus E_{xx}	55.8 GPa	4.1 GPa
Tensile Modulus E_{yy}	55.8 GPa	4.1 GPa
Tensile Modulus E_{zz}	8.1 GPa	32.4 GPa
CLTE α_x	$2.9 \times 10^{-6} \text{ K}^{-1}$	$27 \times 10^{-6} \text{ K}^{-1}$
CLTE α_y	$2.9 \times 10^{-6} \text{ K}^{-1}$	$27 \times 10^{-6} \text{ K}^{-1}$
CLTE α_z	$44 \times 10^{-6} \text{ K}^{-1}$	$3.4 \times 10^{-6} \text{ K}^{-1}$

As the part cures and strengthens, the deflection of the mold will increase; the magnitude of this deflection will depend on the comparison of the mold stiffness with the part stiffness along the z-axis. This ratio is 4:1 with the experimental configuration described above. The simulation case with this ratio has been labeled Case 1.

The sensitivity of simulated results to this ratio was examined by running another simulation labeled Case 2 with a different value of 1.6:1 for the ratio, using $E_{zz}=13.1 \text{ GPa}$, and a correspondingly changed value of $\text{CLTE}=11.4 \times 10^{-6}$, along the z-direction for Case 2. The deformation of the punch or male section of the mold at the end of cure has been presented in Figure 8 for the two cases.

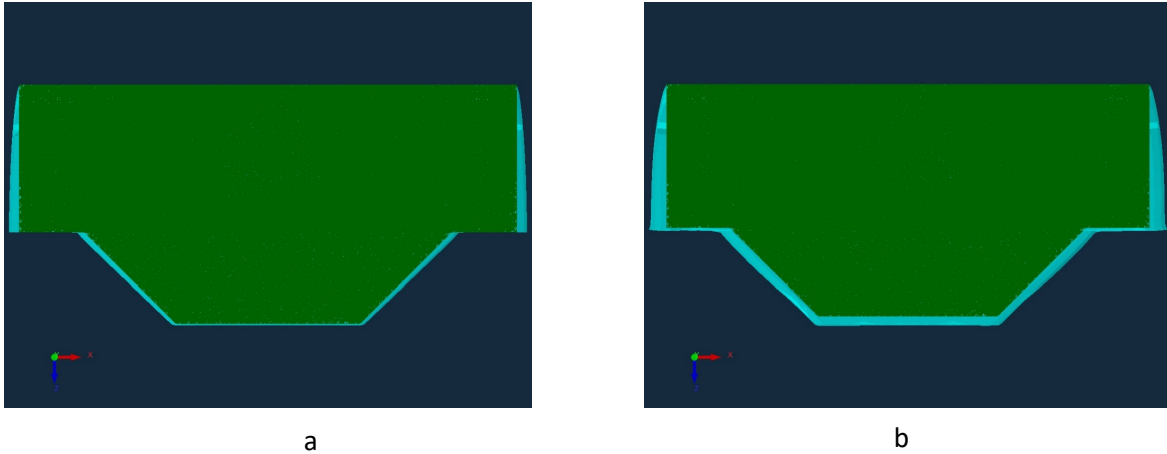


Figure 8. Deformed shape of top mold half or the punch in xz cross section in contact with the part (a) case 1 and (b) case 2. The reference shape appears in dark green and the deformed outline in light blue.

Part distortion after demolding

In the first cooling stage, the part is constrained by the mold; but in the second cooling stage, the part is demolded from the beginning. It was observed that the part deformation was less in case 2 when compared to case 1. Figure 9 displays the z-displacement (with 5X magnification) along the length of the part for the two cases. The angle is pushed in for both cases: by 1.9° in case 1 and by 0.67° in case 2.

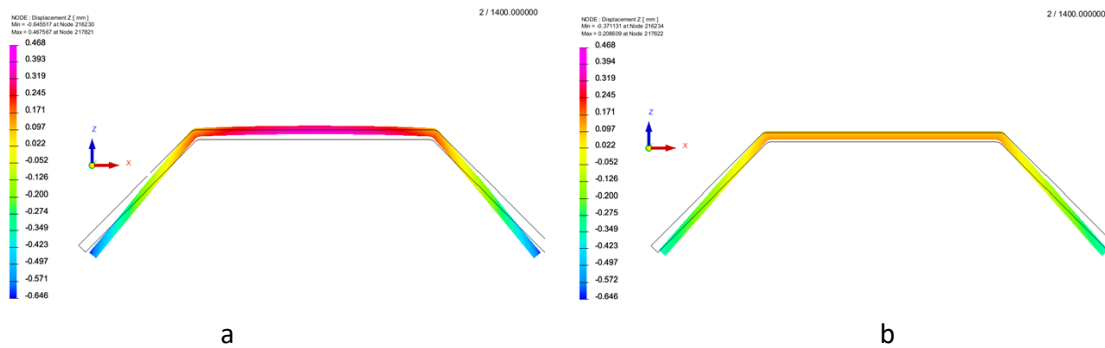


Figure 9. Simulated z - displacement in the part: x-z cross section (a) for Case 1 and (b) for Case 2. The displacement is magnified 5X in the figure.

The arc around the flat portion is also lower for Case 2 and this is presented in detail in Figure 10.

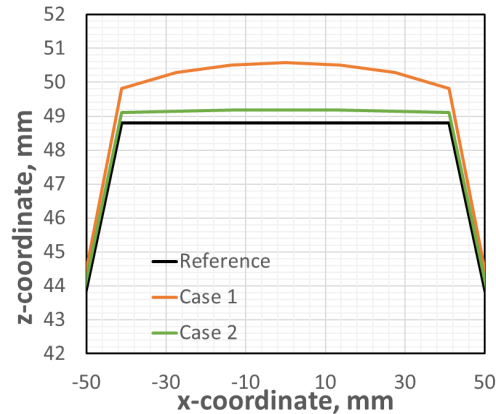


Figure 10: Simulated profile of demolded part in x-z plane with 5X magnified displacement.

6. BENEFITS ASSESSMENT

Additively manufactured composite tooling is much lighter than steel tooling and does not require cranes to mount and dismount the mold from the press. Heating and maintaining the temperature in the mold is easier if the carbon fiber orientation is uniformly along the press closing direction. This feature also allows convenient repair of the mold with 3d-printed blocks for repair.

7. COMMERCIALIZATION

The approach presented here can be used to develop lightweight tooling from a variety of composite materials for compression molding and thermoforming applications demanding a few thousand molded parts. This will be facilitated by the easy reparability of the composite tool. Active cooling may be provided by means of cooling channels to cut down the demolding time. When such cooling is provided, the depth profile of the molded part may extend to 80 mm. Composite tooling can be designed and printed to ensure lower thermal residual stress in the molded part and correspondingly lower part distortion. The printing process can also be designed to obtain better heating of the mold through heated press platens, within limits on the depth of the tool.

8. ACCOMPLISHMENTS

- Publication in conference proceedings:
Jayaraman, K., Shree, S., Ejsmont, D., Dereims, A., Raveendra, R., “Controlling molded part distortion with print orientation in 3d-printed thermoplastic composite mold,” in CAMX – The Composites and Advanced Materials Expo, Dallas, Texas, 2021
- A computer program was developed for predicting viscoelastic effects on the deformation of the anisotropic thermoplastic composite under compression.

9. CONCLUSIONS

In using extrusion deposition additive manufacturing of thermoplastic composite tooling, it is

beneficial to preserve uniform fiber orientation along the press closing direction within the mold. This allows better and quicker heating of the tool in a press and more convenient repair of the mold when required. The tool being much thicker than the molded part produces noticeable deformation of the tool when used for molding carbon fiber reinforced epoxy prepregs.

Anisotropic thermomechanical properties of the composite tooling and of the molded composite part were used in simulations with finite element software. These simulations provided new understanding of the connection between the two sets of properties and the mold deformation as well as the distortion of the molded part. The simulation results for case 1 where the mold was four times as stiff as the part along the press closing direction compared well with experiment. When the mold stiffness was only 1.6 times the part stiffness along with correspondingly higher thermal expansion along the z-direction, the mold deformation during the compression molding increased; but this also lowered the thermal residual stress in the part and resulted in a smaller extent of part distortion.

10. RECOMMENDATIONS

The finite element software used might be coupled with user written subroutines to account for thermoviscoelastic behavior of the thermoplastic composite tool and obtain more accurate representation of the mold deformation and the coupling to the molded part deformation. The code for simulating thermoviscoelastic behavior was written and tested against literature results on a plaque geometry. Transferring this to the company's finite element software could be undertaken by the company as the next step.

11. REFERENCES

- [1] B. K. Post, et al. , "The economics of big area additive manufacturing," in *Solid Freeform Fabrication* 2016, pp. 1176-1182.
- [2] S. R. Huelskamp, Osborn, T, "Large format additively manufactured tooling for out-of-autoclave aerospace composites," in *SAMPE*, Seattle, WA, May 22-25, 2017 2017.
- [3] L. J. Love *et al.*, "The importance of carbon fiber to polymer additive manufacturing," *Journal of Materials Research*, vol. 29, no. 17, pp. 1893-1898, 2014.
- [4] H. L. Tekinalp *et al.*, "Highly oriented carbon fiber-polymer composites via additive manufacturing," *Composites Science and Technology*, vol. 105, pp. 144-150, 2014.
- [5] L. A.A. Hassen, J. et al., "Simulation assisted design for an additively manufactured autoclave tool accounting for an anisotropic expansion," in *CAMX – The Composites and Advanced Materials Expo*, Anaheim, California,, 2019.
- [6] Communication from Oak Ridge National Laboratory *Specific heat capacity and anisotropic thermal conductivity of additively manufactured carbon fiber reinforced thermoplastics.*
- [7] M. Bogdanor, et al., "Design of composite compression molding tools using large scale additive manufacturing," in *SAMPE Conference Proceedings*, Seattle, WA, 2020.