

Large-scale continuous carbon/glass fiber additive-compression molded composites

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ABSTRACT

Additive Manufacturing (AM) or 3-D printing has advanced from small-scale desktop printers to large-scale printers. Most of the present large-scale printers utilize feedstock materials in the form of pellets to create composite structures. To create structurally robust composite parts, reinforcements in the form of short fibers (carbon or glass) are often used to impart mechanical properties to the printed parts. However, poor mechanical properties in Z-direction and high porosity of pellets-based printed composites compared to composite manufactured using traditional methods are serious concerns. The authors report a combined approach in the present work, where fiber reinforced composites are printed with a high-throughput continuous fiber deposition method followed by a secondary compression molding process. A specially designed end-effector mounted on a robotic arm is used to print composite preforms. Continuous comingled fibers (Thermofiber 12K CF-PA12, Thermofiber 12K S2-PA12, and Hybrid Thermofiber 12K CF-PA12+PEEK PA6) embedded in the thermoplastic nylon matrix are printed to create composite preform plaques. The printed preforms were further compression molded (CM) using a hydraulic hot-press to create highly consolidated composite parts. The mechanical properties of the continuous fiber composites produced by this combined approach are improved significantly due to the highly aligned continuous fibers and reduced porosity. Flexural strength, flexural modulus, and tensile modulus of AM-CM Thermofiber 12K CF-PA12 UD sample were 615.37 MPa, 75.65 GPa, and 122.23 GPa, respectively.

Keywords: Continuous Fiber Additive Manufacturing, Integrated Composite Manufacturing, Compression Molding

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1. INTRODUCTION

Additive manufacturing (AM), commonly known as 3D printing, is a technique where the part is modeled using computer-aided design (CAD) software or other methods and further converted using another software called "slicer" to transform the CAD model into a layer-wise representation of the model. This slicer file is fed into so-called "3D printers," which produce the part layer-by-layer [1]. 3D printing is finding its applications in various sectors, but a significant portion is still dedicated to polymer-based 3D printing [2]. AM of polymer composites has come a long way in terms of materials and process advancement. Although AM has proven to be a fantastic tool for prototyping, replacing the traditional manufacturing process with AM is still far-sighted. The main hurdle is the low mechanical properties of 3D printed parts due to the presence of high porosity and poor bead-to-bead interface [3]. However, reinforced polymer AM has the capability to align the fibers in the deposition direction. Extrusion of a reinforced polymer through a nozzle helps in aligning the reinforced fibers in the deposition direction. The alignment comes due to the applied shear force in the nozzle during extrusion. By controlling the deposition direction, preferred fiber alignment can be achieved [4].

Oak Ridge National Laboratory (ORNL) proposed a new manufacturing process, where they combined the advantage of high fiber alignment of AM printed parts with the traditional compression molding (CM) process. This new manufacturing process is termed AM-CM [5]. In AM-CM, high fiber alignment and low porosity can provide exceptional mechanical properties, which were impossible with any other manufacturing processes. AM produced preform can utilize multi-material for more functionalities in the structures, such as over-molding, selective reinforcement, embedding electrically conductive pathways, etc. [6,7].

In this work, we replaced the short-carbon fiber polymer preform with continuous fiber preforms. It is well known that a minimum fiber length is necessary to obtain the best performance of short fiber composites, but during the extrusion process, a lot of fiber length attrition occurs; therefore, to achieve the true benefit of AM-CM process, continuous fiber printed preforms are used in the current study [8]. Orbital Composites Company, 3D printed various AM preforms for CM at ORNL. The experimental setup involved the use of the commercially available Orbital S robotic additive manufacturing system. This state-of-the-art machine was utilized for 3D printing neat nylon samples. Mitsubishi's T700 Continuous Carbon Fibers were also added to some of the samples to provide mechanical reinforcement. Orbital S 3D printer made both neat and continuous fiber samples with a Kuka industrial robot as shown in **Figure 1**.

The ability to print neat polymer, short-fiber reinforced polymer, and continuous fiber give a choice of preparing multi-material composite parts [9]. The integration of AM and CM resulted in significant improvement in material performance due to the control over the microstructure of the composites (fiber orientation, fiber length, and porosity). By designing the integrated AM-CM, energy and material savings will be very high due to the short production cycle time.

2. EXPERIMENTATION

Orbital Composites created the automated tools represented herein. Belilove Company created the precision heater subsystem employed to melt the filament. Two different kinds of robotic tools, or end effectors, were used for continuous fiber preform printing. First is the traditional nozzle with a single central channel used to melt, extrude, and weld with thermoplastic filaments. Continuous comingled fibers (carbon fiber (CF) - Thermofiber 12K CF-PA12, glass fiber (S2) - Thermofiber 12K S2-PA12, and Hybrid (carbon and glass fiber) - Thermofiber 12K CF-

PA12+PEEK PA6) embedded in the thermoplastic nylon matrix are printed to create composite preform plaques. The printed preforms were further compression molded using a hydraulic press to create highly consolidated composite parts. The second method utilizes Orbital *S* end effector, which is a coaxial extrusion nozzle. Coaxial extrusion is used to coextrude insulation onto continuous fibers. One can think of coaxial extrusion of continuous fiber with polymer being similar to the well-known industrial wire insulation process. However, adding the coaxial nozzle on top of a robotic arm allows depositing the continuous fiber with varying fiber volume fractions. The coaxial nozzle is fed thermoplastic filament, and then it extrudes molten thermoplastic around the continuous fibers extruding from the core nozzle. The ratio of coax to the core may be changed quite dynamically during the 3D printing. Consequently, the AM preforms with varying volume fractions ranging between 0% and 60% can be prepared. Plates with unidirectional reinforcement were prepared for this work. Some of the printed test samples are shown in **Figure 2**.

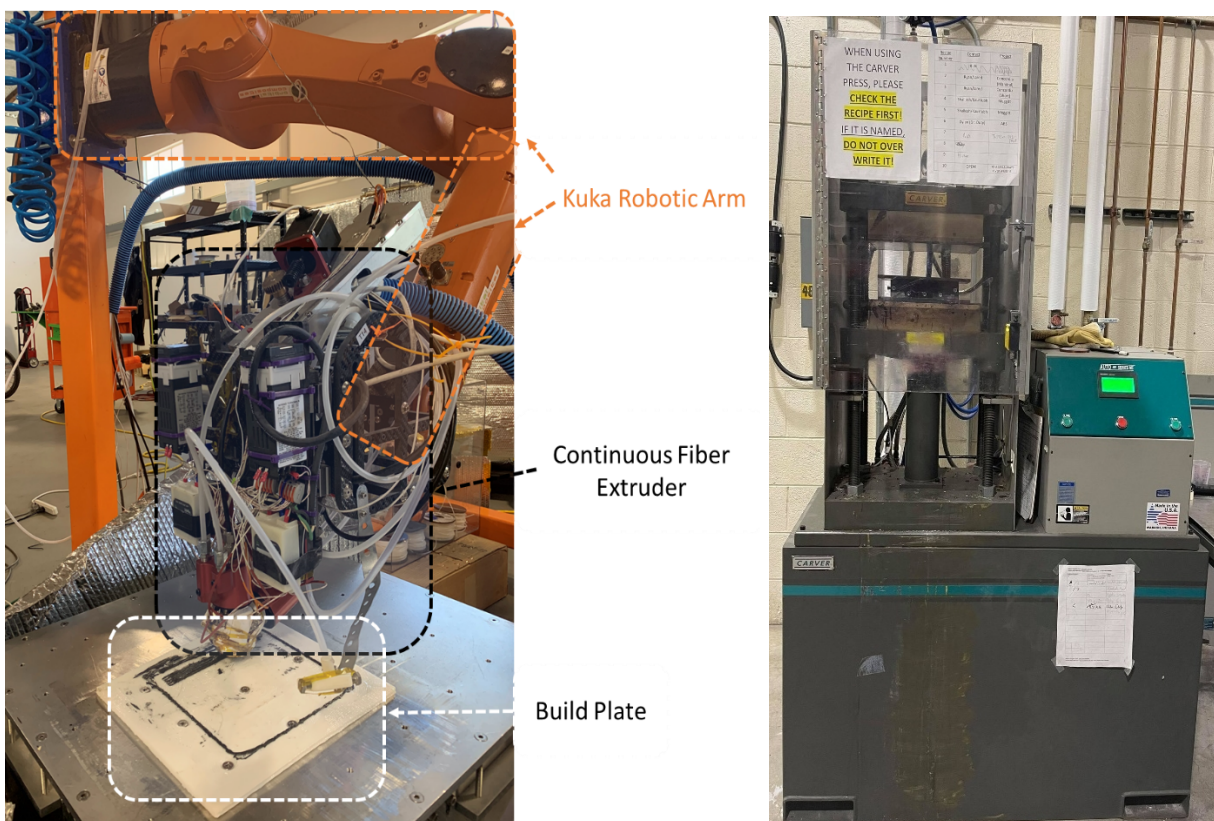


Figure 1. Continuous fiber AM preform printing by coaxial end effector mounted on a Kuka robot. Carver press used for the compression molding.

A hot-press (Carver Press-30 Ton) was utilized for the CM process of the AM preform. The compression parameters for the AM preform were carefully optimized to obtain the desired consolidation of the panels (see **Figure 1**). Thermofiber 12K CF-PA12 and Thermofiber 12K S2-PA12 preforms were hot-pressed at 185°C for 15 min while maintaining a pressure of 5 Tons. Hybrid Thermofiber 12K CF-PA12+PEEK PA6 preforms were hot-pressed at 225°C for 15 min while maintaining a pressure of 5 Ton. Prepared samples are shown in **Figure 3**. The AM preforms panels were then cut for measuring their mechanical properties. Tensile and flexural properties were measured according to ASTM D3039 and ASTM D790 standards, respectively.

A Test Resources Universal Testing Machine (UTM) equipped with a 50 kN load cell was used for mechanical testing. Test machine did not have sufficient load capacity to break the samples; therefore, only the tensile modulus of these samples is reported in this work.

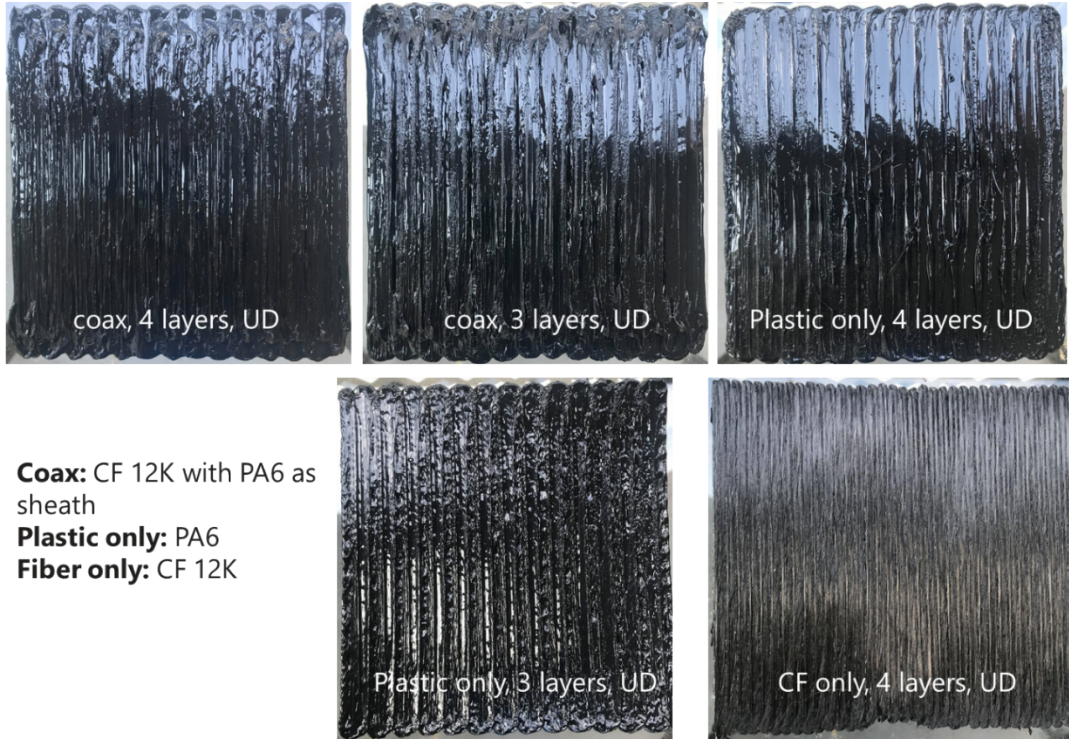


Figure 2. Various example of co-axially printed continuous fiber composites (a) Coax-4 layers-UD, (b) Coax-3 layers-UD, (c) Plastic only-4 layers-UD, (d) Plastic only-3 layers-UD, and (e) CF only-4 layers-UD

3. RESULTS

3.1 Mechanical Properties

Flexure and tensile test results are shown in **Figure 4**. Flexural strength and modulus of AM-CM Thermofiber 12K CF-PA12 UD sample was found to be 615.37 MPa and 75.65 GPa, respectively. The Ultimate tensile strength of carbon composite was not measured as the testing reached the maximum capacity of the Test Resource Frame. Tensile modulus of carbon composite was evaluated to be 122.23 GPa. These numbers are significantly high as compared to any AM-produced continuous CF thermoplastic composites. For comparison, CF epoxy composite with 60% by CF volume of UD composite has a tensile modulus of 135 GPa [10], which means with the current AM-CM approach, we have achieved 91% of the tensile modulus using a more environmental friendly thermoplastic composite. Similarly, the tensile modulus of glass fiber composite was 43.08 GPa, which is 7.5% higher than an epoxy-based E-glass fiber composite [10]. These results confirmed that the AM-CM approach could achieve the maximum performance of fiber-reinforced composites by aligning the fibers in preferred orientation using AM and removing the porosity by CM.

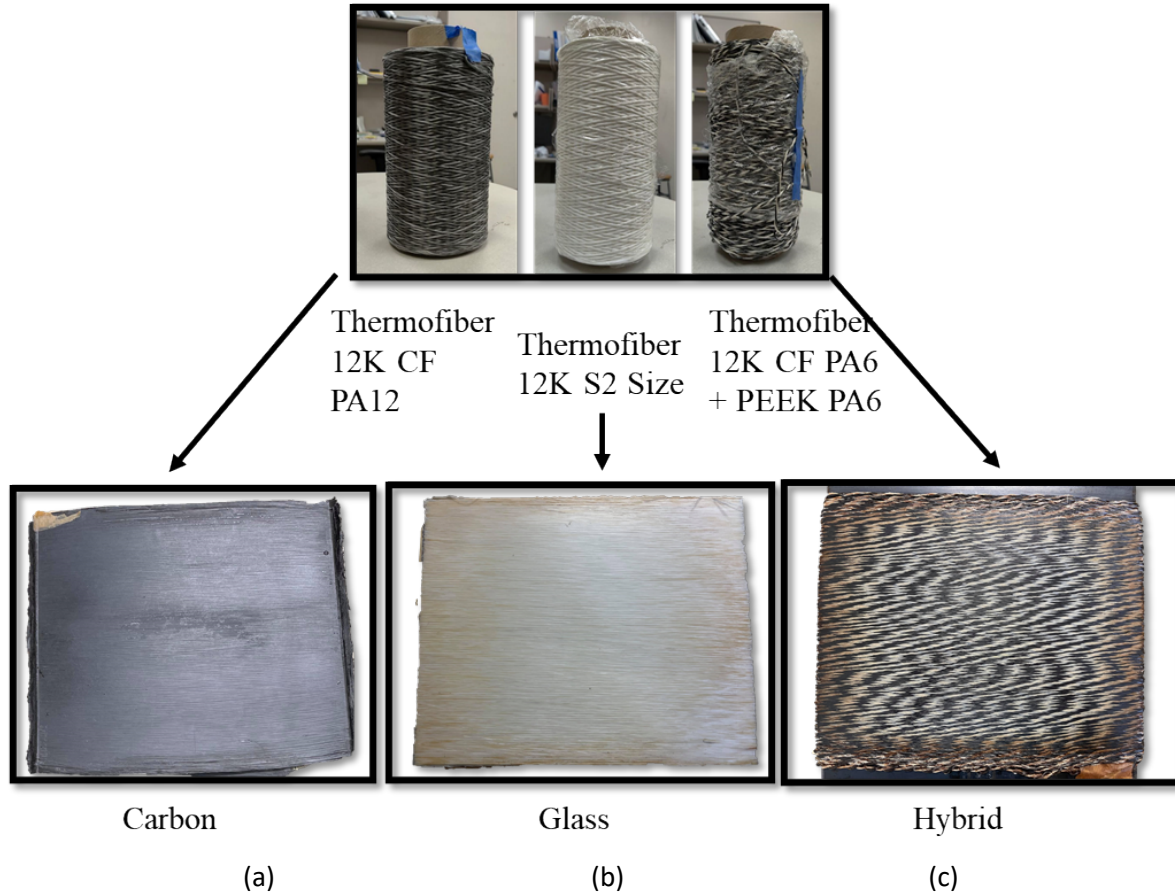
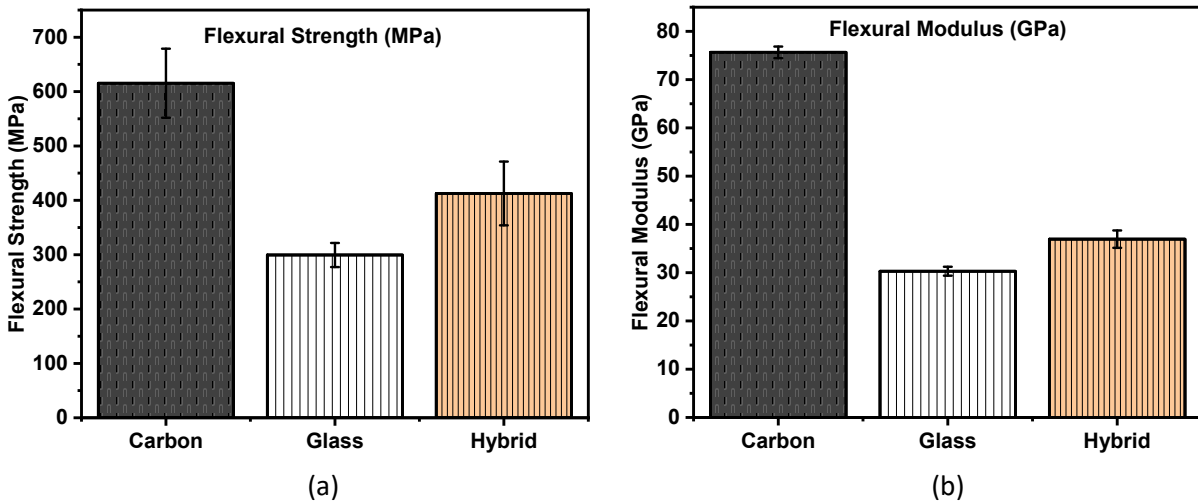


Figure 3. Compression molded AM-preforms; (a) Thermofiber 12K CF-PA12, (b) Thermofiber 12K S2-PA12, and (c) Hybrid Thermofiber 12K CF-PA12+PEEK PA6



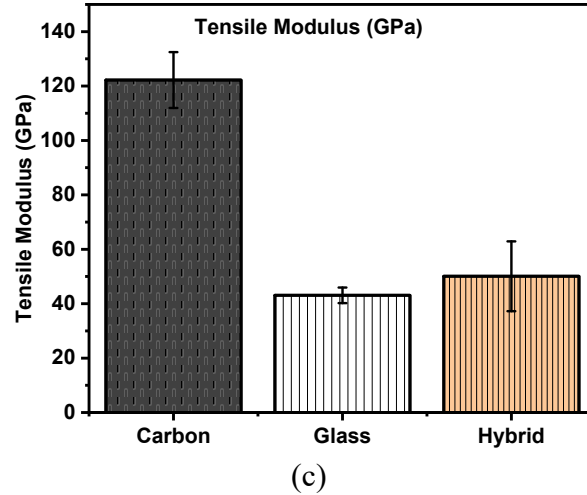


Figure 4. Mechanical properties of Thermofiber 12K CF-PA12, Thermofiber 12K S2-PA12, and Hybrid Thermofiber 12K CF-PA12+PEEK PA6 composites (a) Flexural Strength, (b) Flexural Modulus, and (c) Tensile Modulus.

Another important finding of this work was the ability to print two different fibers embedded in two different polymers together to create a hybrid composite. For this purpose, Hybrid Thermofiber 12K CF-PA12+PEEK PA6 were printed together, and compression molded to obtain a hybrid composite. The mechanical properties measurement of the hybrid samples confirmed that an intermediate performance was achieved. Flexural strength and modulus of AM-CM Hybrid Thermofiber 12K CF-PA12+PEEK PA6 sample were found to be 412.58 MPa and 36.95 GPa. Tensile modulus was 50.08 GPa. This approach can be further utilized to fabricate parts with tailored properties based on the application/requirement.

4. CONCLUSION

The authors report a combined approach of additively manufacturing and compression molding. Various continuous fiber preforms were printed with a high-throughput coaxial extruder mounted on a robotic arm. Comingled fibers (Thermofiber 12K CF-PA12, Thermofiber 12K S2-PA12, and Hybrid Thermofiber 12K CF-PA12+PEEK PA6) embedded in the thermoplastic nylon matrix were printed. The printed preforms were further compression molded using a hydraulic press to create highly consolidated composite parts. Flexural strength, flexural modulus, and tensile modulus of Thermofiber 12K CF-PA12 UD sample were found to be 615.37 MPa, 75.65 GPa, and 122.23 GPa, respectively.

5. ACKNOWLEDGMENTS

Research sponsored by the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, Advanced Manufacturing Office, under contract DE-AC05-00OR22725 with UT-Battelle, LLC. The design and execution of the research was done at the Manufacturing Demonstration Facility of Oak Ridge National Laboratory.

6. REFERENCES

- [1] B. Brenken, E. Barocio, A. Favaloro, V. Kunc, R.B. Pipes, Fused filament fabrication of fiber-reinforced polymers: A review, *Addit. Manuf.* 21 (2018) 1–16. doi:10.1016/j.addma.2018.01.002.
- [2] S.C. Ligon, R. Liska, J. Stampfl, M. Gurr, R. Mülhaupt, Polymers for 3D Printing and Customized Additive Manufacturing, *Chem. Rev.* 117 (2017) 10212–10290. doi:10.1021/acs.chemrev.7b00074.
- [3] P. Yeole, A.A. Hassen, S. Kim, J. Lindahl, V. Kunc, A. Franc, U. Vaidya, Mechanical Characterization of High-Temperature Carbon Fiber-Polyphenylene Sulfide Composites for Large Area Extrusion Deposition Additive Manufacturing, *Addit. Manuf.* 34 (2020). doi:10.1016/j.addma.2020.101255.
- [4] V. Kumar, S. Kim, V. Kishore, K. V Mungale, A. Nowlin, U. Vaidya, C. Blue, V. Kunc, A.A. Hassen, Hybrid manufacturing technique using large-scale additive manufacturing and compression molding for high performance composites, n.d. <http://energy.gov/downloads/doe-public-access-plan>.
- [5] V. Kumar, S.P. Alwekar, V. Kunc, E. Cakmak, V. Kishore, T. Smith, J. Lindahl, U. Vaidya, C. Blue, M. Theodore, S. Kim, A.A. Hassen, High-performance molded composites using additively manufactured preforms with controlled fiber and pore morphology, *Addit. Manuf.* 37 (2021) 101733. doi:10.1016/j.addma.2020.101733.
- [6] S. Alwekar, P. Yeole, V. Kumar, A.A. Hassen, V. Kunc, U.K. Vaidya, Melt extruded versus extrusion compression molded glass-polypropylene long fiber thermoplastic composites, *Compos. Part A Appl. Sci. Manuf.* 144 (2021) 106349. doi:10.1016/j.compositesa.2021.106349.
- [7] V. Kumar, T. Smith, J.C. Condon, P.S. Yeole, A.A. Hassen, V. Kunc, Replacing metal-based lightning strike protection layer of cfrps by 3d printed electrically conductive polymer layer, n.d. <http://energy.gov/downloads/doe-public->.
- [8] A.N. Dickson, J.N. Barry, K.A. McDonnell, D.P. Dowling, Fabrication of continuous carbon, glass and Kevlar fibre reinforced polymer composites using additive manufacturing, *Addit. Manuf.* 16 (2017) 146–152. doi:10.1016/j.addma.2017.06.004.
- [9] R. Singh, R. Kumar, I. Farina, F. Colangelo, L. Feo, F. Fraternali, Multi-material additive manufacturing of sustainable innovative materials and structures, *Polymers (Basel)*. 11 (2019). doi:10.3390/polym11010062.
- [10] Mechanical Properties of Carbon Fibre Composite Materials, (n.d.). http://www.performance-composites.com/carbonfibre/mechanicalproperties_2.asp (accessed June 25, 2021).