Cotton Fibers in 3D Printing

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Abstract

This work explores the materials challenges of cotton-loaded with polymer composites toward sustainable solutions in 3D printed free forms. A key step toward composite filament development is the reduction in size of the original cotton fibers. Mechanical processing of the cotton is introduced as a means of reducing the size of cotton fibers to form a material of an ultra-high aspect ratio (>250) structure that is nanometers in diameter and micrometers in length. Mechanical advantages are low density polyethylene loaded with the high aspect ratio cotton and is observed to maintain a mechanically robust material at loading up to 40 wt%. In addition, attempts to print with 25 wt% cotton fillers (~10-15 aspect ratio) in LDPE is demonstrated. Finally, considerations to processing challenges from a sustainable and practical viewpoint are provided.

Keywords 3D printing, Cotton, Pulverized cotton, low-density polyethylene, nanofibrillated cellulose (NFC)

Introduction

Freeform fabrication and rapid prototyping by additive manufacturing (AM) is an emerging field for creating products and parts for specific applications that have the potential to reduce manufacturing costs. The importance of reducing the consumption of energy becomes evident when as much as one third of U.S. energy cost are devoted to the manufacturing sector [1]. Reducing the amount of energy consumed lessens the reliance on fossil based fuels and other forms of energy harvesting that can negatively impact the environment. However, because of the time necessary to create a product, the energy consumption between AM and traditional manufacturing remains relatively equivalent [2].

Adding different plant-based or cellulose-based materials into traditional filaments can create unique properties that add to the sustainability of AM. Materials such as algae, bamboo, coffee grounds, hemp, and woodfill are commercially available. The mechanical properties tend to be weaker than traditional filaments, such as acrylonitrile butadiene styrene (ABS) and Poly(lactic acid) (PLA). However, when attempting to mimic natural elements (like a pine cone) the properties of natural based filaments exceeded that of synthetic filaments, especially with moisture uptake and curvature detail [3]. These natural filaments tend to contain up to 40% fillers that originated as pulp or cellulose-based nanofibers. These nanoparticles are able to seamlessly blend into powdered synthetic filaments because of a lager surface area, compared the volume of the particles [4].

Cotton based filaments provide a unique set of properties that makes it of interest in 3D printing. For example, cotton is a good conductor of heat, which would allow for a uniform heating of the filament during printing. Additionally, unlike wood, cotton gains strength when wet and therefore can be used in more applications where moisture is involved [5]. The purity and molecular weight are both higher in cotton than in wood products and when processed the crystallinity of cellulose does not decrease [6]. Finally, the size of cotton can be scaled to larger extents in as compared to wood pulp, thus allowing for the high aspect ratio of sized cotton to achieve a composite-like enhancements in the printed freeform. The aim of this work is to first demonstrate procedure for the sizing of cotton and effects of the cotton when used as a plant-based filler in 3D print polymers.

Experimental Design

The cotton materials used in these experiments detailed in this research was obtained as pulverized cotton (pCot) from Solvaira Specialties. This cotton is derived from recycled t-shirts and ground into micro-sized fibers, 350 μm in length and 20-40 μm in width. In comparison, cotton fiber as a typical dimension of X mm in length and 20 μm in diameter. A microgrinding procedure is used to further alter the size of the pCot to a larger aspect ratio. This is conducted using a Masuko Grinder microfluidizer in which a 3 wt % solution of pCot (in water) passes between two rotating stone plates at a gap of -.28 mm rotating at 1500 rpm [7]. The friction between the stones causes the solution to grind the cotton into nanofibrillated cotton (NFC) particles. As previously demonstrated, nine passes of the solution in the microfluidizer results in a uniformly reduced size of cotton. The NFC is then freeze dried in a two-step process, first at -80°C in a glass jar and then at -40°C in a Labconco freeze dryer. Once the fully dried, the NFC is then processed in a blender and shaken through a sieve to produce a powdered product.

At this point two different avenues are pursued low density polyethylene (LDPE) composites with the originated pCot material and also with the NFC material. The LDPE used in these experiments were obtained from Dow Chemical (AsSpun 6850A) and used as received. To compound the polymer with the pCot and NFC a DSM, Xplore, Micro 15 cc Twin Screw Compounder is used at heating of 170 °C, screw speed of 50 rpm, and a mixing time of 10 min. Compounded samples of up to 20 wt. % cotton material were produced. In addition, commercial-grade composite materials were fabricated and produced into pellet sizes with 25% pCOT and LDPE by Techmer PM.

Using a Noztek pro HT filament extruder with a 0.05mm nozzle size the LDPE- pCot pellets were extruded at 185°C then set to rest until cooled. The tensile strength of the filament was tested to compare against LDPE filament without the addition of pCot. Following ASTM D368-14, five different five inch segments of the filament were pulled in tension at 50.82 mm/min. The remaining cooled filament was loaded into a Lulzbot TAZ5 3D printer and extruded at 190°C creating a 127 x 5.1 x 35.6 mm dog bone shape. The print's mechanical properties are then tested to compare to other natural filaments and LDPE without pCot.

Results and Discussion

Filament Extrusion. This effort explores the differences between the LDPE extruded from the Noztek pro HT with and without cotton and define the best extrusion conditions. In these experiments, an initial amount of filament was extruded for approximately 1 min prior to the collection of the samples for collection. Microscope images of the LDPE extruded at 145°C, 165°C, 185°C, and 200°C are provided in Figure 1a, showing the average diameter decreasing as the temperature increased (Table 1). The mechanical strength of the virgin LDPE compared to LDPE with pCot. The strength of the Virgin LDPE is substantially higher than the strength of LDPE with pCot. This is due to the high loading of the cotton fibers decreasing the strength. Additionally, moisture could have infiltrated the composite during the extrusion process.

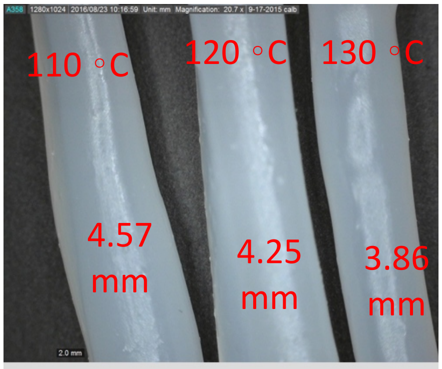

Figure 1. Comparison of LDPE filament extruded at temperatures ranging from 100-130 °C in a Noztek pro HT.

Table 1. Diameters of filaments extruded with the Noztek pro HT.

Virgin LDPE		LDPE with pCot	
Temperature (°C)	Diameter (mm)	Temperature (°C)	Diameter (mm)
145	4.58	110	4.57
165	4.34	120	5.25
185	3.3	130	3.86
200	3.3		

As a comparison, the LDPE extruded with 25 wt% pCOT at the same temperatures are provided in Figure 2. Again, the average diameter is observed to decrease with an increase in temperature and the results are provided in Table 1. In comparison to the virgin LDPE filament, the composite filament is demonstrated to have a rougher surface and similar diameter. The similar diameter is due to the nozzle head on the Noztek pro being the same size. The virgin LDPE was slightly larger due to having no particles impeding the flow.

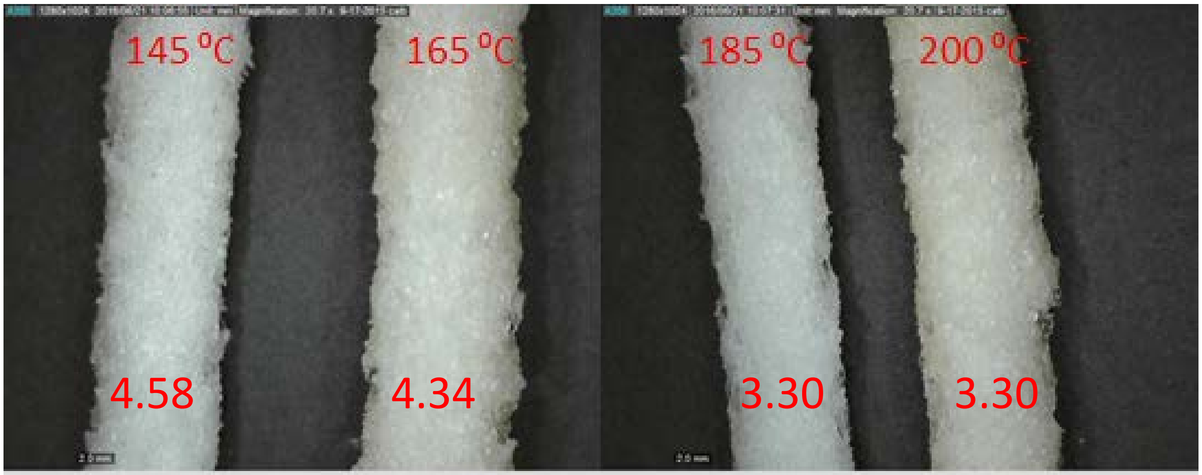

Figure 2. Comparison of the LDPE filament with 25 wt% pulverized cotton (pCOT) extruded at a temperature range of 145-200 °C in a Noztek pro HT.

Mechanical analysis of filaments was performed, with the stress-strain curves presented in Figure 3 and key mechanical values provided in Table 1. With respect to a normalized diameter, the filament extruded at 145 °C had the highest strength value, but proved to be inconsistent with regards to the extruded diameter and quality. Failing at about 45 N, 185 1 was determined to be strongest and most consistent filament. However, all of the extruded filaments with an LDPE pCot mixture were weaker and could withstand less strain than the virgin LDPE filaments.

3D printing of LDPE/pCOT. 3D printing was attempted using the LDPE/pCOT filament extruded at 185 °C. The typical print temperatures of LDPE is most often performed at a range of

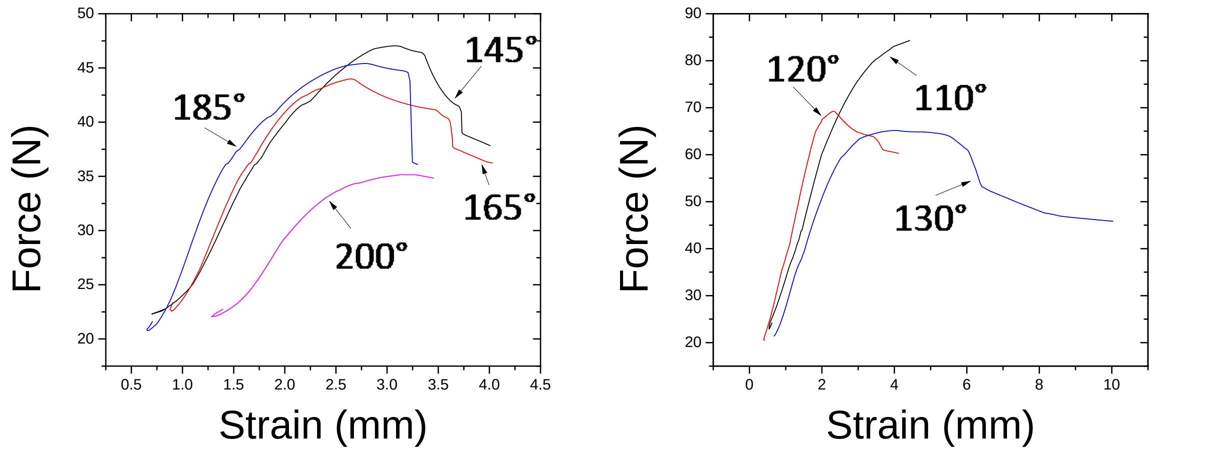

Figure 3. Strain curves of the four tested temperatures for LDPE pCot filament extrusion and **b.** LDPE pellets without pCot extruded at differing temperatures.

160-180 °C. Ideally, a broader range of extrusion temperatures would be selected for printing in this work, due to observations in the change in extrusion temperature observed with the inclusion of pCOT in the LDPE. Still, 3D printing is limited to a high temperature of 200 °C due to thermal limitations to the cotton. The best success of the 3D printing was observed at a temperature of

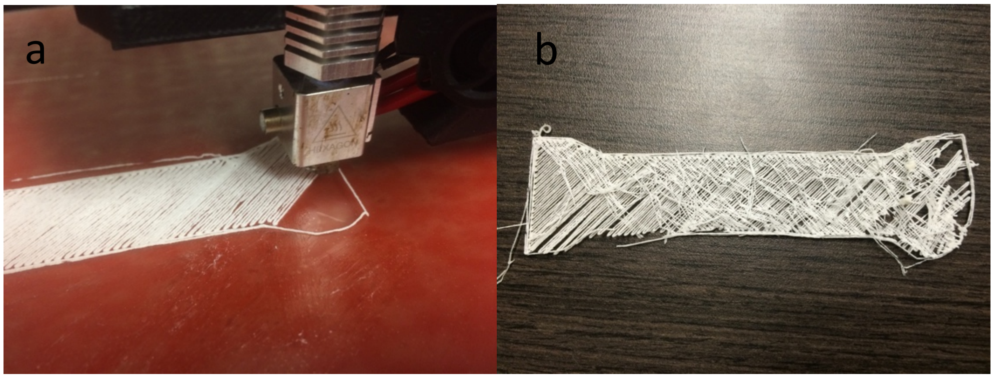

Figure 4. Image of **a.** the printing process of LDPE pCot and **b.** the initial results of the print. Developing NFC

190C (Figure 4). In the printing shown, the print surface was unheated and a glue stick was used to assist in the bonding. The result of the printing showed a successful initial layer of printing, but a multiple layered-structure was unable to be fabricated. Printed virgin LDPE and LDPE with pCot was extruded from the 3D printer and the mechanical strengths were tested. Figure 5 shows LDPE to be the stronger print, which is due to not having a filler polymer. Additionally, the LDPE pCot pellets displayed signs of containing moisture which can detract from the strength.

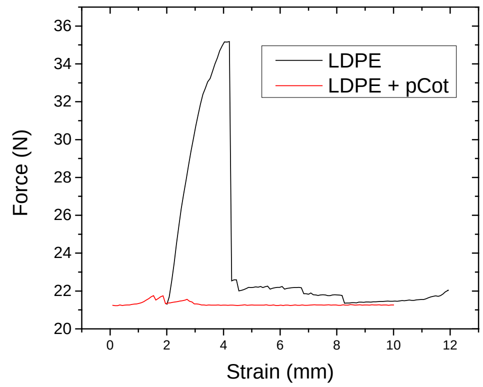

Figure 5. Strain curve of the virgin LDPE compared to the strain curve of LDPE with pCot.

Sizing of cotton to higher aspect ratios. This purpose of this effort is to overview our team's prior effort toward further sizing of cotton to larger aspect ratios.[7] As outlined in the experimental section, the route taken to achieve a larger aspect ratio was through microgrinding. Our results have shown that the nine passes of the pCot through the microgrinding plates, results in a uniform nanofibrilatted cotton structure, has aspect ratios of >250. An SEM and TEM image analysis is of the pCot and NFC materials is provided in Figure 6.

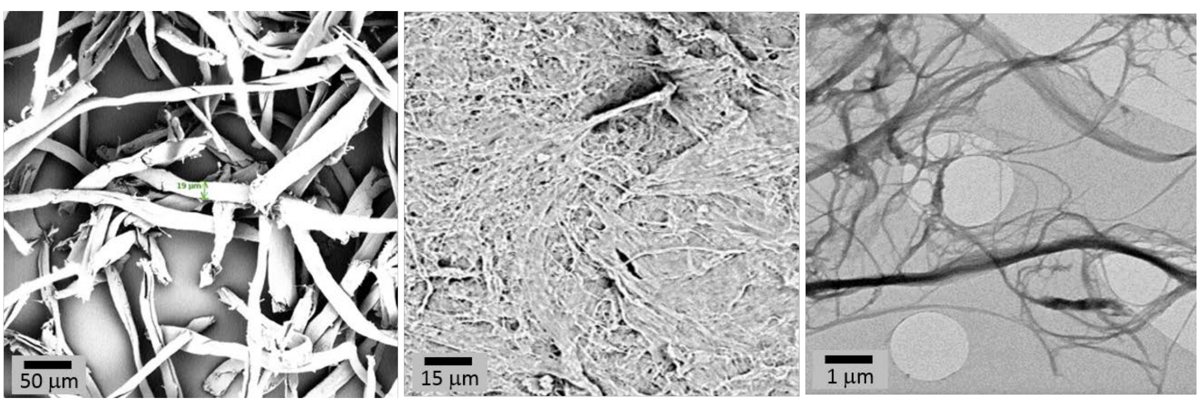

Figure 6. (left) SEM image of pCot before microgrinding. (middle) SEM image of the Cot after 9 microgringinComparison of the LDPE filament with 25 wt% pulverized cotton (pCOT) extruded at a temperature range of 145-200 °C in a Noztek pro HT.

Conclusion

This works examines the creation of nano-sized cotton particles and the integration into additive manufacturing. SEM and TEM images were taken to illustrate the structure of the cotton after two, four, and nine passes in a microgrinder. These images show greater fibrillation of the cotton with more passes and developed structures ranging 5-15 nm in length.

With the help of Techmer PM LDPE and pCot were compounded to create a pellet. An extrusion at 185°C the filament created was strong enough to be printed in a Lulzbot TAZ5 3D printer with success. The results produced a weak print that had trouble binding with other printed layers, however manipulating the heating and print bed should be a remedy. Compounding PLA and NFC proved to be a challenge with a 3:1 weight percent ratio. This work shows that printing with a cotton cellulose material is possible and has the potential to be a mass produced product.

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