Mechanical Properties of Hemp Fiber Reinforced Polylactic Acid Composites

By
Berkeley Skuratowicz

A THESIS

submitted to

Oregon State University

Honors College

in partial fulfillment of
the requirements for the
degree of

Honors Baccalaureate of Science in Mechanical Engineering and Manufacturing Engineering
(Honors Scholar)

Presented March 10, 2023
Commencement June 2023

AN ABSTRACT OF THE THESIS OF

Berkeley Skuratowicz for the degree of Honors Baccalaureate of Science in Mechanical Engineering and Manufacturing Engineering presented on March 10, 2023. Title:
Mechanical Properties of Hemp Fiber Reinforced Polylactic Acid Composites

Abstract approved:

Skip Rochefort

This thesis explores the mechanical properties of hemp fiber reinforced polylactic acid (PLA) composites. Fused filament fabrication (FFF) was used to compare the performance of three different weight percentages of hemp fiber reinforced PLA against PLA and acrylonitrile butadiene styrene (ABS). It was found that as the weight percentage of hemp increased in PLA, the tensile strength decreased and Young's modulus generally increased. The higher weight percent (5 and 10%) hemp fiber reinforced PLA was found to have a significantly higher Young's modulus than PLA but more research needs to be done to measure other mechanical properties to determine appropriate applications of this plastic composite.

Key Words: Plastics, 3D Printing, FDM, Hemp, Sustainability

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Honors Baccalaureate of Science in Mechanical Engineering and Manufacturing Engineering project of Berkeley Skuratowicz presented on March 10, 2023.

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Toni Doolen, Dean, Oregon State University Honors College

I understand that my project will become part of the permanent collection of Oregon State University Honors College. My signature below authorizes release of my project to any reader upon request.

Berkeley Skuratowicz, Author

Acknowledgments

I would like to thank Dr. Skip Rochefort, Dustin Cram at Proto-Pasta, Scott Campbell, Dr. Bryony DuPont, the 3D Printing Club of Oregon State University, Dr. John Simonsen, Dr. Massimo Bionaz, as well as Madeline Pasche, Sydney Nash, and Spencer Mitchell from Dr. Skip Rochefort's Polymer Lab.

Contents

1 Introduction

Over the last decade, the negative impact of petroleum-based plastics on the environment has become more widely known, leading to an increase in the development and production of bioplastics. The growing popularity of bioplastics stems from the biodegradable nature and reduced CO2 emissions when compared to petroleum-based plastics. Bioplastics are derived from agricultural products such as corn, wheat, sugar, rice, potatoes, and soya. These agricultural products absorb CO2 during growth which balances the CO2 emitted when the plastic is produced [1]. This paper focuses on polylactic acid (PLA) which is primarily produced from corn and sugar beet [2].

PLA is a high-strength, high-modulus polymer that is one of the most commercially successful bioplastics. It is commonly used in packaging and single use items and has better mechanical properties and durability than many other biodegradable plastics [3]. In order to further enhance the mechanical properties of PLA, natural fibers such as hemp can be added. Natural fiber reinforced polymers have been of interest in research because of low cost, recyclability, and high strength to weight ratio [2]. This paper will focus on assessing the mechanical properties of hemp reinforced PLA using fused filament fabrication (FFF) and its potential to be used as sustainable material. Hemp reinforced PLA will be compared to PLA and ABS filament in order to determine if it has viable mechanical properties.

2 Previous Work

Hemp reinforced PLA is a relatively new composite material with the majority of research conducted within the past 15 years. Research tends to fall into two sections: properties of hemp fiber reinforced PLA in compression/injection molding and properties of hemp fiber reinforced PLA in FFF.

2.1 Properties of Hemp Fiber Reinforced PLA in Compression and Injection Molding

One 2009 study published in the Journal of Composites looked at the mechanical properties of three different PLA composites using compression molding. These composites all had a fiber mass content of 40% and were PLA mixed with hemp, PLA mixed with Lycell, and PLA mixed with both hemp and Lycell. The results showed that adding a fiber mass content of 40% hemp to PLA increased the tensile strength from 29 N/mm^2 to 58 N/mm^2 and increased Young's modulus from 3800 N/mm^2 to 8050 N/mm^2 [4].

2.2 Properties of Hemp Fiber Reinforced PLA in FFF

Another study published to the Journal of Composites explores hemp and other natural fiber reinforced PLA using fused filament fabrication. This 2017 study used 0, 10, 20, and 30 weight percent hemp and gathered data on each composite's tensile strength. 0% hemp was found to have a tensile strength of 35 MPa while 10% hemp increased the tensile strength to 38 MPa. After 10% hemp, the tensile strength decreased to 29 MPa and 25 MPa for 20% and 30% respectively [5].

3 Methods

3.1 Materials

Spent hemp biomass obtained from the College of Agriculture at Oregon State University was used for the 5, 10, and 15 weight percent (wt%) hemp reinforced PLA filaments. These three filaments were custom made by the company, Proto-Pasta (P-P), and will be referred to as P-P 5 wt% hemp PLA, P-P 10 wt% hemp PLA, and P-P 15 wt% hemp PLA. An additional 4 weight percent hemp reinforced PLA filament (designated as 3DF 4 wt% hemp PLA) and a PLA filament (designated as 3DF PLA) were purchased through the company 3D Fuel (3DF). The ABS filament was obtained through the Oregon State University's 3D Printing Club.

3.2 Preparation of Hemp

In order to develop the P-P 5, 10, and 15 wt% hemp reinforced PLA filament, the hemp biomass needed to be ground down to 150 micrometers or less. To achieve this, a Wiley mill from the Department of Forestry was used. Since the smallest available screen for this mill was 500 microns, the ground hemp had to be sieved after the grinding process to isolate the 150 micron and below particles. Three different sized sieves were used: 425 microns, 149 microns, and 75 microns.

As seen in Figure 1, the sieves were stacked based on particle size with a bottom tray to catch particles less than 75 microns. After grinding, the hemp would be deposited in the 425 micron sieve, the lid would be attached to the top, and the sieves would be put in a vibratory sieve shaker for 15 minutes.

Figure 1: Sieves used to filter hemp particles to 75-149 micrometers.

After being processed through the sieves, the 75-149 micron hemp particles were set aside and the other sizes were also saved for future use (see Figure 2). When 2 pounds of 75-149 micron hemp was collected, it was sent to Proto-Pasta to be mixed to create the hemp reinforced PLA filament.

Figure 2: Hemp particles sorted by size after the grinding and sieving process.

3.3 Analysis of Materials

Differential scanning calorimetry (DSC) was used to determine the glass transition temperatures for cooling and heating, crystallization temperature, and melting temperature of each material. Below (Figure 3) is the DSC for P-P 10 wt% hemp reinforced PLA with the other DSC graphs included in the appendix. The transition temperatures that were found for each material are included in Table 1. 3DF PLA was found to have the lowest melting temperature at 172 °C and the highest crystallization temperature at 112 °C while P-P 5 wt% hemp reinforced PLA had the highest melting temperature at 182 °C and the lowest crystallization temperature at 101 °C.

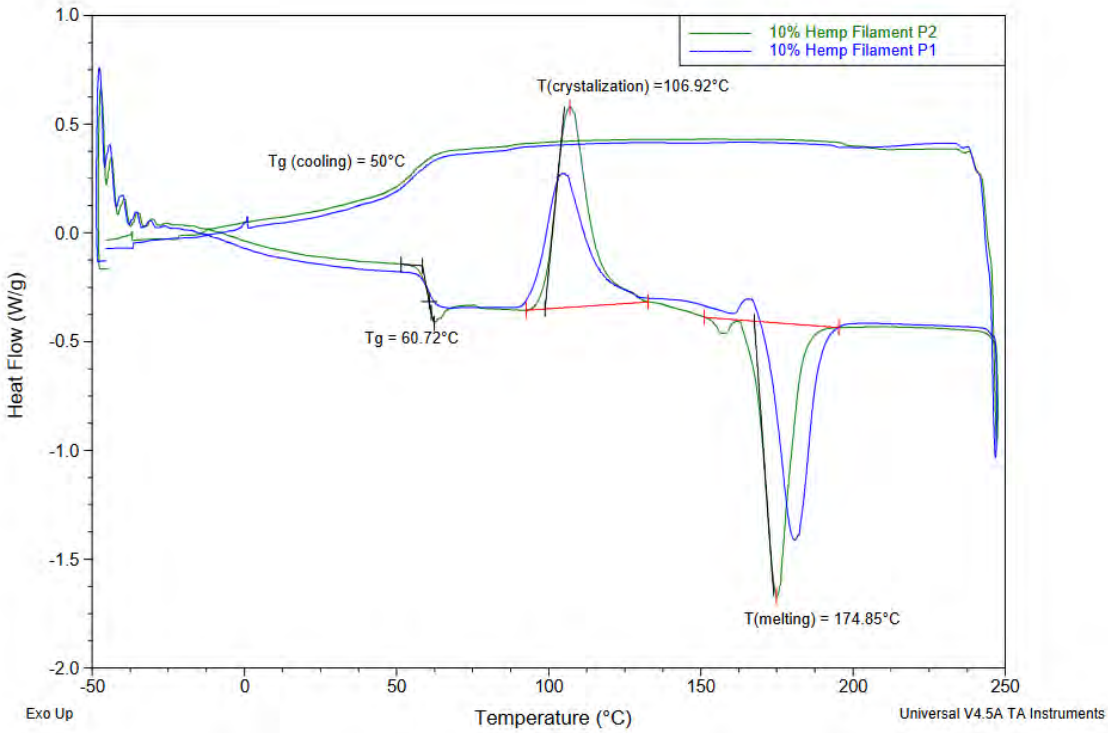

Figure 3: A sample DSC for P-P 10 wt % hemp PLA.

Material	Tg (cooling) (°C)	Tg (°C)	T (crystallization) (°C)	T(melting) (°C)
3DF PLA	105	63	112	172
3DF 4 wt% Hemp	55	64	105	180
P-P 5 wt% Hemp	58	60	101	182
P-P 10 wt% Hemp	50	61	107	175
P-P 15 wt% Hemp	56	61	106	182

Table 1: Results from the DSC.

Additionally, a thermogravimetric analysis (TGA) was run for the 3DF 4 wt%, P-P 5 wt%, P-P 10 wt%, and P-P 15 wt% hemp reinforced PLA. The TGA was used to determine the exact weight percentage of hemp included in each PLA sample.

Figure 4 shows the TGA of P-P 5 wt% hemp and found that there was 5.318 wt% hemp present in the sample. For the TGAs performed, the hemp present in the samples was within 1% of what was expected except for the P-P 15 wt% hemp PLA which was found to be 2.5% lower than expected.

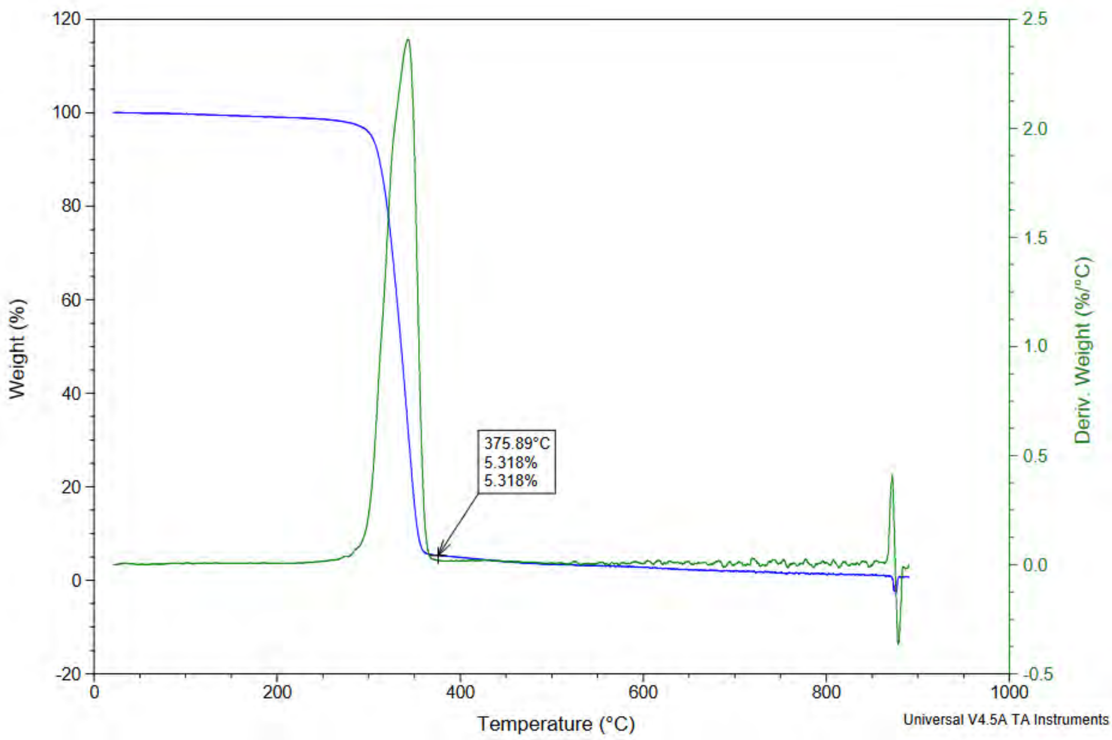

Figure 4: A sample TGA for P-P 5 wt % hemp PLA.

Material	Given Hemp wt%	Hemp wt% Found Through TGA
3DF 4 wt% Hemp	n/a	4.2
P-P 5 wt% Hemp	5	5.3
P-P 10 wt% Hemp	10	11.1
P-P 15 wt% Hemp	15	12.5

Table 2: Hemp wt% found using TGA vs. the hemp wt% that was given.

3.4 Fused Filament Fabrication

Fused Filament Fabrication (FFF) was used to create tensile bars of 4, 5, 10, and 15 wt% hemp PLA as well as PLA and ABS. FFF was decided upon because it is a quick way to get a basic understanding of the mechanical properties of hemp reinforced PLA. In order to print out the tensile bars, a Prusa Mk3 i3 printer from Oregon State University's 3D printing club was used (Figure 5). For each material, 20 tensile bars were printed with 100% infill and a layer height of 0.15mm.

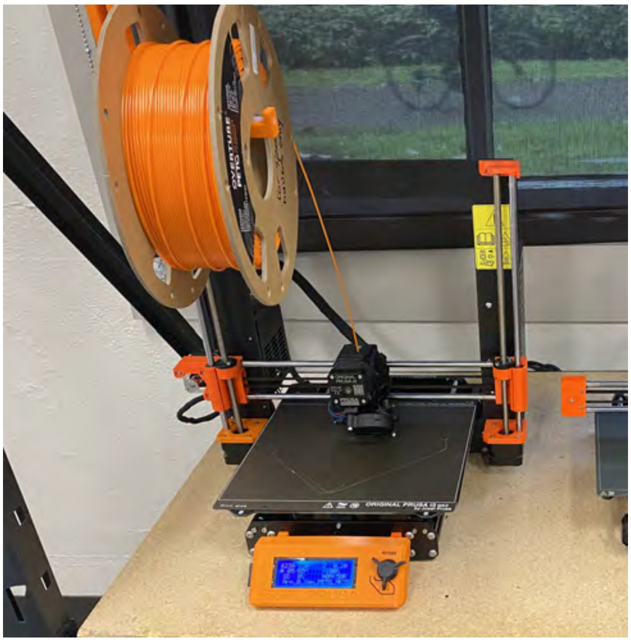

Figure 5: Prusa Mk3 i3 printer used to print tensile bars.

3.4.1 Determining FFF Settings

To get a tensile bar printed with no defects, the proper print settings must be chosen and refined. For 3DF PLA and ABS, no problems were encountered printing at the recommended temperature and other settings (see Table 3). When printing with the 3DF 4 wt% hemp reinforced PLA filament, problems with bed adhesion appeared when using the manufacturer recommended settings. To remedy this, the nozzle temperature was increased from 195°C to 200°C and the printing speed was slowed from 80 mm/s to 64 mm/s.

The P-P 5 and 10 wt% hemp reinforced PLA are custom filaments and have no recommended settings so the nozzle temperature was set to 10°C below the value used for 3DF PLA and the bed temperature was set to the same value as 3DF PLA. To maximize the chance of good bed adhesion, the print speed was set to 60 mm/s, the first layer speed was set to 10 mm/s, and a brim was used. For both filaments, the flow rate had to be increased to 110% in order to get enough filament deposited onto the print bed. It was also found that both the P-P 5 and 10 wt% hemp reinforced PLA filaments were more prone to clogging the print nozzle. The P-P 15 wt% hemp reinforced PLA filament wasn't viable to print as the hemp outgassed too much, causing the strand to expand then collapse into a flat tape.

Material	Nozzle Temp (°C)	Print Speed (mm/s)	First Layer Speed (mm/s)	Bed Temp (°C)	Brim
3DF PLA	210	80	20	60	No
3DF 4 wt% Hemp	200	64	16	60	No
P-P 5 wt% Hemp	200	60	10	60	Yes
P-P 10 wt% Hemp	200	60	10	60	Yes
ABS	255	80	20	110	Yes

Table 3: Successful print settings for tensile bars.

3.5 Heat Press

In order to more closely replicate results from injection molding, heat pressing was attempted. Heat pressing is more comparable to injection molding than FFF because it eliminates the weak bonding in the Z-direction seen in FFF, which can lead to the de-lamination of layers [6].

An aluminum two-part mold was made to create the heat pressed tensile bars as shown in Figure 6. The mold consisted of two sections: a CNC machined cutout of a tensile bar and a bottom block which is attached by screws.

Figure 6: Two part aluminum mold for heat pressing.

To create the heat pressed tensile bars, the two part mold was filled with PLA pellets (Figure 7) and placed in the Carver laboratory press. The hot plates of the press were heated up to the melting point of PLA, 180 °C and a metal sheet cover was put on the top of the mold. The two hot plates were pressed together using a hydraulic press for around 30 seconds. Even with industrial strength high heat mold release, the plastic bonded to the mold and wasn't able to be removed, making this method not viable.

Figure 7: Heat press set up.

3.6 Tensile Testing

When evaluating the mechanical properties of polymers using FFF, the recommended standard is ASTM D682 [7]. In accordance to this standard, the tensile bars used were ASTM D682 type IV with dimensions as shown in Figure 8.

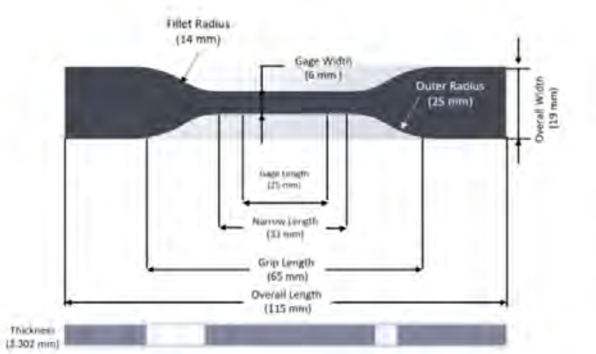

Figure 8: ASTM D6382 Type IV tensile bar specifications [7].

Twenty tensile bars of 3DF 4 wt% hemp reinforced PLA, P-P 5 and 10 wt% hemp reinforced PLA, 3DF PLA, and ABS were produced using FFF. Each of the 100 total tensile bars were loaded into the Instron tensile testing machine and tested using the ASTM D682 determined speed of 5 mm/s (Figure 9). The displacement and force were recorded and saved for analysis. After the tensile testing, the fracture surface of each material was photographed (Figure 10).

Figure 9: Instron tensile test

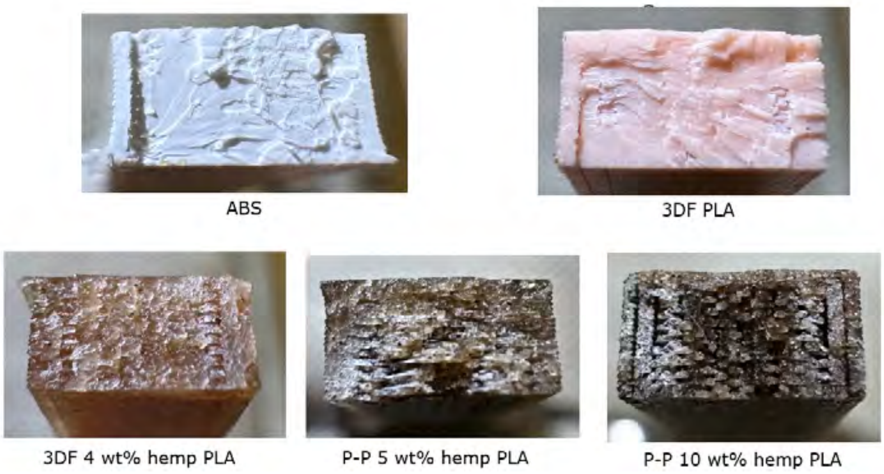

Figure 10: Fracture surface for each tensile bar material.

3.7 Data Analysis

The raw data exported from the Instron testing machine were in kilogram-force (kgf) and displacement (mm). To convert to units of stress, megapascals (MPa), the first step was taking the values for force and dividing them by the cross-sectional area of the tensile bar:

$$Pressure = Force\ (kgf)\ /\ Area\ (19.05\ mm^2)$$

This results in a unit of pressure of kgf/mm^2. To get to MPa, the resultant pressure was multiplied by a factor of 9.807.

In order to find the strain, the displacement data were divided by the initial narrow length. Using the calculated stress and strain data, graphs for each test were plotted (Figures 12, 13, 14) and the Young's modulus was found by calculating the slope of the linear section of each graph (Figure 11).

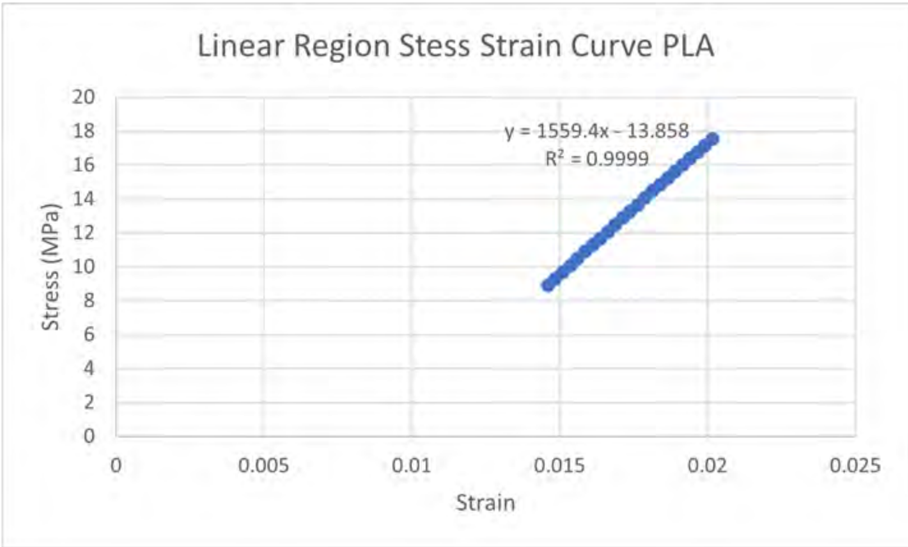

Figure 11: A sample linear region of a stress strain graph for 3DF PLA wth equation for the slope.

4 Results

The average maximum tensile strength and average Young's modulus were found by averaging the data from 20 tensile bars used for each material (Table 4).

For each test, a stress strain graph was created as shown in Figures 12, 13, 14. For each material, 20 graphs were analyzed for the maximum tensile strength and the slope was calculated to determine Young's modulus.

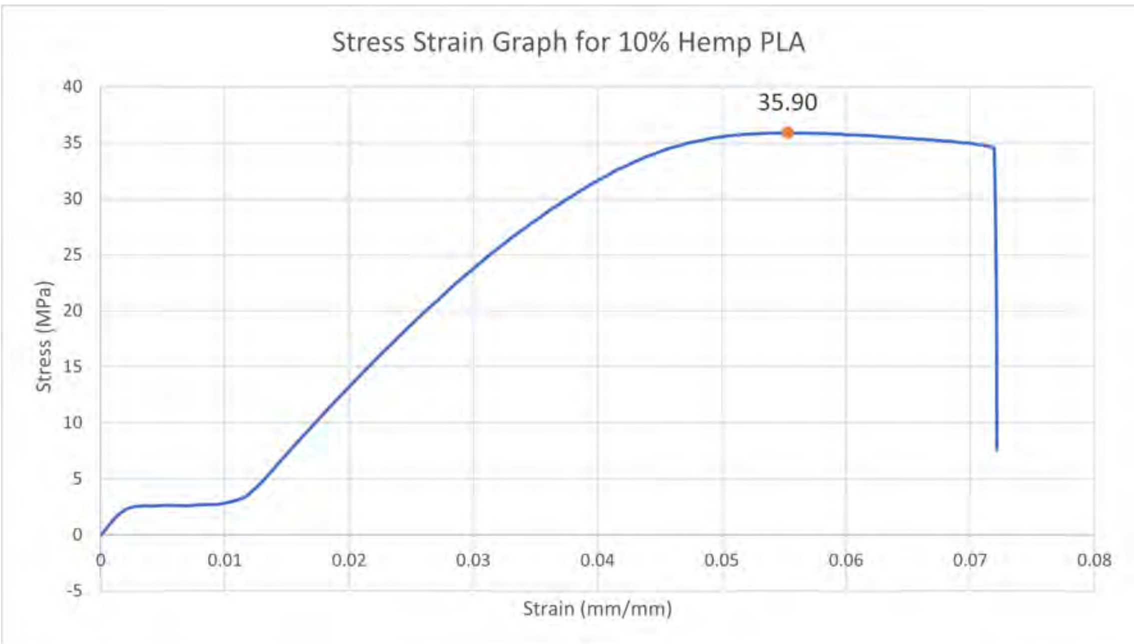

Figure 12: A sample stress strain graph for P-P 10 % hemp with maximum tensile strength marked.

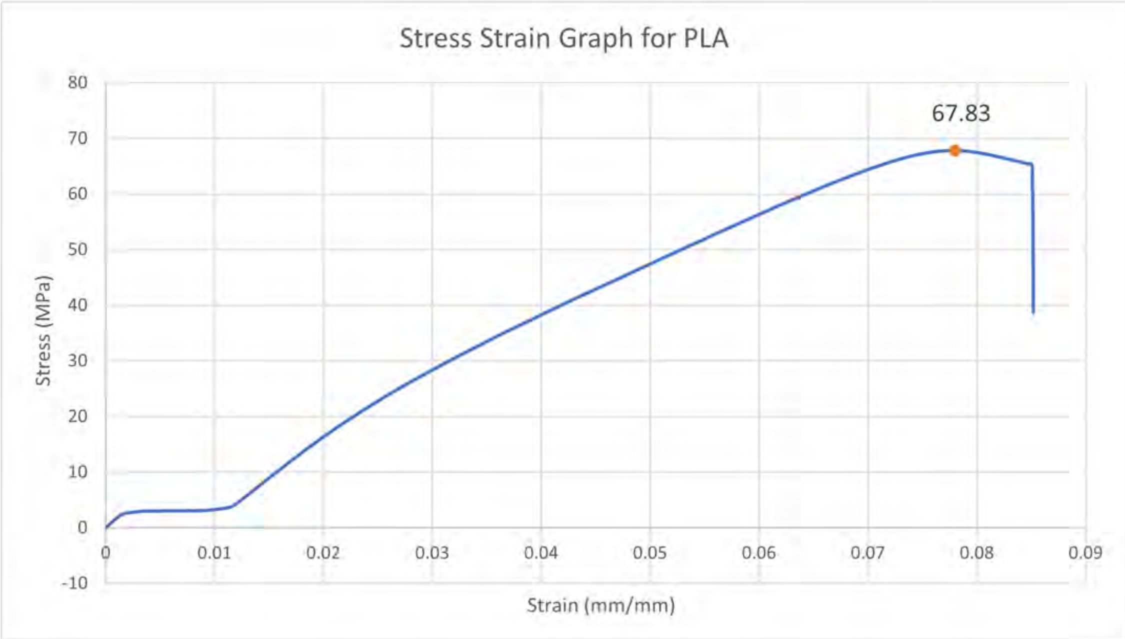

Figure 13: A sample stress strain graph for 3DF PLA with maximum tensile strength marked.

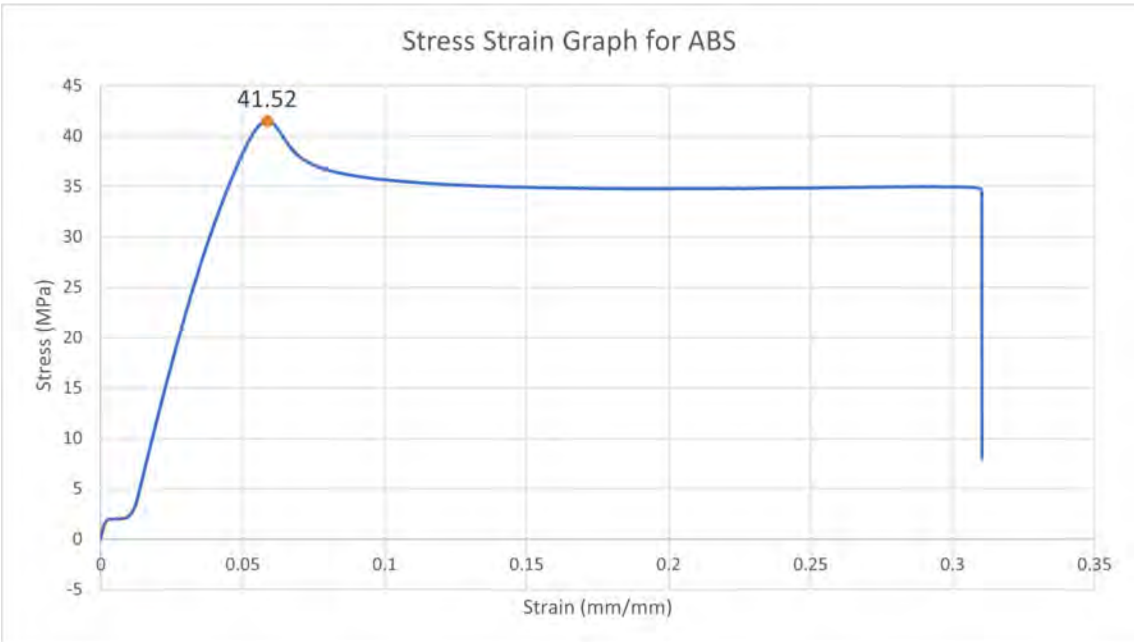

Figure 14: A sample stress strain graph for ABS with maximum tensile strength marked.

Material	Average Max Tensile Strength (MPa)	Average Young's Modulus (MPa)
3DF PLA	67.5	949.1
3DF 4 wt% Hemp	59.1	950.0
P-P 5 wt% Hemp	58.9	1295.4
P-P 10 wt% Hemp	34.5	1037.6
ABS	42.1	918.4

Table 4: This table shows the average maximum tensile strength and the average Young's modulus for each of the five materials. For each material, 20 tensile bars were tested and the results averaged.

5 Discussion

Overall, as the weight percentage of hemp increased in PLA, the maximum tensile strength decreased. 3DF PLA had the highest maximum tensile strength at 67.5 MPa. For 3DF 4 wt% hemp reinforced PLA and P-P 5 wt% hemp reinforced PLA, the maximum tensile strength decreased to around 59 MPa. For the P-P 10 wt% hemp reinforced PLA, the maximum tensile strength decreased significantly down to 34.5 MPa. Compared to ABS, all the samples had higher maximum tensile strengths except for P-P 10 wt% hemp reinforced PLA.

The inverse relationship between maximum tensile strength and weight percentage of hemp is most likely due to the hemp particles having a low length to diameter ratio (L/D) and a size of 75-149 microns. Assuming good adherence with the PLA matrix, fibers with a higher L/D would increase tensile strength and the load could get better distributed through the polymer matrix to the fibers. This would increase the overall tensile strength [8].

Another factor that could cause the maximum tensile strength to decrease as the wt% of hemp increases is under-extrusion of the filament during FFF. As seen in Figure 10, gaps between infill lines become more apparent as the wt% of hemp increases. These gaps can form from not enough filament extruding during FFF. An explanation for this trend could be that as more hemp is added to PLA, it starts to impede the flow of the polymer. This could lead to the filament extruding slower and the gaps seen in Figure 10. As these are assumptions, further testing is needed to determine causes of the gaps between infill lines.

Adding the 75-149 micron hemp particles to PLA to create the P-P 5 and 10 wt% hemp reinforced PLA caused a significant increase in the Young's modulus. Compared to PLA, adding 5 wt% hemp increased the Young's modulus by 346.3 MPa and adding 10 wt% hemp increased the Young's modulus by 88.5 MPa.

The Young's modulus was found to increase overall as the weight percentage of hemp in PLA increased. The exception to that is the decrease from P-P 5 wt% hemp reinforced PLA to P-P 10 wt% hemp reinforced PLA. This could be due to the mixing process as the hemp used was difficult to work with because it was low quality. When being mixed, it was reported from Proto-Pasta that the hemp outgassed making higher percentages of hemp filament more difficult to produce.

6 Conclusion

This study focused on determining the mechanical properties of hemp reinforced PLA composites. P-P 5 wt% hemp reinforced PLA was found to have the best mechanical properties of the three hemp reinforced PLA composites with an overall increase of 36.5% in Young's modulus and a decrease of 12.7% in tensile strength as compared to 3DF PLA. The P-P 10 wt% hemp reinforced PLA was found to have the lowest mechanical properties due to larger amounts of filler material with a low L/D. All of the hemp reinforced PLA composites were found to have inferior tensile strength compared to 3DF PLA which is likely due to the L/D of the hemp particles being close to one.

One area that can be further explored is the effect of a higher L/D on the maximum tensile strength and modulus of elasticity. Since the fibers used in this study had an L/D close to one, they added volume in the entangled polymer network but with no additional tensile strength. A higher L/D could increase the tensile strength and other mechanical properties of the material. Another area that can be explored is using a viable heat pressing method or injection molding to create tensile bars. These methods could help determine how hemp reinforced composites would perform in a large production environment.

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Appendix A

Max Tensile Strength (Mpa)				
PLA	4% Hemp	5% Hemp	10% Hemp	ABS
67.1612	62.4714	58.5897	35.1508	42.0955
68.0518	61.5808	53.3439	34.1675	42.0903
65.6734	62.147	62.3118	35.8972	42.4713
67.4495	60.4945	62.3324	35.6192	42.5639
69.5756	63.9952	62.6207	35.3567	42.5279
67.6606	58.9913	61.9257	36.1031	41.3593
67.4907	60.4533	63.4649	35.0581	41.5189
67.8305	60.3349	62.667	34.6463	42.2036
69.0403	61.4984	57.0453	33.8895	41.7351
68.0673	55.3259	62.2757	34.4249	41.0195
68.0209	61.7043	56.896	36.2782	42.1212
67.0016	56.6541	56.1547	34.5176	41.174
65.9566	56.6695	53.972	34.3786	42.2242
69.1587	56.8446	54.6824	33.4725	42.4404
65.6014	57.1329	56.9115	33.802	42.0749
66.8472	58.1676	55.7635	33.9461	42.9861
66.7133	57.6528	62.5125	31.8509	42.1727
66.945	56.3915	55.0119	34.3065	42.6823
66.9244	55.053	59.7569	34.394	42.2653
69.3749	57.4417	59.7209	32.654	42.5588

Figure A.1: Maximum tensile strength for each of the twenty tensile tests for all five materials.

Young's Modulus (Mpa)				
PLA	4% Hemp	5% Hemp	10% Hemp	ABS
1068.4	917.43	1331	1073.7	936.24
907.07	998.25	1301.5	1058.5	938.41
920.47	978.27	1411.3	1087.1	961.42
954.95	978.47	1293.1	1053.2	937.9
967.24	937.74	1303	1075.2	944.59
955.67	1036.8	1332.8	1078.3	916.9
906.55	996.08	1205.2	1064.7	924.8
925.35	967.46	1315.8	1030.4	925.77
1022.6	940.38	1264.4	1001.3	940.63
940.91	817.58	1288.5	1025.5	899.48
967.11	980.46	1293	1096.3	923.11
948.05	972.46	1278	1043.3	934.73
917.06	968.8	1183.8	1019.8	888.95
956.87	920.38	1304.5	1017.5	901.75
958.33	886.92	1347.9	1014.3	917.22
947.39	975.89	1301.4	1022.7	884.33
944.34	916.37	1382.3	992.31	901.28
918	906.53	1229.8	1034	878.11
907.03	960.44	1265.1	1022.6	901.68
949.21	942.96	1276.4	940.36	910.34

Figure A.2: Young's Modulus for each of the twenty tensile tests for all five materials.

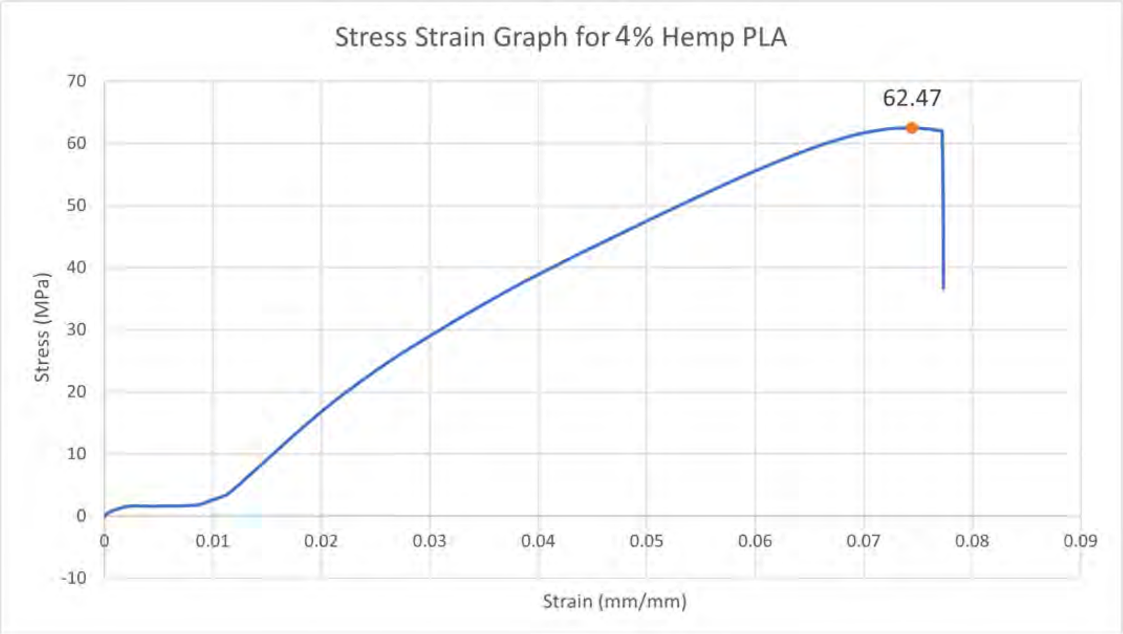

Figure A.3: A sample stress strain graph for 3DF 4% hemp PLA with maximum tensile strength marked.

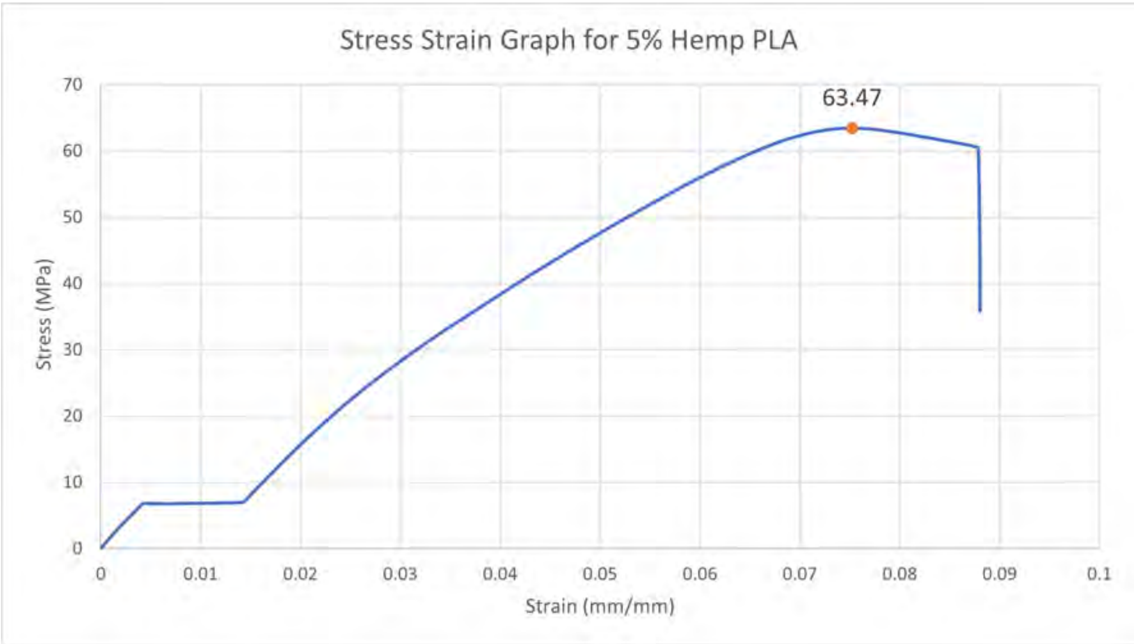

Figure A.4: A sample stress strain graph for P-P 5 % hemp with maximum tensile strength marked.

Appendix B

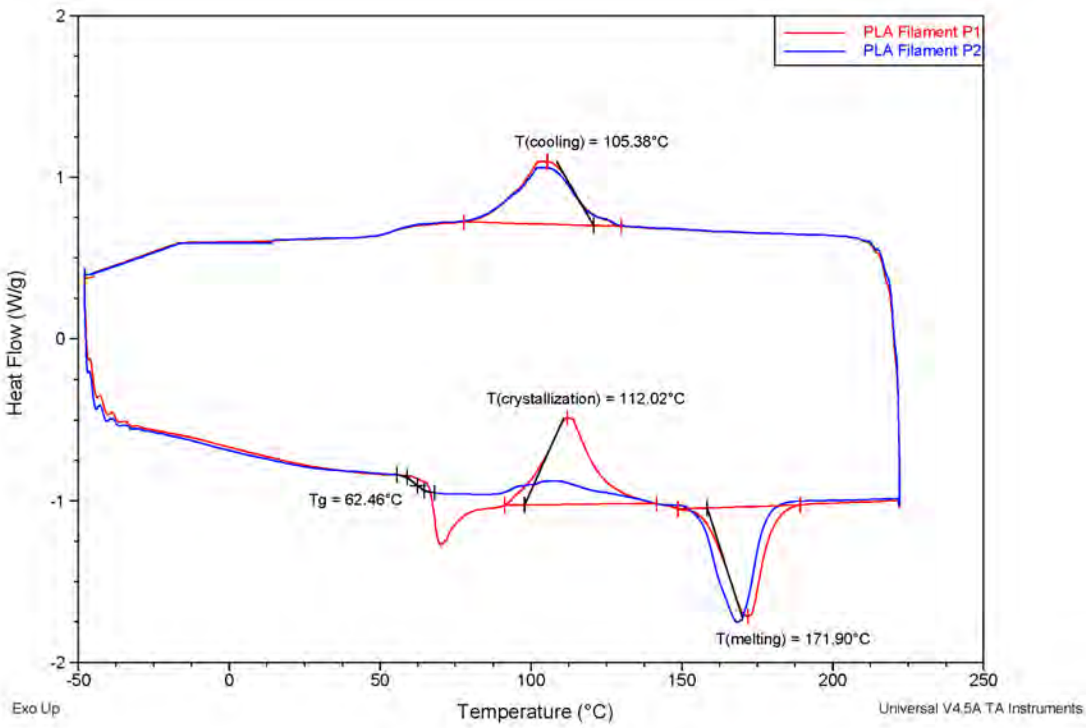

Figure B.1: DSC for 3DF PLA.

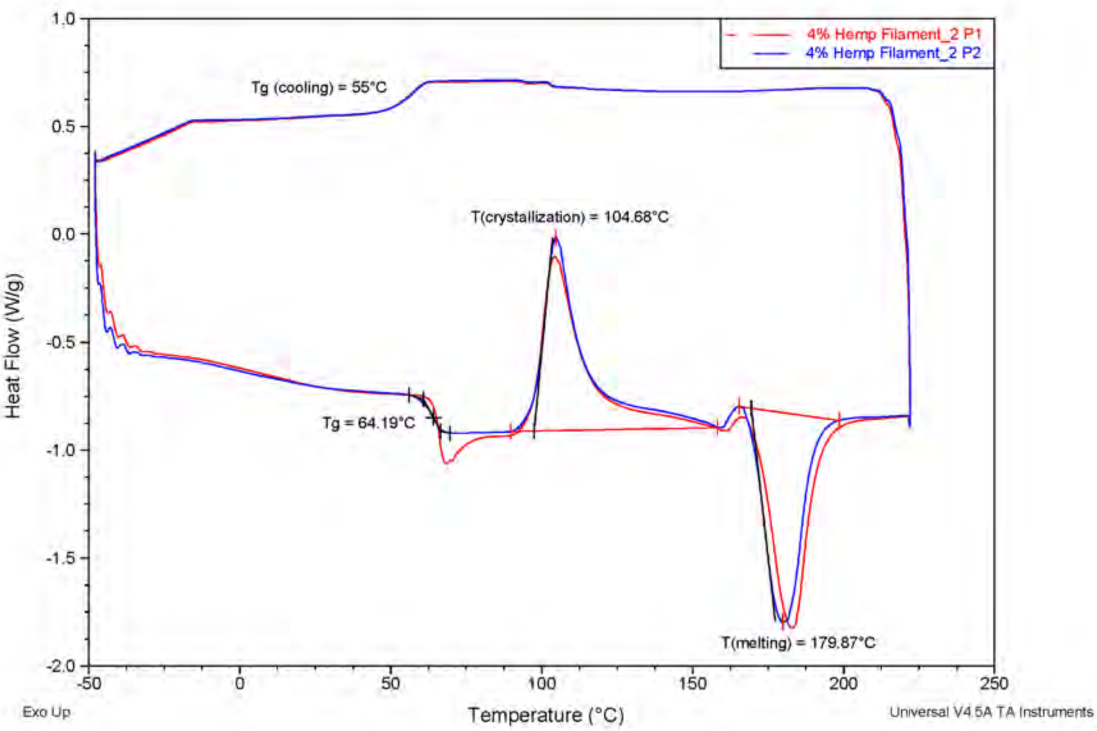

Figure B.2: DSC for 3DF 4 wt % hemp PLA.

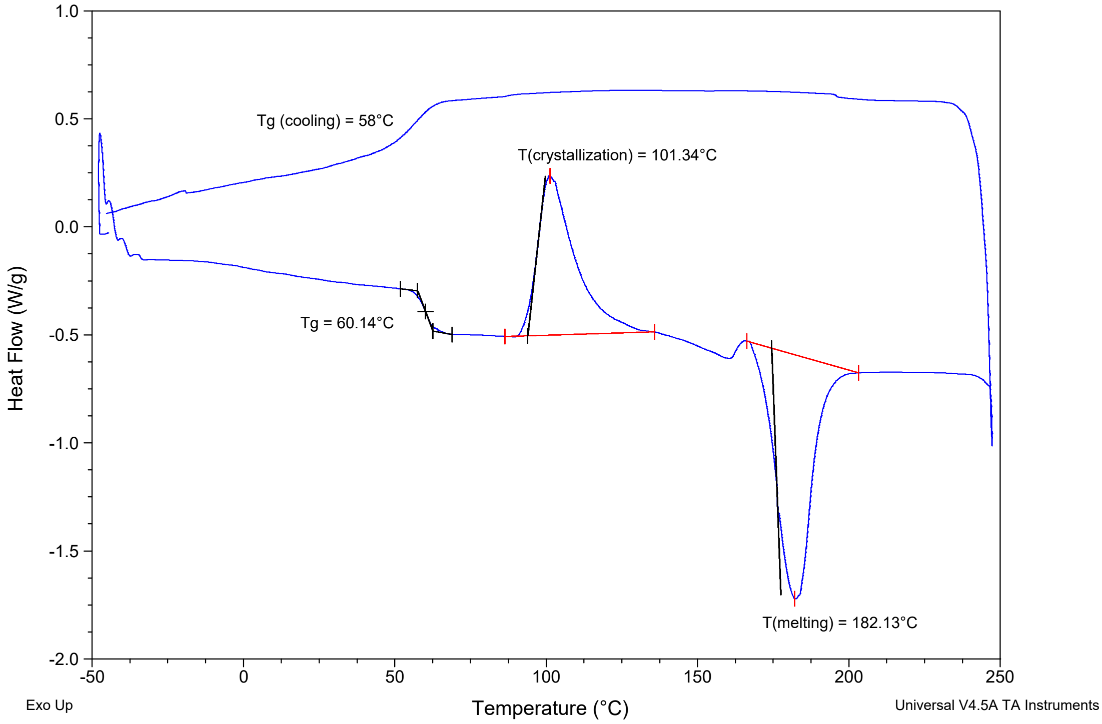

Figure B.3: DSC for P-P 5 wt % hemp PLA.

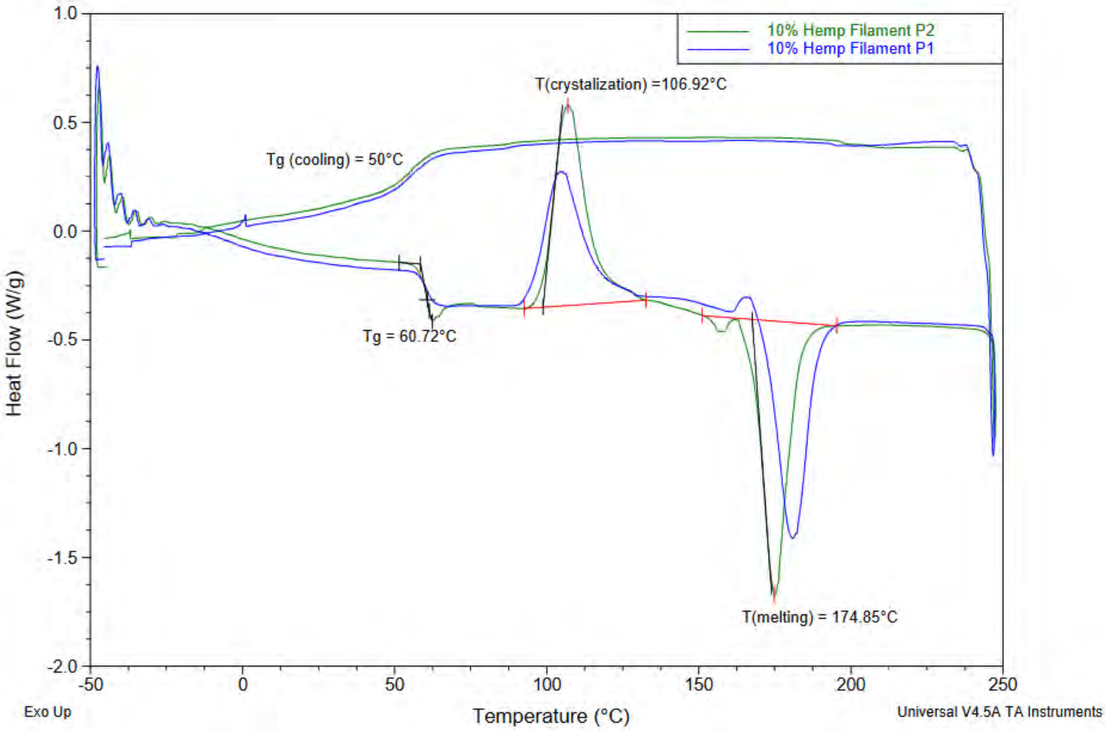

Figure B.4: DSC for P-P 10 wt % hemp PLA.

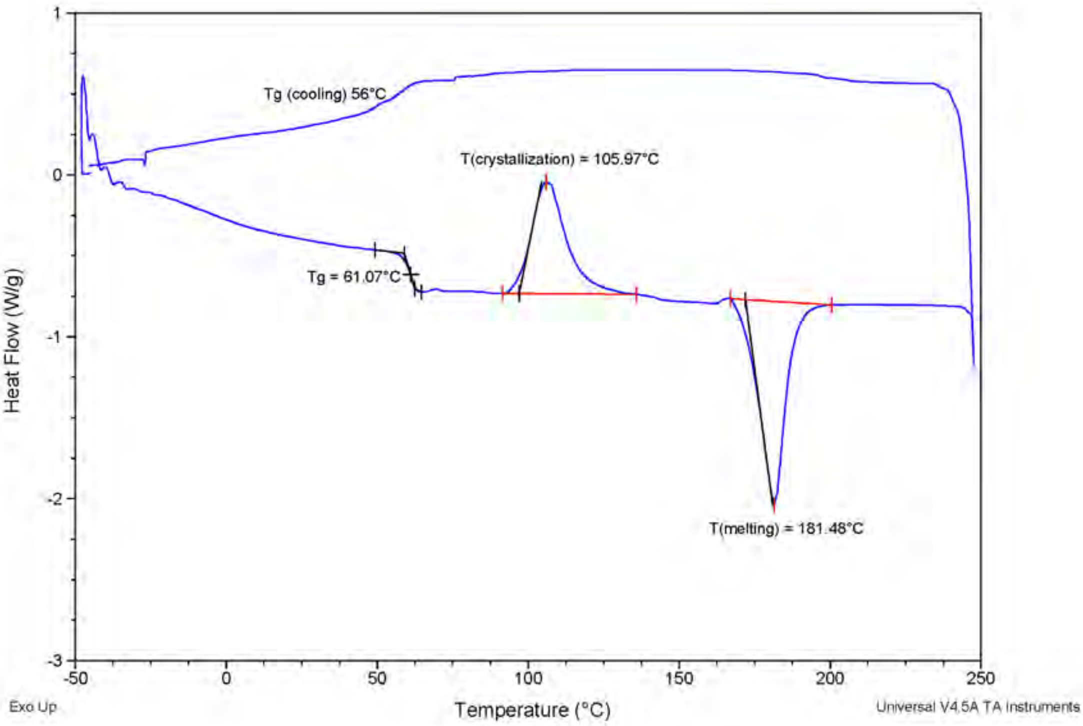

Figure B.5: DSC for P-P 15 wt% hemp PLA.

Appendix C

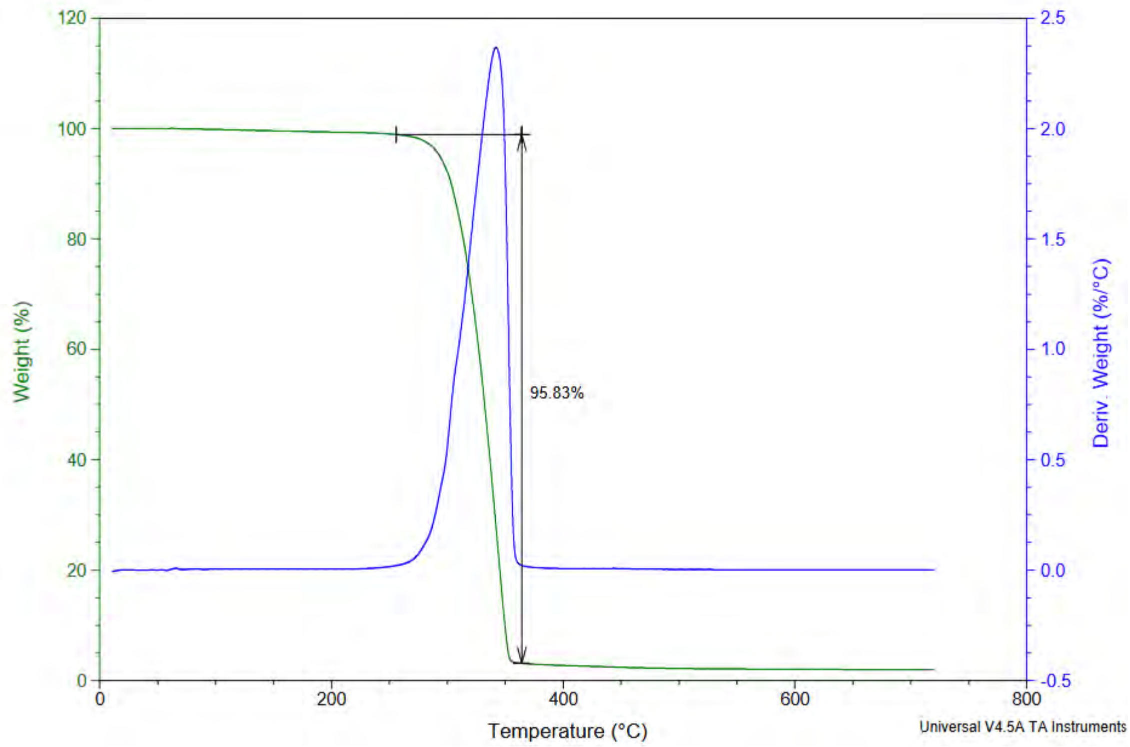

Figure C.1: TGA for 3DF 4 wt% hemp PLA.

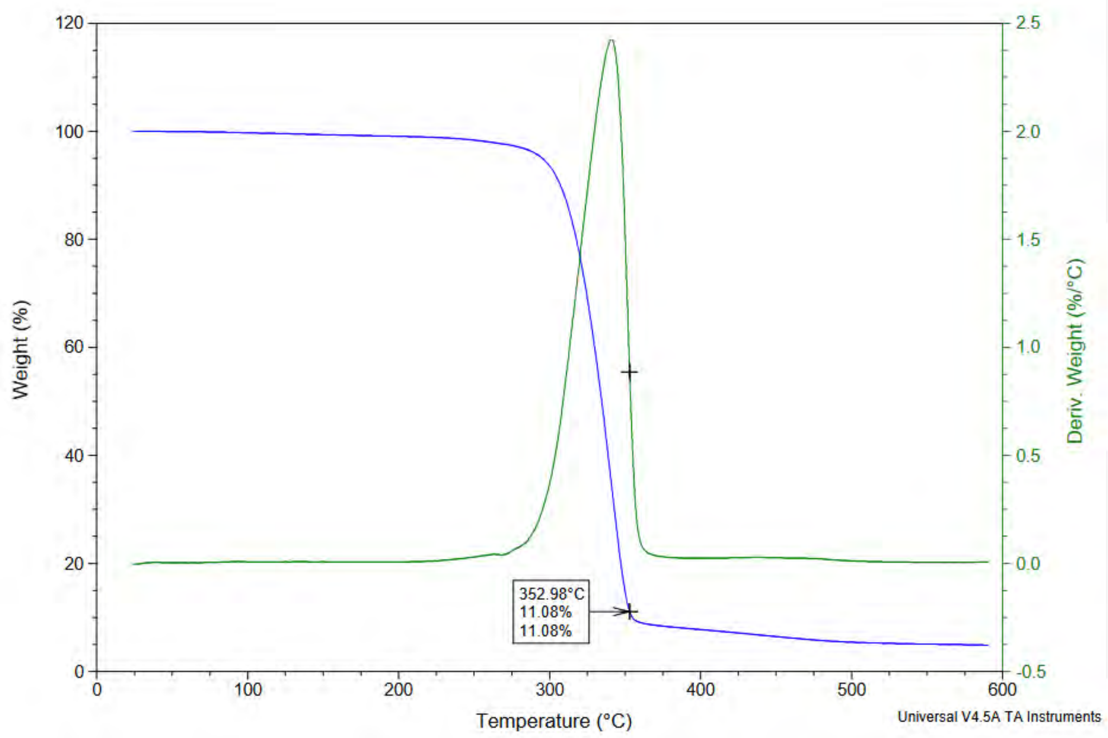

Figure C.2: TGA for P-P 10 wt% hemp PLA.

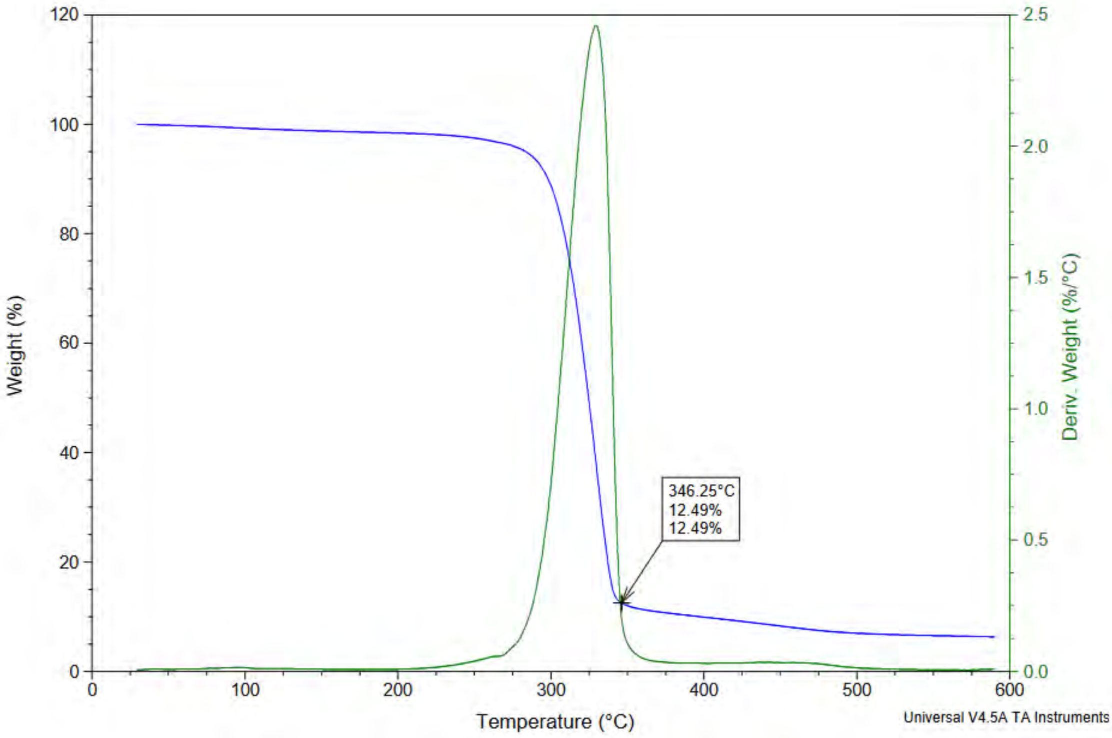

Figure C.3: TGA for P-P 15 wt % hemp PLA.

Appendix D

Figure D.1: 5, 10, 15 wt% hemp PLA (top to bottom) Protopasta custom filament

Figure D.2: 3D-Fuel PLA (Workday PLA) filament

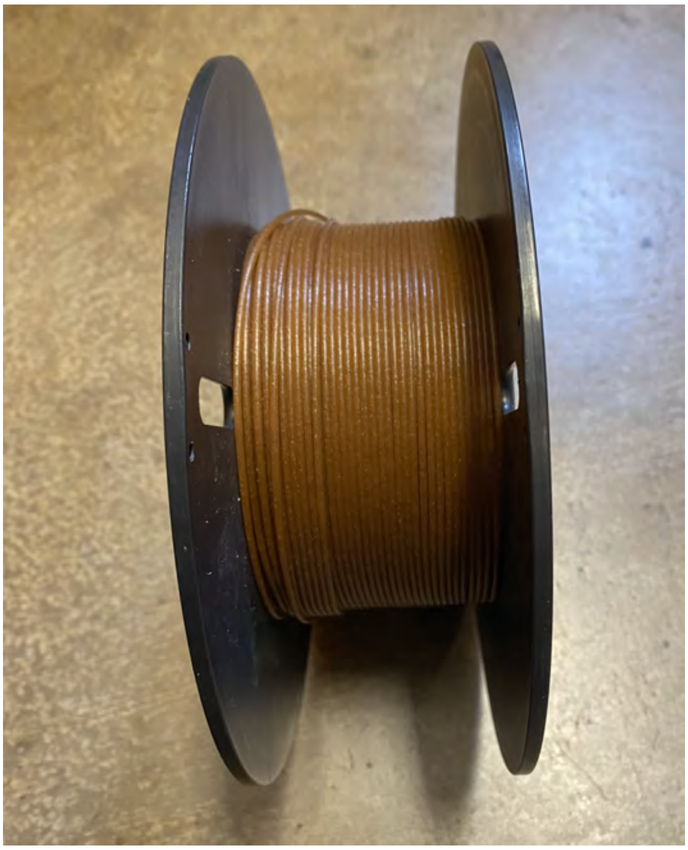

Figure D.3: 3D-Fuel 4 wt% Hemp PLA filament

Figure D.4: Collapsed 15 wt% Protopasta custom filament

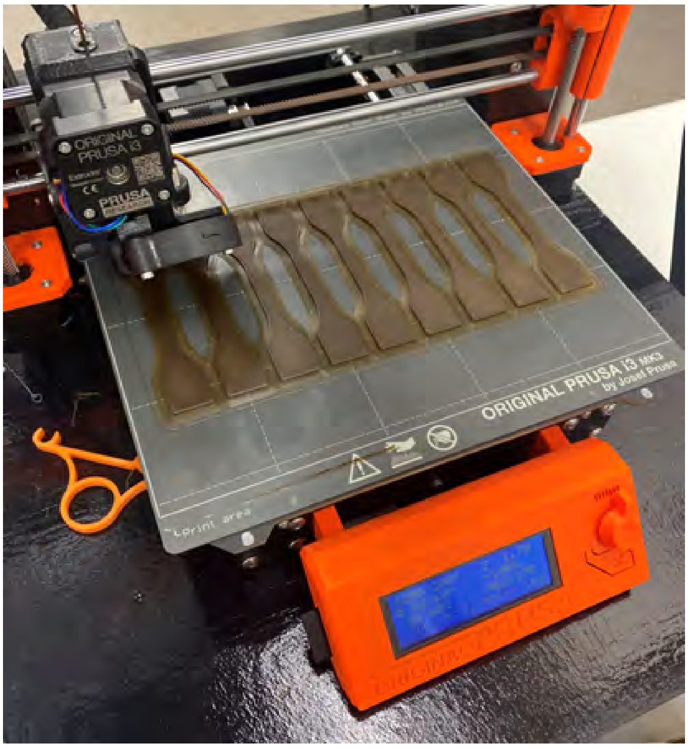

Figure D.5: 3D printing orientation of tensile bars