3D Printing of Natural Fibre Reinforced Recycled Polypropylene

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Abstract

The adverse effects that waste plastics are having on the environment is becoming increasingly apparent. However, the plastics recycling industry in New Zealand is entirely market driven, necessitating the development of new markets to account for increasing quantities of waste. Innovations in additive manufacturing (AM) have presented opportunities to recycle thermoplastics for use as AM feedstock material. Using waste thermoplastic materials to fabricate composites in this way, adds value to the polymer by enhancing mechanical and aesthetic properties. A range of composite filaments with differing fibre and gypsum weight contents were then produced using pre and post-consumer polypropylene (PP). The most successful filaments in terms of tensile properties consisted of 30 wt% harakeke in a post-consumer PP matrix which had a tensile strength and Young's modulus of 41MPa and 3.8 GPa respectively. Comparing these results to those of plain PP filament, showed improvements in tensile strength and Young's modulus of 77% and 275% respectively. Finally, a novel method of measuring shrinkage in 3d printed components was developed and used to compare relative shrinkage of different composites. The composite that showed the least shrinkage consisted of 30 wt% harakeke with a shrinkage value of 0.34% corresponding to a net reduction of 84% relative to plain PP.

Introduction

Almost 30 years since its conception, additive manufacturing also known as 3D printing, has gradually overcome its niche applications and is revolutionising all manner of practices within the manufacturing industry [1]. Fused deposition modelling (FDM) is a type of 3d printing in which a thermoplastic filament is melted and extruded through a circular nozzle. The movement of the nozzle is controlled using a 3-axis system allowing the molten plastic to be deposited onto a print bed. Thermoplastics are the preferred feedstock material, although a wide range of materials, including cement and composites, are compatible with the FDM process [2,3]. Generally, the thermoplastics used for FDM are restricted to amorphous polymers. This is largely because they exhibit a low degree of polymer shrinkage which is pivotal to the accuracy of components produced.

Polypropylene (PP) is a thermoplastic polymer derived from propene, a relatively inexpensive by-product of the oil refining process. As propene is inexpensive, virgin PP is relatively cheap when compared to other virgin materials. Because the recycled plastics industry is entirely market driven and the manufacturers preference for virgin material, the market price for recycled polypropylene varies between $100 -$200 per tonne. Despite the high quantity of PP being used in consumer products, the low market value often does not warrant the cost of collection and sorting resulting in a lot of the polymer going to landfill.

Properties of recycled plastics can be modified and improved in a number of ways. The most common of which is to add a certain percentage of virgin material to the recycled mix. This method increases the average linear chain length, therefore increasing the properties of the material as a whole. Another method of improving the mechanical properties is to blend in other additives such as fibre to form composite materials. Blending in reinforcing fibres offers a range of advantages including increased mechanical properties and reductions in shrinkage [4-7]. This can add value to recycled polymer and potentially open up much needed markets for the waste material.

This paper explores the viability of reinforcing recycled polypropylene with natural fibres and gypsum powder for applications in FDM. Composite feedstock materials with varying reinforcement content were fabricated before tensile testing both feedstock and 3d printed samples. A novel method of measuring the shrinkage effects in 3d printed components was developed and used to obtain relative differences between the different composites.

Materials and methods

Pre-Consumer Recycled Polypropylene: Pre-consumer recycled polypropylene was supplied in granule form by Astron Plastics Group, Auckland. Large woven polypropylene bags, as shown in Figure 1, were supplied by Transpacific Recycling Centre located in Tauranga. The bags are a currently underutalised post-consumer product resulting from a large local salt importer. Although the bags are predominantly made from woven polypropylene, they are stitched together using polyester thread. There was no practical way to remove all traces of this thread, so the two materials were processed in a combined form. In addition to the impracticality of separation, the literature suggests possible mechanical property enhancements when combining polyester fibres in a polypropylene matrix [8-10]. Bags were manually cut into pieces approximately 200mm x 200mm and then granulated using a Castin laboratory scale granulator. The resulting plastic strands (**Error! Reference source not found.**) were then washed with dishwashing liquid in hot water before being rinsed thoroughly and oven dried. Fibres were then compounded in a ThermoPrism TSE-16-TC 16mm screw diameter co-rotating twin screw extruder (later referred to as the 16mm extruder) before being granulated. Before extrusion, polymer samples were dried in a convection oven at 105o C for a minimum of 24 hours.

Figure 1, Post consumer polypropylene woven bags.

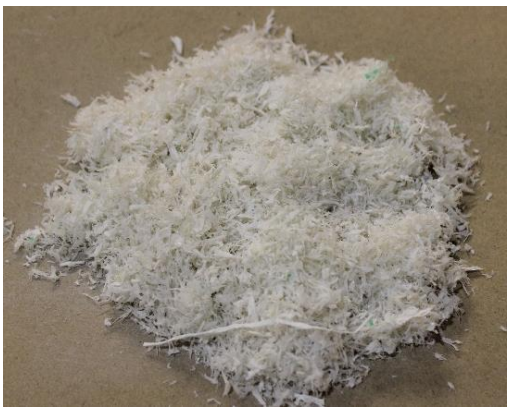


Figure 2, Shredded polypropylene bags

Hemp fibre: Hemp fibre was locally grown from October 2013, harvested in February 2014 after a 120 day cycle, and subsequently donated by the Hemp Farm NZ Ltd. Green hemp stalks were air dried before the bast fibre was separated from the stalks by hand. The bast fibre was thoroughly inspected to ensure it was free of visible defects before being chopped into shorter lengths using a laboratory scale Castin granulator. An 8 mm sieve was used to regulate the size of the resultant fibre which produced an average fibre length of 10mm**.**

Harakeke fibre: Mechanically separated harakeke was air dried and supplied to the University in bundle form by the Templeton Flax Mill in 2014. Fibre bundles were manually cut with scissors into 200mm lengths to avoid entanglement in the granulator blades. The chopped fibres were then fed into a Castin laboratory scale granulator using the same 8mm sieve as used with the hemp fibre giving a similar average material length.

Alkali treatment: The digestion of hemp and harakeke fibre was performed at high temperatures and pressures within a lab scale digester. Sodium hydroxide (NaOH) powder and sodium sulphite ($NaSO_3$) pellets, with a purity level of 98% were acquired from Scharlau Chemie S.A. Oven dried hemp or harakeke fibres were weighed into 90g samples and carefully placed into one of four the steel canisters used to contain the fibre/solution mixtures inside the larger pressurized tank of the digester. For hemp fibres, a solution containing 36 g of NaOH and 684 ml of water was mixed in a conical flask. Four batches of this solution were prepared and combined with the fibres inside the canisters immediately prior to treatment. For harakeke fibres, the solution containing 36 g of NaOH, 14.4 g $NaSO_3$ and 670 ml of water was prepared for use in a similar way.

Figure 3 shows the temperature profiles used for both harakeke and hemp fibre treatment. An investigation conducted by Efendy *et al* [11] had previously determined that these treatments provided the optimal thermal parameters for the aforementioned fibre species.

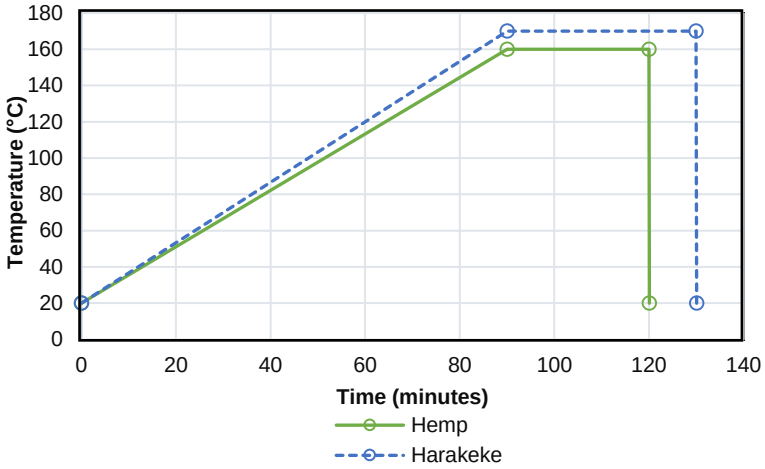


Figure 3, Digester cooking cycles for hemp and harakeke

Once the treatment cycle was completed, the fibres were removed from the canisters and thoroughly washed until all remnants of the alkali solution had been removed. The washed fibre pulp was then separated into smaller pieces by hand before being placed in an oven to be dried at 105^{o} C for a minimum of 24 hours. Dried fibre clusters were then put through the granulator for separation prior to composite fabrication.

Recycled Gypsum Powder: Recycled gypsum board was obtained from building developments underway within the Hamilton area. All gypsum board used in experiments was manufactured by Gib Plasterboards. The constituents for Gib plasterboard are detailed in Table 1.

Table 1, Gib Plasterboard gypsum board constituents

Component	Typical %
Gypsum	93.0%
Paper	6.0%
Acid Modified Wheat or Corn Starch	0.5%
Sugar	> 0.5%
Calcium Naphthalene Sulphonate	> 0.5%
Boric Acid	> 0.5%
Fibreglass	> 0.5%
Vermiculite	> 0.5%
Fly Ash	> 0.5%

To separate the gypsum powder from the paper, boards were cut into 100 x 300 mm sheets and fed into a Filamaker lab scale shredder. The resulting mixture was then put through a 2mm sieve which effectively removed the majority of the paper. The sieved powder was then processed to a smaller size in a Nutribullet food blender for 5 minutes and placed in an oven at 105° C for 24 hours. The resulting particles were measured using a Malvern Mastersizer 2000 and the results are shown in **Error! Reference source not found.**. The instrument was set to analyse with a refractive index of 1.525 which is the recorded standard for measuring gypsum powder.

Table 2, Particle size for recycled gypsum powder

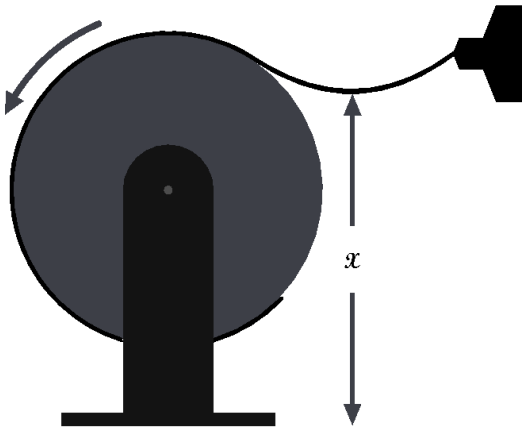
Lower 10% (µm)	Surface weighted mean (µm)	Volume weighted mean (µm)	Upper 10% (µm)
> 3.766	8.429	95.775	< 236.466

Composite Fabrication: Differences in densities between the pre-consumer PP granules, fibre and mineral additives resulted in poor fibre distribution prior to compounding leading to blockages within the extruder and poor fibre adhesion within the matrix. To avoid this, oven dried constituents were weighed and spread in even layers in a square plastic container. A flat scoop was then used to pick up portions of the mixture and force feed it into the 16mm extruder to ensure all fibre was included.

Post-Consumer PP Composite Compounding: Composite constituents were weighed and placed in a sealed plastic bag where they were shaken until relatively even distribution was achieved. The similar uncompressed densities of the hemp, harakeke and PP strands (0.0618, 0.0589 and 0.0686 g/cm^3 respectively) lead to better mixing than obtained with polymer granules. The premixed materials were then force fed into the 16mm extruder in a similar way to pre-consumer PP using the same temperature profile.

Filament Fabrication: Polymer or composite granules of each different combination were stave fed at a rate of 12 rpm into a Labtech 1201-LTE20-44 20mm screw diameter twin screw extruder (later referred to as the 20mm extruder). The screw speed was maintained at 50 rpm for composites up to 30wt% fibre, but decreased with increasing fibre content to maintain a pressure near the end of the barrel below 70% of the allowed maximum pressure.

With a screw speed of 50 rpm, the filament exited the die at approximately 0.05 m/s and was still in a semi molten form. To increase dimensional consistency, a filament spooling machine was designed and built (Figure 5). By matching the rotational speed of the spooling machine with the exit velocity of the filament and controlling the vertical height (labelled x in Figure 4) the filament diameter could be controlled.



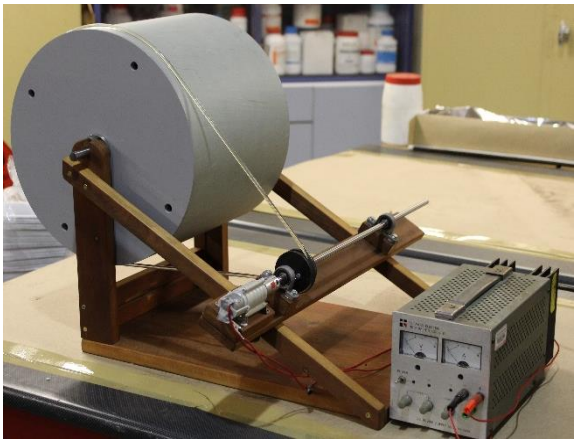


Figure 4, Filament dimension control

Figure 5, Filament spooling machine

Composite filaments were produced containing 10, 20, 30, 40 and 50% hemp and harakeke using both pre and post-consumer polypropylene matricies. The same weight % intervals were also used to produce gypsum composites although these were only produced using pre-consumer recycled polypropylene as a matrix material.

3d printing: The conventional method of printing onto a heated print bed did not work for polypropylene due to lack of adhesion. A 5mm thick polypropylene sheet was retrofitted into the print bed and used for all 3d printing. Prior to printing shrinkage samples, the filament diameter was measured with digital callipers and input into printing software. A rectangular calibration sample measuring 15 x 5 x 1mm was then printed through a 1mm die. As the sample was printed, the width of the extruded bead labelled 'b' in **Error! Reference source not found.** was carefully measured using markings on the print bed and a magnifying glass. The filament diameter parameter in the software was altered until the value of b was measured to be 1.1mm. The software would then print each bead such that distance 'a' was equal to 2mm making the overlapping distance 'c' approximately equal 0.2mm.

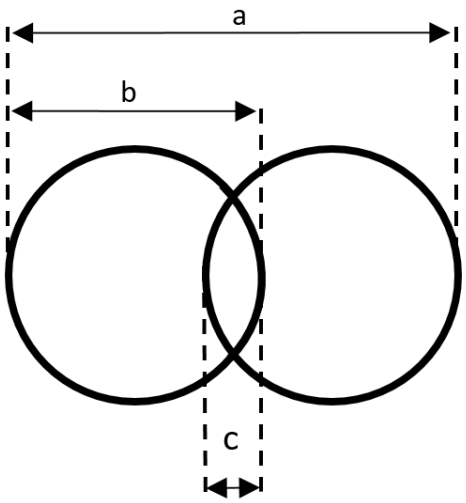


Figure 6, Cross section of 3d printed intersecting beads

Composite Tensile Testing: The ultimate tensile strength and Young's modulus of printed filament specimens were determined using an Instron 33R4204 universal testing machine equipped with a 5 kN load cell. A standard cross-head speed of 1 mm/min was applied to all filament specimens. Strain was measured using an Intron 2630-112 extensometer fitted to the samples. The gauge length of 50 mm was assigned to each of the filament samples. All composite specimens were tested until failure, whereas unfilled polypropylene specimens were only tested to a point of maximum stress due to excessive necking. The average of five measurements was taken as the experimental value.

To investigate fibre-matrix adhesion and fibre alignment, the fracture surfaces of selected tensile test specimens were observed using a Hitachi S-4100 Field Emission Scanning Electron Microscope (SEM)

Shrinkage testing of 3d printed parts: To measure the effects of shrinkage specific to fused deposition modelling, a novel method was developed and introduced simulating a problem found in early testing where printed samples would peel upwards from the bed and eventually become dislodged. This was found to significantly reduce the accuracy of the resulting print.

Test samples with dimensions indicated in Figure 7 were printed in a single layer 0.8mm thick. Because the sample solidified while adhering to the print bed the warping effects of shrinkage were minimised. Once the sample had been printed digital callipers were used to measure the length of each arm. A razor blade was then used to dislodge the arms from the bed while leaving the centre in attached. A second layer was then printed on top of the first sample using a Z offset of 0.8mm.

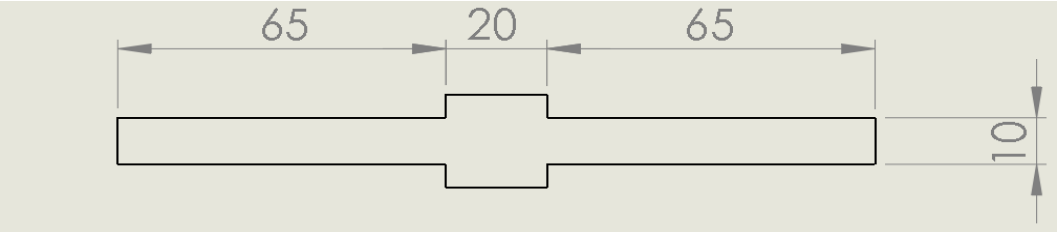


Figure 7, Shrinkage test dimensions (mm)

Figure 8, 3d printed sample showing exaggerated shrinkage effect

As the second layer solidified the shrinkage caused each end to distort in a controlled direction shown in Figure 8. The radius of curvature was used to establish a relative shrinkage value useful for comparing shrinkage properties inherent in each material. Full details of test sample geometry and formulas used are shown in Appendix 1.

Results and discussion

Tensile Strength and Stiffness of Hemp and Harakeke Fibre 3d Printing Filament: The tensile strength and Young's modulus of 3mm diameter composite filaments are compared to plain pre-consumer PP filaments in **Error! Reference source not found.** and 10 respectively. The results clearly show an increase in strength and stiffness as a result of a higher fibre weight fraction. This is supported by highly significant R^2 values and positive gradients of regression lines fitted to both graphs. Hemp and harakeke fibres show similar reinforcing qualities with any variation being well within the standard deviation error. The greatest improvement was for 30 wt% harakeke which had a tensile strength and Young's modulus 52% and 147% higher than plain polypropylene. Increasing the fibre content from 10% to 20 wt% raised the tensile strength and Young's modulus by around 8 and 720 MPa respectively. However, increasing the fibre content from 20% to 30 wt% gave lower increases of 2.5 and 430 MPa. This could possibly be due to reduced fibre wetting with the increased fibre content resulting in underutilised reinforcing fibre.

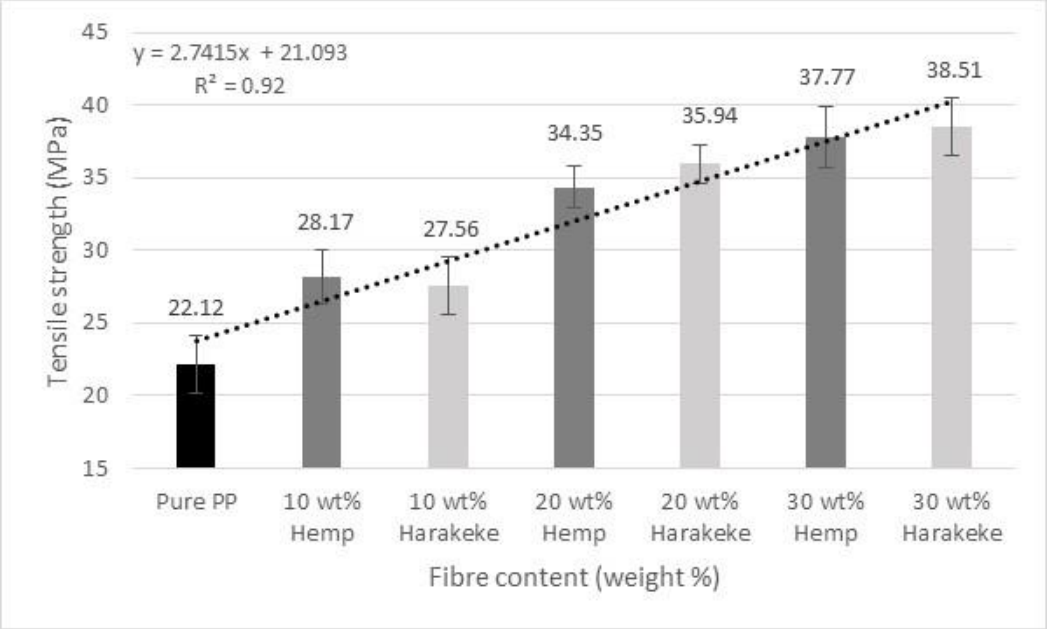


Figure 9, Tensile strengths of the 3mm composite filaments produced using pre-consumer PP as a matrix (MPa).

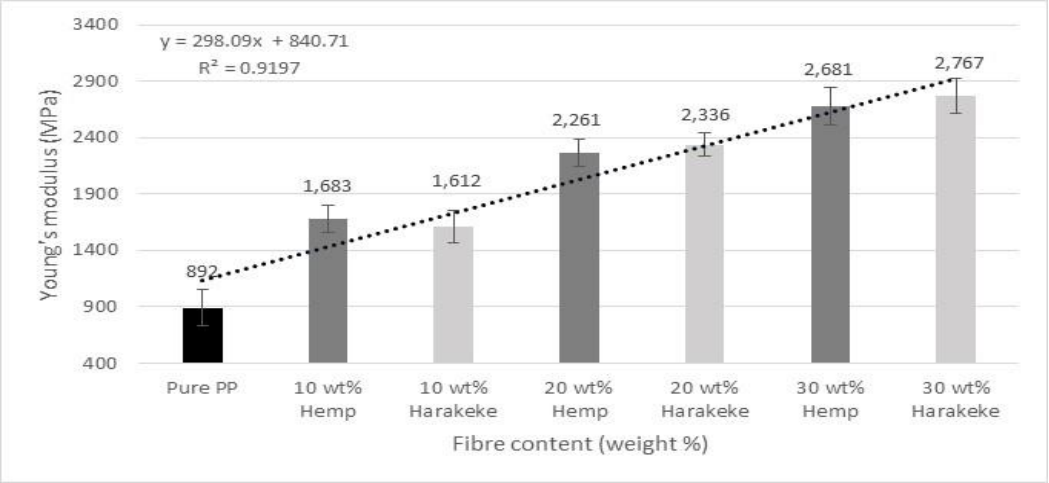


Figure 10, Young's modulus of composites produced using pre-consumer PP as a matrix (MPa).

Research conducted alongside with this research at the University of Waikato investigated the effects of E-glass fibre reinforcement in the same pre-consumer polypropylene; glass fibre/polypropylene 3d printing filament was fabricated using similar temperature profiles and processing machinery [12]. However, the fibre and polypropylene matrix were compounded using a twin screw compound mixer instead of a twin screw extruder. The tensile strength and Young's modulus results for glass fibre/polypropylene 3mm filaments are shown in **Error! Reference source not found.** below. Glass fibre can be seen to result in tensile strength enhancements lower than those of natural fibre composites. However, the Young's modulus values of glass fibre composites are between 18 and 21% higher than those reinforced with harakeke fibre. The study attributed poor mechanical performance to severe fibre length reductions caused by the brittle failure of glass fibres.

Table 3, Mechanical properties of natural fibre compared to glass fibre reinforced 3d printing filament

Sample	Tensile strength (MPa)	Net increase (%)	Young's modulus (MPa)	Net increase (%)
Plain PP	22		892	
10 wt% Glass	29	33	1995	124
20 wt% Glass	32	39	2016	126
30 wt% Glass	36	62	3386	280
10 wt% Harakeke	28	25	1612	81
20 wt% Harakeke	36	62	2336	162
30 wt% Harakeke	39	74	2767	210
10 wt% Hemp	28	27	1683	89
20 wt% Hemp	34	55	2261	153

30 wt% Hemp	38	71	2681	201

Influence of Fibre Content on Filament Fabrication: Increasing the fibre content from 30 to 40 wt% gave rise to a sharp increase in die pressure and screw torque which forced a reduction in screw speed from 50rpm to 30 rpm to avoid damage to the machine. Reducing the screw speed corresponded to a considerable reduction in exit velocity allowing the composite more time to solidify prior to spooling. This rendered the spooling machine ineffective as it relies on the filament being malleable enough to wrap around the cylinder. Given the ineffectiveness of the spooling machine, filament with fibre content in excess of 40 wt% was not dimensionally consistent enough for application in 3d printing.

shows the increasingly coarse surface finish resulting from increased fibre content. Filaments containing 10 wt% fibre content gave a semi-gloss type smooth finish which dulled as the fibre content increased. A 'sharkskin' effect began to develop with the 40 wt% samples and was very severe when fibre content was increased to 50 wt%. This effect could possibly be reduced by increasing the die temperature or modifying the die to reduce shear stress from die walls [13].

Figure 11, Influence of fibre content on surface finish

Tensile Strength and Stiffness of Printed Samples: The trend observed in 3d printed 1mm filament (12 and 13) is very similar to that seen in the 3mm filament providing evidence for the theory that geometry has a significant influence on 3d printed components. However, the results show a significant decrease in tensile properties when compared to the 3mm filament (the only difference being the 3d printing extrusion process). Contrary to predictions of increased mechanical properties as a result of increased fibre alignment. The fact that the plain polypropylene filament showed a drop in strength and Young's modulus similar to that of composite samples indicates that the cause is largely related to issues with the polypropylene.

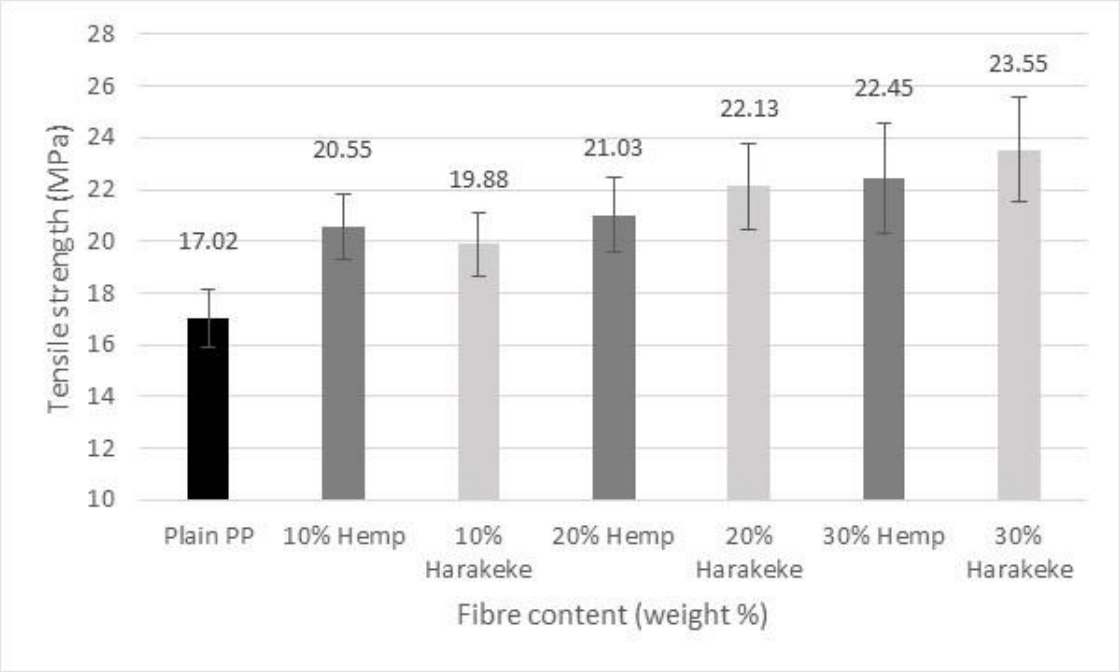


Figure 12, Tensile strength of 3d printed 1mm pre-consumer filament

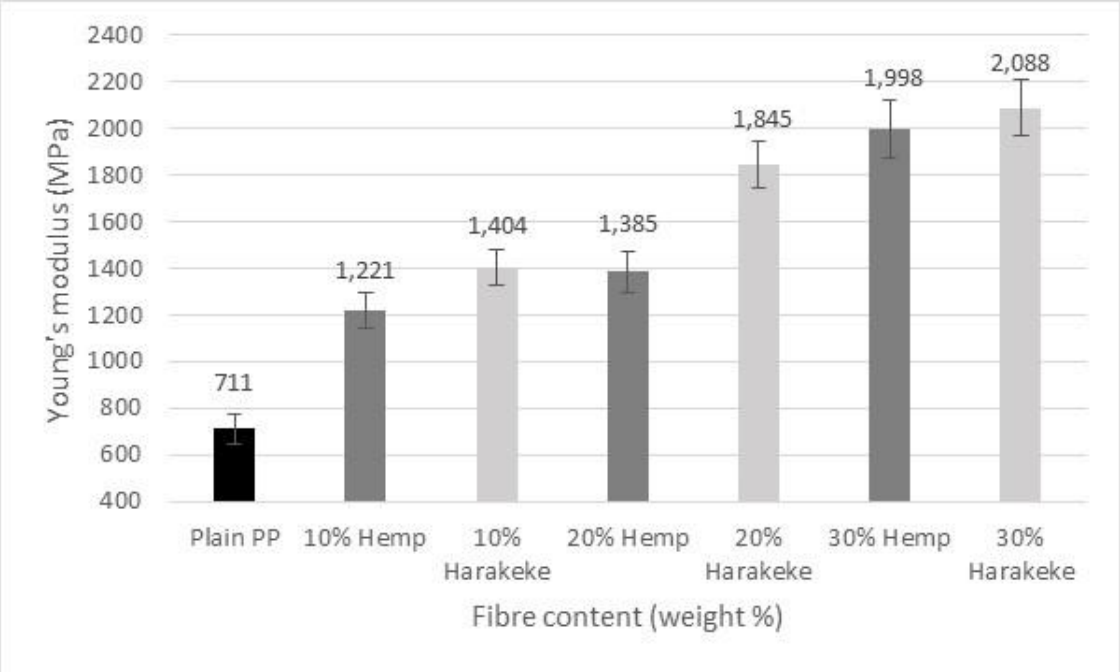


Figure 13, Young's modulus of 3d printed 1mm pre-consumer filament

Tensile Strength and Stiffness of Recycled Gypsum 3d Printing Filament: Gypsum particle reinforced pre-consumer polypropylene composites were fabricated using the same processing temperatures as fibre reinforced composites. Unlike composites containing 40 and 50 wt% fibre, there was no significant increase in torque or pressure when extruding with 40 and 50 wt% gypsum.

The tensile strength and Young's modulus of pre-consumer polypropylene/ gypsum powder composites containing 10 to 50 wt% gypsum are shown in Figure 14 and 15. A small increase for samples in tensile strength of 9% relative to plain PP was observed containing 10 wt% gypsum. As the gypsum content increased further there was a gradual decline in tensile strength to reach a reduction of 2% for the 50 wt% gypsum. The Young's modulus, however, showed a reasonably linear increase with 50 wt% gypsum composites having a maximum improvement of 88%. The decrease in tensile strength could have been caused by the agglomeration of the gypsum particles with increasing gypsum content. Agglomerated particles have the effect of decreasing interfacial adhesion and act as stress concentration points [14]. The gradual increase in stiffness with increased particle content can be explained by a reduction in polymer chain mobility.

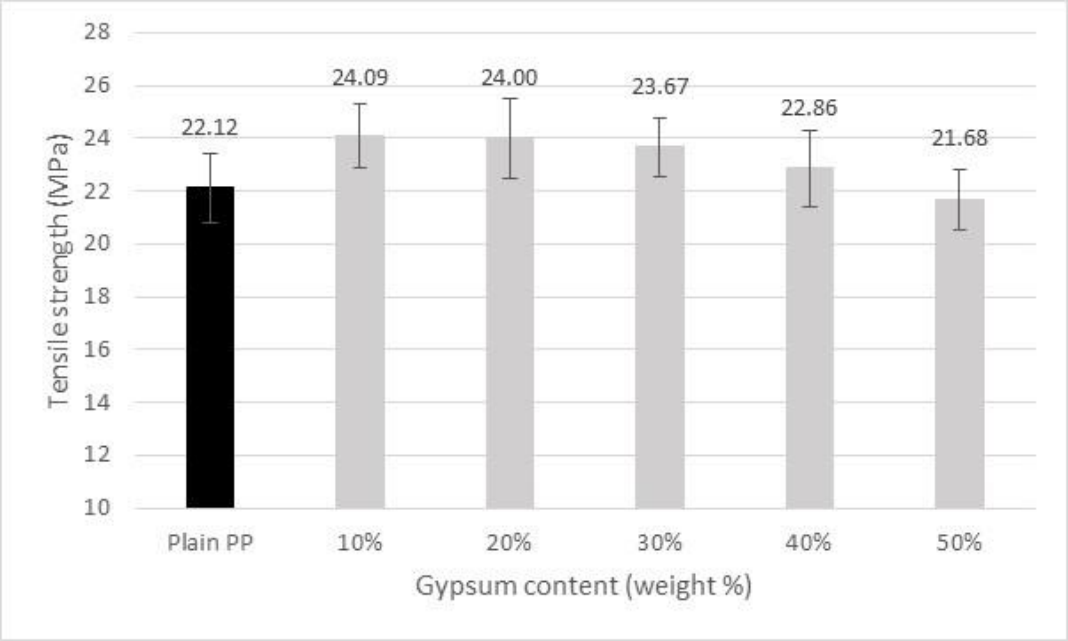


Figure 14, Tensile strength of composites containing gypsum compared to plain PP

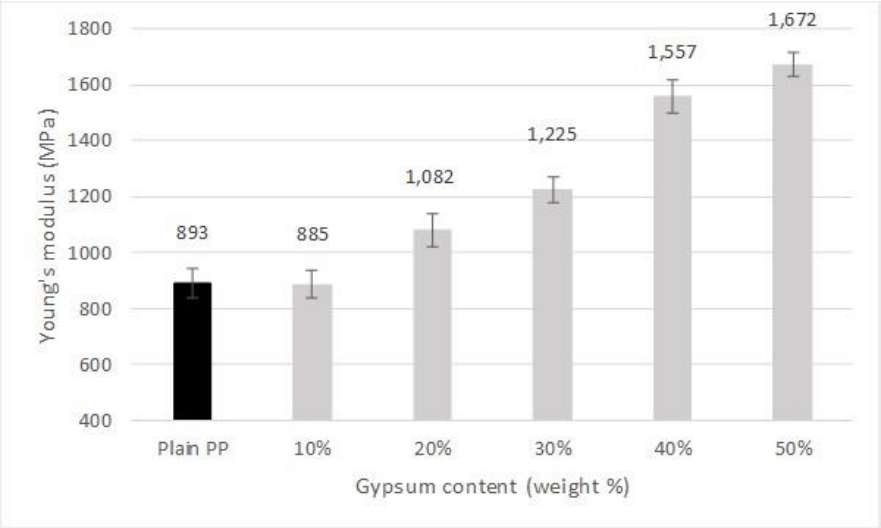


Figure 15, Young's modulus of composites containing gypsum compared to PP

Microscopic Evaluation: At reinforcement content < 30 wt% the gypsum appeared to be broken down and dispersed throughout the matrix, with a mixture of small particles and crystal clusters (Figure 4.18 and 4.19). As the gypsum content increased, the crystals grouped together to form larger conglomerates of gypsum shown in Figure 16 and 17.

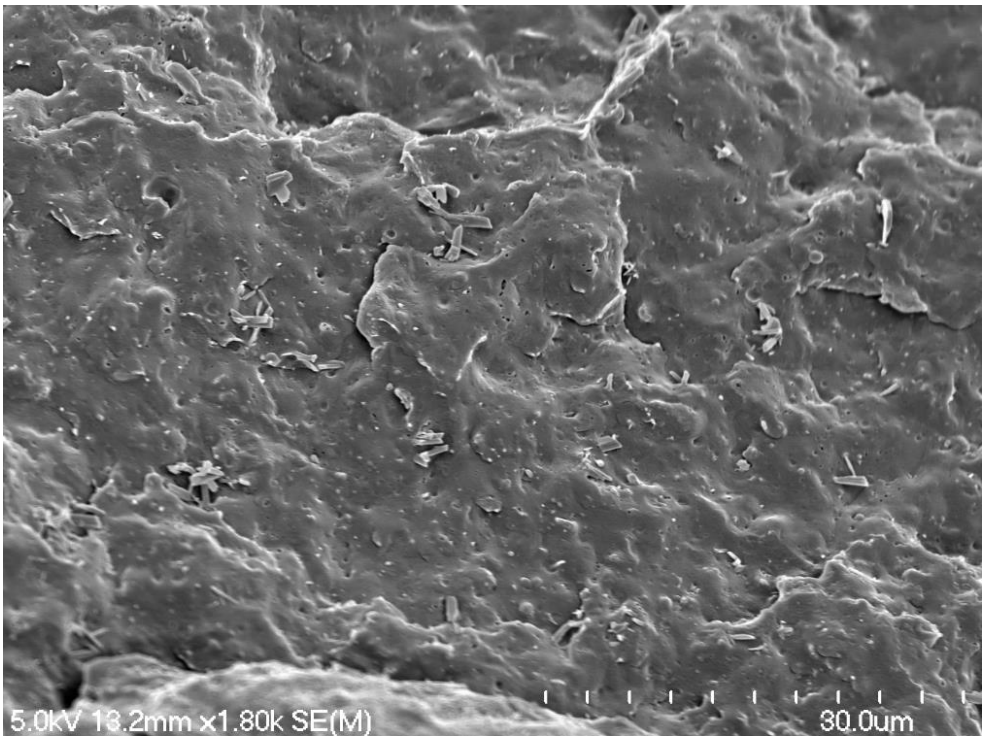


Figure 16, 20 wt% gypsum filament showing particles distributed with gypsum crystal clusters

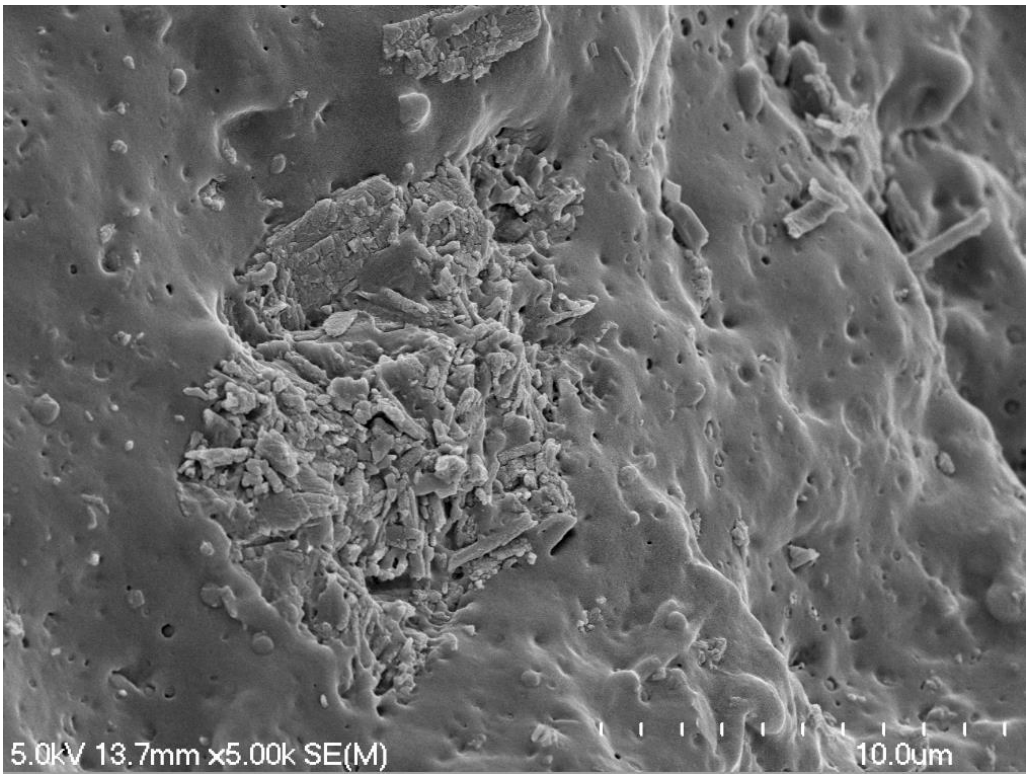


Figure 17, 50 wt% gypsum fracture surface showing crystal cluster.

Shrinkage: The shrinkage results for hemp, harakeke and gypsum composites are shown in below. Natural fibre additives appeared more effective than gypsum in terms of shrinkage reduction. The greatest reduction was that of 30 wt% harakeke filament with a shrinkage of 0.34% corresponding to a 84% reduction compared to plain polypropylene.

Sample	Shrinkage (%)	Net reduction (%)
Pre-consumer PP,	2.13	-
10% Hemp	1.17	45
20% Hemp	0.65	69
30% Hemp	0.47	78
10% Harakeke	1.18	45
20% Harakeke	1.2	44
30% Harakeke	0.34	84
10% Gypsum	1.53	28
20% Gypsum	1.51	29
30% Gypsum	1.39	35
40% Gypsum	1.26	41
50% Gypsum	1.15	46

Sample	Shrinkage (%)	Net reduction (%)
Pre-consumer PP,	2.13	-

Table 4, Shrinkage values for pre-consumer polypropylene composites

10% Hemp	1.17	45
20% Hemp	0.65	69
30% Hemp	0.47	78
10% Harakeke	1.18	45
20% Harakeke	1.2	44
30% Harakeke	0.34	84
10% Gypsum	1.53	28
20% Gypsum	1.51	29
30% Gypsum	1.39	35
40% Gypsum	1.26	41
50% Gypsum	1.15	46

Figure 18, Plain polypropylene shrinkage sample

Figure 19, 30 wt% hemp shrinkage sample

The shrinkage experienced in all composites generally decreased with increased additive content. This can be seen by following the reductions with the addition of 30 wt% hemp shown in Figures 18 and 19. It also implies that if the fibre content is increased beyond 30 wt% further reductions in shrinkage could be achieved; the difficulties in processing higher fibre contents were not

present when processing gypsum composites. This shows that potentially more effective shrinkage reductions could be achieved by fabricating a gypsum/fibre hybrid composite without encountering similar processing difficulties.

Tensile Strength and Stiffness of Hemp and Harakeke Reinforced 3d Printing Filament

The tensile strength and Young's modulus results for post-consumer PP/ fibre composites are shown in Figures 20 and 21 below. The 30 wt% harakeke samples showed the most significant improvements in strength and Young's modulus of 76.8% and 274.9% respectively compared to plain post-consumer PP. The post-consumer PP filament performed better than the pre-consumer with higher strength and Young's modulus values of 6% and 14% respectively. This is believed to be a result of either the polymer being a superior grade and/or the polyester fibre content within the post-consumer PP. The increased performance of all post-consumer composites relative to those fabricated using pre-consumer PP also justify this observation.

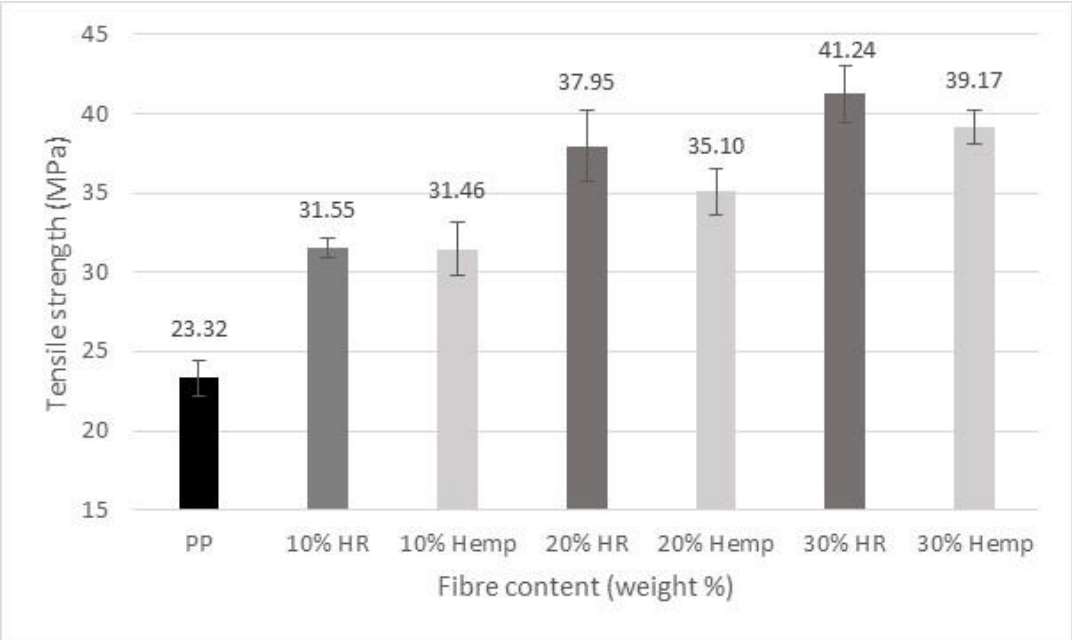


Figure 20, Tensile strength of post-consumer PP/Harakeke and PP/hemp fibre composites (MPa)

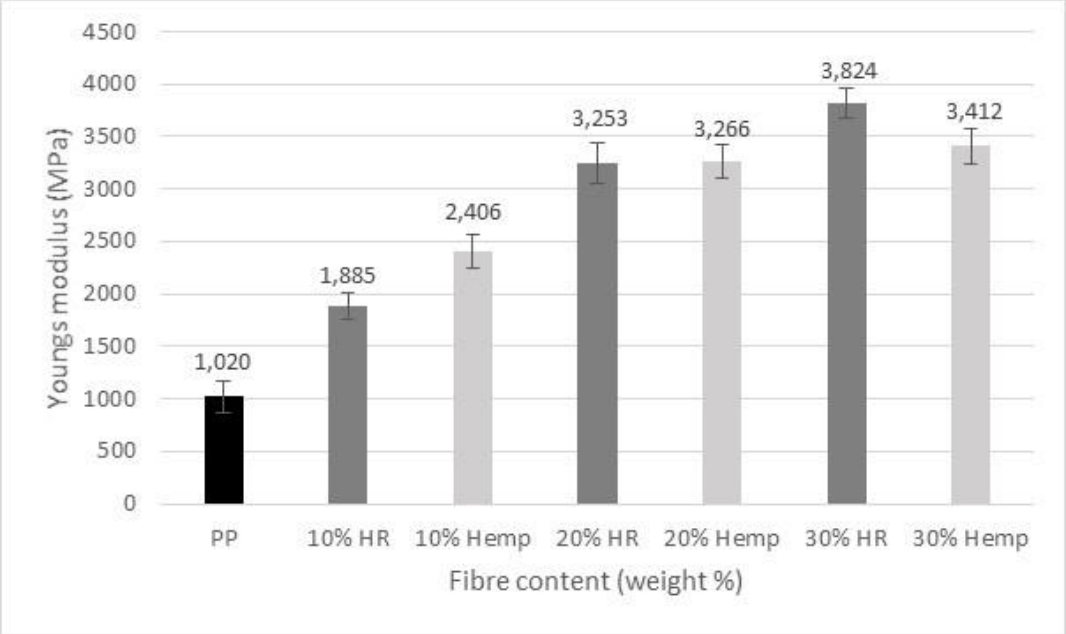


Figure 21, Young's modulus of post-consumer PP/Harakeke composite filament (MPa)

When compared to glass fibre/pre-consumer PP composites in the parallel study mentioned previously, post-consumer PP reinforced with natural fibre showed greater improvements in both strength and Young's modulus as shown in **Error! Reference source not found.** below. The 30 wt% harakeke composite had a tensile strength and Young's modulus 15% and 13% higher than composites fabricated using 30 wt% glass fibre

Table 5, Comparison of pre-consumer PP/ glass fibre composites with post-consumer PP natural fibre composites

Sample	Tensile strength (MPa)	Net increase (%)	Young's modulus (MPa)	Net increase (%)
Plain PP	23.32	-	1020	-
10 wt% Glass	29.33	26	1995	96
20 wt% Glass	30.64	31	2016	98
30 wt% Glass	35.78	53	3386	232
10 wt% Harakeke	31.55	35	1885	85
20 wt% Harakeke	37.95	63	3253	219
30 wt% Harakeke	41.24	77	3824	275
10 wt% Hemp	31.46	35	2406	136
20 wt% Hemp	35.1	51	3266	220
30 wt% Hemp	39.17	68	3412	235

Microscopic Evaluation: The SEM micrograph shown in Figure 22 clearly show polyester fibres protruding from the fracture surface of 30 wt% harakeke/post-consumer PP samples. The length of the protruding fibres is indicative of poor interfacial matrix adhesion between polyester and polypropylene. In addition to interfacial adhesion implications, protruding fibres also indicate a degree of fibre alignment parallel to flow direction. Further evidence of fibre alignment.

The fracture surface shown in **Error! Reference source not found.** shows evidence of harakeke fibre fracture close to the surface but also evidence of fibre pull out. This could suggest that although there was good interfacial adhesion, a proportion of the harakeke fibres used were below the critical reinforcement length. The same figure shows a large void that previously housed a polyester fibre providing a relative size comparison to harakeke fibres.

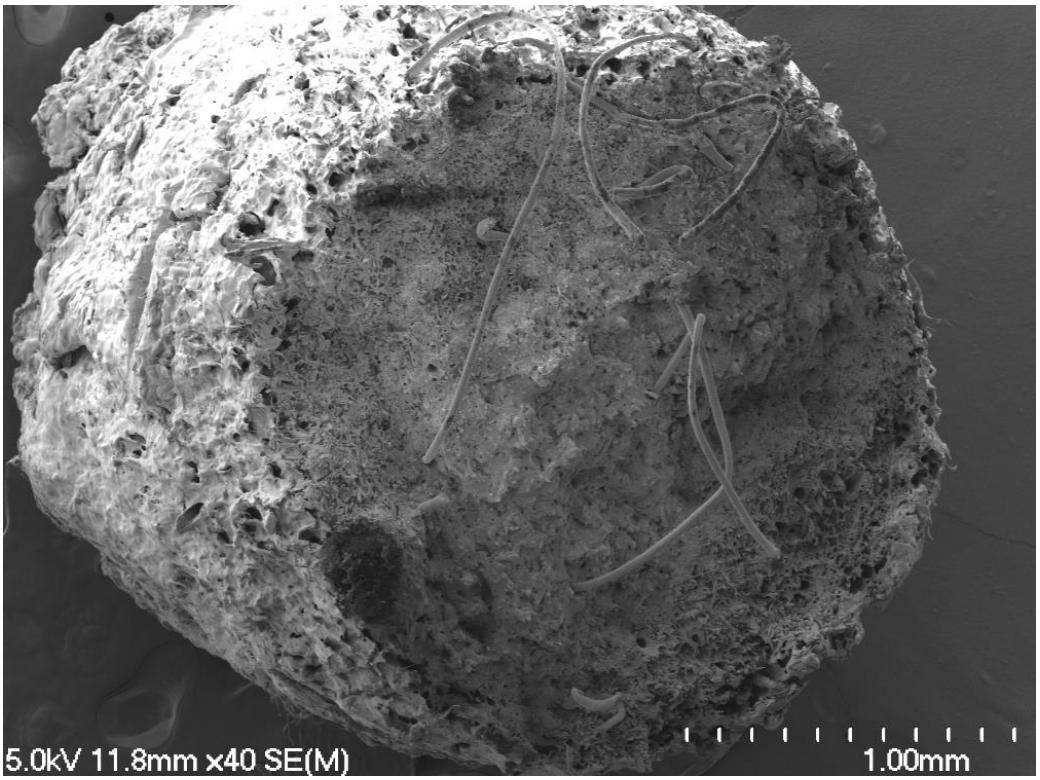


Figure 22, 30 wt% harakeke filament fracture surface showing polyester fibres

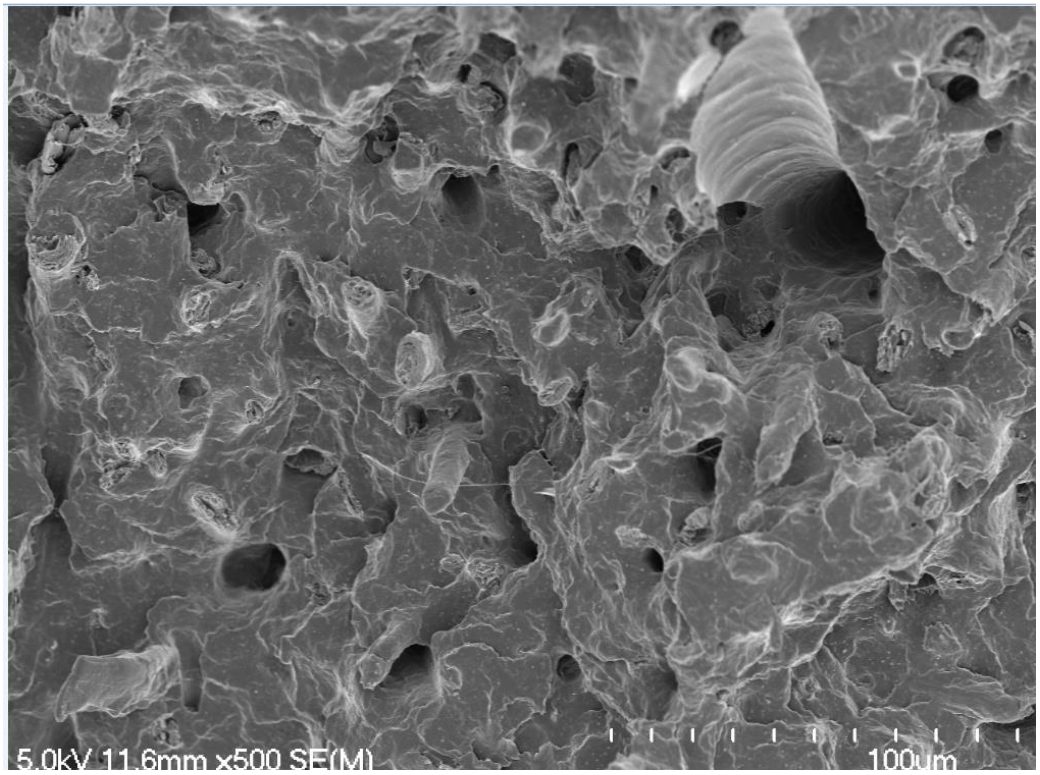


Figure 23, 30 wt% harakeke filament fracture surface showing fibre alignment and fibre pull out

Conclusions

In this research, a series of 3mm composite 3d printing filaments were successfully extruded with varying weight fractions of hemp, harakeke and recycled gypsum. The maximum fibre content that would still allow for accurate filament fabrication was 30 wt%. Exceeding this fraction resulted in a sharkshin surface effect, increased strain on extruder close to its limitations and difficulty using the current spooling machine. For gypsum composites, no such maximum content was found and composites containing up to 50 wt% gypsum were fabricated with little difficulty.

3mm Composite Filament Results: The strongest composite filament produced in this work consisted of 30 wt% harakeke fibre in a post-consumer polypropylene matrix. This composite filament had a tensile strength of 41 MPa and a Young's modulus of 3824 MPa. These values correspond to improvements of 77 % and 275% relative to unfilled polypropylene filament. Using post-consumer PP as a matrix material resulted in better mechanical performance than using pre-consumer PP. This was attributed to either a higher grade of polymer and/or additional reinforcement from polyester fibre content. Although post-consumer polypropylene produced stronger composites than pre-consumer, all composites showed significant improvements in strength and stiffness

SEM micrographs of post-consumer PP composite fracture surfaces supported very poor interfacial adhesion of polyester fibre. This was made apparent by long polyester fibres protruding from fracture surfaces. The protruding fibres were also indicative of fibre alignment along the length of the filament.
Composites containing 10% to 50 wt% recycled gypsum showed an approximately linear decrease in tensile strength while simultaneously showing a linear increase in Young's modulus. This was explained with the aid of SEM micrographs showing conglomeration of gypsum crystals as the particle content increased, creating localised stress concentration points while still restricting polymer chain mobility.

3d Printed Composite Results: A novel method of measuring shrinkage within 3d printed components was developed and used to measure the relative shrinkage of different composites. The composite that displayed the greatest reduction in shrinkage consisted of 30 wt% harakeke with shrinkage reduction of 84% relative to plain polypropylene. Natural fibre proved to be more effective in terms of shrinkage reduction than gypsum with 50 wt% gypsum content achieving approximately the same shrinkage reduction as only 10 wt% natural fibre.

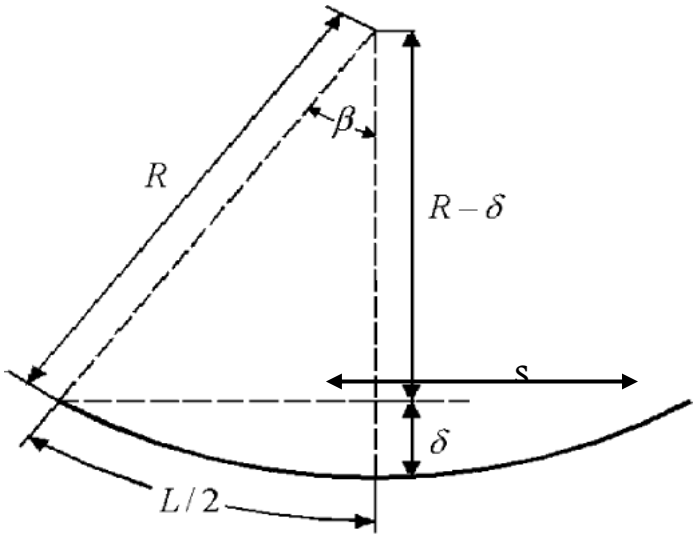
Based on the results shown in this research it can be concluded that recycled polypropylene composites have potential to be used as strong, stiff, cheap and recyclable 3d printing filament. The rapid growth in the 3d printing industry coupled with increased environmental awareness, suggest that these and similar composite filaments are highly relevant in this day and age.

References

1. S.H. Huang, P. Liu, A. Mokasdar and L. Hou, Int. J. Adv. Manuf. Technol., 67, 1191-1203 (2013).
2. H. Brooks, A. Rennie, T. Abram, J. McGovern and F. Caron, Variable fused deposition modelling: Analysis of benefits, concept design and tool path generation, In Innovative Developments in Virtual and Physical Prototyping; P. Bartolo, Ed.; CRC: London, U.K., pp 511-517 (2012).
3. W.H. Zhong, F. Li, Z.G. Zhang, L.L. Song and Z.M. Li, Materials Science and Engineering a-Structural Materials Properties Microstructure and Processing, 301, 125-130 (2001).
4. H.S. Tan, Y.Z. Yu, L.X. Xing, L.Y. Zhao and H.Q. Sun, Polymer-Plastics Technology and Engineering, 52, 257-260 (2013).
5. M.F. D.Dai, Wood fibres as reinforcments in natural fibre composites, In Natural Fibre Composites: Materials, Processes and Properties R.S. A.Hodzic, Ed.; Woodhead Publishing Limited: Cambridge, UK, pp 3-43 (2014).
6. K. Jayaraman and R. Halliwell, Composites Part B-Engineering, 40, 645-649 (2009).
7. D.U. Shah, J. Mater. Sci., 48, 6083-6107 (2013).
8. M. Asgari and M. Masoomi, J. Thermoplast. Compos. Mater., 28, 357-371 (2015).
9. M. Hahm, C.H. Kim and J. Ryu, Polymer-Korea, 32, 7-12 (2008).
10. A. Ujhelyiova, E. Bolhova, A. Marcincin and R. Tino, Fibres Text. East. Eur, 15, 26-29 (2007).
11. M.G.A. Efendy and K.L. Pickering, Composites Part a-Applied Science and Manufacturing, 67, 259-267 (2014).
12. D. Lang, 3D-Printing of Recycled Thermoplastic Composites. MSc Thesis, École Polytechnique Fédérale de Lausanne, The University of Waikato, (2016).
13. R.H. Moynihan, D.G. Baird and R. Ramanathan, J. Non-Newtonian Fluid Mech., 36, 255-263 (1990).
14. S.P. Deshmukh, A.C. Rao, V.R. Gaval and P.A. Mahanwar, J. Thermoplast. Compos. Mater., 24, 583-599 (2011).

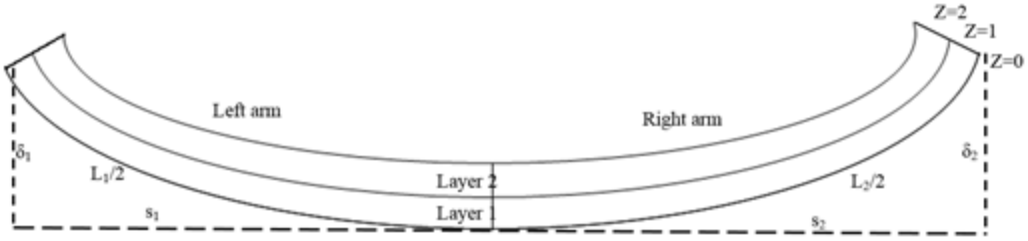
Appendix

Shrinkage test calculations base formulas:



$$R(mm) = \frac{4(\delta^2 + h^2)}{8\delta} \qquad (1)$$

$$\beta(rad) = \frac{L/2}{R};\ \beta(°) = \frac{180 * L/2}{\pi * R} \qquad (2)$$



Assumptions:

1. The arm length *L* at height z=0 is L/2, which is the arm length without shrinkage
2. The layers have the same length at z=1
3. At z=2, the second layer has fully shrunk

On each arm, three measures are taken: the horizontal s, the vertical δ and the arm length L/2. With these, it's possible to determine the central angle β using (1) and (2).
Knowing R, β and the layer height h, it's possible to calculate the shrunk arm length at z=2:

$$L(z=2)(mm) = \frac{(R-2h) * \pi * \beta}{180} \qquad (3)$$

The shrinkage is then the difference between those two lengths

$$Shrinkage\ (\%) = 100 - \frac{100 * L(z=2)}{L(z=0)} \qquad (4)$$