# DEVELOPMENT OF DISCONTINUOUS FIBRE REINFORCED THERMOPLASTIC FEEDSTOCKS FOR HIGH PERFORMANCE 3D PRINTING

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**ABSTRACT**

Fused filament fabrication (FFF) is a 3D printing technique which allows layer-by-layer build-up of a part by the deposition of thermoplastic material through a nozzle. The technique allows for complex shapes to be made with a degree of design freedom unachievable with traditional manufacturing methods. However, the mechanical properties of the thermoplastic materials typically used are low compared to traditional high performance engineering materials. In this work, the development of improved 3D printing feedstocks for FFF is shown, with discontinuous (3mm) carbon fibres embedded in a thermoplastic matrix to reinforce the material. One unique aspect of the proposed approach is that that fibre breakage which limits existing fibre reinforced filament mixing and extrusion techniques is prevented during filament manufacture, such that the fibres are above the critical fibre length for maximum strength, without compromising processing flexibility. One of the critical aspects of this work is selection of the thermoplastic matrix material, as it significantly impacts both processing and performance. This paper therefore focuses on a down-selection process for candidate polymers. First, a pre-selection was made to identify suitable matrices for filament fabrication and printing based on an array of different material, mechanical, and thermodynamic properties. Four matrix materials (ABS, PLA, Nylon, PETG) were then used to manufacture aligned discontinuous fibre thermoplastic (ADFRTP) preforms with a $V_f$ of 12% using an in-house developed continuous consolidation module. Tensile stiffnesses and strengths of ADFRTPs up to 23 GPa and 300 MPa respectively were recorded. This investigation shows that discontinuous fibre filament has the potential to compete with continuous fibre filaments. Future work consists of forming the ADFRTP tapes into a uniform filament and investigations into the interfacial and flow properties of the ADFRTPs.

**Keywords**: composite, thermoplastic, 3D printing, additive manufacture, discontinuous fibres, alignment

## 1 INTRODUCTION

Carbon fibres reinforced plastics (CFRPs) provide excellent mechanical properties and allow for significant design tailorability. A fundamental challenge with manufacture of CFRPs, however, is the consolidation of the reinforcement fibres with the polymer matrix with good control of fibre orientation, low void content, and low cost [1]. While a wide range of manufacturing methods for composites are available, most CFRP parts are currently formed in a two-step process, i.e. material lay-up followed by consolidation. For good consolidation, pressure needs to be applied over the entire part surface area which requires expensive equipment and increases manufacturing costs. In this work, additive manufacture (AM) techniques and more specifically fused filament fabrication (FFF), are investigated as a CFRP manufacturing approach where low investment costs and rapid prototyping ability are important.

Layer-by-layer (LbL) manufacturing techniques, such as FFF, commonly known as 3D printing, have an under-appreciated similarity to traditional composite materials in that they are made from stacking a series of discrete layers. It is therefore reasonable to suggest that successful adaptation of 3D printing technologies to composite materials could enable a simple method for composite manufacture with lower production cost and a high degree of automation. Previous work has identified the current state-of-art in fibre reinforced 3D printing [2], which showed two types of feedstock material; filament reinforced with microfibres (~0.1mm length) or filament with continuous fibres [3]–[5]. The continuous fibre filaments showed the highest mechanical performance, with a tensile strength and stiffness of roughly 900 MPa and 60 GPa respectively. The processing of such filament, however, is more cumbersome as they cannot be freely deposited in tight radii and filament cutting steps are required to finish a print path. Printing with microfibres shows better processing characteristics, but the mechanical properties are typically an order of magnitude lower than continuous fibre 3D prints.

To improve 3D printing of fibre reinforced thermoplastics, a more optimal point in the trade-off between processing and performance needs to be found - as shown schematically in Figure 1. Continuous fibre composites show the highest performance but are harder to process, while composites with microfibres (~0.1mm length) have a low performance as the fibre-matrix interface fails before the fibres do. Increasing the fibre length therefore may be a way forward to a 3D printing filament that can compete with the mechanical performance of continuous fibres, whilst keeping the processing advantages of rapid prototyping. Investigations have shown that aligned discontinuous (3mm) carbon fibre / epoxy composites ($V_f$ =55%) can obtain a strength and stiffness of 1500 MPa and 115 GPa with the HiPerDiF method [6],[7]. The work presented in this paper is part of the HiPerDiF project. It aims to develop a 3D printing filament from aligned discontinuous (3mm) fibre preforms, maintaining fibre length during the filament manufacture process as common techniques (screw extrusion) lead to fibre breakage. This allows the fibres in the 3D printed part to reach their full strength, thereby greatly increasing the mechanical properties of 3D printed parts.

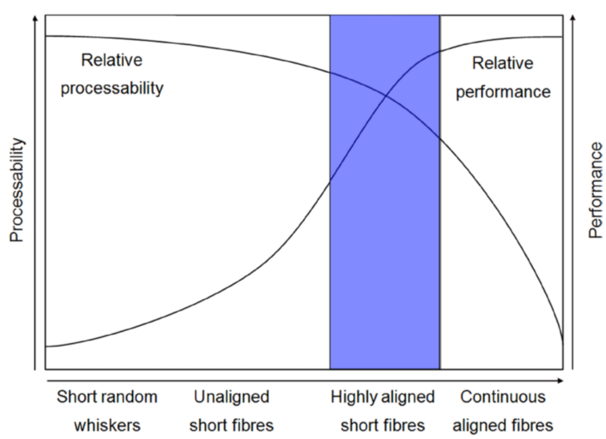



**Figure 1:** Balance between process ability and performance for different fibre architectures [8].

## 2 METHODOLOGY

An overview of the development of aligned discontinuous fibre reinforced thermoplastic (ADFRTP) filaments for high performance 3D printing is given in section 2.1. This paper details the material selection for the ADFRTPs (section 2.2) and the manufacture and testing of the ADFRTP tapes (section 2.3).

### 2.1 Overview

The development of the filament follows four main steps, as shown in Figure 2. The first step is the alignment of 3mm fibres using the HiPerDiF method [6]. The output of the HiPerDiF method is a tape of aligned dry discontinuous fibres with an aerial weight of ~60 g/m$^2$. For this research work, 15cm long tapes are used but the process can be upscaled to create continuous tapes of aligned discontinuous fibres. The second step consolidates the dry fibre tapes with a thermoplastic matrix, similar to pre-pregging. This creates a discontinuous fibre reinforced thermoplastic with a width of 5mm and a thickness of 0.2mm. A third step is required to form the ADFRTP tape into a circular filament for 3D printing without breaking the fibres. This is done by pushing the preform through a custom heated nozzle which changes the cross section from rectangle to circular. This is done with a tight dimensional fit such that the fibres don't coalesce and halt the flow. The last step is deposition of the circular filament using a standard 3D printer.

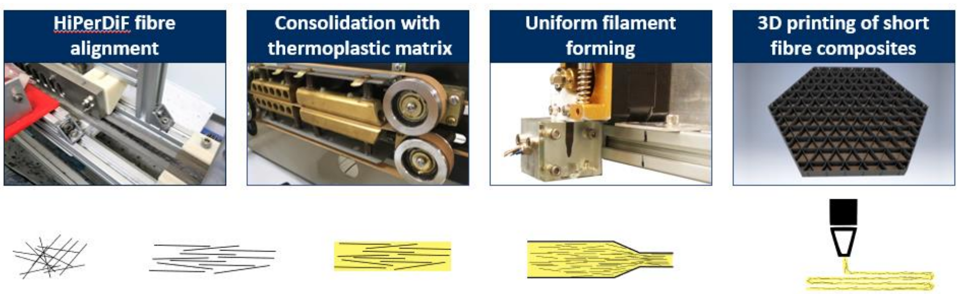



**Figure 2:** Four main processing steps from dry fibres to 3D printing filament.

### 2.2 Matrix selection

Initial trials on ADFRTP manufacture were performed using a polypropylene matrix, as this was shown to have a high potential for recycling [9]. However, it was found that this material has some drawbacks with respect to 3D printing, such as a high degree of shrinkage upon cooling. A material selection was performed over a wide range of thermoplastics (>10), from commodity thermoplastics such as LDPE and HDPE to engineering thermoplastics such as PEEK and PEI.

The main properties considered are the thermal properties, processing properties and the printing characteristics. Ideally, the melt and processing temperature are below 300 °C for easy processing. The coefficient of thermal expansion (CTE), conductivity and heat capacity were considered as these are expected to be important for later printing characteristics. Ideally for 3D printing, the matrix has a low CTE for minimal shrinkage upon cooling, and a high thermal conductivity and a high heat capacity such that more heat can be transported from the nozzle to the printed part to aid the polymer sintering process between printed paths [10]. Both semi-crystalline and amorphous thermoplastics were considered. The viscosity of amorphous materials changes more gradual between room temperature and processing temperature compared to semi-crystalline material which exhibit a distinct melt point. Reference values were obtained from a literature search and from the CES Edupak materials database [11].

From the 15 polymers considered, the four most promising polymers for this application were found to be Nylon, PLA, ABS and PETG. They provide proven printing capability for a processing temperature below 300 °C. PLA, ABS and PETG were acquired from 3D4Makers as a 3D printing filament [12]. The Nylon filament was obtained from MarkForged [13]. The crystallinity of the materials was determined via Digital Scanning Calorimetry (DSC) and the interfacial properties were found in literature.

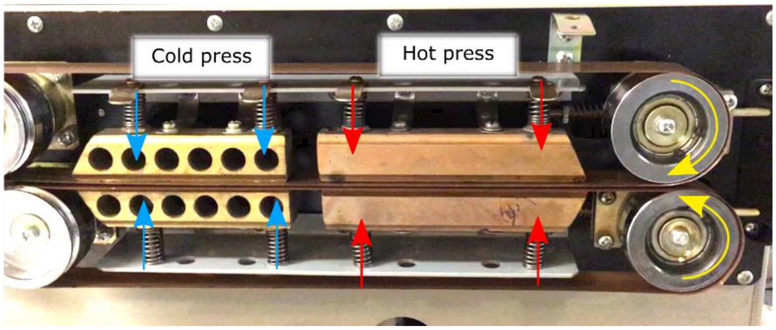
**Table 1:** Selection of matrices, showing criteria and main material candidates and properties obtained from CES Edupak [11].

| | | Nylon | PLA | ABS | PETG |
|---|---|---|---|---|---|
| *GENERAL* | **Costs [GBP / kg]** | 2.4 | 2.6 | 2.0 | 2.0 |
| | **Density [kg/m$^3$]** | 1070 | 1160 | 1050 | 1070 |
| | **Stiffness [GPa] / Strength [MPa]** | 1.1 / 46 | 2.5 / 48 | 2.5 / 36 | 2.1 / 50 |
| *THERMAL* | **Melt temperature [°C]** | 250 | 160 | Amorphous | Amorphous |
| | **Glass transition temperature [°C]** | 60 | 60 | 100 | 85 |
| | **Processing temperature [°C]** | ~260 | 190 | 230 | 270 |
| | **CTE [µstrain/°C]** | 143 | 135 | 170 | 121 |
| | **Thermal conductivity [W/m °C]** | 0.28 | 0.15 | 0.25 | 0.26 |
| | **Specific heat capacity [J/kg °C ]** | 1680 | 1200 | 1400 | 1500 |
| *PROCESSING* | **Crystallinity [-]** | ~50% | ~20% | Amorphous | Amorphous |
| *PRINTING* | **Printing capability** | + | ++ | +- Some warpage | ++ |
| *COMPOSITE* | **Interfacial shear strength [MPa]** | 19.3 [14] | 11-19 [15] | Less than Nylon as addition of Nylon increased tensile properties [16] | Less than Nylon, carbon PETG filament has a lower strength than carbon Nylon filament [17] |

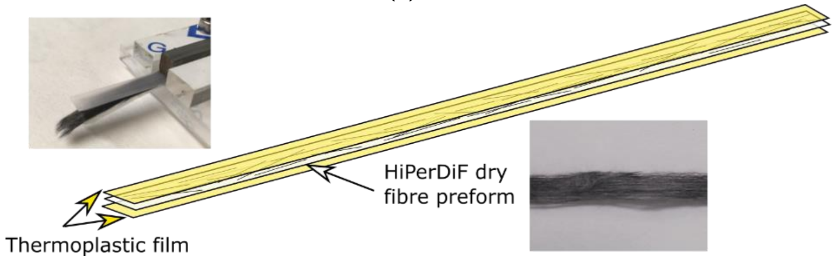
### 2.3 Consolidation and tensile specimens

The second step in the filament development is consolidation of the dry fibre preform with the thermoplastic matrix. This is done with an in-house developed consolidation module as shown in Figure 3a, which applies pressure and heat simultaneously during a hot pressing stage, which is followed by a cold press. The process is suitable to manufacturing continuous ADFRTP tapes.

Thermoplastic films were prepared by 3D printing a single layer with a thickness of 0.125mm using the filament printing recommendation. Dry fibre preforms were prepared using the HiPerDiF method with an aerial weight of about 60g/m$^2$. The fibre preform was enveloped between two films of the polymer matrix as shown in Figure 4b, which were fed through the consolidation module. A Tekscan pressure mapping systems was used to estimate the applied pressure, which was found to be 2 bar. This pressure is applied in total for 25 seconds. All preforms were weighed before and directly after processing, from which the fibre volume fraction $V_f$ was estimated. After processing, the ADFRTPs were trimmed to 10cm length and 4mm width to create uniform tensile test specimens.



**(a)**




**(b)**

**(c)**

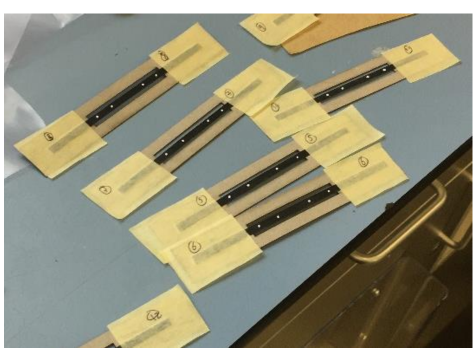
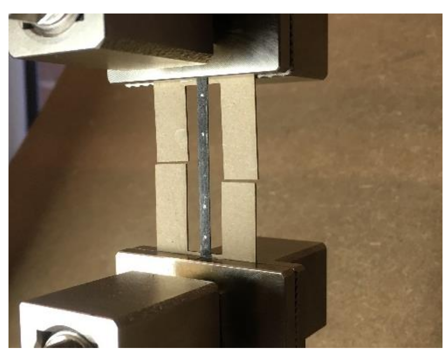
**Figure 4:** (a) Consolidation module showing (b) the fibre and matrix preforms before processing and (c) after processing.

Four different ADFRTPs were made; using PLA, Nylon, ABS and PETG as matrix. For each matrix system, a low and high processing temperature was selected as shown in Table 2 to investigate the effect of processing temperature on the quality and properties of the ADFRTP preforms. These temperatures were chosen as lower and upper bound of the recommend processing temperatures according to CES Edupak [11]. The Nylon processing temperature was based on the printing temperature (260°C) and a lower temperature (200°C) to prevent degradation of the matrix.

**Table 2**: Processing conditions for each TP matrix system.

| **Material** | **$T_{low}$** | **$T_{high}$** |
|---|---|---|
| PLA | 170 °C | 210 °C |
| Nylon | 200 °C | 260 °C |
| ABS | 177 °C | 260 °C |
| PETG | 249 °C | 288 °C |

Tensile testing was performed on samples with dimensions of 100 mm x 4 mm x 0.2 mm. For each configuration, three samples were tested. The ADFRTPs were taped to a paper tab as shown in Figure 5 and clamped between two mechanical grips of an electromechanical Shimadzu test machine with a 1 kN load cell. The specimen was loaded under a displacement rate of 1mm/min and a video extensometer by Imetrum was used to measure the strain during testing.

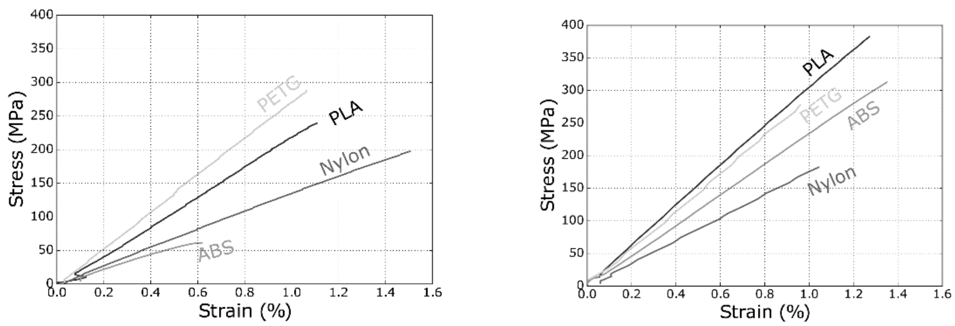
**(a)** **(b)**

**Figure 5:** (a) Preparation of tensile samples and (b) samples loaded between grips.

# 3 RESULTS

The tensile response of the four different ADFRTPs can be seen below for low processing temperatures (Figure 6a) and high processing temperatures (Figure 6b). In general, all specimens showed a linear stress-strain curve. This shows the reinforcing effect of the discontinuous fibres as plastic deformation was prevented and the maximum elongation was limited to about 1%. The best performing specimens had a strength of around 300 MPa and a stiffness of 20 GPa, which included PETG, PLA and ABS specimens

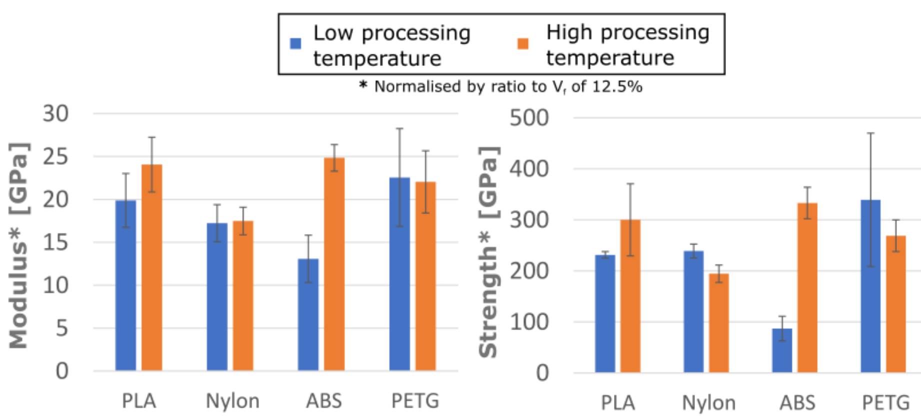
The ABS specimens showed a large increase in performance for higher processing temperatures, which was caused by poor consolidation at the low processing temperatures. For Nylon and PETG, the performance decreased at higher processing temperatures which may be caused by degradation of the matrix.




**(a)** **(b)**

**Figure 6:** Stress-strain curves of ADFRTPs at (a) low processing temperature and (b) high processing temperature.

A comparison of the strength of stiffness of each ADFRTP is shown in Figure 7 which also shows the standard deviation. The properties are normalized to the average fibre volume fraction $V_f$ of 12.5% by ratio. The increase in performance of the ABS specimens at higher processing temperatures can clearly be seen here, as well as the lower overall performance of the Nylon specimens. The lower performance of the Nylon ADFRTPs is attributed to the higher ductility and lower modulus of Nylon.




**(a)** **(b)**

**Figure 7**: Tensile test results of ADFRTP processed at different temperature, normalized by ratio to a $V_f$ of 12.5%.

# 4 CONCLUSIONS AND FUTURE WORK

A 3D printing reinforced thermoplastic filament is being developed using aligned dry 3mm carbon fibre preforms from the HiPerDiF method. The first step in this process is the preparation of aligned discontinuous fibre reinforced thermoplastic (ADFRTP) tapes. The second step is consolidation of the fibres with a thermoplastic matrix. After initial consideration of a wide range of different polymers for the matrix, the four most promising polymers (PLA, Nylon, ABS and PETG) were selected and the mechanical performance of the ADFRTP tapes made from each were assessed with respect to two different processing temperatures. The best performing ADFRTPs had a stiffness of around 23 GPa and a strength of 300 MPa with a $V_f$ of 12.5%. Increasing the fibre volume content above 20% would allow the ADFRTPs to compete with continuous fibre printing solutions.

For PLA and ABS, higher processing temperatures led to an increase in performance while for Nylon and PETG, a decrease in properties was measured. Better understanding of the flow properties of the matrices and the interface between the fibre and matrix may improve the quality and the mechanical properties of the specimens. Future work focuses on measuring the flow properties of the ADFRTPs and how the fibres flow in a thermoplastic melt to optimize as last step in the filament formation process.

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