METHODOLOGY FOR CONTINUOUS FIBRE ADDITIVE MANUFACTURING WITH IN-LINE IMPREGNATED THERMOPLASTIC COMPOSITES

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ABSTRACT

This paper presents a methodology for Continuous Fibre Additive Manufacturing (CFAM) of a glass fibre filled polyamide 12 composite. The whole process, starting from impregnation of dry fibre bundles with polyamide, to deposition of the composite on the build platform is described and relevant parameters such as processing temperatures, pultrusion die diameter, deposition velocity, minimum fibre bending radius, ideal layer thickness and strand width, actual fibre volume fraction and tensile properties of the final composite were determined, calculated or estimated. A composite with a fibre volume fraction of about 40 vol% was successfully produced.

Key Words: ADDITIVE MANUFACTURING, CONTINUOUS FIBRE, THERMOPLASTIC**,** COMPOSITES

1. INTRODUCTION

Extrusion based Additive Manufacturing (AM) techniques for thermoplastic materials have been developed during the last thirty years. In order to avoid limitations in mechanical properties such as strength, stiffness of additively manufactured products compared to their injection moulded counterparts, short fibre filled AM materials have been introduced more recently, as seen in the work of Tekinalp [1], Ning [2], Compton [3], Ferreira [4], Raney [5], Kishore [6], Brenken [7] and Zhang [8]. Depending on the volume fraction of the added fibres, these composites are often more brittle than the unfilled polymer material [9],[10].

To further expand the possibilities of the extrusion based AM materials, different processes which incorporate continuous fibres into polymers were developed. A first category of methods intends to remelt and deposit a fibre bundle, pre-impregnated with a thermoplastic polymer (often referred to as 'prepreg'), as researched by Nunes [11], Gardner [12], Melenka [13], Dickson [14], Eichenhofer [15] and Goh [16]. A second category of methods incorporates both impregnation and deposition in a single process, being researched by Bettini [17], Matsuzaki [18], Nanya [19], Tian [20], Van De Steene [21], Liu [22] and Van De Steene [23]. These processes for Continuous Fibre AM (CFAM) enable the manufacturing of complexly shaped 3D objects that cannot be produced using traditional subtractive production technologies and gives the possibility to fully tailor and control fibre orientation [24], which is not always possible using the classic composite lay-up processes. These two advantages could lead to lighter, stronger and stiffer parts for use in high-end applications.

In this paper, the second methodology for in-line impregnation of a dry fibre bundle with a thermoplastic polymer and the consecutive deposition of this composite material is described. Typical values of the fibre bundle's linear density, layer thickness, road width, achievable fibre bending radii, typical deposition velocity and fibre volume fraction will be discussed and an estimation of the composite's tensile properties will be given. A fibre volume fraction of around 40% will be pursued.

2. MATERIALS AND METHODS

2.1 Materials

Rilsamid AMN O TLD, a polyamide 12 from Arkema, was used as the matrix material for the process. Its tensile properties were determined according to ISO 527-1:2012 and ISO 527-2:2012. The tensile modulus was found to be 1291 MPa (standard deviation: 144 MPa) and tensile strength 43.8 MPa (standard deviation: 2.3 MPa). The density of the material is determined using the immersion method described in ISO 1183-1:2004 and has a value of 1009.2 kg/m³ (standard deviation: 4.5 kg).

StarRov LFTplus Direct Roving 853, a continuous glass fibre roving from Johns Manville was used as the reinforcing material in the composite. The tensile modulus is 57112 MPa (standard deviation: 1247 MPa), tensile strength is 1605 MPa (standard deviation: 135 MPa). Its linear density was determined according to ISO 1889:2009 as 1179 tex (standard deviation: 3 tex), which is equivalent to 2225 single glass fibres in a roving. The density of the fibre material is 2540 kg/m³. The mean single fibre diameter was determined on a micrograph of a polished composite cross section as 16.3 µm (standard deviation: 1.8 µm).

2.2 Continuous Fibre Additive Manufacturing (CFAM) process

Van De Steene [21] proposed and evaluated three methods (Pultrusion, PassivePin and ActivePin) for melt-impregnation of continuous glass fibre bundles with the thermoplastic polyamide 12, as described in the Materials section 2.1. These impregnation methods can be performed in-line with an additive deposition technique. Therefore, the impregnation device (Figure 1) containing a polymer melt chamber (a) and impregnation pins (b) should be equipped with a puller mechanism (c) and a heated deposition nozzle (d) in order to perform an automated lay-up of thermoplastic composite material on the heated build platform (e) of an AM platform, often referred to as '3D-printer'. The relative velocity of the impregnation device and the build platform is v_3. The advantage of the use of impregnation pins is twofold. On the one hand, pulling a fibre bundle over a cylindrical surface spreads out this bundle, decreasing the impregnation distance. On the other hand, the pressure generated between the bundle and the pins's surfaces is the driving force for pushing the thin film of melted polymer between the spreader pin and the bundle into the fibres of the bundle.

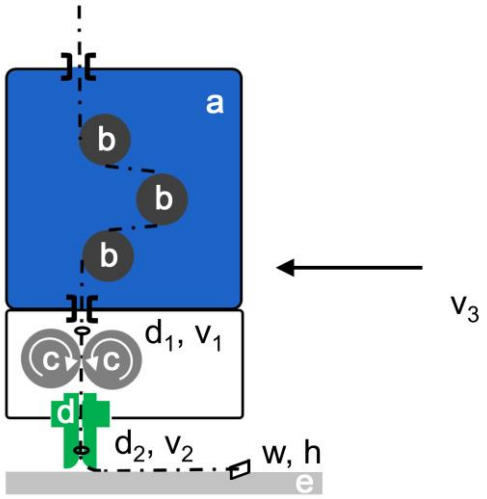

Figure 1. Impregnation device with deposition system.

Before leaving the melt chamber, the impregnated fibre bundle is passed trough a pultrusion die or wipe-off die with diameter d_d, to prevent excess polymer from escaping the melt chamber. The required fibre volume fraction $V_{f,\,req}$ of the composite will be determined by the bore diameter of this pultrusion die d_d (formula 1), in which ρ_f represents the fibre's density and L its linear density.

$$d_d = \sqrt{\frac{4\,L}{\pi\,\rho_f V_{f,req}}} \tag{1}$$

It is important for the deposition nozzle to have a rounded transition to gently guide and smear out the wetted fibre bundle without the occurrence of fibre failure. For an isotropic fibre material with single fibre diameter d, Young's modulus E and bending strength σ_b, the minimum bending radius R to avoid breakage can be estimated according to formula 2:

$$R \geq \frac{2\,E\,d}{3\,\sigma_b} \tag{2}$$

This prerequisite should be taken into account for all applied bends on the fibre bundle throughout the whole impregnation and deposition process, in order to prevent fibre failure and clogging of the pultrusion and/or deposition system.

After being smeared out, the cross section of the impregnated bundle can be seen as rectangular. The height of a deposited strand, and thus the layer height is called h, the width of such a strand is called w. Supposing an equilibrium in the mass balance before and after composite deposition yields formula 3. Note the hyperbolic relationship between layer thickness and strand width.

$$\frac{\pi\,d_d^2}{4} = w\,h \tag{3}$$

Relevant parameters such as processing temperatures, pultrusion die diameter, deposition velocity, minimum bending radius, ideal layer thickness and strand width, fibre volume fraction and tensile properties of the final composite will be determined, calculated or estimated in the 'Results and discussion' section.

2.2 Fibre volume fraction determination

The composites' theoretical fibre volume fraction $V_{f,\,req}$ is a function of the diameter of the pultrusion die d_d, the fibre's density ρ_f and the linear density of the fibre bundle L (formula 1). In practice, the pultrusion velocity might have an influence on the effective fibre volume fraction V_f due to the shear-thinning effects of the polymer melt and thus the fibre-polymer ratio. Due to flow dependent leakage of the polymer between the pultrusion die bore and the fibre bundle, a theoretical calculation of $V_{f,\,req}$ is only an estimate [21]. A more accurate and practical method to determine V_f is incineration of a composite sample (dimensions: 10 x 16 x 0.5 mm) to determine its fibre mass fraction M_m and the fibre mass fraction M_f. Knowing the matrix's density ρ_m and the fibre's density ρ_f, V_f can be calculated using formula 4.

$$V_f = \frac{M_f\,\rho_m}{\rho_f + M_f\,(\rho_m - \rho_f)} \tag{4}$$

2.3 Tensile properties estimation

When the fibre volume fraction V_f , the fibre's tensile modulus E_f, the fibre's tensile strength σ_f, the matrix volume fraction V_m, the matrix's tensile modulus E_m and the matrix's tensile strength σ_m are known, some tensile properties of a unidirectional composite can be estimated using the rules of mixtures (formula 5 to formula 7). In these formulae, the index "x" corresponds to the principal direction parallel to the fibre material, indices "y" and "z" correspond to the principal directions perpendicular to the unidirectional fibres.

$$E_x \approx E_f V_f + E_m V_m = E_f V_f + E_m(1 - V_f) \quad (5)$$

$$\sigma_x \approx \sigma_f V_f + \sigma_m V_m = \sigma_f V_f + \sigma_m (1 - V_f) \quad (6)$$

$$E_y = E_z \approx \frac{E_f E_m}{E_f + V_f (E_m - E_f)} \quad (7)$$

3. RESULTS AND DISCUSSION

3.1 Process parameters determination

During preliminary testing of the CFAM technique, deposition velocities between 0.5 mm/s and 4 mm/s were used. Lowest velocities resulted in a well impregnated and smeared out composite with no fibre breakage. Highest velocities resulted in a moderately impregnated composite and some fractured fibres. A deposition velocity of 2 mm/s is a good trade-off between deposition velocity and composite quality.

Processing temperatures used in the impregnation device have an important effect on the polymer melt viscosity. Choosing high temperatures resulted in a colour change of the matrix material, indicating chemical degradation. For a deposition velocity of 2 mm/s, a chamber temperature of 240°C, a nozzle temperature of 230°C and a bed temperature of 110°C gave optimal results.

In this research, a V_f of around 40 vol% was pursued. Using formula 1, the required pultrusion die diameter was calculated to be 1.2 mm. It was found that the lay-up of a strand at a layer height of 0.5 mm resulted in the best adhesion to the bed or to successive deposited layers. Using formula 3, a required strand width of 2.25 mm was calculated in order to create a theoretically voidless composite.

Filling in the values of the used glass fibre bundle in formula 2, the minimum required bending radius to avoid fibre failure for 95% of the fibres was calculated as 0.7 mm. In this setup, impregnation pins with radii of 4 mm and a nozzle with a radius of 3 mm were chosen to avoid breakage before deposition. A zig-zag motion (figure 2) was chosen as deposition pattern. Sharp changes in the tool path direction were avoided, a path radius of w divided by two, being 1.125 mm, was implemented. From the deposited parts, it can be seen that fibres tend to slightly unbend and curve upwards when being deposited on paths with high curvature (figure 3). This phenomenon was described as 'tow pullup' by [25].

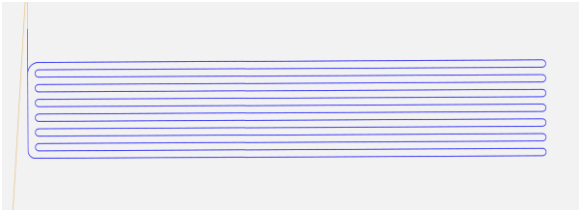

Figure 2. Impregnation device with deposition system.

Figure 3. Tow pullup in regions of high path curvature.

3.2 Fibre volume fraction determination results

Calculation of the composite sample's V_f using formula 4, resulted in a value of 38.84 vol% (standard deviation: 0.46 vol%), which is in line with the pursued value.

3.3 Tensile properties estimation results

Tensile properties tensile modulus and tensile strength were estimated using formulae 5 to 7 and listed in table 1. It is important to validate these predictions through future tensile testing.

Table 1. Estimation of the composite 's tensile properties

	E [GPa]	$\sigma_{tensile}$ [MPa]
glass fibre	57.11	1605
PA12	1.29	44
composite x-direction	22.97	650
composite y- and z-direction	2.08	-

4. CONCLUSIONS AND OUTLOOK

In this paper, a methodology for Continuous Fibre Additive Manufacturing (CFAM) of a glass fibre filled polyamide 12 composite was described, starting from impregnation of dry fibre bundles with polyamide, to deposition of the composite on the build platform. Parameters relevant for this process were determined or calculated and are summarised in table 2. An estimation of the composite's tensile properties was given in table 1 and will be determined through mechanical testing in future research.

Table 2. Summary of relevant CFAM parameters.

Deposition velocity	2 mm/s	Pultrusion die diameter	1.2 mm
Melt chamber temperature	240 °C	Layer height	0.5 mm

Deposition nozzle temperature	230 °C	Layer width	2.25 mm
Build platform temperature	110 °C	Minimum bending radius	0.7 mm
Fibre volume fraction	38.44 vol% (standard deviation: 0.46 vol%)		

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