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Effects of processing conditions on orientation for short-fiber composites in fused filament fabrication

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Effects of processing conditions on orientation for short-fiber composites in fused filament fabrication

Project phase

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Abstract

Fused filament fabrication (FFF) is an additive manufacturing technique that uses polymer extrusion to build a three-dimensional object layer by layer. To improve the mechanical properties of the final product, discrete fibers are added to the matrix material which increases the anisotropy as fibers tend to align in the extrusion direction. A two-dimensional numerical model is developed to better understand the influence of processing conditions such as nozzle geometry and temperature on the orientation of fibers. The possibility of tuning parameters to obtain a more random fiber orientation is investigated, as this might be beneficial for overall strength of the product. The model is validated with a short experimental study, using polyamide 6 with carbon fiber reinforcement.
Acknowledgements

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Last of all, I would like to thank my parents and girlfriend for the support and motivation during my studies and my friends for advice and good company during coffee breaks. This work would have not been possible without their help.
Contents

1 Introduction 1

2 Problem Description & Mathematical Model 2
   2.1 Governing equations 3
   2.2 Boundary and initial conditions 5

3 Numerical Method 6
   3.1 Time integration 6
   3.2 Remeshing 7

4 Validation 8
   4.1 Mesh convergence 8
   4.2 Extrudate Swell Problem 9

5 Results & Discussion 10
   5.1 Free surface height 10
   5.2 Velocity profiles 11
   5.3 Particles simulation: Isothermal vs. non-isothermal 12
   5.4 Effects of substrate velocity on orientation 14
   5.5 Effects of $L_2$ and $W_{gap}$ 16

6 Conclusions 17

References 18

Appendices 21
1 Introduction

Fused filament fabrication (FFF) is an additive manufacturing technique that is used to produce three-dimensional objects based on digital models. Although the technique was invented in the 1980’s, it has gained a lot of popularity over the last few years as a result of the decrease in price of the equipment, the wide range of available materials and the relatively simple operation [1]. The term additive manufacturing refers to a category of techniques that produce an object by layer-wise deposition of material, without the use of specialized tooling or molds. This way of manufacturing is also referred to as three-dimensional (3D) printing and the range of materials that can be used is increasing rapidly. For FFF, a few examples of polymers that can be used are acrylonitrile butadiene styrene (ABS), polylactic acid (PLA) and polyamides like PA6 and PA12. Besides FFF, other polymer 3D printing processes are Selective Laser Sintering (SLS) and Stereolithography (SLA). Selective Laser Melting (SLM) is a process that can be used for 3D printing with metals like aluminum, stainless steel and titanium. Additive manufacturing has the potential to disrupt the current manufacturing industry by reducing inventory, reduce transport costs and enable the manufacturing of customer-specific products at a reduced cost. It is a fast growing business that has already changed the way in which prototypes and highly specialized products such as medical implants and hearing aids are made [2], but recently the technology is more and more implemented in final-product based manufacturing in a great variety of fields.

During the FFF process, a thin thermoplastic wire (filament) is fed into a moving print head that contains a heating element and a converging nozzle. As a result of the internal pressure, the liquefied material is pushed through this nozzle and subsequently deposited on the substrate/platform in a certain path. A schematic representation of the process is depicted in Figure 1. An extra spool of material can be added to print support structures in case of overhanging geometries.

![Figure 1: The basic elements of the Fused Filament Fabrication process [3].](image)

As additive manufacturing is more and more used in final-product based manufacturing, the demand for good overall mechanical properties of 3D printed products is also increasing. For metals, it has been shown that the mechanical properties of 3D printed objects are comparable or even superior to traditionally manufactured objects [4]. Unfortunately, for 3D printed polymers the mechanical properties are still inferior compared to objects produced with traditional processes like injection molding. The demand for high performance materials used in additive manufacturing and more specific, extrusion-based processes like FFF, have lead to research and development of more complex materials such as polymer composites. Recently, advances have been made using short fiber reinforced material, and continuous fiber-reinforced material using a specially designed FFF printer [5, 6]. Results have shown that the modulus and tensile strength in the extrusion direction increased significantly when adding certain weight percentages of either glass or carbon fiber. Furthermore, adding fibers also decreased the amount of distortions as a result of lower shrinkage [7, 8]. The amount of anisotropy is increased by adding fibers, because they have a tendency to align in the extrusion direction.

Although adding fibers can significantly improve the strength of the printed parts, choosing the right processing parameters in combination with a decent hot-end design is of equal importance in obtaining a strong part. Currently the majority of research has shown that short carbon or glass fibers indeed enhance the mechanical properties, but fundamental research on the mechanisms that cause these effects...
and the link to processing conditions are very limited. The importance of understanding the influence of processing conditions on fiber orientation goes well beyond mechanical properties only. For instance, thermal and electrical properties also depend strongly on the degree of anisotropy. For certain types of composites it could also be beneficial to optimize the local micro-structure to induce shape-shifting through external stimuli and in this way enable so called ‘four-dimensional printing’ [9].

Predictions of mechanical behavior and fiber orientation by means of numerical simulation have been done for composites produced with FFF by Heller et al. [10], but results have not been validated experimentally. In their work, they assumed that the fluid was Newtonian and the process was isothermal. A fiber orientation tensor approach was used that was developed by Advani and Tucker [11], mainly to study fiber orientation in highly concentrated suspensions for processes like injection molding and compression molding. The importance of including thermal behavior was discussed by Mackay [12], who concluded that due to the low thermal conductivity the heat distribution in the nozzle is not uniform, especially for high extrusion rates. The material in the center will thus be significantly colder and as a result have a higher viscosity. It is expected that this has serious consequences for the final fiber orientation in the product, since it limits the movement of fibers throughout the nozzle.

Mackay [12] also discussed that extrudate swell effects, commonly encountered in material extrusion, can have significant effects on FFF. It is expected that it limits the dimensional accuracy, has significant influence on the final strength of a printed part and in case of polymer composites it is expected to influence the fiber orientation. Heller et al. [13] and Wang et al. [14] both investigated this effect for fiber filled materials in FFF, and concluded that the final fiber orientation indeed is influenced by the amount of swell but did not consider the effect of deposition on a substrate.

The main objective of this study is to investigate the effect of processing conditions and nozzle geometry on the orientation of fibers after extrusion. The possibility for reducing anisotropy by obtaining a more random orientation is investigated, since this can be important for achieving better overall mechanical properties. The finite element method is used to model the flow behavior and the movement of multiple particles. The approach is fully coupled, including temperature effects and particular attention will be paid to modeling the free surfaces that can show extrudate swell. However, first a simplified model is used where only the effects of extrudate swell are considered without the deposition on the substrate, with a geometry similar to the one that was used by Heller et al. [13] and Wang [14]. This model will be isothermal and only a single fiber is used to qualify the effects of nozzle geometry on extrudate swell. These results are then compared to the results of a short experimental study to validate the numerical method used in this study.

2 Problem Description & Mathematical Model

In Figure 2a a technical drawing of a common FFF nozzle is depicted, including a cross section (right) and a magnification of the outlet (top). The polymer or polymer composite that needs to be deposited enters the nozzle through an opening with diameter $D_1$, and is initially at ambient temperature $T_\infty$. It is assumed that the wall of the nozzle represented by $\Gamma_w$ has a constant temperature of $T_{wall}$, so that the material is heated and consequently the viscosity $\eta$ will decrease. The material wire, in its solid state, acts a piston to push the molten material trough the nozzle, where it leaves via a capillary with diameter $D_2$ and length $L_2$. Typical values of process parameters and dimensions for FFF are shown in Table 1, however these parameters can vary depending on the material that is used and the dimensional accuracy that is requested. Throughout this study, the commercially available Novamid ID 1030-CF10 produced by DSM is used, which is a PA6-66 copolymer with 10 wt % chopped carbon fiber. The material properties of the unfilled material without carbon fiber, Novamid ID 1030, are used since data from this material are known and they are presented in Table 2. A Cross-Arrhenius model is used for fitting the viscosity/shear-rate curve to experimental data in order to obtain the rheological properties, which is shown in Figure 3. In the FFF process, shear rates are in the range of 100-200 s$^{-1}$ [15][16], so according to this data shear thinning must be accounted for in the viscoelastic model.

A two-dimensional representation of the domain $\Omega(t)$ that is of interest is depicted in Figure 2b. Note that the reference frame is changed so that the positive $y$ direction coincides with the extrusion direction from the nozzle. We assume that the velocity field at the inlet $\Gamma_{in}$ is fully developed. After extrusion from the nozzle, the polymer is deposited on a moving substrate $\Gamma_{sub}$ to lay down the material in a specific path, and the width of the gap between nozzle and substrate is denoted by $W_{gap}$. The polymer melt that exits the nozzle will typically show swelling behavior as a result of elastic recovery, and thus the shape of the deposited material will depend on the processing conditions [13]. The free surfaces $\Gamma_{fs1}$ and $\Gamma_{fs2}$, represented by the dotted red lines, change over time as a result of this effect. Note that the term free
Figure 2: (a) Cross section of an FFF nozzle, with parametric dimensions [17]. (b) Domain $\Omega(t)$, representing the flow through the nozzle and the deposition on the substrate.

The boundary $\Gamma_{\text{sub}}$, in combination with the arrow represent the moving substrate and its direction. It is assumed that the temperature of the substrate is fixed at $T_{\text{sub}}$, and as a result the extruded material will cool down at this boundary. At the free surfaces, the material also cools down as a result of convection and radiation.

As explained in the introduction, a simplified domain is considered for the validation of our method and to isolate the effects of extrudate swell on fiber orientation. This domain is described in Appendix I, and the experimental methods are described in Appendix II.

Table 1: Process parameters of the FFF process.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nozzle wall temperature</td>
<td>$T_{\text{wall}}$</td>
<td>280</td>
<td>°C</td>
</tr>
<tr>
<td>Substrate temperature</td>
<td>$T_{\text{sub}}$</td>
<td>100</td>
<td>°C</td>
</tr>
<tr>
<td>Ambient temperature</td>
<td>$T_{\infty}$</td>
<td>40</td>
<td>°C</td>
</tr>
<tr>
<td>Inlet diameter</td>
<td>$D_1$</td>
<td>3.2</td>
<td>mm</td>
</tr>
<tr>
<td>Outlet diameter</td>
<td>$D_2$</td>
<td>0.4</td>
<td>mm</td>
</tr>
<tr>
<td>Total nozzle length</td>
<td>$L_1$</td>
<td>12.5</td>
<td>mm</td>
</tr>
<tr>
<td>Length of capillary</td>
<td>$L_2$</td>
<td>0.6</td>
<td>mm</td>
</tr>
<tr>
<td>Convergence angle</td>
<td>$A_1$</td>
<td>60</td>
<td>°</td>
</tr>
<tr>
<td>Gap width</td>
<td>$W_{\text{gap}}$</td>
<td>0.25</td>
<td>mm</td>
</tr>
</tbody>
</table>

2.1 Governing equations

To solve the flow problem that is described in the previous section, the balance of mass, momentum and energy needs to be solved. If we assume that the fluid is incompressible and the inertia can be neglected, these equations are:

$$\nabla \cdot \sigma = 0 \quad \text{in } \Omega(t),$$  \hspace{1cm} (1)
\[ \nabla \cdot \mathbf{u} = 0 \quad \text{in} \quad \Omega(t), \]  
\[ \rho c_p \frac{D T}{Dt} = k \nabla^2 T + \sigma : \mathbf{D} \quad \text{in} \quad \Omega(t), \]  
\[ \text{where} \quad \mathbf{u} \text{ is the velocity and} \quad \sigma \text{ is the stress tensor.} \]
\[ D/Dt \text{ denotes the material derivative,} \quad \rho \text{ is the density,} \]
\[ c_p \text{ the specific heat,} \quad k \text{ the thermal conductivity,} \quad T \text{ the temperature and the rate of deformation tensor} \]
\[ \mathbf{D} = (\nabla \mathbf{u} + (\nabla \mathbf{u})^T)/2. \]  
\[ \text{Assuming that the domain} \quad \Omega \text{ is two-dimensional, the free surfaces can be described with a one-}
\[ \text{-dimensional height function} \quad H(y, t) \text{ and} \quad H(x, t). \quad \text{The evolution of the flow domain on the first free}
\[ \text{surface} \quad \Gamma_{fs1} \text{ can be determined with the kinematic equations } [18]: \]
\[ \frac{\partial H}{\partial t} + u_x \frac{\partial H}{\partial x} = -u_y \quad \text{on} \quad \Gamma_{fs1}, \]  
\[ \text{and the evolution of the flow domain on the second free surface} \quad \Gamma_{fs2} \text{ can be determined in a similar way,}
\[ \text{however this surfaces swells in the negative} \quad x\text{-direction so this equation reads:} \]
\[ \frac{\partial H}{\partial t} + u_y \frac{\partial H}{\partial y} = -u_x \quad \text{on} \quad \Gamma_{fs2}. \]  
\[ \text{Since the materials that are commonly used in FFF are viscoelastic, the stress tensor of the fluid can}
\[ \text{be written as:} \]
\[ \sigma = -pI + 2\eta_s \mathbf{D} + \tau, \]  
\[ \text{where} \quad p \text{ is the pressure and} \quad \eta_s \text{ is a solvent viscosity component that is added for numerical reasons.} \]
\[ \text{A Giesekus model will be used as a constitutive model for the stress and it is written in terms of the}
\[ \text{conformation tensor} \quad \mathbf{c} \text{ } [19]. \quad \text{The Giesekus model can be written as:} \]
\[ \tau = \frac{\eta_p}{\lambda}(\mathbf{c} - I), \]  
\[ \lambda \nabla \mathbf{c} + \mathbf{c} - I + \alpha(\mathbf{c} - I)^2 = 0, \]  
\[ \text{where} \quad \lambda \text{ is the relaxation time,} \quad \eta_p \text{ is the polymer viscosity and} \quad \alpha \text{ is a material parameter that determines}
\[ \text{the amount of anisotropic drag. The triangle denotes the upper-convected derivative and is defined as:} \]
\[ \nabla \mathbf{c} = \frac{\partial \mathbf{c}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{c} - (\nabla \mathbf{u})^T \cdot \mathbf{c} - \mathbf{c} \cdot \nabla \mathbf{u}. \]  
\[ \text{For better numerical stability, the conformation tensor is changed to a logarithmic equivalent of the}
\[ \text{conformation tensor} \quad \mathbf{s} = \log(\mathbf{c}) \text{ } [20], \quad \text{a detailed explanation of the theory behind this log-conformation is}
\[ \text{beyond the scope of this thesis:} \]
\[ \frac{\partial \mathbf{s}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{s} - g((\nabla \mathbf{u})^T, \mathbf{s}) = 0. \]  
\[ \text{During the process, the temperature is not constant and as result the viscosity will change locally. A}
\[ \text{temperature dependent relaxation time} \quad \lambda(T) \text{ is used to account for this effect:} \]
\[ \lambda = \lambda_0 \ast a_T, \]  
\[ \text{where} \quad \lambda_0 \text{ is the relaxation time at reference temperature} \quad T_{ref}, \text{ and the shift factor} \quad a_T:\]
\[ a_T = \exp \left[ \frac{E_a}{8.31} \left( \frac{1}{T} - \frac{1}{T_{ref}} \right) \right], \]
Table 2: Material Data for Novamid ID 1030.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zero shear viscosity</td>
<td>$\eta_0$</td>
<td>2600</td>
<td>Pa·s</td>
</tr>
<tr>
<td>Activation energy</td>
<td>$E_a$</td>
<td>43</td>
<td>kJ/mol</td>
</tr>
<tr>
<td>Crystallization temperature</td>
<td>$T_c$</td>
<td>170</td>
<td>°C</td>
</tr>
<tr>
<td>Density</td>
<td>$\rho$</td>
<td>1.130</td>
<td>g/cm$^3$</td>
</tr>
<tr>
<td>Mobility parameter</td>
<td>$\alpha$</td>
<td>0.2</td>
<td>-</td>
</tr>
<tr>
<td>Relaxation time at reference temp.</td>
<td>$\lambda_0$</td>
<td>0.006</td>
<td>s</td>
</tr>
<tr>
<td>Reference temperature</td>
<td>$T_{ref}$</td>
<td>260</td>
<td>°C</td>
</tr>
<tr>
<td>Heat capacity</td>
<td>$c_p$</td>
<td>2</td>
<td>kJ/(kg·K)</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>$k$</td>
<td>0.25</td>
<td>W/(m·K)</td>
</tr>
<tr>
<td>Convection coefficient</td>
<td>$h$</td>
<td>30</td>
<td>W/(m$^2$·K)</td>
</tr>
<tr>
<td>Surface emissivity</td>
<td>$\epsilon$</td>
<td>0.95</td>
<td>-</td>
</tr>
</tbody>
</table>

Figure 3: Viscosity versus shear rate data for Novamid ID 1030, obtained by DSM.

with $E_a$ the activation energy. To account for the solidification of the material for $T < T_c$, the relaxation time goes to infinity. It is assumed that all the other material properties are temperature independent.

2.2 Boundary and initial conditions

The boundary conditions of the model are:

\[
\begin{align*}
  u &= 0 \quad \text{on } \Gamma_w \quad (13) \\
  u_y &= 0 \quad \text{on } \Gamma_{sub} \quad (14) \\
  u_x &= u_{sub} \quad \text{on } \Gamma_{sub} \quad (15) \\
  \int_{\Gamma_{in}} u \cdot n ds &= -Q \quad \text{on } \Gamma_{in} \quad (16) \\
  t &= (pI + \tau) \cdot n = 0 \quad \text{on } \Gamma_{out} \quad (17) \\
  t &= (pI + \tau) \cdot n = 0 \quad \text{on } \Gamma_{fs1} \quad (18) \\
  t &= (pI + \tau) \cdot n = 0 \quad \text{on } \Gamma_{fs2} \quad (19) \\
  H(y = 0, t) &= H_0 \quad \text{on } \Gamma_{fs1} \quad (20) \\
  H(x = 0, t) &= H_0 \quad \text{on } \Gamma_{fs2} \quad (21) \\
\end{align*}
\]

where $n$ is the outwardly directed unit normal vector, $Q$ is the flow rate and $H_0$ the initial free surface height. The inlet velocity at $\Gamma_{in}$ is imposed by first solving a Stokes equation for a channel problem with periodical boundary conditions. The solution of the velocity for this channel problem is then used as fully developed inlet velocity. Furthermore, the initial conditions of the model are:
3 Numerical Method

To solve Equation 1-3 described in Section 2 with the appropriate boundary conditions, the finite element method package TFEM is used [21]. The unknowns are the velocity/gradient/pressure, temperature and conformation, which will all be treated as separate problems. An ALE mesh is generated using the open source mesh generator GMSH [22]. For the velocity/gradient/pressure problem, triangular P2/P1 (Taylor-Hood) elements are used and for the temperature triangular P2 elements are used. A continuous DEVSS-G/SUPG algorithm is used for better numerical stability [23].

3.1 Time integration

An iterative procedure is used to solve the coupled system of equations on the domain $\Omega(t)$. All relevant elements of the time stepping procedure are shown in Figure 4.

The position and angle of the particles $P_i$ are updated using a first-order Euler scheme for the first time step and a second-order Adams-Bashforth scheme for all subsequent time steps. Furthermore, the evolution equation of the log-conformation tensor is solved using a semi-implicit Gear scheme similar to [24]. The temperature and position of the free surfaces are updated using a predictor. For the first time step we use:

$$\hat{T}^{n+1} = T^{n},$$

6 Effects of processing conditions on orientation for short-fiber composites in FFF
\( \hat{H}^{n+1} = H^n, \) 

and for all subsequent time steps we use:

\( \hat{T}^{n+1} = 2T^n - T^{n-1}, \)

\( \hat{H}^{n+1} = 2H^n - H^{n-1}. \)

### 3.2 Remeshing

A boundary fitted mesh method (BFMM) is used to describe the particles in the fluid. The mesh around the particle boundary is refined and can move in time, while at the other boundaries the mesh is kept stationary. The motion of the mesh is independent of fluid motion, due to the arbitrary Lagrangian-Eulerian (ALE) formulation that is used [25]. As a result of the change in position of the particle the mesh can become deformed to accurately describe the solution, and in this case a new mesh is created while the outline of the mesh is retained. An example of this is shown in Figure 5.

![Figure 5: Mesh around particle before (left) and after (right) remeshing.](image)

Considering a multiple particle system, the possibility for particles coming close together may arise. To accurately describe the solution at a point between these particles, a minimum number of elements is defined there. In order to do this, the coordinates on the boundary of the particles need to be compared with each other. Unfortunately, the efficiency of this brute force method is limited since the number of nodes that needs to be compared increases exponentially for each extra particle. Therefore, a method with a close resemblance to a Verlet list[26] is used, where the distance of the centers of particles is first compared. Only if this distance is smaller than one particle length plus three element lengths, all coordinates on the particle boundary are compared. A refinement field is defined between these coordinates so that the number of elements is never smaller than three, as can be seen in Figure 6a.

In literature, ellipsoids are often used to represent fibers in materials processing simulations [27]. However, to give an accurate description of the fiber behavior in a fluid, large aspect ratios are necessary because the ability to rotate or tumble depends on this ratio. This gives rise to numerical difficulties as the ends of the ellipsoid tend to become very sharp for large aspect ratios. As a result, the number of elements on the ends needs to be very large, and simulation of multiple particles would require quite long computation times. To solve this problem, the discrete fibers in our model are represented by rectangular particles with semi circular ends, so that only a few elements are necessary on the fiber’s ends. The coordinates are given by an \( x \) and \( y \) position, as well as an angle \( \theta \), as shown in Figure 6b. The long axis of particles used in this study is, unless it is stated otherwise, 0.2 mm and the short axis is 0.01 mm which corresponds to an aspect ratio of 20:1.
4 Validation

4.1 Mesh convergence

The solutions for different meshes and time steps are compared in order to test the convergence. In Table 3, the characteristics of three different meshes are presented. Since the trajectory of the particle is of particular interest, the mesh at the particle boundary is refined for each case, and far away from the particle the mesh is coarse. The meshes M1, M2, and M3 corresponding to the data in Table 3 are depicted in Figure 7.

<table>
<thead>
<tr>
<th>Coarse element size</th>
<th>number of elements on particle</th>
</tr>
</thead>
<tbody>
<tr>
<td>M1 0.1</td>
<td>20</td>
</tr>
<tr>
<td>M2 0.05</td>
<td>32</td>
</tr>
<tr>
<td>M3 0.025</td>
<td>56</td>
</tr>
</tbody>
</table>

The trajectory of a single particle in the domain $\Omega$ is considered as a test case, and the results can be seen in Figure 8a and Figure 8b. It can be concluded that the solution clearly converges since the trajectories nearly overlap for all three meshes and time steps. In order to avoid long computation times for multiple particle systems we choose mesh M1 in combination with a time step $\Delta t = 5e - 4$ at the extrusion velocity of 7.5 mm/s. For higher extrusion velocities the time step must be adjusted so the product of both remains constant. One can expect that since the solutions for different meshes nearly overlap, a

![Figure 6](image-url)  
(a) Example of a refinement field between two particles that are close together. (b) Schematic representation of a single particle, with the angle $\theta$ defined with respect to the $x$-axis.

![Figure 7](image-url)  
(a) Meshes that were used to test mesh convergence, (a) corresponds to M1, (b) corresponds to M2 and (c) corresponds to M3.
coarser mesh might be possible. However, this results in errors during the re-meshing step.

4.2 Extrudate Swell Problem

The simplified extrudate swell problem described in Section 2 is used as a test case to validate the method in this study by comparing the numerical results to experiments. The results of the numerical part are described in detail in Appendix I, where the influence of different processing parameters is tested by plotting the angle of a single particle versus the $x$ position in the domain. The most important conclusion that can be drawn is that the length of the narrow part $L_2$ (see Figure 2a) has the most significant effect on the final fiber orientation. This is also shown in Figure 9, where the final fiber angle in the extrudate is plotted for more variations of $L_2$. Note that an alignment of 1 means that the fiber is fully aligned in the extrusion direction.

This result seems counterintuitive, but can be explained by Jeffery’s theory [28]. In the case that $L_2$ is short, the effects of extensional flow on the particle are dominant, since the particle only encounters shear at the very end of the nozzle. In the case that $L_2$ is long, the particle encounters a relative long period of developed shear flow and according to Jeffery’s theory, an ellipsoid performs a periodic tumbling motion in a shear flow with a period dependent on the aspect ratio. In the narrow section, the particle starts to rotate very slowly and leaves the nozzle at a slight angle dependent on the length $L_2$, after which it is further rotated by elastic recovery effects. This results are in agreement with the numerical simulations performed by Heller et al. [13], who also calculated the fiber orientation in the extrudate for fused filament fabrication based on a decoupled approach using a Folgar Tucker model.

Based on these results, two different nozzles have been designed with different values of $L_2$. Both

Effects of processing conditions on orientation for short-fiber composites in FFF
nozzles are then used to extrude two material sample sets, and the details of this process can be found in Appendix II. These sample sets are analyzed using an optical microscope and to quantify the differences in fiber orientation the image processing software ImageJ is used. Histograms are made for the two sample sets, using the angle data generated with the software. The processed images and histograms are presented in Figure 10 and Figure 11.

![Figure 10](image10.png) ![Figure 11](image11.png)

Figure 10: a) Analyzed image of extruded sample made with nozzle 1 with $L_2 = 1.5D_2$ and b) Histogram of fiber angle data based on this image.

Figure 11: a) Analyzed image of extruded sample made with nozzle 2 with $L_2 = 8D_2$ and b) Histogram of fiber angle data based on this image.

The average fiber orientation for both sample sets is $0^\circ$ with respect to the extrusion direction. However, the deviation from the average orientation for the sample set made with nozzle 2 (large $L_2$) is significantly larger compared to the sample set made with nozzle 1 (Small $L_2$). This is in agreement with the results from the numerical simulations, according to Figure 9, since it was predicted that the longer the narrow section of the nozzle, the more fibers tend to align perpendicular to the extrusion direction.

5 Results & Discussion

5.1 Free surface height

The shape of the free surfaces is highly dependent on the ratio between average extrusion velocity $u_{\text{avg}}$ and substrate velocity $u_{\text{sub}}$. Therefore, a relative substrate velocity $u_{\text{sub}}/u_{\text{avg}}^*$ is defined, where the average extrusion velocity $u_{\text{avg}}^*$ is corrected for the difference in nozzle outlet diameter $D_1$ and gap width $W_{\text{gap}}$ between nozzle and substrate according to:

$$u_{\text{avg}}^* = \frac{u_{\text{avg}} D_1}{W_{\text{gap}}}.$$  \hspace{1cm} (32)

To study the effect on the height $H$ of the deposited layer of material, several simulations are performed with different values of $u_{\text{sub}}/u_{\text{avg}}^*$. Note that the particles are not yet incorporated in this case. The results are shown in Figure 12, where the red dotted line indicates the gap width between nozzle and substrate.

In Figure 13, the steady state solution for three different substrate velocities is shown. It takes several iterations before this steady state is reached depending on the flow conditions, and it can be seen that there is a significant difference in layer height for the three relative substrate velocities.
Figure 12: Layer height versus relative substrate velocity. The red line indicates the initial height.

Figure 13: Steady state solution for a simulation without particles for a) $u_{\text{sub}}/u^*_{\text{avg}} = 1.4$, b) $u_{\text{sub}}/u^*_{\text{avg}} = 1.0$ and c) $u_{\text{sub}}/u^*_{\text{avg}} = 0.6$

In Figure 13b and Figure 13c, a bulge in front of the deposited material can be observed, which can give numerical problems when particles come close to that area. Therefore, to prevent this phenomenon, we choose $u_{\text{sub}}/u^*_{\text{avg}} = 1.2$ for the rest of the simulations unless it is explicitly stated otherwise.

5.2 Velocity profiles

To quantify the differences in the flow for the isothermal versus non-isothermal model the velocity profiles in several parts of the domain are plotted, as can be seen in Figure 14. The dotted lines represent the positions where the velocity is sampled. An average outlet velocity of 7.5 mm/s is imposed and for the non-isothermal simulation an initial material temperature of 200°C Celsius is used. Normally, the material enters the nozzles at ambient temperature but it is assumed that the material is already warmed up considerably to avoid large temperature gradients.

The differences between the two situations are most noticeable in the narrow part (2), where the flow profile of the non-isothermal simulation has a distinct, plug like character. This indicates the importance of incorporating thermal behavior in the simulations, since this flow profile can have major influence on fiber movement.

In order to visualize the effect of particles on the flow in the nozzle, a velocity profile is made at a position in the nozzle where particles are present. In Figure 15a, a section of the flow with multiple particles is shown and the dotted line represents the position where the velocity is sampled. The resulting velocity profile is shown in Figure 15b and although the profile obviously has a parabolic character, some irregularities can be observed at the position of the particles.

The discontinuities in the curve are caused by the particles, since there is no mesh inside and consequently no solution of the flow field. It can be concluded that the presence of a particle has an effect on the velocity close to boundary as a result of the local flow field induced by the particle motion.
5 RESULTS & DISCUSSION

Figure 14: Velocity profiles in the wide and narrow part of the nozzle for an isothermal and non-isothermal simulation.

Figure 15: A section of the flow with multiple particles (a) and the resulting velocity profile(b).

5.3 Particles simulation: Isothermal vs. non-isothermal

To study the influence of temperature gradients in the flow on the orientation behavior of fibers, isothermal simulations are compared to non-isothermal simulations. We consider a case where the fibers are initially aligned with the flow direction in the nozzle, $\theta_0 = 90^\circ$ and have an average extrusion velocity of either 7.5 mm/s or 30 mm/s. We also consider a case where the fibers are initially perpendicular to the flow direction in the nozzle, $\theta_0 = 0^\circ$ with an average extrusion direction of 7.5 mm/s or 30 mm/s. Note that $\theta$ and $\theta_0$ are defined with respect to the $x$-axis. An example of a simulation with particles that are initially aligned is shown in Figure 16, where the particle position is shown at three moments in time.
Comparing Figure 17 to Figure 13, it can be observed that the particles cause a distorted free surface shape. This originates from very small disturbances in the velocity field around the particle, which are discussed in Section 5.2. In this way, the free surfaces height is affected through Equation 4.

In Figure 17, a set of five particles is tracked in the flow domain. It can be observed that after deposition on the substrate, the particles do not rotate and essentially stay at the same relative position. From this we conclude that temperature effects after deposition on the substrate have no effect on the orientation of particles and therefore the influence of substrate temperature and ambient temperature is not tested.

To quantify differences in orientation, histograms are made from particle angle data at a specific point in the extrudate. First, isothermal simulations have been performed and the results are shown in Figure 18. It is assumed that the temperature in the whole domain $\Omega(t)$ is $T = 280^\circ\text{C}$. The differences in orientation are relatively small for all four cases, as most of the fibers have an angle close to $\theta = 0^\circ$ with a similar deviation for all cases.

In Figure 19 the temperature distribution in the domain can be seen for a non-isothermal simulation with an average extrusion velocity $v_{\text{avg}} = 30\text{ mm/s}$, where in the center of the nozzle a significantly lower temperature can be observed.

If we now compare the results for the isothermal simulations to the results of the non-isothermal simulations shown in Figure 20 we can conclude that for the simulations with initial fiber angles parallel to the flow direction there is no clear difference in deviation of the final angle. However, looking at the cases with an initial fiber angle perpendicular to the flow direction, the effect of temperature gradients can be seen in Figure 20c and Figure 20d. The deviation is clearly larger, and this effect can be attributed to the different velocity profiles discussed in the previous section. In this way, it is demonstrated that the non-uniform temperature distribution has a limiting effect on the movement of fibers. Therefore, if high alignment of fibers in the extrusion direction is desired one should pay attention to proper (pre-)heating of the material.
5 RESULTS & DISCUSSION

Figure 18: Normalized histograms of final particle angle data for isothermal simulations with (a) \( u_{avg} = 7.5 \text{ mm/s}, \theta_0 = 90^\circ \), (b) \( u_{avg} = 30 \text{ mm/s}, \theta_0 = 90^\circ \), (c) \( u_{avg} = 7.5 \text{ mm/s}, \theta_0 = 0^\circ \), (b) \( u_{avg} = 30 \text{ mm/s}, \theta_0 = 0^\circ \).

Figure 19: Temperature distribution in the domain \( \Omega(t) \). \( u_{avg}=30 \text{ mm/s} \).

5.4 Effects of substrate velocity on orientation

In Section 5.1 it is discussed that the shape of the extruded layers is highly dependent on the substrate velocity \( u_{sub} \) relative to the average extrusion velocity \( u_{avg} \). The effects of the final shape of the free
surfaces on fiber orientation are investigated by performing simulations with different $u_{sub}$. Histograms are made for the case where $u_{sub}/u_{avg}^* = 0.8$ and $u_{sub}/u_{avg}^* = 1.2$ which can be seen in Figure 21a and Figure 21b, respectively.

Here, the differences are clearly visible as for the case with a relatively low $u_{sub}$, the deviation of particle angles is large compared to case with a relatively high $u_{sub}$. For the latter, the amount of particles that

Figure 20: Normalized histograms of final particle angle data for non-isothermal simulations with (a) $u_{avg} = 7.5$ and $\theta_0 = 90^\circ$, (b) $u_{avg} = 30$ and $\theta_0 = 90^\circ$, (c) $u_{avg} = 7.5$ and $\theta_0 = 0^\circ$, (b) $u_{avg} = 30$ and $\theta_0 = 0^\circ$.

Figure 21: Normalized histograms of the final particle data for a simulation with (a) $u_{sub}/u_{avg}^* = 0.8$ and (b) $u_{sub}/u_{avg}^* = 1.4$. 

Effects of processing conditions on orientation for short-fiber composites in FFF
have an angle close to, or equal to $\theta = 0^{\circ}$ is considerably larger. This can be explained by the fact that there is a bulge of material on the left side of the extrudate, as can be seen in Figure 13c. In this bulge, the rotation of particles is quite random but it generally causes the fiber to be have a large angle with respect to the extrusion direction.

5.5 Effects of $L_2$ and $W_{\text{gap}}$

Since it was demonstrated for the simplified extrudate swell problem that the length of the narrow section of the nozzle $L_2$ has a significant effect on the orientation of particles, this parameter is also studied for non-isothermal simulations on the full domain $\Omega$. We consider a case where $L_2 = 0.75D_2$ mm and a case where $L_2 = 3.00D_2$ mm, and the results are shown in Figure 22.

![Normalized histograms of the final particle data for a simulation with (a) $L_2 = 0.75D_2$ and (b) $L_2 = 3.00D_2$.](image)

Figure 22: Normalized histograms of the final particle data for a simulation with (a) $L_2 = 0.75D_2$ and (b) $L_2 = 3.00D_2$.

Comparing both cases, it can be seen that the deviation for $L_2 = 3.00D_2$ is slightly larger. However, in general the difference is too small to conclude that the length $L_2$ has a significant effect on the final fiber orientation in this case.

Furthermore, the effect of the gap between nozzle and substrate $W_{\text{gap}}$ is studied. The range of values that can be tested for this parameter is limited, since values higher than $W_{\text{gap}} = 0.75D_2$ are not realistic and a value smaller than $W_{\text{gap}} = 0.50D_2$ gives numerical problems as the particles have a similar length scale. The results for the two different cases are shown in Figure 23.

![Normalized histograms of the final particle data for a simulation with (a) $W_{\text{gap}} = 0.50D_2$ and (b) $W_{\text{gap}} = 0.75D_2$.](image)

Figure 23: Normalized histograms of the final particle data for a simulation with (a) $W_{\text{gap}} = 0.50D_2$ and (b) $W_{\text{gap}} = 0.75D_2$.

For $W_{\text{gap}} = 0.50D_2$, it can be observed that the deviation is slightly larger and this effect might be attributed to the fact that the substrate velocity is higher to keep the ratio $u_{\text{sub}}/u_{\text{avg}}$ constant.
6 Conclusions

A numerical model was developed for the simulation of the fused filament fabrication process with short-fiber composites. The problem was split into two parts: First a simplified 2D domain representing the extrusion from the nozzle was considered, to get an understanding of the influence of extrudate swell on fiber orientation. It was shown that the length after the convergent section of the nozzle, $L_2$, has the most significant effect on the orientation after extrusion. Based on this conclusion, a short experimental study was done for validation and the results were in agreement with the simulations.

For the main problem, the deposition on the substrate was also incorporated in the 2D model. The importance of including thermal behavior was demonstrated by comparing isothermal and non-isothermal simulations. Due to the low thermal conductivity, the temperature throughout the nozzle is not constant and this results in a higher viscosity in the center compared with the isothermal case. Based on this conclusion, one can argue that the initial fiber orientation distribution in the feedstock filament is important for the desired orientation in the final product, since the ability for the fibers to align in the extrusion direction is limited by poor heat transfer. Therefore, the production of the filament itself might also be an important factor that has not yet been given attention in research.

We have shown that it is of great importance to accurately model the behavior of both free surfaces, since this has a major impact on the rotation of fibers after extrusion. In this way, it was demonstrated that the substrate velocity has the most significant effect on final fiber orientation. This in contrast to other parameters such as extrusion velocity and gap width, that showed little to no effects. In reality, the swelling of free surfaces is a three-dimensional problem so it is expected that the influence of this phenomenon might be even more dominant in the real life situation.

Overall we have provided a framework to optimize the fused filament fabrication process with short-fiber composites depending on the desired orientation. The fact that it is -to a certain extend- possible to influence the local micro-structure of a product is fairly unique. In most polymer processing applications such as injection molding, one has little to no control over the fiber orientation in the product. For future research, experiments need to be performed to investigate how the guidelines provided in this work can be used to optimize the mechanical properties of printed objects. In particular, the possibility of increasing the strength between consecutive layers by obtaining a random fiber orientation is of interest.

Moreover, the model in this work can be extended to study the influence of crystallization since this has a major influence on the mechanical properties of the final product and it might also affect the fiber orientation. Furthermore, the rheological properties in this study are assumed to be constant, however studying the influence of different matrix materials is also important for future research.
References


18 Effects of processing conditions on orientation for short-fiber composites in FFF


Appendix I: Extrudate Swell Problem

Problem Description & Boundary Conditions

For the simplified extrudate swell problem only one half of the nozzle is considered since the inside of the nozzle is axisymmetric, and the domain is shown in Figure 24. The governing equations are equal to those described in Section 2, except temperature is not included in this case.

The boundary and initial conditions are different than for the main problem, and read:

\[ u = 0 \quad \text{on } \Gamma_w \]  
\[ \int_{\Gamma_{in}} u \cdot n \, ds = -Q \quad \text{on } \Gamma_{in} \]  
\[ t = (pI + \tau) \cdot n = 0 \quad \text{on } \Gamma_{out} \]  
\[ t = (pI + \tau) \cdot n = 0 \quad \text{on } \Gamma_{fs} \]  
\[ t_x = (pI + \tau) \cdot e_x = 0 \quad \text{on } \Gamma_{sym} \]  
\[ u_y = 0 \quad \text{on } \Gamma_{sym} \]  
\[ H(y = 0, t) = H_0 \quad \text{on } \Gamma_{fs} \]  
\[ H(y, t = 0) = H_0 \quad \text{on } \Gamma_{fs} \]  
\[ s(x, t = 0) = s_0(x), \quad \forall x \in \Omega(0), \]  

where \( n \) is the outwardly directed unit normal vector. The boundary conditions on the particle are the same as in the main problem.

Results

An example of the velocity field in the extrudate swell domain \( \Omega(t) \) is shown in Figure 25. In the next subsections, the effect of several parameters is investigated by comparing the final particle angle after extrusion from the nozzle.

Influence of Weissenberg Number

The influence of viscoelasticity is tested by comparing the movement of a single particle in a flow with different Weissenberg numbers. The Weissenberg number is the ratio of elastic time scale versus the convective time scale, and in our case this is defined as:

\[ Wi = \frac{\lambda_{avg}}{D_2}, \]  

where \( v_{avg} \) is the average velocity at the outlet with diameter \( D_2 \). It can be concluded from Figure 26a that for an increasing Weissenberg number, the final fiber angle with respect to the extrusion direction...
increases. This can be explained by the increasing elastic effects in the nozzle, which cause more extrudate swell for higher Weissenberg numbers.

**Influence of Fiber Aspect Ratio**

The same method is applied to test the influence of fiber aspect ratio, and the results can be seen in Figure 26b. It can be observed that the differences are relatively small, but still it can be concluded that the flow has more influence on the movement of fibers with a small aspect ratio. This is in agreement with the theory of Jeffery [28].

**Influence of Nozzle Convergence Angle**

To investigate whether the convergence angle of the nozzle influences the final fiber orientation, the same method as in the previous subsections is applied. Four different conversion angles are tested and the results are shown in Figure 27a, for an initial fiber angle of $\theta_0 = 30^\circ$. In Figure 27b, the same simulation is performed for a particle with an initial angle of $\theta_0 = -30^\circ$. Comparing both figures, it can be concluded that the initial fiber angle in this case is not important since the final angle, at $x = 7$ mm, is nearly the same for all cases. It must be noted that this conclusion might not hold for non-isothermal simulations, where the viscosity is not uniform throughout the nozzle. Furthermore, it seems that the convergence angle has a negligible effect on the final angle.

**Influence of L2**

The length of the narrow section $L_2$ has been varied and the results are shown in Figure 28. Interestingly, for longer values of L2 the final angle is larger with respect to the extrusion direction.
Appendix II: Experimental Methods

For the experiments, two different FFF nozzles have been designed based on the results of the simulations described in Appendix I. The dimensions are shown in Table 4. The nozzles are used in combination with an Airwolf 3D® AXIOM printer. Samples have been made by adjusting the G-code so that the extruded material is not deposited on the substrate but instead extruded vertically. Extruded wires are then sanded and polished so that a smooth cross section is obtained. Visualizing fiber orientation is done by using polarized light microscopy, and the results are presented in Figure 29 and Figure 30.

Table 4: Dimensions of nozzle N1 and nozzle N2.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value N1</th>
<th>Value N2</th>
<th>Unit</th>
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</thead>
<tbody>
<tr>
<td>Inlet diameter</td>
<td>$D_1$</td>
<td>3.2</td>
<td>3.2</td>
<td>mm</td>
</tr>
<tr>
<td>Outlet diameter</td>
<td>$D_2$</td>
<td>0.4</td>
<td>0.4</td>
<td>mm</td>
</tr>
<tr>
<td>Total nozzle length</td>
<td>$L_1$</td>
<td>8.25</td>
<td>8.25</td>
<td>mm</td>
</tr>
<tr>
<td>Length of capillary</td>
<td>$L_2$</td>
<td>0.6</td>
<td>3.2</td>
<td>mm</td>
</tr>
<tr>
<td>Convergence angle</td>
<td>$A_1$</td>
<td>60</td>
<td>60</td>
<td>°</td>
</tr>
</tbody>
</table>

Based on these pictures, we can qualify the differences in fiber orientation for both nozzles. In Figure 29, it can be observed that the alignment of fibers is relatively high. Note that these images are taken from different sample sets for reproducibility. In Figure 30 it can be observed that the fiber orientation is much more random.

Effects of processing conditions on orientation for short-fiber composites in FFF 23
Figure 29: Microscopic images of PA6/66 with 10 weight % Carbon Fiber, extruded with Nozzle N1 at 280° C and 50 mm/s.

Figure 30: Microscopic images of PA6/66 with 10 weight % Carbon Fiber, extruded with Nozzle N2 at 280° C and 50 mm/s.