







## Open Archive Toulouse Archive Ouverte

OATAO is an open access repository that collects the work of Toulouse researchers and makes it freely available over the web where possible

This is an author's version published in: <http://oatao.univ-toulouse.fr/20921>

### To cite this version :

Bakrani Balani, Shahriar  and Cantarel, Arthur  and Chabert, France  and Nassiet, Valérie  *Influence of parameters controlling the extrusion step in fused filament fabrication (FFF) process applied to polymers using numerical simulation.* (2018) In: ESAFORM 2018, 23 April 2018 - 25 April 2018 (Palermo, Italy).

Any correspondence concerning this service should be sent to the repository administrator: [tech-oatao@listes-diff.inp-toulouse.fr](mailto:tech-oatao@listes-diff.inp-toulouse.fr)

# Influence of parameters controlling the extrusion step in Fused Filament Fabrication (FFF) process applied to polymers using numerical simulation

Bakrani Balani Shahriar<sup>1, 2, a)</sup>, Cantarel Arthur<sup>2, b)</sup>, Chabert France<sup>1, c)</sup>, Nassiet Valérie<sup>1, d)</sup>,

<sup>1</sup> LGP-ENIT-INPT, University of Toulouse, 47 Avenue d'Azereix, BP1629-65016 Tarbes Cedex, France  
Web Page: <http://www.enit.fr/>

<sup>2</sup> Institut Clément Ader (ICA), CNRS UMR 5312, University of Toulouse, UPS, France  
Web Page: <http://www.institut-clement-ader.org/>

<sup>a)</sup>Corresponding author: [sbakrani@enit.fr](mailto:sbakrani@enit.fr)

<sup>b)</sup>[arthur.cantarel@iut-tarbes.fr](mailto:arthur.cantarel@iut-tarbes.fr)

<sup>c)</sup>[fchabert@enit.fr](mailto:fchabert@enit.fr)

<sup>d)</sup>[vnassiet@enit.fr](mailto:vnassiet@enit.fr)

**Abstract.** Extrusion is one of the oldest manufacturing processes; it is widely used for manufacturing finished and semi-finished products. Moreover, extrusion is also the main process in additive manufacturing technologies such as Fused Filament Fabrication (FFF). In FFF process, the parts are manufactured layer by layer using thermoplastic material. The latter in form of filament, is melted in the liquefier and then it is extruded and deposited on the previous layer. The mechanical properties of the printed parts rely on the coalescence of each extrudate with another one. The coalescence phenomenon is driven by the flow properties of the melted polymer when it comes out the nozzle just before the deposition step.

This study aims to master the quality of the printed parts by controlling the effect of the parameters of the extruder on the flow properties in the FFF process. In the current study, numerical simulation of the polymer coming out of the extruder was carried out using Computational Fluid Dynamics (CFD) and two phase flow (TPF) simulation Level Set (LS) method by 2D axisymmetric module of COMSOL Multiphysics software. In order to pair the heat transfer with the flow simulation, an advection-diffusion equation was used. Advection-diffusion equation was implemented as a Partial Differential Equation (PDE) in the software. In order to define the variation of viscosity of the polymer with temperature, the rheological behaviors of two thermoplastics were measured by extensional rheometer and using a parallel-plate configuration of an oscillatory rheometer.

The results highlight the influence of the environment temperature and the cooling rate on the temperature and viscosity of the extrudate exiting from the nozzle. Moreover, the temperature and its corresponding viscosity at different times have been determined using numerical simulation. At highest shear rates, the extrudate undergoes deformation from typical cylindrical shape. These results are required to predict the coalescence of filaments, a step towards understanding the mechanical properties of the printed parts.

## INTRODUCTION

Additive manufacturing refers to a wide variety of processes for manufacturing of 3D complex shape parts. These emerging manufacturing technologies are sorted according to various criteria such as the nature of the raw material (metal, ceramic or polymer), the process of material deposition, the energy source, etc. Among these processes, FDM (Fused Deposition Modeling), also called FFF (Fused Filament Fabrication) is the most widely

used technique. In this process, the parts are manufactured layer by layer, in most cases using thermoplastic materials. However, the manufactured parts by FFF lack frequently in mechanical resistance which is mainly due to poor coalescence and interdiffusion between deposited layers and beads. Understanding the parameters influencing the coalescence and optimizing these parameters are very important in order to improve the mechanical properties of printed parts.

This study presents the numerical simulation of the melted polymer flowing from the nozzle up to the time just before the deposition. Then, the filament is deposited close to another one to form the printed part layer by layer. The shape of the filament as well as the thermal field and the viscosity of the filament change during this time. With these results, we gain insight into the temperature and the viscosity variations during the extrusion of the filaments just before its deposition. Studies have been carried out on two polymers, PLA is one the most used polymer in FFF process and PEEK is a high-performance thermoplastic with high viscosity and melting temperature.

## MATERIAL AND METHODOLOGY

In the following study, the numerical simulation of the extrusion step has been carried out by using the “Two phase flow simulation” (TPF) of COMSOL Multiphysics software. Level set (LS) and Navier-Stokes equations are used in order to model the flow of the polymer when it exits from the nozzle. Navier-Stokes equations describe the motion of the fluids in the continuous media while there is conservation of the mass in the system. Level set (LS) method is a transport equation which is added to Navier-Stokes equations in order to track the interface of two immiscible fluids [1]. Equation 1, Equation 2 and Equation 3 show the Navier-Stokes, continuity and level set equations used in TPF module of COMSOL:

$$\rho \frac{\partial u}{\partial t} = \nabla \cdot [-pI + \mu(\nabla u + (\nabla u)^T)] + \rho g + F_{st} + F \quad (1)$$

$$\rho(u \cdot \nabla)u = 0 \quad (2)$$

$$\frac{\partial \phi}{\partial t} + \nabla \cdot (u\phi) = \gamma \nabla \cdot (\varepsilon_{ls} \nabla \phi - \phi(1 - \phi) \frac{\nabla \phi}{|\nabla \phi|}) \quad (3)$$

Where  $\rho$  is the density,  $u$  is the flow velocity,  $P$  is the pressure applied to the fluid, and  $\mu$  is the dynamic viscosity of the fluid. In the level set equation,  $\phi$  is the volume fraction,  $\gamma$  is a re-initialization parameter and  $\varepsilon_{ls}$  is the parameter controlling the interfacial thickness.  $F_{st}$  represents the force resulting from the surface tension and  $F$  is for all other external forces.

The color legend of the volume fraction in TPF varies from 0 to 1 (blue to red): when the value of the color function is 1 this mesh is fully filled by the polymer, otherwise there is air. The interface of the two fluids, polymer and air, is considered as 0.5 value.

For the numerical simulation in COMSOL Multiphysics, free triangular default physics-controlled meshes have been selected. Figure 1 shows used boundary conditions for the numerical simulation of TPF and heat transfer.

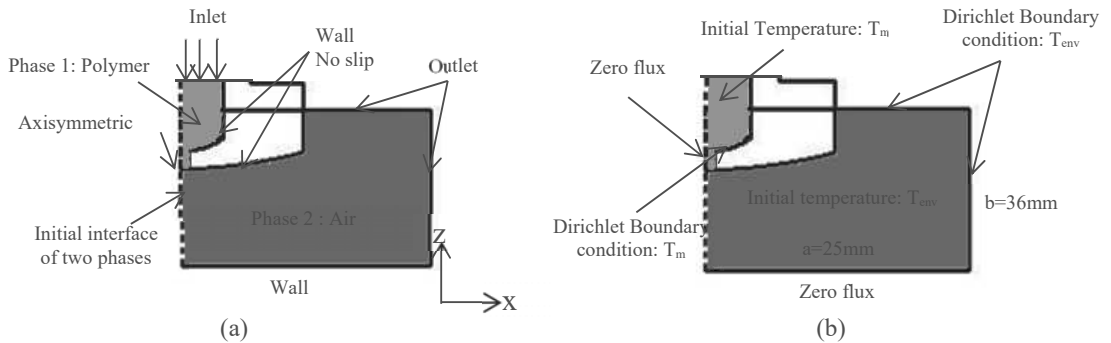


FIGURE 1: Boundary conditions for numerical simulation (a) flow, (b) heat transfer

In order to determine the heat transfer in the flow simulation, a partial differential equation (PDE) [2] has been added to the model.

Equation 4 shows the advection-diffusion equation for determining the variation of the temperature in our model taking into account the movement of the fluid in the system :

$$\frac{\partial T_S}{\partial t} + (\vec{u} \cdot \nabla) T_S = \left( \frac{k}{\rho C_p} \right) \nabla^2 T_S \quad (4)$$

Where T is the temperature and  $\vec{u}$  is the fluid velocity. Here, the fluid velocity is the term that links the flow to PDE, k is the thermal conductivity,  $\rho$  is the density and  $C_p$  is the heat capacity of the fluid. The term  $\left( \frac{k}{\rho C_p} \right)$  represents the thermal diffusivity of the material which is represented in equation 5 as  $\alpha$ . Table 3 shows the properties of air, PLA and PEEK used in the advection-diffusion equation.

**TABLE 1:** Thermal properties of air, PLA and PEEK for determining the thermal diffusivity (from [3] [4] [5] [6])

Phase	Unit	Air	PLA	PEEK
Thermal conductivity	W.m <sup>-1</sup> .K <sup>-1</sup>	0.024	0.205	0.25
Density	kg.m <sup>-3</sup>	1.225	1250	1320
Heat capacity	kJ.kg <sup>-1</sup> .K <sup>-1</sup>	1.005	2.1	1.7
Thermal diffusivity	m <sup>2</sup> .s <sup>-1</sup>	1.95.10 <sup>-5</sup>	7.9.10 <sup>-8</sup>	1.115.10 <sup>-7</sup>
Activation energy	kJ.mol <sup>-1</sup>	-	114	82.8

In order to define the thermal diffusivity for each phase, Equation 5 has been used in advection-diffusion equation:

$$\alpha(\phi) = (\phi * \alpha_{air}) + ((1 - \phi) * \alpha_{polymer}) \quad (5)$$

Where  $\phi$  is the volume fraction determined by TPF module,  $\alpha_{air}$  and  $\alpha_{polymer}$  are the thermal diffusivity of air and polymer respectively. To take into account the metallic parts such as the nozzle in the model, the term  $\vec{u}$  in Equation 4 is neglected since there is no fluid in this region. The viscosity varies with shear rate and temperature, following the Equation 6. The viscosity of shear-thinning fluids is well described with the Carreau model in Equation 7 which takes into account the Newtonian plateau at low shear rate [7][8].

$$\eta(\dot{\gamma}, T) = a_T \eta(\dot{\gamma} a_T, T_0) \quad (6)$$

$$\eta = \eta_{inf} + (\eta_0 - \eta_{inf}) [1 + (\lambda \dot{\gamma})^a]^{\frac{n-1}{a}} \quad (7)$$

Where  $\eta_0$  is the viscosity at zero shear rates and  $\eta_{inf}$  is the viscosity at infinite shear rate,  $\lambda$  is the relaxation time index; n is the power index,  $\dot{\gamma}$  is the shear rate and  $a_T$  is expressed as the Arrhenius law presented in Equation 8 [9]. The viscosity of PEEK (PEEK 450G by Victrex) and PLA (Premium White of 3 mm diameter provided by eMotion Tech company) were measured by rheometry in parallel-plate configuration with frequency sweeps from 0.01 s<sup>-1</sup> to 100 s<sup>-1</sup> within the linear viscoelastic domain, the results have been published elsewhere [10]. Their shear viscosity at high shear rates (up to 10<sup>4</sup> s<sup>-1</sup>) and extensional viscosity have been measured thanks to an extruder equipped with an extensional die from Thermofisher Company. The terms of the Carreau model have been extracted from the fitting of the viscosity curves for both polymers. The terms are presented in table 2.

**TABLE 2:** Values of terms in Carreau model for PEEK and PLA

Polymer	PLA	PEEK
Temperature	485 K	655 K
$\eta_0$	1945	7071
$\eta_{inf}$	0	0
$\lambda$	0.0798	1.4542
a	1.931	0.781
n	0.7032	0.5966

$$a_T = \exp \left[ \frac{E_a}{R} \left( \frac{1}{T} - \frac{1}{T_0} \right) \right] \quad (8)$$

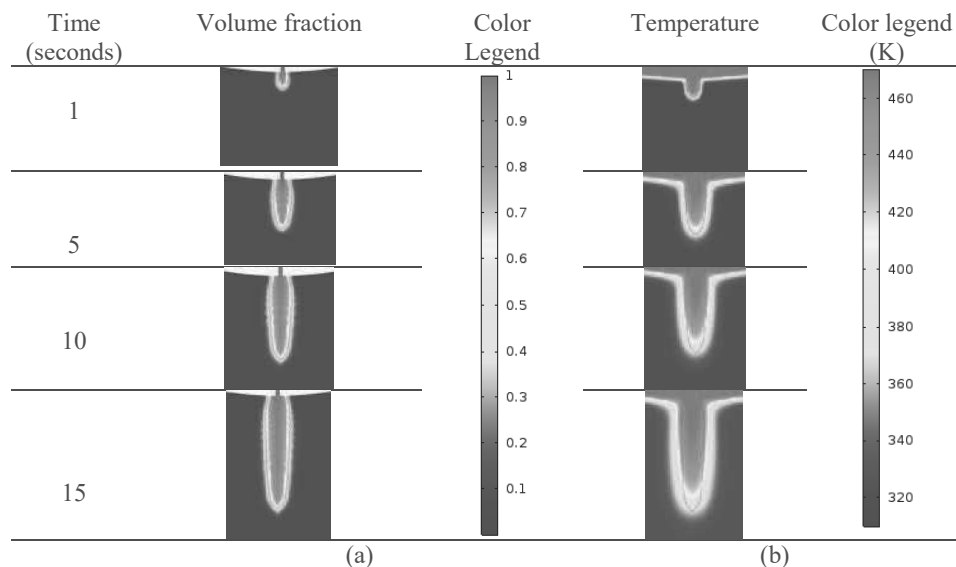
where  $E_a$  is the activation energy [5] [6] and  $R$  is the gas constant [5].

## RESULTS AND DISCUSSION

The numerical simulation of the polymer flow has been determined for PLA and PEEK when it exits from the nozzle at  $t=0$  up to  $t=18$  seconds. The initial temperatures are chosen at  $T_m= 468$  K for PLA and  $T_m= 656$  K for PEEK. At these temperatures, the polymer is fully melted [10]. Two temperatures of environment for each polymer, representing the temperature of the printer's oven are selected as following: for PLA,  $T_{env}= 310$  K (room temperature) and  $T_{env}= 350$  K (glass transition ( $T_g$ ) of PLA). For PEEK,  $T_{env}= 310$  K (room temperature) and  $T_{env}= 428$  K ( $T_g$  of PEEK). The inlet velocity for all the simulations was selected at  $0.36$  mm.s<sup>-1</sup>.

Figure 2 shows the evolution of the volume fraction and temperatures for PLA at  $T_{env}= 310$  K at different times from  $t=0$  to  $t=15$  s. The black line represents the volume fraction of 0.5 in the model which is considered as the interface of two fluids (air and polymer). Figure 3 presents the volume fraction, the temperature and the viscosity for PLA at  $T_{env}= 310$  K at  $t=25$  s. The blue area is the environment or printer's oven.

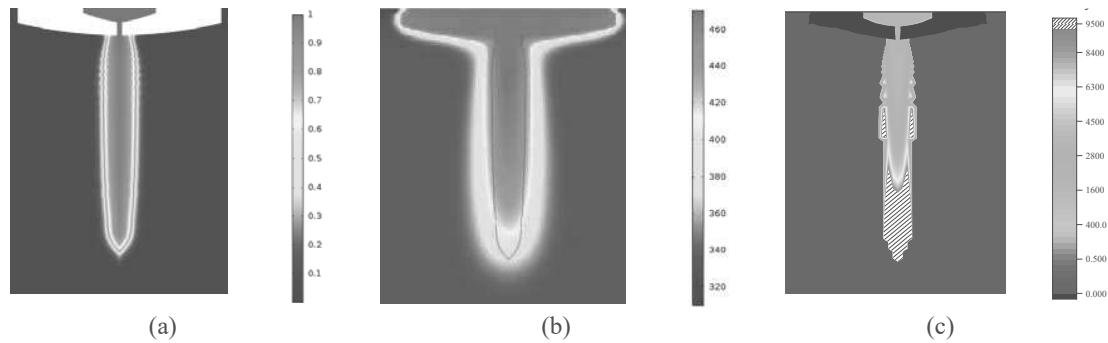
As it is seen in Fig. 3(b), the temperature of the filament decreases while it exits from the nozzle and it is exposed to the environment (room temperature). Furthermore, the numerical simulation shows the gradient of the temperature between the center to the side of the extrudate, the temperature difference is up to 70K. Consequently, while the center of the extrudate is still in the melted state, the external wall of the extrudate has already turned into solid state. Simultaneously, the viscosity in Fig. 3(c) changes from 400 Pa.s in the center to 5000 Pa.s on the sides.



**FIGURE 2:** Evolution of the state of volume fraction and temperature for PLA with time for  $T_m=468$  K and  $T_{env}= 310$  K, (a) volume fraction, (b) temperature (K)

The solid state, for which the viscosity is higher than 9000 Pa.s is represented in hatched area of the extrudate in Fig. 3(c). This region is not well defined because the polymer has changed from liquid state into solid state, so the equations of flow are not valid anymore.

Similar results were obtained with PEEK (not shown). From the numerical simulation of the flow of both PLA and PEEK, the variation of temperature and viscosity of the extrudates according to two environment temperatures are plotted in Fig. 4. For both polymers, the temperature and viscosity are noticed in the center of the extrudate from the distance  $d=0$  mm (point I, just after exiting the nozzle) up to  $d= 20$  mm (point II, at  $t=25$  s for which the volume fraction is 0.5).



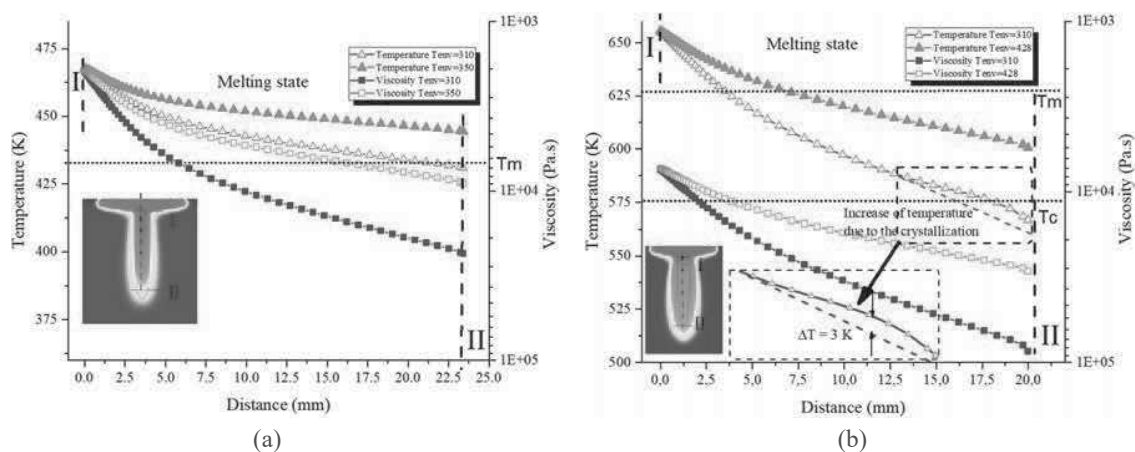
**FIGURE 3:** Numerical simulation for extrusion of PLA at  $T_{env}= 310$  K for  $t= 25$ s  
 (a) volume fraction, (b) temperature (K), (c) viscosity (Pa.s)

The variations of temperature and viscosity for PLA are plotted in Fig. 4(a). When exposed to 310 K (room temperature), the extrudate turns from liquid to solid after a distance of 17.5 mm, whereas the distance of 20 mm is required when the extrudate flows into the air regulated at 350 K.

The variations of temperature and viscosity for PEEK are presented in Fig. 4(b). When exposed to 310 K (room temperature), the cooling of the extrudate is very fast, it turns to solid after 3 mm of extrusion. This distance is higher, about 6 mm when the extrudate exits in an environment regulated at 428 K.

Comparing these results, it is obvious that PLA and PEEK do not behave the same way on cooling: the effect of environment temperature is weak on PLA extrudates and the liquid to solid transition is pushed back to have available the whole distance to deposit the filament. For these reasons, PLA appears to be easily processable which means that a deposited PLA filament has much time to flow and to coalesce with the nearby deposited beads. On the contrary, the PEEK extrudate reach its solid state faster than PLA when it exits from the nozzle. As soon as the distance of 6 mm, the PEEK filament is so rigid that it could not flow to coalesce with the closest deposited beads. As a consequence, PEEK would be very hard to process by FFF, since the time available to deposit the filament is very short. One hypothesis is the gap between the temperature of nozzle and the environment temperature: Comparing the influence of environment temperature on the temperature distribution of PLA and PEEK, it is shown that increasing the environment temperature highly influences the temperature curve of the PEEK while, the influence of environment temperature on the PLA are negligible.

The comparison between the obtained results for the volume fraction show that the length (the distance from points I to II) of PLA extrudate is higher than for PEEK. For the same elapsed time, the length of the PLA extrudate is about 24 mm whereas for the PEEK the maximum length is about 20 mm. These differences are due to the viscosity, the viscosity of PLA being lower than those of PEEK at the same shear rate.



**FIGURE 4:** Determined temperature and temperature related viscosity at 25 s for PLA and 18 s for PEEK.  
 In the center of the extrudate (a) PLA with  $T_m=468$  K (b) PEEK with  $T_m= 656$ K

Our study takes into account the crystallization on cooling. Both PLA and PEEK are semi-crystalline polymers. The crystallization is an exothermic phenomenon: the released heat and the kinetics of crystallization depends on each polymer. Comparison of PLA and PEEK shows that the kinetics of crystallization for PEEK is much faster than PLA. The numerical simulation reveals that for the studied temperatures for PLA, no crystallization occurs during the extrusion. In order to increase the crystallinity of the PLA, the cooling rate must be slower. On the contrary, for PEEK, the polymer undergoes crystallization even though its cooling rate is fast. Our numerical simulation for PEEK at room temperature shows that when the temperature of the extrudate reaches 590 K and below, the crystallization starts. The crystallization increases of 3K the temperature of the extrudate. In the core of the extrudate when it is exposed at room temperature, the crystalline rate reaches up to 20 % at  $t = 25$  s. In the same manner, when the time is increased for PEEK when it is exposed to an environment regulated at its glass transition, the crystalline phase will appear when the polymer reaches its crystallization temperature.

## CONCLUSION

In the current study, the extrusion of two semi-crystalline polymers, PLA and PEEK are modeled in order to optimize the FFF process parameters. The thermal field and the evolution of the viscosity of polymers were calculated. The comparison of the results for PLA and PEEK show that the cooling rate of PEEK is faster than PLA which means the PLA stays longer above its melting temperature than PEEK during the extrusion step. As a consequence, the coalescence of PLA during FFF process is more favorable than for PEEK.

While the temperature in the center of the extrudate remains at melting temperature, the cooling rate at the border of the extrudate close to air is much faster than in the center which is unfavorable to the coalescence of two adjacent filaments during the process.

Hereby, using numerical simulation, we have determined the crystalline rate of both polymers when they exit from the nozzle and the heat release by their crystallization. The numerical simulation for PLA shows that no crystallization appears during the extrusion step. On the contrary, PEEK has a faster tendency to crystallization. Consequently, while the environment temperature does not have much influence on the crystallinity of PLA, for PEEK, the temperature of environment highly influences the crystallinity of the polymer. For these reasons, it is compulsory to regulate very finely the temperature of the oven for succeeding in printing PEEK parts.

This extrusion is the primary step of the process toward modeling of FFF process. We have already studied the coalescence of two beads [10] for the same polymers. In future work, the deposition of beads on a substrate coupled to the coalescence of them will be modeled. These results will be compared to experimental results.

## REFERENCES

1. Olsson, E., Kreiss, G. & Zahedi, S. A conservative level set method for two phase flow II. *J. Comput. Phys.* 225, 785–807 (2007).
2. Landau, L. D. & Lifshitz, E. M. *Fluid Mechanics*, Pergamon Press, (1987).
3. Victrex. Victrex® PEEK 450G. (2014).
4. Garlotta, D., A Literature review of Poly Lactic Acid. *J. Polym. Environ.* 9, 63–84 (2001).
5. Kaseem, M. & Gun, Y. Melt Flow Behavior and Processability of Poly(lactic Acid)/Polystyrene (PLA/PS) Polymer Blends, *J. Polym. Environ.* (2016). doi:10.1007/s10924-016-0873-5
6. Shibuya, N. & Porter, R. S. A kinetic study of PEEK sulfonation in concentrated sulfuric acid by ultraviolet-visible spectroscopy, *Polymer*, 35, 3237–3242 (1994).
7. Dealy, J. M. & Wang, J., *Melt Rheology and its Applications in the Plastics Industry*. Melt Rheology and its Applications in the Plastics Industry, Springer Science, (2013). doi:10.1007/978-94-007-6395-1
8. Schramm, G., *A Practical Approach to Rheology and Rheometry*. Rheology (1994).
9. Lafleur, P. G., *Extrusion des polymères*. (Lavoisier, 2014).
10. Bakrani Balani, S., Chabert, F., Nassiet, V., Cantarel, A. & Garnier, C. Behaviour on the coalescence phenomenon Toward improvement of the properties of parts manufactured by FFF (Fused Filament Fabrication) through understanding the influence of temperature and rheological behaviour on the coalescence phenomenon. in *AIP Conference Proceedings* 1896 40008, (2017).