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Full Length Article

Complex flow and temperature history during melt extrusion in material extrusion additive manufacturing

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ABSTRACT

3D printing using the materials extrusion additive manufacturing (ME-AM) process is highly nonisothermal. In this process, a solid polymer filament is mechanically drawn into a heated hot end (liquefier) where the polymer is ideally melted to a viscous liquid. This melt is extruded through an orifice using applied pressure of the solid filament that is continuously being drawn into the extruder. The extruded filament melt is deposited to build up the desired part. The poor thermal conductivity of most polymers inevitably leads to temperature gradients, in both the radial and axial directions. Here we quantify the temperature evolution of the polymer filament in axial direction using embedded fine thermocouples as a function of process parameters. Information about the radial gradients is obtained by introducing dye markers within the filament through understanding the flow behavior through the extruder by the deformation of the dye from a linear to pseudo parabolic profile. The polymer is heated above the glass transition temperature for less than 30 s for reasonable print conditions with the center of the filament remaining cooler than the liquefier temperature throughout the process. These process measurements provide critical data to enable improved simulation and modeling of the ME-AM process and the properties of the printed parts.

1. Introduction

Material Extrusion(ME) [1], or fused filament fabrication(FFF), is one of the most popular additive manufacturing (AM) techniques due to its low cost and accessibility for the consumer market. ME is a well-defined AM process where molten polymer is deposited onto the build platform to produce three-dimensional objects [2] through control of the x-y motion of the extruder and z (height) position of the build platform. The process parameters associated with this polymer melt based AM technique can significantly impact the properties and quality of the printed part [3,4] This strong dependency on the processing conditions can lead to high variability in part properties if process control is insufficient [5–9].

At the heart of the material extrusion additive manufacturing(ME-AM) process is the extruder where the solid filament is melted and deposited. The solid polymer filament is continuously fed into a temperature-controlled heated barrel (often referred to as liquefier or hot-end), generally using two pinch rollers. Conductive heating transforms the solid filament into a viscous melt in the liquefier, while the incoming cold solid filament pushes the melt through the nozzle to be deposited on the build platform. Self-extruding filament acts as both feedstock and plunger to simplify the design of an ME-AM extruder. This simple design allows the extruder to be integrated with a common desktop CNC gantry for x-y control of the position of the extruder. This ME-AM extrusion process is significantly different from traditional polymer filament extrusion process, such as a single-screw extruder where frictional heating from the screw is the dominant energy source to transform the solid polymer to the melt and the screw homogenizes the polymer melt as well as generates significant pressure to drive isothermal melt flow through the die [10]. Commercial extruders for bulk polymer processing are carefully designed to ensure complete mixing and isothermal properties of the polymer melt prior to exit.

Conversely in an ME-AM extruder, the polymer is heated solely by conduction from the liquefier barrel. The poor thermal conductivity of polymers leads to a large temperature gradient as the polymer filament is pushed through the extruder. This gradient will lead to complexities in flow and thermal histories for the filament from surface to axis center of the filament that are not present in typical single-screw extruders. This temperature history will impact the properties of the printed part, but we have a limited understanding of the temperature profiles associated with the ME-AM process. Understanding the extrusion process could facilitate the development of feedstock material, optimization of

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processing conditions and design of manufacturing hardware.

Due to its importance for ME-AM, mathematical models associated with roller feeding mechanisms [11], heat transfer [12] and pressure drop [13] in the liquefiers have been proposed to understand the extrusion process in ME-AM. Bellini et al. [13] estimated the pressure drops in three geometrically distinct sections of a liquefier by solving the momentum flux balance in each section. This work used a power-law shear-dependent flow to describe the polymer melt, and incorporated a thermal convection model based on a constant heat flux and an Arrhenius temperature-viscosity relationship. However, the Arrhenius law is suitable only for linear semi-crystalline above its melting point or amorphous polymers at temperature significantly greater than its glass transition temperature ($T > T_g + 100\,^{\circ}C$) [14]. An additional limitation is that the power-law model is generally only valid for a limited range of shear rates. An additional challenge in correctly assessing the pressure gradient and flow patterns in the extruder is the temperature gradient induced by conduction from the liquefier that will affect the spatiotemporal flow behavior. The complex temperature profile leads to widely varying polymer melt properties as the filament is melted that makes modeling of the melt flow behavior in the liquefier difficult.

To understand better the temperature complexities, Yardimici et al. [12] employed finite element analysis (FEA) [15] simulations to determine the temperature gradient in the extrusion phase of ME-AM using a computational model. By examining the steady state heat transfer and flow behavior of the ME-AM extrusion process of poly-ε-caprolactone with FEA, Ramathan et al. [16] demonstrated a much smaller pressure drop than predicted from the mathematical models proposed by Bellini [13]. These results provide evidence of the need to understand the temperature profile in order to accurately model the ME-AM process. With the development of more functional materials in ME-AM, such as ABS-Iron composite [17], the melt flow behavior in the ME-AM extrusion process will require understanding of the temperature-heat capacity relationships, such as that proposed by Marcus et al. [18]. Although FEA can effectively analyze the fluid dynamics and the heat-transfer when boundary conditions are known, there are other complexities in the ME-AM process that could lead to discrepancies between the simulated results and the actual process. Notably the abrupt changes in physical properties at melting point or T_g, such as heat-capacity, density and rheological properties, will occur as the filament enters the liquefier, which can lead to challenges in the convergence of the FEA. In addition, as discussed previously, the Power-law model, which is commonly used in FEA to describe the flow properties, works only for a limited range of shear rate and temperature. Moreover, the air gap between the filament and the barrel at liquefier entrance complicates boundary conditions in FEM for examining the polymer flow for the ME-AM process. Carefully understanding the temperature profiles for FEA could help to refine the properties of ME-AM fabricated parts, such as residual stress [19], part distortion [7] and mechanical behaviors [6,20,21].

In order to better understand the ME-AM process, there has been several experimental studies in this area, which include in-situ monitoring of strain and temperature distributions during the ME-AM melt deposition on a platform [22]. Similarly, infrared imaging [23] and thermocouples (TC) [24] have been used to monitor the cooling and reheating during the melt deposition process as the molten fibers are deposited. However, the melt flow behavior of the polymer in the ME-AM extruder has not been reported experimentally to the best of our knowledge.

In this work, we present an experimental method to analyze the flow behavior and temperature history during the extrusion process for ME-AM. To visualize the flow, inorganic pigments are selectively included in the filament as flow indicators to illustrate the flow history of the polymer during the extrusion process. The pigment distribution is tracked both in extruded filament as well as the filament remaining in the extruder nozzle using both optical microscopy and X-ray micro computed tomography (MicroCT). The flow profiles can be rationalized through in-situ real-time temperature measurements during filament extrusion, which uses embedded ultra-fine thermocouples (TC) in the filaments. These measurements provide insight into the flow and temperature evolution through liquefier during ME-AM printing, which can be used to help refine models to better predict the properties of additive manufactured parts with ME-AM.

2. Material & methods

2.1. Materials

The polymer was in this study was a bisphenol-A polycarbonate (PC) (Covestro Inc., Makrolon 3208). Prior to any melt processing, PC pellets (as obtained from Covestro, Inc.) or PC filaments were dried in a vacuum-oven at 110 °C for 12 h to remove residual water. This water can lead to a reduction of the molecular weight of the PC during melt processing. Inorganic pigments (ultramarine, LANSCO UPL-2905) were used as flow indicators to reveal the flow history of PC filament during printing.

2.2. Viscosity measurement

The viscosity of PC at different shear rate was measured using a Rosand RH7 capillary rheometer at 325 °C. Results are shown in Fig. A1. The PC pellets were dried in a vacuum-oven at 110 °C for 12 h to remove residual water prior the measurement.

2.3. Filament extrusion

Filaments of PC were extruded using a HAAKE single screw extruder (Model Rheomex 252p) that was equipped with a gear pump and a simple circular die (diameter = 2.2 mm). This extruder has 3 independently controller temperature zones that can be used to modulate the viscosity of the PC during the extrusion. For the filament extrusion, a temperature profile from the feeding section to metering section was set at 280 °C, 290 °C, 275 °C; while the gear pump and circular die were set to 280 °C and 240 °C. The extruded PC melt was quenched in a room temperature water bath and then drawn onto a take-up wheel. The diameter of extruded filament was drawn down to 1.7 mm. by controlling the take-up speed relative to the extrusion rate. The diameter of the filaments was controlled to 1.70 ± 0.03 mm for these studies.

2.4. 3D printer

For the ME-AM, a customizable 3D printer was used in this study: Cartesio 3D printer (Model: W09). This printer was equipped with an E3D-v6 (1.75 mm-type) hot-end(liquefier) assembly with a 0.4-mm nozzle. The liquefier was heated using a 24 V-40 W cartridge heater (E3D).

2.5. Flow indicator

PC and blue inorganic pigments were dried in vacuum oven for 12 h at 110 °C before mixing. Then PC pellets and blue inorganic pigments were melt blended at a weight ratio of 4:1 in a Brabender mixer at 280 °C and 40 rpm for 10 min. This blue colored polymer was then pelletized, dried, and melt-spun using a capillary rheometer at 280 °C into 0.3 mm-fiber. These blue fibers were inserted into the non-pigmented PC filaments. To facilitate this, holes at intervals of 3 mm were drilled radially through the axis of PC filaments with a drill press equipped with a 0.3 mm-drill bit while utilizing a specially designed holding jig. The blue PC fibers were inserted into the drilled holes and trimmed to length as illustrated in Scheme 1. To eliminate air gaps between the inserted blue fibers and PC filaments, the inserted fiber was lightly heated with hot air. This heat allowed the melt spun fiber to