Print fidelity evaluation of PVA hydrogel using computational fluid dynamics for extrusion dependent 3D printing

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Abstract. This study presents a practical method of print fidelity evaluation for an extrusion-dependent technique of 3D printing. Simulation through computational fluid dynamics (CFD) tool has been used for evaluating the fidelity of printing. The polyvinyl alcohol (PVA) based hydrogel was prepared with deionized (DI) water and PVA powder using a magnetic stirrer at 90 °C for 3D printing. Rheological tests were carried out for checking the viscosity at various shear rates. CFD simulation was done by employing the Bird-Carreau model using rheological values. Velocity, pressure, shearing rate, and viscosity distributions through nozzle were obtained. From the shear rate and viscosity results, the increase in shear rate and decrease in gel's viscosity for both the nozzles prove that the material can be extruded. It was seen that the nozzle with a diameter of 0.51mm shows better results than the 0.41mm diameter, which was concluded from the values of maximum shear rate at the edges of the nozzles. The maximum shear rate value has reached a maximum of up to 326.5102 s⁻¹ whereas for a 0.41 mm diameter nozzle, it is 623.8037 s⁻¹ increasing the chances of developing wavy edges in a 0.41mm diameter nozzle than a 0.51mm diameter nozzle concluding that the nozzle with 0.51mm diameter gives far better results than the 0.41mm diameter nozzle.

1. Introduction

3D printing involves the deposition of material in the form of layers under computer control and its solidification to generate a 3D functional object. This is an addition process and not the same as the traditional removal method of object formation. Therefore, it is also termed as Additive Manufacturing (AM) [1]. Many 3D printing processes like extrusion, light-based vat polymerization, ink-jetting, and laser-based powder bed fusion are available, each having its pros and cons. Out of these technologies, this study is based on direct-write extrusion-dependent 3D printing (ED3DP), in which a paste/gel-like material can be printed utilizing a syringe-like extruder without the involvement of any heating or fusion processes [2]. The ability for a gel/paste to get 3D printed depends on how easily the material (gel/paste) can be extruded through the micron-sized nozzle and how strongly it can bond and solidify [3]. The extruder design and the extrusion of material through the nozzle depend on the complex flow behavior of materials in the extruder during the printing given by its rheological properties.

ED3DP technique has emerged as the competitive alternative in the fields of defense [4], biomedical [5], aerospace [6], food [2], and electronics [7]. Various sectors require different types of materials to be 3D printed as required while using suitable extruders. The extruder designed or chosen for printing should...
bear the behavior of the material in the extruder. The selected material for 3D printing should have fundamental capabilities like pressure-driven extrusion at a required flow rate; post extrusion shape-holding capability; the extruded structure must span a given gap and function as a mechanically strong substrate; dimensional stability while curing [8]. The wide use of ED3DP in various sectors makes studying the printability of material and the system for achieving fine functional objects with less dimensional inaccuracy. Analysis of the flow through the micron-sized nozzles of the technique could help in predicting the print fidelity well.

Prediction of print fidelity plays a vital role in the ED3DP process for achieving a structure-dependent functionality in the field of food, medical, and electronics. Various researchers have adopted different ways of fidelity prediction for accomplishing their requirements. For predicting the print resolution of extrusion-dependent bioprinting, a simple yet precise mathematical model has been developed. The model generated was for understanding the domination of various printing parameters like gauge pressure, printing speed, and nozzle-diameter on the line width while printing a continuous strand of Pluronic F127 hydrogel. The experimental results agreed with the model barring a few exceptions [9]. Apparently, it is found that maximum researchers have performed prediction through experimentation only. For this type of prediction, keeping some parameters constant with varying others is widely accepted. Carbon nanotubes (CNT) and Nafion-based inks' print fidelity was predicted by keeping nozzle diameter (0.29 mm) and printing speed (12 m/s) constant with varying pressure and Nafion concentration [10]. CNT/Polydimethylsiloxane composite ink's print stability has been improved by varying CNT loadings, pressure, and gap span of printed gap-spanning model through several printing trials [11]. However, researchers have adopted software CFD simulation tool also for predicting the print fidelity by analyzing the flow behavior of pastes/gels through the printing nozzle. Five different food grains: mung bean, black rice, buckwheat, job's tear seeds, and brown rice-based pastes' extrusion analysis was made on the nozzle by simulation. The Bird-Carreau model's fitted coefficients were determined using data from the rheological tests, and CFD simulations for viscosity, pressure, and shear rates were carried out to examine the extrusion behavior. It was observed that the simulated pressure values at the central axis being lesser than the maximum applicable pressure by the extruder is the fundamental factor influencing the print fidelity of materials [12].

Conclusively, the print fidelity of a material is determined in three different ways; experimental, mathematical, or analytical, and simulation [9-12]. In the experimental method, a number of printings using different nozzles are carried out for assessing the print quality with the help of specific measuring instruments and visual observations. The second one is the method in which an analytical model is created that describes the output dimensions with the printing parameters, which gives an idea about the print conformity of the material. In the simulation, a mathematical model describing the material's flow behavior is incorporated into the software to produce virtual mapping, showing the distribution of the various properties (viscosity, velocity, pressure, and shear rate), which helps predict the print fidelity [13].

Out of these methods, simulation is the most accurate and convenient method [12]. It helps to analyze the print fidelity of the material without performing the 3D printing process while covering a more extensive range of values of rheological properties compared to rheological tests, which cover a limited range of values. It takes less time and effort than a mathematical model while painting a more comprehensive picture without wasting material [14].

Recent progress on ED3DP has focused on the production of tunable strength synthetic hydrogels with customized structures, recoverable and self-healing properties [15]. Poly (ethylene glycol) [16] and polycaprolactone [17], polyvinyl alcohol (PVA) [15,17], and pluronic F127 [9] are some polymers widely used in the form of hydrogels. Among them, PVA hydrogels have gained more popularity because of their biocompatibility, non-toxicity, swelling stability, low cost, thermal stability, high chemical stability, and mechanical properties [15,18]. According to its potential and usage in various industries like; medical, textile, paper, and food packaging, the extrusion behavior needs to be optimized for flawless deposition over substrates which is a prime requirement for 3D structuring through ED3DP. While research regarding the perfect print material is presently quite prevalent in the biomedical field, the research about
the optimum nozzle is relatively less explored. This serves as an inspiration to carry out suitable analysis to establish the effect of changing print parameters and nozzle geometries on print output quality.

This study determines an accurate method of print fidelity evaluation for ED3DP. For this, the analysis of the flow of visco-elastic PVA hydrogel in the syringe-nozzle system is done. The hydrogel of PVA is prepared by mixing PVA into deionized (DI) water. CFD simulation was carried out using the Bird-Carreau model for the static rheological properties of the printing material. Pressure, velocity, shear rate, and viscosity analysis of syringe-nozzle system are made for two different nozzle-diameters.

2. Materials and Methodology

2.1. Materials and hydrogel preparation

Polyvinyl alcohol (PVA) (Molecular Weight= 88000-97000 g·mol$^{-1}$, 98-99% hydrolyzed) is bought from Alfa Aesar (Thermo Fisher Scientific India Private Limited) in the form of tiny pellets. Deionized (DI) water was prepared in the laboratory.

To prepare a 24 wt% PVA hydrogel, 12 grams of PVA were combined with 50 ml DI water at 90 degrees Celsius with vigorous stirring. Stirring was done using a magnetic stirrer (Tarsons digital spinot, Tarsons Products Pvt. Ltd., Kolkata, India) and magnetic bead at 1200 rpm for 10 hours (figure 1 (a)). The prepared sample is allowed to cool to room temperature (25°C).

2.2. Rheological property determination

Hydrogel’s rheological properties were calculated using Bohlin Gemini 2 (Malvern Instruments, UK), an advanced air-bearing rheometer. An oscillation test and a shear test were conducted. Oscillation tests give results about the dynamic rheological properties, while the shear test gives results about the static rheological properties. The change in viscosity with the shear rate was noted down.

The Bird-Carreau model was used for describing the alter in viscosity of the fluid with shearing rate. It was fit with the results obtained experimentally.

Bird-Carreau model equation:

$$
\eta = \eta_\infty + (\eta_\infty - \eta_0)(1 + (\lambda \gamma)^n)^{1/2}
$$

Here $\eta$ = viscosity of the fluid (Pa·s); $\eta_\infty$ = viscosity of the fluid at infinite shearing rate (Pa·s); $\gamma$ = shearing rate (s$^{-1}$); $\lambda$ = relaxation time(s); $\eta_0$ = viscosity of the fluid at zero shearing rate (Pa·s); $n$ = rheological index [19].
3. Analytical Model
ED3DP CFD simulation was performed with the Mechanical Finite Element Analysis Software version-18.1 (ANSYS Inc, Canonsburg, PA, USA) in order to know the variable properties along the process.

3.1. Physical CAD model
The computational zone was restricted to the nozzle of the syringe-nozzle system. By this, the computational time for simulation was reduced. The simulated geometry was constructed in Design Modeler Geometry of Ansys 18.1 Version. The model has 0.41 mm and 0.51 mm nozzle diameters and 12.7 mm nozzle length for two different sized syringe-nozzle setups (figure 1(b,c)).

3.1.1. Model equations
The PVA hydrogel was laminar and presumed to be an incompressible fluid with a singlephase. ANSYS FLUENT solves the following momentum and continuity equations for an incompressible fluid undertaking isothermal flow via extrusion.

\[ \frac{\partial (\rho \bar{v})}{\partial t} + \nabla \cdot (\rho \bar{v} \bar{v}) = -\nabla P + \nabla \cdot (\bar{\tau}) \]  

\[ \nabla \cdot \bar{v} = 0 \]

Whereas \( \bar{\tau} = \mu [(\nabla \bar{v} + (\nabla \bar{v})^T) - \frac{2}{3} \bar{v} \cdot \bar{v} I] \)

Where, \( P \) is the static pressure (Pa), \( \bar{v} \) is the stress tensor (Pa), \( \rho \) is density (kg/m\(^3\)), and \( I \) is the unit tensor [20].

For solving the governing equations, the following conditions were used:
- There is no slip between the syringe wall and fluid, i.e., velocity near the wall is zero.
- Fluid's inlet velocity is presumed to be 5 x 10\(^{-5}\) m/s
- Outlet conditions gauge pressure is zero.
- Fluid has constant density, i.e., incompressible.
- Pressure is constant at a particular horizontal plane.
- There is viscosity change with respect to shear rate. For this condition, values mentioned in table 1 (\( \eta_0, \eta_\infty, \lambda \) and \( n \)) have been used.

4. Results and Discussions

4.1. Hydrgel's Rheological properties:
The rheological properties of a material have been shown to be a significant predictor of its print fidelity in various studies. The data obtained from the experiment was plotted on the graph, as shown in figure 2(a). A shear-thinning behavior, i.e., when the shear rate is increased, the viscosity decreases, can be observed from the graph of viscosity vs. shear rate, which is a prime requirement for the ED3DP process.

By fitting the Bird-Carreau curve (figure 2(a)) in the data obtained, the coefficients of the Bird-Carreau equation were found by using ORIGIN software are listed in table 1.

Table 1. Co-efficient fitted in Bird- Carreau model

| \( \eta_0 \) (Pa.s) | \( \eta_\infty \) (Pa.s) | \( \lambda \) (s) | \( n \) | \( R^2 \) | \( \rho \) (kg/m\(^3\)) |
|-----------------|-----------------|-----------|-----|--------|----------------|
| 2157.695        | 36.473          | 48.1997   | 0.1407 | 0.992  | 1057.098       |

The coefficient of determination \( (R^2) \) of fitted values was found to be 0.992, which indicates that the PVA Hydrogel viscosity change is accurately described by the Bird-Carreau model.
4.2. CFD Simulation:

CFD simulation was carried out in software ANSYS R18.1 version for nozzles with two different diameters, 0.41mm and 0.51mm. The syringe-nozzle system was designed in the ANSYS drawing module.

Mesh independence tests were carried out to ensure no effect on simulated variables with mesh density. The design consists of 16,150 hexahedral mesh elements (figure 2(b)). The results are given in figures 3, 4, 5 and 6 for 0.41mm diameter nozzle followed by 0.51mm nozzle.

4.2.1. Simulated velocity distribution:

In the simulation (figure 3), it can be seen that the velocity gradually increases from the top to bottom towards the nozzle outlet. It can also be observed that velocity increases while moving from wall to center at the nozzle section.

Figure 2. (a) Viscosity vs. Shear rate plot at different shearing rates with Carreau fit. (b) Mesh geometry of syringe-nozzle section.

Figure 3. Velocity distribution in (a) 0.41mm and (b) 0.51 mm diameter nozzle.
Figure 4. Pressure distribution in (a) 0.41 mm and (b) 0.51 mm diameter nozzle.

Figure 5. Viscosity distribution in (a) 0.41 mm and (b) 0.51 mm diameter nozzle.

Figure 6. Shearing rate distribution in (a) 0.41 mm and (b) 0.51 mm diameter nozzle.
4.2.2. Simulated pressure distribution:
The syringe-nozzle system has given a pertinent level of applied pressure to assure a material. The pressure distribution of the material across the syringe is shown in figure 4. It is observed that with a decrease in pressure while approaching the nozzle, a smooth extrusion of the material occurs. As the material follows the continuity equation, pressure will genuinely decrease with the increase in velocity through the nozzle.

4.2.3. Simulated viscosity distribution:
The distributions of viscosities for both needles were similar, as shown in figure 5. The syringe had higher viscosities, particularly in the corners and top center, where shear rate values were lowest. As the hydrogel is shear thinning, shear rate increasing throughout the nozzle, viscosity decreases through the syringe, ensuring smooth flow of fluid layers one over another.

4.2.4. Simulated local shearing rate distribution:
According to the boundary conditions, the velocity at the walls is zero. Due to this, a different shear rate for the material occurs and increases with the cross-sectional area. It can be observed more shear at the nozzle tip, which ensures more deformation and suitable material extrusion, and also the shear is maximum at the walls and minimum at the center (figure 6).
From the distributions obtained through simulation, it has been observed that velocity, pressure, strain rate, and viscosity follow a similar trend in their change for both the nozzles.

5. Conclusions and outlook
Based on the observations following conclusions can be made:
- CFD simulation of the viscoelastic material employing the Bird-Carreau model shows the material's behavior across the syringe-nozzle system, which can estimate the print fidelity of the material.
- The increasing shear and decreasing viscosity through the nozzle show that the material is extrudable. Also, it was observed that velocity, pressure, shear rate, and viscosity show similar changes for different diameters of nozzles.
- It can be seen that the values of maximum shear rate at the edges of the 0.51mm nozzle have reached a maximum of up to 326.5102 s⁻¹ whereas for a 0.41 mm diameter nozzle, the maximum shear rate value is 623.8037 s⁻¹. So, there are more chances of developing wavy edges in a 0.41mm diameter nozzle than a 0.51mm diameter nozzle. Hence among the two nozzles, 0.51mm diameter gives far better results than the 0.41mm.

In addition, this study also plays a pivotal role in helping to identify the various irregularities in the understandings regarding this field that may exist at the moment. This study also provides an impetus for further exploration down this path to develop optimum parameters for achieving near-flawless ED3DP in the future. Further research should establish a standard procedure for finding out the minimum possible diameter of the nozzle used for the extrusion of a particular material.

6. References.
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