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Research Paper

A Fractal Rheological Model for SiC Paste using a Fractal Derivative

Yuting ZUO[1,2], Hongjun LIU [1,2]

[1] School of Materials Science and Engineering, Lanzhou University of Technology, Lanzhou, 730050, China, Email: 2452767789@qq.com

[2] State Key Laboratory of Advanced Processing and Recycling of Non-ferrous Metals, Lanzhou University of Technology, Lanzhou 730050, China, Email: 28071080@qq.com

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Corresponding author: Yuting ZUO (2452767789@qq.com)

Abstract. The rheological property plays an important role in a free-form extrusion 3D printing process, no rheological model was available in open literature that could effectively take into account effects of both the non-Newtonian viscosity and the concentration of nano/micro particles in a paste. Here a fractal law for non-Newtonian fluids is suggested using a fractal derivative, the law can predict correctly the boundary effect of a viscous flow, and can model effectively the nonlinear velocity distribution across the section. A systematic derivation of a fractal rheological model is suggested using the basic laws in the fluid mechanics, which can provide a deep insight into the two-scale fractal interpretation of non-Newtonian fluids. An experiment was carefully designed to verify the model and to elucidate the relationship between the shear rate and viscosity of the SiC paste. 15wt.%, 25wt.%, 35wt.% and 45wt.% SiC pastes were prepared by using mixing, stirring and ball milling processes. The rheology of the paste can be controlled primarily through the SiC concentration, which affects the fractal order. The fractal model sheds a bright light on a simple but accurate approach to non-Newtonian fluids.

Keywords: Free-form extrusion, 3D printing, SiC paste, rheological properties, fractal derivative, two-scale fractal calculus

1. Introduction

Rheological fluids arise everywhere from a capillary motion to the Earth's inner motion [1-4], and the rheology can be effectively controlled by the concentration of the added particles. The rheological property plays also an important role in 3D printing technology. As the latest development of the free-form extrusion 3D printing technology [5-9], more and more attention has been paid to prepare for an optimal print paste, where the rheological property can be effectively controlled because it plays an important role not only in the printing process but also the product's properties. So far there was not an effective rheological model that could take into account simultaneously effects of the non-Newtonian viscosity and the concentration of nano/micro particles in a paste.

Rheology also affects natural spinning process [10], artificial spinning processes [11,12], polymer filling process [13], and anomalous property [14]. Though there are many rheological models using differential derivatives for various non-Newtonian fluids [15], and many experimental manners to adjust the rheological property of the print paste by adding some nano/ micro particles [16-18], there is not a simple theoretical model in open literature to predict exactly the rheological property by differential models.

Due to the wide applications of fractal calculus [19-22] to various complex phenomena, many researchers found that a fractal model can describe many phenomena which is far beyond the traditional models. This paper is to apply the fractal calculus to establish a fractal rheological model.

Fractal calculus can explain many phenomena beyond the differential models, for examples, Fangzhu's water collection's property [23,24,25], traveling waves in an unsmooth boundary [26-30], natural fiber's biomechanism [31-33], fractal ion release [34-36], snow's thermal insulation property [37], fractal adsorption kinetics [38], fractal oscillators [39], and fractal M/NEMS systems [40,41].

2. Fractal Rheological Model

We begin with Newtonian fluid as illustrated in Fig.1(a), the viscous force can be written in the form

$$\tau=\eta\frac{du}{dx} \tag{1}$$

where τ is the viscous force, η is the viscous coefficient, du/dx is the velocity gradient. The Newtonian fluid assumes that the viscous force arises in intermediate layers interaction. For non-Newtonian fluids as shown in Fig.1(b), Equation (1) has to be modified, for a one-dimensional flow, we have the following modifications:

$$\tau = \eta \frac{du}{dx} + \varepsilon \left(\frac{du}{dx}\right)^3 \tag{2}$$

or

$$\tau + \varepsilon \tau^3 = \eta \frac{du}{dx} \tag{3}$$

where ε is a constant.

The nonlinear term involving in Eq.(2) or Eq.(3) makes the theoretical analysis much difficult and complex. Though the models of Eqs.(2) and (3) can model quantitatively many non-Newtonian fluids, the nonlinear terms have no physical meaning. Fig.1(a) shows a typical Newtonian fluid with a linear velocity distribution, and a non-Newtonian fluid has a nonlinear velocity distribution as shown in Fig.1(b). Using an infinitesimal element in mathematics, we can convert a non-Newtonian fluid into an infinite Newtonian fluid with different viscosities, however the infinite treatment is unserviceable in practical applications, so we cannot take the virtue of the simple Newtonian fluid. To overcome the infinite treatment, a fractal model must be most welcome.

An infinitesimal element leads to a coastline-like velocity distribution, and Mandelbrot's basic concept of his famous fractal theory [42] can be used. That means the fractal dimension of the velocity curve is the main factor affecting the viscous force. Mandelbrot concluded that the length of the coast depends upon the scale used, and it tends to infinity when the scale tends to zero. This can be understood by two animals' walking along the coast, one is a dinosaur with a large step, the other is a cat with a small step. The two animals are walking along the coast with a same velocity, and they will arrive at their destination at different time. That means the motion property depends upon not only the curve geometry of the coast, but also the step. If the step is Δx, then all motion properties scale with $(\Delta x)^\alpha$, where α is the fractal dimension of the coastline.

Similarly, the length of the velocity curve depends on the measured scale as shown in Fig.2. When we use a large scale of AB, an approximate Newtonian model is obtained, different scales (e.g. AB or AC) results in different length of the velocity curve, so the viscous force can be written as

$$\tau = \eta \frac{du}{dx^\alpha} \tag{4}$$

where α is the two-scale fractal dimension, when $\alpha = 1$, we have the Newtonian model, du/dx^α is the fractal derivative defined as [10]

$$\frac{du}{dx^\alpha}(x_0) = \Gamma(1+\alpha) \lim_{\substack{x - x_0 \to \Delta x \\ \Delta x \neq 0}} \frac{u - u_0}{(x - x_0)^\alpha} \tag{5}$$

Eq.(4) can be explained using the two scale mathematics [19-22]. For a large scale, we have an approximate Newtonian model for α =1. The non-Newtonian paste in our study arises in the additive of SiC particles in the fluid, and the Newtonian model cannot take into account the particle size and concentration of the SiC particles. We use the average distance among particles as a smaller scale to model the particles' effect.

Eq.(4) can also describe the boundary effect of a viscous flow, and can model effectively the nonlinear velocity distribution across the section. Taking into account the boundary effect, the velocity distribution can be expressed as $u = kx^\alpha$, where k is a constant.

The rheological property focuses on the relationship between shear stress and viscosity. When a fluid is subject to a shear rate gradient, $\Delta\gamma$, the viscosity will be changed by $\Delta\eta$, see Fig.1. We call $\Delta\eta / \Delta\gamma$ as the rheological gradient. Similar to Eq.(1), the viscous force scales with the rheological gradient in the form

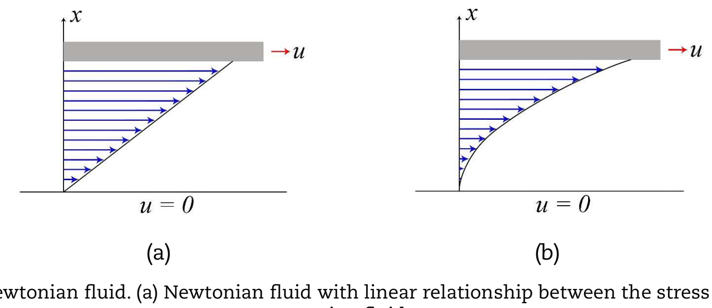

Fig. 1. Newtonian fluid and Non-Newtonian fluid. (a) Newtonian fluid with linear relationship between the stress and strain rate curve; (b) Non-Newtonian fluid.

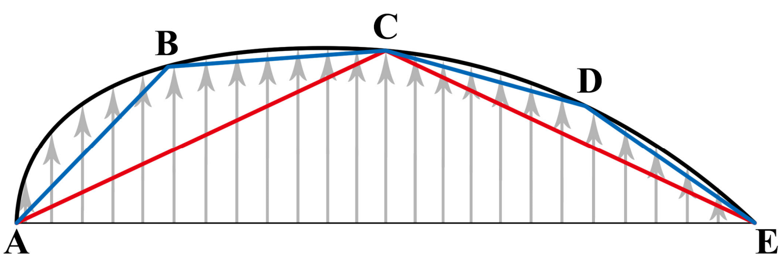

Fig. 2. A coastline-like velocity distribution of a non-Newtonian fluid. It can be decomposed into an infinite Newtonian fluids, and the viscous property depends upon the fractal dimension of the velocity curve (ABCDE) and the measured scale (AE, or AC, or AB).

$$\tau \propto -\frac{d\eta}{d\gamma} \tag{6}$$

Equation (6) implies a higher shear rate leads to a lower viscous force. This phenomenon can be explained as follows: a higher shear rate results in a more order intermediate layers tangential motion.

According to Eq.(1), we have

$$\tau \propto \eta \tag{7}$$

In view of Eqs. (6) and (7), we

$$\frac{d\eta}{d\gamma}+\lambda\eta=0 \qquad \eta(0)=\eta_0 \tag{8}$$

where λ is a constant. Equation (8) has the following solution

$$\eta=\eta_0\exp(-\lambda\gamma) \tag{9}$$

Equation (9) implies that the viscosity decrease exponentially with the increase of γ. This is approximately true, but it cannot model the sudden change of the viscosity when γ tends to zero. For a high viscous fluid, the velocity distribution should follow a non-Newtonian case as illustrated in Fig.3(b). Under this case, Eq. (6) should be modified. Here a fractal modification is adopted

$$\tau \propto -\frac{d\eta}{d\gamma^{\alpha}} \tag{10}$$

where $d\eta / d\gamma^{\alpha}$ is the fractal derivative defined as

$$\frac{d\eta}{d\gamma^{\alpha}}(\eta_0)=\Gamma(1+\alpha)\lim_{\substack{\gamma-\gamma_0\to\Delta\gamma \\ \Delta\gamma\neq 0}}\frac{\eta-\eta_0}{(\gamma-\gamma_0)^{\alpha}} \tag{11}$$

The rheological property can be expressed by the following fractal differential model

$$\frac{d\eta}{d\gamma^{\alpha}}+\lambda\eta=0 \qquad \eta(0)=\eta_0 \tag{12}$$

The solution of Eq.(12) is

$$\eta=\eta_0\exp(-\lambda\gamma^{\alpha}) \tag{13}$$

3. Experimental design

In order to verify the fractal rheological model, an experiment was carefully designed.

3.1 Raw materials

For studying the rheological properties of SiC slurry, α-SiC powder (D50=1.5μm) with a purity above 99% was used in this study, and it was purchased from Zhengzhou Sanmo Superhard Material Co., Ltd., China.

Carrageen (0.7% in water solution) was used as thickener, and it was purchased from Hainan Qionghai Changqing Agar Factory, China. Tetramethylammonium Hydroxide (TMAH), 25% in water solution, was used as dispersant, and it was purchased from Shanghai Macklin Biochemical Co., Ltd., China. Polyethylene Glycol 1500 (PEG 1500) was used as plasticizer, and it was purchased from Shanghai Macklin Biochemical Co., Ltd., China. Glycerol was used as lubricant, and it was purchased from Tianjin Zhiyuan Chemical Reagent Co. Ltd., China. Deionized water was used as solvent, and it was prepared by distillation.

3.2 SiC slurry

The preparation of SiC slurry was described as follows:

A mixed solution was prepared by using 7wt.% TMAH (25% in water solution), 2wt.% PEG 1500, 8wt.% Glycerol, 16wt.% Carrageen (0.7% in water solution) , and 83wt % deionized water, which was then stirred magnetically for 30 min. SiC powders were then added to the solution to the samples with concentrations of 15, 25, 35 and 45wt.%, respectively. Each sample was ball-milled in vertical planetary mill (XQM-04, Changsha Tencan Powder Technology Co. Ltd., China) with pure ZrO_2 balls for 2 hours, so that a uniformly dispersed SiC slurry was obtained. Additionally, a sample without SiC additives was also prepared for comparison.

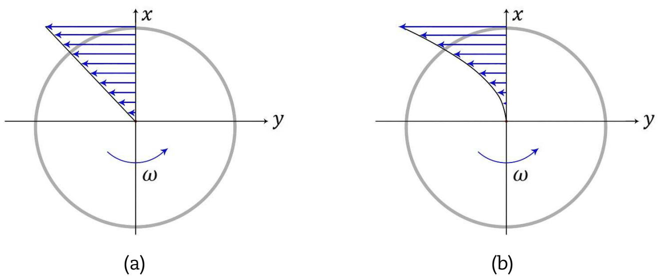

Fig. 3. The rheological property under shearing motion.

Table 1. Samples' viscosity at different shear rate.

	SiC = 0 wt.%	SiC = 15 wt.%	SiC = 25 wt.%	SiC = 35 wt.%	SiC = 45 wt.%
1.5 r/min	127	136	146	156	167
3 r/min	59.2	78	99	123	150
6 r/min	25	45	66	98	136
12 r/min	7	18	38	72	120
30 r/min	0	2	12	41	97

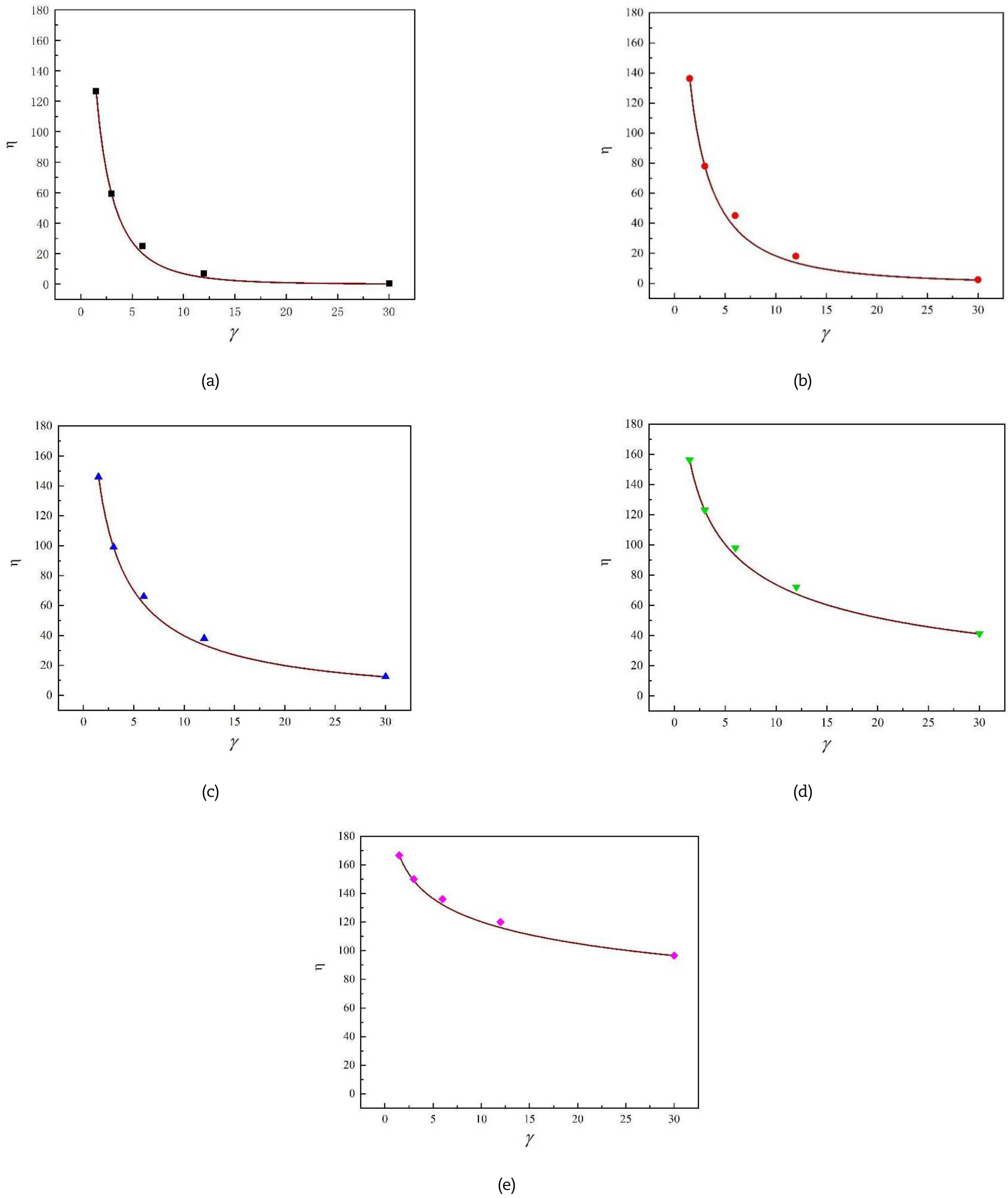

Fig. 4. Rheological properties of the paste with and without SiC powers. (a) without SiC, λ=1.5, α=0.5; (b) 15 wt.% SiC, λ=1.5, α=0.4; (c) 25 wt.% SiC, λ=1.5, α=0.3; (d) 35 wt.% SiC, λ=1.5, α=0.2; (e) 45 wt.% SiC, λ=1.5, α=0.1.

3.3 Rheological properties

The rheological properties of SiC slurry were measured by a rotary viscometer (NDJ-8S, Shanghai Fangrui Instrument Co. Ltd., China). Rotational speed was set as 1.5, 3, 6, 12, 30 r/min, respectively. Five viscosity values under each speed were recorded, and the average value was used for analysis. The test temperature was 25°C, the results were listed in Table 1.

4. Results and Analysis

According to the two-scale fractal theory, all properties in a fractal space depends upon its fractal dimension and its measured scale

$$L \propto (\Delta x)^{\alpha} \tag{14}$$

where L is a property of the studied problem, e.g. the viscous force, Δx is the scale, α is the fractal dimension. Different scales leads to different physical laws. For example, on a micro scale, water can be considered as a continuum, and all laws in fluid mechanics can be applied. However, the continuum assumption becomes invalid for the diffusion of a red ink in water and a fractal model has to be adopted[42]. The Newtonian fluid assumes that the viscous force is a function of coordinates, i.e., $\tau=\tau(x)$, for non-Newtonian fluids, it should be modified as $\tau=\tau(x^{\alpha})$.

Our study focuses on the SiC paste's viscosity, the experiment shows that the rheology of the paste can be effectively controlled through the SiC concentration, which affects the fractal order. Figure 4 shows the relationship between shear rate and viscosity of SiC paste with different SiC concentrations. The viscosity of SiC paste decreases with increasing shear rate, which conforms to the fractal model

$$\eta = 795\exp(-1.5\gamma^{\alpha}) \tag{15}$$

where α=0.5, 0.4, 0.3, 0.2 and 0.1 for, respectively, SiC pastes with concentrations of 0, 15 wt.%, 25 wt.%, 35 wt.% and 45 wt.%. A good agreement between the theoretical prediction given by Eq.(15) and the experiment results shows the reliability of the fractal model. The fractal order, α, strongly depends upon the SiC concentration.

The fractal order can be effectively controlled by the SiC concentration, see Fig.5, a higher SIC concentration leads to a smaller valve of α.

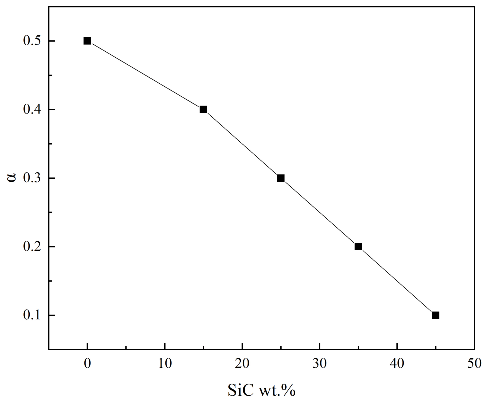

Fig. 5. Rheology can be controlled by the SiC concentration, which has an inverse relationship with the fractal order.

5. Discussion and Conclusion

This paper, for the first time ever, suggests a fractal rheological model for 3D print pastes, and it can be also used for other non-Newtonian fluids. The experimental results show a good agreement with the theoretical prediction, showing the model is reliable and applicable for practical applications. The fractal rheological model can explain well all rheological properties of the 3D print pastes, which cannot be modelled effectively by any a differential model.

Author Contributions

Yuting Zuo planned the scheme, conducted the experiments and developed the mathematical modeling; Hongjun Liu suggested the experiments. All authors discussed the results, reviewed, and approved the final version of the manuscript.

Conflict of Interest

The authors declared no potential conflicts of interest with respect to the research, authorship, and publication of this article.

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ORCID iD

Yuting ZUO https://orcid.org/0000-0001-5260-6926
Hongjun LIU https://orcid.org/0000-0001-6437-4587

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