

Fractional time-dependent apparent viscosity model for semisolid foodstuffs

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Abstract The difficulty in the description of thixotropic behaviors in semisolid foodstuffs is the time dependent nature of apparent viscosity under constant shear rate. In this study, we propose a novel theoretical model via fractional derivative to address the high demand by industries. The present model adopts the critical parameter of fractional derivative order α to describe the corresponding time-dependent thixotropic behavior. More interestingly, the parameter α provides a quantitative insight into discriminating foodstuffs. With the re-exploration of three groups of experimental data (tehineh, balangu, and natillas), the proposed methodology is validated in good applicability and efficiency. The results show that the present fractional apparent viscosity model performs successfully for tested foodstuffs in the shear rate range of 50–150 s⁻¹. The fractional order α decreases with the increase of temperature at low temperature, below 50 °C, but increases with growing shear rate. While the ideal initial viscosity k decreases with the increase of temperature, shear rate, and ingredient content. It is observed that the magnitude of α is capable of characterizing the thixotropy of semisolid foodstuffs.

Keywords Fractional calculus · Semisolid foodstuffs · Apparent viscosity · Thixotropy

1 Introduction

Thixotropy is one of the most classic documented rheological phenomena but remains a challenging research issue in colloid science (Mewis and Wagner 2009). This phenomenon is widely observed in industrial and natural systems, such as crude oil (Petrellis and Flumerfelt 1973; Cheng et al. 1999), mining slurries (Kretser and Boger 2001), decontamination gel (Raber and Mcguire 2002), printing inks (Liang et al. 1996), food products (Coussot and Gaulard 2005), etc. But there still remains confusion concerning its definition, in which

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time-dependence or “memory” of apparent viscosity description with reasonable physical interpretation is the fundamental issue. How to handle this issue directly affects the rationality and applicability of constitutive equations. The traditional apparent viscosity models of time-invariant non-Newtonian fluid, such as the Cross model, the Sisko model (Fischer et al. 2009), the Casson model (Rao 2010) and the F-A model (Falguera and Ibarz 2010), remain being widely used nowadays. Nevertheless, the most of existing models ignore the vital role of “memory”, which causes dilemma when applied.

Semisolid food as typical thixotropic material has received considerable attention in the past decades. Through the study of its thixotropic behavior, the quality of food can be controlled and classified, the related processing equipment can be optimized, so that stirring, cooling, and storage are more scientific and cost-effective. The thixotropic characteristic is an essential aspect for consumers to accept and appreciate the products (oral perception, digestion, and well-being) and improve the quality of semisolid food (stability, taste, perception, etc.). The classical models, however, cannot well describe the thixotropic phenomenon of popular and new foodstuffs, such as compound foodstuffs and three-dimensional (3D) foodstuffs made from 3D food printer. Thus, an effective mechanics model for this problem is highly demanded by the food industries.

As for semi-solid foodstuffs, a widely used time-dependent apparent viscosity model named as structural kinetic model is given by (Abu-Jdayil 2003; Razavi and Karazhiyan 2009; Nguyen et al. 1998)

$$\left[\frac{(\eta - \eta_\infty)}{(\eta_0 - \eta_\infty)} \right]^{1-n} = (n - 1)kt + 1, \quad (1)$$

where η_0 , η_∞ , k , and n are the initial apparent viscosity, the equilibrium apparent viscosity, the rate constant, and the order of the structure breakdown reaction, respectively. But the model (1) is complex and valid only under constant shear rate, because both η_0 and η_∞ are functions of a single variable of applied shear rate. This implies that the applicable scope of structural kinetic model is not applicable to time-variant cases.

Fractional calculus is a promising tool for modeling the history-dependent physical processes or memory phenomena (Nils et al. 2008; Rossikhin and Shitikova 2010; Stiassnie 1979), especially in characterizing viscoelastic behaviors and non-Newtonian fluid properties of time-dependent materials. Compared with the traditional modeling approaches, fractional derivative models usually require fewer parameters. Such models have been applied in various fields, such as viscoelastic materials (El-Shahed 2006; Shah and Qi 2010; Du et al. 2013; Fan et al. 2015), non-Newtonian fluid (Mahmood et al. 2009), anomalous diffusion in complex porous media (Sun et al. 2009; Yu et al. 2015), climate prediction (Yuan et al. 2014), chemotherapy on cancer cells (Namazi et al. 2015), signal processing (Jin et al. 2015), etc. However, to the best of authors’ knowledge, little effort has been done in using fractional derivative constitutive model to model the apparent viscosity of thixotropic materials.

This paper develops a fractional derivative model to characterize the apparent viscosity of thixotropic materials, in which the fractional derivative term is designed to describe its historical dependency. The efficiency and applicability of the proposed fractional apparent viscosity model (FAVM) is validated by re-exploration of three groups of experiments. This paper also offers a discussion on the determination of the model parameter and the relationship between parameters and thixotropic material properties.

2 Methods

2.1 Fractional calculus and its property

To introduce the operator of fractional derivative, here we start from the Cauchy formula for the n -fold integral of function $f(t)$ (Podlubny 1998)

$$f^{(-n)}(t) = \frac{1}{\Gamma(n)} \int_0^t (t - \tau)^{n-1} f(\tau) d\tau \quad (t > 0, \alpha > 0). \tag{2}$$

By assuming that $\alpha = n$ is a non-integer number, we get the definition of fractional integral

$$I^\alpha f(t) = \frac{1}{\Gamma(\alpha)} \int_0^t (t - \tau)^{\alpha-1} f(\tau) d\tau \quad (t > 0, \alpha > 0), \tag{3}$$

where Γ is the gamma function. The definition of fractional derivative can be obtained by conducting differentiation on fractional integral (3). The frequently used Riemann–Liouville fractional derivative is expressed as (Yin et al. 2013)

$$D^\alpha f(t) = \begin{cases} \frac{d^m}{dt^m} \left[\frac{1}{\Gamma(m-\alpha)} \int_0^t \frac{f(\tau)}{(t-\tau)^{\alpha+1-m}} dt \right] & (m - 1 < \alpha < m, m \in N^+) \\ \frac{d^m}{dt^m} f(t) & (\alpha = m, m \in N^+). \end{cases} \tag{4}$$

Notably, the Riemann–Liouville definition has mathematical rigor compared with the other definitions. Also, The choice of this definition facilitates the following modeling process due to its convenience and clarity.

From Eqs. (3) and (4), we can obtain the following basic property:

$$\begin{cases} I^\alpha t^\gamma = \frac{\Gamma(\gamma+1)}{\Gamma(\gamma+1+\alpha)} t^{\gamma+\alpha} \\ D^\alpha t^\gamma = \frac{\Gamma(\gamma+1)}{\Gamma(\gamma+1-\alpha)} t^{\gamma-\alpha} \end{cases} \quad (\alpha > 0, \gamma > -1, t > 0). \tag{5}$$

The α th derivative of a given function $f(t) = C \cdot H(t)$ can be expressed as (Podlubny 1998)

$$D^\alpha C = \frac{C t^{-\alpha}}{\Gamma(1-\alpha)} \quad (\alpha > 0, t > 0). \tag{6}$$

Here, $H(t)$ denotes Heaviside function, also known as unit step function, which can be expressed as follows:

$$H(t) = \begin{cases} 0, & t < 0 \\ 1, & t \geq 0. \end{cases} \tag{7}$$

2.2 Fractional apparent viscosity model for semisolid foodstuffs

Apparent viscosity is an important parameter for non-Newtonian fluid (Mewis and Wagner 2009), numerous experiments have shown that the apparent viscosity under a constant shear rate usually decreases with time (Mcnaught and Wilkinson 1997).

Meanwhile, previous studies have also indicated the apparent viscosity of time-variant non-Newtonian fluid depends not only on shear rate, but also on time of shearing. To describe this property, Cheng (1973) proposed the thixotropic material constitutive equations

$$\tau = \eta(\lambda, \dot{\gamma})\dot{\gamma}, \tag{8}$$

$$\frac{d\lambda}{dt} = g(\lambda, \dot{\gamma}), \quad (9)$$

where η is the apparent viscosity, λ is a structural parameter, $\dot{\gamma}$ is the shear rate, τ is the shear stress, and g denotes the derivative of λ which is a function of λ and $\dot{\gamma}$. Above constitutive equations indicate a relationship between shear rate, shearing time, and the apparent viscosity. Nevertheless, the constitutive equations only contain relevant parameters. Especially involving the apparent viscosity equation (8), only influence factors are given, specific formulas are rare in the literature for semisolid foodstuffs. Also, it is hard to acquire explicitly the structure parameter λ from the experimental measurements, which causes great difficulty in real-world application. In Yang et al. (2017), a system of equations describing time-variant non-Newtonian flow are proposed (including viscosity model (9)). However, due to the complexity of semisolid foodstuffs, the proposed model cannot well address the gap between the model parameters and underlying mechanism in microscopic scale, which will be analyzed in Sect. 4.

In order to enable apparent viscosity model to involve the time-varying factor, based on Eq. (8), we define the fractional derivative apparent viscosity model as

$$\eta(t) = k_1 \frac{d^{1-\alpha} \dot{\gamma}}{dt^{1-\alpha}}, \quad (10)$$

in which k_1 is the initial viscosity, $\dot{\gamma}$ is the shear rate, and α is the fractional derivative order.

3 Results

3.1 Fractional apparent viscosity model (FAVM)

According to the general definition of thixotropy (Abu-Jdayil 2003), the material is sheared at constant shear rate, resulting in the change of the structure with time. The semisolid food containing colloidal sized particles such as solids or immiscible liquids exhibit thixotropic behavior. Thus, we pay particular attention to one condition of shear rate with time, $\dot{\gamma} = C \cdot H(t)$, where C is a constant and $H(t)$ the Heaviside function.

Assuming $k = k_1 \cdot C$, based on Eqs. (6) and (10), the relationship between apparent viscosity and time can be expressed as

$$\eta(t) = k \cdot \frac{t^{\alpha-1}}{\Gamma(\alpha)}, \quad (11)$$

where k denotes the ideal initial viscosity. It is often considered that α varies between 0 and 1 (Yin et al. 2012). Hereby we also provisionally suppose that α is greater than 0 and less than 1. If $\alpha \rightarrow 0$, then $\eta(t) \rightarrow 0$, namely no memory is present, it means that an ideal fluid has no viscosity. If $\alpha \rightarrow 1$, then $\eta(t) \rightarrow k$ (nothing forgotten), apparent viscosity can be seen as an ideal initial viscosity which always keeps full memory and does not vary over time. Thus, we define the fractional order α as the index of historical memory.

The parameters k and α can be obtained through the experimental data fitting. Meanwhile, the ideal initial viscosity parameter k is determined by the initial temperature, pressure, shear stress, and physical ingredients. The fractional derivative order α is largely due to complex internal interactions, such as weak attractive forces between the particles, the strength of the particulate network, and the state of flocs.

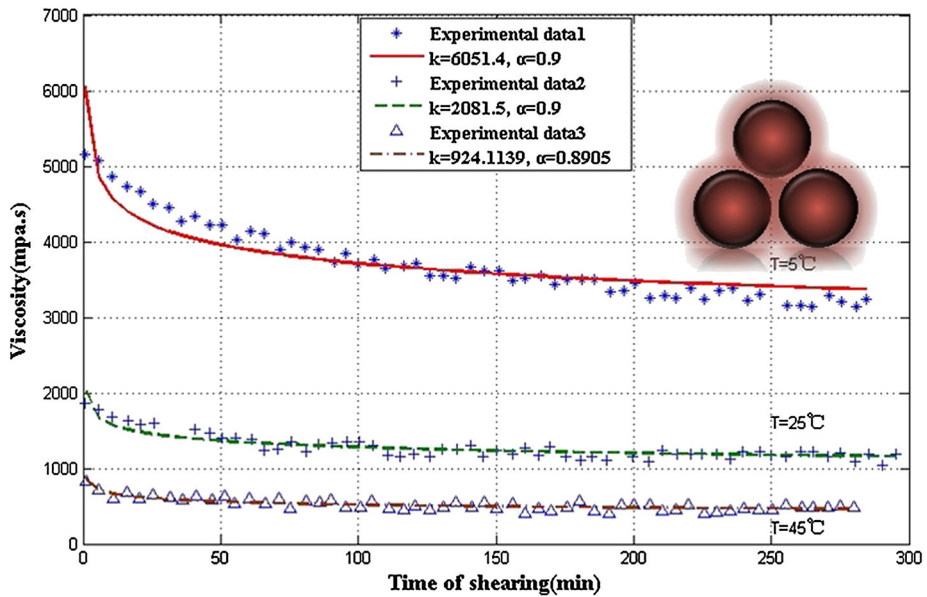


Fig. 1 Effect of temperature on the thixotropic behavior of tehineh at constant shear rate = 102 s^{-1} (the experimental data is quoted from Abu-Jdayil 2003). The red, green, and brown lines are the apparent viscosity of tehineh shearing at 5 °C, 25 °C, and 45 °C, respectively (the blue dot markers, plus signs, and triangles are apparent viscosity experimental data), the apparent viscosity decreases rapidly within the first 100 min (5 °C). The apparent viscosity of tehineh declines obviously from 0–40 min (25 °C). The apparent viscosity of tehineh is easy to reach the equilibrium stage at 45 °C. At the low level of temperature, the apparent viscosity of tehineh did not change sharply, so the size of particles remains the same as before without shearing. Also the larger value of α corresponds to the larger value of k and a larger value of final apparent viscosity. The fractional order α can reflect the extent of thixotropy for tehineh (Color figure online)

3.2 Fitting results and physical interpretation

In this section, the FAVM presented above will be applied to analyze the three groups of experiments conducted under the constant shear rate.

These three groups of apparent viscosity experiments on tehineh, balangu, and natillas were conducted by Abu-Jdayil (2003), Razavi and Karazhiyan (2009), and Tarrega et al. (2004). The corresponding experimental data is shown in Figs. 1–3, respectively. The selected semisolid food samples all exhibited thixotropic behavior under different conditions. In order to validate the fractional apparent viscosity model, model (11) is used to fit these tests.

Figure 1 shows that the apparent viscosity of the tehineh decreases with time shearing at $\dot{\gamma} = 102 \text{ s}^{-1}$, under different temperatures. A good agreement between fitting results of FAVM (solid line) and the experimental apparent viscosity–time data for tehineh can be observed in Fig. 1. k is the ideal initial viscosity and appears sensitive to initial viscosity. The value of k significantly decreases with temperature due to the fact that the colloid particle distribution is heterogeneous during the initial stage. With the increase of temperature and time of shearing, colloid gradually tends to be homogeneous, eventually reaching equilibrium stage. The order α varies from 0.90 to 0.89, slightly decreasing with the temperature, which implies that the order reflects the influence of temperature on apparent viscosity variation. The relative errors of the three curves are 0.0025, 0.0032, and 0.0088 as shown in

Table 1 The parameters of the FAVM

Sample	k (Pa s)	α	Relative error	
tehineh	$T = 5\text{ }^\circ\text{C}$	6.0514	0.9000	0.0025
	$T = 25\text{ }^\circ\text{C}$	2.0815	0.9000	0.0032
	$T = 45\text{ }^\circ\text{C}$	0.9241	0.8905	0.0088
balangu	Shear rate 50/s	4.4031	0.8468	0.0032
	Shear rate 100/s	2.4512	0.8955	0.0003
	Shear rate 150/s	1.0875	0.9689	0.0005
natillas	Sample 7	1.5219	0.8960	0.0005
	Sample 3	1.0482	0.9085	0.0012
	Sample 1	0.8711	0.8416	0.0019

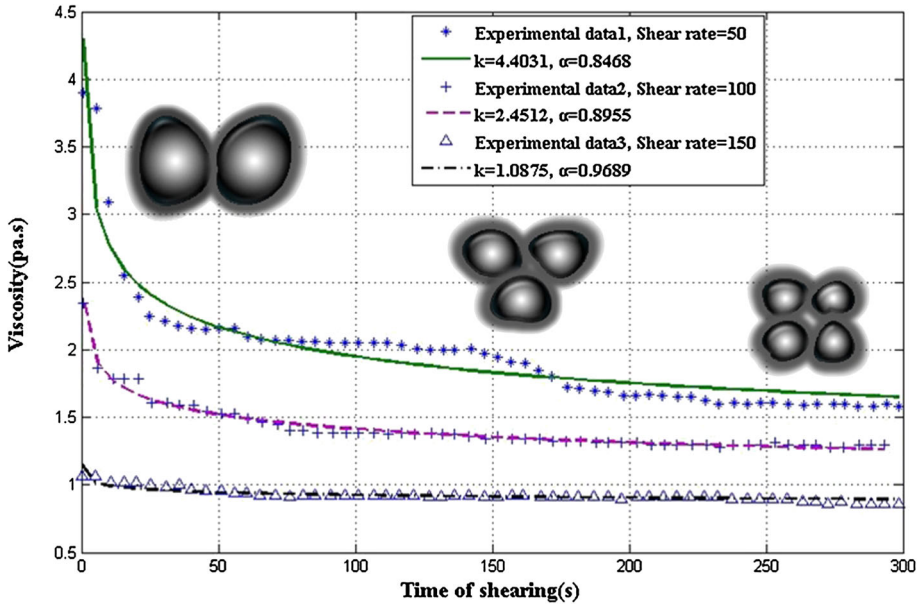


Fig. 2 Apparent viscosity of balangu solution as a function of shearing time at different shear rates (the experimental data is quoted from Razavi and Karazhiyan 2009). The green, pink, and black lines are the apparent viscosity of balangu shearing at 50 s^{-1} , 100 s^{-1} and 150 s^{-1} , respectively (the blue dot markers, plus signs, and triangles are apparent viscosity experimental data), the apparent viscosity decreases sharply within the first 50 s (50 s^{-1}). While the apparent viscosity of balangu changes slowly when shear rate is equal to 100 s^{-1} . The apparent viscosity of balangu contains slight attenuation (150 s^{-1}). At low level of shear rate, the balangu possess larger volume flocs; the size of flocs decreases with increasing value of shear rate. Also a larger value of α corresponds to a smaller value of k and a smaller value of final apparent viscosity. The fractional order α can reflect the extent of thixotropy for balangu solution (Color figure online)

Table 1. What’s more, the apparent viscosity of tehineh tends to decay rapidly at high temperature from the observation of Fig. 1.

The shear test of balangu solution was conducted at different values of constant shear rate as illustrated in Fig. 2. Within the first 50 s, the apparent viscosity decreases dramatically under shearing and reaches a constant value corresponding to a steady state after about 250 s. The ideal initial viscosity k decreases with the increases of shear rate. This fact means that

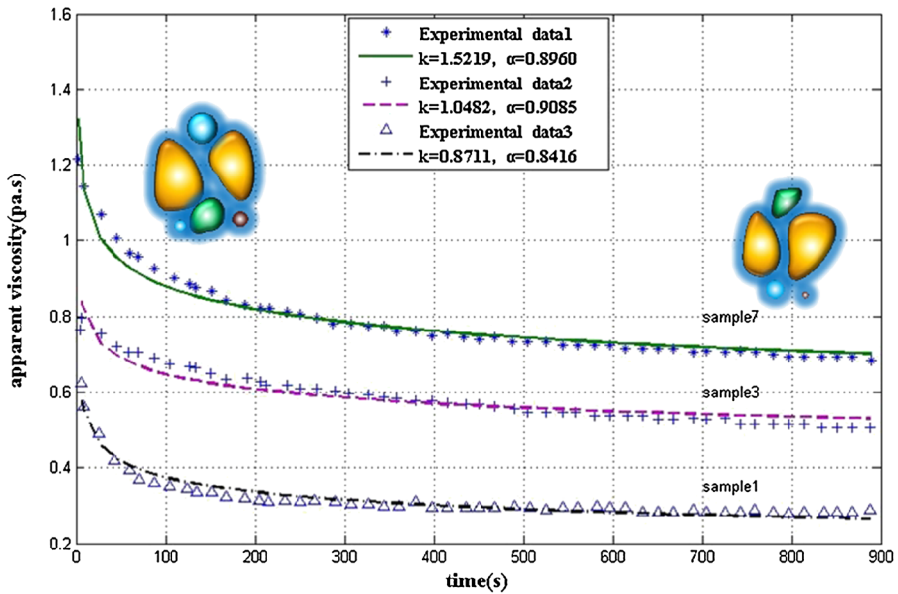


Fig. 3 Apparent viscosity data at a constant shear rate of 100 s^{-1} for some dairy dessert samples at $25 \text{ }^\circ\text{C}$ (the experimental data is quoted from Tarrega et al. 2004). The green, purple, and black lines are the apparent viscosity of natillas shearing at 100 s^{-1} , respectively (the blue dot markers, plus signs, and triangles are apparent viscosity experimental data), the apparent viscosity of natillas decreases sharply within the first 200 s (sample 1 and sample 7), while the apparent viscosity of natillas shows smooth recession (sample 3). A small value of α corresponds to a more drastic reduction of natillas (due to the different condition of every sample, there may be some fitting error for parameters). The fractional order α can reflect the extent of thixotropy for natillas (Color figure online)

the breakdown rate of balangu solution under shear is accelerating at high shear rate. The order α varies between 0.84 and 0.96 and is increasing with the shear rate. It is believed that the magnitude of α relates to the contribution of shear rate to apparent viscosity. Besides, a smaller α corresponds to a higher value of apparent viscosity at equilibrium stage. The relative errors of the three curves are 0.0032, 0.0003, and 0.0005 as shown in Table 1, which means good agreement between numerical results and experimental data.

The fitting result of the FAVM for natillas under the constant shear rate is displayed in Fig. 3, which offers good consistency between the FAVM (solid line) and the experimental apparent viscosity data for the three samples. The three samples contain different dairy ingredients and thickeners, the colloid particle size and distribution of internal condition of each sample are also different. Consequently, the values of k and α appear different. The fractional derivative order α varies from 0.84 to 0.91, which increases with content of soluble solids, while the ideal initial viscosity k is sensitive to the dairy ingredients. Table 1 shows that the relative errors of three curves are 0.0005, 0.0012, and 0.0019.

4 Discussion

In this study, we also use the traditional model of integer order derivative to fit the experimental data. The observation illustrates that the traditional model cannot describe the decay behavior of apparent viscosity. It is mainly because $\alpha = 1$ and $\eta(t) = k$ correspond to Newtonian fluid, $\alpha = 0$ and $\eta(t) = 0$ represent the ideal fluid without viscosity.

Fig. 4 The micro-change of semisolid foodstuffs. The *blue spheres*, *light blue oblate spheres*, and *oval spheres* connected by bonds represent particles, flocs, and networks. All of them are able to exist all at once (Color figure online)

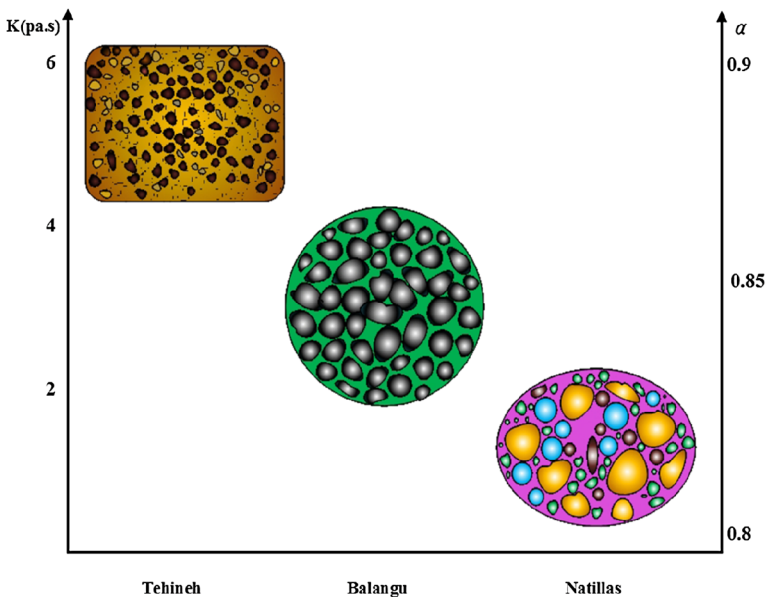
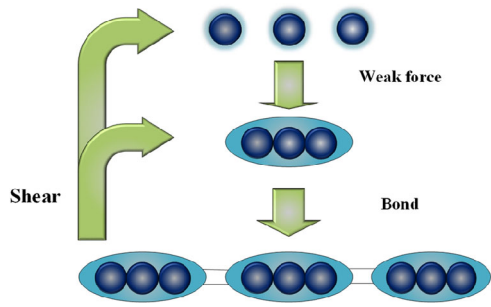


Fig. 5 The parameters of the FAVM. The *brown square*, *green circle*, and *pink oval* represents tehineh, balangu, and natillas, respectively. The particles of tehineh are small with low content. Balangu contains a large number of flocs and small particles, natillas possess larger sized particles, and the *different color of elements in the oval* represents different ingredients (Color figure online)

The thixotropic behavior of semisolid foodstuffs can be comprehended in accordance with microstructure and shear history. From the viewpoint of composite materials, the thixotropic behavior of semisolid foodstuffs results from additional effects from the interaction between all ingredients, i.e., relative weak attractive forces between food particles. The microstructures such as flocs and networks come into being due to the weak attractive forces. However, the bonds among networks are weak enough to be destroyed by the shear stresses under flow, resulting in a number of flocs, which causes the apparent viscosity decline with time from macroscopic angle. Meanwhile, the structure reconstruction requires a certain period of time and stress, and gives rise to thixotropic behavior as seen in Fig. 4. Since the varying microstructures of different foodstuffs and shear history, the ideal apparent viscosity k , together with the magnitude of α , is unique for a given kind of foodstuff. The corresponding values of the above two parameters are presented in Fig. 5 and can be recognized as unique indexes for foodstuffs.

It should be pointed out that the proposed model adopts a constant fractional order corresponding to a constant shear rate. In order to involve possible time-variant factors, such as temperature, pressure, etc., the variable-order fractional models can be taken into consideration in the future study. Moreover, some abnormal phenomena still puzzle researchers, for instance, viscosity bifurcate, and our theoretical scheme could also provide a new perspective.

As a matter of fact, many kinds of semisolid food contain inherent history-dependent feature which manifest in terms of apparent viscosity and the constitutive relationship. This study makes an attempt to build a fractional constitutive theory for semisolid food. Thixotropic phenomenon is a hotspot in soft matter mechanics, and the proposed model might be applied to characterize other soft matters, such as natural muds, biological fluid, clay/ceramic suspensions, cement/concrete, greases, etc. Hereby, more potential applications for the proposed model can be explored in the future study.

5 Conclusions

In summary, this study presented a fractional apparent viscosity model to describe the historical dependency of some semisolid foodstuffs, which exhibits thixotropic characteristics. Comparisons between experimental data and numerical results of fractional derivative model confirm that the FAVM with two parameters can well describe the apparent viscosity of thixotropic foodstuffs. Further research may concern on the constitutive relationship of time-invariant non-Newtonian substances.

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