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Original Research Paper

Rheological and viscoelastic behavior of concentrated colloidal suspensions of silica nanoparticles: A response surface methodology approach

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ABSTRACT

This study investigates the rheological behavior of fumed silica nanoparticle suspended in polyethylene glycol (PEG) at steady and oscillatory shear stress using a stress controlled rheometer. The suspension was prepared at different concentration from 25 to 45 wt.% silica nanoparticle and studied under the temperature of 20–60 °C. It is found that the fumed silica/PEG suspension exhibits shear-thickening behavior. Effects of silica nanoparticle concentration and solution temperature on the rheological behavior of silica suspensions in terms of initial viscosity, critical viscosity, and peak viscosity were quantitatively studied through the response surface method (RSM). The individual and interaction effects of silica concentration and temperature on viscosity of suspensions showed significant increase of critical viscosity versus silica concentration. On the contrary, a montonic decrease was observed as a function of solution temperature. Moreover, it is found that the onset of shear thickening for the sample with high silica content appeared at lower shear rate. Furthermore, increasing the temperature of suspension leads to an increasing the critical shear rate.

1. Introduction

Fumed silica, as an extremely versatile material, have become attractive and been used for preparation of shear thickening fluids (STFs) [1–5]. The STF is a specific type of non-Newtonian fluid whose viscosity increases with increasing shear rate or applied stress. In other words, under the high-speed shear rate the viscosity of STFs increases dramatically and acts like a solid. However, after removal of applied stress, the STF reverses to initial liquid-like status as soon as shear removes. This phenomenon has been observed in concentrated colloidal suspensions [6].

According to the literature, there are several theories proposed by researchers to explain the shear thickening behavior. A detail review shows that the "order–disorder" transition (ODT) [7–11] and the "hydrocluster" mechanism [12–14] give a better explanation compare to other theories. The ODT theory describes the breaking of particle arrangements in the fluid by the application of shear rate. The hydrocluster mechanism is based on particle interactions in a liquid medium. Based on this, the shear thickening occurs when hydrodynamic shear forces overcome repulsive steric and Brownian forces.

The rheological behavior of concentrated colloidal suspensions is controlled by a variety of parameters. The most important of which are the preparation methods, particles shapes and sizes, particles volume fraction, temperature, surfactant concentration and properties of continuous phase [15–19]. Barnes [15] showed a drastic change versus shear rate while particle volume fraction is more than 0.5. The more the volume fraction of particle, the lower critical shear rate (the shear rate at which shear-thickening begins) occurs. Similarly, Kang et al. [20] studied the effect of silica concentration on shear thickening behavior of silica suspension and found that the critical shear rates of STFs appears at lower shear rates for the higher concentrations of silica particle. Moreover, the results show a highly sudden increase in suspension viscosity as a function of the silica particle concentration. In another study, Wetzel et al. [21] observed the decreases in required particle volume fraction to reach the critical shear rate as the aspect ratio of the particles increases. Maranzano and Wagner [18] reported that particle size has great impact on reversible shear thickening transition in dense colloidal suspensions. They also observed the systematic shift of flow curves to lower shear stresses with increasing particle

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size. Similar behavior was also observed by Lee et al. [22]. Upon temperature effect on critical shear rate and viscosity of the STF, Kang et al. [20] reported that, the hydrodynamic force inducing the dilatancy phenomenon increases due to the increased thermal Brownian motion of silica particles in liquid medium, and therefore, the critical shear rate appears at higher shear rates as temperature of STFs increases.

Nowadays, several numerical methods are widely used for either modeling or optimizing the complex and non-linear processes generally found in nanoscience and technology. Response surface methodology (RSM) is a combination of mathematical and statistical techniques used to design experiments, evaluate the relationship between a set of controllable experimental factors and observed results, and also search optimum conditions of factors for desirable responses [23–25]. Therefore, the application of RSM to the colloidal suspensions should be helpful in an effort to find and optimize the rheological properties. Numerous studies have been published on the modeling and optimization of the rheological characteristics of the suspensions. However, to best of our knowledge, there is no report regarding the modeling and optimization of the rheological properties of STFs. Hence, the present study was conducted to explore the rheological behavior of fumed silica nanoparticles suspended in polyethylene glycol (PEG) and to investigate the effects of concentration and temperature on the viscosity of silica suspensions. For this purpose, the response surface model was employed to quantitatively investigate the effect of these variables on the response.

2. Experimental details

2.1. Materials and preparation

Fumed silica nanoparticles with a primary particle size about 12 nm and approximate specific surface area of 200 m^2/g were purchased from Degussa (Germany) and used as a suspending phase. Polyethylene glycol (PEG) with average molecular weight of 200 g/mol was purchased from Merck (Darmstadt, Germany) and used as continuous phase without further treatment. The properties of continuous phase could have a significant impact both on the rheological behavior and potential application of the STF. Low volatility, high thermal stability and close refractive index to the silica nanoparticles, which provide enhanced colloidal stability, makes PEG a good candidate as a continues phase in silica suspension. Moreover, the presence of OH bonds on the surface of PEG and also the presence of this type of bond in the silica particles leads to the formation of hydrogen bonds between PEG chains and silica nanoparticles and consequently hydrocluster formation becomes easier [26,27].

The fumed silica nanoparticles were gradually added into PEG and mixed using a mechanical mixer rotating at 3000 rpm. The mixing process was continued until a homogeneous and stable suspension was obtained. The suspension was then left undisturbed at room temperature for 48 h to remove air bubbles. The concentration of silica in the suspensions was selected 25, 30, 35, 40, and 45 wt.%.

2.2. Rheological measurements

The rheological measurements were conducted using a stress controlled rheometer (MCR-300, Physica/Anton Paar) with a cone-and-plate geometry (24.94 mm in diameter and 2° in cone angle). The samples were placed on the plate of rheometer and the cone placed into it on top of sample with a measuring gap at 1 mm. The temperature control device was utilized for measurements of the rheological properties of STF samples with different silica concentration were measured at the temperature of 20, 40, and 60 °C.

The linear viscoelastic region (LVR) for each sample was determined by amplitude sweep tests at a constant frequency of 10 Hz. The viscoelastic behavior measurements were performed by a frequency sweep from 0.01 to 1000 Hz at 25 °C within the LVR of all samples.

2.3. Experimental design and optimization

The response surface methodology (RSM) was used for the experimental design and optimization. The effects of silica concentration (wt.%) and temperature (°C) on the viscosity of STFs were investigated using RSM based historical design. The coded values were calculated according to the following equation:

$$X_i = \frac{\xi_i - [\xi_{Hi} + \xi_{Li}]/2}{[\xi_{Hi} - \xi_{Li}]/2} \quad (1)$$

where ξ_{Hi} and ξ_{Li} refer to the high and low levels of the variables ξ_i (i = 1,2), respectively. The behavior of the system is explained by the following quadratic polynomial equation:

$$Y = \beta_0 + \sum_{i=1}^{k}\beta_i X_i + \sum_{i=1}^{k}\beta_{ii} X_i^2 + \sum_{i=1}\sum_{j=i+1}\beta_{ij} X_i X_j + \varepsilon \quad (2)$$

where Y is the predicted response, X_i and X_j are the independent variables, β_0 is constant coefficient, β_i, β_{ii}, β_{ij} are coefficients estimated from the regression, ε and is the error [23]. RSM was applied to the experimental data using the statistical software, Design-expert (Version 8.0.3, Stat-Ease, 2010, USA). The experimental parameters and their levels are given in Table 1.

3. Result and discussion

3.1. Rheological behavior

The effects of silica nanoparticles concentration and temperature on the rheological behavior of STFs were studied. Fig. 1 illustrates the viscosity vs. shear rate for (a) 20 °C, (b) 40 °C, and (c) 60 °C at different silica content of 25, 30, 35, 40, and 45 wt.%, respectively. Fig. 1 represented three distinct regions: relative shear thinning at low shear rates, followed by sudden transition with an increase in viscosity over a critical shear rate, and finally shear thinning like behavior at high shear rates.

Fig. 1 shows that the critical viscosity (the viscosity which corresponds to the critical shear rate) increases with an increase of nanoparticle concentration and decrease with temperature accordingly. The increasing trend of critical viscosity was almost similar disregarding to temperature value. Moreover, results indicates that the onset of shear thickening for 25 wt.% containing silica nanoparticle appears at a higher shear rate compared to suspension containing 45 wt.%. In other words, the viscosity value for 45 wt.% suspension shows higher value compared to 25 wt.% at the same shear rate and temperature (Fig. 1). Furthermore, by increasing the temperature of suspension from 20 °C to 60 °C shear thickening behavior was observed at higher shear rate due to the increased thermal Brownian motion of silica nanoparticles in PEG. This can

Table 1
Coded and actual values of variables.

Factors	Coded levels of variables		
	Low level	Center level	High level
Silica concentration (wt.%)	25	35	45
Temperature (°C)	20	40	60

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