

Viscosity of dilute Na-montmorillonite suspensions in electrostatically stable condition under low shear stress

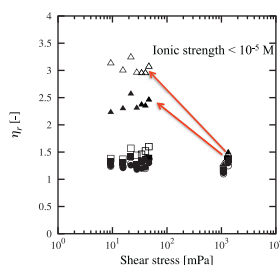
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HIGHLIGHTS

- ▶ We focused on the electroviscous effects of dilute Na-montmorillonite suspensions.
- ▶ The viscosity was measured using an original spiral type viscometer.
- ▶ An enhancement of viscosity was remarkable in the limit of salt free condition.
- ▶ We analyzed this effects based on the effective collision radius.

GRAPHICAL ABSTRACT



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ABSTRACT

We have studied the rheological behavior of dilute suspensions of electrostatically dispersed Na-montmorillonite to elucidate the importance of the electroviscous effect under the condition of zero stress limits. The viscosities of the suspensions were measured using an original spiral-type viscometer that consisted of two measuring cylinders connected with a one-meter-long capillary tube. Extremely dilute suspensions, whose volume fractions ranged from 2.0×10^{-4} to 2.0×10^{-3} , were used for the experiments. The ionic strength of the suspensions was controlled to be less than 0.001 M. Thus, a fully developed electrical double layer was formed. It was confirmed that the measured viscosities of the suspensions increased with a decrease in their ionic strength, in accordance with a manifestation of the electroviscous effect. The viscosity increased markedly owing to the secondary electroviscous effect under the limit of the salt-free condition. To analyze this effect, we focused on the effective radii of the suspended montmorillonite particles. We estimated their effective radii by fitting the Dougherty–Krieger equation to the viscosity data. We found that the obtained effective radii depended significantly on the ionic strength in the low Peclet number regime, which was similar to the theoretical dependence of the effective radii determined from the interaction balance between the particles governed by hydrodynamic, electrostatic, and diffusive energies. Thus, the increase in the effective radius with a decrease in the ionic strength gives rise to the expansion of the electric repulsive force, resulting in an increase of the viscosity of this clay suspension. In addition, the magnitudes of the effective radii were greater than the Debye length. This indicates that even when the distance between the montmorillonite particles is greater than the Debye length, the electrical repulsive force affects the interaction between the particles.

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1. Introduction

Montmorillonite is a clay mineral that has been used in many industries and has received many interests in terms of practical application [1]. Rheological behavior of this clay suspension is important for various applications such as oil drilling and is also related to adsorption or transportation of pollutants in

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underground water. It is known that the unit of this clay is quite thin layer with a thickness of 10 angstrom. The layer consists of two different sheets. One is silica tetrahedral sheet located out side of unit clay sheet. The other is alumina octahedral sheet hold between two silica sheets. Isomorphous substitution from Al^{3+} to Mg^{2+} in the alumina sheet yields permanent negative charges at the face of the layer. On one hand, the broken bonds located at the edge of the layer have a capacity to adsorb H^+ or OH^- . As a result, such charging property gives rise to different types of floc structure as well as colloidal stability [1–4]. These charging and flocculation–dispersion properties of montmorillonite affect the rheological characteristics of this clay suspension. Understanding macroscopic properties of montmorillonite suspensions in terms of microscopic colloidal interactions has been regarded as a major issue. The flow properties and sol–gel transition of montmorillonite suspensions are dependent on ionic strength, solid contents and particle size [4–6]. Abend and Lagaly carried out rheological measurements of montmorillonite suspensions as functions of these factors and revealed that the existence of two types of gelation, i.e., attractive gel and repulsive gels [4]. These two types of gel are formed by different mechanisms. One is the so-called card-house structure due to electrostatic attractive interaction between edge and face of clay, which provides attractive gel [1,3]. Laxton and Berg investigated clay yield stress relating to zeta potential as colloidal parameter using laponite suspensions and reported that yield stress is due to the development of microstructure through electrostatic attraction between the oppositely charged edge and face of clay particles [7]. The other mechanism is induced by the development of the electrical double layer (EDL) around the clay particles. The EDL induces the electrostatic repulsive force that causes gelation through the formation of the lamellar crystal like structure for the repulsive gel. Under electrostatically stabilized condition, the increase of viscosity and the appearance of yield stress occur with decreasing ionic strength. Our previous measurements on the yield stress of montmorillonite suspension using a vane spindle with torque sensor also revealed the increase of resistance against the deformation of suspensions with decreasing ionic strength [8]. The experimental condition was at higher pH where edge–face interaction is negligible. A simple model assuming parallel array of thin clay layer successfully explained the dependence of yield stress on the volume fraction of clay and the ionic strength on the basis of the repulsive force. According to their assumption, the estimated mean distance between clay layers at 1% volume fraction of clay is about 100 nm and is thus significantly longer than the thickness of EDL. They concluded that the electrostatic repulsive force is effective for long-range over the Debye length, and controls the appearance of the yield stress and gelation.

The effects of charging properties of clay particles and the development of EDL are also reflected in the viscosity of suspensions. So-called electroviscous effects of colloidal suspension can be ascribed to the additional distortion energy of fluid due to the formation of EDL around colloidal particles. The deformation of EDL against fluid distortion of shear causes the primary electroviscous effect, which appears as an increment of the intrinsic viscosity that was also affected by the shape of particles. This effect was investigated in detail by many scientists, in particular by Booth [9]. The secondary electroviscous effect is caused by the additional energy dissipation generated by the overlapping of EDL when two charged particles are conveyed to a close position by an external fluid motion. It was treated theoretically by Russel [10,11] and has been regarded as one of the major factor of non-Newtonian behavior. Krieger and Eguiluz pointed out that the secondary electroviscous effects are extremely important in modifying the rheological behavior of a colloidal suspension where the relative viscosity at a given shear rate can vary by several orders of magnitude [12]. Additionally, they demonstrated that this effect is

related to shear thinning behavior. Buscall analyzed this rheological behavior of concentrated lattices based on the Dougherty–Krieger equation (D–K equation) [13]. Furthermore, Takamura and van de Ven incorporated the particle interaction parameter into the D–K equation [14]. They revealed that the parameter was given as a function of Peclet number and the secondary electroviscous effects became negligible at high shear rate. For montmorillonite suspensions, there are a few experimental studies of the electroviscous effects. Egashira found that the intrinsic viscosity of montmorillonite is 190, which is much larger than that of the theoretical value of spherical particles [15] but he did not refer to electroviscous effects. Tamaki et al. measured the viscosity of dilute suspension of montmorillonite at high pH using the Ostwald viscometer and found that the magnitude of these electroviscous effects increases with a decrease of ionic strength, especially in the region of very low ionic strength, c.a. NaCl concentration less than 1.0×10^{-2} M [16]. However, experimental data for the dependence of the secondary electroviscous effect of montmorillonite suspension on the applied shear stress are still lacking. Therefore, it is desirable to analyze the secondary electroviscous effect of dilute montmorillonite suspension in the limit of zero shear stress. Kobayashi et al. developed a viscometer enabling us to measure the viscosity under zero shear limits and suggested the rheological behavior is non-Newtonian even if the suspension is so dilute that it is usually regarded as Newtonian fluid [17,18]. However, they did not mention the electroviscous effects in dispersed state under low shear stress. Thus, in this study, we analyze the electroviscous effects in dilute Na-montmorillonite suspensions under low shear stress on the basis of experimental results obtained by using this original viscometer as well as standard capillary viscometer.

2. Theory

2.1. Viscosity of colloidal suspension

The viscosity of colloidal suspension is expressed as a function of volume fraction, ϕ as follows,

$$\eta_r = \frac{\eta_s}{\eta_0} = 1 + K_1\phi + K_2\phi^2 \quad (1)$$

where η_r , η_s and η_0 denote the relative viscosity, the viscosity of suspension and the viscosity of solvent, respectively. K_1 is determined by the interaction between a single particle and surrounding fluid, which is corresponding to the intrinsic viscosity. On the other hand, K_2 is determined on the basis of the hydrodynamic interaction between two particles. The increment of K_1 and K_2 can be related to as the primary and the secondary electroviscous effects, respectively. Not only volume fraction but also Peclet number, Pe , affects the viscosity of suspensions. This is the dimensionless group given by the timescale for diffusive motion relative to that for convection of flow and defined as follows [10,11]

$$Pe = \frac{6\pi a^3 \tau}{kT} \quad (2)$$

where τ indicates the shear stress, and $2a$ indicates the hydrodynamic diameter. Boltzmann constant $k = 1.38 \times 10^{-23}$ J/K and absolute temperature $T = 298$ K, respectively.

Russel reported that the rheological behavior of dilute suspensions composed of monodispersed sphere is affected by the interaction between particles governed by viscous, electrostatic and van der Waals forces, etc. The balance among these factors is qualitatively described by a characteristic length which is called the effective collision radius (effective radius). He worked out theoretical treatment on the secondary electroviscous effect assuming

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