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# Effective viscosity measurement of interfacial bubble and particle layers at high volume fraction



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#### ABSTRACT

An experimental method for measuring the effective viscosity of two dimensional dispersion systems is proposed. The method is based on interfacial rheology, which was originally developed to investigate surface active adsorption layers such as protein film formed at liquid–liquid interfaces. Bubbles or rigid particles at around 50% of volume fraction in liquid are positioned in the gap between a rotating disk and a stationary cylindrical container. With this configuration, shear-rate dependent effective viscosities of bubble and particle layers were investigated. Steep shear-thinning property is observed for spherical bubble systems in the shear rate regime from  $10^{-1}$  to  $10^2$  s<sup>-1</sup>. This is explained by topological transition from regular to random arrangement of the bubbles at the interface. For rigid particle systems, the viscosity starts from high value due to solid contact friction, then decreases sharply due to fluidization process until inter-particle collision lead to an increase of the viscosity.

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### 1. Introduction

The effective viscosity of liquids containing bubbles and particles is already an over one century old research topic since its first appearance in literature [1,2]. As the disperse phase is suspended in the continuous liquid phase of viscosity  $\mu_0$ , the effective viscosity of the dilute suspension  $\mu$  is given by the following formula.

$$\frac{\mu}{\mu_0} = 1 + f\alpha, \begin{cases} f = 5/2 & \text{(solid)} \\ f = 1 & \text{(gas)} \end{cases}, \tag{1}$$

where  $\alpha$  denotes volume fraction of the dispersion phase. This formula is valid for spherical dispersion at small volume fraction,  $\alpha < 0.05$ . Schowalter et al. [3] and Choi and Schowalter [4] obtained the following formula valid for deformable bubbles and for higher volume fraction

$$\frac{\mu}{\mu_0} = \frac{1 + (1 + 4\alpha) \left(1 + (20/3)\alpha\right) \left((6/5)Ca\right)^2}{1 + \left(1 + (20/3)\alpha\right)^2 \left((6/5)Ca\right)^2} \left(1 + \alpha + \frac{5}{2}\alpha^2\right),\tag{2}$$

where Ca denoted the Capillary number defined by

$$Ca = \frac{\mu_0 \dot{\gamma} r}{\sigma},\tag{3}$$

http://dx.doi.org/10.1016/j.flowmeasinst.2014.10.006 0955-5986/© 2014 Elsevier Ltd. All rights reserved. in which  $\sigma$ ,  $\dot{\gamma}$ , and *r* being surface tension coefficient, shear rate, and bubble radius. Temporal transition from spherical bubble regime to shear-yield bubble regime was theoretically modeled by Pal [5]. The validity of these theoretical equations for various bubbly liquids were confirmed with circular Couette flow by Rust and Manga [6], Müller-Fischer et al. [7], and Gutam and Mehandia [8]. The similar trend was reported for pipe flows by Llewellin and Manga [9]. Murai and Oiwa [10] studied the influence of non-equilibrium deformation of bubbles in unsteady shear flow and found significant increase in effective viscosity compared to steady shear rate values. With higher gas volume fractions, the effective viscosity is significantly controlled by the complex liquid film dominating the bulk properties [11,12]. Pronounced viscoelastic properties occur in foam flow, which is influenced by surfactants and electrochemical interfacial properties [13–19].

Back to the early research, Mooney [20] suggested the following formula to describe the increase of effective viscosity at high volume fraction in the case of spheres with non-slip surfaces, i.e. rigid spherical particles:

$$\frac{\mu}{\mu_0} = \exp\left(\frac{A\alpha}{1-k\alpha}\right) = 1 + A\alpha + \frac{1}{2}A(A+2k)\alpha^2 + O(\alpha^3),\tag{4}$$

where *A* and *k* denote the dimensionless values, which approximate the measured viscosity and the spatial arrangement. The value *A* is given by A=1 for uncontaminated bubbles while A=5/2 for rigid particles and well-contaminated bubbles. The value *k* is a factor that describes the influence of spatial arrangement pattern of the spheres.

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Theoretically, the value *k* takes the range of 1.35 < k < 1.91 in accordance with two limits between a face-centered cubic lattice and simple cubic arrangement. Comparison of the polynomial terms in Eq. (4) with Eq. (2) yields that k=2 was employed in Eq. (2) at Ca=0, which exceeds the highest limit of k for spherical dispersion. Batchelor and Green [21] theoretically obtained the proportional factor to  $\alpha^2$  in case of rigid particle suspension to be 5.2 as the further higher order terms were neglected. This corresponds to k=0.83, which is lower than the above-mentioned lowest limit. A common issue in these theoretical works is uncapability to address the spatial fluctuation of local dispersion arrangement that naturually happens to real systems. Numerical simulations by Kuwagi and Ozoe [22] adapted a representative value of the factor at k=1.43 assuming a random arrangement. From the experiment of Darton and Harrison [23] and Tsuchiya et al. [24], average values of k for particle suspension at high volume fraction (0.45 <  $\alpha$  < 0.55) were k=1.2. This value is, on the other hand, being smaller than the theoretical smallest limit. Different from the theory, the spatial arrangement pattern of the spheres is hardly controlled in experiments. It varies with shear rate like a fluidization process of packed rigid spheres. Such transition of the arrangement produces dramatic shear rate dependence in viscosity whereas deformation of the dispersion is insignificant. Morris and Katyal [25] and Morris [26] found strong shear-thickening as a result of inhomogeneous particle distribution, which is triggered by direct contact among rigid spheres. Any case in the above previous works deals with 3D system of dispersion distribution subject to shear rate. Since 3D systems allow individual dispersions to move in three directions, shear-dependent viscosity correlates to transitions of three-dimensional arrangement pattern of dispersions. In contrast, rheological property of dispersion system confined in two dimensions has not been studied experimentally yet.

In the present research, we investigate the effective viscosity of interfacial bubble and particle layers at high interfacial volume fraction, i.e. from 0.2 to the packing limit (inter-lock condition of spheres). We focus on spherical shapes of the bubbles where deformability is almost negligible, thus Ca < 0.05. These conditions are attractive to engineers who aim at flow control by means of small dispersions [27-29]. Especially, in turbulent boundary layers, the dispersions interact with coherent structures in turbulence and form clusters within the layers. The clusters can have local volume fraction much higher than 0.2 and often reach the closest packing limit. This happens because strong congregation force acts on the dispersion due to steep local pressure gradient within coherent structures [30]. Thus, effective viscosity of dispersion at high volume fraction takes a primary issue rather in turbulences than in laminar flows [31–33]. A theoretical and numerical approach to such densely arranged spheres in simplified flow geometry was reported by Kang et al. [34]. Direct numerical simulation by Yeo and Maxey [35] showed particles selfdiffusion which results in shear-thickening characteristics.

In this paper, we desribe a new methodology to acquire the effective viscosity of interfacial bubble and particle layers at high interfacial volume fraction. The measurement principle is based on interfacial rheometry which was established for shear viscosity assessment of interfacial adsorption layers. General configuration and application of interfacial rheometry is reported elsewhere [36– 40]. The original purpose of the method is to quantify the rheological properties of a very thin material layers formed at liquid interfaces such as a protein films and surfactant adsorption layers. The present study extends this technique to the measurement of the viscosity of bubble/particle multiphase layers just by changing the shape of a rotating cone. In this article, the working principle of the present rheometry, the advantages in experimental handling of bubble/particle suspension layers, and their applications to bubble/ particle multiphase layer are reported accompanied with a brief discussion on the measured results.

#### 2. Interfacial rheometry

Fig. 1 shows the schematic diagrams of the experimental setups. The measurement system is constructed by the combination of a rotating disk attached to a commercial rheometer (Physica MCR300, Anton Paar) and a cylindrical fluid container. The setup (a) is used to measure the interfacial rheology of adsorption layers. The working principle of interfacial rheology is reviewed in the literature [36]. The setup (b) used in the present study is applicable for bubble and particle layer with a finite thickness. In both types, shear flow is driven by a rotating disk that is separately installed from a stationary cylindrical container. Viscosity of the sub-phase liquid,  $\mu_0$ , filled in the container is accounted for in the full flow analysis performed by the rheometer software [36]. In the setup (b), the structure of the fluid interface is additionally observed and recorded by a video camera (Sony DFW V500) from the top and the side of the container. With this combination, spatial distribution and motion of floating dispersions are monitored to find their correlations to bulk viscosity to be measured. The dimensions,  $R_1$ ,  $R_2$ , H, and  $\delta$  in Fig. 1 can be modified in accordance with the target material of measurement as well as the range of shear rate to be studied.

## 2.1. Original interfacial rheometry

Through a normal force transducer the biconical disk is positioned exactly at the interface. A ring-shaped gap between the rotating disk and the cylindrical container forms the free surface area to be measured. When the disk rotates, a circular Couette flow on the liquid surface within the gap is induced. In standard interfacial rheometry, the sharp edge of a rotating biconical disk drives the shear flow as shown in Fig. 1(a). The thickness of the material is ordinarily treated as unknown since this methodology is designed for adsorption layers with molecular dimensions. The interfacial viscosity has the dimension of [Pa s m]. Only in the case that the target layer is uniformly formed with a constant film thickness, the length scale multiplied in the dimension coincides with the film thickness. Dependent on the target material (e.g. surfactant, protein), the film



Fig. 1. Schematic diagram of the measurement system based on interfacial rheometry. Target fluid is set in a thin layer on the surface of liquid with known viscosity. (a) Rotating bi-cone with sharp edge for measurement of e.g. protein adsorption layers. (b) Rotating conical disk with a step for measurement of bubble/particle multiphase layer systems.

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