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Volume fraction dependence of viscosity curves for dilute and dense suspensions of gel-like microbeads fabricated in aerial process

空中生成マイクロゲルビーズ分散液のビーズ分率による流動特性変化

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1. Introduction

Viscosity and elasticity are important factors in any liquid handling fields for achademic studies as well as industrial applications. They are also the fundamental material properties for determining ultrasonic propagation and attenuation. In most cases, they show relaxation behaviors and/or resonance phenomena rather than constant values with respect to the time scale of dynamic response of the materials. Therefore, viscosity curve; a graph wherein viscosity and shear rate are plotted, is a useful means to directly compare a difference in the sound wave characteristics, since the shear rate has the dimmension of the inverse of time.

We recently developed an innovative system based on an electromagnetically spinning (EMS) method[1,2] for invsetigating the shear rate dependence of viscosity[3]. This system has a feature of remote driving and sensing the rotational motion of a rotor. Deu to this feature, more accurate measument can be achieved especially for low visocosity and in low shear rate, comapred with the other conventional viscometers. In addition, a completely sealed condition and a contamination-free operation are technically feasible.

These advantages in the EMS system will be solutions for evaluating the fluidity of biofluids such as tear, sweat, saliva, blood, and so on. In fact, the viscosity curves of human blooods were measured, and their downward tendencies with increasing shear rate (shear thinning) were found to significantly differ according to the owners of each measured bloods[3]. These differences are due to the mechanical properties and the volume percentage of blood cells, which are provably associsted with a health condition.

For more precise study of the relationship between the blood fluidity and the health condition, a suitable procedure for the measurement range and the analysis method must be determined. If possible, in the initial stage, to use control samples of soft-microbeads suspension with the varied material properties is preferred. However, there are no commercial products satisfying such a request, because of the absence of ready-made device for fabricating soft and micron-size structure at a practicable throughput.

Then, we tried to generate gel-like particles such as artificial roe with the diameter of about 10 microns using an originally developed microdroplet manupulation technique[4]. A liquid jet is emitted and in periodically brought up to a series of microdroplets by adding an ultrasonic vibration to the injection nozzle. The noteworthy feature of this technique is that the entire process; ejection, collision, capsulation, and gelation, is conducted in the air with no walled channel. Compared to other methods using microfluidics, the throughput of microbeads fabrication can be increased by more than 20 times. Incidentally, this technique can also be used for measureing dynamic surface and interfacial tention wih high time resolution[5,6].

First in this study, the originally developed technique for fabricating gel-like microbeads is introduced. Second, the typical results of viscosity curves for the suspensions of the self-made microbeads are shown. Finally, the differences in the obtained data are discussed with regards to the volume fraction of thses suspensions.

2. Experimentals

The used materials for fabrication process are two aqueous solutions of sodium alginates and calcium chlorides. The former is the main material for gelation, and the latter is the cross-linking agent. The latter is mixed with a small amount of ethanol to control the balance of surface and interfacial tension. As a result, a capsular structure wherein the latter encloses the former can be obtained spontaneously, when the both of microdroplets collide in the air. The gelation progresses with the diffusion of calcium ions from the outer to inner side, so the thickness of

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the gel-like layer might be controlled with adjusting the concentration of materials and/or the volume of ejected droplet. The fabricated microbeads were collected by plunging into distilled water in a test tube.

Figure 1 shows a typical photo image of the self-made suspension. The sample liquid was clearly separated into transparent and clouded layers after still standing for 1 hour. Then, the volume fraction of microgels was determined to be the percentage of occupation of the clouded layer. By adding or removing the water layer, the volume fraction of the measured sample was adjusted.

Note here that the separated sample was reverted to a homogeneous state by stirring. The homogeneity was confirmed by a microscope observation, and a typical microscopic image of the suspension is also shown in Fig. 1. Each microgel keeps its original shape without sticking together.

The measurements of viscosity curve were conducted by the EMS system using an auto-standing rotor[3]. This type of rotor can provide superior performance when the sample volume was set to be an appropriate amount, which was 0.6 ml in this study. The viscosity curves were obtained with sweeping shear rate form high to low in all the measurements.

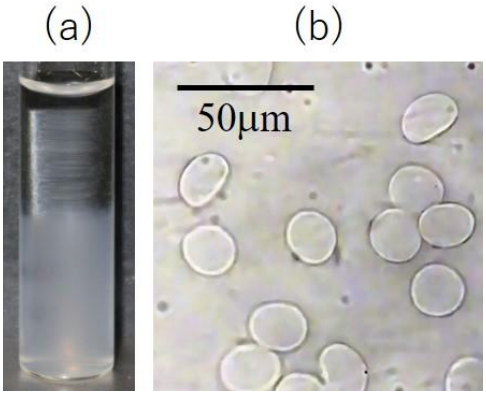

Fig. 1 (a) Macro and (b) microscopic photo images of separated and homogenous states, respectively.

3. Results and discussions

The comparison data of viscosity curves for the suspension samples with characteristic three kinds of percentages are shown in Fig. 2. The suspension of 30 vol%, being a dilute state, represented minute shear rate dependence. On the other hand, the suspension of 60 vol%, which is a dense state, showed a clear shear thinning behavior. The results that the sample with higher concentration show the stronger thinning tendency is reasonable compared with other suspensions of soft particles.

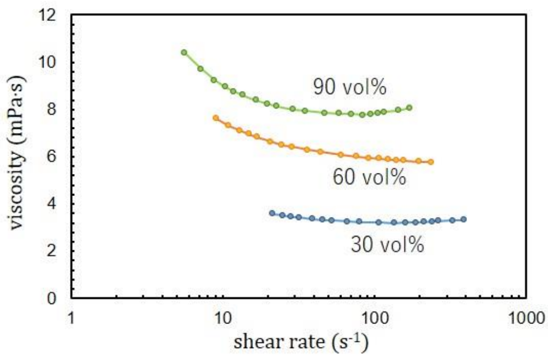

Fig. 2 Viscosity curves for self-made microgel suspensions with various volume fractions.

We already reported the degree of change of viscosity curve is probably due to the softness of gel-like beads[4]. A well-fitted function to these viscosity curves for evaluating the material properties is now under the consideration.

Here, let us focus on the data for the suspension of 90 vol%. Such very high concentration can be available only in soft suspensions. It is well known for hard particles there should be the upper limit of occupation, so called the closest packing of 74%. However, for the soft particles, the limit of packing ratio can increase according to the deformability of particles. In such extraordinary dense state, a shear thickening behavior is occasionally observed[7]. The shear thickening is tendency in opposition to the shear thinning, and then viscosity increases with the shear rate increasing. The data of 90 vol% in the high shear rate region was appeared to be a shear thickening. We will also discuss the border of percentage whether shear thickening occurs or not.

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