Experimental investigation into forming condition of airborne gel-like microparticles and preparing method for aqueous dispersion of them

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1. Introduction

The viscosity and/or viscoelasticity are the fundamental physical properties determining the hydrodynamic response. For example, they are needed to calculate the stress applied to a pipe wall and the pressure loss due to flow. Even for a complex situation for which the solution cannot be found, it is possible to predict the time evolution of a fluid behavior with using them as a main parameter for numerical simulation. Therefore, the viscotic and/or viscoelastic properties are in high demand for all industrial fields that handle fluids.

On the other hand, their properties are also the mechanical quantities reflecting the dissipation and propagation of energy. Namely, they have the possibility of changing according to the number of degrees of freedom of microscopic molecular motion as well as the magnitude of interactions between degrees of freedom. To have such characteristics, in most cases, they show relaxation or resonance phenomena with respect to frequency. Thus, spectroscopic study is a useful tool to reveal hydrodynamic behavior from the view point of an academic interest.

Although blood must be the most familiar fluid for human beings, its flow mechanism is still less well understood. While an angiographic technology and a hemodynamic simulation are advancing, viscometry techniques suitable for blood measurement have made slow progress. Why the development of measurement method for blood viscosity is lagging behind?

A human blood is known to show a non-Newtonian property such as shear thinning^{1,2)}, which means the viscosity decreases with increasing the shear rate. In short, we need to know the viscosities corresponding to every shear rates of blood flow in focus. Here, the shear rate is the same as the velocity gradient, and then it becomes higher when blood flows through narrower vessels. As the shear rate has the unit of inverse of seconds, the shear thinning is similar to viscous relaxation depending on ultrasonic frequency.

In such a situation, the viscosity of blood becomes only several to 10 times higher than that of water. It is extraordinary that a lowly viscous fluid has a clear tendency of shear thinning during the range of shear rate of 20 to 600 s⁻¹, which corresponds to the actual blood flow through the thickest to thinnest vessels in human body.

If we pay attention to the measurable range of the above-mentioned shear rates, a rotational-type rheometer must be the first candidate. In general, the commercially available rheometers, however, have insufficient accuracy in measuring low viscosity less than 10 mPa·s. On the other hand, a capillary-type viscometer that performs the highest measurement accuracy can not sweep the shear rate as expected.

Recently, the authors have developed a rheometry system equipped with the Electro-Magnetically Spinning (EMS) method^{3,4}). This EMS rheometer is a torque-controlled rotational-type device in noncontact manner, and then achieves the more accurate measurement rather than conventional high-end rheometers. In addition, a sealed condition, easy temperature control, and disposable usage are feasible owing to the noncontact technique, and also a contamination-free operation for preparing and removing samples is available. These features might be a powerful tool for examining large number of blood samples without spending time and cost.

Blood is a typical dense system, where red blood cells (RBCs) are dispersed in aqueous solution including some ions and proteins. The increase in viscosity stays at such degrees is due to that RBCs have some soft and flexible characters. Then, the flexibility and softness of RBCs can be estimated by investigating the viscosity curve in detail, which is a well-known graph plotting the relationship between viscosity and shear rate.

The final goal of this study is to obtain the quantitative data about the mechanical properties of RBCs by analyzing the viscosity curve of blood. It is preferred in the initial step towards achieving this goal to use an aqueous dispersion of microparticles covered with soft membrane as the model sample. Unfortunately, there are no commercial products satisfying such a request. Therefore, the authors also tried to airborne fabricate a salmon roe-like structure with the 10 micron-order diameters by using originally developed inkjet technique^{4,5)}.

In this presentation, our microfabrication technique is introduced, and appropriate condition

for forming capsule structure is investigated. In addition, preparing method of aqueous dispersion sample is discussed, and also the obtained data of viscosity curves are considered with regards to the volume occupation fraction of the self-made microparticles

2. Sample preparation

A schematic form of RBCs can be regarded as a capsular structure containing an inner aqueous solution and an outer gel-like membrane. Then, a microparticle of artificial roe will be useful as a substitute for RBCs. The raw materials are two aqueous solutions of sodium alginates and calcium chlorides. The former is the main material forming gel networks, and the latter is the curing agent for cross-linking reaction.

Both solutions are ejected into the air as microdroplets with the synchronized frequency so that each droplet of different solutions collides one by one. To spontaneously generate a capsular structure due to the balance of surface and interfacial tensions, ethanol is added only in the solution of calcium chlorides. Gelation reaction progresses at the contacting area towards the inside during airborne flying. The flying particles are finally caught by sufficiently large amount of distilled water in a test tube.

The typical flight time of fabrication process is approximately 10 ms. The thickness and softness of the gel-like membrane can be controlled with adjusting the concentration and/or molecular length of sodium alginates. Figure 1 shows a schematic image of micro fabrication system and the photo image of generated particles. It can be confirmed that microparticles with almost uniform diameters are dispersed individually.

The key feature of this self-made aqueous dispersion is to show sedimentation and redispersion properties similar to those of blood. The dispersion

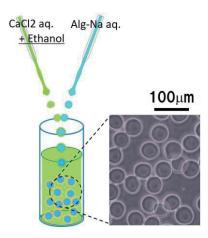


Fig. 1 Schematic image of microfabrication and microscope image of generated particles.

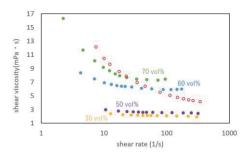


Fig. 2 Viscosity curves for the originally made dispersions (closed circles) and the real blood (open circle).

sample is separated into two parts; condensation layer of particles and supernatant liquid, after still-standing for several minutes. Then, the separated sample can be reverted to the homogenous state just by stirring. Therefore, the volume occupation ratio of the sample is readily adjusted by increasing or decreasing the volume of the supernatant liquid.

3. Viscosity curve measurements

The viscosity curves were measured with the EMS rheometry device using an auto-balancing rotor (ABR20003-ALE, TRIPLE EYE). In this type of setups, the sample volume should not be too much or little. The appropriate volume is 0.5 ml, which is acceptable to future practical usage for medical test of blood. All the measurements were conducted with sweeping shear rate from high to low after the preshear process, in which a few minutes of high shear flow was applied to the set sample.

Some examples of measurement data are shown in Fig. 2. These samples contain the self-made microparticles with the diameter of about 50 microns. There is significant difference in the magnitude of viscosity modulation for the volume occupation ratio above or below 50%. For comparison, measured data for my own blood are included as the closed symbols. From the view point of particle size and volume ratio, the difference in curve shape of shear thinning will be discussed in detail.

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