Transactions of the Materials Research Society of Japan 27 [1] 73-76 (2002)

Effect of Particle Rotation on Electrorheological (ER) Effect and Dielectric Properties in ER Suspensions

Yasuhito Misono, Junji Shinagawa and Keishi Negita
Department of Chemistry, Faculty of Science, Fukuoka University
8-19-1, Nanakuma, Jonan-ku, Fukuoka 814-0180, Japan
Fax: 81-92-865-6030, e-mail: misono@fukuoka-u.ac.jp

Relationship between electrorheological (ER) effect, effect of an applied electric field on rheology, and dielectric property is investigated on barium titanate ($BaTiO_3$) and carbon suspensions. Varying the frequency of the electric field (a few kV mm^{-1}), the ER effects and the first-order and third-order dielectric permittivities are measured simultaneously. In the ER measurements of the $BaTiO_3$ suspension, a decrease of the ER effect is observed above a frequency with its frequency increasing with the shear rate. While in the dielectric measurements, a small positive peak is observed in the first-order dielectric permittivity and a large negative peak in the third-order dielectric permittivity with the peak frequency nearly proportional to (shear rate)/4π. Referring to the theoretical result that under a steady shear flow the particle in the suspension rotates at a frequency of (shear rate)/4π, the observed dielectric peak indicates that the particle in the suspension rotates in spite of the field-induced chain-like structure formation. Similar results are obtained in the carbon suspension, suggesting that the shear-induced particle rotation would be a general property and should be considered for developing high-performance ER fluids.

Key words: nonlinear dielectric property, electrorheological fluid, particle rotation, barium titanate suspension, carbon suspension

1. INTRODUCTION

Under an application of a high electric field of a few kV mm^{-1}, a suspension composed of a micrometer-sized particle and insulating liquid shows a change of rheological property. This phenomenon, electrorheological (ER) effect, was first discovered by Winslow [1] over a half century ago. In his study, a dramatic change of viscosity due to a change from a liquid-like state to a solid-like state was found in some suspensions dispersing water-adsorbed particles into oil. The ER fluids, thus, have been expected as a potential functional material for mechanical devices such as shock-absorber, clutch, valve and so on, but, it was suggested that the suspensions containing water-adsorbed particles were not adequate for the ER fluids, since such suspensions are limited to a temperature range somewhere between -20 and 70 °C and have a high current level eventually leading to the electrical breakdown [2]. These inadequate properties were overcome by replacing the water-adsorbed particles by anhydrous semiconducting or ion-conducting particles [2, 3]. For practical use of the ER fluids, it is still required that increase of the shear stress is much larger than that of the present ER fluids. In order to achieve such higher performance, the fundamental understanding of the ER effect must be advanced.

The mechanism of the ER effect in suspensions has been recognized as follows. The application of the electric field induces polarization on the particle to form a chain-like structure by the dipolar interaction, which gives rise to an increase of the shear stress of the fluid. Thus, for developing higher-performance ER fluids, the dielectric properties responsible for the formation of the effective chain-like structure should be understood. For this purpose, a lot of dielectric measurements have been successfully performed at low field, but measurements at high field have been scarcely made. In our studies to clarify the electrical properties of the ER suspensions [4], we observed that the waveform of the current passing through the suspensions largely distorted from a sinusoidal one, suggesting that the nonlinear dielectric property is also associated with the ER effect.

In the present study, the ER and the nonlinear dielectric properties under the steady shear flow are measured simultaneously in two types of suspensions, barium titanate ($BaTiO_3$) and carbon suspensions. Barium titanate has been well known as a ferroelectric material having a permanent dipole, and carbon is a successful particle for the ER suspension. On the basis of the results of dielectric and rheological measurements, relationships between the ER effect and the nonlinear dielectric properties under a steady shear flow are discussed.

2. EXPERIMENTAL

The $BaTiO_3$ suspension was prepared by dispersing 10 vol% barium titanate particle having the particle size of ca. 0.3 μm (BT-01, Sakai Chemical Engineering Co., Ltd., Japan) into 50 cS silicone oil (TSF-451-50, Toshiba Silicone Co., Ltd., Japan). Before dispersion, the $BaTiO_3$ particle was annealed at 1073 K for four hours to improve the crystallinity. After dispersion, the suspension was homogenized by using a planetary ball mill (P-7, Fritsch GmbH, Germany). The carbon suspension was a commercial ER suspension (BA-1, Bridgestone Co., Ltd., Japan), containing ca. 30 vol% carbon particle, and used as received. In these ER fluids, the ER effect and the current were measured simultaneously by using a viscometer of a double cylinder type by applying a high voltage across 1 mm gap between the outer and the inner cylinders made of stainless steel 304 [5]. A small AC voltage from a multifunction generator (1946, NF electric instruments, Co., Ltd., Japan) was amplified up to 2 kV by using a high voltage amplifier (664, Trek Inc., U.S.). The current passing through the suspensions was monitored by using a lock-in amplifier (7260, EG&G instruments, U.K.); the first and the third harmonics contained in the current were converted to the first-order and the third-order dielectric permittivities, respectively [6]. All measurements were carried out at 300.0 K. AC electric field strength, *E*, used in this paper is expressed by rms.

3. RESULTS

3.1 ER effect and dielectric properties in the $BaTiO_3$ suspension

Varying the shear rate, the ER effect of the $BaTiO_3$ suspension is measured as a function of the frequency of the applied electric field. As is obvious in Fig. 1, a yield stress, an increment of the shear stress induced by the application of the electric field, decreases above a frequency. Such a behavior is recognized in other ER suspensions, but in this suspension it should be noted that the frequency dependence of the yield stress depends on the shear rate: the yield stress decreases above a frequency with its frequency increasing with the shear rate. At a constant shear rate, the frequency dependence of the yield stress is measured at some electric field strength (Fig. 2), indicating that the frequency dependence of the ER effect is independent of the electric field strength. In order to examine the dielectric properties responsible for these ER effects in detail, frequency dependence of the first-order and third-order dielectric permittivities was obtained at a constant electric field strength (Fig. 3) and at a constant shear rate (Fig. 4). As Fig. 3 (A) shows, the first-order dielectric

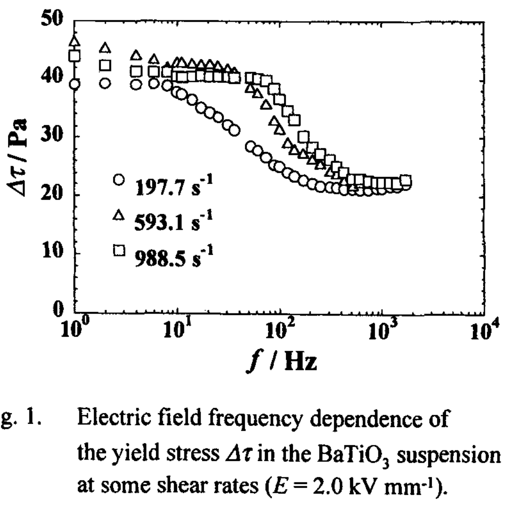

Fig. 1. Electric field frequency dependence of the yield stress $\Delta\tau$ in the $BaTiO_3$ suspension at some shear rates ($E = 2.0$ kV mm^{-1}).

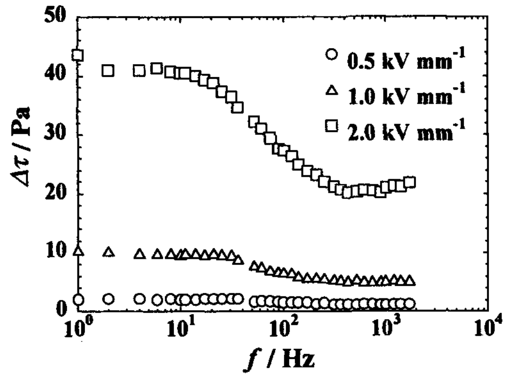

Fig. 2. Frequency dependence of the yield stress $\Delta\tau$ in the $BaTiO_3$ suspension at some electric field strength (shear rate = 329.5 s^{-1}).

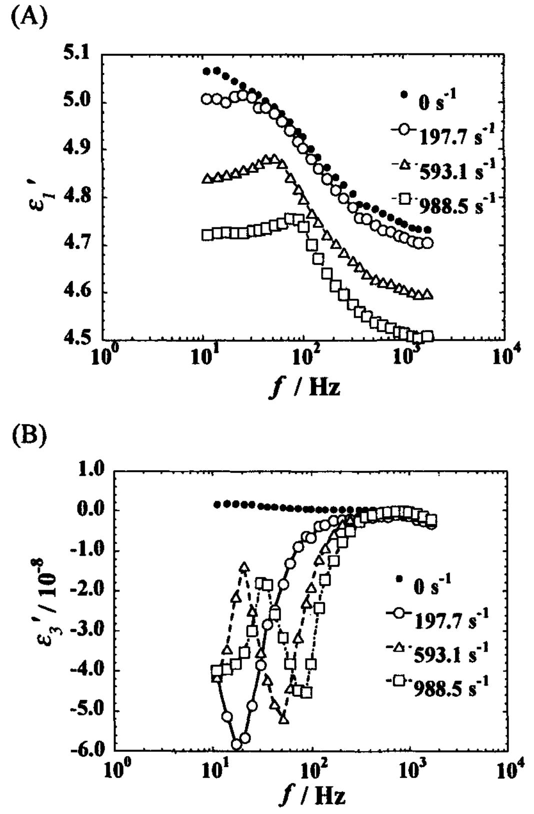

Fig. 3. Frequency dependence of the dielectric permittivities in the $BaTiO_3$ suspension at some shear rates ($E = 2.0$ kV mm^{-1}).
(A) first-order dielectric permittivity.
(B) third-order dielectric permittivity.

permittivity is modified by applying the shear deformation. In a quiescent state, the first-order dielectric permittivity monotonously decreases with the frequency. On the other hand, under the steady shear flow the first-order dielectric permittivity has a peak with its frequency (f_p) proportional to the shear rate with a relation of f_p = (shear rate)/4π. Such shear rate dependence is more clearly seen in the third-order dielectric permittivity as shown in Fig. 3 (B); under the steady shear flow, a clear negative peak is obtained with its frequency nearly equal to that of the first-order dielectric permittivity. From Fig. 4, it is recognized that the peak frequency in the first-order and third-order dielectric permittivities is not altered by changing the electric field strength. These results indicate that the peak is due to some shear-induced motion under the steady shear flow.

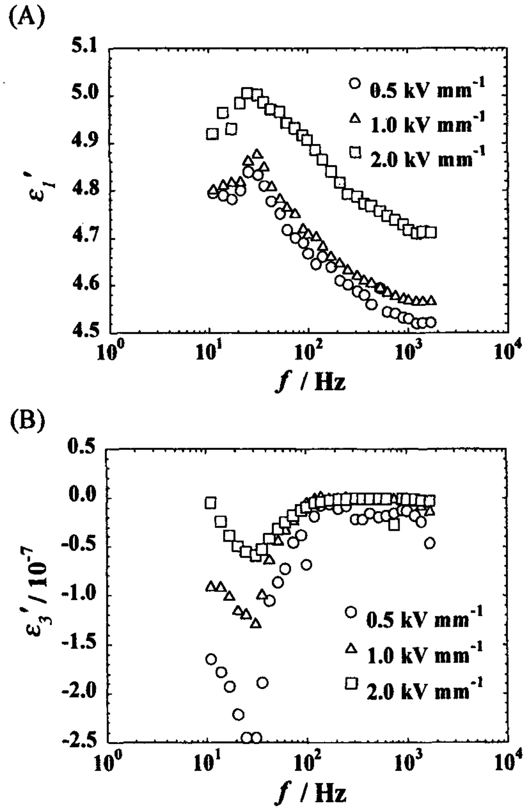

Fig. 4. Frequency dependence of the dielectric permittivities in the BaTiO3 suspension at some electric field strength
(shear rate = 329.5 s^{-1}).
(A) first-order dielectric permittivity.
(B) third-order dielectric permittivity.

3.2 ER effect and dielectric properties in the carbon suspension

ER and dielectric properties under the steady shear flow were also investigated on the carbon suspension. In Fig. 5, frequency dependence of the yield stress is given. As this figure shows, there are two processes responsible for the ER effect. The first one appears in the frequency region below 100 Hz; in a similar manner to the $BaTiO_3$ suspension, a small decrease of the yield stress is observed above a frequency with its frequency depending on the shear rate as (shear rate)/4π. The second one appears above 100 Hz without depending on the shear rate. In these processes, the second one can be explained by our results of the dielectric permittivity

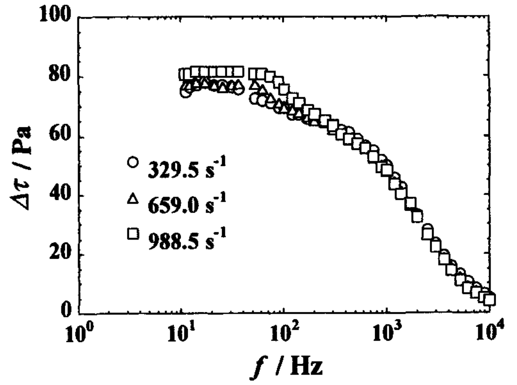

Fig. 5. Frequency dependence of the yield stress $\Delta\tau$ in the carbon suspension at some shear rates (E = 1.0 kV mm^{-1}).

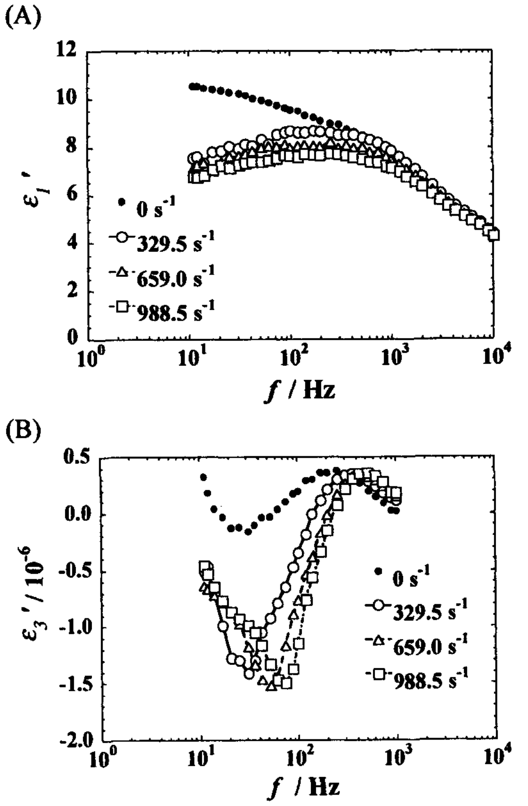

Fig. 6. Frequency dependence of the dielectric permittivities in the carbon suspension at some shear rates. (E = 1.0 kV mm^{-1}).
(A) first-order dielectric permittivity.
(B) third-order dielectric permittivity.

observed at low electric field [7]: a clear dielectric dispersion is observed around 1 kHz. This dielectric dispersion is also confirmed in the first-order dielectric permittivity at high field (Fig. 6 (A)), indicating that the second process in the ER effect is due to the relaxation of the interfacial polarization. With respect to the first process, the frequency dependence of the third-order dielectric permittivity (Fig. 6 (B)) is suggestive. A clear negative peak is also observed in this suspension, which indicates that the first process is associated with some shear-induced motion similar to that of the $BaTiO_3$ suspension.

4. DISCUSSION

In our present study, it is found that at high electric fields there appears a negative resonance peak in the third-order dielectric permittivity in the $BaTiO_3$ and the carbon suspensions and a positive resonance peak is in the first-order dielectric permittivity in the $BaTiO_3$ suspension. The peak frequency of these resonance peaks is dependent on the shear rate with a relation of (shear rate)/4π, indicating that some shear-induced particle motion is responsible for the resonance peaks.

For understanding this particle motion, theoretical study on the particle motion and dielectric studies of the suspension under a steady shear flow are suggestive. It is theoretically clarified that under a steady shear flow the particles in the suspension rotate with an angular frequency of (shear rate)/2 [8]. This is experimentally confirmed in a dielectric measurement conducted at a lower electric field [2]; a resonance peak appears in the dielectric permittivity when the frequency of the electric field matches that of the particle rotation. This behavior has been known well as flow-modified permittivity (FMP).

The theoretical and experimental results indicate that the observed resonance peaks at high electric fields are due to particle rotation under a steady shear flow. This implies that each particle can rotate in spite of the formation of the chain-like structure, giving rise to some modification in the ER effect. Considering that similar dielectric and ER properties are observed in the $BaTiO_3$ and the carbon suspensions, the particle rotation, which is induced even at high electric fields, would be a general property of the ER suspensions, and this property should be taken into consideration for developing higher-performance ER fluids.

In our study, the effect of the particle rotation appears clearly in the third-order dielectric permittivity rather than the first-order one. For clarifying such a behavior, a theory to make clear the nonlinear dielectric response in a suspension under steady shear flow and high electric field is necessary, but studies of the nonlinear dielectric properties have been made only in some homogeneous systems such as liquid crystals [6]. Further accumulation of experimental results and a new theoretical approach should be required. Analysis by applying some theoretical models [9, 10] is now in progress.

ACKNOWLEDGEMENT

This study was supported by Special Coordination Funds for Promoting Science and Technology from the Japanese Ministry of Education, Culture, Sports, Science and Technology, to whom we are deeply indebted. The authors wish to thank Professor Hiroyuki Ikawa and Mr. Kōji Nakamura of Kanagawa Institute of Technology for supplying the $BaTiO_3$ suspension generously.

REFERENCES

1. W. M. Winslow, *J. Appl. Phys.*, 20, 1137 (1958).
2. H. Block, J. P. Kelly, A. Qin, and J. Watson, *Langmuir*, 6, 6 (1990).
3. F. E. Filisko and L. H. Radzilowski, *J. Rheol.*, 34, 539 (1990).
4. Y. Misono, N. Shigematsu, T. Yamaguchi, and K. Negita, "Studies in Surface and Catalysis 132", Ed. by Y. Iwasawa, N. Oyama, and H. Kunieda, Elsevier (2001), pp. 391-394.
5. K. Negita, *Netsu Sokutei*, 22, 137 (1995).
6. Y. Kimura, *Ekisho*, 4, 73 (2000).
7. Y. Misono, T. Yamaguchi, and K. Negita, *J. Mol. Liquids*, 90, 333 (2000).
8. G. B. Jeffery, *Proc. R. Soc. London A*, 102, 161 (1923).
9. O. Nakada, *J. Phys. Soc. Jpn.*, 15, 2280 (1960).
10. Y. Kimura and R. Hayakawa, *Jpn. J. Appl. Phys.*, 31, 3387 (1992).

(Received December 21, 2001; Accepted January 30, 2002)