*Transactions of the Materials Research Society of Japan* 25 [1] 159-162 (2000)

# Concerted Amplification of the Viscosity in Some Complex Fluids under application of Electric Field

Keishi Negita, Yasuhito Misono, and Takashi Yamaguchi
Department of Chemistry, Faculty of Science, Fukuoka University,
Nanakuma 8-19-1, Jonan-ku, Fukuoka 814-0180, Japan
Fax: 81-92-871-6030, e-mail: negita@ssat.fukuoka-u.ac.jp

Effect of electric field on the viscosity, electrorheological (ER) effect, of complex fluids such as suspensions and liquid crystals is discussed from a viewpoint of the concerted amplification. Of some ER properties observed for carbon, barium titanate, and zinc oxide suspensions, interesting to shed new light on the ER mechanism is the difference in the electric field frequency dependence of the ER effect among these suspensions. A decrease of the ER effect with the electric field frequency is observed in the carbon suspension, but an increase in the barium titanate suspension and a decrease followed by an increase in the zinc oxide suspension. While in the liquid crystals, various ER effects of nematic (Ne), chiral nematic (Ne*), smectic A (SmA), and chiral smectic C (SmC*) phases can be observed. From these results, a possibility of the concerted amplification of the viscosity in the complex fluids is discussed.

Key Words: electrorheological effect, suspensions, liquid crystals, concerted amplification, viscosity

## 1. INTRODUCTION

Some complex fluids show an increase of the viscosity under an application of an electric field of a few kV/mm. This phenomenon, electrorheological (ER) effect, was first discovered by Winslow [1] about a half century ago in suspensions composed of water-adsorbed particles of a few μm and electrically non-conducting liquids. In such a fluid, the application of the electric field makes the viscosity of the fluids change dramatically, making a liquid-like fluid change to a solid-like one. The ER fluids, thus, have been expected to be a potential material for some mechanical devices such as shock-absorber, clutch, valve and so on. In recent years, fundamental and technological studies of this fluid have become more extensive after findings of more practical anhydrous ER fluid, where semi-conductive or ionic-conductive particles are used [2,3]. The ER fluids, thus, are fascinating functional materials, but the viscosity (shear stress) change due to the electric field is not enough for the practical application to the mechanical devices. At present, the shear stress change of the most effective fluids is at most a few kPa. If it becomes one hundred times larger, the ER fluids would be widely used in various mechanical devices.

The ER effect is caused by a formation of a chain-like structure across the electrodes; the electric field induces a polarization on the particle and the resultant dipole-dipole interaction between the particles is responsible for the structure formation [4,5]. Thus, for developing a higher perfomance ER fluids, inductions of a larger polarization on the particle and a more effective fluid structure are necessary. For these purposes, a concept of concerted amplification, which means a non-linear amplification of some physical quantity by a field induced cooperative behavior of the constituents, is fruitful. In the present study, a possibility of a concerted amplification of the viscosity, which would be accomplished by a concerted amplification of the polarization and a concerted formation of the structure, is discussed through the verification of the ER behaviors and the ER mechanism of some complex fluids.

## 2. EXPERIMENTAL

Measurements were made on anhydrous suspensions and liquid crystals. The suspensions investigated were carbon (30 vol %) /silicone oil, barium titanate (5 vol %) / silicone oil, and zinc oxide (20 vol %) / silicone oil suspensions, of which the carbon suspension is a commercial one (BA-1, Bridgestone). For liquid crystals, some substances are used to measure the ER properties of various liquid crystalline phases; pentylcyanobiphenyl(5CB) and octyloxycyano biphenyl (8OCB) for the nematic (Ne) phase, octyloxy phenyl(methybutyl)biphenylcarboxylate (8OBE) for the chiral nematic (Ne*) phase, octylcyanobiphenyl (8CB) for the smectic A (SmA) phase, and octyl resorcylidene octylaniline (MORA-8) for the smectic C* (SmC*) phase. The ER effect was measured with a viscometer of a double cylinder type. The electric field was generated by applying an AC high voltage of a few kV to the gap (1 mm) between the inner and the outer cylinders. The electric field, thus, is applied along a direction perpendicular to the flow. The AC electric field strength is expressed by rms.

## 3. RESULTS

### 3.1 ER effects in suspensions

Effect of the electric field on the rheology (shear stress vs. shear rate) for three suspensions is given in Fig. 1. The electric field induces an increase in the shear stress (viscosity) with somewhat different behavior among three suspensions; the increment of the shear stress due to the electric field , yield stress, is almost independent of the shear rate in the carbon suspension, whereas a decrease with the shear rate is observed in the barium titanate [6] and an increase in the zinc oxide suspensions. The magnitude of the yield stress for the carbon suspension is larger than those for the other two suspensions.

In Fig. 2, electric field frequency dependence of the yield stress is given. The carbon suspension shows a decrease of the yield stress above c.a. 300 Hz and becomes smaller at frequencies higher than a few kHz [6]. Such a behavior has been recognized in other ER suspensions [7,8]. On the other hand, the yield stress monotonously increases with the frequency in the barium titanate suspension [6]. In the zinc oxide suspension, the yield stress decreases at lower frequencies, but increases at higher frequencies with its behavior depending on the shear rate.

The yield stress in these suspensions is almost proportional to square of the electric field strength. A representative result, which is obtained in the carbon

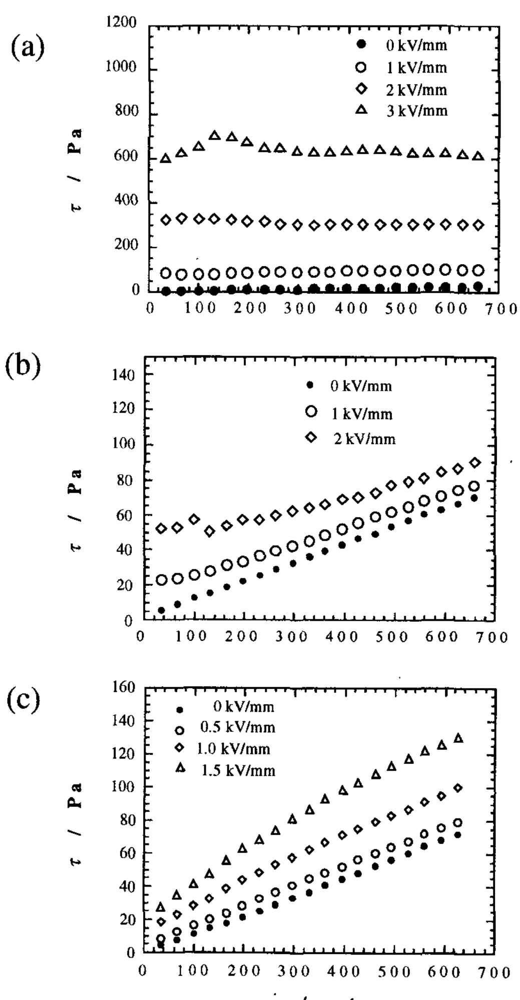


Fig.1 Effect of the electric field (20 Hz) on the rheology, shear stress vs. shear rate, at 300 K. (a) carbon, (b) barium titanate, and (c) zinc oxide suspensions.

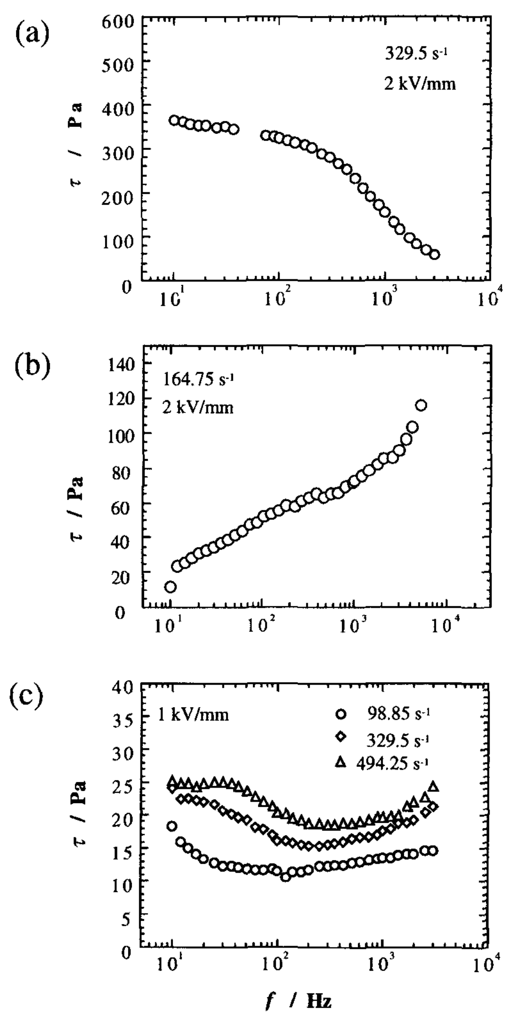


Fig. 2 Electric field frequency dependence of the yield stress $\Delta\tau$ at 300 K. (a) carbon, (b) barium titanate, and (c) zinc oxide suspensions. Conditions of the measurements (shear rate and electric field strength) are given in the figures.

suspensions, is depicted in Fig. 3. Such electric field dependence indicates that a dipolar interaction is responsible for the increase of the viscosity.

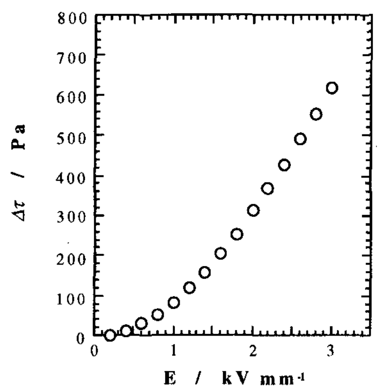


Fig. 3 Electric field dependence of the yield stress in the carbon suspensions. Shear rate: 329.5 $s^{-1}$, electric field: 20 Hz.

3.2 ER effects in liquid crystals

Unlike the ER behavior in the suspensions, a variety of ER effects are observed in the liquid crystals owing to the characteristic structures of the respective liquid crystalline phases. (a) In the Ne phase, the viscosity monotonously increases with the electric field strength and saturates at higher fields (Fig. 4-(a)) [9]. (b) In the close vicinity of the SmA - Ne phase transition point, $T_{AN}$, the viscosity increases to give a maximum, and then decreases to a constant value at higher fields (Fig. 4-(b)) [10]. (c) In the chiral Ne (Ne*) phase, the shear stress does not show a distinctive change until rather high field, but above a threshold electric field, at which the helical cholesteric structure is unwound, the shear stress increases with the electric field strength (Fig. 4-(c)). (d) The shear stress decreases with increasing the electric field strength in the SmA phase (Fig. 4-(d)) [11]. (e) In the ferroelectric liquid crystalline (SmC*) phase, the shear stress increases at lower fields, but at a moderate electric field strength it discontinuously decreases to a lower value owing to a change from a multi-domain to a mono-domain structure, which is followed by an increase of the shear stress at higher fields [12].

The ER mechanism in liquid crystals is thoroughly understood in the Ne phase [9], but is not well characterized in other liquid crystalline phases. However, it has been clarified that the change of the viscosity is brought about by changes of the molecular orientation, layer, and domain structure. The dipolar interaction, which mainly contributes to the ER effect in the suspension, does not play an essential role in the liquid crystals.

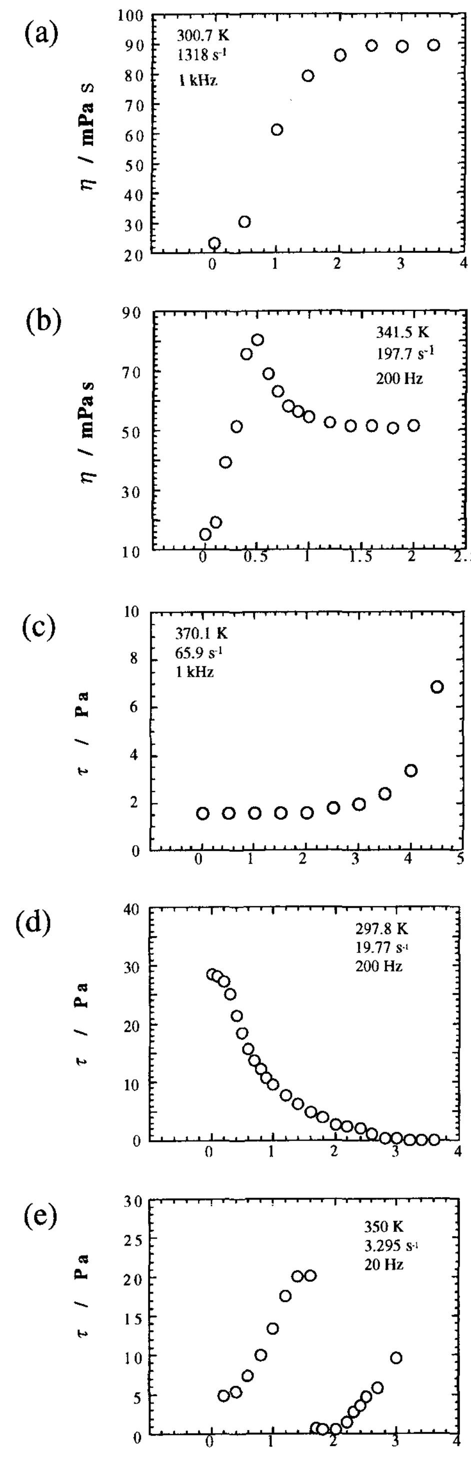


Fig. 4 Electric field dependence of the viscosity (shear stress) in liquid crystals. (a) Ne phase (5CB), (b) Ne phase near $T_{AN}$ (8OCB), (c) Ne* phase (8OBE), (d) SmA phase (8CB), and (e) SmC* phase (MORA-8). Conditions of the measurements (temperature, shear rate, and frequency of the electric fields) are given in the figures.

## 4. DISCUSSION

### 4. 1 Amplification of the polarization

Although the mechanism how the polarization is induced by the electric field is not well understood, the results given above and accumulated until now show us some suggestions to get a fluid exhibiting a larger polarization, i.e. a larger ER effect. In the case like the carbon suspension, where the ER effect decreases with increasing the frequency of the electric field, following factors should be considered. (i) The dielectric constant of the particle, $\varepsilon_p$, should be much larger than that of the liquid. This can be simply understood from the relation derived by Maxwell and Wagner; the interfacial polarization **P** induced by an electric field **E** is expressed by $\mathbf{P} \propto a^3\varepsilon_l(\varepsilon_p - \varepsilon_l)/(\varepsilon_p + 2\varepsilon_l)\,\mathbf{E}$, where $a$ is a radius of the particle, and $\varepsilon_p$ and $\varepsilon_l$ are dielectric constants of the particle and the liquid, respectively[13]. (ii) The relaxation time of the polarization should be shorter than $(2\pi f)^{-1}$ and $\Omega^{-1}$, where $f$ is the frequency of the electric field and $\Omega$ is an angular velocity of the particle rotating under a shear flow, which is given by a shear rate $\dot{\gamma}$ as $\dot{\gamma}/2$. If these conditions are not fulfilled, the polarization along the electric field diminishes, since the polarization can not follow the alternating electric field or its direction is rotated away form the direction along the electric field [7]. (iii) The existence of mobile charges near the interface of the particle. This needs further studies, but our recent measurements of the electric current vs. the electric field for the carbon suspension indicates an interesting result. In the low frequency region, where the ER effect is large, the waveform of the current distorts from the sinusoidal wave and the current increases non-linearly with the electric field strength. On the other hand, in the high frequency region where the ER effect is small, a linear relationship is observed between the current and the electric field strength. Although the mechanism of such a non-ohmic behavior observed in the low frequency region should be clarified scientifically, it is similar to that observed in the cellulose, where an interstitial conduction occurs; mobile charges near the interface give rise to a non-linear relationship between the current and the electric field [14]. Our result on the carbon suspension, thus, indicates that mobile charges near the interface is strongly associated with the ER effect.

On the other hand, the ER effect characterized by an increase of the yield stress with the frequency, which is found in the barium titanate suspension, needs further investigations. Although an idea incorporating frequency dependent polarizability, which suggests a larger polarizability at higher frequencies, is developed [15], our result that the dielectric constant of this suspension decreases with increasing the electric field frequency [6] does not support this idea. The result in the zinc oxide suspension (Fig. 2-(c)), where the frequecy dependence of the ER effect depends on the shear rate, may indicate that an effect of shear rate, e. g. particle rotation, is associated with such frequency dependence.

### 4.2 Formation of concerted structure

As described in section 3.2, the structure of the liquid crystals is largely modified upon application of the electric field. If the particles are dispersed in liquid crystals, a complex structure, which may be called a concerted structure, is formed. For this kind of fluid, it is not so irrelevant to expect that the ER effect is concertedly amplified to make the effect larger than sum of each ER effect due to the particles and the liquid crystals. The study of such a concerted structure may also give us a suggestion for development of a high performance ER fluid, and is in progress.

## ACKNOWLEDGEMENT

This study was supported by Special Coordination Funds for Promoting Science and Technology from the Japanese Science and Technology Agency, to whom we are deeply indebted.

## REFERENCES

1. W. M. Winslow, *J. Appl. Phys.*, 20, 1137 (1958).
2. H. Block, J. P. Kelley, A. Qin, and J. Watson, *Langmuir*, 6, 6 (1990).
3. F. E. Filisko and L. H. Radzilowski, *J. Rheol.*, 34, 539 (1990).
4. A. P. Gast and C. F. Zukoski, *Adv. Colloid Interface Sci.*, 30, 153 (1989)
5. D. J. Klingenberg, F. van Swol, and C. F. Zukoski, *J. Chem. Phys.*, 91, 7888 (1989).
6. Y. Misono, T. Yamaguchi, and K. Negita, to be published in *J. Mol. Liq* (2000).
7. K. Negita, *J. Phys. II France*, 5, 883 (1995).
8. D. K. Klass and T. W. Matrinek, *J. Appl. Phys.*, 38, 67 (1967).
9. K. Negita, *J. Chem. Phys.*, 105, 7837 (1996).
10. K. Negita, Electro-Rheological Fluids, Magneto-Rheological suspensions and their Applications, Ed. by M. Nakano and K. Koyama, World Sci. (1998), pp. 423-428.
11. K. Negita, *Mol. Crys. Liq. Crys.*, 300, 163 (1997).
12. S. Fukayama and K. Negita, to be published in *J. Mol. Liq* (2000).
13. R. W. Sillars, *J. Instr. Elect. Eng.* (London), 80, 278 (1937).
14. E. J. Mulphy and H. H. Lowry, *J. Phys. Chem.*, 34, 598 (1930).
15. P. J. Rankin and D. J. Klingenberg, *J. Rheol.*, 42, 639 (1998).

(Received December 17,1999 ; Accepted February 15,2000)