Ferroelectrics, 2002, Vol. 270, pp. 93–98
Reprints available directly from the publisher
Photocopying permitted by license only

THERMAL PROPERTIES OF AN ELECTRO-RHEOLOGICAL FLUID BASED ON $Bi_4Ti_3O_{12}$ CERAMIC POWDERS

F. DONADO, M.E. MENDOZA, V. DOSSETTI, E. LOPEZ-CRUZ,
AND J.L. CARRILLO
Instituto de Física, Universidad Autónoma de Puebla, A.P. J-48, Puebla,
72570 México.

(Received in final form November 24, 2001)

Thermal diffusivity of a ferroelectric based electro-rheological fluid is studied. The rheological fluid is prepared by dispersing powders of $Bi_4Ti_3O_{12}$ in silicon oil. The thermal diffusivity of the sample, under different intensities of applied electric field, was measured by means of an open cell photo-acoustic technique. We have found that the main contribution to the measured photo-acoustic signal, comes from the thermo-elastic bending effect. It was clearly observed that thermal diffusivity increases with the intensity of the applied electric field, this fact is explained in terms of the formation of clusters of $Bi_4Ti_3O_{12}$ particles.

Keywords: electro-rheological fluid; ferroelectric; bismuth titanate; thermal diffusivity; photo-acoustic.

INTRODUCTION

Electro-rheological (ER) fluids are suspensions of particles in an inert fluid which undergo reversible changes in its rheological properties such as viscosity, plasticity and elasticity, when it is subjected to the application of the electric field (AC and/or DC), also heat transfer is changed under application of the field. This behavior is due to the formation of a columnar structure of the particles [1]. ER fluids have been proposed for a wide range of applications in the electrical control of systems such as hydraulic valves, clutches, heat exchangers and suspensions systems.

The substances commonly used as particles are metals like aluminum; glass; ceramic powders like potassium niobate, lithium niobate, barium titanate, strontium titanate or even polymers like polyacrylonitrile [2-6]. In this context, ferroelectric materials are interesting as particles due to the presence of spontaneous polarization and high dielectric constant. One expects that they must be more sensitive to the applied electric field than the above mentioned materials. There are few reports concerning the use of ferroelectric particles in ER fluids, most of them are focused on barium titanate [7-9]. Generally, the activity of ER fluids is explained in terms of dielectric constants and conductivity of the solid and the liquid material. Particle polarization could involve primarily a particle-fluid conductivity mismatch or a dielectric constant mismatch [10].

Bismuth titanate, $Bi_4Ti_3O_{12}$, (BTO), is a displacive bidimensional ferroelectric mixed oxide, the transition from the paraelectric tetragonal phase, 4/mmm, to the ferroelectric monoclinic phase, m, occurs at 675°C. The ferroelectric phase is easily described as pseudo-orthorhombic, by the point group 2mm; at room temperature lattice parameters are a = 5.450 Å, b = 5.4059 Å and c = 32.832 Å, so the typical morphology is lamellar [11,12]. The ER fluid studied in this work is constituted by BTO particles ($\varepsilon = 150$, 1 kHz) [13] at a volume fraction of 7 % in silicone oil (poly(dimethyl siloxane)), ($\varepsilon = 2.5$) [10].

EXPERIMENTAL

BTO ceramic powders were obtained by coprecipitation of bismuth nitrate and ammonia titanyl solutions, followed by a thermal decomposition at 1000°C , as we reported elsewhere [14]. The ER fluid was prepared by dispersing these powders in silicone oil (Aldrich), in a square cell (8.5 mm by side, 1 mm height) built with cover glass, the electrodes were made with silver paint on both sides of the cell and one of them has been also painted in black, in order to improve thermal contact.

Thermal diffusivity was measured by the photo-acoustic (PA) open cell technique [15], the system is composed of a 250 W incandescent tungsten lamp, a chopper (Stanford Research Inc. mod. SR540), and a conventional optics to focalize the beam into the sample. The light impinges the sample and the heat generated perturbs the air between the sample and the diaphragm of an electret microphone, the signal was detected by means of a lock-in amplifier (Stanford Research Systems Inc. mod. SR510) in order to register the amplitude of the signal. DC electric field was applied using a high voltage power supply (246 Keithley Instruments).

RESULTS AND DISCUSSION

According to the PA spectroscopy theory, it is commonly accepted that the acoustic signal has two contributions, one coming from the properly called thermal diffusion and the other one from the thermo-elastic bending effect [15,16]. To elucidate which one of these contributions dominates in our measured PA signal, we have compared the experimental results with the well known expressions that describe those contributions. We have found from this comparison, that thermo-elastic effect is clearly dominant. Furthermore, the difference between the thermal expansion coefficient of BTO, 11×10^{-6} /K [17], and of silicone oil, 8×10^{-4} /°C [18], is relatively large. It has been found that in solid samples, this condition propitiates the predominance of the thermo-elastic bending mechanism [15]. Then, to evaluate α from the modulation frequency dependence of either, the signal amplitude or signal phase, we shall use the following expression for the pressure fluctuations induced by thermo-elastic effect, in the thermally thick regime

$$p_{el}=(3\alpha_T R^4 \gamma P_0 I_0 \alpha_s)/(4\pi R^2{}_c l^2{}_c l_g k_s f)[(1- 1/x)^2 + (1/x^2)]^{1/2} e^{j[\omega t + (\pi/2) + \delta)]}, \quad (1)$$

where $x = l_s a_s = l_s (\pi f / \alpha_s)^{1/2}$, α_T is the linear thermal expansion coefficient, R is the microphone inlet hole radius, γ is the air specific heat ratio, R_C is the radius of the PA chamber in front of the diaphragm, P_0 is the ambient pressure, I_0 is the absorbed light intensity, f is the modulation frequency, and l_i , k_i *and* α_i are the length, thermal conductivity, and thermal diffusivity of material *i*, respectively and $\tan \delta = 1/(x-1)$. Equation (1) means that the thermo-elastic contribution, at high modulation frequencies such that $x >> 1$, varies as f^{-1}, and its effective phase ϕ_{el} approaches 90° as $\phi_{el} \approx \pi/2 + \arctan [1/(x-1)]$.

Fig.1 shows the amplitude and the phase of the PA signal for the bismuth titanate based ER fluid as a function of the chopper frequency, for a representative value of the applied electric voltage. The continuous line is obtained by using the expression (1), circles are our measured values. We observe a good agreement between the theoretical predictions and the measured values for the amplitude of the PA signal, but the fitting for the phase case is relatively less accurate. This poorer fitting can be associated to the interference effects due to the thinness of the cell which propitiate backwards propagation of secondary signal.

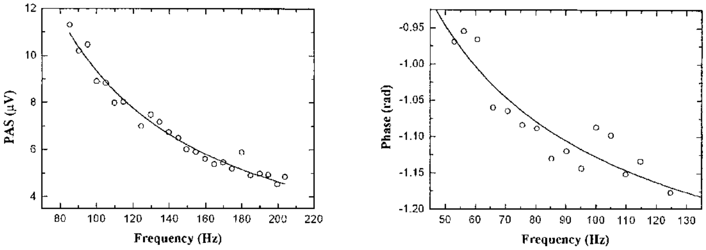

FIGURE 1 Fitting of (a) the amplitude and (b) phase of the PA signal for the bismuth titanate based ER fluid under applied voltage of 750 V.

With the calculated fitting parameters for the thermo-elastic function, and since $l_s = 965\ \mu$, it is possible to estimate the thermal diffusivity α. The α values obtained are 0.08, 0.17 and 0.19 cm^2/sec , for the increasing values of the applied voltage, 250, 500 and 750 V, respectively. The measured α value for the plain silicone oil is 0.04 cm^2/sec.

We shall focus our attention on the general trend that these results show, namely, that the ER effect increases the thermal diffusivity of the complex fluid. In order to discuss this, we will consider the possible mechanisms which mediate to produce the observed behavior, the clustering of particles and the propagation of the acoustic signal through the resulting clusters-oil composite media.

Field induced polarization is responsible for the ER effect, it is generally accepted that particle polarization could involve primarily, a particle-fluid conductivity mismatch or a dielectric constant mismatch [10]. The dielectric mismatch, namely, the ratio of the dielectric constant of the particles to that of the fluid, in our bismuth titanate based ER fluid has a value of 60, that is favorable for the ER effect, as it has been observed in strontium titanate suspension [8]. Moreover, recently it has been observed that the ER activity in perovskite-based fluids is driven by the particle dielectric response [9].

In absence of an electric field, and for low particle concentration, the particles in the ER fluid are homogeneously dispersed in the oil. When the field is turned-on, at low intensities, we have observed that the particles aggregate forming clusters. The weak interaction among them provides an additional energy transport mechanism. We have discussed the analogous situation for a magneto-rheological slurry [19]. Thus, the clustering of particles explains the larger value of α at low voltage, in comparison to the corresponding value for the plain silicon oil.

By increasing the applied voltage, the clusters of particles evolve into larger structures going from one extreme to the other of the cell, as it can be appreciated in the microphotograph shown in Fig. 2. In this situation, the contribution to α from the network of particles becomes more important for increasing applied voltages.

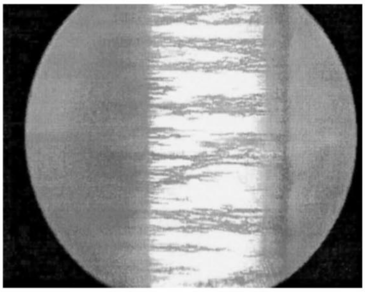

FIGURE 2 Fibrillar structure formed by clusters of BTO particles in silicone oil in the presence of an electric field.

CONCLUSIONS

Thermal diffusivity of bismuth titanate based electro-rheological fluid was experimentally studied by the open cell photoacoustic technique, for different values of applied voltage, 250, 500 and 750 V . For increasing voltages, thermal diffusivity increases too, this fact can be explained in terms of the changes induced by the voltage in the BTO particles cluster structure. In our system, the difference between the thermal expansion coefficients of BTO and silicone oil, is relatively large, this leaded us to assume that the dominant contribution to the PA signal comes from a thermo-elastic bending effect, this assumption agrees, of course, with the fitting to the equation (1). It is worthy of mention that the thermal diffusivity values determined by the procedure we used here, are not pure values, because in the PA measurements it is always present a contribution of thermo-diffusive nature.

Acknowledgements
This work was partially supported by CONACYT-México, under the grant 32100-E. F.D. and V.D. received a scholarship from CONACyT.

References

[1] P.P. Phule and J.M. Ginder, *MRS Bull.* 19, August (1998).
[2] W.Wen and L. Lu, *Appl. Phys. Lett.* **67**, 2147(1995).
[3] W.Wen and K. Lu, *Appl. Phys. Lett.* **68**, 3659(1996).
[4] K.Lu, W. Wen, C. Li, and S. Xie, *Phys. Rev. E* **52**, 6329(1995).
[5] T. Hao and Y. Xu, *Appl. Phys. Lett.* **69**, 2668 (1996).
[6] T. Hao, *Appl. Phys. Lett.* **70**, 1956 (1997).
[7] C.A. Randall, D.E. McCauley, C.P. Bowen, T.R. Shrout and G.L. Messing, *Proc 4th. Int.Conf. on ER Fluids*, R.Tao and G.D. Roy Eds. (1994), p.60.
[8] Y. Zhang, Y. Ma, Y. Lan, K. Lu, and W. Liu, *Appl. Phys. Lett.* **73**, 1326 (1998).
[9] H. Bose and A. Trendler, *Proc. 7th.Int.Conf.on ER Fluids and MR suspensions*, R. Tao Ed.,(World Scientific, Singapore, 2000), p.80.
[10] J.M. Ginder, L.C. Davis and S.L. Ceccio, in *Progress in Electrorheology,* Eds. K.O. Havelka and F.E. Filisko, (Plenum Press, New York, 1995), p.281.
[11] S.E. Cummins and L.E. Cross, *J. Appl. Phys.* **39**, 2268 (1968).
[12] D. Rae, J.G. Thompson, R.L. Whiters, and A.C. Willis *Acta Cryst.B* **46**, 474 (1990).
[13] A. Fouskova and L.E. Cross, *J. Appl. Phys.* **41**, 2834 (1970).
[14] F.Donado, M.E.Mendoza and J.L. Carrillo, 7th ECERS Conference Brugge, 9-13 Sept. 2001.
[15] L.F. Perondi and L.C.M. Miranda, *J. Appl. Phys.* **62**, 2955 (1987).
[16] A. Rosencwaig and A. Gersho, *J. Appl. Phys.* **47**, 65 (1976).
[17] E.C. Subbarao, *Phys. Rev.* **122**, 804 (1961).
[18] < www http://www.satron.com/hps.htm>
[19] F. Donado, M.E. Mendoza, and J.L. Carrillo, *Physica A* **295**, 81(2001).