Magnetic materials-based electrorheological fluids

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A type of electrorheological (ER) system, denoted the magnetic materials-based electrorheological fluids, is introduced. The solid particles of this system are 40–50 μ-microspheres obtained by the sol–gel processing of a ferroelectric material containing a ferromagnetic component. Since the solid material is magnetic, the presence of a small magnetic field, such as that from a small permanent magnet, can suspend the microspheres in liquid. The incorporation of a small amount of magnetic materials thereby solves the long standing problem of particle sedimentation in ER fluids. It is found that this type of ER fluid is very stable and exhibits a strong ER effect at low electric field. © *1997 American Institute of Physics.* [S0003-6951(97)01843-3]

Electrorheological (ER) fluid is a two-phase mixture whose rheological properties are controllable by the application of an electric field. In the past decade, research in ER fluids has attracted much renewed attention because of its wide-ranging potential applications.[1] However, realization of ER fluids' application potential requires the solution of a host of problems, one of which is the sedimentation of the solid phase. In this letter, a type of material, denoted ''magnetic materials-based ER fluids,'' is introduced.[2] The solid component of this material is made by adding a ferromagnetic component to the ferroelectric sol–gel solution during the gelation process, and shaped into spherical particles with a high-temperature spouting device. Since the spheres are ferromagnetic, the application of a small magnetic field, such as that from a small permanent magnet, is sufficient to suspend the spheres in liquid. The incorporation of a small amount of magnetic materials in the solid particles thereby solves the traditional problem of particle sedimentation. It is found that this type of ER suspension also displays a strong ER effect in low electric field, sometimes surpasses 2 kPa at 1 kV/mm.

The flow diagram for the fabrication of magnetic materials-based lead zirconate titanate (MBPZT) microspheres is illustrated in Fig. 1. The steps are as follows: (1) Preparation of lead zirconate titanate (PZT) stock solution:[3] Lead acetate $[\mathrm{Pb(CH_3CO_2)_2 \cdot 3H_2O}]$, Zirconium propoxide, $[\mathrm{Zr(C_3H_7O)_4}]$, and titanium isopropoxide $[\mathrm{Ti[(CH_3)_2CHO]_4}]$ were used as precursor materials. 12 gm of lead acetate were dissolved into 12 ml of acetic acid, which had been preheated over 100 °C to remove any trace of water. After cooling to below 80 °C, 6.55 gm of Zirconium propoxide and 2.18 gm of titanium isopropoxide were then added sequentially. The solution was agitated by ultrasonic cleaner at 60 °C until all solids were dissolved. Finally, the solution was diluted with 50 ml of distilled water. (2) Preparation of MBPZT solid gel: 1 gm of ferric sesquioxide, $\mathrm{Fe_2O_3}$ (0.1 mm in particle size) and 3 gm of polyethylenglycol were added to the PZT solution. The solution was agitated with a magnetic stirrer at 80 °C for about 1 h until gelation started. Then, it was heated in an oven at 200 °C to form a solid gel. (3) Fabrication of MBPZT microspheres: The solid gel was ground and sieved. The sieved particles were then flame treated using a spouting device at about 1000–1200 °C to form microspheres.[4] The microspheres were then coated with a thin layer of PZT film using diluted PZT stock solution and the modified sol–gel method reported recently.[5] The coated microspheres were annealed at 620 °C for about 2 h to obtain a high dielectric constant, better adhesion, and hardness.

Figure 2 shows the scanning electron microscope (SEM) micrographs of MBPZT particles before and after the flame treatment to form microspheres. The insets are x-ray diffraction (XRD) results showing the amorphous state of MBPZT particles [Fig. 2(a)] and the crystalline tetragonal phase of the MBPZT microspheres [Fig. 2(b)].

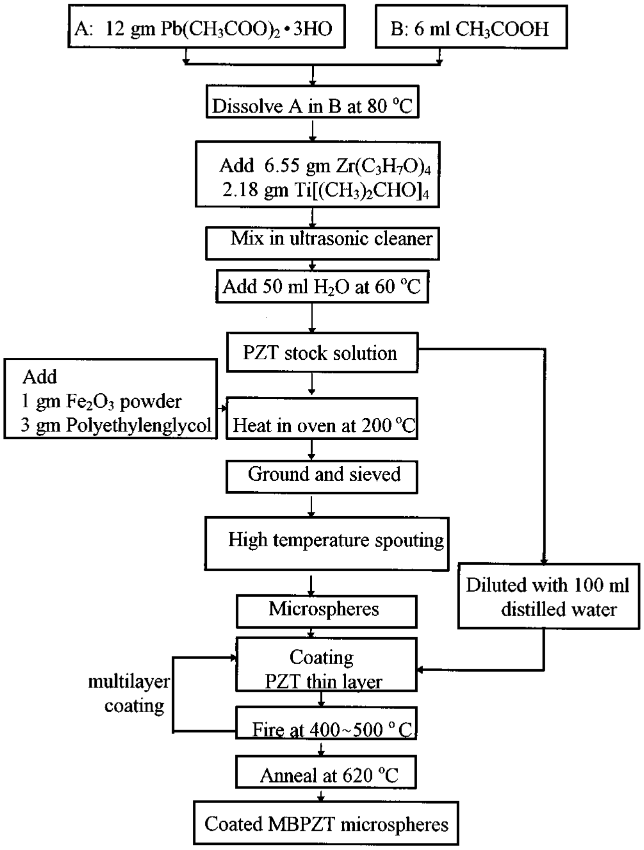

FIG. 1. Flow diagram for the fabrication of MBPZT microspheres.

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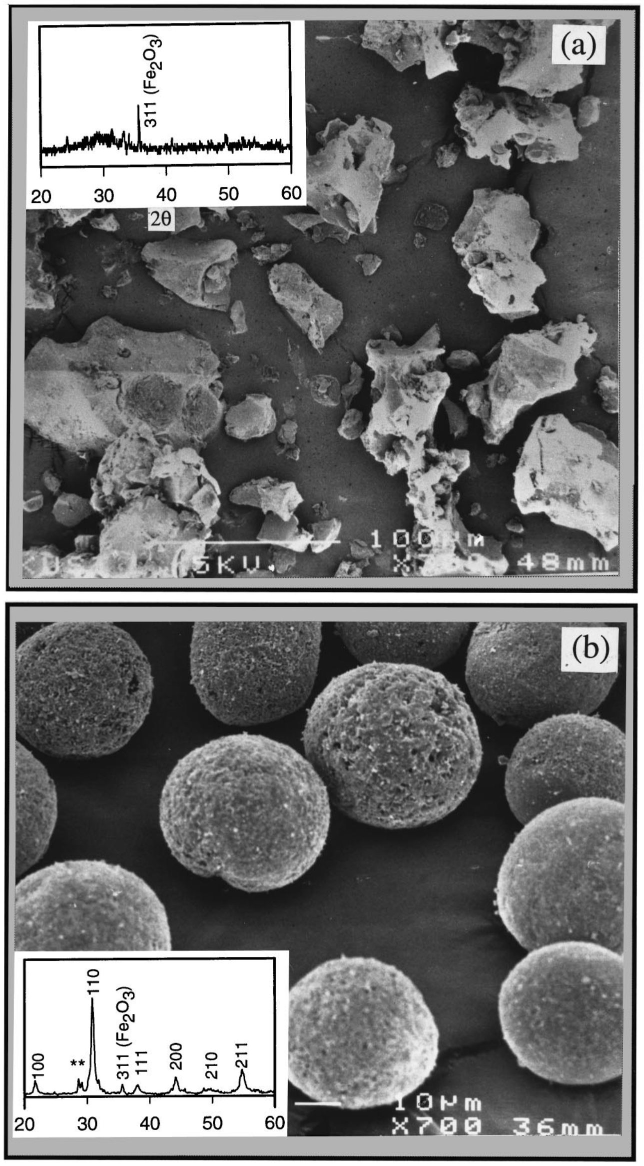

FIG. 2. SEM micrographs of MBPZT particles before (a) and after (b) flame treatment. Insets are the corresponding XRD results.

The ER fluids used in our experiments consisted of 45 ±5 μ-coated MBPZT microspheres dispersed in a silicon oil (DOW Corning No. 705). Both the oil and microspheres were heated at 100 °C for 5 h to remove any trace of water before testing. Figure 3 illustrates the suspension of the MBPZT microspheres under a magnetic field. The experiment was conducted as follows: A permanent magnet (with a vertical field and strength less than 1 G) mounted below the bottom plate of a $10\times10\times10\ \mathrm{cm}^3$ glass cell containing 0.1 volume fraction of MBPZT particles, was used to levitate the microspheres. Separated columns were clearly visible even under such a small magnetic field. The SEM micrograph as shown in Fig. 3 was obtained by removing the oil from the sample and depositing a layer of gold (through evaporation) onto the acetone-cleaned surface of the columns.

The setup for the static yield stress measurement is shown in the inset of Fig. 4. It consists of the MBPZT ER fluid sandwiched between two plates of a standard torsional device. The lower plate was driven by a motor with speed controlled by a computer. A small permanent magnet mounted below the bottom plate provided the magnetic field for particles suspension as described above, while a pair of Helmholtz coils was used to provide an additional vertical magnetic field. To start the measurement, the lower plate was rotated very slowly at 0.005 Hz, dragging the top plate, which was attached to a torque meter. The static yield stress was determined as the point where slipping just occurred between the two plates. The measured static yield stress of our samples under different field strengths is shown in Fig. 4. As can be seen from the plot, the static yield stress increases rapidly. At 1 kV/mm the ER strength reached over 2 kPa. However, the static yield stress saturated at fields over 2 kV/mm. At low fields, the static yield stress shows a linear E dependence, in contrast to the E^2 dependence observed in the

FIG. 3. SEM micrograph of columns of MBPZT microspheres suspended by a small magnetic field.

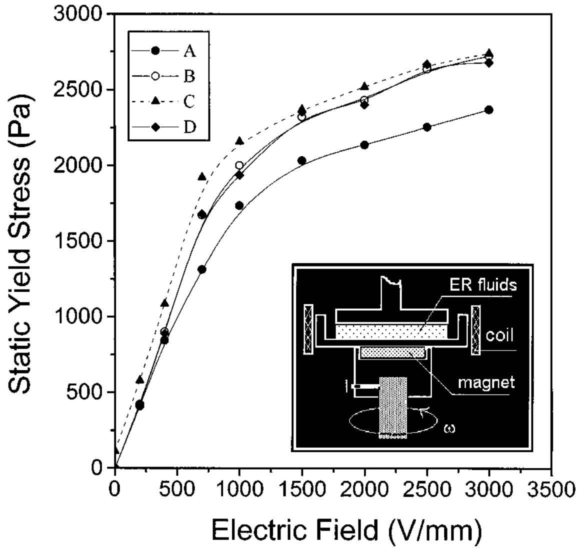

FIG. 4. Static yield stress of MBPZT ER fluids with different volume fraction ϕ vs electric field with (curve C) and without an additional magnetic field (curves A, B, and D) where A: $H=0$, $\phi=0.28$; B: $H=0$, $\phi=0.34$; C: $H=100$ G, $\phi=0.34$; and D: $H=0$, $\phi=0.34$. Curve D was obtained using sample B stored for a month in a bottle and redispersed before use. Inset shows the experimental setup.

usual ER systems. This may be explained by the fact that the origin of yield stress lies in the interaction between the polarization P of the microspheres. As a result, it is expected that the yield stress should be $\propto P^2$. In the usual case, $P \propto E$, so that yield stress $\propto E^2$. However, in the present case since the microspheres are ferroelectric, $P \propto E^{1/2}$ for PZT materials at low fields.[6,7] Consequently, the yield stress $\propto E$ for $E < 1$ kV/mm. Saturation of the yield stress is reached when the increase in P slows down at higher field strengths. The dashed line shown in Fig. 4 gives the static yield stress when an additional magnetic field was applied to the ER fluids in the same direction. Only a small enhancement is seen since the material is only weakly magnetic. It should be noted that the ER fluids made from MBPZT spheres are very stable, as no detectable change in the static yield stress was found a month later (see Fig. 4).

In summary, the fabrication of a type of ER material, denoted magnetic materials-based PZT microspheres, is introduced. The MBPZT microspheres can be suspended in oil with a very small magnetic field. The traditional problem of sedimentation in ER fluids is, thus, solved in this ER system. The MBPZT ER fluids are very stable and exhibit a strong ER effect at low electric-field strength.

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[1] K. O. Havelka and F. E. Filisko, *Progress in Electrorheology* (Plenum, New York, 1995).

[2] Electromagnetorheological fluid has been studied, for example, in T. Fujita, J. Mochizuki, and I. J. Lin, J. Magn. Magn. Mater. **122**, 29 (1993); K. Minagawa, T. Watanabe, K. Koyama, and M. Sasaki, Langmuir **10**, 3926 (1994); and K. Koyama, K. Minagawa, T. Watanabe, Y. Kumakura, and J. Takimoto, J. Non-Newtonian Fluid Mech. **58**, 195 (1995).

[3] G. Yi, Z. Wu, and M. Sayer, J. Appl. Phys. **64**, 2717 (1988).

[4] W. Wen and K. Lu, J. Cryst. Growth **165**, 191 (1996).

[5] W. Y. Tam, G. Yi, W. Wen, H. Ma, M. M. T. Loy, and P. Sheng, Phys. Rev. Lett. **78**, 2987 (1997).

[6] S. A. Mansour, G. L. Liedl, and R. W. Vest, J. Am. Ceram. Soc. **78**, 1617 (1995).

[7] Y. Xu, *Ferroelectric Materials and Their Applications* (Elsevier Science, Amsterdam, 1991), p. 151.