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Electrorheological behavior of kaolinite–polar liquid intercalation composites

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In this paper, two kaolinite hybrids have been synthesized using an intercalation method and characterized by XRD and FTIR. The result shows that DMSO (dimethyl sulfoxide) intercalated the interlayer of kaolinite and the basal spacing of hybrids was swelled from 0.715 to 1.110 nm, so that the kaolinite–organic matter intercalation compounds were formed. Then the dielectric and rheological properties of the ERF (electrorheological fluid) were measured. The results show that in electric fields of 3 kV mm^{-1}, the shear stress of DMSO/kaolinite ERFs can be 600 Pa (shear rate 5 s^{-1}) which is 2.14 times higher than that of pure kaolinite ERFs. In the meanwhile, the sedimentation and temperature effect is excellent. The intrinsic reason for enhancement of the rheological properties is the improvement of the dielectric properties of the intercalation hybrid. The properties of glycerol/kaolinite ERFs were enhanced also, but an optimum electrorheological effect zone appeared at a glycerol content of 6%.

Introduction

Electrorheological fluids (ERFs) have been regarded as a smart material, which could respond rapidly (in a few milliseconds), adjustably and reversibly to the external electric field, resulting in a sharp increase in viscosity.[1–3] But some shortcomings of the material, such as low shear stress, poor temperature stability, easy sedimentation and high cost, have led to much restriction of their industrial development.[4–6] Especially the market price of ERFs in Europe and America is over $800 per litre. Hence, researching the ERFs that have low cost, good properties and high cost performance was the key to electrorheological technology. Previously, studies on ERF materials were focussed on those of inorganic oxides and organic polymers. The inorganic oxide ERF materials, such as titania,[7] barium titanate,[8] zeolite[9,10] and alumisilicate,[11] have some shortcomings which include high density, high friction and low shear stress. Meanwhile, organic polymers, for example, polyaniline,[12,13] polyurethane,[14] poly(*p*-phenylene)[15] and poly(acene quinone),,[16] also have some limitations such as toxicity, *etc.* So recent study of ERFs materials was mainly focused on the organic–inorganic hybrids. Katayama adopts hybrid particles prepared by substituting the skeleton of an inorganic polymer comprising M–O–M bonds (M = metal or semi-metal) with O–[–Si–(R)$_2$–O–]$_n$ groups (R = organic group).[17] the *in-situ* preparation method was used by Edamura to prepare inorganic/organic composite particles. These were composed of a core of an inorganic polymeric compound and a shell of organic material particles; furthermore, the inorganic–organic composite particles are preferably produced by means of a method in which the core and the shell are formed simultaneously.[18] In order to enhance the properties of ERFs, a sol–gel method was used by our research team to prepare the ERFs of modified titania (TiO_2) doped with rare earth ions. In electric fields of 3 kV mm^{-1}, when a shear rate of 1.441 s^{-1} was applied to the ERFs at 25 °C the shear stress was 1.7 kPa. However, the preparation process is complex, the cost is high and the sedimentation is rapid.[19,20] At the same time, a new type of organic–inorganic hybrid material, made of carboxyl methyl starch (CMS) and titanium dioxide (TiO_2), was synthesized by means of a modified *in-situ* sol–gel technique. The suspensions of (CMS/TiO_2) hybrids in silicone oil display a remarkable ER effect, but the cost is still high and the CMS can be scorched easily.[21] Kaolinite is a hydrated aluminosiliate possessing the ideal composition $Al_2Si_2O_5(OH)_4$. Moreover, kaolinite is a 1 : 1 dioctahedral clay mineral whose structure is composed of interstratified $AlO_2(OH)_4$ octahedral sheets and SiO_4 tetrahedral sheets. Some examples of kaolinite intercalation with small polar molecules such as DMSO, potassium acetate and hydrazine are frequently reported in the literature.[22] Frost *et al.* reported that differences in the position and number of bands were observed for DMSO-intercalated low and high defect kaolinites by the use of a Raman microprobe, and that additional bands of DMSO were attributed to the different molecular structures of DMSO in the DMSO-intercalated kaolinites.[23] The kaolinite–organic matter intercalation material was prepared for ERFs by use of the kaolinite, which is in plentiful supply and at low cost. Owing to many advantages, such as process simplicity, low cost, good temperature stability and high anti-sedimentation, kaolinite may provide a new route for the preparation of ERF materials. Parameters in connection with particle polarization, *i.e.* dielectric constant, conductivity and dielectric loss, have been regarded as basic factors dominating the ER effect.[24–27] The choice of intercalationg kaolinite with a polar liquid is aimed at modifying the dielectric and polarization properties of kaolinite, so as to improve its ER activity.

To this end, using a combination of physical and chemistry designs,[20,21,28] polar liquid/kaolinite hybrids were prepared by means of an intercalation method. The hybrids were used as the particle phase and were mixed with silicone oil to prepare the ERFs. The dielectric properties of polar liquid/kaolinite were measured and the electrorheological behavior of the ERFs was studied.

1 Experimental

Kaolinite used in this study was from Shanghai, China. It was received as a finely divided pale powder of great purity, and possible phases of the kaolinite were examined by the XRD technique. The median particle size of the kaolinite was 2 μm.

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The kaolinite used to prepare the hybrid composites was used without further purification. Particles of polar liquid/kaolinite composites were prepared by the intercalation method of Zhong *et al.*[29] First the kaolinite must be dehydrated 2 h at 150 °C. Then the correct amount of DMSO or glycerol (such as 3.5%, 5.5% *etc.*, in order to compensate for the loss of polar liquid during the reaction) was added to the kaolinite and reacted at 80 °C for 20 h. The product was dried in a vacuum oven at 60 °C for a period of time (normally 3–4 h, when the product was dried, the hybrids were weighed at the same time to ensure the right amount of polar liquid) to obtain a composite of the correct DMSO or glycerol content (such as 2%, 6%, *etc.*). The particles were then milled.

Firstly silicone oil was dried at 150 °C for 2 h, and then ER fluids were prepared by dispersing the hybrid particles (at 30 wt%) in dimethyl silicone oil ($\varepsilon_f \approx 7.6$–$7.8$, $\sigma \approx 10^{-12}$–10^{-13} S m^{-1}, $\rho \approx 0.9$–1.0 g cm^{-3}, $\eta = 500$ mPa.s, 25 °C).[30]

A rotary viscometer (NXS-11A; the gap between the outer cup and the inner bob is 2 mm; China) and high-voltage DC power source (GYW-0/0; China) were used to research the rheological properties of ER fluids. A rotational viscometer equipped with a thermal instrument was used to measure the temperature dependence of shear stress (the thermal range was from 0 to 120 °C, and the thermal rate was *ca.* 1 °C min^{-1}).

2 Results and discussion

2.1 X-Ray characterization

Fig. 1(a), (b) and (c) are the X-ray diffraction (D/IIIX-γA, CuKα, 45 kV/80 mA; Japan) patterns of kaolinite, kaolinite/DMSO hybrids, and kaolinite/glycerol hybrids, respectively. The result shows that the peak of 7.15 Å (d_{001}) of kaolinite disappears completely after kaolinite is reacted with DMSO, and that a new peaks at 11.10 Å is observed. This result indicates that DMSO is intercalated into the interlayer space of kaolinite and that the kaolinite–DMSO composite is formed. In the meantime the peak of 7.15 Å of kaolinite was transformed into the peak of 7.17 Å after the reaction of kaolinite with glycerol, hence the interlayer space was enlarged by 0.02 Å. At the same time, the diffraction intensity of 7.17 Å is decreased by over 50%. This result shows that glycerol is not intercalated into the interlayer space of kaolinite and it may be simply adsorbed on the edges.

2.2 FTIR characterization

Fig. 2 shows the FTIR spectra (Nicolet 60 SXR FTIR spectrometer; Germany) of the kaolinite–DMSO, kaolinite–glycerol, and pure kaolinite particles. The IR absorption bands of kaolinite at 3694, 3669, 3654, and 3621 cm^{-1} are attributed to O–H stretching vibrations, those at 1114, 1032, 913 cm^{-1} to Si–O stretching vibrations. The 3694, 3669, and 3654 cm^{-1} bands are O–H stretching vibration bands of the interlayer surface of layered silicates and 3621 cm^{-1} band is the O–H stretching vibration of the internal structure of layered silicates. When the intercalation composites of DMSO/kaolinite were formed, the 1429 cm^{-1} vibration band is attributed to the CH_3 stretching vibrations and the 1037 cm^{-1} vibration band is due to the S=O stretching vibrations. But the 3694 cm^{-1} peak diminished notably, the 3654 cm^{-1} peak disappeared and the intensity of the 3669 cm^{-1} peak increased remarkably. In addition, the 1114 cm^{-1} peak of the Si–O stretching vibration is divided into peaks at 1123 and 1101 cm^{-1}, and it is noteworthy that new peaks of 3539 and 3504 cm^{-1} were formed; the results show that the O–H bands of the interlayer surface were disturbed so the peaks moved to lower wavelength. The hydroxy deformation of DMSO–kaolinite hybrids was characterized by one intense band in the FTIR at 905 cm^{-1}; this result was in accord with the research of Frost *et al.*[31] So DMSO is indeed intercalated into the kaolinite interlayer space. Meanwhile the IR absorption bands of the O–H stretching vibration for glycerol/kaolinite particles were slightly varied; for example, the 3654 cm^{-1} peak was increased, but the 3694 and 3669 cm^{-1} peaks were diminished. The broad peak of 3390 cm^{-1} is an O–H stretching vibration of glycerol, yet no new peaks were seen at *ca.* 3550–3500 cm^{-1}. Hence, the glycerol molecules were reacted partly with kaolinite and it is simply adsorbed on the edges of kaolinite.[32]

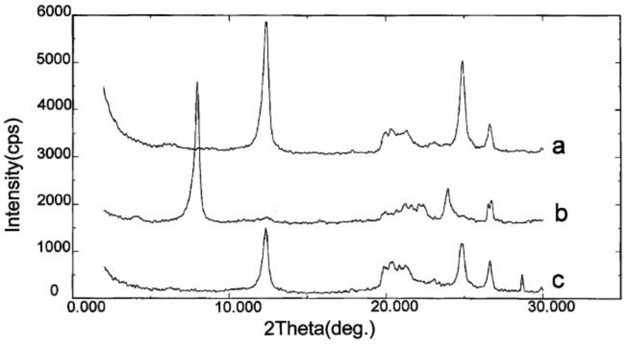

Fig. 1 XRD curves for (a) kaolinite, (b) kaolinite/DMSO, (c) kaolinite/glycerol.

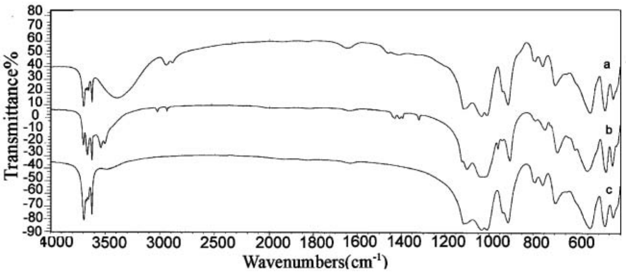

Fig. 2 FTIR spectra of (a) kaolinite/glycerol, (b) kaolinite/DMSO, (c) kaolinite.

2.3 Dielectric properties of kaolinite hybrids

The dielectric properties of the ERFs were measured using an automatic LCR meter (WK-4225; Germany) at frequencies of 100, 1 k and 10 kHz. For the ER fluid of different DMSO contents intercalating kaolinite, the dielectric constant (ε), conductivity (σ) and dielectric loss (tgδ) were measured as shown respectively in Fig. 3, 4 and 5. From these figures it can be seen that ε, σ and tgδ were increased by different degrees with the increase of DMSO content. As is shown in Fig. 3, the dielectric constant of 14% DMSO/kaolinite ER fluid is 1.72 times than that of the pure kaolinite ER fluid (100 Hz, 25 °C). In Fig. 4, the conductivity of same content DMSO/kaolinite ER fluid is 2.6 times than that of the pure kaolinite ER fluid (10 kHz, 25 °C). In Fig. 5, dielectric loss of the same hybrid ER fluid is 2.59 times than that of the pure kaolinlite ERF (1 kHz, 25 °C). These results show that the dielectric properties of the

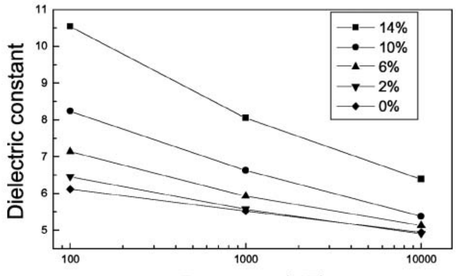

Fig. 3 Dielectric constant *vs.* frequency for DMSO/kaolinite ERFs.

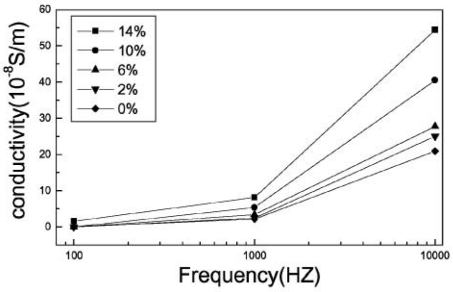

Fig. 4 Conductivity *vs.* frequency for DMSO/kaolinite ERFs.

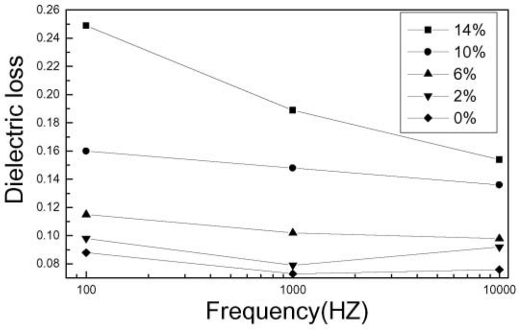

Fig. 5 Dielectric loss *vs.* frequency for DMSO/kaolinite ERFs.

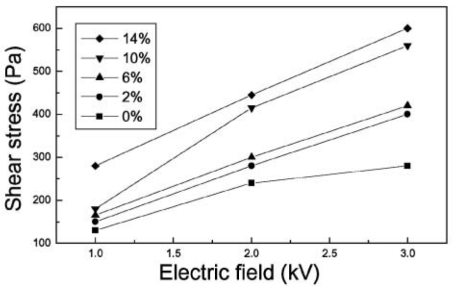

Fig. 6 Dependence of the shear stress *vs.* electric field for DMSO/kaolinite ERFs.

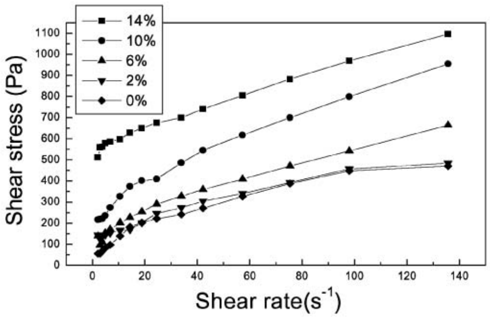

Fig. 7 Dependence of shear stress *vs.* shear rate for DMSO/kaolinite ERFs (3 kV mm^{-1}).

DMSO/kaolinite ER fluid at high a intercalation ratio were increased markedly compared with the pure kaolinite ER fluid. From the dielectric properties it can be seen that the dielectric constant decreased quickly, but that the conductivity was increased rapidly with an increase in frequency. Owing to interfacial polarization, the dielectric constant at low frequency is usually large, but with the emergence of loss peak at critical frequency, ε values at high frequency were completely declined.[33,34] It is well known that a high dielectric constant and dielectric loss, proper conductivity is the physical base to obtain the critical optimum electrorheological effect.[35] In this paper, the dielectric constant and dielectric loss of the DMSO/kaolinite ER fluid were both increased notably and the conductivity remained at 10^{-8}–10^{-7} S m^{-1}. These properties are customary for the electrorheological effect.

At the same time, the dielectric constant, conductivity and dielectric loss of glycerol/kaolinite hybrid ER fluids with various glycerol contents were also measured (not shown). With an increase inglycerol content, the dielectric properties were increased to a different degree. The dielectric constant of the 10% glycerol/kaolinite composite ER fluid ($\varepsilon = 7.958$) is 1.3 times of that of the pure kaolinite ER fluid (100 Hz, 25 °C, $\varepsilon = 6.118$). The conductivity of the same glycerol content kaolinite hybrid ER fluid ($\sigma = 3.789 \times 10^{-7}$ S m^{-1}) is 1.93 times of that of the pure kaolinite ER fluid (10 kHz, 25 °C, $\sigma = 2.095 \times 10^{-7}$ S m^{-1}). Its dielectric loss ($\mathrm{tg}\delta = 0.120$) is 1.64 times that of the pure kaolinite ER fluid (1 kHz, 25 °C, $\mathrm{tg}\delta = 0.073$). Therefore, it can be seen that for high glycerol content hybrids, the ER fluids have higher dielectric properties than the pure kaolinite ER fluid.

2.4 Rheological properties of kaolinite composite ER fluids

The DMSO/kaolinite hybrid particles were milled to a fine powder and then mixed with dimethyl silicone oil (30wt%). Fig. 6 shows the shear stress of different DMSO content kaolinite composite ER fluids with an increase of DC electric field under a fixed shear rate (5 s^{-1}, with a parallel-plate force transducer). It can be seen from the curves that the shear stress of hybrid ERFs at high DMSO contents are much higher than that of a pure kaolinite ERF. The shear stress is above 600 Pa (14% DMSO content) under a 3 kV mm^{-1} DC field at 20 °C, with leaking current density limited to less than 20 μA cm^{-2}. However, the shear stress of the pure kaolinite ERF is only 280 Pa, which is 7/15 of that of the hybrid ERF. This result shows that the shear stress of ERFs for high DMSO content kaolinite hybrids was increased notably over that of the pure kaolinite ERF.

The shear stresses of different DMSO content kaolinite ERFs *versus* a change in shear rate were measured using a rotary viscometer under an applied electric field (*ca.* 0–3 kV mm^{-1}); here, only the 3 kV mm^{-1} field results are shown (see Fig. 7). It can be seen that the ERFs behave as a Bingham fluid immediately and show notable electrorheological effects when a DC electric field was applied.[36] The widely accepted ER mechanisms suggest that particle polarization is responsible for the interaction forces between particles which leads to rheological change in the ERFs. The parameters in connection with particle polarization, such as high dielectric constant, dielectric loss and suitable conductivity, play an important role for high performance ER materials. The dielectric properties of DMSO/kaolinite hybrid ERFs were improved by the intercalation of DMSO compared with pure kaolinite ERFs, so the shear stress of DMSO/kaolinite hybrid ERFs was increased notably.

The shear stresses of different glycerol content kaolinite ERFs with a change of the electric field under a fixed shear rate (5 s^{-1}, with a parallel-plate force transducer) were also measured. The shear stress is above 560 Pa for the 6% glycerol content hybrid ERFs. On the contrary, the shear stress of the pure kaolinite ERF is only 280 Pa. The former is twice the latter.

The shear stresses of different glycerol content kaolinite ERFs with a change of shear rate were measured using a rotary viscometer under a DC electric field (*ca.* 0–3 kV mm^{-1}). The electrorheological effects under the DC electric field were also observed. Over the rangel of glycerol contents used, the optimum electrorheological effect was noted at the 6% glycerol/kaolinite ERF. Increasing the glycerol content above 6% (such as 10%) resulted in a decreased electrorheological effect due to the increased leaking current of the ERFs. This result can also be seen in Fig. 8.

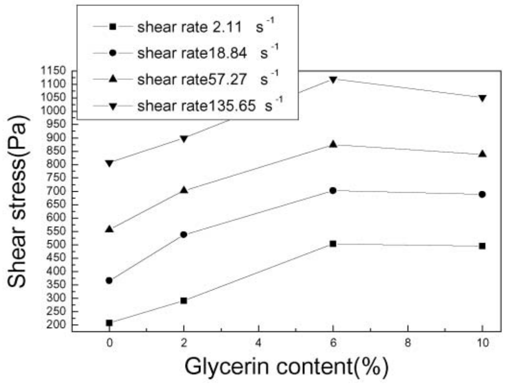

Fig. 8 Shear stress *vs.* glycerol content (3 kV mm^{-1}).

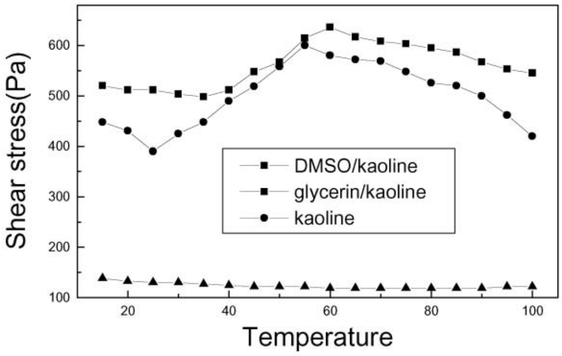

Fig. 9 Temperature dependence of kaolinite hybrid ERFs.

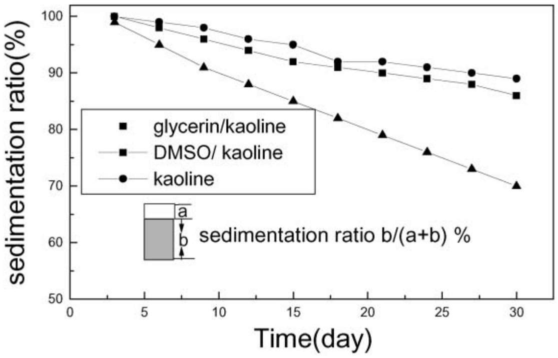

Fig. 10 ERF sedimentation ratio for glycerol/kaolinite, DMSO/kaolinite and kaolinite *vs.* time.

2.5 Temperature effects of kaolinite intercalation hybrid ERFs

The temperature dependence of kaolinite/DMSO hybrid ERFs, as shown in Fig. 9, shows obvious optimization. The operating temperature was measured over the range of 10–100 °C. The shear stress of kaolinite/DMSO hybrid ERFs increases with an increase of temperature at first, then at a critical temperature it starts to drop. Its maximum stress appears at around 60 °C, and remains at a siginificant value even at 90 °C. As the temperature is increased, the polarization strength is enhanced. However, with the increasing temperature, the polarization rate becomes faster and the polarization stability becomes poorer. When the above two factors reach an equilibrium at the temperature, the field-induced shear stress reaches its maximum accordingly.

Fig. 9 also shows the temperature dependence of kaolinite/glycerol hybrid ERFs. It can be seen that there is a peak in the curve, where its maximum stress appears at *ca.* 55 °C. In the meantime, with the enhancement of temperature, the leaking current density is also increased. The reason for this is that the surface conductivity, thermal ionic conductivity and thermal electronic launching current are increased with the increase in temperature which leads to the enhancement of electric conduction. However, with the variation of temperature the shear stress of the pure kaolinite ERFs was changed only slightly and declined slowly with an increase of temperature.[37]

2.6 Sedimentation properties of kaolinite hybrid ERFs

The DMSO/kaolinite hybrids ERFs, as is shown in Fig. 10, have excellent sedimentation stability. Because kaolinite clay is a good suspension stabilizer due to its lamellar and open structure[38–40] its density was decreased for the DMSO intercalation effect; moreover, through milling the hybrid diameter was also decreased. From Fig. 10, we can see that the sedimentation ratio of DMSO/kaolinite ERFs is above 85% after 30 days and that the sedimentation stability is increased notably over that of the pure kaolinite ERF. Furthermore, the glycerol/kaolinite hybrid ERFs also have the same good sedimentation stability, which to a certain degree is due to the viscosity of glycerol (20 °C, η = 1069 mPa.s).

2.7 Comparison between the two kinds of hybrids

As mentioned above we can see that differences exist between the two kinds of hybrids. Firstly, from XRD curves the basal spacing of the DMSO/kaolinite hybrids (d_{001}) was swelled to 1.110 nm, compared with 0.717 nm of that of the glycerol/kaolinite hybrids. This shows that the intercalation effect of glycerol/kaolinite hybrids was not observed. Secondly, the dielectric properties of DMSO/kaolinite hybrids are superior to those of glycerol/kaolinite hybrids. Finally, the electrorheological effect is increased for the DMSO/kaolinite hybrids of increasing DMSO content from 0 to 14%, but the glycerol/kaolinite ERFs have an optimum electrorheological effect at *ca.* 6% glycerol content and the effect is decreased at a 10% glycerol content. In the meantime the mechanical properties of DMSO/kaolinite hybrid ERFs are also superior to those of glycerol/kaolinite hybrid ERFs.

From Table 1, we can see that the polarity of DMSO is higher than that of glycerol, so the hydrogen bond is formed more easily between DMSO and the interlayer of kaolinite. On the contrary, the hydrogen bond for glycerol was difficult to form. Owing to the higher dielectric constant and conductivity of DMSO, the dielectric properties of the DMSO/kaolinite hybrid were improved notably. Finally the molecules of glycerol were unfavorable for hydrogen bonding with kaolinite due to the viscosity of glycerol. So it is mainly physical absorption and mechanical adhesion within the glycerol/kaolinite hybrids, and the conductivity mechanism of charge carrying in the glycerol/kaolinite hybrid ERFs was different from that of the DMSO/kaolinite ERFs. This point can be highlighted in that the leaking current density of glycerol/kaolinite ERFs is over 30 μA cm^{-2} at DC 3 kV mm^{-1} for 10% glycerol

Table 1 Physical properties of polar liquids (25 °C)

Compound	Dielectric constant, ε	Conductivity, δ/S m^{-1}	Dipole moment/D	Viscosity/mPa.s	Boiling point/°C
DMSO	45	3.0×10^{-6}	3.9	7.20	189
Glycerol	42.5	6.4×10^{-8}	2	1069	290

content, but under the same conditions the leaking current density of the DMSO/kaolinite ERFs is no more than 6 μA cm^{-2}.

3 Conclusions

A direct intercalating method was used to prepare the polar liquid/kaolinite hybrids, such that the kaolinite–organic matter intercalation materials were formed, and then the composite particles were mixed with silicone oil to prepare the ERFs. The experiments show that the properties of kaolinite/DMSO ERFs were enhanced notably and that the shear stress of the kaolinite/DMSO ERFs was 2.14 times higher than that of the pure kaolinite ERF; also, this material exhibits good temperature stability and sedimentation properties. The properties of glycerol/kaolinite ERFs were also enhanced, but an optimum electrorheological effect zone appeared for glycerol content of 6%. All of these results can provide a new way for the preparation of ERF materials with a good cost performance.

Acknowledgements

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