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# The evaporation of silicone oil in electrorheological fluids

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## Abstract

A study on the evaporation of electrorheological (ER) fluids consisting of CTO nanoparticles and silicone oil is performed. The serious evaporation observed in giant ER fluids is mainly due to the small size of particles contained. The weight losses of the ER fluids under different experimental conditions were measured and the systematic results on the relationships of type of silicone oil, weight fraction of particles, surface area and depth of samples were obtained. Those evaporating phenomena have been explained mainly based on the Kelvin equation. The understanding on the behaviors of evaporation in ER fluids should be beneficial for applying and storing the ER fluids.

## 1. Introduction

Electrorheological (ER) fluid consists of dielectric particles and insulating liquid, of which the yield stress is controllable by applying an external electric field and the state can be transformed from liquid-like to solid-like reversibly [1–4]. In the last decade researchers have devoted efforts to enhance the shear stress of ER fluids by preparing nanoparticles with polar molecules adsorbed. Several kinds of ER fluids with high yield stress (even over 200 kPa at 5 kV mm<sup>-1</sup>) have been fabricated in the systems containing TiO<sub>2</sub>, Ca–Ti–O (CTO) or Sr–Ti–O (STO), etc, nanoparticles, referred to as giant ER fluids or polar molecule dominating ER fluids (PM-ER) [5–11]. This outstanding achievement will hopefully bring ER fluids to a bright application foreground. However, in both manufacture and applications the stability of ER fluids is one of the important issues we must pay attention to. Many insulating liquids such as silicone oil (SO), mineral oil and hydrocarbon oil can be chosen as dispersing media for preparing ER fluids. The mostly used one is SO because of its permanently stable properties. A problem often met is that the prepared giant ER fluids will become thicker and even completely dried if they are exposed to air after a period of time due to the evaporation of oil. Also it can be observed

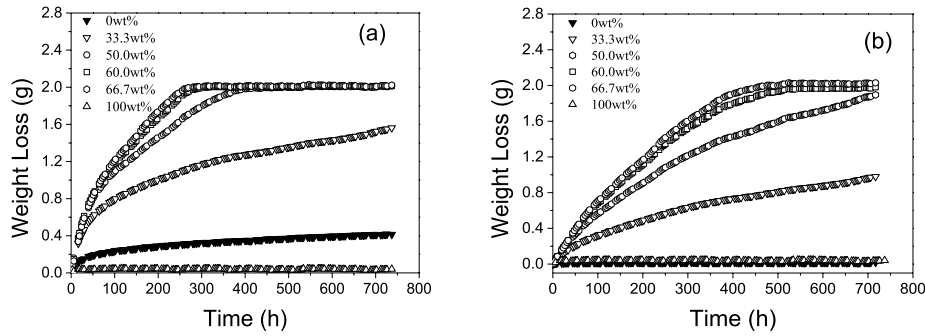
that the oil is seriously evaporated when the ER fluid is contained in an incompletely sealed vessel kept in a dryer for several months. A better comprehension of such evaporation is needed for the preparation, storage and application of ER fluids. Although a preliminary study on the phenomenon of nanoparticle enhanced evaporation in suspensions has been performed [12], a systematic investigation on the evaporation of SO in ER fluids is still absent.

In this paper, we present the measured results on the evaporation of SO in ER fluids under different conditions. The relationships of evaporation rate versus different factors, such as the viscosity of SO, concentration of particles, surface area and depth of samples, are given. The observed phenomena can be explained qualitatively with the Kelvin effect.

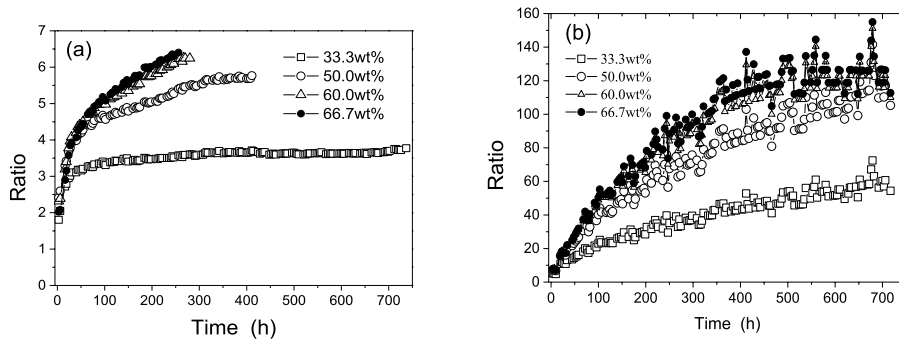
## 2. Experimental details

Ca–Ti–O (CTO) nanoparticles were synthesized with the co-precipitation technique [7]. The size of the CTO particles is in the range of 50–100 nm measured with a granulometer (Zetasizer Nano-ZS, Malvern Instruments Ltd, UK) and the density is 2.089 g cm<sup>-3</sup> measured with a pycnometer (AccuPyc II 1340 Pycnometer). Two different SO (Beijing Huaer Co. Ltd) were used as dispersing phases of which the kinetic viscosities were 10 mm<sup>2</sup> s<sup>-1</sup> (10<sup>#</sup>) and 200 mm<sup>2</sup> s<sup>-1</sup> (200<sup>#</sup>) respectively. CTO particles with different weights were

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**Figure 1.** The weight loss of SO versus time in ER fluids with different CTO weight fractions. (a) For samples mixed with 10<sup>#</sup> SO (2g), (b) For samples mixed with 200<sup>#</sup> SO (2g).



**Figure 2.** Relative weight loss in the samples of CTO/10<sup>#</sup> SO (a) and CTO/200<sup>#</sup>SO (b).

mixed with 2g SO and then grinded sufficiently to form homogeneous suspensions. The CTO/SO suspension shows good giant ER effect [7]. The suspensions with different weight percentages of CTO particles were respectively filled in glass dishes of 35 mm diameter and 25 mm in height, or filled in glass dishes with different diameters for various experimental targets. All the samples were put in a drying oven at 80 °C for 30 days. The temperature was set at 80 °C instead of room temperature because the evaporation rate of SO at room temperature is very low and hard to measure. In order to measure the dependence of surface area on the evaporation, glass dishes with different diameters were utilized, in which the thickness of filled samples were all 2 mm. By measuring the weight loss of the samples with an electronic balance (Shimadzu Co. Japan, 10<sup>-4</sup> g resolution) during the period, the relationships of evaporation of SO versus time for all samples were obtained.

### 3. Results and discussions

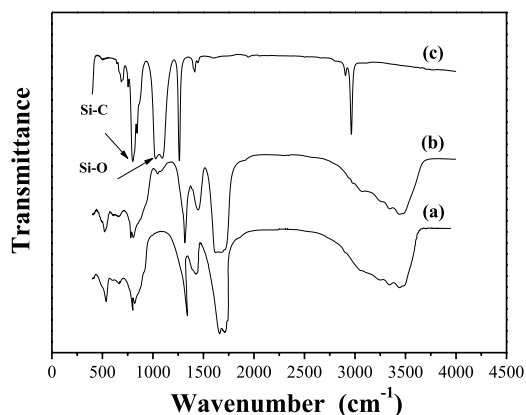
#### 3.1. The influence of CTO weight percentage on the evaporation rate of SO

Figure 1 shows weight loss of 10<sup>#</sup> and 200<sup>#</sup> SO under 80 °C in different samples, of which the weight percentages of CTO particles are  $\phi_w = 0, 33.3, 50.0, 60.0, 66.7, 100\%$  respectively. It can be seen from figure 1(a) that in 700 h the weight loss for pure 10<sup>#</sup> SO ( $\phi_w = 0$ ) of 2g is about 20%, while the weight of pure CTO particles ( $\phi_w = 100\%$ ) remains

unchanged. However, the evaporation rate of SO in other samples enhances with  $\phi_w$  increasing during a same period. In the sample of  $\phi_w = 33.3\%$ , 75% of SO is lost in 700 h. For the samples with  $\phi_w = 50\%$  and  $\phi_w = 60\%$ ,  $\phi_w = 66.7\%$ , all the SO is evaporated in about 400 h and 250 h respectively, and those samples all then become dry powder.

Figure 1(b) shows a same tendency for the samples of CTO mixed with 200<sup>#</sup> SO, but the time for losing all 200<sup>#</sup> silicone in the samples of  $\phi_w = 60\%$  and  $\phi_w = 66.7\%$  is about 500 h, which is almost twice as much as in the samples with 10<sup>#</sup> SO mixed. On the other hand, the pure 200<sup>#</sup> SO has hardly evaporated at 700 h, thus the evaporation rate is markedly lower than that of 10<sup>#</sup> SO. This is due to the smaller mean molecule size and thus lower vapor pressure of 10<sup>#</sup> SO.

However, it is observed that the ratio of weight loss of CTO/200<sup>#</sup>SO divided by that of 200<sup>#</sup>SO alone is larger than the ratio in the samples with 10<sup>#</sup>SO mixed. Figures 2(a) and (b) illustrate the effect of CTO particles on the evaporation of oil in the samples of CTO/SO. The vertical axis shows the relative weight loss, i.e. the ratio of the weight loss of CTO/SO suspensions divided by that of pure SO. Obviously, the higher particle fraction in the samples can cause more SO to be evaporated although the weights of SO contained in all samples are same as 2g. On the other hand, the evaporation of 200<sup>#</sup>SO in the samples of CTO/200<sup>#</sup>SO is more enhanced by adding CTO particles compared to that in CTO/10<sup>#</sup>SO. The former is about an order of magnitude larger than the latter. The reason that adding CTO particles can cause the evaporation in CTO/200<sup>#</sup>SO to be more effective than in CTO/10<sup>#</sup>SO will be discussed later.



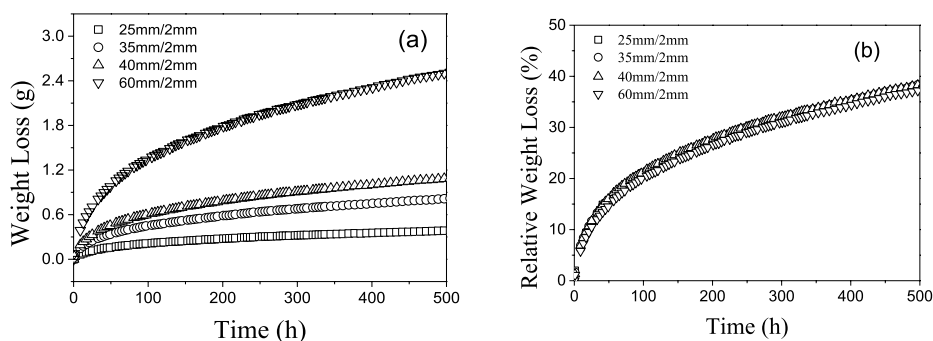
**Figure 3.** Infrared spectra of CTO nanoparticles (a); dried CTO ER fluids (b); pure SO (c).

It can also be seen from figure 1 that the evaporation rate in the samples is rapid in the beginning and then slows down until all the oil has completely evaporated. It is well known that SO is a mixture of polydimethylsilane (PDMS) of different chain length. The short-chain molecules find it easier to escape from the liquid phase than the long-chain ones. In the earlier time the smaller SO molecules in the samples are evaporated easily. The mean chain lengths of the remaining SO molecules as well as their mean molecule weight become larger gradually, hence the evaporation rate must decrease with time. The rare mass loss of CTO powder in the period was neglected in the measurements.

When 2g SO in samples are evaporated as seen in figure 1, the remaining dried CTO powder does not contain any SO. The infrared spectra shown in figure 3 have confirmed that the Si-C and Si-O peaks from SO have disappeared in the dried samples and those samples are as same as the pure CTO powder.

### 3.2. The influence of liquid–vapor interface area on the evaporation rate of SO

The measured evaporation rates of silicone oil in ER fluids for various areas of liquid–vapor interface are illustrated in figure 4. The samples of CTO/10<sup>#</sup>SO (50 wt%) were placed



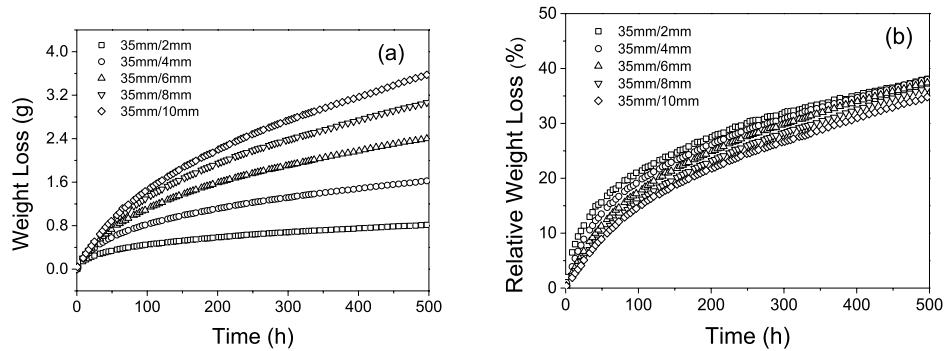
**Figure 4.** Weight loss (a) and relative weight loss (b) of CTO/10<sup>#</sup>SO ER fluids (50 wt%) in glass dishes with different diameters (25, 35, 40, 60 mm) and same depth of 2 mm.

in glass dishes of 25 mm, 35 mm, 40 mm, 60 mm in diameters respectively with same thickness of 2 mm. It can be seen from figure 4(a) that the weight losses of samples increase with the increase of surface area. Figure 4(b) plots the relative weight losses of samples (measured weight loss divided by the total weight of 10<sup>#</sup>SO). The fact that all curves almost overlap in figure 4(b) indicates that the evaporation of SO per unit surface area for all samples with the same particle fraction is the same and the weight loss of oil is nearly proportional to the area of free surface of glass dishes. These results demonstrate that the dependence of the evaporation in ER fluids on the surface area is the same as in common liquids, even with nanoparticles added.

### 3.3. The influence of sample depth on the evaporation rate of SO

Figure 5(a) shows the weight loss of CTO/10<sup>#</sup>SO ER fluids with a same weight fraction (50 wt%) and same liquid–vapor interface area but the depths of samples are different in glass dishes (35 mm in diameter). The depths of samples are 2 mm, 4 mm, 6 mm, 8 mm and 10 mm. The larger weight loss for deeper sample is due to larger weight of oil in the sample. Figure 5(b) shows the relative weight losses of samples, calculated with the lost weight of each sample divided by its total SO weight. It is obvious that the relative weight loss of SO depends on the depth of sample, i.e. more SO evaporates in a shallower sample. The diffusion process of the silicone molecules in the samples causes this effect. For deeper samples the molecules of oil in the fluids need more time to move to the surface through Brown motion and then evaporate, while the molecules in shallow samples find it easy to reach the surface.

All the results observed above demonstrate the evaporation of SO in the ER fluids at a certain temperature mainly determined by four factors: the vapor pressure of oil, concentration of particles, the area of free surface and the depth of the sample. Now we are going to analyze the physical origins of all those behaviors. In ER suspensions partial nanoparticles locate at the interface between liquid and vapor phase. At the surface the silicone oil adsorbed on the particles forms a lot of convex liquid surface with small diameter, of which the curvature is the same as that of



**Figure 5.** (a) Weight losses of 10<sup>#</sup> silicone oil in ER samples (50 wt%) with same surface area (35 mm in diameter) but different depths. (b) Relative weight loss of 10<sup>#</sup> silicone oil in ER samples with different depths.

the particles. According to the Kelvin equation principle, the saturated vapor pressure of SO on the convex liquid surface is much higher than that at the normal liquid surface. The Kelvin equation is expressed as [13]

$$\ln \frac{p}{p_0} = \frac{2\gamma M}{RT r \rho}, \quad (1)$$

where  $P$  is the vapor pressure for a curved surface,  $P_0$  is the vapor pressure for a flat surface,  $\gamma$  is the surface tension,  $M$  and  $\rho$  are the molar mass and mass density of oil,  $R$  is the universal gas constant,  $r$  is the radius of the droplet, and  $T$  is temperature. Therefore, that the SO in ER fluid is lost faster than in the pure one is due to the protuberances caused by CTO particles at the surface. We can estimate the difference of vapor pressure for different oils with equation (1). The mean molecule weights of 10<sup>#</sup> silicone oil and 200<sup>#</sup> silicone oil are 1432 and 11 896 respectively [14], while their surface tensions and densities are nearly the same. For a protuberance with the same curvature at the surface, the enhancement of vapor pressure of 10<sup>#</sup> SO is several orders of magnitude lower than that of 200<sup>#</sup> SO. This can explain why the evaporation rate enhancement of SO in CTO/200<sup>#</sup>SO is much higher than that in CTO/10<sup>#</sup>SO as seen in figures 1 and 2. With increasing concentration of CTO particles, more protuberances emerge at the liquid–vapor interface and cause the evaporation rate to increase.

On the other hand, molecular sizes in the same SO are different, and the smaller molecules find it easier to evaporate. As time goes on, smaller molecules at the surface are evaporated and the size of molecules remaining in the SO gradually increases. The evaporation rate of the oil must decrease with time. Consequently, the weight losses of silicone oil in all samples are faster at the beginning and become slower with the passage of time.

From the behaviors of evaporation of silicone oil in ER fluids described above, it is known that the evaporation rate of oil at a certain temperature mainly depends on the size and fraction of particles, the molecular mass of oil and the free surface of sample. Smaller size and higher concentration of particles can significantly enhance the vapor pressure of oil at the surface. The higher vapor pressure must induce higher evaporation rate at the surface of ER fluids. In giant ER fluids the size of particles ( $\sim 100$  nm) is usually much smaller than

that ( $\sim 10$   $\mu$ m) in conventional dielectric ER fluids, therefore the vapor pressure of oil at the surface of giant ER fluids is orders of magnitude higher than that in conventional ER fluids. Hence the evaporation rate of oil in giant ER fluids is remarkable. On the other hand, by using an oil with smaller molecules the evaporation of oil can be reduced. For instance, mineral oil is one choice, according to our experience, for which the evaporation in giant ER fluids is much less than that of silicone oil. Nevertheless, to avoid the evaporation of oil, the giant ER fluids should be put in a well sealed container in the measuring process or for storage.

#### 4. Conclusions

The evaporation behavior of silicone oil in CTO/SO ER fluids and the effective factors on the evaporation have been systematically studied. The results reveal that the evaporation of oil in giant ER fluids is a problem worth noting. Smaller size and high concentration of particles as well as larger size of molecule of oil can enhance the evaporation rate. The observed phenomena can be well explained qualitatively with the Kelvin rule. The study provides a better understanding and methods for reducing the evaporation in applications and storage of giant ER fluids.

#### Acknowledgments

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