

## 電氣流變流體의 粘彈性 特性에 관한 研究

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### Viscoelastic properties of electrorheological fluids

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**요 약** 전기유변유체(electrorheological fluid : ERF)는 크기가  $10^{-6}$ m 정도를 가진 분극성이 강한 입자 분말을 유전유체속에 혼합하여 만든 콜로이드용액으로서 외부전기장에 의하여 액체상태로부터 겔(gel)상태로 상태변환을 한다. 상태변환과정은 가역과정이며  $10^{-3}$ 초 정도의 빠른 시간내에 이루어진다. 본 논문에서는 주기적응력이나 주기적 변형이 가해지는 강제진동실험을 통하여 ERF의 점탄성 특성을 조사하였다. 점탄성은 복소수로서 실수부인 동적전단탄성율(storage shear modulus,  $G'$ )과 허수부인 동적손실율(loss modulus,  $G''$ )의 합으로 표현된다. 동적전단탄성율은 강제진동의 진폭과 주파수의 함수로 외부전기장에 의하여 크게 변화되었으나 손실계수(loss factor,  $\eta = G''/G'$ )는 진동의 진폭, 진동수 및 외부전기장에 거의 독립적이었다. 실험(1)은 강제진동의 진폭을 고정하고 진동수를 변화하는 방법(frequency sweep)과 실험(2)는 진동 진폭을 변화시키는 방법(amplitude sweep)으로 실행했다. 본 실험에서는 cornstarch입자를 corn oil에 혼합한 것과 zeolite입자를 silicone oil에 혼합한 콜로이드 용액이 ERF로서 사용되었으며 인가된 전기장은 약  $(1\sim 3) \times 10^6$ V/m의 직류전기장이 가하여졌다.

**Abstract** Electrorheological(ER) fluid's storage shear modulus( $G'$ ) and loss factor( $\eta$ ) have been directly measured using small amplitude forced oscillating rheometer as a function of oscillating frequency, strain amplitude and applied electric field. Two types of experiment were performed ; (a) frequency sweep and (b) amplitude sweep. Two kinds of sample were employed for this experiment ; cornstarch particles in corn oil and zeolite particles in silicone oil. The storage shear modulus was a strong function of driving frequency. Generally, the modulus increased with driving frequency. On the other hand, the loss factor was not well behaved as storage modulus, but as the driving frequency increases the loss factor slightly decreases was the trend of the material's characteristics. Also the modulus was a strong function of strain amplitude. Generally, modulus decreased with increasing strain, but loss factor increases slightly with increasing strain amplitude. For  $G'$ , cornstarch in corn oil ER fluid has higher values than zeolite based fluid as we increased applied electric field. On the other hand, zeolite based fluid has higher values for  $\eta$ . There is a reasonable agreement between theoretical calculation and experiment.

### 1. Introduction

The fact that suspensions of hydrophilic particles(e. g. cornstarch or activated silica) in low viscosity dielectric oils exhibit a marked reversible increase in apparent viscosity on application of an electric field of a  $10^6$ V/m( $1\text{kV}/\text{mm}$ ) was first reported by Winslow<sup>1)</sup>. This

phenomena can be attributed to electric field induced fiberization of the particles to the field direction, thereby requiring an additional shear stress for flow. This kind of fluid is called electrorheological(ER) fluid in order to distinguish it from electroviscous effect since the material property itself changes due to the formation of structure of the oriented particles

rather than viscosity change of fluid. Recently, many attempts have been made to apply the ER fluid to various industrial fields utilizing its quick response and easy control characteristics.

Selecting an ER fluid for a particular application requires an accurate knowledge of its dynamic mechanical behavior. In addition to the shear stress, viscoelastic properties of an ER fluid are also important parameters especially for vibration damping application. Most devices which use ER fluids operate under dynamic conditions thus small-amplitude oscillatory measurements are appropriate from a practical point of view. There are many techniques to measure the complex shear modulus of viscoelastic materials<sup>2,3</sup>. General idea of these technique is based on the different response of viscous and elastic parts to a sinusoidally varying stress or strain. Of these, the best method to measure ER fluid has not been determined.

Recently, dynamic studies performed with sufficiently small sinusoidal shear strains suggest that ER materials behave as linear viscoelastic materials do<sup>4</sup>. From conducting low frequency(0.01-1.0Hz) dynamic studies on ER fluids composed of diatomite and transformer oil at several concentrations(10~60% by weight of solid phase), they observed, the storage shear modulus( $G'$ ) and loss modulus( $G''$ ) are related to the applied electric field(0-0.6kV/mm). They showed that  $G'$  and  $G''$  were of similar magnitude and that both increased with increasing electric field. It was further noted that the behavior of  $G'$  and  $G''$  as related to the electric field is dependent on the concentration of the particles, i.e.,  $G'$  and  $G''$  increase with the particle concentration. They also reported that moduli were strong function of strain amplitude.

Coulter and Duclos<sup>5</sup>) measured the complex shear modulus using the concentric cylinder annular pumping technique. According to their work, the storage shear modulus( $G'$ ) ranging

from 0 to 70kPa, increases with increasing applied electric field and driving frequency. The loss factor( $\eta=G''/G'$ ) also increases with driving frequency but decreases with applied electric field, which reflects the increasing elastic behavior of the materials. Typical values for their loss factor were between 0 to 4.0 and their electric field was 0 to 4kV/mm.

In this paper, ER fluid's storage shear modulus( $G'$ ) and loss factor( $\eta$ ) have been directly measured using small amplitude forced oscillating rheometer as a function of oscillating frequency, strain amplitude and applied electric field. Two types of experiments were performed ; (a) driving frequency was swept from 10 to 50 Hz at fixed strain amplitude of 0.08 and (b) strain amplitude was swept from 0.02 to 0.2 at fixed driving frequency of 30Hz. We used 2 kinds of sample for this experiment ; cornstarch particles in corn oil and zeolite particles in silicone oil. The electric fields of 1-3 kV/mm were employed.

## 2. Materials

Materials used for present work are ; (1) constarch in corn oil and (2) zeolite in silicone oil. A typical size for constarch particles was about 10 $\mu$ m and that of zeolite was 2 $\mu$ m in diameter on average. Some characteristics and sources of the material are summarized in Table 1. As we know, water content in the particle is important for hydrobased ER fluids

Table 1. Some characteristics and sources of materials

ER fluid (particle/vehicle)	$W_s$ (wt%)	$W_w$ (wt%)	$K_p$	$K_t$
Cornstarch/corn oil	34	8.0	31	3.2
Z3125/silicone oil	34	8.0	12	2.6

Cornstarch : CPC International Inc.(specific gravity=1.38)

Corn oil : CPC International Inc.(specific gravity=0.95)

Z3125 : Sigma Chemical Co. : (specific gravi-

$\eta = 1.99$ )

Silicone oil : Dow Corning 200 (specific gravity = 0.96)

$W_s$ : weight wet particle / (weight wet particle + weight vehicle)

$W_w$ : (weight wet particle - weight dry particle) / weight dry particle

$K_p$ : dielectric constant of particle (measured by using rule of mixture)

$K_t$ : dielectric constant of oil (directly measured)

such as corstarch and zeolite based ER fluids. For cornstarch fluid, cornstarch was dried at 80°C and put into the humidity chamber to absorb 8.0wt% of water and corn oil was also dried at 80°C to remove water inside. For zeolite fluid, zeolite was oven dried at 150°C for 6 hours, in that case it was subsequently allowed to absorb 8.0wt% of water and silicone oil was also heated at 150°C to remove water inside. Those who want to know more about the sample preparation, discuss with the reference by Conrad et al.<sup>6)</sup>

### 3. Experimental

To measure the storage shear modulus,  $G'$  and loss factor,  $\eta$  of the ER fluid the Couette cell type oscillating rheometer was employed. The principle of this measurement is comparing the oscillating shear strain signal and response shear stress signal; forced oscillation devices apply a sinusoidal stress or strain of known amplitude and frequency and measure the resulting strain or stress. The dynamic properties are calculated from the relation between the two. If a sinusoidal strain  $\gamma = \gamma_0 \sin \omega t$  is applied to a linear spring, the resulting stress,  $\tau = G\gamma_0 \sin \omega t$ , is in phase with the strain (since  $\tau = G\gamma$ ). But for a linear dashpot, because the stress is proportional to the strain rate,  $\tau = \eta \dot{\gamma} = \eta \omega \gamma_0 \cos \omega t$ , the stress is 90° out of phase with the strain. As expected, viscoelastic materials exhibit intermediate response which might look like Fig. 1. This can be ex-

plained as being a projection of two vectors,  $\tau^*$  and  $\gamma^*$ , rotating in complex plane. The angle between these two vectors is the phase angle  $\delta$  ( $\delta = 0$  for purely elastic materials and  $\delta = 90^\circ$  for purely viscous materials).

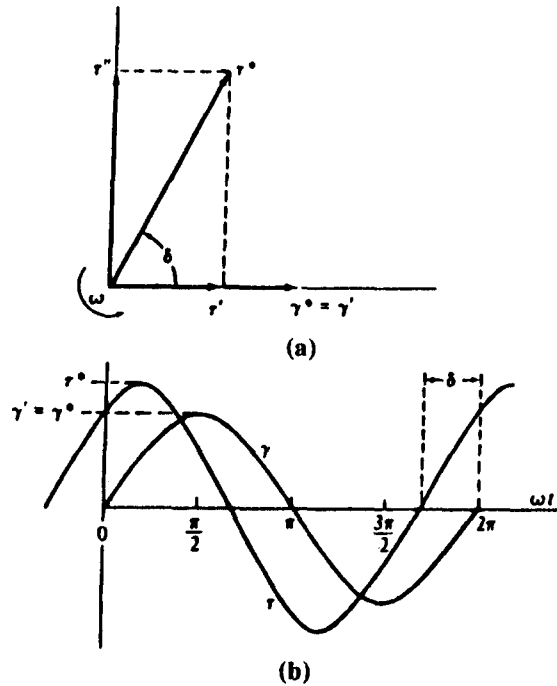


Fig 1. Relationship between stress and strain in viscoelastic material.

It is customary to resolve the vector representing the dependent variable into in phase ( $\tau'$ ) and 90° out of phase ( $\tau''$ ) components with the independent variable in this case being the applied strain. The stress vector ( $\tau^*$ ) is resolved into its in phase ( $\tau'$ ) and out of phase ( $\tau''$ ) components and the strain vector ( $\gamma^*$ ) is resolved into  $|\gamma^*| = \gamma'$  and  $\gamma'' = 0$ . A storage shear modulus (in phase) is defined by

$$G' = \frac{\tau'}{\gamma'} \tag{1}$$

this value represents a measure for the size of the strain energy reversibly stored in the substance, which can be regained. And a loss modulus (out of phase) is defined by

$$G' = \frac{\tau''}{r'} \quad (2)$$

this value represents a measure for the size of energy irreversibly given off to the surroundings by the materials, which is lost. The complex modulus  $G^*$  is defined as the vector sum of in phase and out of phase moduli :

$$G^* = G' + iG'' = (\tau' + i\tau'')/r' = \tau^*/r^* \quad (3)$$

From Fig. 1 and relations above, the loss tangent,  $\tan \delta$ , is defined as

$$\tan \delta = \tau''/\tau' = G''/G' \quad (4)$$

To simplify the equation, the loss tangent,  $\tan \delta$  which defined above will be called loss factor,  $\eta$  hereafter.

The principle of the Couette flow viscometer is well known, and consists of an asynchronous electrical motor to generate a

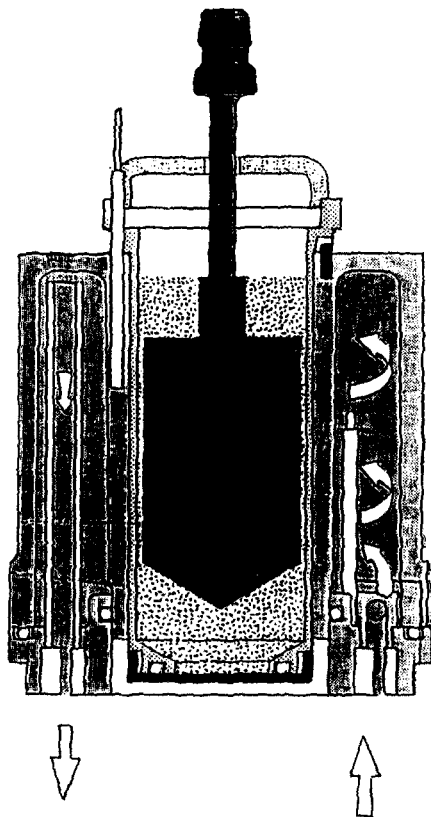


Fig 2. Schematic diagram of the Couette cell type oscillating rheometer.

defined torque or shear rate(or driving frequency). A sensor can be used as a motor and a transducer at the same time. Using Physica universal measuring system and OS200 software, the material under investigation is placed in the gap between the stationary outer measuring cup and the rotating measuring bob. The electric field is applied by high voltage power supply to the cylindrical cup, the outer cup is positive electrode and the bob becomes ground. The details of the cylindrical measuring system are shown in Fig. 2 and schematic diagram of the experimental set up is in Fig. 3.

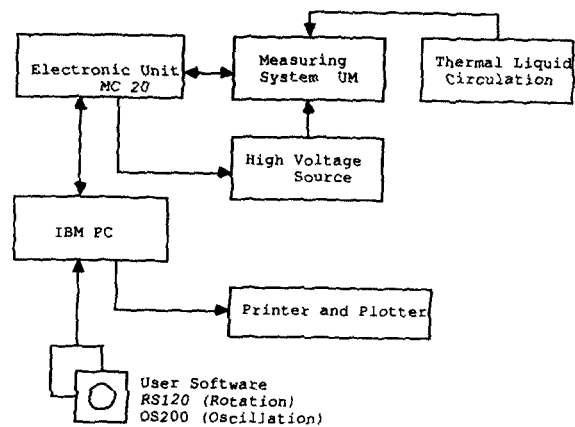


Fig 3. Schematic diagram of experimental set up to measure modulus and loss factor

By selecting different rotational speeds, the shear rate may be varied. The resistance of the material to flow leads to a drag on the rotating bob, which is measured as a torque on the drive shaft. The measured torque is related to the shear stress and viscosity. Physica MS-Z3DIN concentric cylinder measuring system was used which has shear rate range from  $1.291$  to  $1033s^{-1}$ , minimum shear strain amplitude was  $1$  mili rad( $strain=0.02$ ), driving frequency range of  $0.01$  to  $100Hz$  and sample volume was  $17ml$ . The diameter of the cup and bob were  $27mm$  and  $25mm$  respectively (i. e., the gap was  $1mm$ ).

The sinusoidal shear strains were applied to the ER fluid by driving the cylindrical bob. The above parameters shown in the equations ( $G'$ ,  $G''$  and  $\eta$ ) were automatically measured. Using the Physica universal measuring system described above and OS 200 software two types of experiment were performed :

(1) Frequency sweep mode, the strain amplitude was fixed at the intermediate strains (0.08) and driving frequency was varied from 10Hz to 50Hz.

(2) Amplitude sweep mode, driving frequency was fixed at medium frequency (30Hz) and the strain amplitude was varied from 0.02 to 0.2.

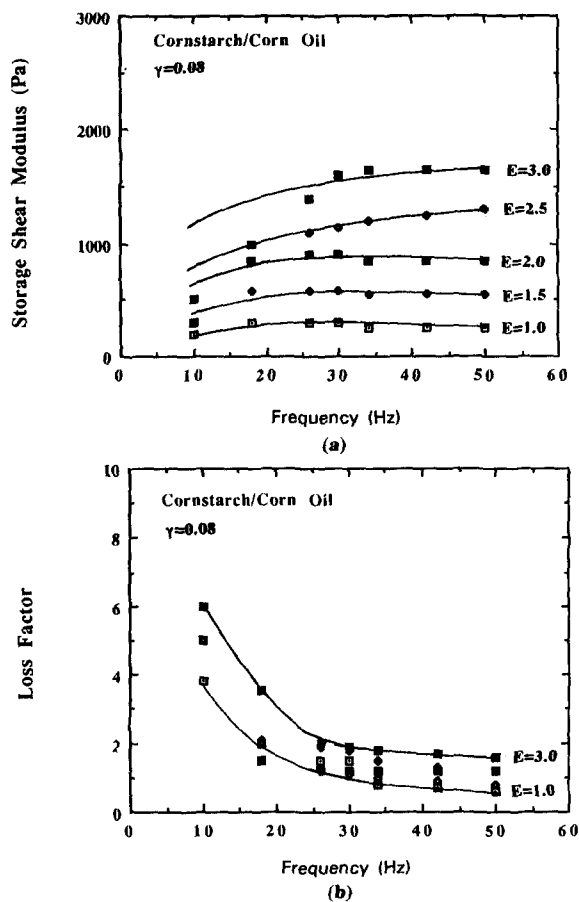


Fig 4. Frequency dependence of (a) storage shear modulus,  $G'$  and (b) loss factor,  $\eta$  at  $\gamma=0.08$  for cornstarch in corn oil ER fluid.

#### 4. Result and Discussion

Fig 4. shows (a) storage shear modulus,  $G'$  and (b) loss factor,  $\eta$  vs frequency,  $f$ , at strain of 0.08 for cornstarch in corn oil at different electric field applied. Fig 5. shows those data for zeolite in silicone oil ER fluid. These data were obtained by frequency sweep method. As we applied electric field, the ER fluid become more elastic. The magnitude of the storage shear modulus increased as high as 1.6kPa at  $E=3\text{kV/mm}$  for cornstarch in corn oil ER fluid at oscillation frequency of 50 Hz. Generally, storage modulus increase with driving frequency as typical polymeric material does. On the other hand loss factors are not well behaved as storage modulus, but as the driving

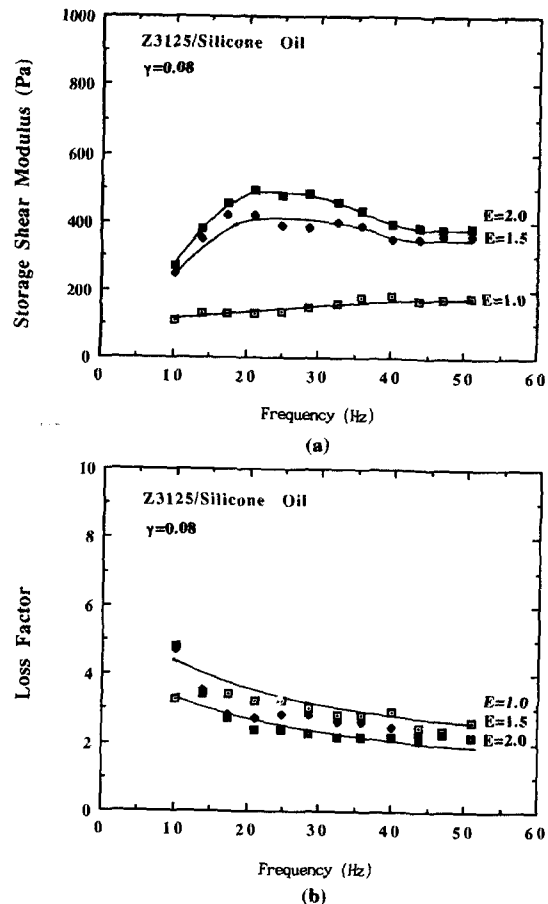


Fig 5. Frequency dependence of (a) storage shear modulus,  $G'$  and (b) loss factor,  $\eta$  at  $\gamma=0.08$  for zeolite in silicone oil ER fluid.

frequency driving frequency increases the loss part decreases was the trend of the material. A typical value for the loss factor was about 2. Both storage modulus and loss factor are strong function of strain amplitude, the strain amplitude which we applied was 0.08 for this measurement. We believe this value is in the linear viscoelastic region.

Fig. 6 shows (a) storage shear modulus,  $G'$  and (b) loss factor,  $\eta$  vs strain amplitude,  $\gamma$  at fixed driving frequency of 30 Hz for cornstarch in corn oil ER fluid at different electric field. Fig 7. shows those data for zeolite in silicone oil ER fluid. These data were measured by strain amplitude sweep method. As can be

seen in the figure storage shear modulus increases as strain amplitude becomes smaller because chains formed were not broken due to small displacement. Due to the limitation of the measuring system the smallest strain we can go was strain of 0.02. The magnitude of the  $G'$  was 4.2kPa at  $E=3\text{kV/mm}$  for cornstarch in corn oil ER fluid at oscillation frequency of 30Hz and that of zeolite fluid was about 2.3 kPa at  $E=2\text{kV/mm}$ , respectively. The loss factor was about 2 for both fluid at the same conditions Korobko and Shulman<sup>4)</sup> studied on ER fluid composed of diatomite and

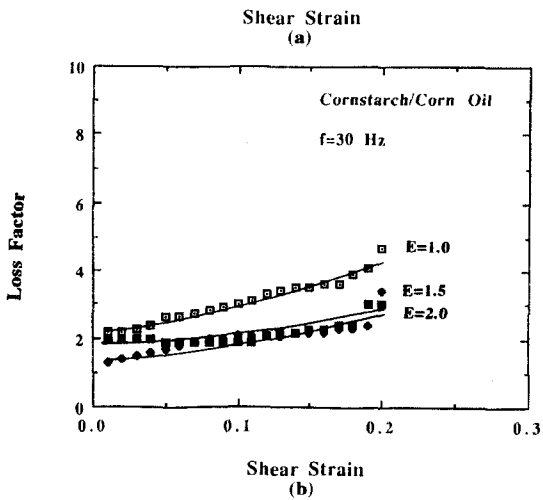
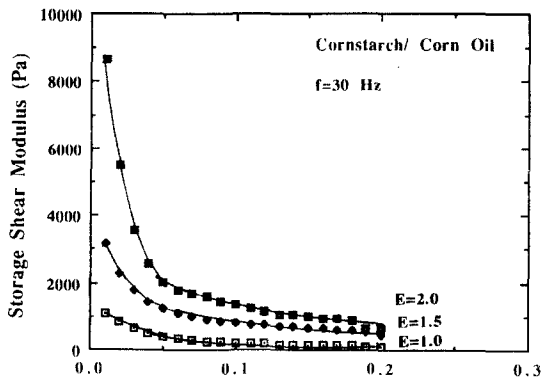


Fig 6 (a) storage shear modulus,  $G'$  and (b) loss factor,  $\eta$  vs strain amplitude,  $\gamma$  at  $f=30\text{Hz}$  for cornstarch in corn oil ER fluid.

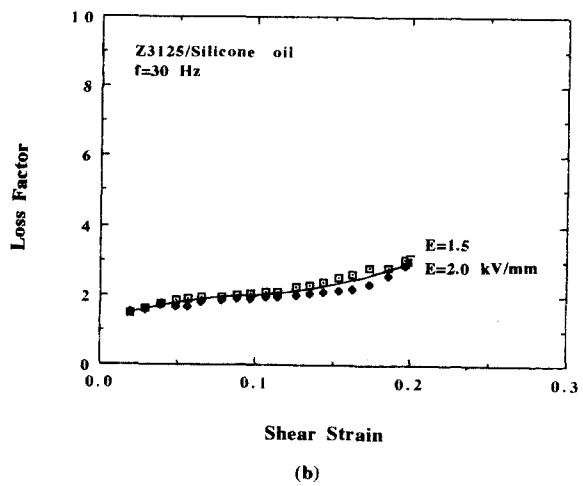
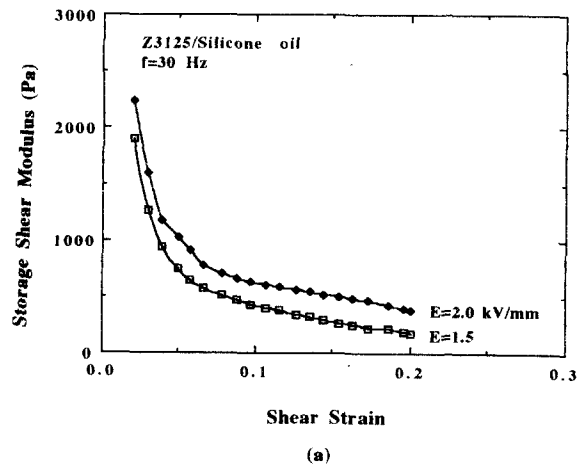


Fig 7 (a) storage shear modulus,  $G'$  and (b) loss factor,  $\eta$  vs strain amplitude,  $\gamma$  at  $f=30\text{Hz}$  for zeolite in silicone oil ER fluid.

transformer oil at several concentrations, they also observed the storage shear modulus ( $G'$ ) and loss modulus ( $G''$ ) were strong function of strain amplitude. Storage modulus of 100kPa was reported for their ER fluid.

Fig 8. shows (a) storage shear modulus,  $G'$  and (b) loss factor,  $\eta$  vs electric field,  $E$ , at driving frequency of 30Hz and strain amplitude of 0.08. Data were picked from Fig 4. and 5. As can be seen in Fig. 8a storage shear modulus increases in an approximately linear fashion with electric field. Data for loss factor were independent with electric field as shown in Fig. 8b.

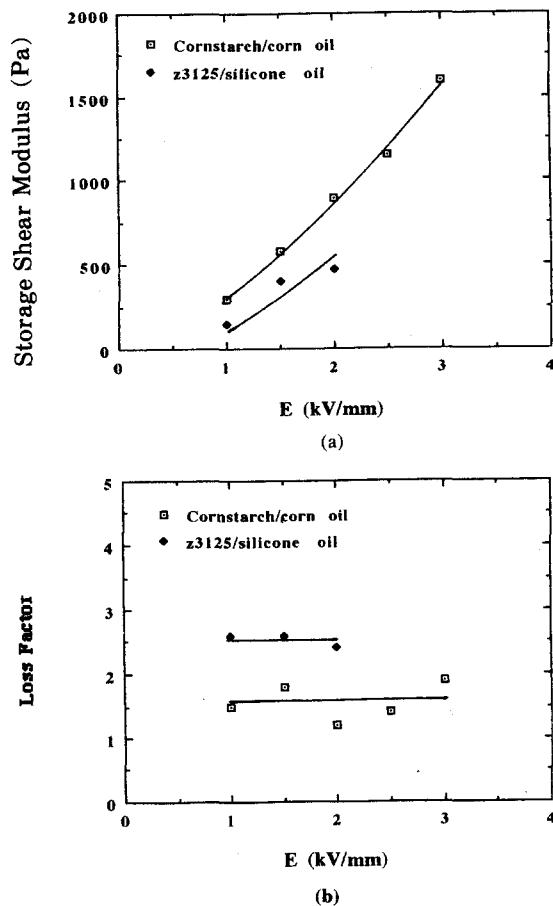


Fig 8. Electric field dependence of (a) storage shear modulus,  $G'$  and (b) loss factor,  $\eta$ . Data were picked from Fig 4 and 5 at  $f=30\text{Hz}$ .

As discussed in reference by Chen et al.<sup>7)</sup>, the theoretical stress can be expressed in terms of shear stress,  $\tau$  vs. shear strain,  $\gamma$ . Rewriting that Eq.

$$\tau = 9A_s \phi \pi \epsilon_0 K_t (E\beta)^2 (a/R)^4 [1.56 \cdot \exp\{[14.84 - 6.16(R/a)\beta^2]\}] \gamma \quad (5)$$

Where  $\tau$ =shear stress,  $A_s$ =structure factor due to many chains in ER fluids,  $\phi$ =volume fraction of particle phase,  $\epsilon_0$ =permittivity of free space,  $\beta = (K_p - K_t)/(K_p + 2K_t)$ =dielectric mismatch,  $a$ =radius of the particle and  $R$ =center to center distance of two particles. This calculation will give the shear modulus  $G' = \tau/\gamma$ , which can then be compared with that measured. For the cornstarch in corn oil case ( $A_s = 11$ ,  $\phi = 0.26$  ( $W_s = 34\text{wt}\%$ ),  $K_t = 3.2$ ,  $K_p = 31$  and  $\beta^2 = 0.554$ ), theoretical calculation gives  $G' = 3.5\text{kPa}$  at strain of 0.02. This value is compared with the measured value of 4.2 kPa. With above theoretical consideration we can predict the yield stress and the shear modulus of an ER fluid from basic physics and chemistry consideration.

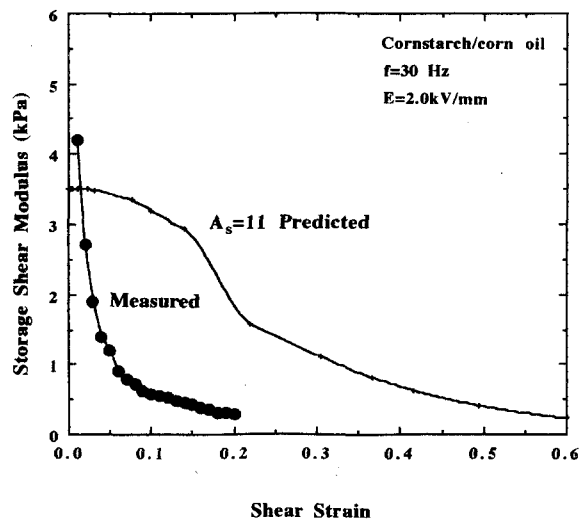


Fig 9. Comparisons between theoretical calculation and experiment (Theoretical curve was drawn using equation 5). Data measured by the oscillating rheometer are included ( $f=30\text{Hz}$ ,  $E=2\text{kV/mm}$ ).

The plot of storage shear modulus,  $G'$  vs shear strain,  $\gamma$  obtained by Eq.5 for cornstarch in corn oil ER fluid at  $E=2\text{kV/mm}$  is shown in Fig 9 along with the measured values. There is a reasonable agreement between the two for low strain amplitude.

### 5. Summary and Conclusions

ER fluid's storage shear modulus,  $G'$  and loss factor,  $\eta$  have been directly measured using small amplitude forced oscillating rheometer as a function of oscillating frequency, strain amplitude and applied electric field. The following summarizes the results and conclusions :

(a) The magnitude of  $G'$  increased as high as 1.6 kPa at  $E=3\text{kV/mm}$  for cornstarch in corn oil ER fluid at oscillation frequency of 50 Hz and strain amplitude of 0.08, but data for zeolite fluid were much lower at the same conditions. The magnitude of the  $\eta$  was about 1.5 for the cornstarch in corn oil ER fluid, and that of zeolite fluid was about 2.5. Generally, storage shear modulus increase with driving frequency. On the other hand the loss factor are not well behaved as storage modulus, but as the driving frequency increases the loss factor slightly decrease was the trend of the material.

(b) Storage shear modulus and loss factor of the ER fluid were measured using amplitude sweep method. The storage shear modulus decreases with the increase of strain amplitude because the rate of chain destruction of the ER fluid increase. Generally, loss factor increases with increasing strain amplitude. There is a reasonable agreement between the

ory and experiment.

(c) Storage shear modulus,  $G'$  was a strong function of applied electric field, but loss factor,  $\eta$  was independent with field (i.e,  $G'$  behaves similarly with  $G'$ ). As we applied electric field ER fluid become more elastic. For  $G'$ , cornstarch in corn oil ER fluid has higher values than zeolite based fluid. On the other hand, zeolite in silicone oil fluid has higher values for  $\eta$ .

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