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Normal forces of magnetorheological fluids under oscillatory shear

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ABSTRACT

The normal forces of magnetorheological fluids under oscillatory shear are investigated by a commercial magneto-rheometer with plate–plate geometry. At the constant strain amplitude and frequency, the normal forces almost keep a steady value with the testing time if the strain amplitude is smaller than the critical value. When a larger strain is applied, they will fluctuate periodically. Under the strain sweep mode, the relationships between normal forces and strain amplitude can be divided into three regions: linear viscoelastic region, nonlinear viscoelastic region and the viscoplastic region. Under the frequency sweep method, it is found that the angular frequency show little influence on the normal forces. At last, the normal forces increase with increasing of the temperature under a low magnetic field, while they decrease under a high magnetic field.

1. Introduction

Magnetorehological (MR) fluid, which is a type of intelligent material consisting of magnetic particle dispersed in non-magnetic liquid such as oil or water, has attracted considerable attentions as their rheological properties could be changed fast and reversibly under magnetic field. During the past decades, various works have been done to develop high performance MR fluids and their outstanding property makes them to be superior candidates for applications in semi-active control systems or mechanical transmission systems. The main applications of MR fluids are attributed to their controllable shear rheological properties, thus tremendous researches have been focused on the shear properties, such as viscosity, shear yield stress and shear modulus. Thanks to many years of effort from various groups, many interesting phenomena and applications of the MR fluids have been intensively studied [1–9].

Normal force of MR fluids, which is perpendicular to the surface of the contact and along the magnetic field, is also important rheological property for evaluating the material and understanding the structure deformation of the magnetorheological fluids. Shulmana et al. [10] firstly calculated the first/second difference of normal stresses in shear flow model by utilizing the tensor of excessive stress for diluted suspensions. Then, Shkel and Klingenberg [11,12] pointed out that the normal stress σ_{33} and the magnetic field intensity H can be expressed as $\sigma_{33} \propto \alpha H^2$ under stationary condition, based on a model of the MR fluid as an anisotropic continuum and using the equilibrium thermodynamics. Vicente et al. [13] have done the pioneer experimental research on the normal force of the MR suspension using a commercial rheometer. They found the normal force could be generated when two conditions were satisfied: the magnetic field must reach a critical value and the MRS must be under shearing. The positive and strain dependent normal force reached a maximum with the increasing of the shearing strain and then decreased sharply. However, See and Tanner [14] and Laun et al. [15] presented the different experimental results that the non-negligible normal force generated even when the MRS was not subject to any deformation. Obviously, more works should be done to clarify the relationship between the normal forces and the magnetic flux density. Very recently, López-López et al. [16] also reported that there were three regions in the normal forces *vs.* shear rate. Especially, they took into account the inhomogeneity of the applied field and calculated the normal force as the integral of the stress over the total surface of the rotational plate. Their results successfully explained the disagreement between the work by Vicente et al. [13] and these by See and Tanner [14] and Laun et al. [15].

The normal forces of MR fluids are complex and they are influenced by many factors. Chan et al. [17] presented that the normal forces can be increased by the addition of a steady-relative speed compared to the stationary plates. Tian and his colleagues [18] found the normal stress increased considerably with increase of the shear rate and magnetic field, and decreased suddenly and significantly upon the onset of shear thickening in MR fluids. Until now, all the works were focused on investigating the normal forces of the MRS in stationary or shear flow. For most of the MR devices operated in dynamic modes, the fluids are subjected to a oscillatory shear [19–22]. Therefore, the normal forces under oscillatory shear are very important to comprehend the characteristic of the devices. However, the normal forces of MR fluids under oscillatory shear have not been reported.

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In this work, the normal forces under oscillatory shear mode are firstly systematically investigated. The time, magnetic field, strain amplitude, frequency and temperature influences on the normal forces under oscillatory shear are studied and analyzed. In linear viscoelastic region the normal forces keep a steady value with the testing time while in nonlinear viscoelastic region the normal forces fluctuate obviously. Three regions can be obtained through the plot of normal forces with strain amplitude. The angular frequency shows little influence on the normal forces. Moreover, the temperature effect on the normal forces is opposite for the low and high magnetic field. These give us a comprehensive understanding of the normal forces of MR fluids and they could be utilized to conduct the designing of the MR device.

2. Experimental

2.1. Materials

MR fluids used in this study were composed of carbonyl iron powder particles (mode CN, BASF, chemical composition (wt%): $>$ 99.5% Fe, $<$0.04% C, $<$0.01% N, 0.2% O; the average particle size was about 6 μm) in silicone oil (H201, Sinopharm Chemical Reagent Co. Ltd and the viscosity was about 20 mPa s). The particles were fully mixed with the silicone oil with a strong electromagnetic mixer. Two MR samples were prepared with iron particle volume fractions of 20% and 30%. A small amount of stearic acid (2 wt%) was added to improve sedimentary stability due to the large density mismatch of the particles and the base oil. The samples were vigorously shaken to ensure the required homogeneity before every measurement.

2.2. Apparatus and methods

The plate–plate magneto-rheometer (Physica MCR301, Anton Paar, Austria) was used to test the normal force of the MR fluids with a temperature controller. The normal force could be measured with a sensor built into air bearing and it could be recorded from -50 to 50 N with an accuracy of 0.03 N. However, the time dependent normal force under oscillatory shear in one period cannot be directly obtained through the system software package. A dynamic signal analyzer (SignalCalc ACE, Data Physics, USA) was used to get the transient normal forces from rheometer output (Fig. 1). After running the rheometer with the MR fluids, a DC voltage through the dynamic signal analyzer is supplied to the magnetorheological unit (Physica MRD 180) to generate the controllable magnetic field other than using the system software. Then the voltage signal can be acquired from rheometer configuration M1 and M2 output with normal force and absolute displacement with a sampling rate of 400 Hz, respectively. The voltage signal about normal force and strain can be standardized with the data obtained from the system software and then the true values can be obtained.

The diameter of the plate used is 20 mm. The MR fluids are placed between the two plates and the gap is fixed at 1 mm. The oscillatory shear mode with the constant amplitude and frequency is applied to the sample through the rheometer, the normal force and strain with time is acquired by the dynamic signal analyzer simultaneously. Then two typical modes, amplitude sweep mode and frequency mode, are utilized to carry out the experiments. The effects of strain amplitude, oscillatory frequency and the magnetic field on the normal forces of MR fluids are tested. The temperature is controlled using the water bath. Before the testing, the samples are sheared without a magnetic field at 50/s for 150 s to ensure good dispersion. Then, the shearing is stopped and the magnetic field is applied at the same time for 30 s to equilibrate the sample. At last the oscillatory shear testing begins. All the tests are repeated for three times to guarantee the validity of the results and the temperature is fixed at 25 °C except for studying the temperature effect on normal forces.

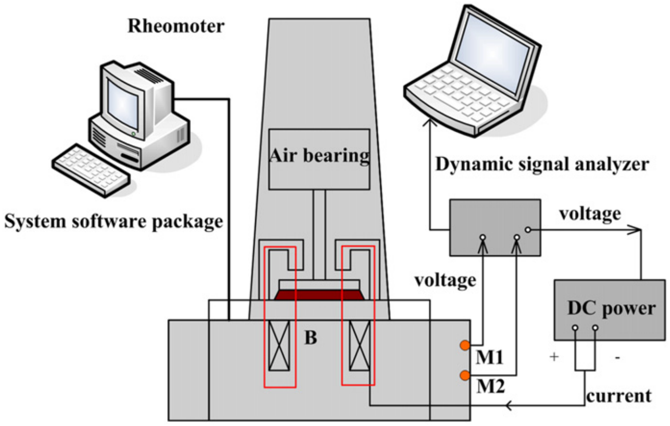

Fig. 1. Schematic of the measuring system with the magneto-rheometer and data process system.

3. Results and discussion

3.1. The dynamic behavior of MR fluids

Firstly, with the strain sweep mode, the storage modulus of MR fluid as a function of strain amplitude is measured under different magnetic field (Fig. 2). The driven angular frequency is kept at 5 rad/s

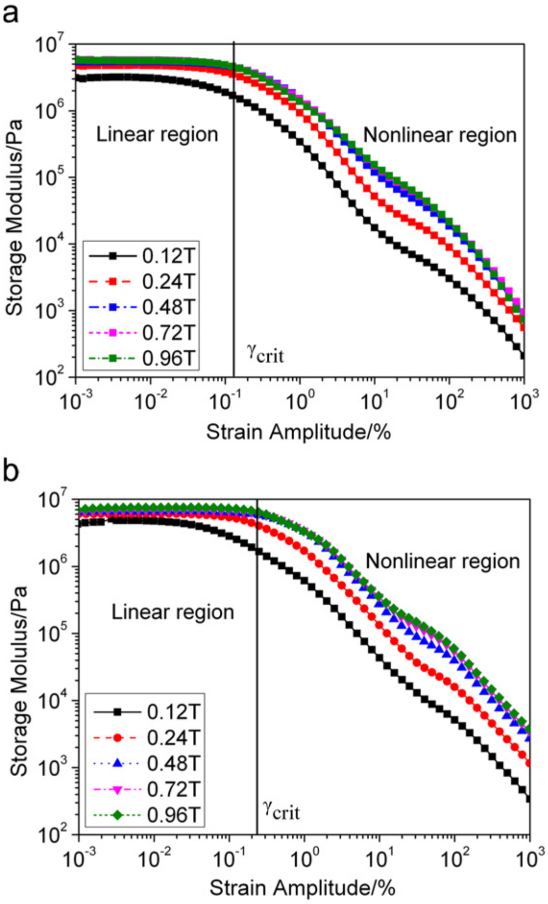

Fig. 2. Storage modulus as a function of the strain amplitude for (a) 20% MR fluid and (b) 30% MR fluid at 5 rad/s and different magnetic field.

and the strain amplitude is swept logarithmically from 0.001% to 1000%. As shown in Fig. 2, when the strain amplitude is smaller than the critical value (γ_{crti}), the storage modulus keeps almost constant and the dynamic behavior MR fluids belong to a linear viscoelastic region. As soon as the strain amplitude exceeds the critical value, the storage modulus decreases sharply. In this case, the changes of the storage modulus enter into a non-linear region. The critical strain values are about 0.1%–0.5% and the value increases with the increasing of the magnetic field and the fraction of MR fluids, similar to the previous reports [23–25]. It is reported that the storage modulus is highly dependent on the microstructure of the MR fluids, thus these transitions are related to the structure transformation [26,27]. After applying the magnetic field, the randomly dispersed iron particles re-assembled to form chains or clusters. Under very small strain amplitudes, particle displacements from equilibrium are small and the response is linear. When the strain amplitude reaches the critical value, the slight rearrangements of the unstable clusters under shearing happen and the transition from linear viscoelastic to the nonlinear viscoelastic deformation occurs. At even larger strain amplitude, more drastic rearrangements occur (including chain rupture). Then, the MR fluids exhibit a viscoplastic behavior (This phenomenon cannot be shown directly from Fig. 2 and the magnitude of the complex dynamic viscosity would be a better choice for representing the transition from viscoelastic to viscoplastic deformation [27]).

3.2. The time dependent normal forces

Using the dynamic signal analyzer, the time dependent normal forces during one oscillatory period are obtained. As shown in Fig. 3, the applied strain is sinusoidal and the beginning is at the maximum strain (zero shear rate). The normal forces under oscillatory shear in linear viscoelastic and nonlinear viscoelastic region are investigated in Figs. 4–6. As shown in Fig. 4, the applied strain amplitude is 0.01% under different oscillatory frequency (Fig. 4 (a) $f=0.1$ Hz and (b) $f=1$ Hz) for the 20% MR fluid, which is smaller than the critical value (γ_{crti}). The response of MR fluids is in the linear viscoelastic region. Upon application of the magnetic field, the positive normal forces generate and push the plates apart. Under a constant magnetic field, the normal forces keep a constant value and the testing time do not show any influence on them. With the increasing of the magnetic field the normal forces increase monotonously. In previous reports [14,15], it is claimed that the formed chains or clusters are the origin of normal forces. With increasing of the magnetic flux density, the magnetostatic interactions between the particles become stronger, and hence the normal forces increase. The data is fitted with a power law equation $F_N=kB^n$, where k and n is constant (the inserts of Fig. 4). In this work, $n=1.14$ ($f=0.1$ Hz) and 1.18 ($f=1$ Hz), which is much smaller than that predicated by Shkel and Klingenberg [11,12]. Under simple shear deformation γ, the leading order γ for the normal force or normal stress simplifies to $F_N \propto \sigma_{33} \propto \alpha H^2$, α relates to the material magnetostriction coefficient and magnetic susceptibility. That is, the normal force under small deformation (linear viscoelastic region) is independent on strain. Therefore, the normal forces measured under different testing time are almost steady values. Here the exponent n is smaller than 2, which maybe relate to the inhomogeneity of the applied field [16,28,29] and that

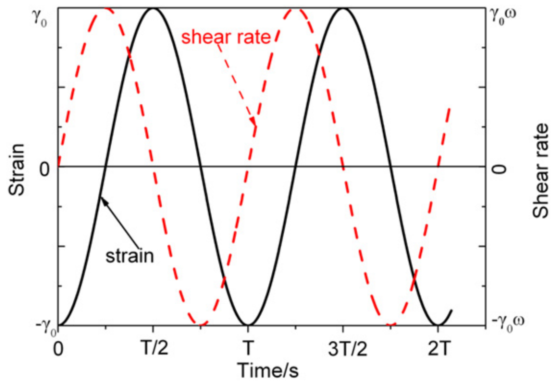

Fig. 3. Applied sinusoidal strain with time (the beginning of strain is at the maximum shear strain and zero shear rate).

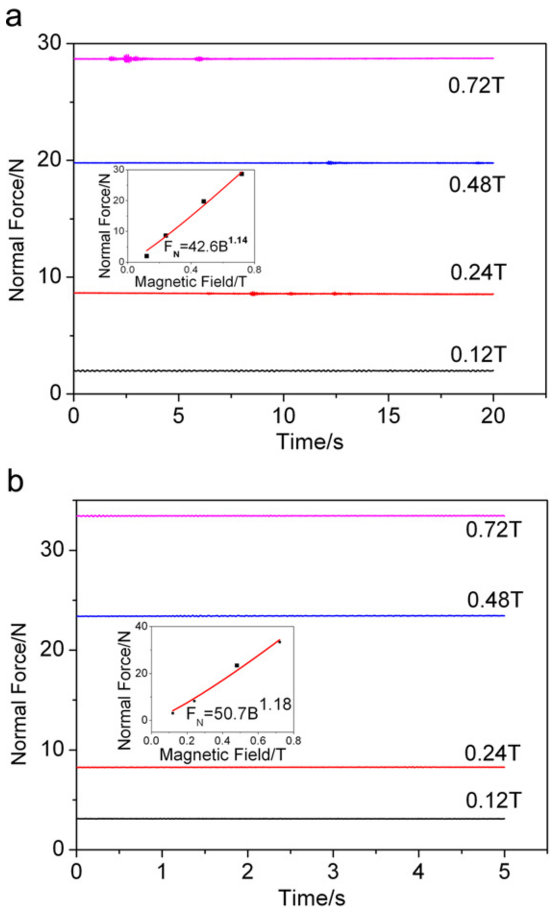

Fig. 4. Normal forces of 20% MR fluids with time under sinusoidal strain at $\gamma=0.01\%$ for (a) $f=0.1$ Hz and (b) $f=1$ Hz (the inserts show the power law relation between normal forces and magnetic field).

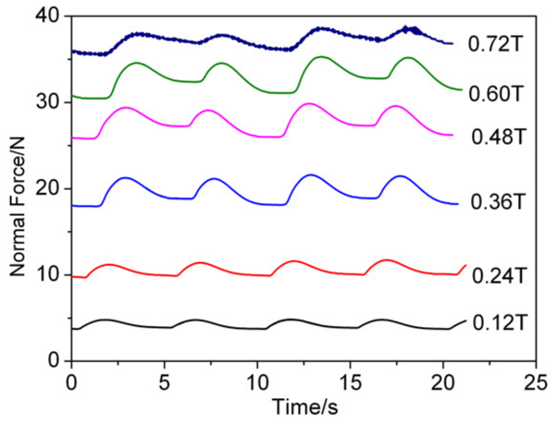

Fig. 5. Normal forces of 30% MR fluid with different magnetic field at $\gamma=10\%$ and $f=0.1$ Hz.

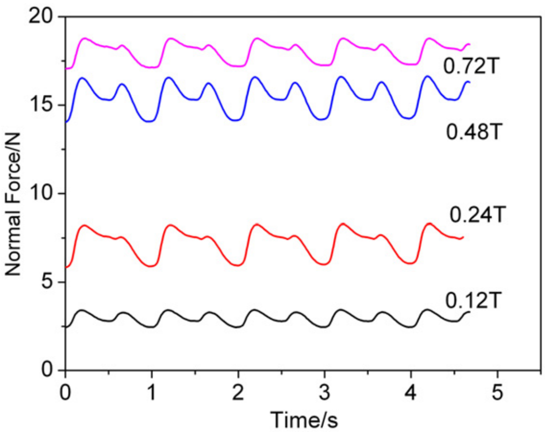

Fig. 6. Normal forces of 20% MR fluid with different magnetic field at $\gamma=10\%$ and $f=1$ Hz.

the MR fluids are not perfect transverse isotropic materials. Similar phenomenon can also be found for the 30% MR fluids (not shown here for brevity).

Figs. 5 and 6 show the responses of the MR fluids in the nonlinear viscoelastic region where the applied strain amplitude is 10%. As soon as the magnetic field is applied, the positive normal forces are obtained. The normal forces increase with increasing of the applied magnetic field. Under a constant magnetic field the normal forces fluctuate with the testing time but not in a sinusoidal way. More specifically, at the beginning of the applying strain, the shear rate is zero (Fig. 3); in this case for 30% MR fluid (Fig. 5), the normal forces are the minimum and keep almost constant values. Once the shear rates reach a critical value, with the increasing of the shear rate, the normal forces increase to the maximum where shear rate is about at the maximum. Then the normal forces begin to decrease and then settle at a steady value until the shear rates decrease to zero. The same tendency happens in the other half period and another maximum normal force exists in the negative maximum shear rates. This progress can be recycled in the following period. For the 20% MR fluid (Fig. 6), the similar fluctuant normal forces exist. However, the normal forces in the first half cycle and last half cycle differ obviously compared to 30% MR fluid (Fig. 5), as the oscillatory frequency for the 20% MR fluid is more higher and there is not sufficient time to change the microstructure during every half cycle of oscillatory shear.

The results show an imposed shear can increase the normal forces under oscillatory shear. The normal forces increase with increasing of the shear rate and the maximum values occur in the maximum shear rate where the strain is zero. That is different from the normal force under steady shear where the maximum normal forces exist in the critical tilt angle of the chains [13,16,17]. As in the nonlinear viscoelastic region the clusters formed by the magnetic field rupture and rearrange dynamically and the full chain does not exist here. A new model needs to be developed to capture the normal force under oscillatory shear.

3.3. *Strain amplitude dependent normal forces*

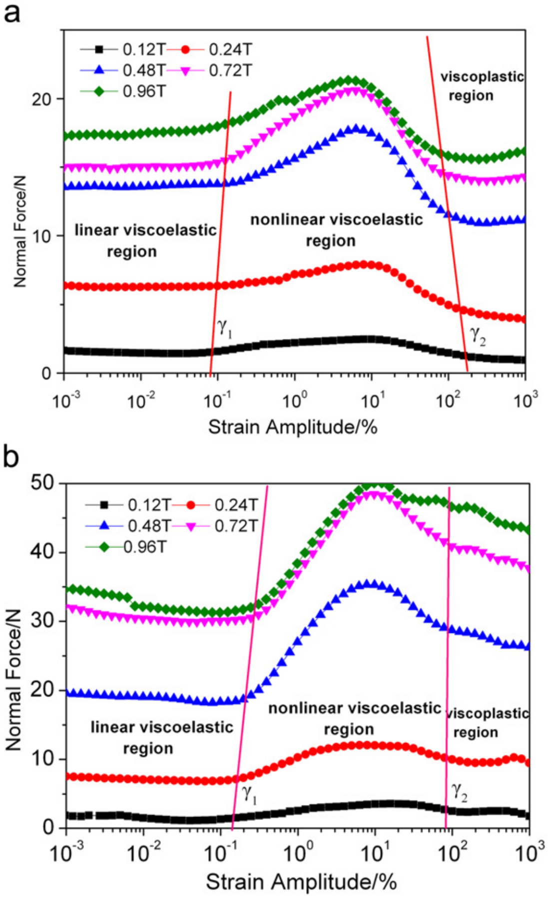

Fig. 7. Normal forces with strain sweep mode for (a) 20% MR fluid and (b) 30% MR fluid under various magnetic field and at $\omega=5$ rad/s.

The average normal forces of one oscillatory period with strain amplitude swept logarithmically from 0.001% to 1000% for the 20% and 30% MR fluids are measured under different magnetic field (Fig. 7). The oscillatory frequency is 5 rad/s. Keep the strain amplitude constant, the normal forces increase with increasing of the applied magnetic field. When the magnetic field is kept as a constant value, the relations between the normal forces and strain amplitude are complex and they could be divided into three regions. At first, when the strain amplitude is smaller than the critical value γ_1 (about 0.1–0.5%), the normal forces keep almost steady values. This can be defined as a linear viscoelastic region. The iron particle chains or clusters keep almost intact and the strain amplitude has little effect on the normal force. For the second region, when the applied strain amplitude exceeds the critical value γ_1, the normal forces increase firstly and then decrease until to steady values with increasing of the stain amplitude. The maximum normal forces occur when the strain amplitude is about 10%. This is in the nonlinear viscoelastic region where the particle clusters rupture and rearrange slightly. At the beginning of the nonlinear viscoelastic region the clusters keep almost intact. With increasing of the strain amplitude the shear rate is increased. Therefore, the normal forces increase simultaneously. However, at the larger strain amplitude, the chains and clusters rupture more obviously and the normal forces would decrease with increasing of the strain amplitude. The critical value γ_1 equals the value γ_{crti} from Fig. 2. Finally, when the strain amplitude exceeds the critical value γ_2, the normal forces settle a relatively steady value and the response is in the viscoplastic region where more drastic rearrangements occur and a new dynamic equilibrium for normal force was reached. The critical value γ_2 is about 100%–300%. For the 20% and 30% MR fluids the normal force with the strain amplitude is almost the same except that the critical value is a little different.

The strain amplitude dependent normal forces supply more obvious and comprehensive understanding for the dynamic behavior of the MR fluids. In comparison to the storage modulus-strain amplitude plot (Fig. 2), the plot of normal force *vs.* strain amplitude should be a better way to study the dynamic behavior. The three dynamic behaviors can be obtained directly from the strain amplitude dependent normal forces. Therefore, it could be considered as a better method to study the rheological property of MR fluid.

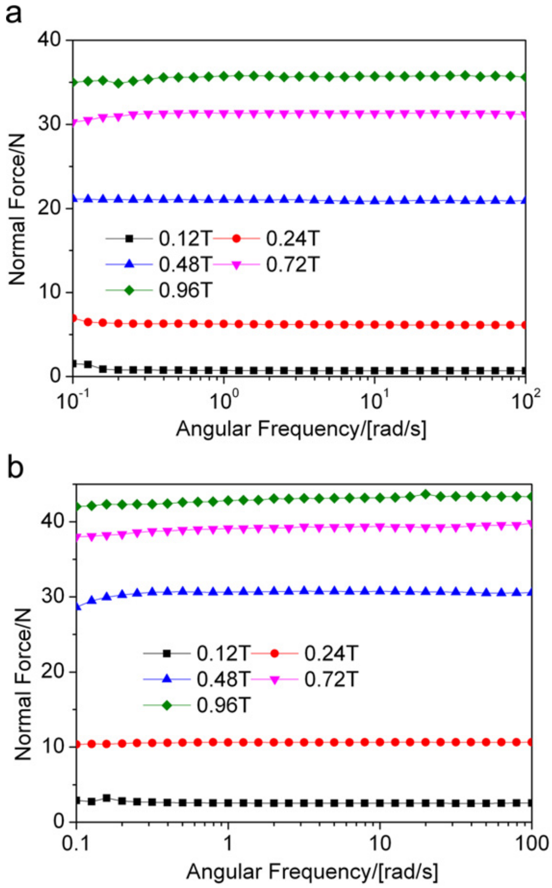

Fig. 8. Normal forces of 30% MR fluids with frequency sweep mode under various magnetic field for (a) $\gamma=0.01\%$ and (b) $\gamma=1\%$.

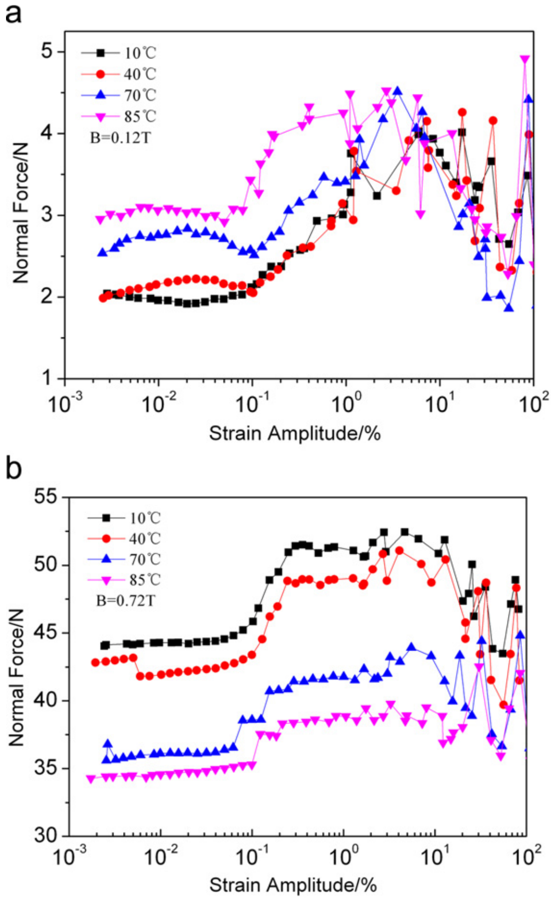

Fig. 9. Strain amplitude dependent normal forces of 30% MR fluid for various temperatures at (a) $B=0.12$ T and (b) $B=0.72$ T.

3.4. *Frequency dependent normal forces*

The frequency sweep mode is utilized to measure the average frequency dependent normal forces (Fig. 8). The angular frequency is swept logarithmically from 0.1 to 100 rad/s at $\gamma=0.01\%$ and $\gamma=1\%$. The normal forces increase with the increasing of the magnetic field at constant angular frequency. Under a constant magnetic field the normal forces keep almost steady values. The normal forces decrease at low magnetic field while increase a little at high applied field, with increasing of the frequency. At such a range of angular frequency and constant strain amplitude, the MR fluids lie in a determinate region. For example, the response of MR fluid with oscillatory frequency from 0.1 to 100 rad/s and amplitude strain 0.01% is in the linear viscoelastic region (Fig. 8(a)) while the dynamic behavior of MR fluid with oscillatory frequency from 0.1 to 100 rad/s and amplitude strain 1% is nonlinear viscoelastic region (Fig. 8(b)). The microstructures have no too much alteration at the region of frequency from 0.1 to 100 rad/s. However, if the frequency was increased to a critical value, the normal forces will change remarkably as the MR fluids behavior enters the Newtonian region [27]. Besides, the oscillatory normal forces almost equal to the normal forces under stationary when the ME fluids is in the linear viscoelastic zone while the oscillatory normal force is larger than the static normal forces in nonlinear viscoelastic zone.

3.5. *Temperature effect on the normal forces*

The temperature effects on the normal forces are measured under oscillatory shear. As shown in Fig. 9, the strain amplitude dependent normal forces of 30% MR fluid are tested at 10 °C, 40 °C, 70 °C and 85 °C under different magnetic field. The driven angular frequency is 5 rad/s and the strain amplitude is swept logarithmically from 0.001% to 100%. At different temperature, the tendency of the normal forces with strain amplitude is the same and it can also be divided into three regions. At low magnetic field of $B=0.12$ T (Fig. 9(a)), the strain dependent normal forces increase with the increasing of the temperature. However, under high magnetic field of $B=0.72$ T (Fig. 9(b)), the strain dependent normal forces decrease with the increasing of the temperature. At moderate magnetic field the strain amplitude normal forces keep almost steady with various temperatures (not shown here). Therefore the strain dependent normal forces are dependent on the temperature and the magnetic field and there are competitions between temperature effect (Brownian force) and magnetic field effect (magnetostatic attractive force). At low magnetic field the attractive forces between particles are not so strong and the incomplete and free clusters exist in the carrier oil. With the increasing of the temperature, the viscosity of the carrier oil will decrease and the Brownian motion will become severe. It will increase the opportunity to form more complete cluster to strengthen the normal forces. However, under high magnetic field, all the particles are fully constrained to form chains and with the increase of temperature the separation of particles from the clusters may become more severely and the normal force caused by the particles clusters would decrease.

Then, the temperature effect on the angular frequency dependent normal forces of 30% MR fluid for various temperatures at $B=0.12$ T and $B=0.72$ T are studied, as shown in Fig. 10. The strain amplitude is 1% and the angular frequency is swept logarithmically from 1 to 100 rad/s. Similarly, at low magnetic field (Fig. 10(a)) the angular frequency dependent normal forces increase while under high

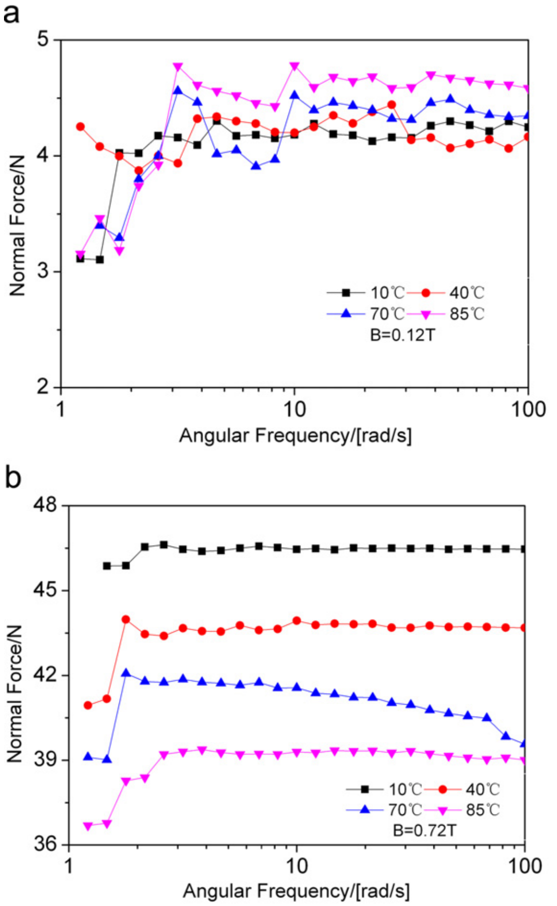

Fig. 10. Angular frequency dependent normal forces of 30% MR fluid for various temperatures at (a) $B=0.12$ T and (b) $B=0.72$ T.

magnetic field (Fig. 10(b)) the angular frequency dependent normal forces decrease with the increasing of the temperature. All can be attributed to the competition between the temperature and magnetic field as above. Moreover, it can be found that at low magnetic field the normal forces fluctuate obviously because the particles move easily and the chains are not steady.

4. Conclusions

In this work, the normal forces of MR fluids under oscillatory shear are measured and analyzed. Using a plate–plate magneto-rheometer and dynamic signal analyzer, the normal forces at a constant strain amplitude and angular frequency are obtained. When the strain amplitude is smaller than the transition value from linear viscoelastic to nonlinear viscoelastic region, the normal forces keep almost steady values with the testing time while they fluctuate obviously when the strain amplitude exceeds the critical value. However, the normal forces always increase with increasing of the magnetic field.

With the strain sweep and frequency sweep mode, the strain amplitude dependent normal force and the angular frequency normal forces are investigated. The first one can be divided into three regions: *linear viscoelastic region*-the normal forces keep steady value where the strain value is less than a critical value γ_1 (0.1–0.5%); *nonlinear viscoelastic region*-the normal forces firstly increase and then decrease between the critical values γ_1 and γ_2 (100–300%); *viscoplastic region*-the normal forces settle another stable value where the strain amplitude exceeds the critical value γ_2. These regions relate to their microstructure transitions. However, at a constant strain the normal forces with angular frequency settle stable values, as the dynamic behavior of MR fluids is in a determinate region at such a region of the testing angular frequency.

Under the low magnetic field, both the strain and angular frequency dependent normal forces increase with the increasing of the temperature. However, under high magnetic field, they decrease with the increasing of the temperature. These can be attributed to the competition between the temperature and magnetic field.

Acknowledgments

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