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Study of Magnetorheological Grease's Thermomagnetic Coupling Rheology

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Abstract: The controllable rheological properties of magnetorheological grease offer significant application prospects in regulating the lubrication behavior of frictional substrates. A novel nano-magnetorheological grease was prepared using nanoscale manganese ferrite as magnetic particles. The prepared magnetorheological grease underwent magnetic field scanning and rate scanning studies under thermomagnetic coupling, and we investigated the variation patterns of rheological parameters under different temperatures and magnetic field intensities. The Herschel–Bulkley rheological model was utilized for data fitting to determine the shear yield stress of the magnetorheological grease. Furthermore, the variation patterns of shear yield stress with increasing magnetic field intensity were explored. The results demonstrated that the apparent viscosity and shear stress of the magnetorheological grease decreased with increasing temperature, while they increased with enhanced magnetic field intensity. The apparent viscosity of the magnetorheological grease decreased with increasing shear rate. Additionally, the shear yield stress of the magnetorheological grease decreased with a temperature rise, but increased when an external magnetic field was applied. The adverse effects of high temperature on the magnetorheological grease could be mitigated by the application of an external magnetic field.

Keywords: magnetorheological grease; rheological properties; thermomagnetic coupling; apparent viscosity

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1. Introduction

Magnetorheological grease is a colloidal dispersion system composed of magnetic particles and a lubricating grease matrix. Currently, the most commonly used magnetic particles are micron-sized carbonyl iron powders. Lubricating greases with different viscosity grades are selected as the matrix based on the viscosity requirements of practical working conditions. The base carrier fluid in the grease can prevent the sedimentation of magnetic particles [1,2]. Under the action of an external magnetic field, the magnetic particles arrange themselves into a chain-like structure along the direction of the magnetic field due to the magnetic torque, giving rise to a magnetorheological effect in the magnetorheological grease. The thickening agent in the grease causes the magnetorheological grease to exhibit non-Newtonian flow characteristics [3]. Owing to its remarkable sedimentation stability and magnetorheological effect, magnetorheological grease holds significant research value and promising application prospects.

Researchers both domestically and internationally have primarily focused on the preparation and rheological properties of magnetorheological grease. Mohamad et al. [4–7] investigated the influence of differently shaped magnetic particles on the rheological properties of magnetorheological grease and found that plate-like particles exhibited superior yield stress and magnetorheological effects. Hu et al. [8,9] studied the effects of different timings of magnetic particle addition and cooling conditions on the performance of magnetorheological grease. The research results indicated that adding magnetic particles during

the dilution stage improved the stability and flowability of magnetorheological grease, and faster cooling rates resulted in lower grease consistency. Ye et al. [10] investigated the effect of temperature and magnetic field on the rheological properties of magneto-rheological grease and showed that the apparent viscosity of magneto-rheological grease became less affected by temperature after the application of the magnetic field. Bahiuddin et al. [11] proposed a simulation model based on machine learning methods, which was then compared with experimental results. The comparison between rheological data and experimental data demonstrated good accuracy. Wang et al. [12] prepared magnetorheological grease with different mass fractions of hydroxyl iron powder and studied the variation in rheological parameters with shear rate and magnetic field intensity. The results showed that the magnetorheological grease prepared with hydroxyl iron powder exhibited excellent settling stability, and MRG-70 had an outstanding magnetorheological effect at a saturated magnetization intensity. Ye et al. [13] investigated the normal force characteristics of a prepared magnetorheological grease and found that the normal force gradually increased with increasing temperature, mass fraction of the magnetic particles, and magnetic field intensity. Yang et al. [14,15] studied the thermal-magnetic coupling shear stability of prepared magnetorheological grease and observed that as the magnetic field intensity increased, the formation of magnetic chains among the magnetic particles in the magnetorheological grease affected its rheological performance, resulting in good shear stability. In addition, foreign scholars have carried out some research on the thermal effect of nanofluids. Alhadri et al. [16] proposed a Cu–Al_2O_3/water hybrid nanofluid with the Darcy–Forchheimer effect and studied the local Nusselt number and skin friction, which were predicted by an artificial neural network under different temperatures and speed conditions. Zhu et al. [17] proposed a numerical study on the heat and mass transfer characteristics of a non-Newtonian Williamson nanofluid over a stretching/shrinking sheet, considering thermophoresis and Brownian effects. The analysis found that increasing the stretching parameter reduces fluid velocity and increases thermal boundary layer thickness. Alahmadi et al. [18] proposed an optimization study on the magnetohydrodynamic flow of a radiative micropolar nanofluid in a channel using RSM and sensitivity analysis. The analysis focused on the skin friction coefficient's sensitivity to the Reynolds number and magnetic parameter, revealing their direct and inversely proportional relationships with the micropolar parameter. Raza et al. proposed nonsimilarity simulations for the heat and mass transfer phenomena of a micropolar nanofluid with microrotation effects, presenting numerical results and analyzing the influence of physical parameters on velocity, microrotation, heat, and concentration. They also investigated thermal radiation and natural convection in the flow of a hybrid nanofluid across a permeable longitudinal moving fin using the TOPSIS method, aiming to optimize the heat transfer rate by selecting suitable parameter values. Additionally, Jawairia et al. conducted sensitivity analysis and optimization of the heat transfer rate in a moving porous fin under radiation and natural convection by utilizing response surface methodology, highlighting the impacts of various parameters on heat transmission [19–21].

The research conducted by scholars both domestically and internationally has established a mature understanding of magnetorheological grease synthesized from lithium-based lubricating grease and carbonyl iron powder. Previous studies have demonstrated that the application of an external magnetic field can alleviate the adverse effects of high temperature on magnetorheological grease, enabling long-lasting lubrication over a wide temperature range. However, the current studies predominantly employ micron-sized carbonyl iron powder as the magnetic particles for magnetorheological grease synthesis, with limited exploration of nano-particles possessing micro-ball and wear repair effects [22]. Consequently, this study introduces nano-magnetic particles into the lubricating grease to synthesize nano-magnetorheological grease, aiming to investigate its rheological properties under different temperatures through the application of an external magnetic field. The applied magnetic field is used to reduce the influence of high temperature on the rheological properties of magnetorheological grease, solve the problem of lubricant failure under

high-temperature conditions, and realize the long-term service of magnetorheological grease in a wide temperature range.

2. Preparation of $MnFe_2O_4$ Magnetorheological Grease

The magnetic particles used in this study were $MnFe_2O_4$ particles manufactured by Shanghai Macklin Biochemical Technology Co., Ltd., with an average particle size of approximately 40 nm (LOT: F062701, CAS: 68086-94-7). In order to prevent sedimentation of the magnetic particles in the base carrier liquid and ensure that they could form chains by breaking through the fibrous network structure of the grease soap under the influence of an external magnetic field, a base carrier liquid of NLGI (National Lubricating Grease Institute) grade 00 lithium-based lubricating grease was utilized. The manufacturer of the lubricating grease was the Tianjin Branch of Sinopec Corporation, Tianjin, China, and the main composition and technical parameters are presented in Table 1.

Table 1. Main composition and technical parameters of grease.

Item	Parameter
Base oil	Mineral oil
Type of thickening agent	Lithium 12-hydroxystearate
Thickener content/%	4.8 (mass fraction)
Viscosity (20 °C, 10 s^{-1})/$mm^2 \cdot s^{-1}$	13
Dropping point/°C	173
Consistency class	00
Working cone penetration/dmm	417

The preparation of magnetorheological grease involved the combination and heating of nanoscale manganese ferrite ($MnFe_2O_4$) with lubricating grease. The specific preparation method was based on the approach put forward by Wang et al. [23]. Initially, the measured quantity of lubricating grease was placed into a beaker and subjected to water bath heating using a digital constant-temperature water bath pot until it reached a temperature of 80 °C. Subsequently, an electric stirrer was employed with a rotational speed of 500 rpm for a duration of 10 min to ensure uniform heating. Following this, the measured quantity of nanoscale manganese ferrite particles was added to the lubricating grease. The rotational speed of the electric stirrer was then adjusted to 800 rpm and stirring was continued for 1 h, enabling the even dispersion of the nanoscale manganese ferrite particles within the lubricating grease. This process yielded magnetorheological grease characterized by exceptional superparamagnetic properties and sedimentation stability. The mass fraction of magnetic particles in this experiment was 15%.

3. Rheological Experiment of $MnFe_2O_4$ Magnetorheological Grease

The rheological test of magnetorheological grease was conducted using an Anton Paar MCR301 rotational rheometer, manufactured by Anton Paar GmbH, Ostfildern, Germany. The experimental setup, as shown in Figure 1, employed a flat plate testing configuration. The temperature control module featured a water bath heating system, capable of maintaining temperatures within the range of 5 °C to 85 °C. The magnetic field control module consisted of an energized coil, capable of delivering a maximum output current of 5 A, corresponding to a magnetic field intensity of 1.1 T. In the figure, the upper platform represents the lower plate of a PP20/MRD test head with a test diameter of 10 mm. During the experimental procedure, an appropriate quantity of magnetorheological grease was placed between the lower plate of the PP20/MRD test head and the lower platform. The desired temperature and magnetic field intensity were achieved by adjusting the water bath temperature and current magnitude, respectively. Real-time data captured by the PP20/MRD measuring system during the test were fed back to the main unit, allowing for the acquisition of experimental results.

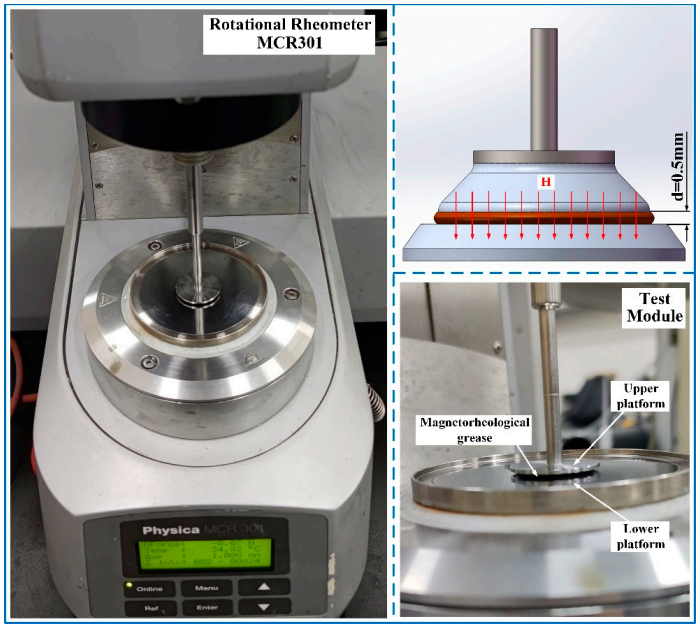


Figure 1. Rotational rheometer.

During the experiments, we considered uncertainties such as variations in grease volume, ambient temperature differences, and vibrations. To address these, we carefully controlled the operating process, recorded ambient temperature, and monitored the experimental platform for disturbances. We made efforts to minimize their impact on the data.

To investigate the rheological behavior of magnetorheological grease under the influence of thermomagnetic coupling, a rotational rheometer was utilized to conduct magnetic field scanning experiments at different temperatures. Additionally, rate scanning experiments were performed on the magnetorheological grease under various temperature and magnetic field intensity conditions. The magnetic field saturation intensity of the magnetorheological grease was found to be 440 mT. Consequently, five magnetic field intensities (0 mT, 55 mT, 110 mT, 220 mT, and 440 mT) were selected for the rheological tests, with the corresponding current values for the magnetic field control module presented in Table 2. The temperature control module of the rheometer employed a water bath heating system, providing a temperature range of 5 °C to 85 °C. Hence, four temperature conditions (20 °C, 40 °C, 60 °C, and 80 °C) were chosen for the experiments.

Table 2. The magnetic field intensity of the rotary rheometer corresponds to the current.

Magnetic Field Intensity/mT	Current/A
0	0
55	0.25
110	0.5
220	1
440	2

The experimental procedure for the magnetic field scanning of the magnetorheological grease was in accordance with the schematic diagram shown in Figure 2. At a shear rate of 10 s^{-1}, the experiments were conducted at four different temperature conditions: 20 °C, 40 °C, 60 °C, and 80 °C. The current was linearly increased from 0 to 5 A, resulting in a linear increase in magnetic field intensity from 0 to 1.1 T. To ensure the accuracy of the experimental data, a pre-shearing process was performed for 72 s at 0 current, followed by a 480 s ramp-up period of the current. Data points were collected every 12 s to examine the changes in shear stress and apparent viscosity with variations in magnetic field intensity at each temperature.

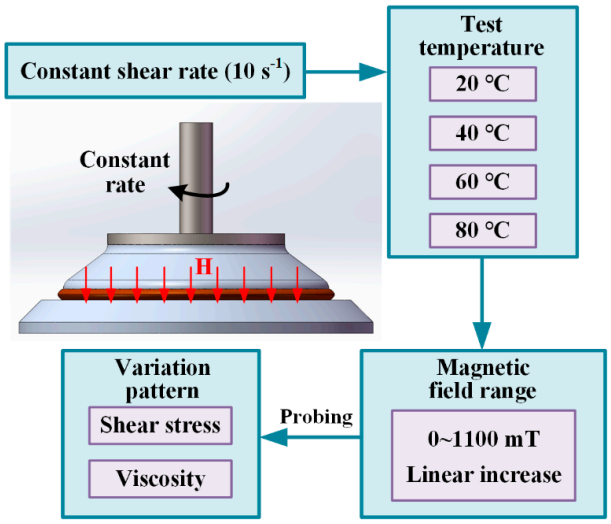


Figure 2. Magnetorheological grease magnetic field scanning scheme.

The rate scanning experiments for the magnetorheological grease were conducted according to the experimental design shown in Figure 3. The experiments were performed at four temperature conditions: 20 °C, 40 °C, 60 °C, and 80 °C, and five magnetic field intensity conditions: 0 mT, 55 mT, 110 mT, 220 mT, and 440 mT, resulting in a comprehensive cross-trial. The shear rate exponentially increased from 0.01 s^{-1} to 100 s^{-1} over time. To ensure the accuracy of the experimental data, a pre-shearing process was carried out at a shear rate of 0.01 s^{-1} for 72 s, followed by a 480 s increase in shear rate. Data points were recorded every 12 s to investigate the variation in shear stress and apparent viscosity with changes in the shear rate at each temperature.

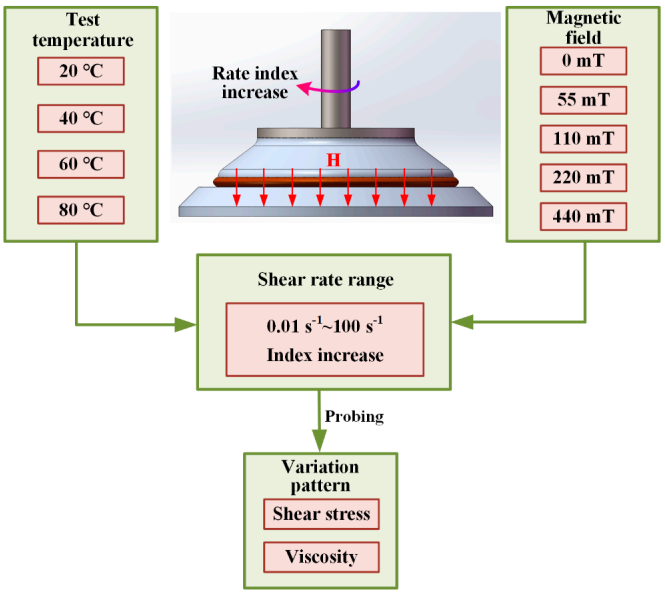


Figure 3. Magnetorheological grease rate scanning scheme.

4. Results and Discussion

In Figure 4, the magnetic field scanning curves of the magnetorheological grease at different temperatures are depicted. From the graph, it can be observed that the shear stress of the magnetorheological grease decreased gradually as the temperature increased under zero field intensity. This phenomenon could be attributed to the reduction in the entanglement degree of the fibrous network structure formed by the base carrier fluid and the lubricating grease soap fibers, resulting in a decrease in the damping force during the

shearing process of the rotational rheometer. With an increase in magnetic field intensity, the shear stress of the magnetorheological grease gradually increased at each temperature [14]. However, when the magnetic field intensity reached 400 mT, the shear stress tended to stabilize and exhibited a slight decrease. This behavior could be explained by the alignment of magnetic particles in the magnetorheological grease along the magnetic field direction, forming chain-like structures with a certain yield strength. Consequently, the structural strength of the magnetorheological grease increased, leading to an increase in the damping force during the shearing process of the rotational rheometer. However, at 400 mT, the magnetic particles in the magnetorheological grease reached saturation magnetization, and the chain-like structures no longer strengthened. Furthermore, due to the continuous shear action, the degree of entanglement of grease soap fiber decreased, the phenomenon of shear thinning occurred, and the damping force in the shear process of the rotary rheometer decreased slightly. This was because, under the influence of an external magnetic field, the magnetic particles within the magnetorheological grease aligned themselves in chain-like structures along the direction of the magnetic field due to magnetic torque. With the increase in magnetic field intensity, the number of magnetic chains increased, and their structural strength was enhanced. The effect of these magnetic chains was similar to that of the soap fibers, leading to an increase in the apparent viscosity of the magnetorheological grease. Therefore, under high-temperature conditions, the application of an external magnetic field could mitigate the shear thinning phenomenon of magnetorheological grease.

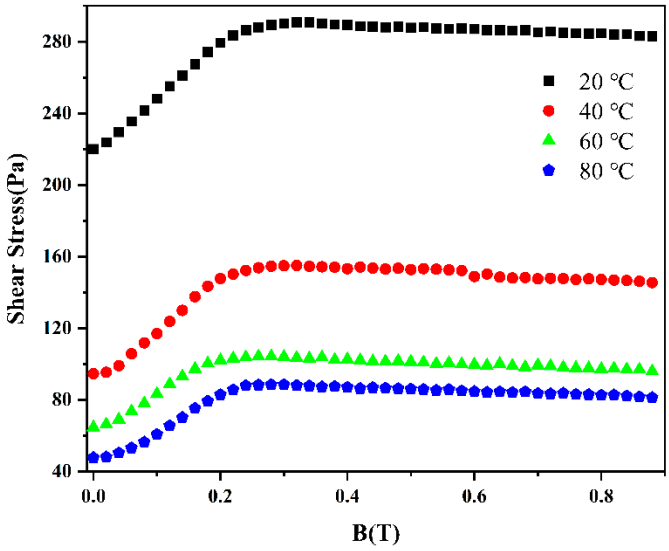


Figure 4. Magnetorheological grease magnetic field scanning curve.

Viscosity served as a significant parameter for quantifying the frictional forces within magnetorheological grease. In the context of Newtonian fluids, the viscosity remained constant regardless of changes in shear rate. Conversely, non-Newtonian fluids exhibited varying viscosity with alterations in shear rate. Magnetorheological grease falls under the category of non-Newtonian fluids, and its viscosity is referred to as apparent viscosity. The formula for apparent viscosity was derived by referencing the viscosity formula applicable to Newtonian fluids [24,25].

$$\eta = \frac{\tau}{\dot{\gamma}} \tag{1}$$

where η is apparent viscosity, unit Pa · s; τ is shear stress, unit Pa; and $\dot{\gamma}$ is the shear rate, unit s^{-1}.

Please refer to Equation (1). It was observed that the apparent viscosity of magnetorheological grease increased linearly with the increase in shear stress when a constant shear rate was applied. As shown in Figure 4, it can be inferred that at zero field intensity, the apparent viscosity of magnetorheological grease gradually decreased with increasing

temperature. Furthermore, as the magnetic field intensity increased, the apparent viscosity of magnetorheological grease gradually increased at all temperatures. When the magnetic field intensity reached 400 mT, the apparent viscosity tended to stabilize and exhibited a slight decrease. The reduction in the viscosity of magnetorheological grease after reaching saturation magnetization was attributed to its shear thinning effect.

In Figure 5, the change in the viscosity of magnetorheological grease with the shear rate at different temperatures is depicted. It is observed from the graph that the viscosity of magnetorheological grease decreased gradually with the increase in shear rate, indicating a shear thinning phenomenon. Without a magnetic field, the viscosity of magnetorheological grease decreased gradually with increasing temperature. This could be attributed to the decrease in the entanglement degree of soap fibers within the magnetorheological grease, allowing the base oil to flow out more easily and resulting in viscosity reduction. At the same temperature, with the enhancement of magnetic field intensity, the apparent viscosity of the magnetorheological grease increased progressively. At 60 °C, the apparent viscosity of the magnetorheological fluid showed a relatively small increase with increasing magnetic field strength. It was observed that at 440 mT, the apparent viscosity of the magnetorheological fluid was the lowest compared to 20 °C, 40 °C, and 80 °C. This phenomenon can be primarily attributed to the reduced entanglement of soap fibers within the magnetorheological fluid at 60 °C due to the elevated temperature, resulting in a decrease in apparent viscosity compared to 20 °C and 40 °C. Furthermore, although the entanglement degree of soap fibers within the magnetorheological fluid decreased at 60 °C compared to 80 °C, the residual entanglement structure inhibited the chain formation behavior of magnetic particles, leading to a reduction in apparent viscosity. Conversely, at 80 °C, the entanglement degree of soap fibers within the magnetorheological fluid reduced significantly, resulting in a significant increase in the quantity of chain-formed magnetic particles, and consequently, an increase in apparent viscosity [15].

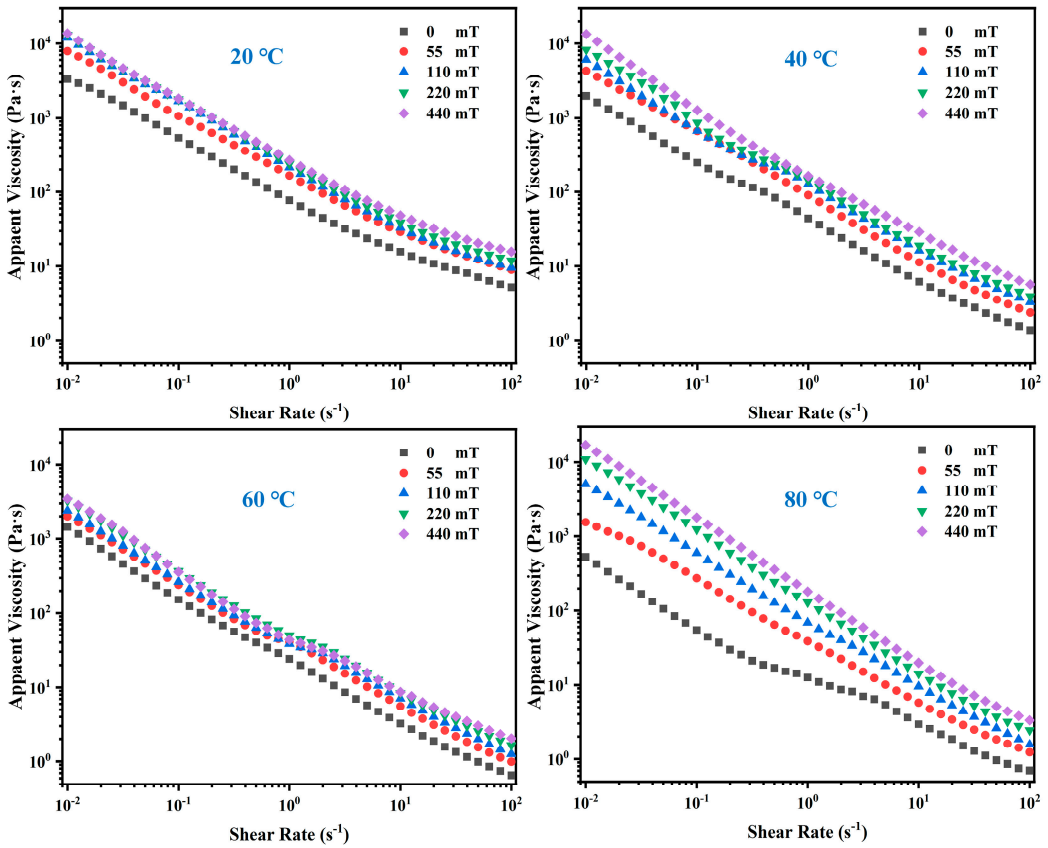


Figure 5. Magnetorheological grease apparent viscosity curve.

The flow characteristics of magnetorheological grease under varying temperatures and magnetic fields are shown in Figure 6. The graph indicates a gradual decrease in shear stress during the pre-shear stage as the temperature increased. This behavior can be attributed to the non-Newtonian properties of the grease, which result in a nonlinear increase in shear stress with higher shear rates. Notably, increasing magnetic field intensity led to a more pronounced increase in shear stress within the magnetorheological grease. It can be noted from the figure that the shear stress at 60 °C was disorganized as the shear rate changed. This was because the degree of entanglement of soap fibers in magnetorheological grease decreased at 60 °C. The shear stress was supported by soap fibers and magnetic chains, and the disorganization phenomenon occurred due to the obstruction of soap fibers in the chain formation process [26]. At elevated temperatures (80 °C), the disruption phenomenon of shear stress within the grease became less evident. This observation can be primarily attributed to the comparatively higher concentration of magnetic particles. When subjected to an external magnetic field, numerous magnetic chains were formed, exhibiting a certain level of structural strength. As the magnetic field intensity strengthened, the clustered magnetic chains progressively increased in structural integrity, reaching maximum strength at the saturation magnetization intensity. Consequently, this resulted in an increasing damping force throughout the shearing process, peaking at the saturation magnetization intensity. The graph further reveals that within a specific range of shear rates, the shear stress of the grease remained relatively constant. This phenomenon was predominantly associated with the occurrence of shear yield in the magnetorheological grease; thus, the consistent shear stress value represented the shear yield stress [25]. Moreover, the graph indicates a gradual increase in shear yield stress with stronger magnetic field intensity, while it decreased with rising temperatures.

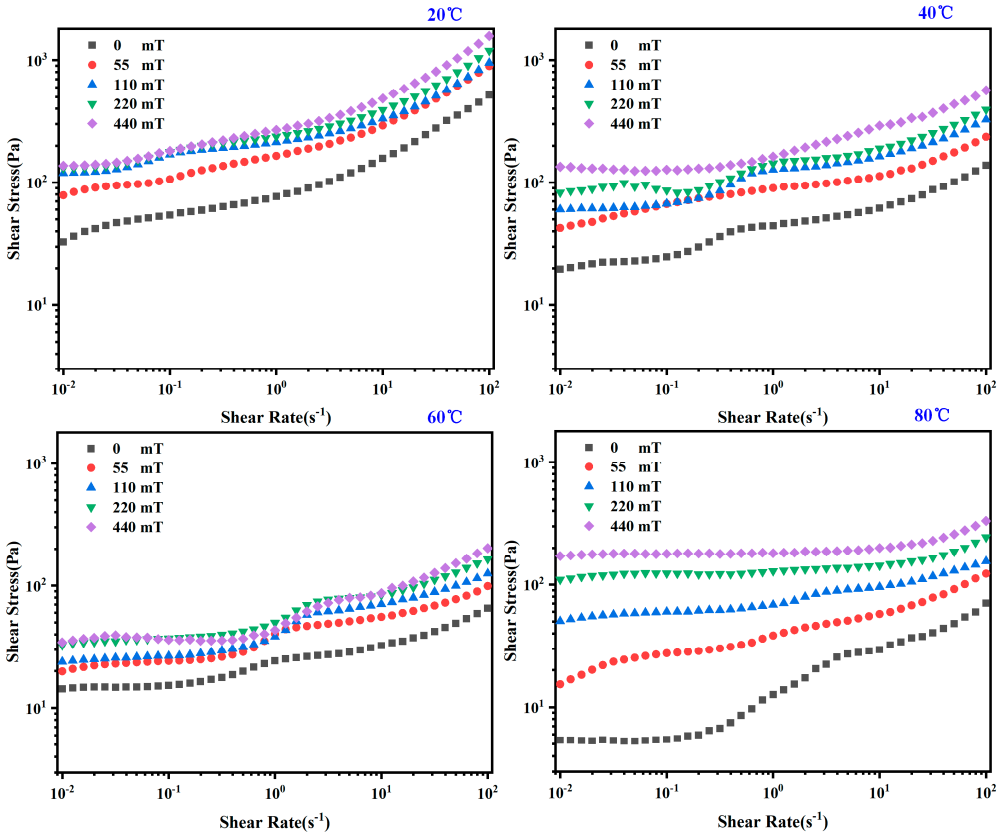


Figure 6. Magnetorheological lipid flow curve of magnetic particles.

Due to the inability of Figure 6 to clearly depict the region where shear stress remained relatively constant with increasing shear rate, making the shear yield phenomenon of the magnetorheological grease less apparent, the extrapolation method based on the curve

was not feasible. However, the Herschel–Bulkley (H-B) rheological model could be used by fitting the data to determine the shear yield stress of the magnetorheological grease. Formula 2 represents the H-B rheological model [27].

$$\tau = \tau_0 + k\dot{\gamma}^n \quad (2)$$

where τ is the shear stress, unit Pa; τ_0 is the shear yield stress, unit Pa; k is the consistency coefficient, unit Pa · s^n; $\dot{\gamma}$ is the shear rate, unit s^{-1}; and n is the shear thinning index.

The rheological data were substituted into the H-B rheological model for data fitting, and rheological parameters at different temperatures and magnetic field intensities were obtained, as shown in Table 3.

Table 3. Parameters of magnetic particle magnetorheological grease H-B model.

Temperature/°C	Rheological Parameter	Magnetic Field Intensity/mT				
		0	**55**	**110**	**220**	**440**
20	Yield stress/Pa	44.75	98.03	149.6	161.1	163.3
	Consistency coefficient/Pa · s^n	28.42	51.44	44.05	54.51	77.2
	Shear thinning index	0.3774	0.3555	0.3416	0.3156	0.3354
40	Yield stress/Pa	19.51	51.65	42.68	80.32	110.2
	Consistency coefficient/Pa · s^n	19.31	28.45	44.78	43.65	59.96
	Shear thinning index	0.4344	0.4291	0.4145	0.4135	0.4061
60	Yield stress/Pa	12.99	21.62	27.75	28.85	30.95
	Consistency coefficient/Pa · s^n	8.333	25	31.37	27.39	28.97
	Shear thinning index	0.5412	0.5296	0.5311	0.5342	0.5132
80	Yield stress/Pa	8.223	12.78	18.68	21.26	21.57
	Consistency coefficient/Pa · s^n	7.852	17	22.39	23.94	22.27
	Shear thinning index	0.6396	0.5915	0.6027	0.6129	0.6048

As shown in Figure 7, a trend of variation in the H-B model parameters of the magnetorheological grease with changing magnetic field intensity at different temperatures was observed. The results showed that the rheological parameters of the magnetorheological grease exhibited a certain trend with the changes in temperature and magnetic field intensity.

1. At the same temperature, with an increase in magnetic field intensity, the yield stress and consistency coefficient of the magnetorheological grease gradually increased, while the shear thinning index exhibited a slight decrease. This phenomenon can be attributed to the enhanced magnetic field intensity, which aligned a greater number of magnetic particles in a chain-like structure along the direction of the magnetic field, resulting in a certain yield strength of the magnetorheological grease [14]. The coupling effect between this chain-like structure and the fibrous structure of the grease soap led to an increase in the grease's consistency and consistency coefficient. As the magnetic field intensity increased, not only did the number of magnetic particle chain-like structures increase, but their structural strength also intensified. Consequently, the shear thinning effect of the magnetorheological grease weakened.
2. At a constant magnetic field intensity, an increase in temperature led to a gradual decrease in the yield stress and consistency coefficient of the magnetorheological grease, while the shear thinning index showed an upward trend. This phenomenon could be attributed to the rising temperature, which resulted in a decrease in the entanglement degree of the soap fibers present in the base carrier lubricant of the magnetorheological grease. As a consequence, the strength of the fibrous structure weakened, facilitating the flow of the oil component within the magnetorheological grease. Consequently, the yield strength and consistency of the grease decreased. The decrease in the entanglement degree of the soap fibers, coupled with the characteristic

shear thinning behavior of the magnetorheological grease, resulted in an intensified shear thinning effect.

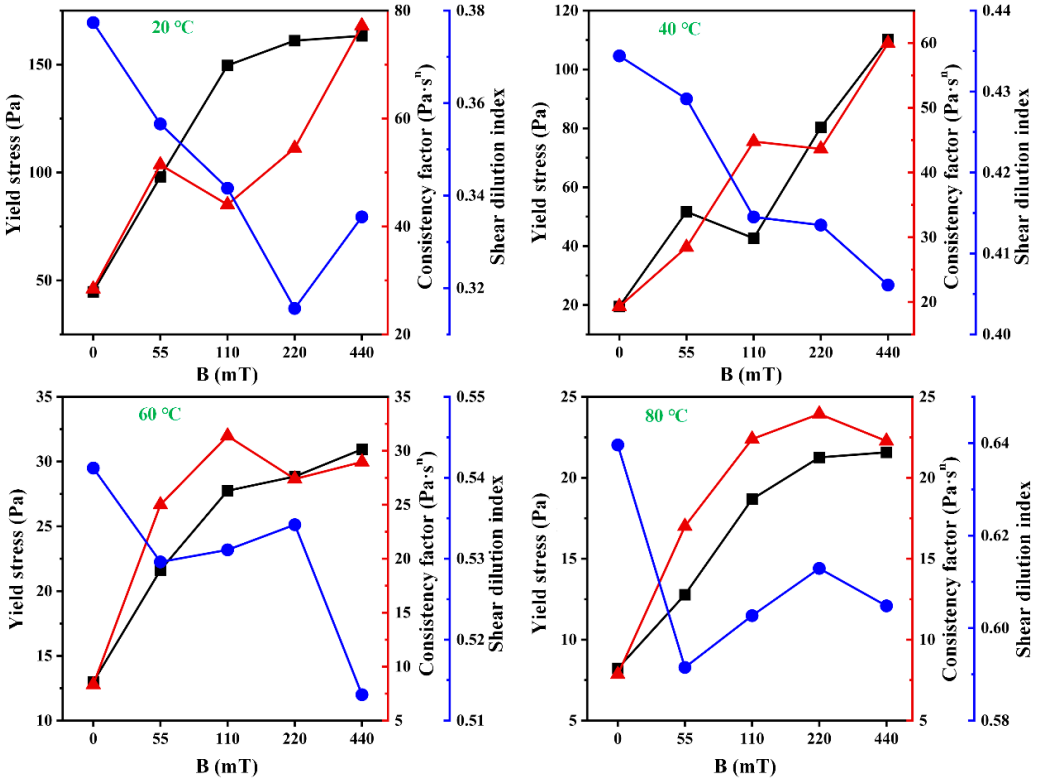


Figure 7. Parameters of magnetic particle magnetorheological grease H-B model. Black represents yield stress, red represents consistency coefficient, and blue represents shear thinning index.

Based on the above observations, it can be concluded that within a certain range, an increase in magnetic field intensity corresponded to an increase in the shear yield stress and consistency index, accompanied by a decreased shear thinning effect. This implies that a higher magnetic field intensity could impede the flow properties of the magnetorheological grease. Conversely, a decrease in temperature resulted in a reduced shear yield stress and consistency index, leading to an improved shear thinning effect. Therefore, an increased magnetic field intensity could enhance the flowability of the magnetorheological grease.

5. Conclusions

This study utilized nanoscale $MnFe_2O_4$ particles as magnetic fillers to prepare a 15% mass fraction of magnetorheological grease, utilizing NLGI00 lithium-based grease as the base fluid. The prepared magnetorheological grease underwent a comprehensive investigation of its thermomagnetic coupled rheological properties. The variation patterns of shear yield stress with increased magnetic field intensity at different temperatures were explored by fitting the rheological data using the Herschel–Bulkley (H-B) rheological model. The main conclusions obtained were as follows.

1. At a constant shear rate without the presence of an applied magnetic field, an increase in temperature resulted in a decrease in both the shear stress and apparent viscosity of the magnetorheological grease. Under constant shear rate and temperature conditions, the shear stress and apparent viscosity of the magnetorheological grease increased with the intensification of the magnetic field intensity. However, once the magnetic field intensity reached 400 mT, further enhancement of the magnetic field had a negligible effect on the shear stress and apparent viscosity. Furthermore, under the same temperature and magnetic field conditions, an increase in shear rate resulted in a gradual reduction in the apparent viscosity of the magnetorheological grease.

2. Under a constant applied magnetic field intensity, raising the temperature resulted in a decrease in the extent of the entanglement of soap fibers within the magnetorheological grease, leading to a reduction in damping force and shear stress. In the same temperature range, with the intensification of the applied magnetic field intensity, the load-bearing capacity of magnetic chains increased, resulting in an elevation of shear stress.
3. Under the same magnetic field intensity, with the increase in temperature, the shear yield stress and consistency coefficient of the magnetorheological grease gradually decreased, while the shear thickening index progressively increased. Similarly, with the augmentation of the externally applied magnetic field intensity at a constant temperature, the shear yield stress and consistency coefficient of the magnetorheological grease gradually increased, whereas the shear thickening index gradually decreased.

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