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Abstract: The rheological properties of lithium grease and polyurea grease at different temperatures and consistencies were determined with a rotary rheometer. The plateau moduli of the greases were calculated, and the mechanism of influence of consistency and temperature on the rheological properties of the greases was explained. The tribological and wear properties of the two greases were measured by high-temperature friction and wear tester. The friction and wear mechanisms are probed by the rheological properties of lubricating grease. The results show that the plateau modulus G_N can be used to assess the structural strength of different greases, but it can only assess the degree of entanglement of the same grease. The higher the consistency of the grease, the larger the apparent viscosity, structural strength, and yield stress. The rheological properties of PAO-polyurea grease are greatly affected by temperature, but its structural strength is better than that of mineral oil-lithium grease. The consistency of mineral oil-lithium grease is expected to affect the friction coefficient and wear through its influence on the grease's structural strength and film-forming ability. For PAO-polyurea, the consistency in a certain range has little effect on the friction coefficient and wear resistance.

Keywords: grease; consistency; thickener fiber; rheology; tribology; boundary lubrication

1. Introduction

The use of lubricating grease can reduce friction and wear on the relative moving surface. Consistency, as a key parameter pertaining to any grease, is affected by thickener concentration, manufacturing processes and other factors. Grease is widely used in the lubrication of 90% of ball bearings due to its advantages of sealing gaps and preventing pollutants from entering equipment [1,2]. Lithium grease and polyurea grease are the two most widely used types [3], with good mechanical stability. Lubrication is critical for reducing frictional losses in electric motors and transmissions, and as the trend toward electric vehicles drives, the performance of greases has become more complex [4,5]. However, the mechanism of the influence of consistency on the rheological properties and friction and wear properties of lubricating greases are yet to be elucidated. It is necessary to understand the rheological, tribological and wear properties of greases.

Lubricating grease is a colloidal dispersion system, in which the thickener fiber traps the base oil [6–8]. Through the study of the rheological properties of lithium grease, many factors, such as additives, thickener type and concentration were found to affect the rheological behavior of grease [7,9–12]. On this basis, Mao et al. [13], Couronne et al. [14], and Sánchez et al. [15] studied the relationship between the rheological properties and microstructure of lithium grease. Xu et al. [16] evaluated the effects of the amine molecular structure on the rheological properties of polyurea grease and found that the concentration of thickener and the viscosity of base oil will significantly affect the microstructure of grease. Maciej et al. [17] assessed the boundary characteristics of lithium-based grease



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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). and polyurea grease on different materials, finding that different surface materials have different adsorption capacities for thickener fibers and form different boundary layers. The microstructure of grease will further affect its tribological properties. Fan et al. [3] compared the tribological properties of lithium grease and polyurea grease and found that thickener not only determines the physical properties of grease but is was also involved in the lubrication process for reducing friction and wear. In steel/steel friction pairs, the lubrication performance of polyurea grease is better than that of lithium grease. Cousseau et al. [18] found that the interaction between grease thickener and base oil exerts a significant influence on bearing friction torque. Gonçalves et al. [19] studied the influence of grease thickener content on friction torque on rolling bearings, finding that the increase of thickener content will reduce the sliding coefficient of friction and oil-bleeding capacity of grease. Some studies show that good additives can improve the tribological properties of grease [1,20,21]. Li et al. [1] found that by adding a few layers of graphene (FLG) to lithium grease, it can improve its anti-friction and anti-wear properties. Wu et al. [21] found that CuO nanoparticles can improve the tribological properties of polyurea grease, inhibiting bearing vibration. The tribological properties of lubricating grease are rarely combined with rheological properties and are rarely described systematically.

Lubricating grease tends to separate out the lubricating oil under certain temperatures or shear stresses. The separation of soap and oil directly leads to a change of consistency and loss of grease. Trace oil separation can keep equipment lubricated, but excessive oil separation caused by increasing temperature will both thicken and harden a grease, resulting in thermal aging [22]. The thermal-aging temperatures of lithium grease and polyurea grease at different temperatures are 130 °C and 280 °C, respectively. The high temperature in the running process of roller bearings will lead to the thermal aging of grease, which is one of the most important reasons for severe local wear of roller bearings [23]. Pan et al. [24] simulated the static thermal degradation of grease in a drying oven; the results show that lithium grease has better anti-friction and anti-wear properties after heat treatment at 120 °C. The temperature will affect the rheological properties and microstructure of grease [15]; it is, therefore, important to explore the rheological properties of grease at different temperatures.

In the present work, the rheological properties and tribological properties of lubricating greases were measured. The influence mechanism of consistency and temperature on the rheological properties and tribological properties and the correlation between the rheological properties and tribological properties were investigated. The research results based on the rheological and tribological characteristics of grease will provide a theoretical basis for selecting the most appropriate grease for use under different working conditions.

2. Materials and Methods

2.1. Materials

Seven types of grease samples were selected as research objects. According to ISO 2137-1985 (E), the cone penetration of grease is the depth after the standard cone falls freely in a standard grease cup filled with the tested grease for 5 s at 25 °C, and the unit is 0.1 mm. Cone penetration data are used to indicate the consistency of grease. All grease samples were provided by Sinopec Lubricating Oil Co., Ltd. (Tianjin, China). The viscosity of the base oil is 48 mm²/s at 40 °C. The nomenclature and basic parameters of these seven grease samples are listed in Table 1. The basic parameters of base oil are given in Table 2. The selected greases are mainly used for sealed bearings. In NGLI grades 2 and 3, four cone penetration values are selected as the research object. Lithium grease with different consistencies is named Li-230 to Li-300, respectively. The polyurea greases with different consistencies are named Po-230 to Po-300, respectively. The samples are milky white with a hard texture and good stability.

Grease	Base Oil	Thickener Type	Viscosity of Base Oil (mm²/s)	Unworked Penetration (0.1 mm)
Li-230 Li-250 Li-270 Li-300	Paraffin based mineral oil	Lithium 12 Hydroxystearate	48 ± 0.1	$\begin{array}{c} 230 \pm 6.7 \\ 250 \pm 7.0 \\ 270 \pm 6.5 \\ 300 \pm 7.1 \end{array}$
Po-230 Po-250 Po-300	PAO	Cyclohexylamine, 18 amine and isocyanate	48 ± 0.1	$\begin{array}{c} 230 \pm 6.8 \\ 250 \pm 7.2 \\ 300 \pm 7.0 \end{array}$

Table 1. Main components and properties of the tested greases.

Table 2. The basic parameters of base oil at 40 °C.

Base Oil	Dynamic Viscosity η (mPa·s)	Viscosity-Pressure Coefficient α (10 ⁻⁸ Pa ⁻¹)
Paraffin based mineral oil	42.24 ± 0.08	1.94
PAO	39.36 ± 0.09	1.65

2.2. Microstructure Characterization

The microstructures of the greases were observed by scanning electron microscopy (SEM, Hitachi Regulus 8220, Tokyo, Japan). Before observation, the grease was soaked with petroleum ether and a mixing of grease and the organic solvent was promoted by means of ultrasonic dispersion. Static 2H was used to facilitate the extraction of the base oil from grease by petroleum ether. Further centripetal separation of the mixture of grease and petroleum ether was conducted by means of a high-speed centrifuge. The separated thickener was then dissolved in petroleum ether and a suspension was prepared by ultrasonic dispersion; this step was repeated three or four times until the base oil of the grease was completely extracted; the specimen was then coated with gold in preparation for examination by SEM.

2.3. Rheological Test

To investigate the effects of temperature, consistency and the type of grease on the rheological properties, steady-state rheological and dynamic rheological tests of grease were conducted using a rheometer (MCR302, Anton Paar, Graz, Austria) at 30 °C, 70 °C, and 130 °C. The maximum torque applied by the rheometer was 200 mN·m with a torque resolution of 0.1 nN·m. The steady-state rheological experiment was conducted at a controlled shear strain rate, and the main parameters were apparent viscosity η_{ap} , shear stress τ , and shear strain rate $\dot{\gamma}$. Dynamic rheological experiments were conducted under sinusoidal vibration of the sample within a certain frequency range to study dynamic effects thereon. Cone plate cp-25 and flat plate pp-25 were selected for the rotor. At the beginning of the test, the grease was pre-sheared for 120 s, then allowed to stand for 30 min to eliminate damage to the grease when the rotor was pressed down onto the specimen.

In steady-state tests, the shear strain rate ranged from 0.1 to 1000 s^{-1} , apparent viscosities η_{ap} at different shear strain rates were obtained, the flow curve was generated, and the yield stress τ_y was determined. In dynamic rheological experiments, the strain ranged from 0.01% to 100%, the constant frequency was 10 rad/s, and the linear viscoelastic region of the grease was determined. In the linear viscoelastic state, a frequency-scanning experiment was conducted, and the frequency ranged from 0.1 to 100 rad/s, and the constant strain was 0.1%. All specimens were taken from the same sampling position in the same batch, and each was tested at least three times to reduce the influence of errors in the testing process.

2.4. Tribological Test

Before the experiment, the film thickness ratio λ is obtained by calculating the ratio of the minimum oil film thickness to the surface roughness of the friction pair, as shown in Equation (1).

$$\lambda = h_{\min} / \sqrt{\sigma_1^2 + \sigma_2^2},\tag{1}$$

In Equation (1): h_{\min} is the minimum oil film thickness of the grease (µm), σ_1 and σ_2 is the roughness Rq of the surfaces of the two friction pairs. The minimum oil film thickness under point contact can be estimated by Hamrock–Dowson formula [25].

$$h_{\min} = 3.63 \overline{U}^{0.68} \overline{G}^{0.49} \overline{W}^{-0.073} (1 - e^{-0.68k}) R,$$
⁽²⁾

In Equation (2): $U(= \eta U/(E^*R))$ is the dimensionless speed parameter, $G(= \alpha E^*)$ is the dimensionless material parameter, $\overline{W}(= W/(E^*R^2))$ is the dimensionless load parameter. k is the ellipticity. E^* is the equivalent elastic shear modulus. R is the equivalent radius of curvature.

The tribological properties of greases with different consistencies were measured by high-temperature friction and wear tester (CSM THT 1000 °C, Anton Paar, Graz, Austria). The contact form of the friction pair was of the ball-disk-type. The lower specimen was a GCr15 metal disk measuring 55.0 mm × 10 mm, and the upper specimen was a Q195 carbon steel ball with a diameter of 6 mm, on which the load W was 5 N, the maximum Hertz contact stress was 1.17 GPa, the rolling speed U was 1.26 m/s, the rotation radius was 20 mm, the applied frequency was 10 Hz, for a duration of 1 h at a temperature of 25 °C. The surface roughness Rq of the ball and disk measured by the 3D white light interference profilometer was about 0.40 µm and 0.26 µm, respectively. The film thickness ratios of the two greases were 0.23 and 0.21, respectively. It is generally believed that $\lambda < 1$, the lubrication status is boundary lubrication.

Before the experiment, we placed the ball and disk specimens into petroleum ether for ultrasonic cleaning (three times), then fixed the disk specimens and evenly coated an appropriate amount of grease over the friction surface before starting the test after fixing the ball specimen. The morphologies of the worn surfaces were examined in optical microscopy (MIT500, Cnoptec, Chongqing, China), and the wear spot area was analyzed to measure the anti-wear performance of grease in a more intuitive manner. Tribological experiments were carried out three times to avoid randomness in the experiment.

3. Results and Discussion

3.1. Microstructure and Plateau Modulus of Greases and Their Correlation

The microstructure of the grease is shown in Figure 1. The lithium grease, with a consistency of 230 and 300, is demonstrated in Figure 1a,b, respectively. The thickener fiber of the lithium grease shows a highly entangled reticulated structure. The microstructure of polyurea grease is very different from lithium grease, as shown in Figure 1c,d. Its thickener fiber is strip-shaped or rod-shaped and contains more oil. It is difficult to clean the base oil in polyurea grease with an organic solvent. The greater the consistency (the smaller the cone penetration) of lithium grease and polyurea grease, the thicker the fibers that were present per unit volume, the finer the structure, and the higher the range of entanglement between fibers.

The better to describe the microstructures of soap fibers, the plateau modulus G_N is introduced to represent the degree of entanglement between grease soap fibers. G_N can be obtained by frequency-scanning experiments and extrapolation from the data. This gives the storage modulus corresponding to when the loss coefficient reaches the minimum value, as shown in Equation (3).

$$G_{\rm N}^0 = \left[G'\right]_{\tan\delta \to \rm minimum} \tag{3}$$



Figure 1. Microstructure of greases with different consistencies: (a) Li-230, (b) Li-300, (c) Po-230, (d) Po-300.

Figure 2a shows the plateau modulus values of lithium greases at 30 °C, 70 °C, and 130 °C, and Figure 2b shows the plateau modulus values of polyurea greases at 30 °C, 70 °C, and 130 °C. The plateau moduli of the two greases increase with increasing consistency at the same temperature because the greater the consistency, the more soap fibers and the higher the degree of entanglement between fibers, as evinced by the SEM images. This shows that the plateau modulus can be used to characterize the microstructure of lubricating grease. The larger the plateau modulus, the more entangled the grease thickener fiber. At the same time, the plateau modulus of the grease at low temperature is larger than that at high temperature because, with the increase in temperature, the intermolecular force decreases, and the degree of entanglement of the grease fiber decreases, and the plateau modulus decreases. Comparing Figure 2a,b, the plateau modulus of lithium grease is much lower than that of polyurea grease, but the SEM image (Figure 1) does not show the degree of entanglement of lithium grease soap fiber as lower than that of polyurea grease. For the same type of grease, the degree of entanglement of soap fibers is high, the probability of chemical and physical cross-linking between fibers is greater, the structural framework is more stable, and the structural strength is greater. Through the results of Figures 1 and 2, we obtained new conclusions that are different from past literature: The plateau modulus $G_{\rm N}$ can be used to assess the structural strength of different greases, but it can only assess the degree of entanglement of the same grease. For example, in Figure 1. In Figure 2, the structural strength of the polyurea greases is stronger than that of lithium greases, but the degree of entanglement of the polyurea greases is not as good as that of the lithium greases. At different temperatures, the plateau modulus of lithium grease is slightly different; the plateau modulus of polyurea decreased with the increasing temperature. This shows that, with the increase in temperature, the degree of entanglement of the soap fiber of polyurea grease decreases to a significant extent, while that of the soap fiber of lithium grease decreases slightly, which shows that the degree of thickener fiber entanglement of mineral oil-lithium grease is less affected by temperature, and the degree of thickener fiber entanglement of PAO-polyurea grease is greatly affected by temperature.



Figure 2. Plateau modulus G_N of greases at different temperatures: (a) lithium greases, (b) polyurea greases.

3.2. Apparent Viscosity of Grease

Consistency is an important factor that affects the performance of the grease. The consistency of grease is expressed by cone penetration. Figures 3–8 shows the maximum relative standard deviation (RSD) of each curve. Figures 3 and 4 show the variation of the apparent viscosity of the lithium greases and polyurea with the shear strain rate at different temperatures: the apparent viscosity of grease decreases with the increase in shear strain rate (i.e., shear-thinning). With the increase in shear strain rate, the resistance of thickener fibers along the flow direction is small, and the viscosity decreases. With increasing shear strain rate, the height, width and length of the thickener fibers may decrease, and the thickener fibers themselves may fracture and rupture, gradually orienting their alignment, whereupon the shear-thinning effect gradually decreases, especially at very high shear strain rates (the apparent viscosity of the grease decreases slightly).



Figure 3. Variations in apparent viscosity with shear strain rate of lithium grease with different consistencies at different temperatures: (a) $30 \degree C$, (b) $70 \degree C$, (c) $130 \degree C$.

As can be seen from Figures 3 and 4, the higher the consistency (the smaller the cone penetration), the higher the apparent viscosity at the same shear strain rate. That is, under the premise of the same base oil viscosity, the increase in grease consistency will increase the apparent viscosity of the grease. From the SEM images and G_N values, it can be found that the high consistency of soap fibers has a high degree of entanglement, and the resistance of soap fibers is large along the direction of flow, thus increasing the apparent viscosity. The

shear-thinning rate of lubricating grease with different consistencies is similar, that is, the consistency exerts little influence on the shear-thinning rate.

Temperature is also an important factor affecting the rheological properties of grease. With the increase in temperature, the apparent viscosity of grease decreases, because even if the chemical structure of grease used at a higher temperature does not change, the change of physical entanglement of the associated colloidal dispersion system will weaken the original performance of the grease, resulting in the decrease of viscosity. With the increase in shear strain rate, the curve shows an unstable change, which is due to the shear yielding of the grease; the fiber structure of the grease is cut, and the apparent viscosity changes suddenly. The difference is that the apparent viscosity of polyurea grease is very unstable after shear yield at high temperature, and the data fluctuate, which may be related to the structure of polyurea grease having been significantly affected by temperature (the plateau modulus is significantly affected by temperature). At room temperature, the apparent viscosity of lithium grease is lower than that of polyurea grease. When the temperature is greater than 70 $^{\circ}$ C, the plateau modulus of polyurea grease decreases greatly, that is, the degree of entanglement of soap fiber decreases greatly. However, the plateau modulus of lithium grease decreases very little, that is, the degree of entanglement of soap fiber decreases very little. When the temperature is 70 °C or 130 °C, the apparent viscosity of polyurea lubricated grease decreases far too much and it tends to be less than the apparent viscosity of lithium grease. This is because the structural strength of PAO-polyurea is greatly affected by temperature. Because the apparent viscosity is related to the filmforming properties and friction characteristics of the grease, the apparent viscosity of PAO-polyurea grease is more affected by temperature than mineral oil-lithium grease. The film-forming properties and friction properties of PAO-polyurea grease are more affected by temperature than mineral oil-lithium grease. Polyurea is easier to form a film, but the friction is relatively large.



Figure 4. Variations in apparent viscosity with a shear strain rate of polyurea grease with different consistencies at different temperatures: (a) $30 \degree C$, (b) $70 \degree C$, (c) $130 \degree C$.

3.3. Rheological Properties of Grease

In the steady-state test, Figures 5 and 6 show the changes in shear stress with the shear strain rate of lithium grease and polyurea grease, which reflects the resistance of the fluidity. It can be seen that lithium grease and polyurea grease have common characteristics: the shear stress increases with the consistency of the grease due to enhancement in inner compositions interactions. When the temperature is 30 °C the interaction between grease molecules is strong, the entanglement degree of soap fiber is high, and the wall surface only has weak adsorption to grease molecules; the wall slip will occur, that is, the shear stress will decrease in the initial stage. When the shear stress reaches the platform area, the corresponding value is the yield stress τ_y under the steady-state test of grease, and

then the shear stress increases with the increase of the shear strain rate. At this stage, the grease begins to flow. In this process, the grease has experienced wall slip—a solid-like deformation-yield-flow. When the temperature is 70 °C and 130 °C, the intermolecular force in the grease decreases, and the influence of the wall surface on the adsorption of grease molecules increases, so there is no wall-slip phenomenon at the beginning. However, with the increase of the shear strain rate, the shear stress increases, and the adsorption of the molecules on the wall surface is difficult to maintain, thus the wall surface slip begins to appear after the grease reaches the first turning point (upper yield point). When the second turning point (lower yield point) is reached, the grease begins to flow, and the shear stress at this turning point is the yield stress τ_y . In this process, the grease has experienced a solid-like deformation-yield coupling with wall slip-flow. The yield stress of lithium and polyurea grease increases with the increase of consistency, which means that the greater the consistency of the grease, the worse the fluidity. It should be pointed out that the entanglement between the thickener fibers of polyurea with a consistency of 300 is lower at 30 °C, so the curve shape is similar to that at the high temperature shown in Figure 6.



Figure 5. Variations in shear stress of lithium grease with shear strain rate at different temperatures: (a) $30 \degree C$, (b) $70 \degree C$, (c) $130 \degree C$.



Figure 6. Variations in shear stress of polyurea grease with shear strain rate at different temperatures: (a) $30 \degree C$, (b) $70 \degree C$, (c) $130 \degree C$.

In the dynamic test, the variation of storage modulus G' and loss modulus G' with strain or stress can be obtained. G'reflects the internal elastic potential energy of the material during deformation and is related to the retention capacity of the grease; G"refers to the

energy dissipated as heat during deformation. Both are key indices used to measure the viscoelastic properties of lubricating greases. The higher the viscoelasticity, the worse the fluidity. In dynamic experiments, the region before the storage modulus is decreased by 10% and is defined as the maximum linear viscoelastic region Ld. The strain-scanning curves of lithium grease and polyurea grease with different consistencies at different temperatures are shown in Figures 7 and 8, respectively: in the L_d region, the storage modulus of the grease at all temperatures is higher than the loss modulus, and it is in a solid state. The greater the consistency, the higher the storage modulus and loss modulus. It can be found that the storage modulus and loss modulus of polyurea grease are higher than those of lithium grease for the same consistency by comparing Figures 7 and 8.



Figure 7. Strain-scanning curves of lithium grease with different consistencies at different temperatures: (**a**) $30 \degree C$, (**b**) $70 \degree C$, (**c**) $130 \degree C$.



Figure 8. Strain-scanning curves of polyurea grease with different consistencies at different temperatures: (**a**) 30 °C, (**b**) 70 °C, (**c**) 130 °C.

With the increase in shear strain, the storage modulus decreases, and the loss modulus increases, indicating that the grease is undergoing a transition to a liquid state. The intersection of the storage modulus curve and the loss modulus curve is defined as the flow point, and the stress corresponding to the flow point is defined as the cross-stress τ_{co} . There is a positive correlation between the magnitude of the cross-stress and the resistance to flow, which is of great significance to the study of the fluidity of grease. Figure 9a shows the cross-stress of lithium grease with different consistencies at 30, 70, and 130 °C. Figure 9b shows the cross-stresses of polyurea greases with different consistencies at 30, 70, and 130 °C. The two greases have similar characteristics, such that, at the same temperature, the

greater the consistency, the greater the cross-stress τ_{co} ; at different temperatures, the crossstress of the grease with the same consistency decreases with the increase of temperature. The difference between the two greases is such that at 30 °C and 70 °C, the cross-stress of polyurea grease is much greater than that of lithium grease; however when the temperature rises to 130 °C, the cross-stress of polyurea grease is an approach to that of lithium grease, which means, the fluidity of lithium grease is better than the polyurea grease. The higher the temperature, the smaller the difference in fluidity between the two greases.



Figure 9. Cross-stress τ_{co} of greases with different consistencies at different temperatures: (a) lithium grease, (b) polyurea grease.

It can be seen from the above analysis that the higher the consistency of lithium grease and polyurea grease or the lower the temperature, the greater the shear stress and viscoelasticity. The difference is such that the fluidity of polyurea grease is lower than that of lithium grease, and its viscoelasticity is higher than that of lithium grease. With the increase in temperature, the fluidity of the two greases gradually increases, and their viscoelasticity gradually decreases.

3.4. Yield Stress and Structural Strength of Grease

Accurate structural strength assessments cannot be obtained only by means of the modulus of the plateau: the shear strength of the grease must be expressed by the magnitude of the yield stress. From the analysis of rheological properties, a yield stress exists in both steady-state and dynamic tests. The two yield stresses were compared, and the structural strength of the grease was measured.

In steady-state experiments, the yield stress τ_y of lithium grease and polyurea grease at various temperatures is shown in Figure 10a,b, respectively. At 30 °C, the yield stress of polyurea grease is much greater than that of lithium grease, indicating that the fluidity of lithium grease is better than that of polyurea grease at room temperature. At 70 °C, the yield stress of polyurea grease is close to that of lithium grease. At 130 °C, which is beyond the working temperature of the lithium grease, the yield stress of lithium grease with different consistencies is similar, which indicates that the structure of lithium grease soap fibers has been destroyed. At this temperature, the yield stress decreases, but it has no physical significance. The yield stress of polyurea grease with different consistencies remains quite different, that is, the microstructure of the polyurea grease is not destroyed at 130 °C.



Figure 10. Yield stress τ_y of greases under steady-state test conditions: (a) lithium grease, (b) polyurea grease.

In dynamic tests, the shear stress corresponding to L_d represents the yield stress under dynamic test conditions τ_d (Figure 11a,b): similar to the yield stress measured under steady-state conditions, the higher the consistency, the greater the yield stress. Comparing the yield stress τ_d in the dynamic test with the yield stress τ_y in the steady-state test, τ_d is shown to be much less than τ_y . In the steady-state experiment, the soap fiber is pulled in one direction; in the dynamic test, the soap fiber oscillates dynamically and shears back and forth. In practice, dynamic reciprocating shear is often easier to make the grease flow, than single direction shear, so the yield stress in the dynamic test will be less than that in a steady-state test. The comparison between Figure 11a,b shows that the dynamic yield stress of polyurea is greater than that of lithium grease in the experimental temperature range, which indicates the structural strength of polyurea grease is higher than that of lithium grease.



Figure 11. Yield stress τ_d of greases under dynamic test conditions: (**a**) lithium grease, (**b**) polyurea grease.

It can be seen from the above analysis that the similarities between lithium grease and polyurea grease are as follows: the higher the consistency or the lower the temperature, the higher the yield stress and structural strength. When the temperature rises to 130 °C, the performance of polyurea grease remains good. The structural strength of PAO-polyurea grease is higher than that of mineral oil-lithium grease; the higher the temperature, the smaller the structural strength between the two greases.

3.5. Tribological Properties of Grease

The grease forms a thin film on the friction surface, which can prevent direct contact between elements of the friction pair and reduce the friction and wear. The friction and wear characteristics of greases with different consistencies were studied, and the influence of the rheological properties of greases on friction and wear characteristics were assessed.

Under the conditions of 5 N, 10 Hz, 25 °C, for 60 min, the tribological properties of lithium grease and polyurea grease on bearing steel/carbon steel friction pairs were measured. Figure 12 shows the average value of three friction experiments. The wear spot diameter (WSD) of the ball specimen can better reflect the anti-wear performance of greases. The wear surfaces of the ball specimens lubricated with lithium grease and polyurea grease were compared, and the average values of the three measurements were taken to represent the wear spot diameter (Figure 13). Figure 14 shows the relationship between the ball wear rate and consistency, as well as the relationship between the ball wear rate and consistency.



Figure 12. Coefficient of friction of greases with different consistencies: (a) lithium greases, (b) polyurea greases.



Figure 13. Wear spot of greases with different consistencies: (**a**) Li-230, (**b**) Li-250, (**c**) Li-300, (**d**)Po-230, (**e**) Po-250, (**f**) Po-300.



Figure 14. Wear rates and WSD of greases with different consistencies: (a) WSD, (b) wear rates.

The friction test results for lithium grease are shown in Figure 12a: For Li-230, the COF peak at the initial run-in stage is almost absent, while the peak for Li-300 is the largest. This shows that at the beginning of the test, Li-230 forms a more stable grease film compared to Li-300. After a short initial period, the friction and wear on the friction pair surfaces enter into a steady-state period for Li-230 and Li-250, and the COF values for Li-230 and Li-250 tend to be stable, and their coefficient of friction (COF) is about 0.11, showing good friction stability. It is worth noting that the COF value for Li-300 is larger than normal in the whole test, which indicates that a stable grease film is not formed. The wear spots and wear rate for Li-230, Li-250 and Li-300 are shown in Figures 13a-c and 14, respectively; the smaller the consistency, the larger the wear spot diameter and wear rate, and the parallel groove for Li-300 is significantly larger than that for Li-230 and Li-250. According to the calculation results of the film thickness ratio λ in Section 2.4, the lubrication state between the friction pairs in the friction experiment is boundary lubrication. The frictional stress arises from asperity interaction and the shear stress in the grease. The increase in thickener consistency promotes better separation of surfaces. The thickener consistency for Li-300 is too small compared to Li-230 and Li-250, to form a thick thickener particle layer on the ball and disk surfaces, which results in more asperity contact and greater friction and wear. The stability of the friction coefficient during the whole experiment time for Li-230 and Li-250 shows that a thick layer of thickener particles is formed on the surface of the disk to reduce asperity interaction.

From the rheological results, it can be seen that the yield stress τ_y and the storage modulus G' characterizing the strength of grease structure for Li-230 is slightly larger than that for Li-250, which means the shear resistance for Li-230 is slightly larger, therefore, the COF of Li-230 is slightly larger than that of Li-250. However, the wear spot diameter and wear rate for Li-230 are smaller than that for Li-250, which results from the better separation of surfaces of friction pair by the thicker grease film formed through Li-230 with a greater consistency.

The friction test results for polyurea grease are illustrated in Figure 12b: compared with lithium grease, its consistency has little effect on its friction performance. As shown in Figures 13d–f and 14, the reduction in the consistency did not cause a significant change in the diameter of the wear scar and wear rate. The friction curves for polyurea greases also fluctuate, and the wear for polyurea greases is larger than that for lithium grease. These indicate that the three polyurea greases did not form stable oil film, which is related to the rheological properties of polyurea grease. High-structural stability of polyurea grease, which is reflected in high storage modulus and yield stress values, leads to a decrease in oil-bleeding ability, which causes a reduction in contact replenishment, which may result in a film thickness much lower than the estimated value.

4. Conclusions

- 1. Through the plateau modulus G_N and yield stress τ_y in the rheological experiment, the structural strength of grease can be obtained, the degree of entanglement of the same type of grease can be assessed, and then the tribological and wear properties of the grease can be predicted.
- 2. The increase in temperature will decrease the degree of fiber entanglement, apparent viscosity, storage modulus and structural strength of the greases. The rheological properties of PAO oil-polyurea grease are greatly affected by temperature, but its structural strength is better than that of mineral oil-lithium grease, and its fluidity is lower than that of mineral oil-lithium grease.
- 3. The higher the consistency (the smaller the cone penetration), the higher the apparent viscosity, yield stress (structural strength), and cross-stress (the worse the fluidity) due to the enhancement in inner compositions interactions of greases. The consistency exerts little influence on the shear-thinning rate.
- 4. In boundary lubrication state: The consistency of mineral oil-lithium grease is expected to affect the friction coefficient through its influence on the grease's structural strength and the film-forming ability. Higher consistency promotes better separation of the surfaces of friction pair to result in a reducing wear rate. For PAO-polyurea grease, the consistency, within a certain range, has little effect on the coefficient of friction and wear. The wear for PAO-polyurea grease is larger than that for lithium grease because the higher storage modulus and yield stress of PAO-polyurea grease lead to worse contact replenishment.

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