

## Article

# Elaboration of Ionic Liquids on the Anti-Wear Performance of the Reinforced Steel-Steel Contact Surface

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**Abstract:** This study conducted a tribological investigation of base oil (PAO6 and 5W 40) and ionic liquids (IL)-modified lubricants through a four-ball tribometer for 30 min. The lubricants were fabricated via a two-step method using stirring magnetic and ultrasonic dispersion. IL, base oil, and lubricants were, respectively, characterized by XRD and FTIR analysis. In addition, multiple characterizations such as EDS, 3D morphology, and SEM were carried out to evaluate the wear and friction performance of steel balls. Ultimately, the results showed that the coefficient of friction (COF) and wear scar diameter (WSD) of wear scar lubricated by IL-modified lubricants were greatly decreased than that by base oil. IL can well improve the tribological properties of PAO 6 oil and 5W-40 oil due to the tribo-film appearance on the friction surface of wear scar by the effective role of IL. Fascinatingly, this investigation comprehensively and elaborately put a new sight into the lubrication mechanism of how IL reacted with a base oil and enhanced the tribological characteristics.

**Keywords:** anti-wear; chemical tribo-film; lubricants; ionic liquids



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## 1. Introduction

Most mechanical components have two movable parts, which exist in planes, cars, and trains, so the friction and wear phenomenon happen all the time. Friction and wear are inevitable phenomena in the operation of mechanical parts in industrial production, especially in the automobile engine system, where the friction and wear between the piston ring and cylinder liner are the most serious. Proper friction is normal and beneficial to the development of industry, but excessive and severe wear will place a burden on mechanical components such as the increasing friction heat and reduced total mechanical power, which will weaken the power performance, fuel economy, and so on. Therefore, the optimization of friction between components has become a very important issue. The quality of lubricants is of great significance for mechanical components and needs to be efficiently enhanced [1]. There are many kinds of lubricants including solid lubricants, lubricating oils, and greases, among which lubricating oil is the most widely used one with good properties as it tends to generate the stable tribo-film between contact surfaces, thus preventing and delaying the earlier contact of steel–steel parts. Suitable lubricants can play a positive role in the tribological performance of mechanical components while it will be undermined if using ineffective lubricants. In addition, brilliant lubricants should not only have good thermal stability, thermal conductivity, anti-friction and wear properties, but also be compatible with the friction pair material, especially the corrosion phenomenon between lubricants and friction pair should be well avoided. Multiple researchers indicated that base oil exhibited inferior tribological performance than those containing additives. Several approaches were used to optimize the tribological performance of base liquids, among which the additives employment was the widely popular one.

In recent years, the direct use of IL as base oil additives is becoming more popular because of their excellent thermal stability, thermal conductivity, and tribological peculiarities. ILs were regarded as potentially remarkable additives of base liquids as the research gradually deepened [2–14]. IL is a salt compound entirely consisting of anions and cations that are liquid at room temperature. The cations are mainly imidazole cations, and the anions are predominately halogen ions and other inorganic acid ions. The thermochemical and tribological properties of ILs dispersing into different base liquids including PAO oil, 5W 40, 5W 30, 10W 40 and PEG are widely investigated. Li et al. [15] compared the tribological characteristics of three kinds of ILs (LL108, LB108, LP108) using 10W 40 and PEG400 as base liquids. Ultimately, the results suggested that the addition of IL into base liquids well improves the tribological performance due to the formation of a chemical film on the surface of the friction contact point via XPS characterization. Battez et al. [16] investigated the rheological and tribological performance of mineral oil, respectively. The worn scar surfaces were characterized by SEM and EDS, which asserted that the COF and WSD values were reduced owing to the use of ILs under the optimal concentration and further facilitated the anti-wear effects. Jiang et al. [17] conducted an experiment concerning the rheological properties and thermal stability of hybrid nanoparticles modified with IL, the results suggest the synergetic role between the IL and nanoparticles generates a complex reaction and finally enhances the thermal properties of base oil. Syahir et al. [18] investigated the tribological behavior of two kinds of base oil (PAO8 and bio-based oil) adding different ILs with various concentrations. They ultimately found that not all ILs can improve the tribological characteristics and only suitable ionic liquids can achieve the best anti-wear and anti-friction properties at the best concentration. In summary, these studies all indicated that IL can play a positive role in improving the tribological peculiarities of the contact surface, which also provides us with potential ideas [19–21] for our following research. In addition, some convincing mechanisms for reducing the wear and friction of IL were proposed by some experts [22–25]. Previous reviews of the effect of IL on tribological properties were listed in Table 1.

**Table 1.** Previous reviews of the effect of IL on tribological properties.

Lubricants Information			Conditions			Reduction	Friction Pair	
Ref./Year	Base Oil	Ionic Liquids	Wt%	Speed	Temperature (°C)	Load (N)	COF(%)	
[2]/2019	MF (DIOS-Fe <sub>3</sub> O <sub>4</sub> )	MIL ((C <sub>6</sub> mim) <sub>5</sub> [Dy(SCN) <sub>8</sub> ])	1.22 g/cm <sup>3</sup>	\	25–150	100–200	Reduced	Ball on disk
[3]/2020	Mineral oil	DCI	1	0.05 m/s	25–100	2	29–35.5	Ball on flat
[5]/2017	Priolube 1936(A2)	[P <sub>66614</sub> ][NTf <sub>2</sub> ]	0.25–1	\	25	80	1.37	Ball on plate
[6]/2017	Yubase4/Group III	[P <sub>66614</sub> ][[iC8] <sub>2</sub> PO <sub>2</sub> ], [P <sub>66614</sub> ][BEHP]	0.5–1	0.01–3 m/s	40–100	\	Reduced	Ball on disc
[7]/2015	Coffee bean oil	[THTDP][Deca], [THTDP][NTf <sub>2</sub> ]	1–5	\	25	2	Reduced	Block on flat
[8]/2020	PP and LiX greases	[P <sub>66614</sub> ][BOB], [P <sub>66614</sub> ][BMB], [P <sub>66614</sub> ][BMPP], [P <sub>66614</sub> ][DCA]	2–10	0.2 m/s	25–130	\	60	Pin on disk
[10]/2019	Priolube 3970 (BO)	[N <sub>8881</sub> ][C <sub>8</sub> 0], ([N <sub>8881</sub> ][C <sub>12</sub> 0]), ([N <sub>8881</sub> ][C <sub>16</sub> 0])	0.5–2	10–2500 mm/s	25–100	30–50	4	Ball on disc
[13]/2012	Glycerol	BuMepyrr-MeSO <sub>4</sub> , Et <sub>3</sub> MeN-MeSO <sub>4</sub>	0.625–8	0.1 m/s	100	10	Reduced	Ball on flat
[15]/2021	SAE 10W-40, PEG400	L-L108, L-P108, L-B108	0.5–4	200 rpm	25	5	6–24.3	Ball on disk
[18]/2020	PAO8, TMPTO	[P <sub>14,6,6,6</sub> ][TMPP], [P <sub>14,6,6,6</sub> ][Deca], [BMIM][BF <sub>4</sub> ]	0.5–1.5	\	75	100	28	Four ball
[20]/2019	OPAG, Estisol 240(ME)	P-BMB	0–20	0.2 m/s	90	21	10	Ball on plate
[26]/2018	Avocado oil	P <sub>66614</sub> Tf <sub>2</sub> N, C <sub>10</sub> mimTf <sub>2</sub> N	0–100	36 mm/s	23	10	11.24–68.88	Pin on disk
[27]/2019	Glycerol	1-n-butyl-1-methylpyrrolidinium methylsulfate	0.6–8	0.1 m/s	25–100	10	20–56	Ball on disc
[28]/2017	Jatropha Trimethylolpropane ester (MJO)	Methyltrioctylammonium bis(trifluoromethylsulfonfyl)imide (AII)	1–10	1200 rpm	75	392	48	Four ball
[29]/2021	Glycerol	M-16-EDHPA	0.3–1.5	\	25	100	Reduced	Ball on disk
[30]/2017	Priolube 1936(A2)	[N <sub>1888</sub> ][NTf <sub>2</sub> ]	1.25–5	\	Room temperature	40–120	1.45	Ball on disk
[31]/2019	diester oil(A1)	[N <sub>4441</sub> ][NTf <sub>2</sub> ]	1.5	\	25–100	30–70	Reduced	Ball on disk
[32]/2014	Safflower oil (SO)	P <sub>1444</sub> DPP	0.005 mol/kg	0.1 m/s	50	50–350	Reduced	Pin on disc

**Table 1.** *Cont.*

Lubricants Information			Conditions			Reduction	Friction Pair	
Ref./Year	Base Oil	Ionic Liquids	Wt%	Speed	Temperature (°C)	Load (N)	COF(%)	
[33]/2021	DOP, PEG200	TMG-DMP, TMG-DEP, TMG-DPP, TMG-DBP	0.5–5	\	25, 100	300	Reduced	Ball on disc
[34]/2021	PEG400	NP44, NP88	0.5–3	7–3200 mm/s	25–100	20	Reduced	Ball on disc
[35]/2020	Mineral oil	Eet, Met, Det	1	0.03 m/s	25	3	63.3	Ball on flat
[36]/2011	PAO6	ethyl-dimethyl-2-methoxyethylammonium tris(pentafluoroethyl)trifluorophosphate	1	0.02 m/s	Room temperature	20–40	\	Ball on plate
[37]/2006	Mineral oil	1-n-alkyl-3-methylimidazolium	1	\	\	2.45	70	Pin on disc
[38]/2002	\	PEPE, XIP, L108	\	0.1 m/s	20	0.5–400	\	Dy-sialon disc

To verify the effect of IL as additives dispersing into base oils on the tribological performance, this study used two kinds of base oils (PAO6 and 5W 40) and IL (1-vinyl-3-ethylimidazoliumtetrafluoroborate) via a four-ball tribometer for 30 min duration. Then, a series of characterizations such as FESEM, 3D profiler, and EDS were employed to evaluate the wear scar conditions. Currently, in spite of the fact that there are multiple studies of ILs as additives dispersing water-based and oil-based fluids, the direct addition of IL into PAO 6 and 5W 40 oil are far fewer, which is also the reason rendering us to carry out some correlated investigation. In this article, we aim to explore the effect of IL modification on the base oil properties including rheological and tribological performance. The underlying mechanism of ionic liquids to increase viscosity and improve friction performance is analyzed theoretically, and a convincing analysis is provided. In addition, many directly use pure ionic liquids as lubricants for the mechanical components, which are too expensive to achieve industrial use. For the consideration of economic effect, this paper uses two kinds of base oils modified with ionic liquid, which improves the friction and lubrication performance, and at the same time meets the economic and practicality and has the potential to be applied in the industrial field.

## 2. Experimental Section

### 2.1. Materials

5W 40 oil and PAO 6 oil, as synthetic oil and crude oil, respectively, with potentially great physicochemical characteristics especially higher viscosity and viscosity index, were used as the base oil in this study. According to previous investigations and engineering practice, the lubricating effect of base oil alone seems not to be satisfied. Hence, IL was opted as a modifier to enhance the potential properties of base oil. The IL used in this study was obtained from the Shanghai Aladdin Biochemical Technology Co., Ltd., Shanghai, China. The label of lubricants was shown in Table 2. The key physicochemical parameters of IL were separately listed in Table 3.

**Table 2.** The label of lubricants used.

Base Oil	Viscosity Index	IL Concentration	Sample Label
PAO6	138	1, 2, 5 wt%	PIL
5W-40	174	1, 2, 5 wt%	WIL

**Table 3.** The physicochemical details of IL used.

IL Used	Molecular Weight	Molecular Formula	CAS Number	Melting Point	Density
(1-vinyl-3-ethylimidazoliumtetrafluoroborate)	209.9	C <sub>7</sub> H <sub>11</sub> BF <sub>4</sub> N <sub>2</sub>	936030-51-2	−3.7	1.25 (50 °C)

## 2.2. Preparation Details of Lubricants

Miscibility between base oil and IL as an important indicator should be considered preferentially since some significant physicochemical properties such as thermal stability, rheological properties, and friction properties may be affected unless the IL is well soluble with the base oil and then a stratified (unstable) state was formed. In this study, the fabrication method for lubricants similar to the two-step method was employed. Some proportional IL was preliminarily added into base oil via a 20-min magnetic stirring and then a 30 min ultrasonic dispersion. In the end, ultrasonic bath was used to stabilize the lubricants. The use of ultrasonic aims to further render IL wrap tightly with base oil due to the cavitation formation caused by ultrasonic waves. Magnetic agitation alone may be visually complete fusion between IL and base oils, but in essence, the interior will soon layer. Hence, the combination between magnetic stir and ultrasonic dispersion can achieve the optimal dispersion status. In this step, keeping the temperature not fluctuating is of key significance because the Brown motion tends to be influenced by temperature, leading to the agglomerations occurrence between nanoparticles in the fluids. Therefore, thermostatic water baths are used to control the temperature for each sample.

## 2.3. Tribological Test Condition

In the process of lubrication, a four-ball tribometer in Figure 1 was employed in this study. The material of the steel balls (including three lower balls and one upper ball) is Gcr15. The two important indicators, COF and WSD value, can be calculated from the software of tribometer. The testing working conditions are under a load of 392 N, 75 °C temperature, and 1200 RPM rotating speed, an industrial standard. The friction duration is 30 min.



**Figure 1.** The real images of four-ball tribometer used.

## 2.4. Experiment Devices Utilized

In the lubricant's preparation stage, we use an electric balance (METTLER TOLEDO ME204, Greifensee, Switzerland) to accurately weigh the mass of each material used. Meanwhile, magnetic stir equipment (JOANLABHS, Huzhou, China) and ultrasonic disperser (DW-SD20-1200, Dowell Ultrasonic Technology Co., Ltd., Hangzhou, China) were applied to effectively blend lubricants. Thermostatic water bath was used to control the temperature in case of overheating. In the materials characterization process, XRD (D8 Advance, BRUKER AXSG MBH, Karlsruhe, Germany) was used to seize the crystal structure and

FTIR (NEXUS, Thermo Nicolet, Waltham, MA, USA) was utilized to detect the formation and change in the chemical bonds. In rheological investigation, a viscometer (Brookfield DV-II+Pro, Middleboro, MA, USA) was employed to explore the viscosity of lubricants and the type of fluids. In tribological test procedure, a four-ball tribometer (MS-10A, Xiamen Tenkey Automation Co., LTD, Xiamen, China) was used to evaluate the friction and wear situation. In micro-morphology investigation of wear scar, we take advantage of FE-SEM (Zeiss Ultra Plus, Oberkochen, Germany) to find the detail of micro-morphology and use 3D profiler (Countour GT-X, Karlsruhe, Germany) to detect the surface roughness, depth curve as well as 3D profile of wear scar. Finally, for the purpose of detecting the microelement composition of wear scars, the EDS was employed.

### 3. Results and Discussion

#### 3.1. Characterization of Base Oil and IL

The XRD and FTIR characterizations were used to detect the crystal structure and chemical bond change in IL and base oil. The XRD investigation of base oil and IL was shown in Figure 2, where the peaks occurred at the  $20.1^\circ$  and  $20.9^\circ$  of  $2\theta$  value for PAO6 and 5W 40 oil while  $24.1^\circ$  for IL, which may indicate that many organic matters exist in the base oil and IL. Since they have a non-crystal phase structure, there are few peaks that can be detected by XRD. Figure 3a,b, respectively, showed the FTIR analysis of base oil, IL, and corresponding lubricants. The FTIR spectrum of 5W 40 and PAO6 oil was greatly similar due to their similar organic composition, and take PAO 6 for example, it is exhibited that two distinct peaks noted at approximately  $2923.5\text{ cm}^{-1}$  and  $2855.5\text{ cm}^{-1}$  for PAO6 oil were corresponding to the C–H bond vibrations of the fat carbon chain, and two other peaks at  $1466.24\text{ cm}^{-1}$  and  $1380.24\text{ cm}^{-1}$ , which were attributed to the C–H bond deformation of the methyl. While there were a large number of peaks for IL, the characteristic peaks located at about  $851.46\text{ cm}^{-1}$  are ascribed to the vibration from the P–F chemical bond. O–H bond formation resulted in symmetrical peaks situated at  $3159.1\text{ cm}^{-1}$  and  $3565.39\text{ cm}^{-1}$ . The peak occurred at  $1061.1\text{ cm}^{-1}$  owing to the  $\text{BF}_4^-$  vibration. In addition, C=C bond and C=N were found at the peaks of about  $1575.48\text{ cm}^{-1}$  and  $1658.25\text{ cm}^{-1}$ . C–H bond was detected at the band of approximately  $1382.8\text{ cm}^{-1}$ . In addition, the new peaks from IL occurred for WIL and PIL (at about  $3630.1\text{ cm}^{-1}$ ,  $3428.5\text{ cm}^{-1}$ ,  $1048.1\text{ cm}^{-1}$ , and  $1660.8\text{ cm}^{-1}$ ) compared with base oils, which strongly confirmed the base oils were successfully functionalized by IL.

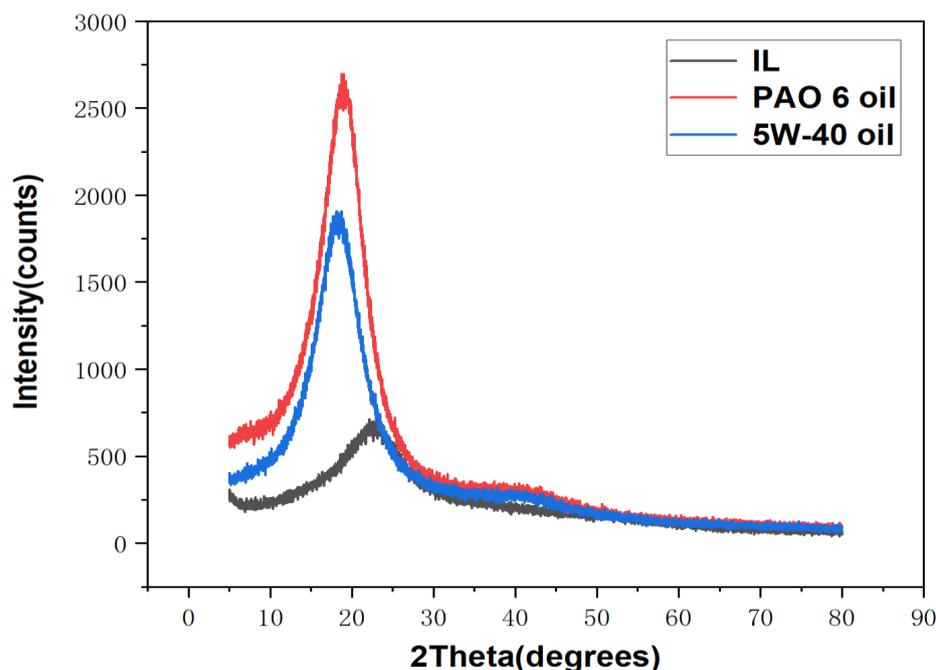
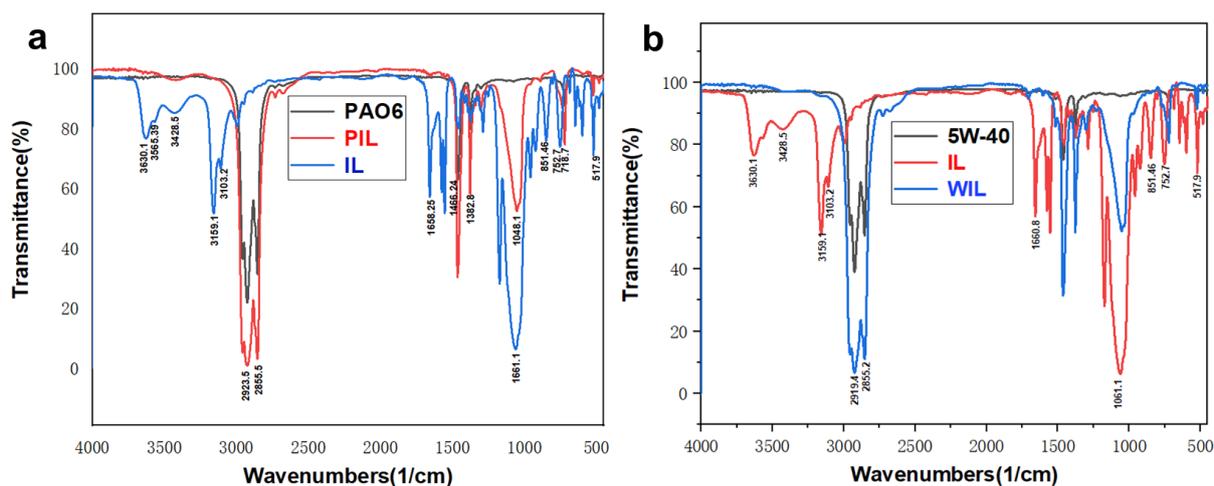


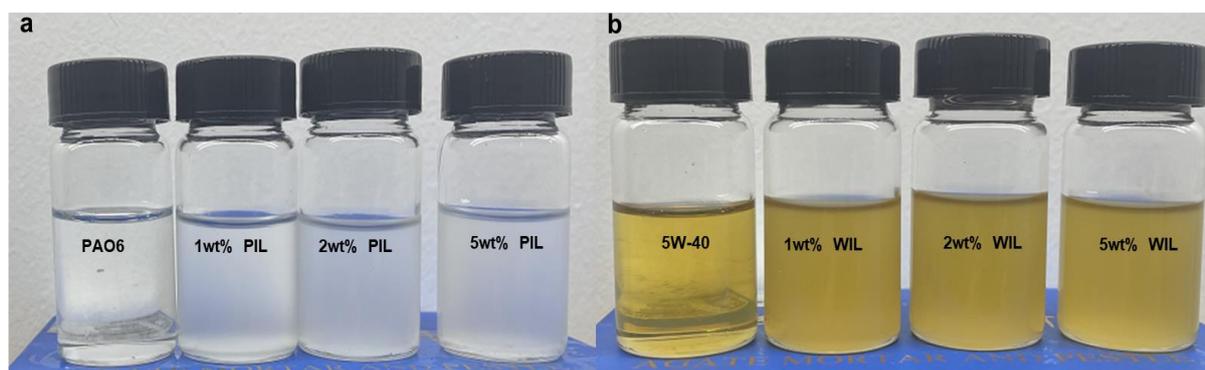
Figure 2. The XRD characterization of IL, PAO oil and 5W 40 oil.



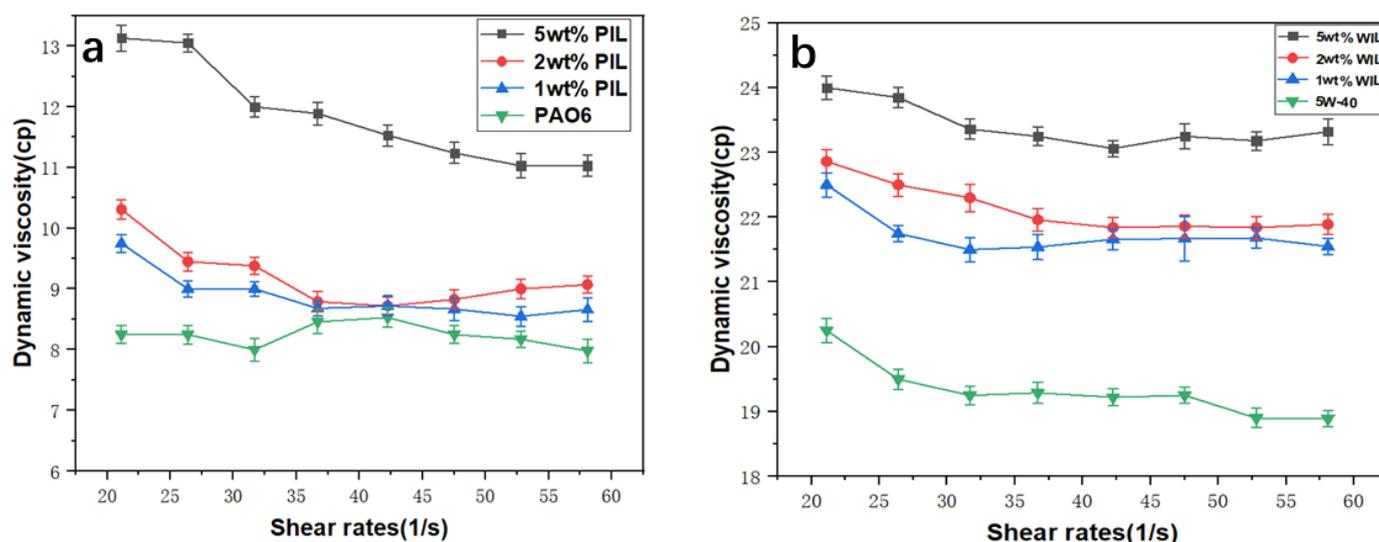
**Figure 3.** The FTIR characterization of (a) IL, PAO oil, and PIL, (b) IL, 5W 40 oil, and WIL.

### 3.2. The Rheological Behavior of Lubricants

Figure 4 displayed the natural pictures of each lubricant, among which Figure 4a showed the visual photo of PAO6 and PIL with different concentrations while Figure 4b exhibited the 5W 40 and WIL with diverse concentrations. The phenomenon that all lubricants can keep stable within several weeks can be visually observed. Furthermore, viscosity as the physical parameters is of great significance for lubricants as it can affect multiple physicochemical characteristics such as dispersion stability, heat transfer, thermal stability, and tribological performance. Figure 5 showed the viscosities of each sample. The influence of different shear rates ( $21.16\text{--}58.08\text{ s}^{-1}$ ) on the dynamic viscosity of PAO6, PIL (Figure 5a), 5w 40, and WIL (Figure 5b) was elaborately investigated under a temperature of  $75\text{ }^{\circ}\text{C}$  via a viscometer, where it can be noted that the viscosity of PAO 6 oil is lower than the 5W 40 due to their natural properties. With the modification of IL on PAO 6 oil, the viscosity of 2 wt% PIL was enhanced. Identically, the dynamic viscosity of 5W 40 oil-based lubricants modified by IL was improved compared with 5W 40 oil alone. Furthermore, with the improvement of shear rates, the dynamic viscosity of each sample was changed, which means that regardless of base oil or IL-modified lubricants (PIL and WIL), all behaved as the non-Newtonian fluids, that is, the shear thinning behavior was detected under the boundary lubrication status. Hence, IL plays a positive role in improving the viscosity of PAO6 oil and 5W 40 oil, which can be attributed to the fact that the entanglement and wrapping role caused by the introduction of IL. Viscosity value can affect the dispersion stability, heat transfer, and thermal stability of lubricants, thereby influencing the tribological properties indirectly. Hence, lubricants with higher viscosity may not be beneficial for the reducing friction characteristics.



**Figure 4.** The real image of base oils and lubricants. (a), PAO 6, 1 wt%, 2 wt%, 5 wt% PIL, (b), 5W 40, 1 wt%, 2 wt%, 5 wt% WIL.



**Figure 5.** Shear rates/dynamic viscosity correlation for (a) PAO6, 1 wt%, 2 wt%, and 5 wt% PIL (b) 5W 40, 1 wt%, 2 wt%, and 5 wt% WIL under 75 °C.

### 3.3. Tribology Characteristics

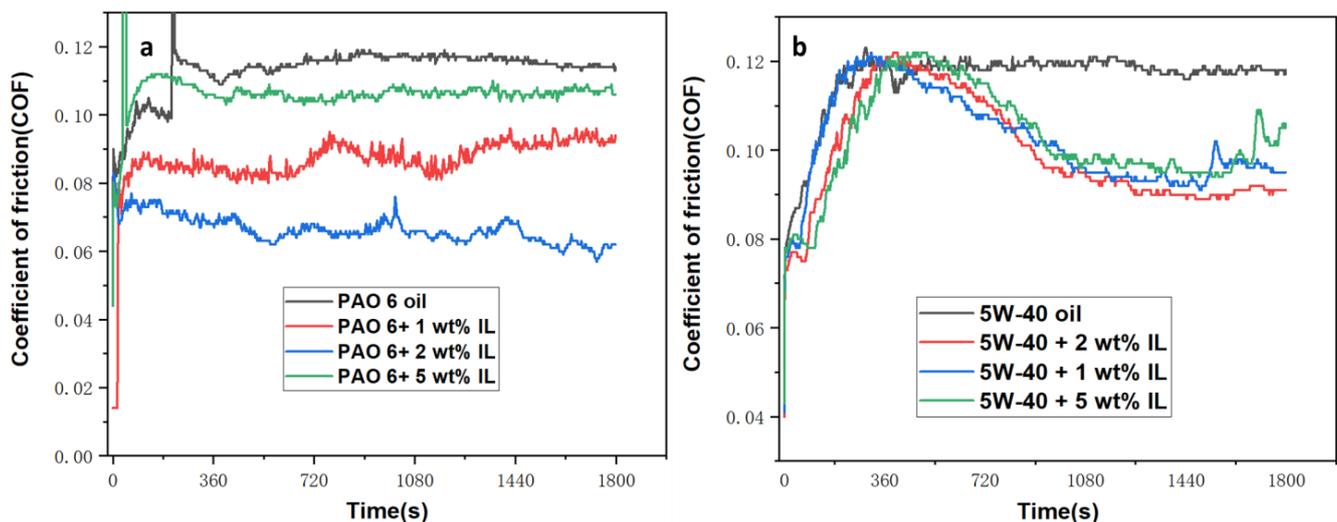
#### 3.3.1. The COF and WSD Measurement

The wear as well as friction characteristics for pure PAO 6 oil, 5W 40 oil, PIL, and WIL with different concentrations, respectively, were evaluated via a four-ball tribometer under a testing condition of 392 N, 1200 RPM, 75 °C. Figure 6 showed the COF–time curve of wear scar lubricated by PAO 6 oil, 5W 40 oil, 1 wt%, 2 wt%, and 5 wt% (PIL and WIL). In Figure 6a, the COF curve of PAO 6 oil is situated the highest and the value kept the largest compared with the other samples. The lowest COF curve was found when lubricated by 2 wt% PIL, followed by 1 wt% PIL and 5 wt% PIL. When the friction time of PAO 6 oil and 5 wt% PIL reached approximately 200 s and 80 s, the COF curve shows a short abrupt change and quickly returns to normal value, which may be due to external interference and does not affect the overall friction performance. While in Figure 6b, there is also a clear difference in the COF curve like what happened in Figure 6a between the 5w 40 oil and WIL with different concentrations. The COF curve of 5W 40 oil is located at the top position among all specimens. Those lubricated by IL-modified lubricants exhibited a lower COF curve, especially for the concentration at 2 wt%. Unlike the PIL, there is no significant difference between the curves of WIL and base oil before the friction duration of about 360 s. After the friction duration of approximately 360 s, the COF curves of WIL show a significant decrease. Additionally, the COF curve of 2 wt% WIL was located in the lowest position. However, the difference between the effect of WIL on the COF curve in the concentration is less obvious than that of PIL. Figure 7 showed the morphology of the wear scar of the lower balls captured by microscopic in the tribometer system, which agreed well with the COF results.

Figure 8 displayed the average COF and WSD values of wear scar lubricated by base oil, PIL, and WIL. It can be clearly noted from Figure 8a that the COF values of wear scar lubricated by 1 wt% and 2 wt% PIL were apparently decreased by approximately 21.2% and 32.2% than PAO 6 base oil while 5 wt% PIL showed higher COF value than those at 1 wt% and 2 wt% PIL. Additionally, the WSD values of 1 wt% and 2 wt% PIL displayed a lower value of about 0.6855 and 0.6325 mm, which is better than the PAO 6 oil of 0.707 mm. The 5 wt% PIL exhibited the highest WSD values among all samples. Comparatively, in Figure 8b, although the difference between samples is not as obvious as in Figure 8a, the pattern is similar, that is, 2 wt% WIL shows the smallest COF and WSD values. The COF and WSD values of 2 wt% PIL were decreased by 41.7% and 10.5% compared with PAO 6 oil while 14.5% and 2.8% for 2 wt% WIL compared with 5W 40 oil. In addition, the

COF value of 2 wt% PIL is 0.06636 and the 2 wt% WIL is 0.09939, while the WSD value of 2 wt% PIL is 0.6325 mm and the 2 wt% WIL is 0.418 mm, indicating that the PAO6 oil-based lubricant has better anti-friction performance, and the 5W 40 oil-based lubricant has better anti-wear performance. Moreover, the excessive increase in IL concentration is not conducive to the improvement of friction properties. In summary, in spite of the fact that IL improves the frictional properties of both base oils, the improvement effect on PAO6 is more obvious, which is related to the physicochemical properties of the base oils, such as viscosity because the annual value of 5W 40 is much higher than that of PAO 6. After IL modification, the viscosity increase amplitude of 5W 40 is also larger than that of PAO6. Figure 9 seized the pictures of the wear scar on the upper balls. Different with the lower balls, the wear scar of the upper balls is a circle arc but a spot so the microscope can only pick up part of the arc. The wear evaluation index becomes the width of the arc rather than the WSD value of the lower ball. Similar to the lower balls, there is a lowest arc width for the 2 wt% PIL and WIL. An amount of 5 wt% PIL and WIL are not conversely beneficial for the tribological properties, which agreed well with the results of Figures 6–8. Through Figures 6–9, a preliminary finding can be detected that for the single base oil, regardless of PAO 6 oil and 5W 40 oil, the addition of IL can effectively reduce the COF value within the suitable concentration. Nevertheless, excess concentration of IL will in turn undermine the tribological properties. This phenomenon should be explained more deeply in the following investigation.

In addition, Figure 10a–d, and a'–h', respectively, showed the 3D profile and depth curve of the wear scars lubricated by PAO6, 2 wt% PIL, 5W-40, and 2 wt% WIL. In Figure 10, it is apparent that the wear scar lubricated by base oil (PAO and 5W-40) is larger than that lubricated by 2 wt% PIL and WIL. Moreover, the average depth of wear scar with PAO 6 and 5W-40 lubrication is 13.2  $\mu\text{m}$  and 5.02  $\mu\text{m}$  while 9.12  $\mu\text{m}$  and 4.35  $\mu\text{m}$  with 2 wt% PIL and WIL lubrication in Figure 10b1,d1,f1,h1, and the corresponding decrease rates are 30.9% and 13.3%, which agreed well with the morphology investigation and tribological test results.



**Figure 6.** The coefficient of friction (COF)–time curve of diverse specimens. (a) PAO 6 oil, 1 wt%, 2 wt%, and 5 wt% PIL, (b) 5W 40 oil, 1 wt%, 2 wt%, and 5% WIL.

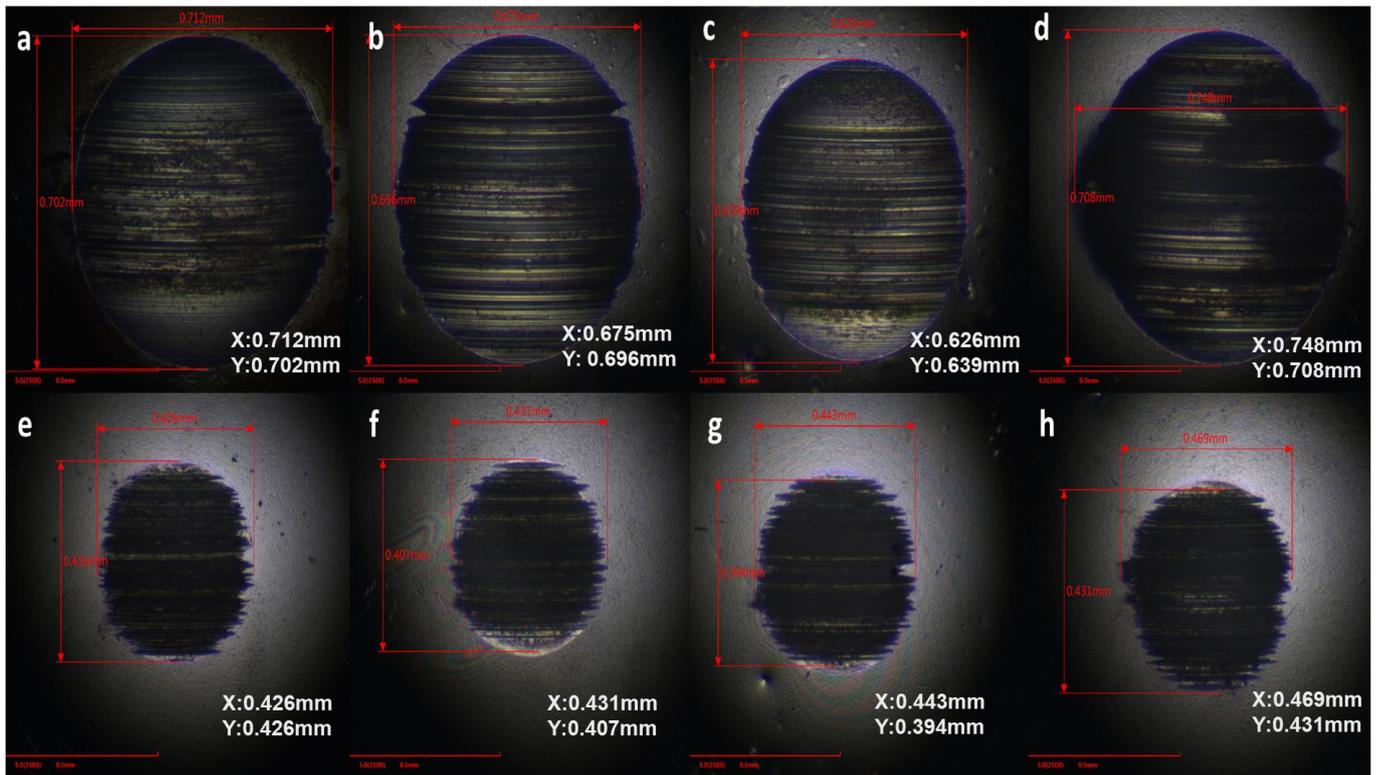


Figure 7. The wear morphology of the lower steel balls lubricated by (a) PAO 6 oil, (b–d) 1 wt%, 2 wt%, and 5 wt% PIL. (e) 5W 40 oil, (f–h) 1 wt%, 2 wt%, and 5 wt% WIL.

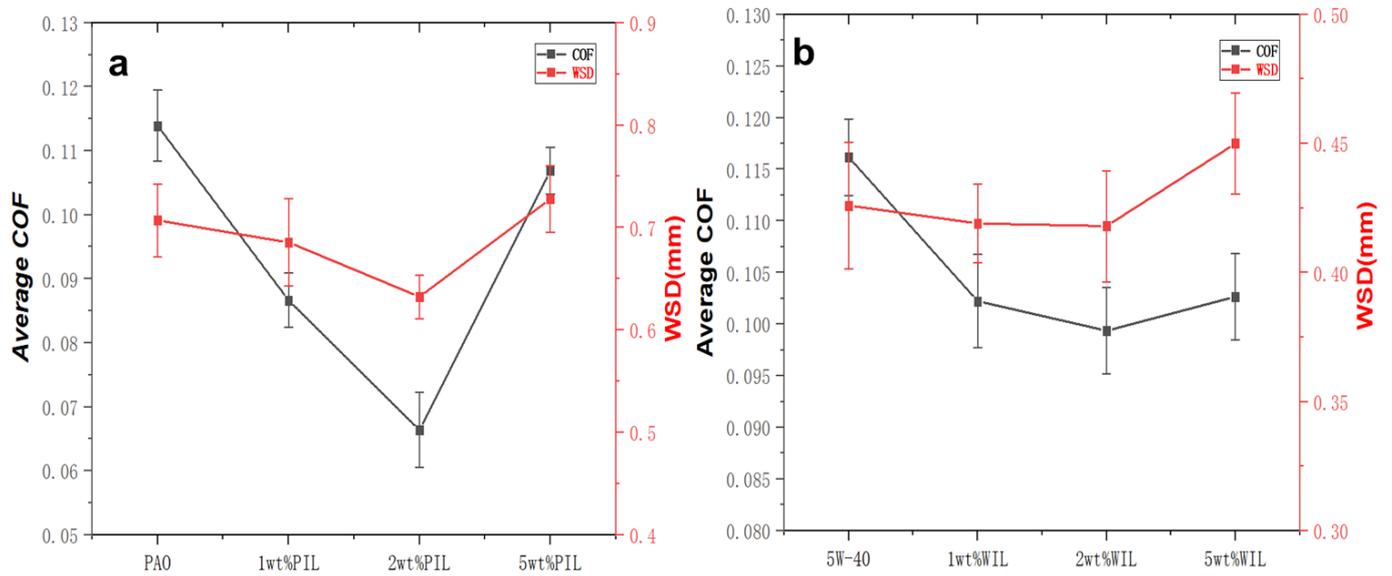
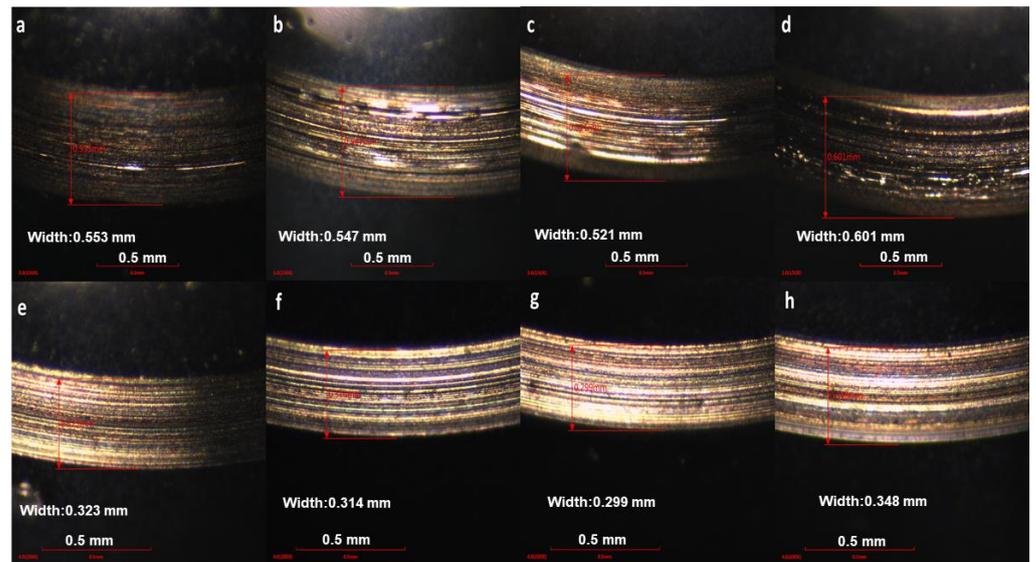
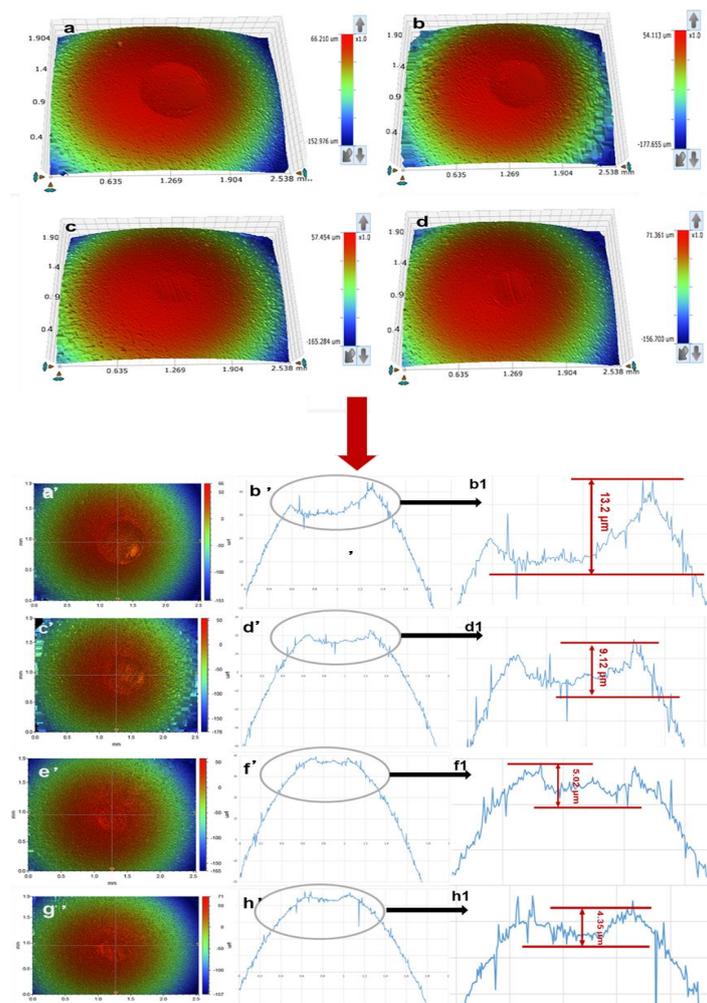


Figure 8. The average COF and WSD values of wear scar lubricated by (a) PAO 6 oil, 1 wt%, 2 wt%, and 5 wt% PIL, (b) 5W 40 oil, 1 wt%, 2 wt%, and 5 wt% WIL.



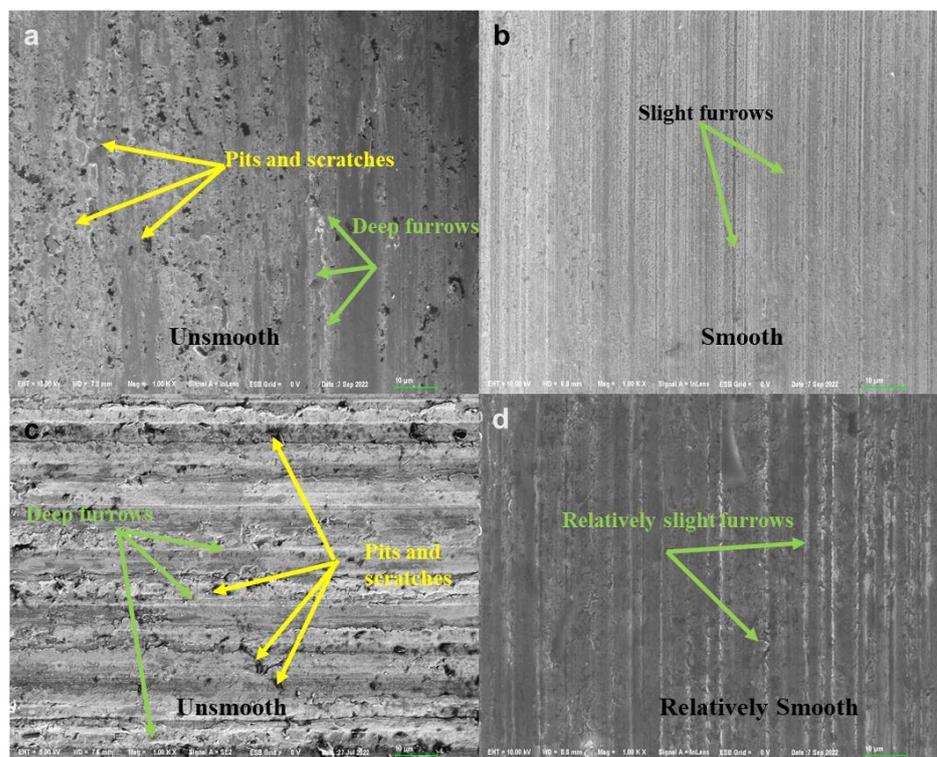
**Figure 9.** The wear morphology of the upper steel balls lubricated by (a) PAO 6 oil, (b–d) 1 wt%, 2 wt%, and 5 wt% PIL, (e) 5W 40 oil, (f–h) 1 wt%, 2 wt%, and 5 wt% WIL.



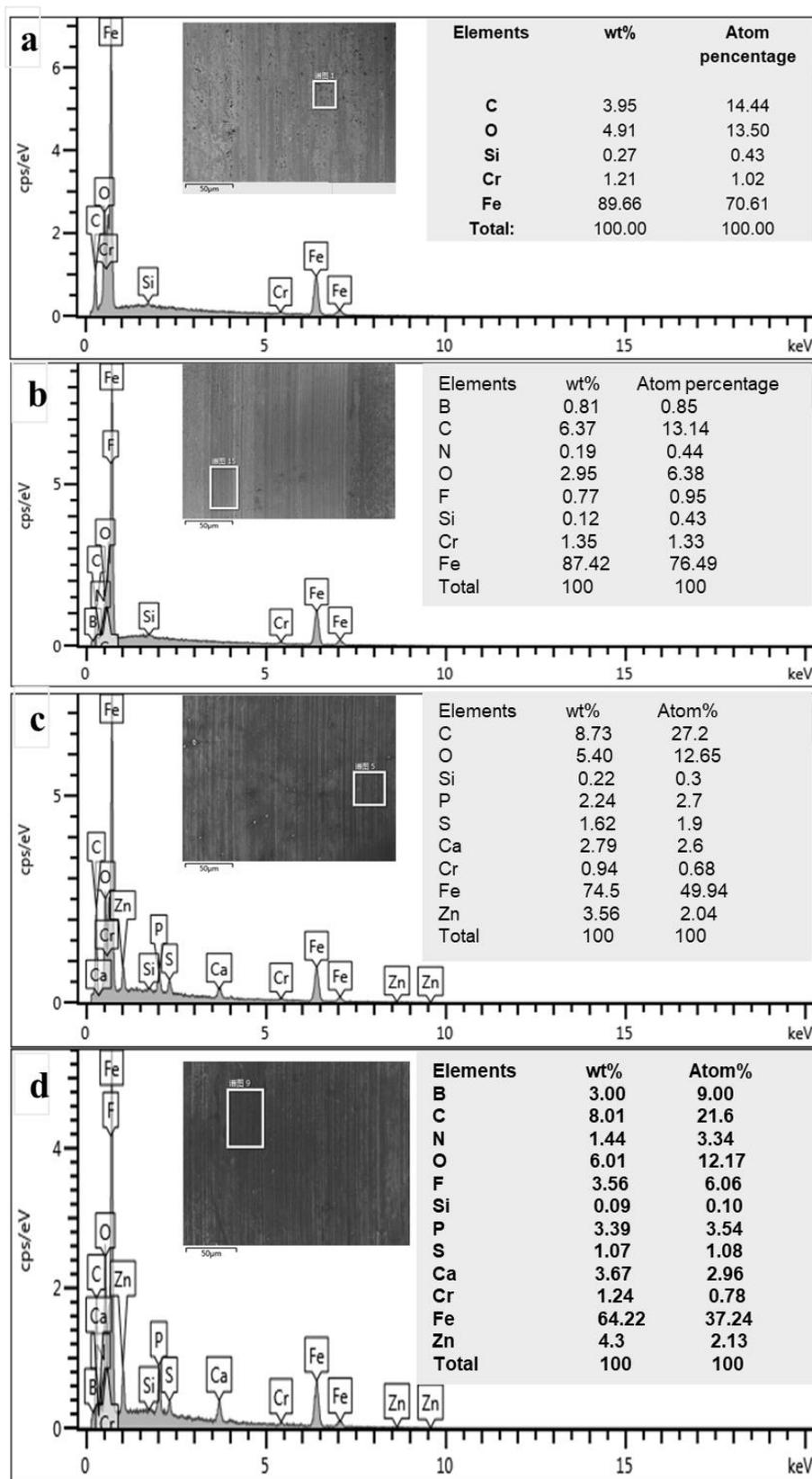
**Figure 10.** The 3D profile morphology of lower steel balls lubricated by (a) PAO 6 oil, (b) 2 wt% PIL, (c) 5W40, (d) 2 wt% WIL and the depth curve of the (a',b') PAO 6 oil, (c',d') 2 wt% PIL, (e',f') 5W 40, (g',h') 2 wt% WIL.

### 3.3.2. The Micro Morphology Characterization of Wear Scar

In addition, Figure 11 depicts the FE–SEM analysis of wear scars with various lubricants lubrication. It was connoted in Figure 11a,b that the wear scar lubricated by PAO 6 oil has severe furrows, pits, and scratches compared with that lubricated by PIL. There is a smooth friction surface when lubricated by PIL, which can be attributed to the effective role of IL on PAO 6 oil. While Figure 11c showed the severe scratches, deep furrows as well as the pits, and the relatively slight furrows can be observed in Figure 11d. Additionally, the pits and scratches of wear scar lubricated by WIL have a significant improvement to the lubrication by 5W 40 oil alone. In spite of the fact that IL additions have both improved the smooth degree of contact interface lubricated by PAO 6 and 5W 40, the effect of IL on the modification of PAO6 oil is superior to that of 5W 40 oil in improving the microstructure of friction pairs, which may be related to the original properties of base oil and the chemical changes during the modification of base oil by IL. The micromorphology as part of the independent investigation should also be compared with the friction test results (COF and WSD) for comprehensive analysis. It can be precisely detected in Figure 12a,c that Fe, Cr, Si, C, and O were found when lubricated by PAO 6 while the Fe, Cr, Si, C, O, P, S, Ca, and Zn were detected for 5W 40 oil. The extra B, F, and N elements were captured in Figure 12b,d, in addition to the detected element before, and after the lubrication of PIL and WIL, confirming the IL not only works but exerts a significantly key role in the tribological lubrication procedure. The IL attached to the surface of the base oil to improve the physicochemical properties of the base oil and the tribo-film containing the oxidation product such as  $\text{FeF}_3$ , and  $\text{B}_2\text{O}_3$  [16] was formed via a chemical reaction process. The tribo-film preventing the direct contact of steel-steel surface and alleviating the wear degree during the friction process was ascribed to the modification of IL on base oil. Base oils have few polar functional groups while IL is polar liquid. For lubricants modified with ionic liquids, anions and cations can be adsorbed to the friction surface through the Coulomb force between molecules. The interaction between ions and long alkyl chains in base oils makes the innermost layer closer, thus enhancing the lubrication performance [14].



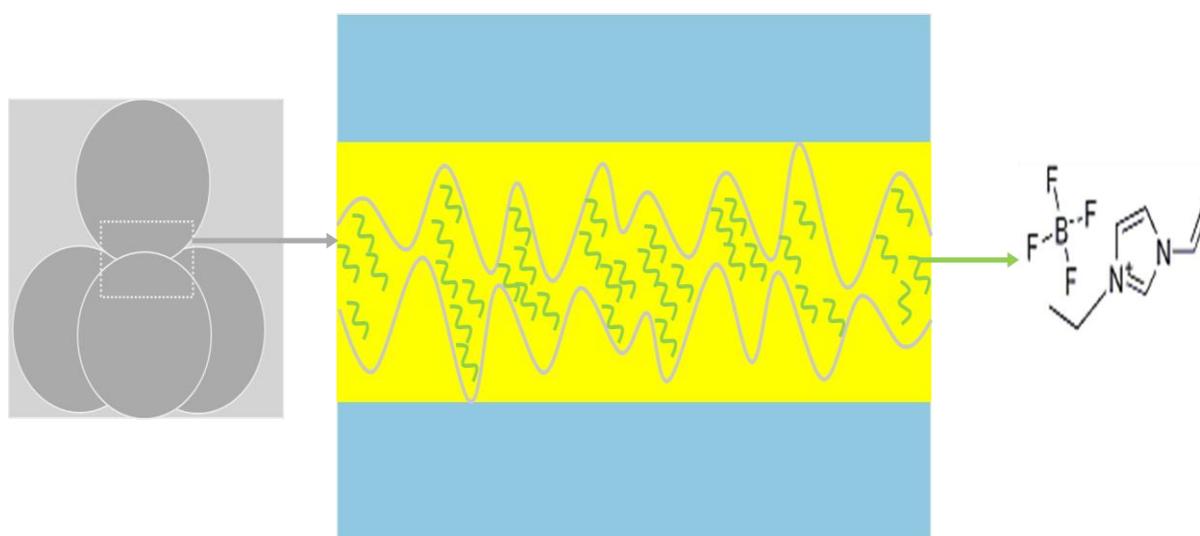
**Figure 11.** The FESEM characterization of wear scar lubricated by (a) PAO 6 oil, (b) 2 wt% PIL, (c) 5W 40 oil, (d) 2 wt% WIL, under the magnification of 1000 $\times$ .



**Figure 12.** EDS characterization of wear scar lubricated by (a) PAO6, (b) 2 wt% PIL, (c) 5W 40 and (d) 2 wt% WIL.

### 3.3.3. Lubrication Mechanism

As seen in Figure 13, where it was vividly described the lubrication mechanism scheme of base oil and IL-modified lubricants and it is suggested that when the lubrication process proceeds, the lubricating oil modified by the IL begins to work. Moreover, the IL is wrapped on the lubricating oil surface and enters into the friction pair. Under high temperatures, an oxidation reaction occurs on the friction surface, and a dense tribological film due to the chemical reaction interacting with the oxides on the friction surface is generated, which prevented direct contact between the steel–steel contact surface and further improved the tribological properties. The tribological properties are well improved by the IL additions under boundary lubrication conditions. In addition, the physical absorption of IL on the surface of oils molecule is also responsible for the lubricating mechanism. This positive tribological behavior caused by ILs can be ascribed to the polar characteristics of IL itself, making them chemically adsorb to the substrate surface. Meanwhile, friction-resistant protective chemical films were formed during friction [25].



**Figure 13.** The anti-wear mechanism scheme of IL-modified oil.

## 4. Conclusions

This study investigated the rheological and tribological properties of two base oils modified by IL at different concentrations. Multiple characterizations were elaborately conducted. Based on the results and discussion, the important conclusions can be drawn as follows:

1. The COF and WSD values of 2 wt% PIL were decreased by 41.7% and 10.5% while 14.5% and 2.8% for 2 wt% WIL compared with their corresponding base oils.
2. IL addition is conducive to the viscosity enhancement of base oil, especially the 5W 40 oil. In addition, all lubricants behave like non-Newtonian fluids.
3. The COF and WSD values of base oil can be greatly reduced by the modification of IL (within suitable concentration), especially the PAO6 oil. In this study, 2 wt% PIL and WIL showed the best tribological performance. An amount of 5 wt% PIL and WIL will in turn undermine the anti-friction properties due to the rising viscosity.
4. Although IL can also improve the friction properties of 5W 40, the improvement effect of WIL is not as significant as that of PIL, which can be attributed to the higher viscosity of 5W 40, especially after being modified by IL.
5. EDS characterization has confirmed that the key elements such as F, B, and N from IL in the surface of the wear scar were detected, strongly indicating the IL does work in the process of friction lubrication and is responsible for the generation of tribo-film in the contact surface due to the polar effect of IL.

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