



Phosphonium-based ionic liquids as grease additives in rolling bearing tests

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ABSTRACT

Two phosphonium-derived ionic liquids: trihexyltetradecylphosphonium bis(2-ethylhexyl)phosphate (IL1) and trihexyltetradecylphosphonium tricyanomethanide (IL2) were used as additives in lithium complex- (G1) and anhydrous calcium-based (G2) greases at 5 wt%. Friction torque and wear tests were performed using a modified four-ball machine for testing rolling bearings in order to determine the friction and wear reducing properties of these grease samples in a real component. The IL2 improved the friction reduction performance of both greases, especially G1. Both ILs improved the antiwear behaviour of grease G2. Grease G2 showed higher oxidation and thermal ageing levels than G1, but the addition of the ILs, IL2 in particular, improved this issue.

1. Introduction

Lubricating greases are the most commonly used lubricants in rolling bearings. More than 90% of rolling bearings are sealed for life, and they use grease to ensure lubrication [1], as its semi-solid state makes it much less likely to leak out from the bearing [2]. Mineral oils and triglycerides are commonly used as base oils, although synthetic oils are required for some applications. Among synthetic oils, poly- α -olefins (PAO), perfluoropolyalkylether fluids (PFPE), polyalkylene glycols, silicones and synthetic esters are generally used. The substances most commonly used to thicken the grease are: soap thickeners, simple or complex; inorganic thickeners such as clay, silica or polyurea; and mixed soap thickeners comprised of several cations. Grease lubrication with synthetic base oils is most often used for bearing applications in extreme environments, but many synthetic base oils are not compatible with soap thickeners, so teflon, polyurea, clay or fumed silica thickening systems must be used to form grease lubricants [1].

Since 2001, ionic liquids (ILs) have been widely studied as lubricant additives, but most of the research on this topic involves liquid lubricants [3,4]. The ILs can be grouped according to different properties, such as the cation or anion on which they are based and their miscibility in organic compounds, among others. ILs that are immiscible in non-polar hydrocarbon oils were used as additive in earlier research

[5–24]. Despite their immiscibility, they can enhance friction and wear reduction. Further research was carried out into oil-soluble phosphonium cation-based ILs [25–50] and ammonium cation-based ILs [11,18,50–61], as additives in lubricant oils or organic compounds. Several ILs based on imidazolium and pyrrolidinium cations, which are only miscible in base oils of a polar nature, have been studied [11,38,57,58,62–74]. ILs have also been studied as additives in water-based fluids, an environmentally friendly alternative to petroleum oil-based lubricants, especially in fire-resistant hydraulic fluids and metal-working fluids [75–85].

From 2010 onwards, researchers started to study the use of ILs as additives in lubricant greases [85–89], showing them to possess good antiwear and friction reduction properties. In these studies, standard tribological tests (pin-on-disk, ball-on-disk, four-ball or reciprocating configurations) were used to test the influence of the ILs on the tribological properties of greases. Polyurea grease was additised with five different alkyl imidazolium ILs at 1 wt% [85] and with imidazolium bearing a benzotriazole group at 2 wt% [87]. The authors found that the 1 wt% alkyl imidazolium IL additives performed better at high temperature conditions, while imidazolium bearing a benzotriazole group, at a concentration of 2 wt%, had excellent friction reduction and antiwear performance. Wang et al. [86] tested a lithium lubricating grease based on a polyalphaolefin (PAO 10) additised with three phosphonium

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ILs at 5 wt%. The results showed that the friction-reducing and antiwear properties were enhanced in all cases. More recently, Ploss et al. [88] tested four non-halogenated ILs containing the trihexyl(tetradecyl) phosphonium cation as an additive in polypropylene (PP) and lithium complex (LiX) greases at concentrations of 2–10 wt%. In this case, a reduction in wear of up to 60% and in traction of up to 40% were found.

Although the main use of lubricant greases is in rolling bearings, few studies have described the performance of greases in this practical application [90–97]. Some of these studies explored topics like lubrication film thickness, wear or friction [90–92]; while others were focused on power loss, heating or lifetime [93–97]. None of them dealt with the performance of greases additised with ILs in practical applications (e.g., rolling bearings).

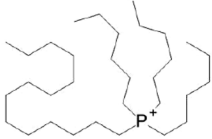
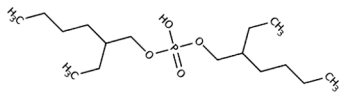
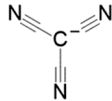
In previous works [89,98], thermal conductivity, thermal stability and tribological performance of two greases (lithium complex- and anhydrous calcium-based) were studied using three phosphonium-based ILs as additives: trihexyltetradecylphosphonium bis(2-ethylhexyl) phosphate, $[P_{6,6,6,14}][BEHP]$ (designated as IL1); trihexyltetradecylphosphonium tricyanomethanide, $[P_{6,6,6,14}][TCM]$ (designated as IL2); and trihexyltetradecylphosphonium decanoate, $[P_{6,6,6,14}][DEC]$ (designated as IL3). Rheology, lubricant film thickness and friction behaviour experiments were also carried out. The results showed improvements in friction reduction and lubricant film thickness for some of the mixtures [89]. Meanwhile, the higher friction and wear reductions were obtained with the use of $[P_{6,6,6,14}][BEHP]$ and $[P_{6,6,6,14}][TCM]$. Now, this work aims to determine how the additivation of the above-mentioned greases with these two better-performing ILs affect friction torque and wear in a real component (bearing) tested under real operating conditions (rolling/sliding motion). In addition, greases were analysed after tribological tests with FTIR and ferrometry.

2. Methodology

2.1. Greases and ionic liquids

Two non-additised greases (lithium complex-, G1, and anhydrous calcium-based, G2) provided by Axel Christiernsson International and two ILs (trihexyltetradecylphosphonium bis(2-ethylhexyl)phosphate or $[P_{6,6,6,14}][BEHP]$, coded as IL1, and trihexyltetradecylphosphonium tricyanomethanide or $[P_{6,6,6,14}][TCM]$, coded as IL2) provided by IOLITEC GmbH were used in this work. The chemical formulae and structures of these ILs are shown in Table 1. The physicochemical properties of the greases and the mixing procedure of the base greases and the ILs at 5 wt% were described in [89]. Apart from the non-additised greases, only the 5 wt% IL mixtures were tested in this work, as their performance was much better than that of greases with 2 wt% of ILs in previous tests.

Table 1
Chemical structure and empirical formula of the ionic liquids.

Ionic liquid	Cation	Anion
Trihexyltetradecylphosphonium bis(2-ethylhexyl)phosphate $[P_{6,6,6,14}][BEHP]$		Bis(2-ethylhexyl)phosphate
Empirical formula: $C_{48}H_{102}O_4P_2$ Designation: IL1		
Trihexyltetradecylphosphonium tricyanomethanide $[P_{6,6,6,14}][TCM]$		Tricyanomethanide
Empirical formula: $C_{36}H_{68}N_3P$ Designation: IL2		

2.2. Rolling bearing assembly and test procedures

Rolling bearing tests were conducted in a modified Cameron-Plint TE 82/7752 four-ball machine (Fig. 1) to study the tribological behaviour of the greases and their mixtures with ILs in a real component. For this purpose, the four-ball arrangement was replaced by a rolling bearing assembly on which different bearing tests can be performed and friction torque measurements can be obtained at different test temperatures. This procedure allows the performance of tests under rolling/sliding conditions, which correspond with the real operating conditions of bearings. A complete explanation of the development of this procedure can be found in references [93–95,99–101]. This test performed on a full bearing configuration is better than the well-known 4-ball test (ASTM D2266), which simulates smearing under pure sliding and is not relevant for rolling bearings, so the bearing and grease industry recommend full bearing tests [102].

During the test, the bearing was under a constant axial load (P) of approximately 7000 N, which was applied from bottom to top using a dead weight system. An electric motor was used to set the different test speeds as needed. The power was transmitted to the rolling bearing shaft with a belt-pulley arrangement, so that it rotates the upper bearing track, while the lower track was fixed to the housing. A KISTLER® 9339A (Kistler Group, Winterthur, Switzerland) piezoelectric reaction torque cell, coupled to the housing, was used to measure the friction torque.

A thrust ball bearing (TBB) reference 51107 from SKF (Sweden) was chosen for testing, and a new one was used for each test. This rolling bearing has 21 rolling elements of 6 mm of diameter, and the raceways have a mean diameter of 43.5 mm.

Two different rolling bearing tests were performed:

- Friction torque tests: a short test performed at constant load and temperature, where the friction torque is measured at different rotational speeds;
- Wear tests: a long test (3 × 24 h) performed at constant load, temperature and rotational speed. After the test, the roughness of the lower raceway was analysed, as well as the mass loss of the rolling bearing. A grease sample was collected after the test for evaluation by FTIR and Ferrometry.

2.2.1. Friction torque tests

These tests started with a period of 15 h, during which testing was performed at the established axial load of 7000 N, at a temperature of 50 °C in the bearing, and at 500 rpm rotational speed, to allow running-in and churning / grease distribution. To reach and maintain the temperature, an external thermal bath was used as described by Marques et al. [104]. After the churning period, the temperature in the rolling bearing was set at 80 °C and the rotational speed was reduced to 250 rpm. Once the temperature stabilized, after about two hours, the

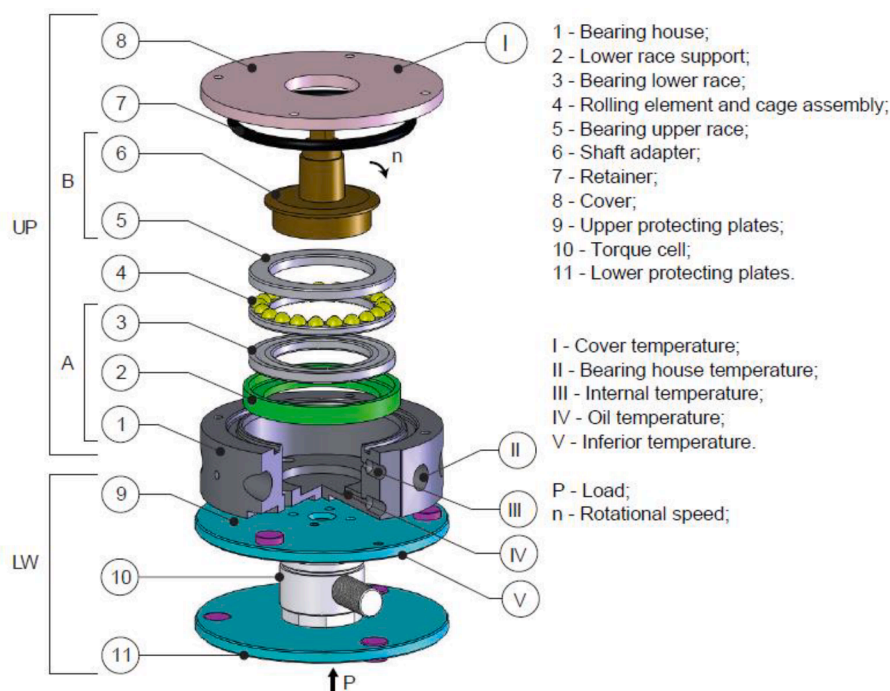


Fig. 1. Schematic diagram of rolling bearing assembly [103].

frictional torque was measured. Then the speed was increased to 750 rpm until the temperature stabilized (about 1 h) and friction torque was measured again. Finally, the speed was increased to 1500 rpm and the torque measured one last time after the temperature had stabilized. At each speed step, five friction torque measurements were taken and then averaged. 2 mL of grease was used to lubricate the rolling bearings (which corresponds to approximately 30% of the rolling bearings free volume). For each grease, the test was performed twice.

2.2.2. Wear tests

Wear tests were performed in the same device used in the friction torque tests. In these tests, the rolling bearing operated for 3 days (72 h), at constant load (7000 N), constant temperature (80 °C) and constant rotational speed (250 rpm). To increase the severity of the tests, three balls were removed from each bearing, which put the maximum hertzian pressure at 2.5 GPa. The operating conditions were chosen in order to promote boundary lubrication conditions.

The mass loss of each rolling bearing was measured on a scale with a precision of 0.001 mg. In addition, the infrared spectra of each grease before and after the wear tests were obtained on an Agilent Cary 630 FTIR device, using an ATR (Attenuated Total Reflectance) accessory to determine molecular alterations in the greases due to thermal aging. All the spectra shown in this work were taken directly from the device's software without smoothing, and a very good reproducibility was achieved. All spectra were normalized to the same peak's height at 1460 cm^{-1} [105], allowing the comparison between the relative height of the sample spectra. After each test, a grease sample was also analysed by Direct Reading Ferrography, obtaining the DS (wear particles $<5 \mu\text{m}$) and DL (wear particles $>5 \mu\text{m}$) parameters. The severity of wear particles index (ISUC) and the concentration of wear particles index (CPUC), defined by the Eqs. (1) and (2), were calculated from these parameters [106].

$$ISUC = \frac{(DL^2 - DS^2)}{d^2} \quad (1)$$

$$CPUC = \frac{(DL + DS)}{d} \quad (2)$$

where d is the dilution of the grease sample.

The roughness of the lower (fixed) racetrack of each rolling bearing was measured by interferometry using a BRUKER NPFLEX. A total area of $1.5 \times 1.5 \text{ mm}$ was collected. Although it is not possible to measure the roughness profiles at exactly the same position before and after test, the data collection was performed in the same region, which was assured by marking the bearing ring before the tests.

As it is possible to observe in the Fig. 2, the raceway is curved in the (y) direction. This curvature was removed and then the surface roughness (Ra) was determined according to ISO 4287, filtering the data with a Gaussian filter with a cut-off length of 0.25 mm. The roughness results were analyzed in the rolling direction (x) only because the curvature removal might show misleading results in the filtered roughness profile in the y direction.

3. Results and discussion

3.1. Rolling bearing tests

3.1.1. Friction torque results

The frictional torque of the TBB lubricated with the above-mentioned greases is shown in Fig. 3. The values represent the mean of two replicates performed for each lubricant mixture. The greases without IL (G1 and G2) showed the highest friction torque values, which were similar and approximately constant under the tested speeds. Grease G1 additised with IL2 showed the lowest values of friction torque, these being more significant at the lowest speed. The mixture of grease G1 with IL1 showed friction torque values similar to neat G1, always bearing in mind the uncertainty of the measurements. Furthermore, IL2 also conferred better friction reduction properties when mixed with grease G2, while the mixture of G2 + IL1 showed friction torque values that were similar to those of grease G2 at the tested speeds.

3.1.2. Wear results

Fig. 4 shows the wear results from the tests performed with all the lubricant samples. The addition of both ILs to grease G1 increased wear, but the wear values were very small and the differences observed between samples are within the combined uncertainties of the bearing test

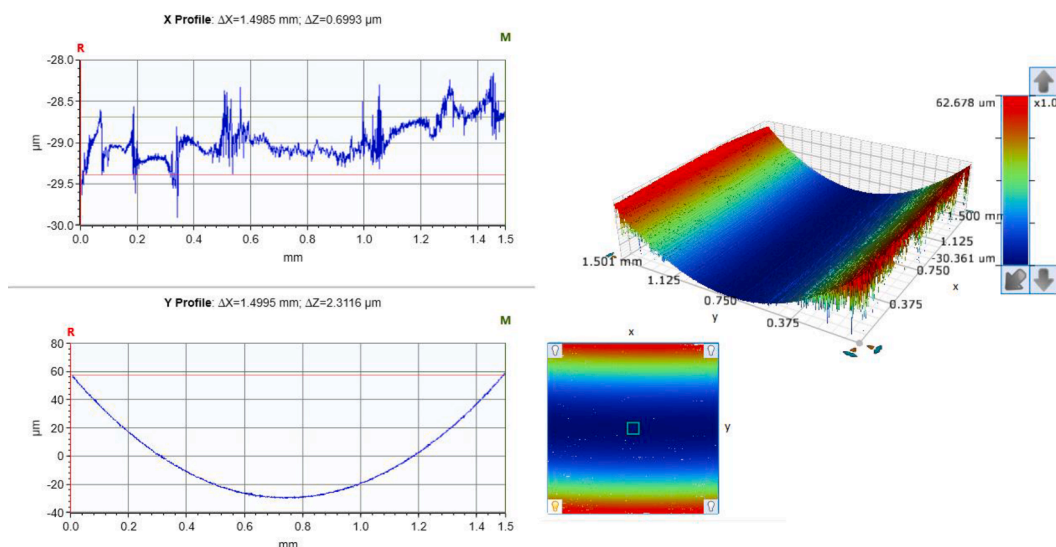


Fig. 2. Roughness of the bearing surface in the perpendicular (y) and longitudinal (x) directions of movement.

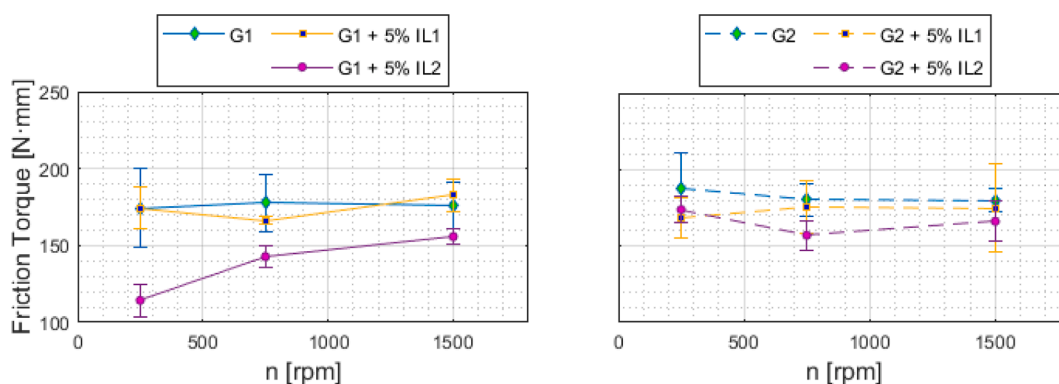


Fig. 3. Friction torque for the different blends of G1 (left) and G2 (right).

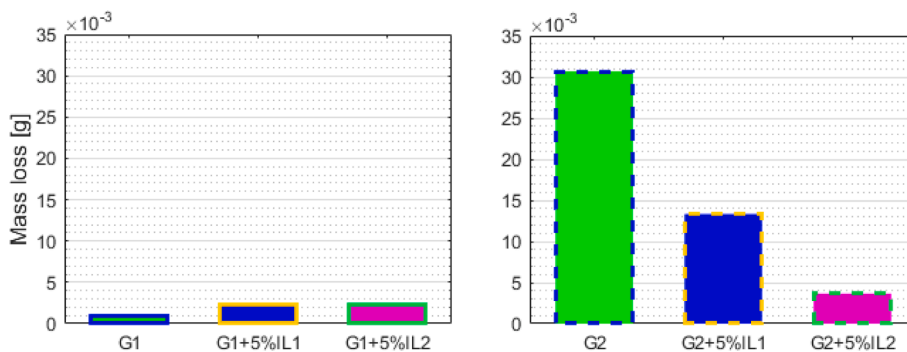


Fig. 4. Mass loss for the different blends of grease G1 (left) and grease G2 (right).

and the mass measurement. However, the addition of the ILs to grease G2 decreased wear considerably in all cases.

These results were different to those reported in [98], where the same lubricant samples were tested under pure sliding motion with both four-ball (constant speed) and reciprocating (variable speed) tests. In such cases, wear decreased with the addition of the ILs to the grease (G1) in both tribological tests, while wear increased in the case of grease G2 in the four-ball tests. On the other hand, friction behaved differently with the addition of the ILs to grease G1 (increased) and to grease G2 (decreased). Although the antiwear behaviour found in that case was

correlated with the presence of phosphorus on the wear surface, the results obtained in this study could be related to the different motion configuration (rolling/sliding), where the sliding is typically 5% [102]. The tribological improvement not only depends on the concentration and chemistry of the IL but on the tribosystem also, as was stated by Zhou et al. [3].

The extreme complexity of grease lubrication is closely related to the high number of variables involved (base oil viscosity/nature, additive package, thickener type and/or content, consistency, etc.) and the numerous performance requisites (low friction, low and high

temperature properties, improved oil bleeding, etc.) [102]. In fact, Gonçalves et al. [107] found that greases showing better performances in single contact tests can provide worse wear protection or rolling bearing life. In this study, the tests were performed after the churning or fully flooded film thickness period. Then, the bleeding period took place, where the bearing is mainly lubricated by the base oil or the mixture base oil-IL, and the film thickness decreases, which is known as the starvation phenomenon. This phenomenon occurs due to the lack of replenishment or the insufficient filling of the inlet region of the lubricated contact, which can decrease the lubricant film thickness by around 75% with respect to the values of the fully flooded film thickness [108], resulting in a lower load carrying capacity, asperity contacts taking place, and the appearance of wear mechanisms, like scuffing [109]. Under normal operation and depending on the initial grease volume and distribution, the lubricated contacts can starve due to several causes: side flow, centrifugal effects, surface tension, oil bleeding from the grease and evaporation [107]. In addition, lubricant loss may be caused by oxidation, polymerization, evaporation, centrifugal force induced thin film flow or droplet formation in the outlet of the contacts [110].

Table 2 shows the ferrometric parameters and indexes. In general, the number of wear particles larger than 5 μm was higher than those smaller than 5 μm . Grease G2 without IL had the highest CPUC and ISUC values, indicating that the use of this grease resulted in higher wear. The wear reduction found when IL2 was added to grease G2 can also be observed in the ferrometric parameters and index values shown in Table 2. However, grease G1 showed smaller values for both the CPUC and ISUC indexes than its mixtures with the ILs, corresponding with the wear results reported in Fig. 4.

Fig. 5 shows the average roughness evaluated in the rolling direction for all the 12 tests. It is possible to observe that the initial roughness is slightly different between the new bearings (average of 0.082 μm with a standard deviation of 0.008 μm) so instead of comparing just the final roughness value after the tests between samples, it is more suitable to analyze the difference relative to the initial roughness for each sample.

From Fig. 5 it is clear that the roughness increased after the testing for all samples, due to the severe operating conditions, namely boundary lubrication (3 days running at 250 rpm, 80 °C and 2.5 GPa). However, and despite the differences in the initial roughness of each bearing, it is also possible to observe that the increase of roughness after the wear test is higher for all G2 greases, particularly for the neat G2. Given that grease G1 and grease G2 are formulated with a base oil of the same nature, the differences in the variation of the roughness should be due to the slight smaller viscosity of the base oil used in grease G2 and the influence it might have on lubricant film generation. This was shown in the film thickness tests reported in [89], where grease G1 showed generally higher lubricant film thickness.

It is also interesting to notice that the addition of both ionic liquids improve the antiwear behaviour for grease G2, but this does not happen to grease G1. Nevertheless, it is clear that between the greases with ionic liquids, the ones which contain IL2 show a better behavior (smaller roughness increase) than those containing IL1. The reason for this behavior cannot be inferred from the tests performed in this work, but the results reported in [35,67] support that ionic liquids generate tribofilms on the metallic surfaces contributing to the reduction of friction and improving the antiwear behaviour [98].

Table 2
Ferrometric parameters and indexes.

Grease sample	DL	DS	CPUC $\times 10^{-3}$	ISUC $\times 10^{-6}$
G1	7.4	4.9	0.123	0.003
G1 + 5% IL1	46.5	18.2	0.647	0.183
G1 + 5% IL2	57.2	24.3	0.815	0.268
G2	119.8	79.8	19.960	79.840
G2 + 5% IL1	77.4	49.8	12.720	35.107
G2 + 5% IL2	25.3	11.8	3.710	5.009

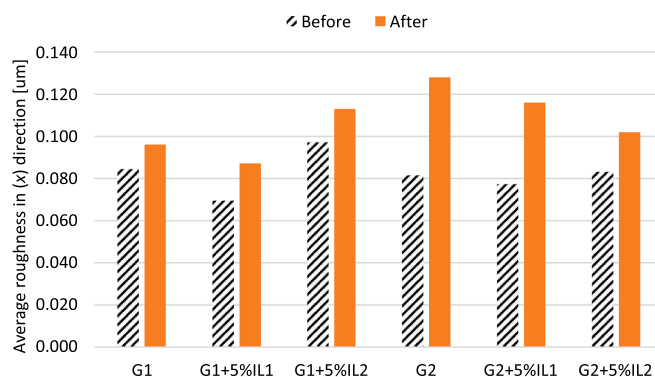


Fig. 5. Surface roughness of the rolling bearing before and after the wear tests.

Fig. 6 shows the variation of the average roughness (R_a) of the bearing raceways in the rolling direction versus the mass loss of the rolling bearing. According to Fig. 6, the greater the mass loss, the higher is the increase in roughness. Only the bearing lubricated with the mixture G1 + 5%IL1 broke that tendency, but the difference with the G2 + 5%IL2 counterpart was minimum (about 10^{-3} g of mass loss and 2% of variation of R_a , which is within the uncertainties of the wear test and the mass loss measurement).

Cen and Lugt [111] reported that the only relevant physical property that determines the film thickness, and thus the probability of asperity contacts and wear, in the early lifetime of a grease is the base oil viscosity, and not the bleed rate or any grease rheological properties. Considering that the greases G1 and G2 were formulated with base oils of similar viscosities, their original consistency and yield stress are also similar [89], and were studied under the same testing conditions, their different antiwear behavior could be related to oxidation. Changes on EHL film over long times, which are related to tribological behaviour, are given by mechanical and chemical degradation [102]. Chemical degradation is primarily given by oxidation, and also by evaporation, although other phenomena occur such as acid formation, thermo-oxidative degradation of the thickener and the base oil, varnish and sludge formation, etc. The oxidation of the base oil and thickener are not

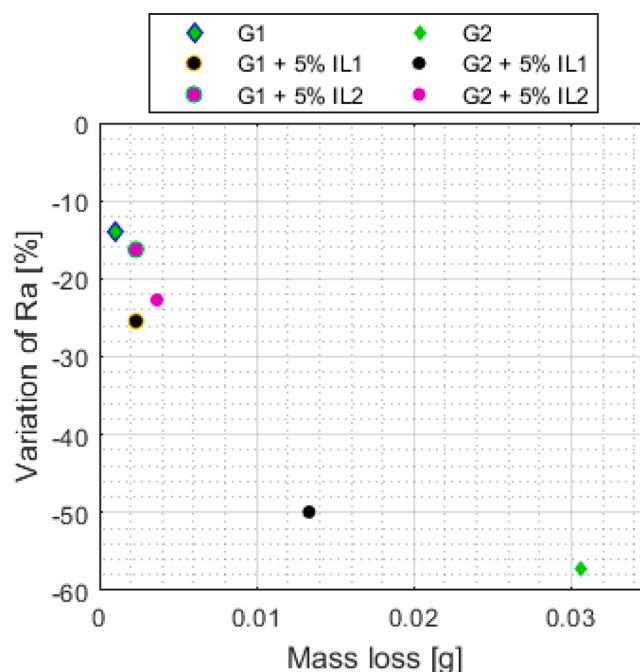


Fig. 6. Variation of surface roughness ($\frac{R_{a_{\text{before}}}-R_{a_{\text{after}}}}{R_{a_{\text{before}}}}$) in the rolling direction.

fully independent problems. However, studies on grease thickener oxidation are rare and most oxidation research has been made on lubricating oils, being generally accepted that their corresponding results can be applied to lubricating greases [1]. Fig. 7 shows the FTIR spectra of the fresh grease (before wear testing) and the used grease (after the wear test, with the suffix “a”). From comparison of the spectra, it is clear that the calcium-based grease (G2) seems to be more sensitive to oxidation than the lithium-based grease (G1), due to its higher peak around 1750 cm^{-1} and also a clear offset in the whole fingerprint region ($1800\text{--}650\text{ cm}^{-1}$). The G1-containing samples also show some oxidation, but much less than their G2 counterparts. The oxidation level was diminished with the addition of IL2 to both G1 and G2 greases, and IL2 confers higher resistance to aging/oxidation than IL1. The higher improvement on thermal stability of these greases with the addition of the IL2 was also reported in a previous work [98], which is related to evaporation and hence to chemical degradation. The fact that anhydrous calcium soap greases (G2) can be used up to temperatures of $110\text{ }^{\circ}\text{C}$, while the operating temperature range of the lithium complex greases (G1) is between $-30\text{ }^{\circ}\text{C}$ and $140\text{ }^{\circ}\text{C}$ [1], explains the higher contribution of the ILs on the wear protection properties of the grease G2 (Fig. 4) and the better tribological behavior of the grease G1. In summary, these data may explain the worse antiwear protection of grease G2 and its mixtures with the ILs. In addition, the polar nature of the oxidation products increase the polarity of the grease over time, which lead to an increase of water absorption from the air [112,113]. This phenomenon can cause corrosion impacting negatively in the grease wear protection.

4. Conclusions

Friction torque and wear tests in a real component (bearing) were carried out to evaluate the tribological behaviour of two greases (lithium complex- and anhydrous calcium-based) additised separately with two phosphonium-based cation ILs. The mass and roughness variations of the bearing were evaluated, and the greases were analysed after wear

tests with FTIR and ferrometry. The main conclusions of this study are the following:

- IL2 improved the friction reduction performance of both greases, with the lowest friction torque value in the case of grease G1.
- The addition of both ILs to grease G1 resulted in a slight wear increase, while the addition of the ILs to grease G2 improved its antiwear performance, especially in the case of IL2.
- The ferrometric results were in concordance with the mass loss (wear) results. Furthermore, in general the decrease in surface roughness of the bearing raceways was also closely related to wear.
- Regarding oxidation and thermal aging, the FTIR spectra showed higher oxidation of grease G2 than grease G1, and the addition of IL2 provided higher resistance to oxidation than IL1 in both greases.
- The different tribological behaviour of the tested ILs is probably related to their antioxidant action. The exact mechanism taking place is unclear, but these ILs might be a suitable additive for grease G2 in bearing applications.

CRediT authorship contribution statement

M. Bartolomé: Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Visualization, Funding acquisition. **D. Gonçalves:** Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – original draft, Writing – review & editing, Visualization. **A. García Tuero:** Validation, Formal analysis, Investigation, Data curation, Writing – review & editing, Visualization. **R. González:** Conceptualization, Methodology, Resources, Data curation, Writing – review & editing, Visualization, Supervision. **A. Hernández Battez:** Conceptualization, Methodology, Validation, Formal analysis, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition. **J.H.O. Seabra:** Conceptualization, Methodology,

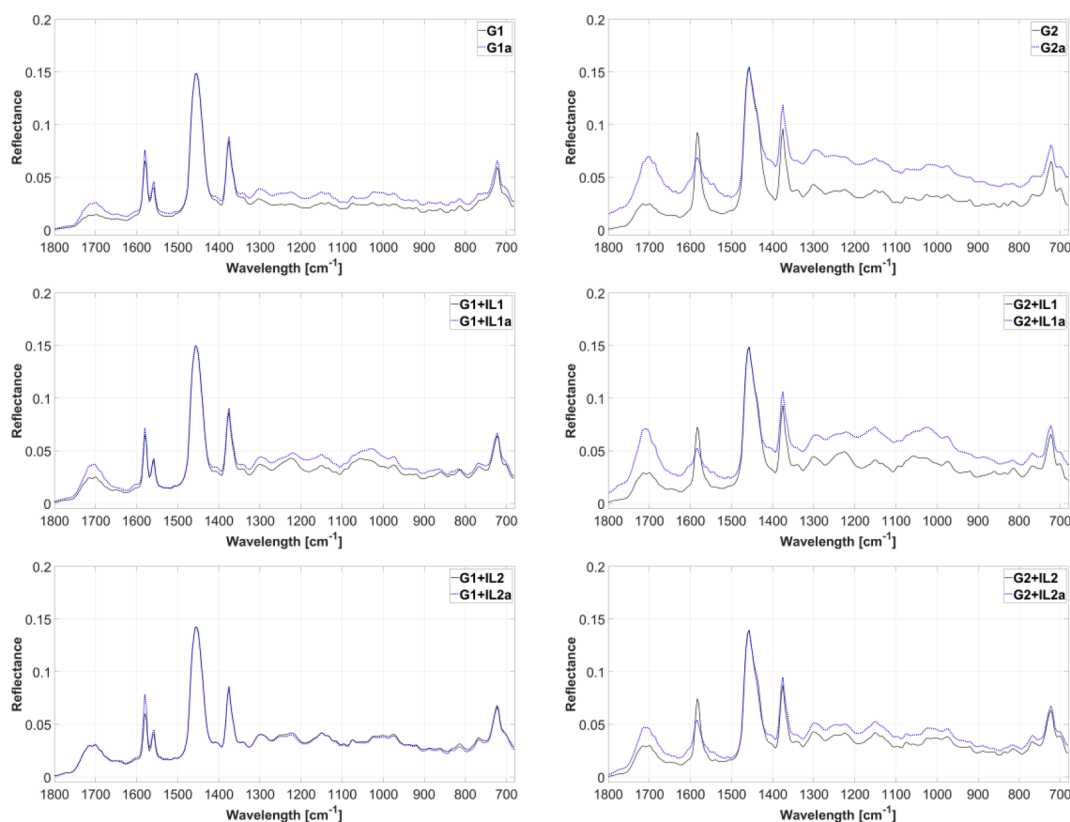


Fig. 7. FTIR spectra of the grease samples before and after tests.

Validation, Formal analysis, Resources, Data curation, Writing – review & editing, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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