

Two fatty acid anion-based ionic liquids - part II: Effectiveness as an additive to a polyol ester

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Abstract

This work is about the use of two ionic liquids obtained from fatty acids (FAILs) as an additive at 0.5 and 2 wt.% to a polyol ester (POE) base oil. Tribological tests under two different configurations (reciprocating and sliding/rolling "ball-on-disc") were performed at different temperatures and loads. Worn surface was measured and analyzed after reciprocating experiments by confocal and scanning electron microscopies and by X-ray photoelectron spectroscopy (XPS). Main results showed that these FAILs at low concentration did not change the viscosity of the POE and hence the tribological behaviors of the mixtures and the POE under elastohydrodynamic lubrication (EHL) in rolling/sliding tests were similar, but using mixtures resulted in lower friction regarding the POE under mixed lubrication regime; the friction behavior in reciprocating tests was similar with the use of all mixtures and the POE at 25 °C for the two loads used, while the mixtures outperformed the antiwear behavior of the POE; the antifricition behavior of the mixtures under both loads at 100 °C was slightly worse than that of the POE and all mixtures had better antiwear performance than the POE.

Keywords: ionic liquids; additive; polyol ester base oil; lubrication; friction; wear

1. Introduction

In the early years of the twenty century, the ethylammonium nitrate $[(C_2H_5)NH_3][NO_3]$ was reported as the first ionic liquid (IL) [1]. The research interest of these liquid salts, which melt at temperatures lower than 100 °C, started to grow in the 70s using pyridinium/imidazolium cations and halide/tetrahalogenoaluminate anions for synthetizing ILs and employ them as electrolytes in batteries [2,3]. The first known drawback of ILs was the possible basicity or acidity of these novel substances, which could represent a problem in many applications. This issue was solved in the 90s when Wilkes and Zawarotko [4] obtained new ILs formed by neutral weakly coordinating anions, such as tetrafluoroborate $[BF_4]^-$ and hexafluorophosphate $[PF_6]^-$. These new ILs allowed a significant growth of research in numerous industrial applications: catalysts, liquid crystals, extraction technology, solvents for both organic or inorganic materials and synthesis [5-8]. The findings of the promising lubricant behaviour of ILs was firstly reported in 2001 [9]. This lubricant property is related to some interesting features of these novel salts: non-volatility, high thermo-oxidative stability and wide liquid range. Along the next 10 years, numerous journal papers and patents about tribological application of ILs have been published, demonstrating the huge interest on this topic [10-21].

1 The possibility of using these novel substances in lubricant formulation is mainly related to their capacity
2 to form tribofilms on the contact surface, resulting in both antifricition and antiwear improvements [22-26].
3 Despite all these advantages of the ILs, two major problems appeared regarding the use of ILs in lubrication.
4 Although these substances were firstly considered as green lubricants, the majority of ILs contain elements
5 that are toxic or hazardous to the environment [27-31]. In addition, the price of these substances remains
6 too high, so the use of ILs as neat lubricants are limited to very specific applications [32-35]. This above-
7 mentioned fact causes that the use of ILs as lubricant additive is for now more reasonable than using them
8 as base fluid or pure lubricant [36-44]. However, the inherent polarity of ILs provoked that a lot of
9 researches using ILs as additive have been carried out over the last two decades in order to overcome the
10 solubility issue. The first research works using non-polar hydrocarbon oils as lubricant base stock were
11 very low IL-concentrations mixtures or even oil-IL emulsions that need to be stabilized [45-49]. This
12 approach could take advantage of aditising non-polar oils with low-concentration polar additive [50-52].
13 Taking into account the solubility problem in non-polar base oils, the ILs were generally found having
14 better compatibility with polar oils [53-59].
15 The toxicity problem of the ILs have been addressed throughout selecting proper constituent ions for
16 searching of new environmentally friendly compounds that contribute to a more sustainable development
17 [60, 61]. According to this purpose, not only bio-based lubricants and additives [62-66], but also FAILs are
18 being studied. The synthesis of FAILs via metathesis reaction was firstly reported in 2013 [67]. From that
19 moment, the research works focused on using these novel compounds in lubrication increased exponentially
20 in recent years [68-82].
21 Two novel FAILs (tetrahexylammonium octanoate, $[N_{6,6,6,6}][C_{8:0}]$, and tetrahexylammonium palmitate,
22 $[N_{6,6,6,6}][C_{16:0}]$) were studied as neat lubricant in the first part of this work [83]. Now, this paper addresses
23 the use of them as an additive to a polyol ester with the goal of assessing their tribological performance.

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2. Methodology

2.1 Fatty Acid anion-based Ionic Liquids (FAILs)

28 The tetrahexylammonium octanoate $[N_{6,6,6,6}][C_{8:0}]$ and tetrahexylammonium palmitate $[N_{6,6,6,6}][C_{16:0}]$ fatty
29 acid anion-based ionic liquids (FAILs) were synthetized using caprylic acid (octanoic) for the former and
30 palmitic acid (hexadecenoic) for the latter as anionic precursor (water content of both FAILs: <2 wt.%). In
31 addition, tetrahexylammonium bromide was employed as cationic precursor and acquired from Sigma-

1 Aldrich S.A. The process of synthesis was performed through a double replacement reaction. Besides, ^1H
2 and ^{13}C nuclear magnetic resonance (NMR) and FTIR spectroscopy were executed to ascertain the chemical
3 structure. The synthesis/identification procedure of these FAILs were described extensively in the first part
4 of this research, as well as their density and viscosity data [83].

5 6 *2.2 Preparation of mixtures*

7 With the aim of analysing the miscibility of the two studied FAILs in different commercial oils, 5 wt.%
8 samples were prepared using three different base oils kindly supplied by REPSOL: a Group IV synthetic
9 base oil (PAO 4), a Group I mineral base oil (SN-150) and a Group V polyol ester base oil (Priolube 3970).
10 Density and viscosity at reference temperatures of these base oils are reported as Supplementary
11 Information. A simple experiment was conducted looking for the most appropriate base oil from a
12 miscibility point of view: all 6 blends (each FAIL at 5 wt.% in the three base oils) were mixed manually.
13 The progress of the mixtures' stability was followed by visual inspection along 48 h and only the mixtures
14 with the polyol ester (POE) were miscible, while the others showed two immiscible phases. Finally, FAILs
15 were separately mixed at two different concentrations (0.5 and 2 wt.%) in the POE using an ultrasonic
16 probe (Bandelin Sonoplus HD2200) at 70% of amplitude for 5 min and controlling the blend temperature
17 below 60 °C.

18 19 *2.3 Density and viscosity*

20 The density and dynamic viscosity of the blends were obtained according to ASTM D7042 between 20 and
21 100 °C (atmospheric pressure), taking measurements every 10 °C in a Couette rotational viscometer (SVM
22 3001 Stabinger Viscometer). The equipment automatically calculates the kinematic viscosity and the
23 viscosity index according to the ASTM D445 and ASTM D2270 standards, respectively.

24 25 *2.4 Friction and wear tests*

26 Two different tribometers were used in order to characterize the tribological behaviour of the FAIL-
27 containing samples. The PCS Instruments Mini-Traction Machine (MTM) with a ball-on-disc setup was
28 employed for performing a test at 50% of slide-to-roll ratio (SRR) and varying the mean entrainment speed
29 in the 2500-10 mm/s range (using decreasing steps of 100 mm/s until reaching a speed of 100 mm/s and at
30 10 mm/s steps from a speed of 100 mm/s until 10 mm/s) while electrical contact resistance (ECR) and

1 friction coefficient were recorded. A lubricant volume of 10 mL, 30 N-load (0.95 GPa of maximum contact
2 pressure) and 40, 60, 80 and 100 °C of temperature were used in this test. Before the test, both MTM
3 specimens (discs and balls) were cleaned with petroleum ether for 20 min using an ultrasonic bath, then
4 rinsed in ethanol and finally dried with air. The lower specimen is a disc of AISI 52100 steel with 720-780
5 HV₃₀ of hardness and surface roughness (Ra < 0.02 μm). The upper specimen is a ball of AISI 52100 steel
6 with hardness of 800-920 HV₃₀ and Ra < 0.02 μm of surface roughness.

7 In addition, a Bruker UMT-3 tribometer with a ball-on-plate reciprocating configuration was also used for
8 measuring the lubricant properties of the mixtures. These tests were conducted during 30 min, loads of 30
9 and 50 N (2.06 and 2.43 GPa of maximum contact pressures, respectively), frequency of 15 Hz, stroke
10 length of 4 mm, 4 mL of sample volume and temperatures of 25 and 100 °C. At least 3 replicates for every
11 test condition were made involving a relative error less than 10%. An ultrasonic bath was used for cleaning
12 the specimens with petroleum ether during 25 min, then rinsed in ethanol and finally dried with air before
13 and after the tests. In this case, the lower specimen was a disc of AISI 52100 steel with 225 HV₃₀ of hardness
14 and Ra = 0.018 μm of surface roughness, and the upper specimen was a ball of AISI 52100 steel (58-66
15 HRC of hardness, Ra = 0.05 μm of surface roughness).

16 17 *2.5 Surface characterization*

18 All lower specimens (discs) employed in the tribological tests performed with the UMT-3 were studied by
19 confocal microscopy. This equipment can capture multiple two-dimensional images at different depths in
20 the sample enabling the reconstruction of a three-dimensional wear scar topography and calculating its
21 volume from a selected reference plane. In addition, different techniques such as: scanning electron
22 microscopy (SEM) and both energy dispersive and X-ray photoelectron spectroscopies (EDS-XPS) were
23 employed in order to evaluate the chemical interaction between the surface and the lubricant samples.

24 A JSM-5600LV SEM (JEOL) device operating in low vacuum (LV) with imaging mode detection of
25 secondary and backscattered electrons was employed. The equipment was also fitted with an energy
26 dispersive X-ray microanalysis (EDX) Oxford Inca Energy 200 apparatus allowing elementary mapping of
27 the sample and qualitative-quantitative structural determination for elements between Be and U thanks to
28 its X-ray based characterization capabilities. The SEM microscope was set to work at 20 kV voltage and
29 images were taken at x100 magnification.

XPS tests were carried out with a SPECS spectrometer. A K_{α} (Al) monochromatized X-ray source at 45° (1486.7 eV) was employed for the experiments. The final X-ray spot (area of 3.5×1 mm²) was always focused on the wear scar. In addition, binding energy was corrected using the position of the adventitious carbon (284.6 eV). At least 0 scans were taken for every analysis (step energy of 0.1 eV and pass energy 30 eV). The total number of scans depended on each sample and element.

3. Results and discussion

3.1. Density and viscosity

These properties were measured in the first part of this research [83], exhibiting the same behavior that other FAILs employed in previous research [84-87]. In addition, Table 1 shows density and viscosity values at reference temperatures (additional data can be found as Supplementary Information) of the blends resulting of mixing the FAILs at 0.5 and 2% in POE. In view of the results, it can be concluded that the use of ionic liquids at the concentrations employed in this research study hardly change some initial base oil properties, such as density, viscosity and viscosity index.

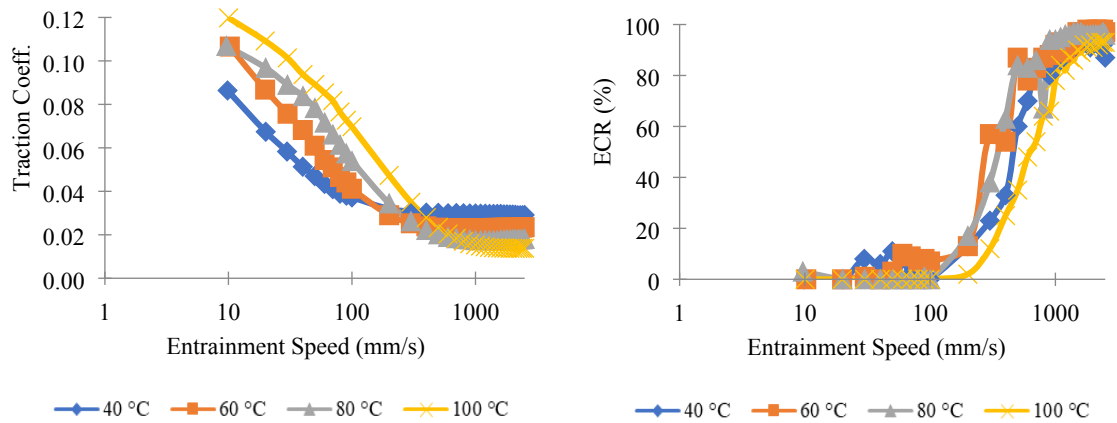
Table 1. Density and viscosity parameters of the FAIL-containing mixtures.

Lubricant samples	Density at 20 °C (g·cm ⁻³)	Kin. Viscosity (mm ² ·s ⁻¹)		Viscosity Index
		40 °C	100 °C	
POE + 0.5% [N _{6,6,6,6}][C _{8:0}]	0.9417	20.3	4.52	140
POE + 2.0% [N _{6,6,6,6}][C _{8:0}]	0.9412	21.3	4.63	138
POE + 0.5% [N _{6,6,6,6}][C _{16:0}]	0.9411	20.4	4.52	140
POE + 2.0% [N _{6,6,6,6}][C _{16:0}]	0.9407	21.2	4.63	139

3.2. Tribological properties

Fig. 1 shows both friction and ECR performance from tests lubricated with the POE under sliding/rolling conditions (SRR=50%) at different speeds and temperatures. The test performed under EHL regime in the 2500-1000 mm/s range at all temperatures with decreasing friction at increasing temperature. This friction behavior takes place because of viscosity control friction under EHL regime and viscosity decrease with temperature. The thicker lubricant film under this lubrication regime results in higher ECR values. From a speed of 1000 mm/s downwards, the lubrication regime changed from elastohydrodynamic to mixed corresponding to a decrease in ECR due to a thinner lubricant film.

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Fig. 1 Traction coefficient and electrical contact resistance (ECR) behavior versus speed at different temperatures for the base oil (polyol ester).

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The addition of these FAILs to the POE in the concentrations used hardly changed the friction behavior of the POE (Figs. 2-3) under elastohydrodynamic lubrication regime (see also the Supplementary Information) and the speeds at which the lubrication regime changed from elastohydrodynamic to mixed. These results can be associated with the similar viscosity values of the mixtures and POE (Table 1). Under mixed lubrication regime, as expected, the test made at higher temperature showed the higher friction values according to the lower viscosity, which result in thinner lubricant films (see ECR values in this case) and hence much more asperities contact. However, the addition with the ionic liquids decreased friction values with regard to the base oil at decreasing speeds (under mixed lubrication regime) in most of the cases as can be observed in the Supplementary Information.

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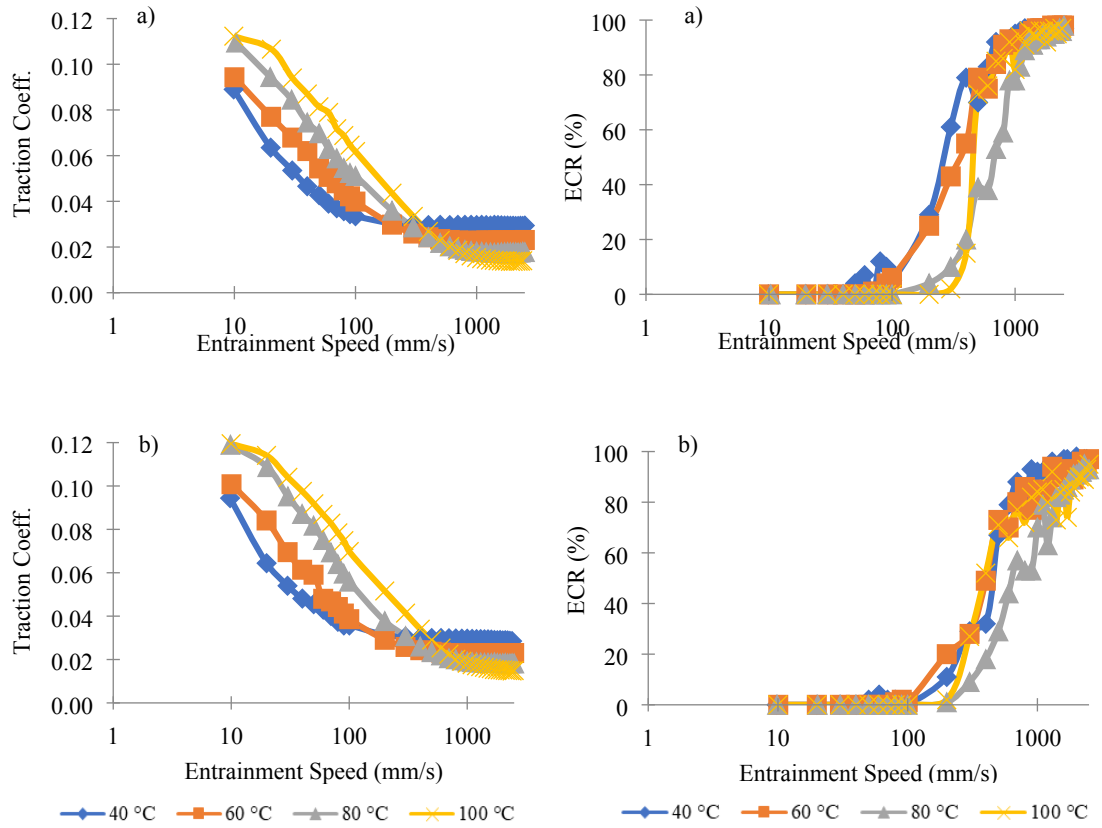


Fig. 2 Traction coefficient and ECR behavior versus speed for: a) POE + 0.5% [N_{6,6,6,6}][C_{8:0}] and b) POE + 2% [N_{6,6,6,6}][C_{8:0}].

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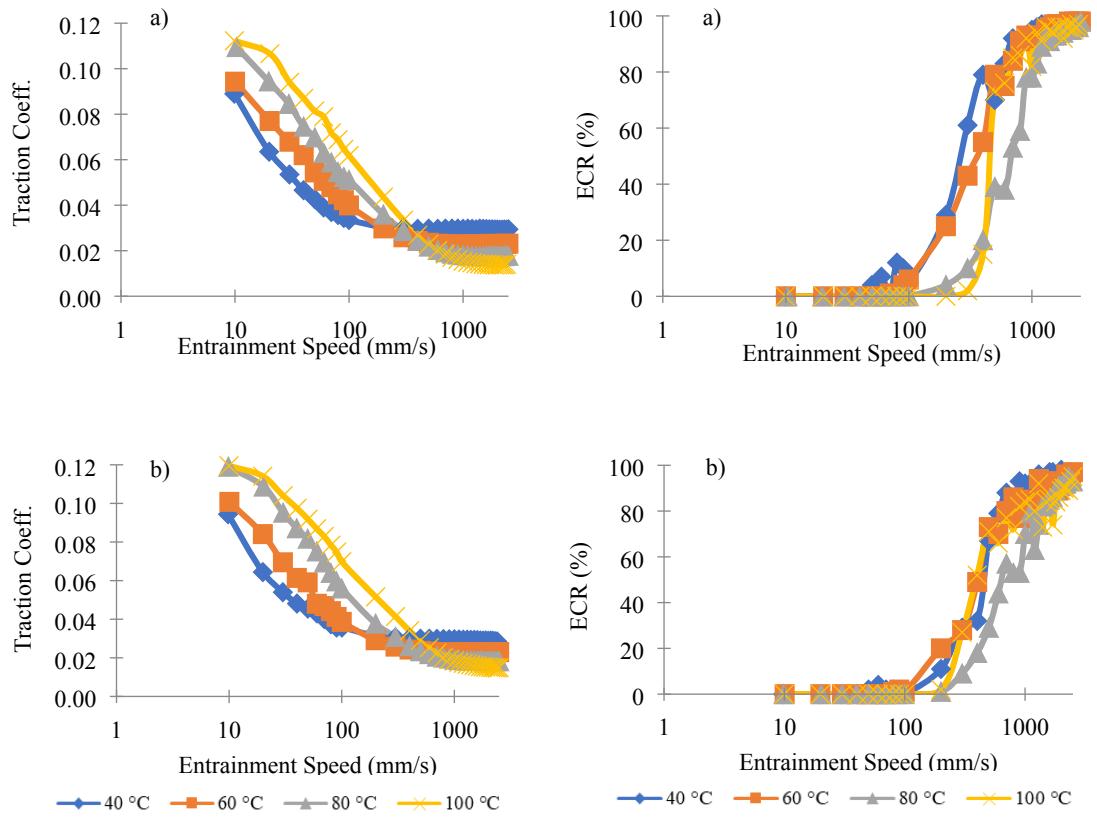
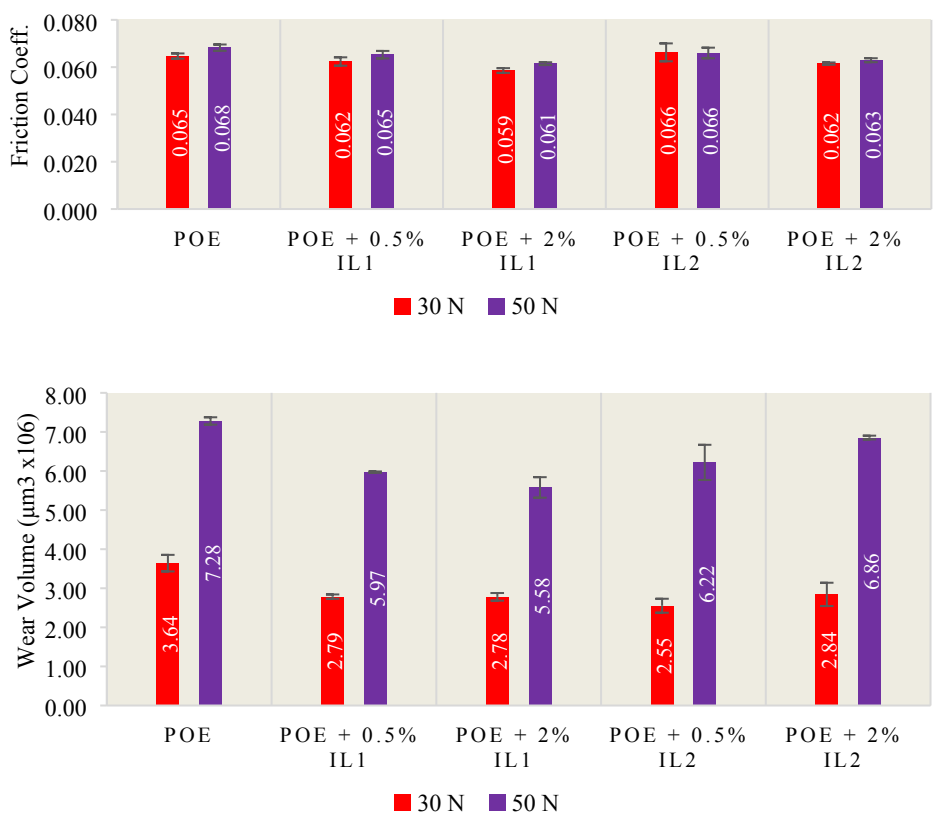


Fig. 3 Traction coefficient and ECR behavior versus speed for: a) POE + 0.5% [N_{6,6,6,6}][C_{16:0}] and b) POE + 2% [N_{6,6,6,6}][C_{16:0}].

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The results from tests made under reciprocating ball-on-disc configuration at 25 °C shows that mean friction coefficient is quite similar for both loads in the tests performed with mixtures and pure POE, Fig. 4. However, wear increased with load and the mixtures outperformed the antiwear behavior of POE. The antiwear behavior of all mixtures was quite similar at 30 N-load, meanwhile the [N_{6,6,6,6}][C_{8:0}]-containing mixtures outperformed the antiwear behavior of the [N_{6,6,6,6}][C_{16:0}] counterparts at 50 N.



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Fig. 4 Friction coefficient and wear volume from tests at 25 °C (IL1: [N_{6,6,6,6}][C_{8:0}], IL2: [N_{6,6,6,6}][C_{16:0}]).

16 For reciprocating tests at 100 °C, the antifriction behavior of the blends was slightly worse than that of the
17 base oil, Fig. 5. Analogously to tests performed at 25 °C, the antiwear behavior of the blends was better
18 than that of POE and wear increased with load. All mixtures had similar wear reduction properties at 30 N-
19 load; but the best antiwear behavior at 50 N-load was found for mixture with 0.5% of [N_{6,6,6,6}][C_{8:0}]. This
20 antiwear behavior of the FAILs as an additive at 25 and 100 °C-tests was very similar to that found in the
21 tests performed for these ionic liquids used as neat lubricant at the same temperatures [83].

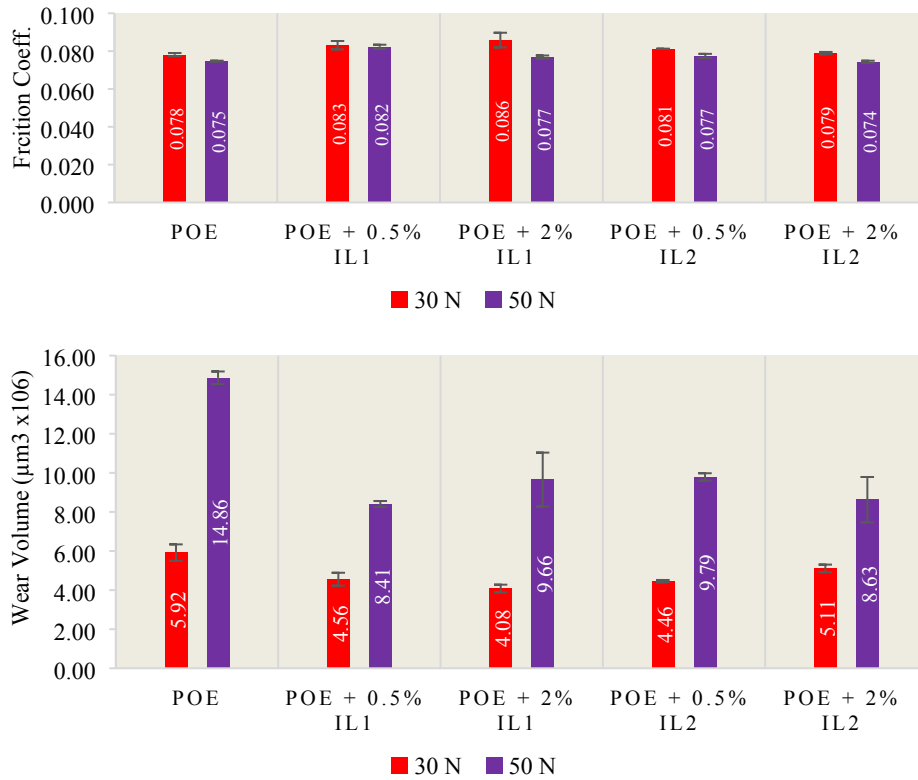
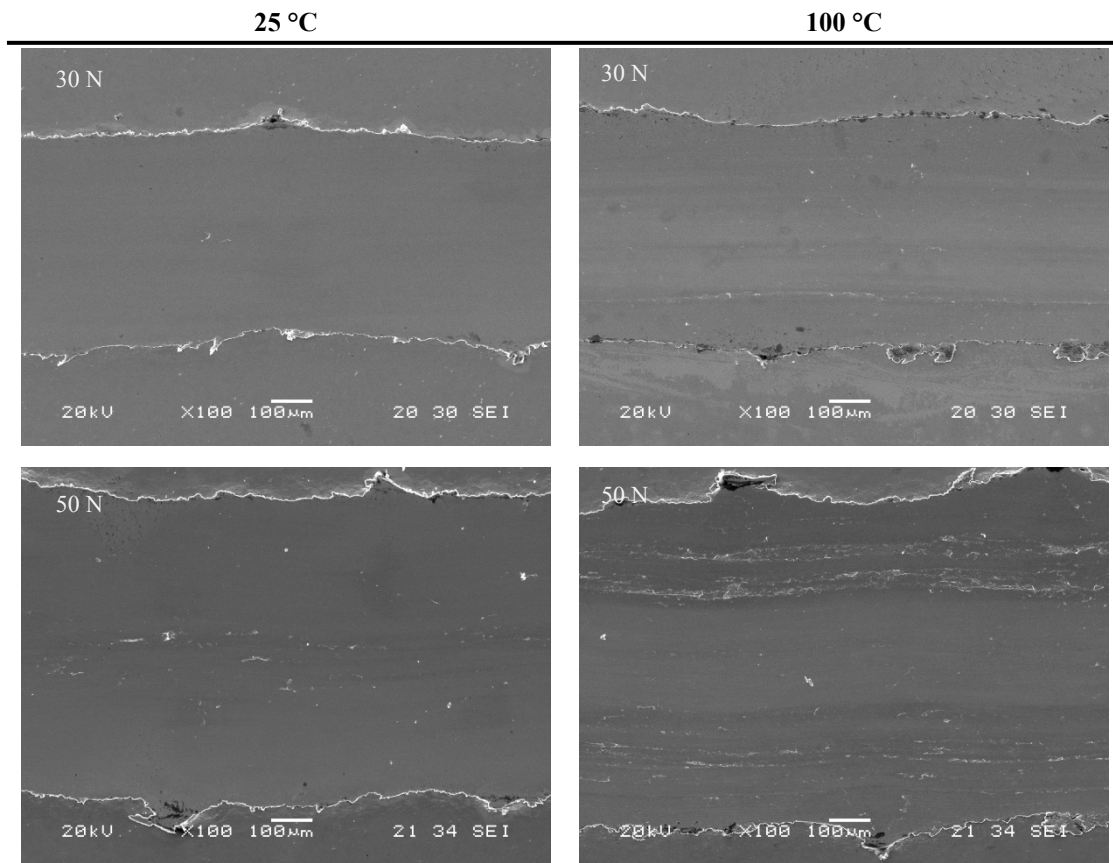


Fig. 5 Friction coefficient and wear volume from tests at 100 °C (IL1: $[\text{N}_{6,6,6,6}][\text{C}_{8:0}]$, IL2: $[\text{N}_{6,6,6,6}][\text{C}_{16:0}]$).

3.3. Surface characterization

The wear scar on the discs after reciprocating tests was studied by SEM and they showed that the predominant wear mechanism was mainly adhesive with plastic deformation (Figs. 6-10). As expected, wear increased with load and temperature. Taking into account the state of the worn surface, it seems that the $[\text{N}_{6,6,6,6}][\text{C}_{16:0}]$ behaves better at high temperature and load, maybe because its better thermal stability.



1 Fig. 6 SEM images of the wear track on the disc after tribological tests with pure POE at 30 and 50 N.

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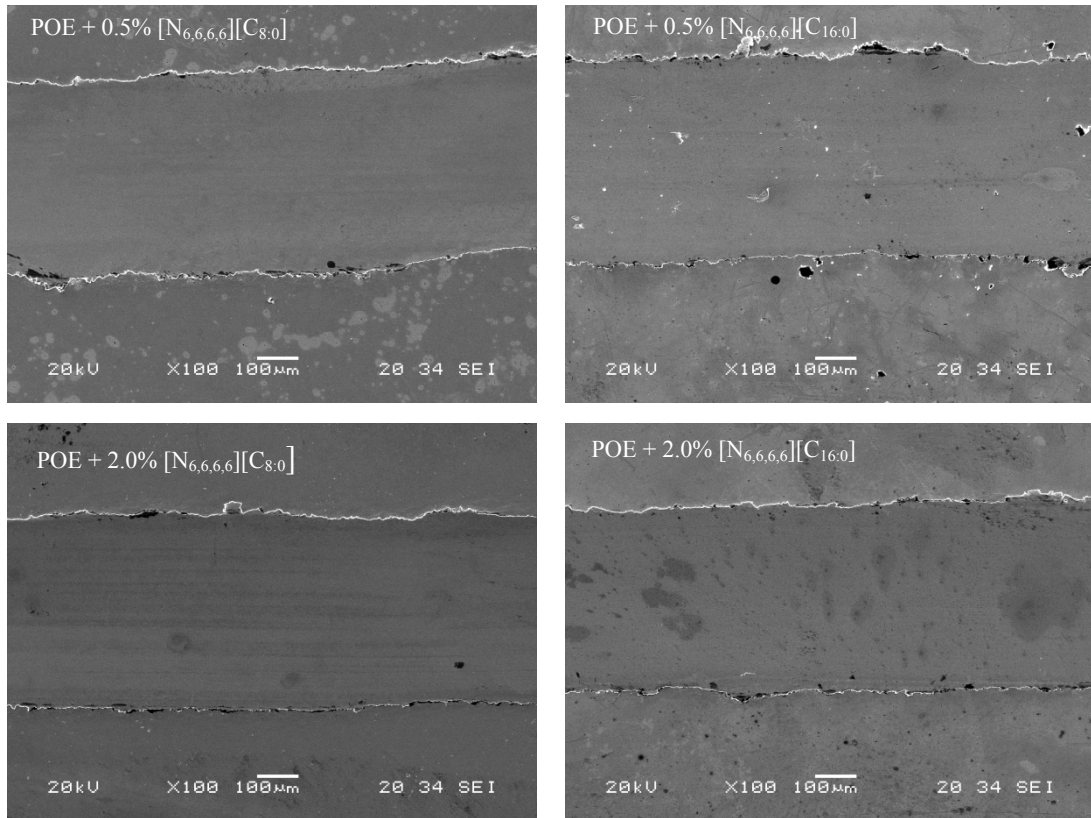


Fig. 7 SEM images of the wear track on the disc after tests with $[N_{6,6,6,6}][C_{8:0}]$ - and $[N_{6,6,6,6}][C_{16:0}]$ -containing mixtures at 25 °C and 30 N.

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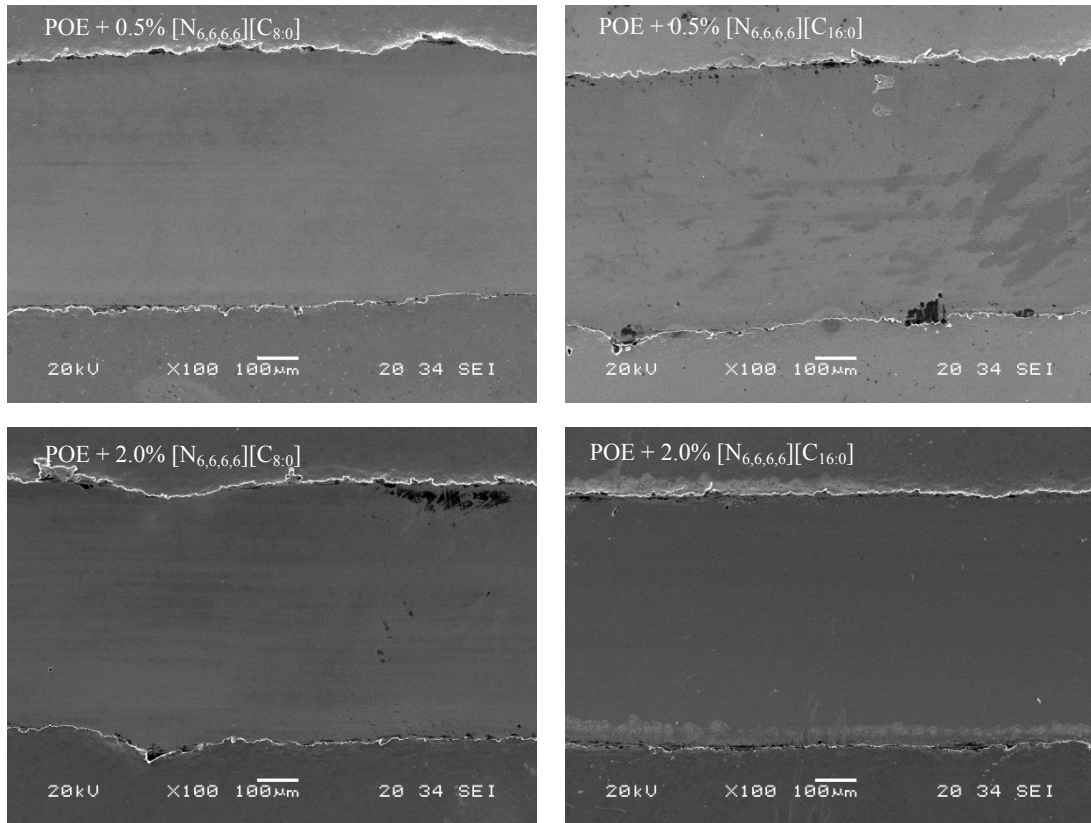


Fig. 8 SEM images of the wear track on the disc after tribological with [N_{6,6,6,6}][C_{8:0}]- and [N_{6,6,6,6}][C_{16:0}]-containing mixtures at 25 °C and 50 N.

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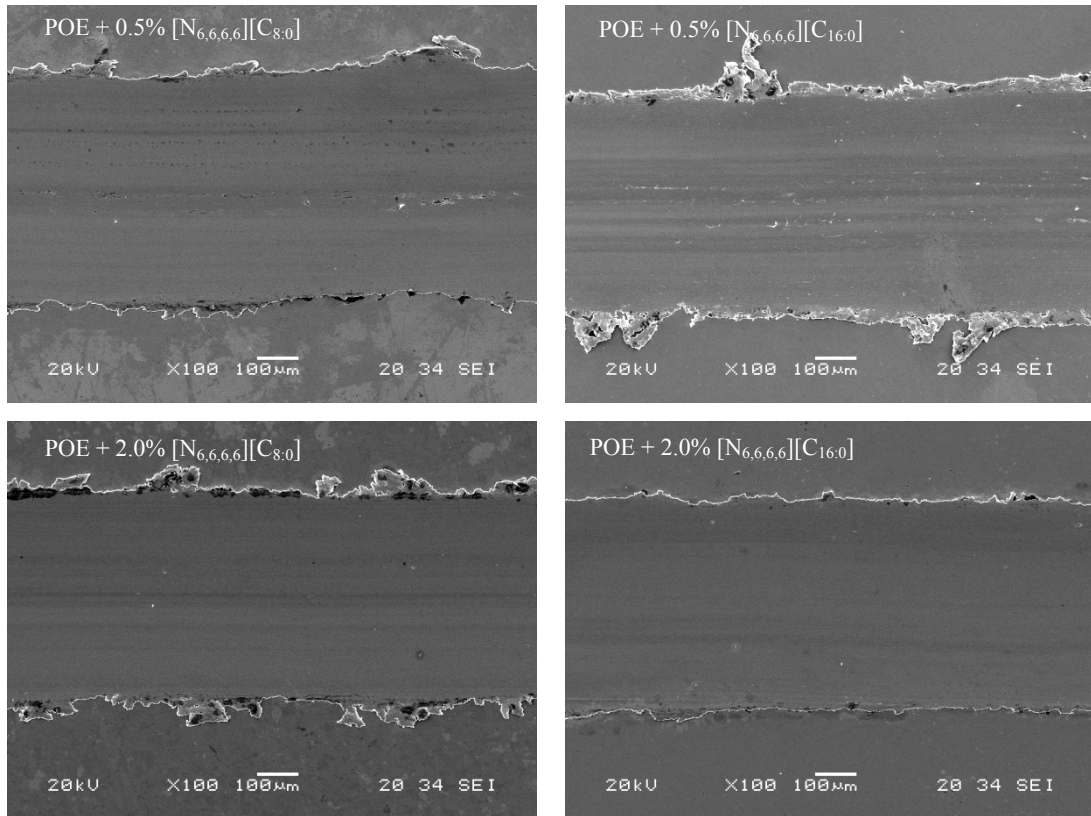
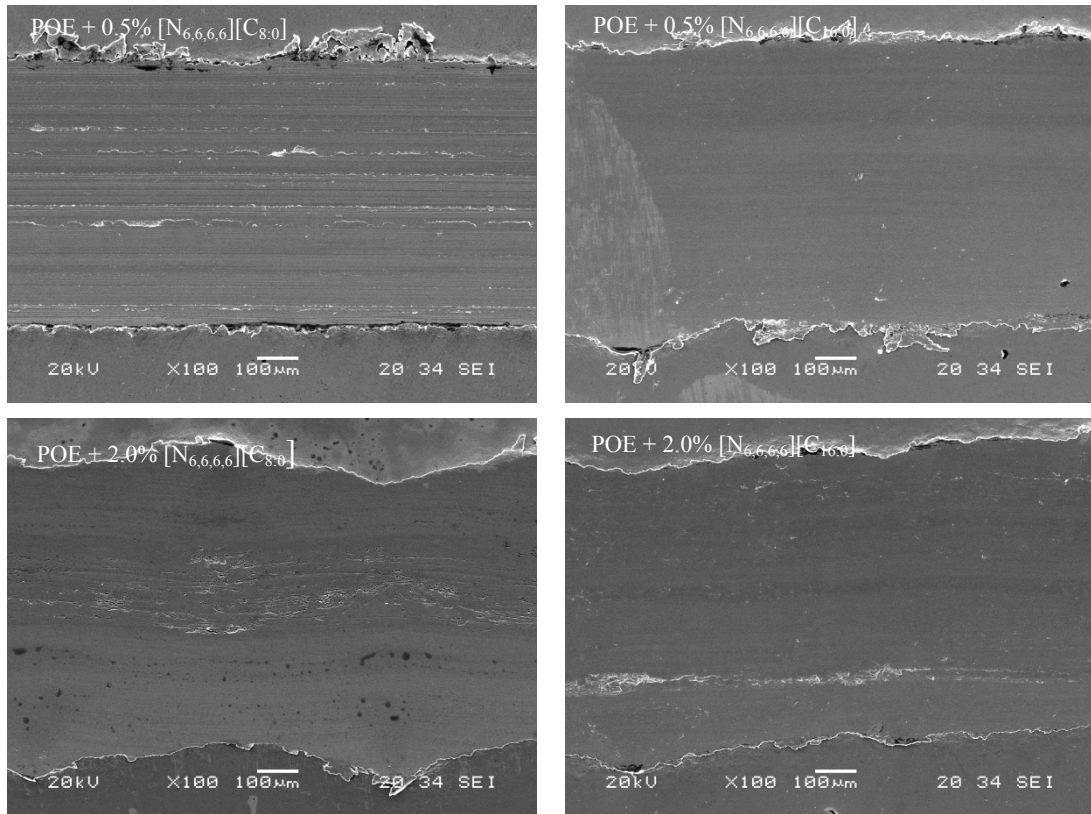


Fig. 9 SEM images of the wear track on the disc after tribological tests with [N_{6,6,6,6}][C_{8:0}]- and [N_{6,6,6,6}][C_{16:0}]-containing mixtures at 100 °C and 30 N.

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2 Fig. 10 SEM images of the wear track on the disc after tribological tests with [N_{6,6,6}][C_{8:0}]- and
3 [N_{6,6,6}][C_{16:0}]-containing mixtures at 100 °C and 50 N.
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5 For clarifying the tribological performance of these FAILs as an additive to a polyol ester at high
6 temperature (more probable scenario in a real application), the worn surface of the discs from some tests
7 were analyzed by XPS. Curve fitting for Fe2p^{3/2} (Table 2) shows four different peaks, which can be assigned
8 to Fe(0) (Gaussian-Lorentzian 70:30 with an exponential blend tail k=0.65), FeO (Gaussian-Lorentzian
9 70:30), Fe₂O₃ (Gaussian-Lorentzian 70:30 with an exponential blend tail k=1.5) and FeOOH (Gaussian-
10 Lorentzian 70:30) [56,88,89]. Samples with the lowest wear volume (those tested at 30 N) showed very
11 low amounts of Fe(0) on the wear track. Steel surface is more accessible to air oxygen than the inner part
12 of the sample and, therefore, it is more likely to find Fe(0) in the inner layers and oxidized Fe in the more
13 surface layers. This becomes evident when analyzing the iron composition on the wear track in tests at 30
14 N and 50 N. The ones tested with the lowest load showed less damage and, therefore, the Fe(0) inner layers
15 are not yet reached and only iron oxides can be found. However, the higher wear damage obtained in tests
16 at 50 N led to reach the Fe(0) layers below the surface.

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Table 2. XPS bands of the Fe₂p^{3/2} (Temp.: 100 °C).

Load: 50 N								
Lubricant samples	Peak position (eV)				Peak contribution (%)			
	Fe(0)	FeO	Fe(III)	FeOOH	Fe(0)	FeO	Fe(III)	FeOOH
POE	706.6	709.5	711.2	713.7	21	33	38	8
POE + 0.5% [N _{6,6,6,6}][C _{8:0}]	707.1	709.9	710.8	713.8	21	19	51	10
POE + 0.5% [N _{6,6,6,6}][C _{16:0}]	707.0	709.8	710.4	713.4	22	1	65	12
POE + 2.0% [N _{6,6,6,6}][C _{8:0}]	707.0	710.0	710.5	714.1	19	3	69	9
POE + 2.0% [N _{6,6,6,6}][C _{16:0}]	706.9	-	709.7	712.4	21	-	63	15

Load: 30 N								
Lubricant samples	Peak position (eV)				Peak contribution (%)			
	Fe(0)	FeO	Fe(III)	FeOOH	Fe(0)	FeO	Fe(III)	FeOOH
POE	704.3	707.2	709.6	711.4	4	15	60	21
POE + 0.5% [N _{6,6,6,6}][C _{8:0}]	-	707.2	710.0	711.9	-	11	73	16
POE + 0.5% [N _{6,6,6,6}][C _{16:0}]	703.3	706.7	709.3	711.2	2	14	68	16

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4. Conclusions

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Two FAILs were studied as an additive to a polyol ester at two concentrations (0.5 and 2 wt%) and the

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main conclusions obtained are the following:

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- The use of these FAILs at low concentration did not change the viscosity of the POE and hence the tribological behaviors of the mixtures and the POE under elastohydrodynamic lubrication in rolling/sliding tests were similar. However, the mixtures showed lower friction values under mixed lubrication regime in most of the cases.

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- The friction behavior in reciprocating tests was similar with the use of all mixtures and the POE at 25 °C for the two loads used, while the mixtures outperformed the antiwear behavior of the POE.

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- The antifriction behavior of the mixtures under both loads at 100 °C in reciprocating tests was slightly worse than that of the POE and all mixtures had better antiwear performance than the POE.

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Conflict of Interest and Authorship Conformation Form

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- x All authors have participated in (a) conception and design, or analysis and interpretation of the data; (b) drafting the article or revising it critically for important intellectual content; and (c) approval of the final version.
- x This manuscript has not been submitted to, nor is under review at, another journal or other publishing venue.
- x The authors have no affiliation with any organization with a direct or indirect financial interest in the subject matter discussed in the manuscript.

Author Statement

M. Sernaglia: Investigation, Writing- original draft; **D. Blanco:** Conceptualization, Investigation, Data curation, Writing-review and editing, Resources; **A. Hernández Battez:** Conceptualization, Methodology, Writing-review and editing, Project administration, Funding acquisition; **A. Fernández-González:** Investigation, Data curation; **R. González:** Project administration, Funding acquisition, Supervision; **M. Bartolomé:** Data curation, Supervision.

SUPPLEMENTARY INFORMATION

Table 1. Density and viscosity of the different base oils.

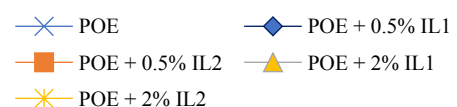
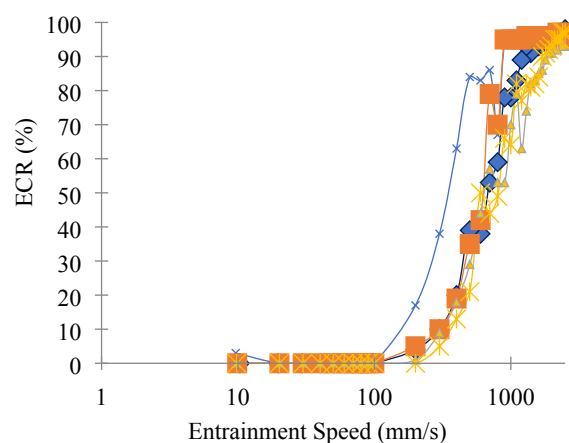
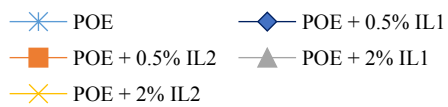
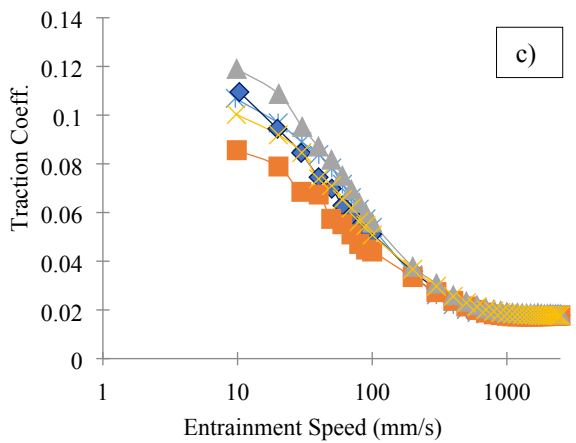
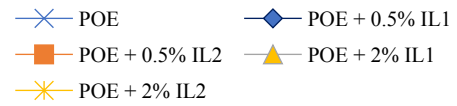
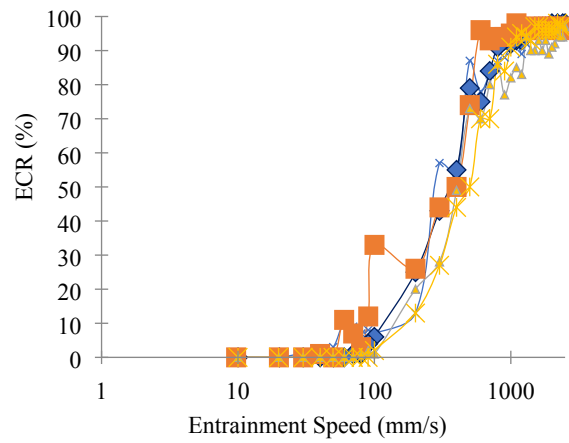
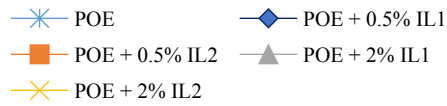
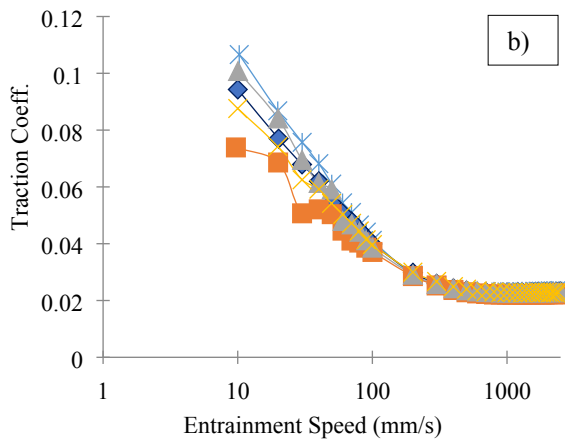
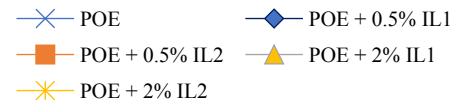
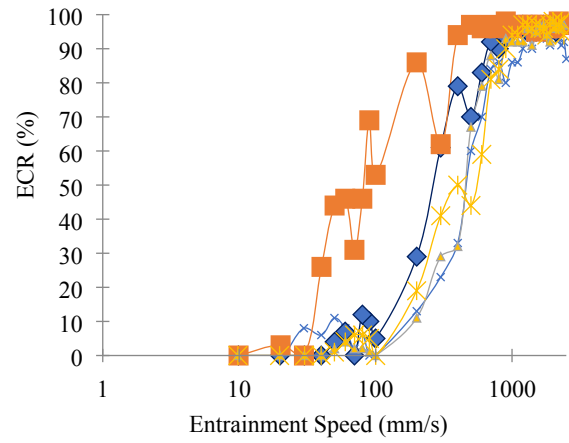
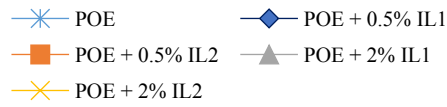
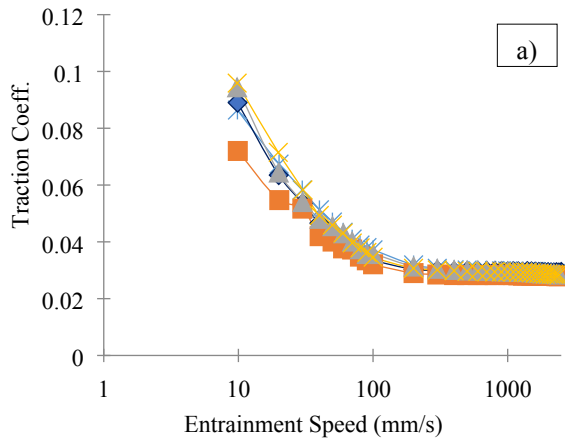
Base Oils	Density at 15 °C (g.cm ⁻³)	Kin. Viscosity (mm ² .s ⁻¹)		Viscosity Index
		40 °C	100 °C	
PRIOLUBE 3970 (POE)	0.9417	20.5	4.54	140
SN-150	0.8750	32.0	5.20	98
PAO 4	0.8193	17.8	3.99	124

Table 2. Density values of the FAIL-containing mixtures at different temperatures.

Samples	Density (g.cm ⁻³)								
	20 °C	30 °C	40 °C	50 °C	60 °C	70 °C	80 °C	90 °C	100 °C
POE + 0.5% IL1	0.9417	0.9344	0.9272	0.9201	0.9109	0.9058	0.8987	0.8916	0.8846
POE + 2.0% IL1	0.9412	0.9340	0.9268	0.9196	0.9125	0.9054	0.8983	0.8913	0.8842
POE + 0.5% IL2	0.9411	0.9338	0.9266	0.9195	0.9124	0.9052	0.8981	0.8911	0.8840
POE + 2.0% IL2	0.9407	0.9335	0.9263	0.9192	0.9120	0.9050	0.8979	0.8908	0.8837

Table 3. Kinematic viscosity of the FAIL-containing mixtures at different temperatures.

Samples	Kinematic viscosity (mm ² .s ⁻¹)								
	20 °C	30 °C	40 °C	50 °C	60 °C	70 °C	80 °C	90 °C	100 °C
POE + 0.5% IL1	45.34	29.66	20.33	14.64	10.92	8.44	6.70	5.46	4.52
POE + 2.0% IL1	48.16	31.26	21.31	15.28	11.35	8.74	6.92	5.61	4.63
POE + 0.5% IL2	45.34	29.70	20.36	14.66	10.93	8.45	6.71	5.46	4.52
POE + 2.0% IL2	47.72	31.05	21.19	15.21	11.31	8.72	6.91	5.60	4.63



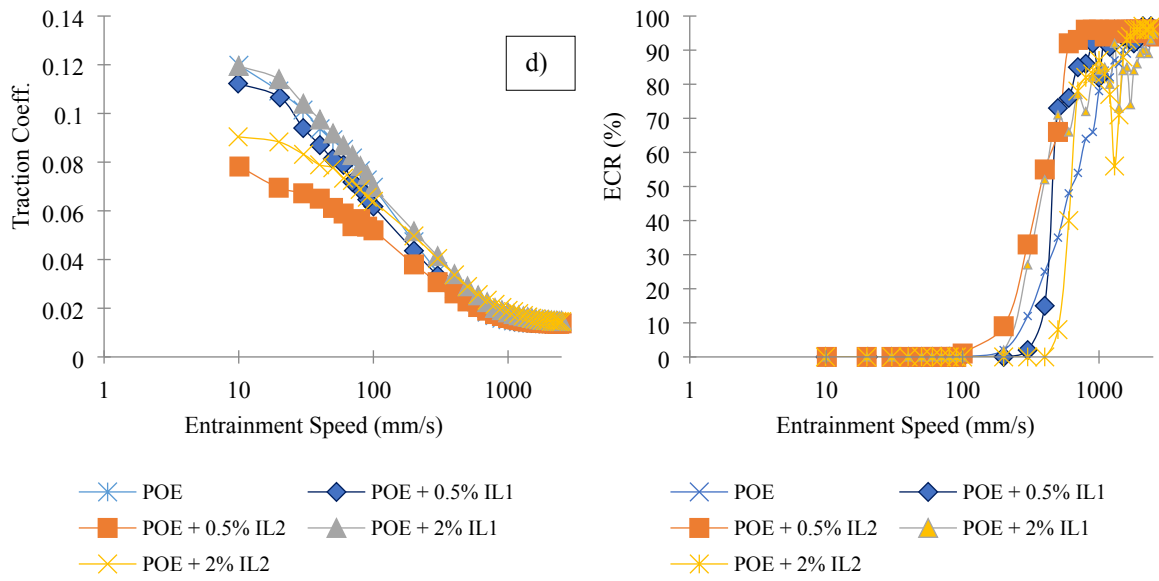


Fig. 1 Traction coefficient and electrical contact resistance (ECR) behavior versus speed for the pure base oil (polyol ester) and the base oil plus ionic liquids (IL1: $[N_{6,6,6,6}][C_{8:0}]$, IL2: $[N_{6,6,6,6}][C_{16:0}]$) at the temperature of: a) 40 °C; b) 60 °C; c) 80 °C; d) 100 °C.