Supporting Information

Formation of boundary film from ionic liquids enhanced by additives

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This file includes:

1. Lubricant data

2. Additional results

- a. $C\{150 \text{ N}/25 \text{ °C}/30 \text{ min}\}$ Wear scars
- b. $V\{100-300 \text{ N}/25 \text{ °C}/120 \text{ min}\}$ Wear scars
- c. V{150 N/40-100 °C/80 min} EDS spectra
- d. $V{300 N/40-80 °C/60 min}$ Disc wear profile map
- e. C{300 N/80 °C/30 min} Wear scars

1. Lubricant data

Five P-SiSO have been synthesized by Nisshinbo Holdings Inc. (Tokyo). The chemical structures is given by the formula $[(n-C_{\alpha}H_{2\alpha+1})_3(n-C_{\beta}H_{2\beta+1})P][(C_{\chi}H_{2\chi+1})_3SiC_{\delta}H_{2\delta}SO_3]$, where the variables α , β , χ , and δ determine the number of carbon atoms in the alkyl groups. Reported properties can be seen in Table S 1**Error! Reference source not found.** The bulkier cations, [4-16], and [6-14], have lower viscosities than the smaller [4-12] cation, as expected due to the weaker ion bonds in bulkier ions. In this work, only P-SiSO [6-14][1-2] have been evaluated. The PFPE sample was supplied by Aldrich and used as received, the properties are shown in Table S 2, with the full name of PFPE being Fomblin Y 317950. A fully formulated gear oil, Shell Spirax S6 AXME 75W-90 GL-5 is used in test V{100–300 N/25 °C/120 min}.

Table S 1. Properties of P-SiSO.

Code	Density, g cm ⁻³	Viscosity, mPa⋅s		Melting	10% weight loss at.	Water content,	
	25 °C	25 °C	60 °C	point, C	°C	ppm	Chemical formula
P-SiSO[4-12][1-2]	0.90	2570	208	-6	311	68	$[(n-C_4H_9)_3(n-C_{12}H_{25})P][(CH_3)_3SiC_2H_4SO_3]$
P-SiSO[4-12][1-3]	0.93	2429	104	2	291	93	$[(n-C_4H_9)_3(n-C_{12}H_{25})P][(CH_3)_3SiC_3H_6SO_3]$
P-SiSO[4-16][1-2]	0.91	1932	269	5	316	137	$[(n-C_4H_9)_3(n-C_{16}H_{33})P][(CH_3)_3SiC_2H_4SO_3]$
P-SiSO[4-16][1-3]	0.90	1735	142	-5	299	68	$[(n-C_4H_9)_3(n-C_{16}H_{33})P][(CH_3)_3SiC_3H_6SO_3]$
P-SiSO[6-14][1-2]	0.92	1879	200	-19	310	83	$[(n-C_6H_{13})_3(n-C_{14}H_{29})P][(CH_3)_3SiC_2H_4SO_3]$

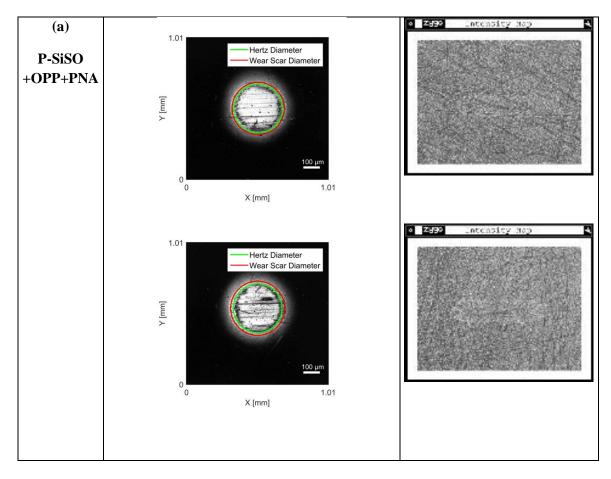
Table S 2. Properties of PFPE.

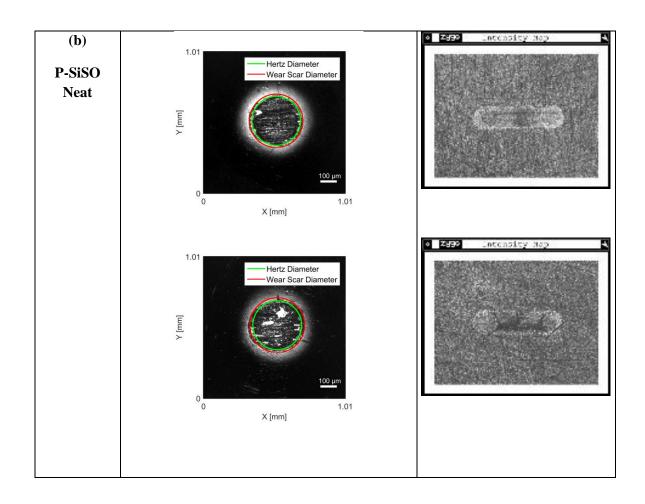
Code	Density, g cm ⁻³	Viscosity	Flash Poin	
	25 °C	20 °C	60 °C	
PFPE (Fomblin)	1.90	513	х	>113

2. Additional tribotest data

a. Scenario I

Wear scar results from C{150 N/25 °C/30 min}.





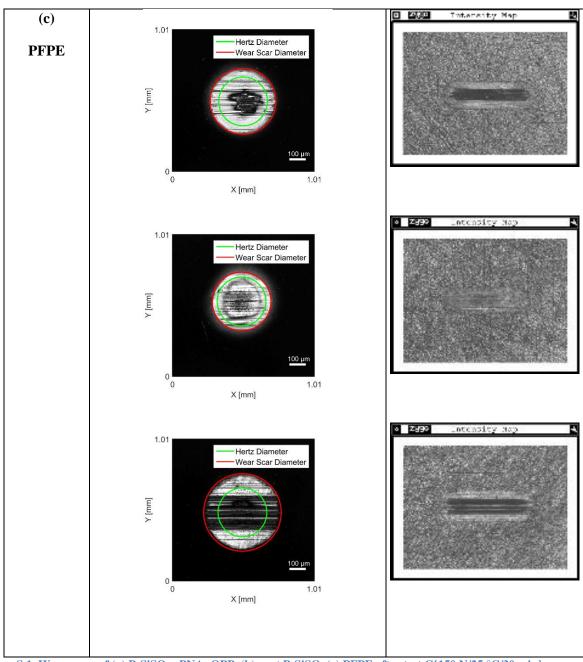
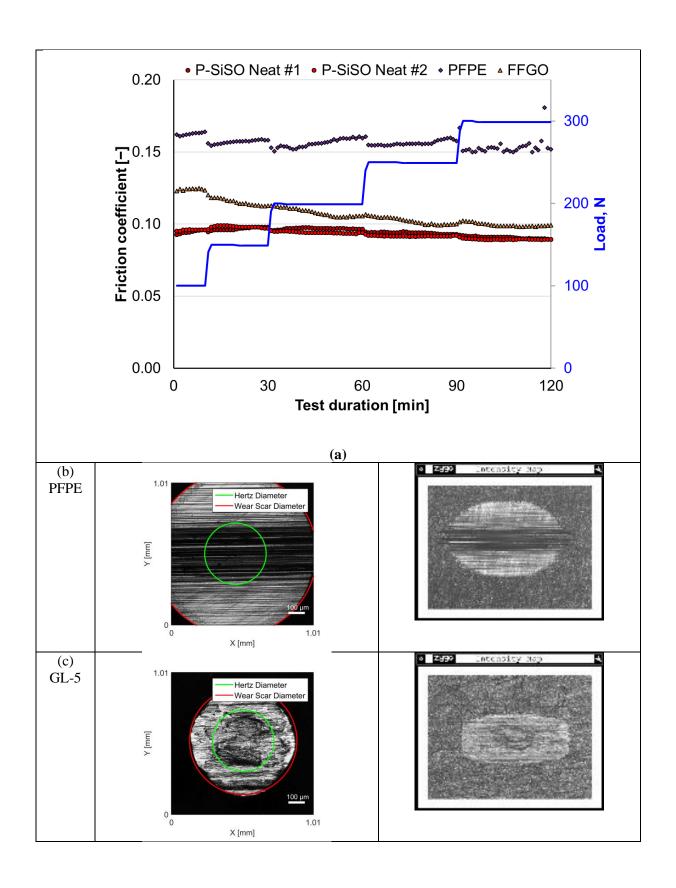


Figure S 1. Wear scars of (a) P-SiSO +PNA+OPP, (b) neat P-SiSO, (c) PFPE after test $C\{150 \text{ N}/25 \text{ °C}/30 \text{ min}\}$ (Corresponds to Figure 2). Disc image depicts reflected intensity for field of view 2.53 mm X 1.90 mm.

b. Scenario II

V{100-300 N/25 °C/120 min}



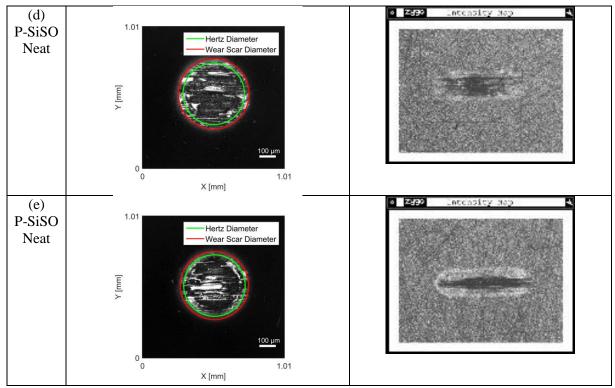


Figure S 2. (a) Friction trace V{100–300 N/25 °C/120 min}, (b) and (c), ball wear scars of neat P-SiSO, (d), wear scar of PFPE, (e), wear scar of GL-5 fully formulated gear oil.

V{150 N/40-100 °C/80 min}

EDS point mapping of tribofilm in Figure 5 (c). The electron beam is concentrated first at the center of the largest dark patch seen in Figure 5 (c), then at an adjacent bright region where no boundary film is visible. Spectrum show dark patch is rich in Si and O, P or S is not detected. The bright region is similar to an unused reference steel disc (labeled Ref: AISI52100), but with less O, as shown by the spectra in Figure S 3.

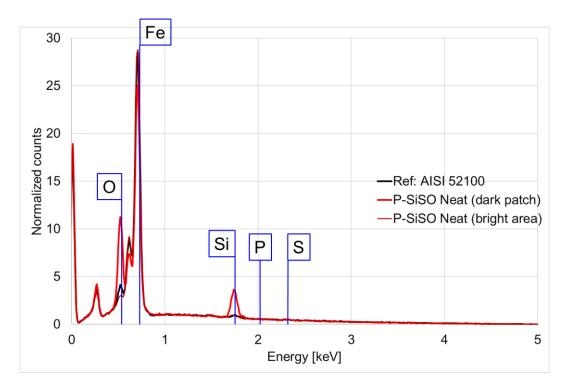
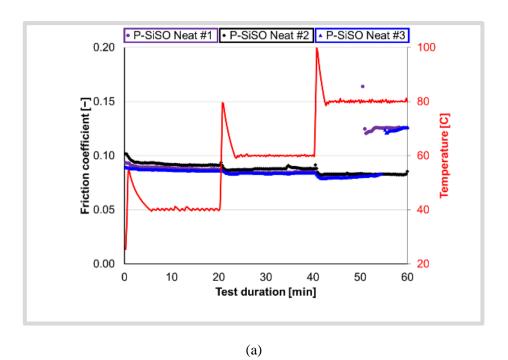


Figure S 3. EDS spectra corresponding to Figure 5 (c).

V{300 N/40-80 °C/60 min}



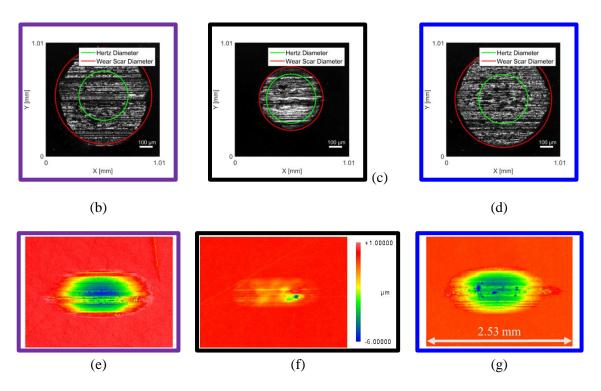


Figure S 4. Results from tribotest at condition V{300 N/40–80 $^{\circ}$ C/60 min}: (a), Abrupt change in friction at high temperature in for neat P-SiSO. (b)-(d) ball wear scars, (e)-(g) disc wear profile map, same scale is used between images.

c. Scenario III

C{300 N/80 °C/30 min}

Table 1. Ball wear scars for tests at C{300 N/80 $^{\circ}\text{C/30 min}}.$

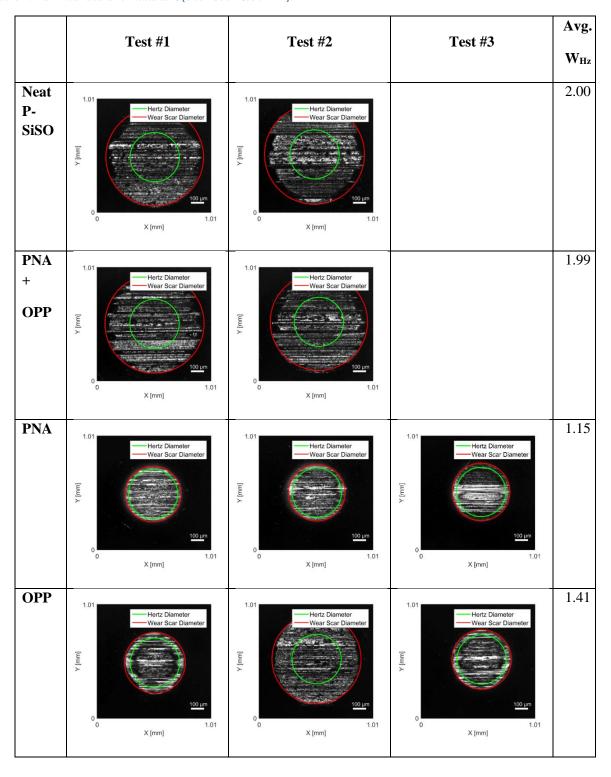


Table 2. Disc wear scars for tests at C{300 N/80 $^{\circ}\text{C/30 min}}.$

