Original Research Article

Synergistic effect of ionic liquid on additive performance of octyl acrylate-vinyl acetate copolymer and their comparison when added to lube oil

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ABSTRACT

Ionic liquids (ILs) have come out as a promising lubricant additive in base oil during the past few years due to their low volatility, non-flammability, thermal stability, moisture stability and miscibility with organic compounds. In the present work, an investigation was carried out to determine whether the flow improvement property and shear stability of a polymeric additive can be affected when blended with an ionic liquid. We prepared a copolymer of vinyl acetate and octyl acrylate and blended it with imidazolium–based IL. Their performance as pour point depressant (PPD), viscosity index improver (VII) and also their stability against the mechanical shear in terms of permanent shear stability index (PSSI) were determined when added to the base oil as solo additive or as blend. The results revealed that the blend outweighs the polymer and the IL in all respect as a lubricant additive. As PPD, the best result was obtained in case of the blend at 4% concentration in both the base oils i.e. –12.1 °C and –13.7 °C in B01 and B02 respectively. Kinematic viscosity loss was least when the base oils were doped with the blend. This was supported by the PSSI data: 24.22 at 1% concentration in B01 and 11.82 at 3% concentration in B02.

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Introduction

The automotive industry is getting flourished day by day with new inventions and applications of lubrication science which is itself an ever exploring research area. A typical lubricant is composed of mineral base oils doped with different categories of additives such as antioxidants, detergents, dispersants, friction modifiers, pour point depressants (PPDs), antiwear additives, extreme-pressure additives, and viscosity modifiers as per requirement [1]. Recent advances with ionic liquids (ILs) have provoked many studies on the application of ILs in the lubrication industry.

Ionic liquids are molten salts composed of large, asymmetric organic cations like imidazolium, ammonium, phosphonium etc and usually an inorganic anion such as tetrafluoroborate and hexafluorophosphate. The electrostatic force between the ions is highly diminished as the charges on the ions are usually diffuse over the large molecule and hence they fail to acquire a regular crystalline shape, rather they opt for the liquid state at room temperature mostly. ILs are unique to exhibit versatile properties such as low volatility, non-flammability, thermal stability, moisture stability, miscibility with organic compounds, good electronic and ionic conductivity and its broad application in electrochemical field [2]. They are often referred to as "designer solvents" due to their ability to dissolve a variety of organic, inorganic and organometallic compounds by simply choosing the proper combination of cations and anions. In addition ILs are considered to be more environmentally friendly in comparison to current alternative solvents and electrolytes [3] and for this reason are treated as green solvents [4]. Most importantly, due to the availability of various combinations of cation and anion enriched with unique properties, ILs have attained enormous attention over the decade [5] and today variety of ILs are explored with versatile application.

Such a promising range of properties along with their potential for controlled reactivity and also their low evaporation, researchers are provoked to explore their application in corrosion protection [6, 7] and tribology. Especially their extremely low vapor pressure, non-flammability, and thermal stability even up to temperatures above 200 °C are accounting for their capability of lubricating even at increased temperatures and pressure. So far most of the IL based lubricants were investigated at a more fundamental level; however, some studies have explored their
applications such as in engine lubrication [8–11] and micro–electromechanical machines (MEMs) [12–14]. M. Anand et al. [15] have explored the high miscibility and non corrosive behavior of two phosphonium based ILs i.e. Trihexyltetradecyl phosphonium bis(2, 4, 4–trimethylpentyl) phosphate and Trihexyltetradecyl phosphonium bis(2–ethylhexyl) phosphate and used them successfully as additive in the fully formulated diesel engine lubricants. This study provided a pathway for the used lubricants to recover their tribological performance for further use at the end of service life. Patrick Rohlmann et al. [16] evaluated phosphonium orthoborate ionic liquid as a wear reducing additive in biodegradable oils at steel–steel surfaces in the boundary lubrication regime. Again 1, 4–bis(2–ethylhexyl) sulfo succinate anion–based ILs containing different cation–imidazolium and organoammonium were synthesized and characterized as candidate lubricant additives in synthetic ester oils by Zeyun Wang [17]. Tribological tests on these synthesized ILs which were fully miscible with synthetic ester oil and noncorrosive to steel, revealed effective friction reduction and antiwear functionality. In addition, they worked well with the other typical additives, such as viscosity improver, antioxidant and pour point depressant. In another work, three phosphonate ionic liquids such as 1–butyl–3–methylimidazolium O–butyl phosphonate, N, N–dibutylammonium O–butyl phosphonate and N, N–dibutylammonium O–ethyl phosphonate were used as additives in polyethylene glycol (PEG) for steel/steel contact by Yunyan Han et al. [18]. Their characterization showed that these ionic liquids improved the tribological performances of the base oil significantly. The reason was the competitive adsorption between the base oils and the additives and also the chemical reactions between the ionic liquids and the metal substrate. V Pejakovic et al. [19] studied the influence of temperature on tribological behaviour of a sulfate–based ionic liquid with a pyrrolidinium cation as neat lubricant and as additive for glycerol in lubrication of steel–steel contacts and demonstrated that the ionic liquid played a significant role in the friction and wear reduction, as well as in the smoothening of the worn surface. Shuyan Yang et al. investigated the tribological performance of IL additives in the mixed lubrication and the elasto–hydrodynamic lubrication regimes and evaluated that IL additives could reduce friction and metal wear compared to pure PEG in mild conditions [20]. In another study, ionic liquids were investigated as a possible base oil of next–generation space grease to solve mainly temperature related issues and compatibility with the space environment [21]. The newly synthesized perfluoropolyether (PFPE) ionic liquid whose terminal group is an ammonium salt with a carboxylic acid has better frictional properties and has been used for magnetic tapes for about a quarter century because of their good lubricity [22]. Ammonium–based protic ionic liquids (PILs) were tested as lubricant fluids for aluminum–steel contacts by Maria Rita Ortega Vega et al. and they showed that the increase of the anion carbon chain length in the PIL structure reduced significantly the coefficient of friction value [23]. Jun Qu et al. evaluated the anti–wear behavior of an oil–miscible ionic liquid, i.e. trihexyltetradecylphosphoniumbis (2–ethylhexyl) phosphate in a base oil at 1.0 wt% concentration under both room and elevated temperatures and reported that the IL and the conventional anti–wear additive, zinc dialkyl–dithiophosphate (ZDDP) worked equally well to prevent scuffing and reduce wear in the room temperature tests. However, the IL significantly outperformed ZDDP in the tests performed at 100 °C [24]. Patrícia M. Amorim et al. [25] tested
imidazolium–based ILs as additives to the base oil PEG to lubricate Si surfaces and found that almost all the additives (2 wt%) led to a decrease in friction coefficient as well as an increase in viscosity. W. John et al. have successfully utilized 1-ethyl-3-methylimidazolium acetate and 1-buthyl-3-methylimidazolium acetate ionic liquids as solvent and stabilizing agent for the preparation of cobalt nanoparticles via chemical reduction method [26]. Mohammad Jahidul Islam et al. have synthesized biologically active morpholinium based Ionic Liquid having inorganic anions like nitrate and nitrite [27].

Imidazolium based ILs were studied extensively owing to their higher stability than a conventional synthetic oil at 200 °C [28] easy adsorption onto a surface, such as phosphonyls and esters [29] and easy variation of alkyl chain length to tune up wear and friction [30, 31] according to requirement. From various studies it was determined that the longer the alkyl chain length in the imidazolium group, the lower the wear [29, 32]. Again number of studies suggested that ILs having highly fluorinated anions possess higher thermal stability. Thus imidazolium tetrafluroborates were hugely utilised due to their ability to exhibit lower friction and wear than traditional lubricants for a number of systems [33]. These immense applications of imidazolium tetrafluoroborates encouraged us to utilise it in our present work.

However, the ILs are very much expensive compared to mineral base oils and synthetic base oils and hence they cannot be used solely in large volume. On the other hand, they may be used as a lubricant additive where a small amount of IL is doped into base oil, but provides a large difference to the friction and wear [34]. But there comes a problem with solubility of the IL in base oil. Generally ILs due to their polarity, are not so easily soluble in non polar base oils [10, 35] but it is seen that longer the alkyl chain length on the imidazolium group, higher is the solubility of IL in base oils [1]. Therefore, we opted for 1-decyl-3-methylimidazolium tetrafluoroborate (Figure 1) for this present work.

![Figure 1. 1-decyl-3-methylimidazolium tetrafluoroborate](image)

Number of polymeric additives have been synthesized and evaluated in this field of lubrication. Versatile studies on modification of polymers according to requirement have been reported. Mohammad H. Nasirtabrizi and S. J. Mousavi have shown that the chemical modification of maleic anhydride copolymers with 2-amino ethyl benzoate substituents as side chains decreased the free volume of the polymers as well as the rigidity whereas glass transition temperature is increased [36]. Here we have prepared a copolymer of vinyl acetate and octyl acrylate and blended it with the said IL. Vinyl acetate and acrylate–based polymers are broadly applicable to improve the flow ability of even very waxy crude oil, diesel fuel to other base oils at low temperature [37, 38]. Vinyl acetate copolymers satisfy most of the properties, a good additive must possess. This reason actually prompted us to explore it in this present study where we have investigated the effect of the IL under consideration on the flow improvement properties of acetate–acrylate based copolymer.

**Experimental**

**Materials and methods**
Octyl alcohol, Vinyl acetate (VA, 99%, S. d Fine Cheme Ltd.), Acrylic acid were used without purification. Toluene (99.5%, Merck Specialties Pvt. Ltd.), Hexane (99.5%, S. d Fine Cheme Ltd.) and Methanol (98%, Thomas Baker Pvt. Ltd.) were used after distillation. Hydroquinone (99%, Merck Specialties Pvt. Ltd.) and Benzoyl peroxide (BZP, 98% Thomas Baker Pvt. Ltd.) were purified by recrystallization before use. Conc. H$_2$SO$_4$ (98%, Merck Specialties Pvt. Ltd.) was utilised as received. The preferred IL 1–decyl–3–methylimidazolium tetrafluoroborate was obtained from TCI Chemicals (India) Pvt. Ltd. Properties of the mineral base oils are tabulated in Table 1.

**Table 1. Base oil (BO) properties**

| Physical Property                  | BO1   | BO2   |
|-----------------------------------|-------|-------|
| Density (g/cm$^3$ at 40°C)        | 0.83  | 0.95  |
| Viscosity at 40 °C in cSt         | 6.698 | 24.211|
| Viscosity at 100 °C in cSt        | 2.02  | 4.47  |
| Viscosity Index (VI)              | 81.13 | 89.87 |
| Pour Point (PP in °C)             | -3    | -6    |
| Cloud Point (°C)                  | -10   | -8    |

BO1, BO2 collected from IOCL, Dhakuria, West Bengal

**Preparation of esters and their purification**

The ester (octyl acrylate, OA) was prepared by reacting 1.1 mol of acrylic acid with 1 mol of octyl alcohol in presence of concentrated sulfuric acid as a catalyst, 0.25% hydroquinone with respect to the total reactants as polymerization inhibitor and toluene as a solvent. The method of esterification and its purification were carried out by the process as reported in the earlier publication [39].

**Preparation of polymer and its blend with il**

The copolymer of vinyl acetate and octyl acrylate was prepared by free radical polymerisation in presence of BZP as initiator (0.5% w/w, with respect to the total monomer) following the same process as described in our earlier publication [39]. The prepared and purified copolymer was blended with the ionic liquid in 1:1 ratio via mechanical blending process in a magnetic stirrer at 60 °C to obtain a homogeneous blend.

**Measurements**

The number average molecular weight ($M_n$) and weight average molecular weight ($M_w$) were determined using the GPC method (Water 2414, polystyrene calibration) in HPLC grade THF (0.4%, w/v) at 308K temperature at a flow rate of 1 mL/min.

**Spectroscopic measurements**

IR spectra were recorded on a Shimadzu FT–IR 8300 spectrometer using 0.1 mm KBr cells at room temperature within the wave number range (400 to 4000) cm$^{-1}$. NMR spectra were recorded in Bruker Avance 300 MHz FT–NMR spectrometer using a 5 mm BBO probe. CDCl$_3$ was used as solvent and tetramethylsilane (TMS) as reference material.

**Performance evaluation as pour point depressants in base oil**

The effect of additive concentration on pour point of the base oils was evaluated using different doping concentrations ranging from 1% to 6% (w/w) for each of the prepared
copolymers, IL and their blend. According to the ASTM D 97–09 method the pour points of the prepared solutions were measured using cloud and pour point tester model WIL–471 (India).

Performance evaluation as viscosity index improver (VI)

According to the ASTM D 2270–10 method viscometric parameters were determined at 313 and 373 K using different doping concentrations (% w/w) from 1% to 6% of the additives in the base oils. All of the experimental data, given in Table 3 & 4 were noted by taking an average of three experimental results under identical conditions in each case.

Shear stability measurement

Stability against mechanical shear is expressed in terms of shear stability index or PSSI values. It is basically measurement of viscosity loss under shearing condition which may be of two kinds such as Temporary Viscosity Loss (TVL) and Permanent Viscosity Loss (PVL) [40]. The required equations are as follows:

\[
PVL (\%) = \left( \frac{V_i - V_s}{V_i} \right) \times 100
\]
\[
PSSI (\%) = \left( \frac{(V_i - V_s) / (V_i - V_o)} \right) \times 100
\]

Where, \( V \) refers to Kinematic Viscosity (KV) at 100 °C.
\( V_o \) = KV of the oil before addition of polymer
\( V_i \) = KV of the oil after addition of polymer
\( V_s \) = KV of the oil doped with polymer after shearing

The PVL and PSSI values are measured for the solutions prepared using 1% – 6% (% w/w) doping concentration of the prepared copolymer, IL and their blend in the base oils as per ASTM D–3945 method and are given in Table 5.

Results and Discussion

Molecular weight analysis

The obtained values of \( M_n, M_w \) and PDI of the prepared copolymer are 25339, 53374 and 2.106 respectively. In general, these values are kind of a proof for a successful polymerisation.

Spectroscopic analysis

In IR spectrum for the copolymer of octyl acrylate and vinyl acetate (Figure 2) peak at 1732 cm\(^{-1}\) indicates the presence of ester carbonyl group and peak around 1165 cm\(^{-1}\) appeared for the carbonyl C–O stretching. No peak has been observed around 3100 cm\(^{-1}\) associated with alkenyl C–H stretching which supported the complete polymerization.

Figure 2. FT-IR spectrum of the polymer

In the \(^1\)H NMR spectrum of the polymer (Figure 3), the methyl and methylene protons appeared in the range of 0.88 to 1.93 ppm. The peak appeared in the range of 3.93 ppm indicated the presence of \(–OCH_2\) protons. Absence of any peak in the range of 5-6 ppm confirms the complete polymerisation.

In \(^13\)C NMR spectrum (Figure 4) the peaks around 174.7–177.8 ppm indicated the presence of ester carbonyl carbon. The peaks at 63.1–65.4 ppm corresponded to the \(–OCH_2\) carbons, peaks ranging from 14.1–45.1 ppm for all other sp\(^3\) carbons. Again no peak in the range of 120–150 ppm indicated the absence of unsaturation and confirmed complete polymerization.
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Performance evaluation as pour point depressant

Pour point data obtained from Table 2 implies that the copolymer and its blend with IL are effective pour point depressant in both the base oils. High molecular weight of the polymer accounts for its effectiveness as pour point depressant. However, the IL alone is hardly effective as PPD for the base oils. Since a polymeric backbone is a vital condition to be an effective pour point depressant the IL is lacking behind in this case. Due to the presence of a hydrophobic long alkyl chain which may interact with the wax coming out at a reduced temperature from the base oil, a little change is observed in the pour point of the base oils. With increasing concentration the depression slightly enhances and gets fixed at some point due to lower solubility at higher concentrations as clearly shown in Figure 5.
Table 2. Pour point data with respect to the different concentrations of the additives in base oil (BO)

| Additive | BO  | 0%  | 1%  | 2%  | 3%  | 4%  | 5%  | 6%  |
|----------|-----|-----|-----|-----|-----|-----|-----|-----|
| Polymer  | BO1 | -3  | -10.0 | -10.9 | -11.5 | -11.9 | -11.0 | -10.5 |
|          | BO2 | -6  | -12.0 | -12.5 | -13.2 | -11.9 | -10.8 | -10.7 |
| IL       | BO1 | -3  | -7.0  | -7.5  | -9.0  | -8.4  | -8.1  | -7.0  |
|          | BO2 | -6  | -8.1  | -9.3  | -9.5  | -10.2 | -10.1 | -10.1 |
|          | BO1 | -3  | -10.7 | -11.1 | -11.7 | -12.1 | -11.9 | -11.8 |
| Blend    | BO2 | -6  | -12.4 | -12.8 | -13.3 | -13.7 | -13.6 | -13.5 |

Figure 5. Variation of pour point with respect to the different concentrations of the additives

Better interaction was observed in case of the blend of the polymer and the IL which is supported from the pour point data given in Table 2. Comparing the data for the polymer and its blend with the IL, it was observed that they are more or less equally active as PPD. However, the blend was slightly better due to the fact that the IL under consideration also possessed a long alky chain which enhanced the activity of the polymer.

Performance evaluation as viscosity index improver

As seen in Table 3 and 4, the copolymer, IL and their blend acted as moderate to good viscosity index improvers in both the base oils. If compared between IL and the copolymer of octyl acrylate and vinyl acetate, the later actually outweighs the IL. Actually the polymers exist as a coil in the base oils at normal temperature. At elevated temperature this coil unfolds itself into an open chain structure. Thus it gets higher in volume and as a result exhibits higher surface area which is responsible for higher viscosity index of doped base oil. Therefore, the temperature effect was reversed in case of polymers and as a result polymers act as better viscosity index improver in base oil.

Comparing all the data (Figure 6) it was observed that IL and the polymer blend acted as the best viscosity index improver among all three. It simply suggested that the IL under consideration actually enhanced the viscometric property of the polymer. The observation is not in any contrary to the other base oil also. Irrespective of all the additives and base oil, viscosity index gradually increased with increasing concentration of the additive which may be due to an increase in the total volume of the micelles in the oil solutions [41].
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Table 3. Viscosity Index (VI) data with respect to the different concentrations of the additives in base oil 1 (BO1)

| Conc | Polymer KV1 | Polymer KV2 | VI  | IL KV1 | IL KV2 | VI  | Blend KV1 | Blend KV2 | VI  |
|------|-------------|-------------|-----|--------|--------|-----|-----------|----------|-----|
| 1    | 6.705       | 2.120       | 113 | 6.700  | 2.033  | 86  | 6.721     | 2.181    | 135 |
| 2    | 6.714       | 2.160       | 127 | 6.706  | 2.051  | 91  | 6.737     | 2.223    | 147 |
| 3    | 6.727       | 2.200       | 141 | 6.712  | 2.072  | 98  | 6.763     | 2.274    | 162 |
| 4    | 6.750       | 2.250       | 157 | 6.720  | 2.094  | 104 | 6.792     | 2.325    | 176 |
| 5    | 6.780       | 2.300       | 170 | 6.729  | 2.125  | 113 | 6.822     | 2.355    | 184 |
| 6    | 6.810       | 2.340       | 184 | 6.738  | 2.143  | 118 | 6.841     | 2.391    | 197 |

Conc represents concentration in % w/w; KV1 & KV2 represent kinematic viscosity at 40 °C & 100 °C respectively.

Table 4. Viscosity Index data with respect to the different concentrations of the additives in base oil 2 (BO2)

| Conc | Polymer KV1 | Polymer KV2 | VI  | IL KV1 | IL KV2 | VI  | Blend KV1 | Blend KV2 | VI  |
|------|-------------|-------------|-----|--------|--------|-----|-----------|----------|-----|
| 1    | 24.320      | 4.850       | 122 | 24.235 | 4.512  | 94  | 24.388    | 5.113    | 142 |
| 2    | 24.340      | 5.000       | 134 | 24.244 | 4.593  | 102 | 24.442    | 5.234    | 151 |
| 3    | 24.390      | 5.250       | 153 | 24.257 | 4.714  | 111 | 24.563    | 5.502    | 170 |
| 4    | 24.480      | 5.370       | 161 | 24.269 | 4.875  | 124 | 24.601    | 5.686    | 183 |
| 5    | 24.570      | 5.560       | 174 | 24.281 | 4.983  | 133 | 24.655    | 5.782    | 189 |
| 6    | 24.650      | 5.780       | 189 | 24.291 | 5.107  | 142 | 24.712    | 5.891    | 196 |

Conc represents concentration in % w/w; KV1 & KV2 represent kinematic viscosity at 40 °C & 100 °C respectively.

Figure 6. Variation of viscosity index with respect to the different concentrations of the additives

Shear stability index analysis

The polymer–IL blend showed highest stability against mechanical shear at all concentrations, which clearly supports its better mechanical and thermal property in both the base oils (Table 5). The PSSI data also suggested that the polymer is itself a good shear stable additive; however, when blended with the IL under consideration, the mechanical stability is enhanced much more. It implies that

[Graphs showing VI variation vs concentration]
the blend undergoes molecular degradation under mechanical shear at much lower rate compared to the polymer and the IL alone. Higher the concentration of the additive in the base oil, higher is the volume exposed to the mechanical shear. Thus the PSSI values increases with increasing concentration of the additive in both the base oils; however, this relationship is not totally linear since some other factors also come into play under severe mechanical shear like molecular weight of the polymer and composition of the base oil.

Graphical representation (Figure 7) provides clear comparison of shear stability among the additives. It clearly shows that IL alone is comparatively poor shear stable than the polymer and the blend. The blend of the polymer and the IL here has been proved to possess better mechanical stability.

### Table 5. PVL & PSSI data with respect to the different concentrations of the additives in base oil (BO)

| Base Oil | Conc<sup>n</sup> | KV<sub>2</sub> after shear | PVL (%) | PSSI (%) |
|----------|------------------|--------------------------|---------|----------|
|          | Polymer  | IL  | Blend | Polymer  | IL  | Blend | Polymer  | IL  | Blend |
| BO1      | 1        | 2.081 | 2.022 | 2.142 | 1.84 | 0.54 | 1.79 | 39.00 | 84.61 | 24.22 |
|          | 2        | 2.100 | 2.022 | 2.176 | 2.78 | 1.41 | 2.11 | 42.86 | 93.55 | 23.15 |
|          | 3        | 2.120 | 2.031 | 2.215 | 3.64 | 1.98 | 2.59 | 44.44 | 78.85 | 23.23 |
|          | 4        | 2.140 | 2.042 | 2.252 | 4.89 | 2.48 | 3.14 | 47.83 | 70.27 | 23.93 |
|          | 5        | 2.150 | 2.047 | 2.271 | 6.52 | 3.67 | 3.57 | 53.57 | 74.28 | 25.07 |
|          | 6        | 2.160 | 2.055 | 2.295 | 7.69 | 4.11 | 4.02 | 56.25 | 71.54 | 25.88 |
| BO2      | 1        | 4.655 | 4.476 | 5.032 | 4.02 | 0.79 | 1.58 | 51.32 | 85.71 | 14.15 |
|          | 2        | 4.710 | 4.492 | 5.137 | 5.80 | 2.19 | 1.85 | 54.72 | 82.11 | 12.69 |
|          | 3        | 4.800 | 4.502 | 5.380 | 8.57 | 4.48 | 2.21 | 57.69 | 86.88 | 11.82 |
|          | 4        | 4.837 | 4.528 | 5.545 | 9.93 | 7.12 | 2.48 | 59.22 | 85.68 | 12.84 |
|          | 5        | 4.890 | 4.565 | 5.614 | 12.05 | 8.39 | 2.91 | 61.47 | 81.48 | 12.80 |
|          | 6        | 4.930 | 4.635 | 5.709 | 14.71 | 9.24 | 3.09 | 64.89 | 74.09 | 12.81 |

<sup>n</sup> Conc represents concentration in % w/w; KV.<sub>2</sub> represents kinematic viscosity at 100 °C

**Figure 7.** Variation of PSSI with respect to the different concentrations of the additives
Conclusions

This research study found that the IL under consideration does play an effective role when blended with acetate–acylate copolymer. The polymer is itself a good pour point depressant, viscosity index improver, and at the same time, it is quite stable against mechanical shear under severe conditions. The IL alone is not as effective as the polymer in terms of pour point depressant; however, its viscometric properties are moderate. Above all, the blend of the polymer and the IL predominates in every property, i.e. pour point depression, viscosity index and shear stability. Though IL was proved to be a promising lubricating agent, its high cost retards its solo application. When blended with proper polymer, lower amount of IL is required which lowers the cost to a considerable amount and at the same time the blend exhibits better property than even the parent polymer also. As PPD, the best result was obtained in case of the blend at 4% concentration in both the base oils i.e. –12.1 °C and –13.7 °C in BO1 and BO2 respectively. Kinematic viscosity loss was least when the base oils were doped with the blend. Obtained PSSI data supported this observation: 24.22 at 1% concentration in BO1 and 11.82 at 3% concentration in BO2.

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Disclosure Statement

No potential conflict of interest was reported by the authors.

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