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New graphene/ionic liquid nanolubricants

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Abstract

In this work, we have prepared new graphene/ionic liquid dispersions by adding a 0.1wt.% proportion of 1-2 layers graphene (G1) or 1-10 layers graphene (G2) to the ionic liquid 1-octyl-3-methylimidazolium tetrafluoroborate (IL). The new dispersions (IL+G1) and (IL+G2) have been used as external lubricants in polymer-steel and ceramic-steel contacts. For AISI 316L stainless steel/epoxy resin, the order of friction reduction is (IL+G2)>IL>(IL+G1). The wear reducing order is the same, as abrasive wear takes place for (IL+G1), while G2 prevents any degree of surface damage on both materials, even the very mild wear observed for neat IL. The poor performance of G1 is related to the formation of abrasive graphene-IL aggregates. The (IL+G2) dispersion also shows superior friction reducing and anti-wear behavior under more severe contact conditions, for AISI316L against sapphire.

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Keywords: Epoxy resin; stainless steel; sapphire; ionic liquid; graphene; nanolubricants; friction; wear.

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1. Introduction

Graphene, constituted by one-atom thick layers of sp^2 carbon [1], has risen and extraordinary scientific and technical interest [2]. At the present moment, there exist diverse preparation methods [3] and very different commercial qualities of graphene. The tribological and surface science applications of graphene are some of the most front edge lines of research at the present moment [4, 5]. There exist very few precedents of the use of graphene in friction and wear reduction [4-9]. A 26% friction reduction and a 9% wear reduction were reported by Choudhary et al. [9], for steel by lubrication with hexadecane containing chemically modified graphene, by intercalation between the sliding surfaces. However, a diversity of friction behaviours have been found between different bi-layer graphenes [10]. The use of hybrid graphene nanocomposites has increased due to its effect in the improvement of polymer properties.

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The wear rate of polytetrafluoroethylene (PTFE) is reduced in four orders of magnitude by addition of a 10% of graphene sheets of 3-4 layers as nano-reinforcements [11, 12]. These authors have proposed that the graphene sheets could reduce wear by inhibiting crack propagation. Recent studies have shown that room temperature ionic liquids (ILs) can lower wear and friction in steel-polymer contacts [13, 14] among many other high performance tribological applications [15-19]. The effect of the use of ionic liquid (IL) as an internal lubricant in epoxy-IL composites in pin-on-disk friction tests has been previously studied [13]. Chemical modification of graphene has been described by covalent and non-covalent interactions [20]. Covalent modifications could modify the graphene conjugation system and its properties. Non-covalent modification by π–π stacking and van der Waals interactions, are believed to maintain the structure and properties of graphene. The combination of graphene and ionic liquids is being explored as novel lubricants for friction and wear reduction [21, 22]. We have recently described [23] the good tribological performance of epoxy matrix nanocomposites modified by dispersion of graphene modified by ionic liquid. In the present study, graphene-IL dispersions with non-covalent interactions between two different commercial graphene qualities and IL have been produced under mild conditions and used as external lubricants, and compared with the neat IL. The IL selected for the present study has previously demonstrated an excellent lubricanting performance [15] combines a high thermal stability with a high molecular polarity, which allows the formation of adsorbed layers on metal surfaces, and the presence of a long alkyl side chain which could improve the compatibility of nanophases with the polymer network. These properties have been tested, in the present work, in epoxy resin(ER)-steel contacts. The high load carrying ability of graphene and the high thermal stability of the IL lubricant anticipate a good tribological performance of graphene-IL dispersions also under the more severe conditions, thus we also present the results obtained for steel-ceramic lubrication.

2. Experimental

The IL 1-octyl-3-methylimidazolium tetrafluoroborate (purity >99%) from Iolitec, GmbH (Germany) and two commercially available graphene qualities were used, G1 (1-2 layers graphene from Avanzare Nanotechnology Spain) and G2 (1-10 layers graphene from Iolitec, Germany). To obtain the dispersions (IL+G1) and (IL+G2), G1 or G2 were added to the IL in a 0.1wt.% proportion, the mixtures were mechanically stirred in an agate mortar for five minutes and then at 1600 rpm for 30 seconds [23], finally the mixture was subjected under sonication for 30 min, 100%A and 30ºC. Epoxy resin disks were processed as previously described [23]. Transmission electron microscopy (TEM) and scanning electron microscopy (SEM), Raman spectroscopy and X-ray photoelectron spectroscopy (XPS) analysis techniques have been previously described [23]. For TEM, Raman and XPS, excess IL was removed from IL+G2 by repeatedly washing with acetonitrile and drying. Pin-on-disk tests were carried out with an ISC 200-PC pin-on-disk tribometer. For the polymer-metal contact, ER disks (diameter 40 mm; thickness 4 mm) against AISI 316L stainless steel balls (hardness 210 HV) with a 0.8 mm sphere radius were used under a normal applied load was 4.9 N (mean contact pressure 0.7 GPa), with a sliding velocity of 0.10 ms^{-1} and a sliding distance 500 m, for a sliding radius of 9 mm. For the ceramic-metal contact, sapphire (Al_2O_3, 99.9%) balls (Goodfellow Cambridge Ltd. UK) (Al_2O_3; 99.9 %; HV 2,750; Young’s modulus 445 GPa; Poisson’s ratio 0.24) of 0.75 mm sphere radius were tested against AISI 316L stainless steel disks (HV 200; Young modulus 197 GPa; Poisson’s ratio 0.27; surface roughness R_a 0.05 μm) of 25 mm diameter and 5 mm thickness, under a normal applied load of 0.98 N (mean contact pressure 1.3 GPa), with a sliding velocity of 0.10 ms^{-1} and a sliding distance 500 m. Optical micrographs were obtained with a Leica DMRX microscope. Wear rates were determined by means of a Talysurf CLI profilometer. At least three tests were performed under the same experimental conditions for each material, at room temperature with a relative humidity of 40±5%.

3. Results and discussion

3.1. Characterization of IL+G2.

TEM microscopy and Raman spectroscopy studies of G1 and IL+G1 have been previously described [23]. Figure 1 shows TEM micrographs of as-received G2 (figure 1a) and IL+G2 (figure 1b), after removing the excess IL. Energy dispersive X-ray (EDX) analyses show the presence of up to a 12 wt.% of fluorine from the tetrafluoroborate anion in G2 after being treated with IL. Fig. 1c compares the Raman spectra of G2 as received, and after treatment with IL and removal of IL excess. No significant effect is observed due to the IL in the characteristic D (1361.2 cm^{-}