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Hybrid effect of ZnS sub-micrometer particles and reinforcing fibers on tribological performance of polyimide under oil lubrication conditions

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ABSTRACT

In response to the challenges posed by friction and wear of tribo-components exposed to boundary and mixed lubrication conditions, there is increasing interest in the development of polymer-based, self-lubricating composites. This investigation addressed the effect of various fillers (i.e. short carbon fibers (SCF), short glass fibers (SGF), ZnS sub-micrometer particles and combinations of respective fibers with ZnS particles), on the friction and wear of polyimide (PI) when lubricated by poly-alpha-olefins (PAO). A sliding plate-on-ring geometry, using GCr15 steel rings, was used. It was demonstrated that the reinforcing fibers greatly improved the composites' wear resistance. SGF was observed to be more effective than SCF in respect to friction- and wear-reduction. Moreover, an obvious synergism between SGF and ZnS was identified. The hybrid composite ZnS/SGF/PI exhibited excellent tribological performance under oil lubrication. With respect to the hybrid formulation, the glass fibers imparted their high abrasion resistance to the composite; while ZnS led to complex tribo-chemical reactions. A stable tribofilm consisting of transferred materials and various tribo-chemical products improved the boundary lubrication capability of the sliding system.

1. Introduction

With the development of automotive engines and industrial machineries, more and more mechanical motion components operate often under mixed and even boundary lubrication regimes owing to frequent start-stop and extremely high load etc [1,2]. Traditionally, in order to minimize friction and wear of metal-metal contacts subjected to boundary and mixed lubrication regimes, some proportions of friction modifiers and anti-wear additives were introduced into the lubricant [3]. During the rubbing process, the generation of a high-performance tribofilm was believed important for improving the boundary lubricity of the sliding pair [4]. However, seizure problems can still occur during long term engagement of metal-metal friction pairs with an unusual high load or low viscosity oil [5]. In order to enhance the reliability and life span of such motion systems, one potential solution is to replace conventional metal-metal friction pairs with high performance nonmetal-metal ones [6]. In this context, polymer-based materials are increasingly studied as tribo-materials because of their outstanding properties, such as light weight and self-lubrication capabilities [7–10].

In the last decades, tribological behaviors of polymer-based materials were extensively investigated under dry sliding conditions. Series of self-lubrication composites were developed successfully for satisfying numerous "non-lubricant" tribo-applications [11–14]. Moreover, the tribological mechanisms of polymer-based materials have been studied in-depth based on tribofilm and interface analyses [15–17]. However, in comparison to vast number of investigations of polymer-based composites under dry friction conditions, much less researches were conducted under oil lubrication conditions [18,19]. Our previous studies demonstrated that the addition of abrasion resistant fibers enhanced significantly the wear resistance of epoxy under oil lubrication [1,6]. It was found that besides the abrasion resistance, formation of a high-performance tribofilm was also of great importance for the tribological performance of the polymer composites. However, the presence of oil molecules on the interface complicated the formation of tribofilm and exerted an important role in the tribological performance of polymer-based composites [2]. In spite of the previous efforts, the tribological mechanisms of polymer-based composites under oil lubrication conditions are still not well

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understood and hence required further elucidation.

Polyimide (PI) as one of high-performance engineering polymers can be processed by regular molding techniques and offers excellent mechanical, chemical, and thermal stabilities [20]. PI has attracted attentions for tribo-engineering applications under high normal loads and/or high temperatures [21]. Nevertheless, the long running-in duration, high friction coefficient and poor wear resistance of neat PI restrict its application in the field of tribology [22]. In order to enhance the tribological performance of neat PI, various functional fillers and fiber reinforcement were added into PI matrix [23]. ZnS is a kind of metal compound solid lubricant, and possesses close-packed hexagonal lattice structure with low shearing strength [24]. Some studies reported that ZnS nanoparticles used as additive in lubricating oil reduced the friction coefficient and enhanced the wear resistance of metal-metal sliding pairs. For instance, Liu and Cheng et al. [25,26] found that addition of ZnS nanoparticles coated by diakydithiophosphate effectively enhanced the friction-reduction and antiwear abilities of oils. Wang et al. [27] investigated the tribological properties of surface-modified ZnS nanoparticles as an additive in polyethylene glycol. The results demonstrated that the addition of ZnS particles led to an obvious improvement of the tribological properties due to the formation of a tribofilm formed on the frictional surfaces. On the other hand, some researchers [28–30] reported that with the addition of ZnS particles into polymer-based composites, the friction coefficient and wear rate of the composites were further reduced under dry friction conditions. The authors attributed the improved performance to the load-carrying capacity and a possible "rolling effect" of the ZnS particles.

In response to the challenges posed by friction and wear of tribo-components exposed to boundary and mixed lubrication conditions, development of polymer-based self-lubricating composites attracts more and more interests. Nevertheless, till now, only few results on the tribological behaviors of PI composites under oil lubrication have been reported, especially tribological behaviors of polymer composites filled with hybrid functional fillers have been rarely investigated in published works. In the present study, series of PI-based materials filled with short carbon fibers (SCF), short glass fibers (SGF), sub-micrometer ZnS particles and their combinations were prepared and their tribological properties when exposed to poly-alpha-olefins (PAO) oil lubrication were investigated comprehensively. The main objective of this study was to investigate the hybrid effect of ZnS sub-micrometer particles with reinforcing fibers on the tribological properties and tribofilm formation of PI-based materials. The mechanisms of tribofilm formation were analyzed based on worn surface characterizations and tribo-chemical analyses. It is expected that this work will pave a route for formulating high-performance polymer-based composites for applications under oil lubrication conditions.

2. Experimental

2.1. Preparations of materials

PI (YS-20) powders (< 75 μm) were commercially obtained from Shanghai Synthetic Resin Institute (Shanghai, China) and used as matrix material. Polyacrylonitrile (PAN)-based SCF were supplied by Nantong Senyou Carbon Fiber Co. Ltd. (Jiangsu, China) with a length of 20–50 μm and a diameter of 7 μm. SGF were provided by Nanjing Fiberglass R&D Institute with a diameter of 10 μm and aspect ratio of 10:1. Representative Scanning Electron Microscope (SEM) graphs of SCF and SGF can be found in a previous paper [31]. ZnS sub-micrometer particles (Sachtolith HDS) were supplied by Sachtleben Chemie GmbH (Germany) with an average diameter of 300 nm. A typical SEM image of the ZnS particles is shown in Fig. 1.

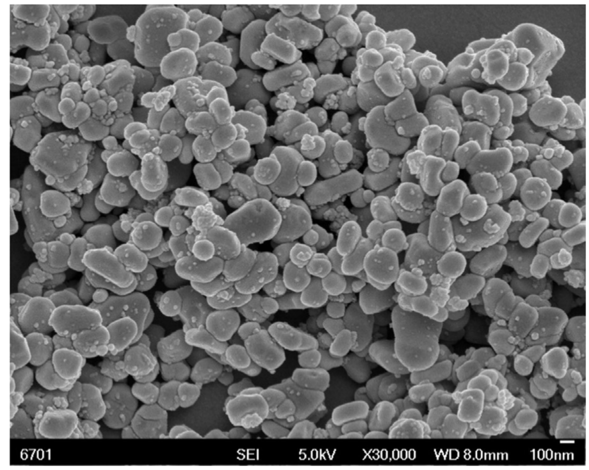

Fig. 1. SEM image of sub-micrometer ZnS particles.

Table 1
Compositions of PI-based materials (vol%).

Sample Abbreviation	Matrix	SCF	SGF	ZnS
PI	100	–	–	–
ZnS/PI	95	–	–	5
SCF/PI	90	10	–	–
SGF/PI	90	–	10	–
ZnS/SCF/PI	85	10	–	5
ZnS/SGF/PI	85	–	10	5

Four series of PI-based materials were investigated, i.e. pure PI, PI filled with ZnS, PI reinforced with short fibers (SCF or SGF) and PI filled with hybrid fillers (SCF or SGF combined with ZnS). The compositions and abbreviation of PI-based materials are listed in Table 1. Hot press molding technique was used to fabricate PI-based materials. Firstly, desired amount of fibers and ZnS particles were mixed with PI powders using a high speed mixer. And then the mixtures were compressed and heated up to 380 °C in a stainless steel mold with a dimension of $80 \times 80 \times 10$ mm^3. The pressure was held at 20 MPa for 60 min to allow a complete compression sintering. At last, the specimens were cooled under pressure to room temperature and cut into test samples with a dimension of $50 \times 10 \times 4$ mm^3 for testing (Fig. 2).

2.2. Tribology tests

Tribological tests were performed using a Plate-On-Ring (POR) apparatus (MRH-1A, Jinan Yihua, China). The schematic diagram of the POR contact configuration is shown in Fig. 2. The PI samples had a dimension of 50 10 4mm^3. The counterpart was a GCr15 (C 0.95–1.05 wt%, Mn 0.25–0.45, Si 0.15–0.35, $P \leq 0.025$, $S \leq 0.025$, Cr 1.40–1.65) steel ring with a diameter of 60 mm and a roughness, R_a of 0.1–0.2 μm. Before tribological tests, the steel ring and PI sample were ground by W20 SiC metallographic abrasive papers for getting the desired surface roughness and hereafter thoroughly cleaned in petroleum ether by ultrasonic cleaner. The roughness Ra of PI sample's original surface was around 0.14 μm. At first, load was applied on the PI samples before starting the experiment. And then the composite samples and steel counterparts were totally immersed in PAO4 base oil lubricant (provided by Mobile, China) (Fig. 2). The sliding time for each test was 3 h. The friction coefficient was recorded online by a force transducer during the entire sliding test. In practical applications, load and speed are important parameters influencing directly the hydrodynamic pressure of the