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Investigation of the running-in process of silicon nitride sliding in aqueous solutions of ethylene glycol



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ABSTRACT

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1. Introduction

In 1987, a very low friction coefficient (0.002) of silicon nitride against itself in water was discovered by Tomizawa and Fischer [1]. Since then, silicon nitride has gained attention due to its excellent tribological properties in an aqueous environment, such as low friction coefficient, high corrosion resistance and low critical sliding speed to achieve hydrodynamic lubrication [2–16]. Compared with mineral oil, water has unique advantages, such as environmentally friendly, low cost and nonflammable. Therefore, the combination of Si_3N_4 and water demonstrates promise in replacing the metal/oil system in bearings.

However, the load carrying ability of water is limited by its low viscosity and low pressure-viscosity coefficient [17,18]. To address this problem, different additives were added in water. Hartung et al. [19] added poly(L-lysine)-graft-poly(ethylene glycol) in water, and this brush-like polymer additive significantly reduced the friction coefficient of self-mated Si₃N₄ in the slow-speed regime (≤ 20 mm/s). And they found that a running-in process could improve enhance the effect even more. Li et al. [20] added phosphoric acid in water, and a low friction coefficient (0.004) between a glass/Si₃N₄ tribopair was obtained at high contact pressure (up to 1.65 GPa). Ma et al. [21] added glycerol and boric acid in water, and the friction pair (the Si₃N₄ ball sliding on the silicate glass) displayed a very low friction coefficient (0.0028) under a pressure of 300 MPa. Liu et al. [22] used fullerenol – a watersoluble nanoparticle – in water to increase the tribological properties of an Al₂O₃/Si₃N₄ tribopair, and the result showed that the load carrying

http://dx.doi.org/10.1016/j.triboint.2015.04.040 0301-679X/© 2015 Elsevier Ltd. All rights reserved. To investigate the tribological properties of self-mated silicon nitride in aqueous solutions of ethylene glycol, running-in tests in different solutions were conducted in a ball-on-disk apparatus. The results showed that the running-in performance depends on the concentration of ethylene glycol. When the concentration was greater than 10%, ethylene glycol could improve almost all of the tribological properties. However, when the concentration of ethylene glycol was around 5%, the most violent fluctuation of friction coefficient and the largest diameter of the wear scar was observed. The results suggest that the high concentration ethylene glycol solutions have great potential to be used in self-mated silicon nitride sliding bearings.

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ability of water film was increased. Nalam et al. [23] investigated the tribological properties of surfaces with PLL-g-PEG sliding in aqueous of glycerol or ethylene glycol solutions under different lubrication regimes, and they found that the combination of polymer layers on surfaces with aqueous phases of enhanced viscosity greatly reduced the friction. Shi et al. [24] studied the boundary and elastohydrody-namic lubrication of glycerol aqueous solutions, and their results showed that glycerol aqueous solutions have great potential to replace rapeseed oils. Yan et al. [25] added different additives in water, and the tribological properties of self-mated Si₃N₄ in the "green water-based lubricant" were tested.

Besides the above water-based lubricants, the water-glycol solution is a widely used water-based hydraulic fluid in industry due to its wonderful properties, such us low-temperature performance, good resistance to oxidation and good wear resistance [26,27]. The tribological properties of Si₃N₄ in alcohols and their aqueous solutions have been studied by researchers [23,28–34]. Hibi and Enomoto [28-30] systematically investigated the friction and wear characteristics of Si₃N₄ in linear alcohols or normal alcohols, and they found that the friction and wear of Si₃N₄ were influenced by the reactivity of the mechanochemical reaction producing silicon alkoxide. Wei and Xue [31] compared the friction and wear properties of Si₃N₄ in water and ethanol, and Si₃N₄ displayed lower friction coefficient and worn volume in ethanol. Qu et al. [32] investigated the tribological properties of Si₃N₄ in ethylene glycol aqueous solutions using a reciprocating pin-on-block tester. And they found that the wear volume decreased with increase in the concentration of ethylene glycol. Gates and Hsu [33] studied the lubrication mechanism of Si₃N₄ in alcohols under boundary lubrication conditions, and the formation of silicon alkoxides was revealed by dynamic wear tests and static chemical reaction studies. Zhang et al. [34] investigated the

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friction and wear behaviors of (Ca, Mg)-sialon/SAE 52100 steel pair in aqueous solutions of propanetriol, and the results showed that the highest wear volume occured when the concentration of propanetriol was 10%.

From the above results, the concentration of ethylene glycol plays an essential role when Si_3N_4 sliding in water-glycol solutions. In this investigation, the running-in tests were conducted to show the influence of concentration of ethylene glycol on the tribological properties of Si_3N_4 .

2. Materials and methods

2.1. Materials

The water used in the experiments was filtered and deionized. The ethylene glycol used in the experiments was a commercial product (Jiangtian Chemical Company, Tianjin, China) with a purity of greater than 99%, and different volume percentages of ethylene glycol in water (0, 5, 10, 20, 30, 40, 50, 70 and 100% v/v) were used. Dynamic viscosities and pressure-viscosity coefficient values of the mixtures according to their volume fractions are shown in Table 1 [17,26,27]. The material properties of Si₃N₄ balls and discs are presented in Table 2. The diameter of Si₃N₄ balls was 9.525 mm.

2.2. Experimental procedures

The unidirectional sliding running-in test is a simple and effective method to investigate the tribological properties of tribopairs [2,3,5,35,36]. In this study, running-in tests of Si_3N_4 in different aqueous solutions of ethylene glycol were conducted in a MMW-1 type ball-on-disc tribometer (Puye Mechanical & Electrical Technology Co., Ltd., China).

Before each test, the ball and disk were cleaned with ethanol and acetone for 15 min each in an ultrasonic bath, and then washed thoroughly with the deionized water. During the test, the ball and disc were submerged in aqueous solutions of ethylene glycol, and the temperature of solutions was controlled at 30 ± 1 °C. The normal load was 10 N, which could generate a maximum contact pressure of 1.3 GPa according to the Hertz contact theory. The sliding speed was 0.5 m/s, and the test time was 3600 s, corresponding to a sliding distance of 1800 m. The friction coefficient was recorded throughout the experiments. After each test, worn surface of the ball was observed by optical microscopy and SEM. The wear scar diameter on the ball was measured by optical microscopy. Then the wear volume was calculated according to the ASTM-G 99 Standard. All running-in tests were repeated four times under the same conditions.

Table 1

Dynamic viscosities and pressure-viscosity coefficient values for different percentages of ethylene glycol in water at 30 $^\circ\text{C}.$

Concentration of ethylene glycol in water (vol%)	Dynamic viscosity at 30 °C (mPa s)	Pressure-viscosity coefficient $(\times 10^{-9} \text{ m}^2 \text{ N}^{-1})$
0	0.8	0.75
5	0.89	0.76
10	0.97	0.78
20	1.3	0.83
30	1.69	1.05
40	2.26	1.35
50	2.94	1.85
70	5.23	3
100	13.76	9.6

3. Results and discussion

3.1. Friction coefficient

The friction coefficient during the whole test has been plotted as a function of time for different ethylene glycol solutions (Fig. 1). As shown in Fig. 1, the friction coefficient for all ethylene glycol solutions showed a sharp rise to a maximum value in the initial period (about 15 s). After different running-in time, the friction coefficient curves tend to a steady state at about 0.02. It must be noted that when the concentration of ethylene glycol was 5%, the fluctuations in friction coefficient were observed for longer durations at 0.07 (Fig. 1a). Meanwhile, similar fluctuations could be observed when the concentration of ethylene glycol was 10%. However, this kind of fluctuation could not be found in pure water and in high concentration solutions. The fluctuations of the 5% and 10% ethylene glycol solutions are classic symptoms of incomplete film formation on the tribopair surface, which known as the slipstick phenomenon. This is very typical when the system is in a lubricant or additive starvation regime.

The maximum value of friction coefficient during each test has been shown in Fig. 2. The maximum value of friction coefficient was observed to decrease with increasing concentration of ethylene glycol from 0.8 (pure water) to 0.1 (pure ethylene glycol).

Table 2	
Material	properties

Material Si ₃ N ₄ ball Si ₃ N ₄ disc Vickers hardness (H_{ν}) 1580 1420 Fracture toughness (MPa m ^{1/2}) 6.0 6.18 Flexural strength (MPa) 900 791 Density (g/cm ³) 3.25 3.27 Initial roughness Ra (µm) 0.014 0.020			
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$111111 \text{ roughness } Ra (\mu m) \qquad 0.014 \qquad 0.020$	Vickers hardness (H_{ν}) Fracture toughness (MPa m ^{1/2}) Flexural strength (MPa) Density (g/cm ³)	1580 6.0 900 3.25 0.014	1420 6.18 791 3.27
	Initial roughness <i>ka</i> (µm)	0.014	0.020



Fig. 1. Friction coefficient as a function of test time for different ethylene glycol aqueous solutions (10 N, 0.5 m/s, 3600 s).

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