Gradual degeneration of liquid superlubricity: Transition from superlubricity to ordinary lubrication, and lubrication failure

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ABSTRACT

The step-by-step evolution of the friction coefficient and substrate damage during the superlubricity degeneration of Si_3N_4/glass pair lubricated with H_3PO_4 solution was investigated. The experimental results indicate that the degeneration process experiences two stages after superlubricity failure. The superlubricity state initially degenerates to the ordinary lubrication state, gradually destabilizes, and eventually fails. Further analysis demonstrates that superlubricity degeneration is mainly attributed to serious substrate damage; however, the subsequent lubrication failure is mainly caused by the insufficient amount of lubricant. Moreover, load, not speed, plays a pivotal role in the wear behavior of substrate surface and further affects the superlubricity stability. The results may reveal the degeneration mechanism of liquid superlubricity and optimize the operation and design of superlubricant systems.

1. Introduction

More than 33% of the energy consumption and 60% of machine failure worldwide are attributed to the severe friction and wear issues, thus, friction and wear reduction will greatly improve energy efficiency and create massive economic benefits [1]. The superlubricity phenomenon has attracted considerable attention since it was first defined by Hirano in the early 1990s due to the enormous demand of the mechanical industry for improved lubricity, energy savings, wear protection, and emission reduction [2,3]. The applications of superlubricity state, in which the friction coefficient at tribological interface is less than 0.01, is one of the most effective approaches for managing the issues in machine failure and energy consumption [4–6].

Over the last two decades, drastic progress and achievements in superlubricant systems have been obtained, particularly in the development of solid and liquid superlubricants. Solid superlubricants mainly contain molybdenum disulfide [7,8], diamond-like carbon film [9,10], graphene [11,12], and graphite [13,14] due to the incommensurate contact or the interlayer with weak shear strength induced by tribochemical reactions [15–17]. Liquid superlubricants mainly contain ceramic materials under water [18], molecular 'brush' [19,20], mixed solution of polyol and acid [21,22], and polysaccharide slime extracted from plants [23]. Among them, water-based superlubricants are one of the optimal choices given their low-cost, high

reproducibility, and environmental friendliness [24]. Nevertheless, superlubricant systems realized in laboratory environments are difficult to apply in actual products because these systems require strict working conditions, such as limited load capacity.

Recently, it is reported that Si_3N_4/glass interfaces lubricated with acids in aqueous solutions or mixed with polyol solutions can achieve and maintain an ultralow friction coefficient of 0.004 for more than 10 h [21,22]. For example, the superlubricity mechanism of H_3PO_4 solution (pH = 1.5) is mainly ascribed to the formation of a superlubricating film at tribological interfaces. This film can separate rubbing surfaces effectively, provide a composite hydration layer with ultralow shear strength, and prevent direct tribological contact [22]. Most related studies have focused on achieving superlubricity. However, information on the failure behavior of superlubricity state remains limited. Previously, our group reported that the surface damage of substrate considerably influences the stability of liquid lubrication, for example, probable high flash temperature at running-in process under high speed may destroy the substrate surface and result in lubrication failure [25]. Under low load, superlubricant systems exhibit excellent stability even at high speed as long as the superlubricity is realized after a gentle running-in period [25]. Thus, the degeneration process of superlubricity state has not been observed. The surface damage of the tribopair increases drastically with increased load [26], which may provide the opportunity to study the evolution process of superlubricity

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degeneration at specific substrate conditions. Further understanding of the load capacity and dependence of superlubricity stability will promote the practical application of liquid superlubricants and help develop novel superlubricant systems. Therefore, revealing the mechanism that underlies superlubricity degeneration is essential for improving the operation reliability and lifetime of superlubricant systems.

In this study, the superlubricity stability of Si_3N_4/glass pair with H_3PO_4 solution under various applied loads was investigated. The close relationship between the lubricating performances and wear behaviors of glass substrates was discussed. Specifically, the characteristics of every step of the superlubricity generation process were observed. Further analysis demonstrated that superlubricity degeneration is mainly contributed by serious substrate damage. However, subsequent lubrication failure is mainly caused by the insufficient amount of lubricant. Based on these results, the load capacity of the liquid superlubricity system is improved by enhancing the mechanical properties of the substrate surface. The results may help understand the mechanism that underlies the degeneration of liquid superlubricity and optimize the operation and design of superlubricant systems.

2. Materials and methods

H_3PO_4 solution with a mass fraction of about 0.3 wt% (pH = 1.5) was used as the superlubricant in all frictional tests, which was diluted by the concentrated solution purchased from Sinopharm Chemical Reagent Co., Ltd. The upper samples were Si_3N_4 ball with a radius of 2 mm and the substrates were common glass slides with the root-mean-square (RMS) roughness of approximately 0.05 μm over an area of 100 μm × 100 μm as measured by a white-light interferometer (MFT-3000, Rtec, USA). To remove surface contamination on the tribopair surfaces which may affect the superlubricity behavior, experimental samples were ultrasonically cleaned sequentially in acetone and ethanol solutions, finally rinsed by plenty of deionized water and dry nitrogen gas.

All the friction tests were performed by a universal micro-tribotester (UMT-V, Bruker, USA) with a ball-on-disk rotation mode. 10 μL of H_3PO_4 solution was added at the Si_3N_4/glass interface before started the equipment. In accordance with Hertzian contact theory, normal load was applied over the range of 1–15 N to provide the maximum contact pressure of 0.55–1.44 GPa [27]. The rotating speed was varied in the range of 36–3600 rpm with a track radius of 4 mm, and the corresponding liner speed was 0.15–1.5 m/s. During the whole tests, the temperature was controlled at 22 ± 2 °C and the relative humidity was approximately 30%. To ensure that friction was measured accurately, measurement errors were reduced through pre-commission. Details about that process can be found in previous literature [28]. Using white-light interferometer, the topographies and RMS roughness of the wear tracks on tribopair surfaces were measured. The elemental characterizations of severely damaged area in the lubrication failure state and original area on glass substrates were conducted by energy disperse spectroscopy (EDX, SEM, ZEISS, EVO18).

3. Experimental results

3.1. Gradual degeneration of liquid superlubricity with H_3PO_4 solution

Load strongly affects friction and wear behaviors at nearly all tribological interfaces, and the load capacity is the key parameter used to select the appropriate lubrication system for actual industrial production [26,29]. Fig. 1 displays the friction coefficient evolution of Si_3N_4/glass pair with H_3PO_4 solution under six different loads. When the load is in the range of 1–5 N, the superlubricity state is achieved after a running-in period over 200s, and remains stable through the entirety of the experimental period. Moreover, the friction coefficient further decreases from 0.007 to 0.004 and the sliding time of running-in period
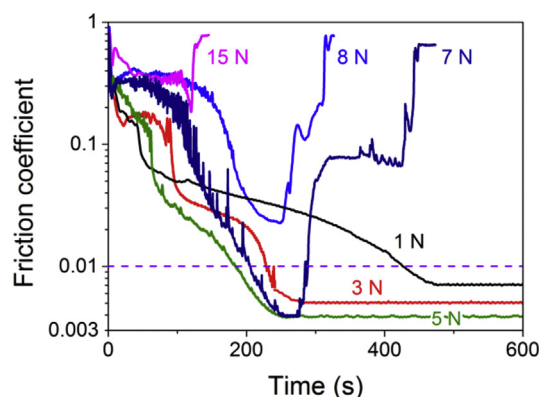

Fig. 1. Friction coefficient of Si_3N_4/glass pair lubricated with H_3PO_4 solution under various loads. The speed is 0.15 m/s.

also decreases as load increases to 5 N. It is found that the load capacity of the superlubricity system is approximately 7 N, and there exists a transition period (under the loads of 5–8 N) between superlubricity and lubrication failure, which is different from the results of speed effect [25]. Under the load of 7 N, the superlubricity state is realized after ~250s of the running-in period but it only maintains for ~50s, and then fails until the end of the test. When the load is further increased to 8 N, the friction coefficient decreases to 0.03 after running-in period and then increases sharply to 0.8. The temporal evolution of the frictional behavior under the load of 15 N is similar to that under 8 N, however, the running time is shorter and the lowest friction coefficient achieved is larger. Moreover, the load capacity of about 7 N is measured under above given operating environment. Such load capacity will decrease with increasing speed [25], which may be mainly attributed to the stronger impact force at higher speed.

These friction results indicate that the lubrication states of Si_3N_4/glass pair with H_3PO_4 solution strongly depends on the applied load, and the superlubricity state is unrealizable when loads exceed 7 N. Fig. 2 shows that the friction coefficient as a function of load initially decreases, reaches a minimum value, and then increases. The transition point for superlubricity failure is approximately 7 N, and the corresponding initial contact pressure is ~1.1 GPa. When the load exceeds 7 N, the lubrication failure (μ > 0.1) occurs after a short unstable ordinary lubrication period (0.01 < μ < 0.1). In this case, the superlubricity state is unprocurable and even the ordinary lubrication state failed quickly. Notably, the transition period under the load of 5–8 N completely exhibits the whole process of superlubricity to failure, which may provide a perfect opportunity to further understand the superlubricity failure.
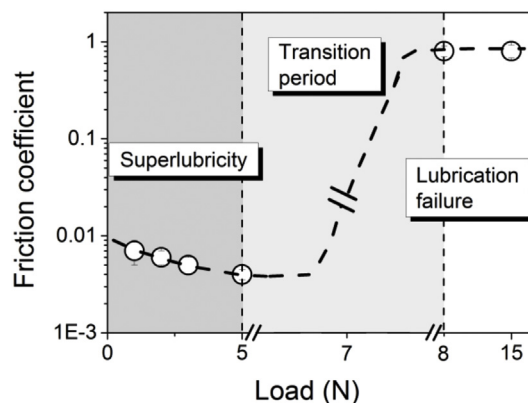

Fig. 2. Comparison of the final friction coefficients of Si_3N_4/glass pair with loads after running 600s.