

Analysis on mechanism of thin film lubrication

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Abstract It is an important concern to explore the properties and principles of lubrication at nano or molecular scale. For a long time, measurement apparatus for film thickness of thin film lubrication (TFL) at nano scale have been devised on the basis of superthin interferometry technique. Many experiments were carried out to study the lubrication principles of TFL by taking advantages of aforementioned techniques, in an attempt to unveil the mechanism of TFL. Comprehensive experiments were conducted to explore the distinctive characteristics of TFL. Results show that TFL is a distinctive lubrication state other than any known lubrication ones, and serves as a bridge between elastohydrodynamic lubrication (EHL) and boundary lubrication (BL). Two main influence factors of TFL are the solid surface effects and the molecular properties of the lubricant, whose combination effects result in alignment of liquid molecules near the solid surfaces and subsequently lubrication with ordered film emerged. Results of theoretical analysis considering microstructure are consistent with experimental outcomes, thus validating the proposed mechanism.

Keywords: thin film lubrication, super thin interferometry, lubrication property, ordered film, microstructure.

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It is a main concern to interpose a lubrication film of low shear strength between two solid surfaces in relative motion intending to efficiently reduce friction and wear and to prevent seizure. There is a veiled regime in lubrication theories: film thicknesses of elastohydrodynamic lubrication (EHL) are typically of the order of 0.1–1 μm , while film thicknesses of boundary lubrication (BL) are usually of the order of several nano meters^[1]. Thus the unknown span of film thickness constitutes the study object of TFL investigation. Furthermore, it is widely accepted that EHL is featured with viscous fluid films, in addition to piezo-viscous effect and solid elastic deformation, and it is based upon continuum mechanism. Boundary lubrication, however, featured with adsorption films, either due to physisorption or chemisorption, is based on surface physical/chemical properties^[2]. It will be of great importance to bridging the gap between EHL and BL in aspects of work mechanism and study methods, by taking

TFL as a research intention.

Essentially, TFL is ascribed to a lubrication regime where the liquid film thickness is of the order of nano meters or molecules. In literature, super or partial or molecular thin film lubrication is used to describe this new lubrication featured with sub-micron or nanometer scale film thicknesses^[3,4]. However, the term ‘thin film lubrication’ comes into being dominant, for instance, Wen^[5] and Tichy^[6]. To date, it is widely accepted that TFL is a distinctive lubrication state, bridging the gap between EHL and BL, which has its own distinctive lubrication traits. With the lubricant film thickness stepping down, lubricating state will undergo following changes: hydrodynamic lubrication \rightarrow elasto-hydrodynamic lubrication \rightarrow thin film lubrication \rightarrow boundary lubrication \rightarrow dry friction. Recently, new lubrication map was proposed accordingly^[7], as shown in Fig. 1, where h is the lubrication film thickness, R_a the combined roughness of the two surfaces in relative motion, and R_g the effective radius of lubricant molecules, respectively. TFL bridges the gap between EHL and BL.

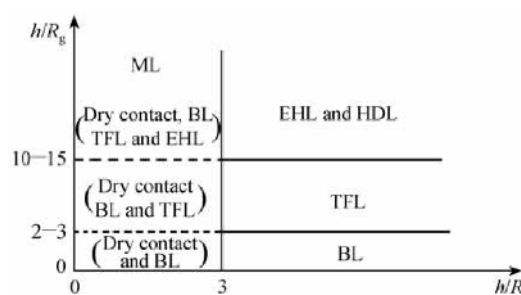


Fig. 1. Lubrication map^[7].

In the 1990s of last century, researchers extended conventional optical interference technique so as to provide measurement ability of film thickness down to a few nanometers or molecular scale. With the help of these methods, profound experiments were carried out to investigate the specific features of TFL, some comprehensive references were summarized by the first author^[8]. At present, extensive investigations have been conducted to experimentally study the properties of TFL, however, the other side of the same coin is, it is a far cry to obtain mathematically feasible predictive model of TFL, leaving aside the ordered model has been put forward for a long time. It is the main purpose to give a preliminary analysis on mechanism of TFL on the basis of previous works.

1 Superthin interferometry makes it possible to study properties of TFL

A key problem to investigate the characteristics of TFL is to devise an available rig in favor of measuring film thickness purpose. The family of techniques based on optical interferometry is one of the most important contributors to experimental research. Conventional optical interferometry has two major limitations that prevent it being

able to measure very thin films. One is that, for optical interference to occur, the optical path difference must be at least half a wavelength of light, which means that it is not practical to measure films less than about one-quarter of wavelength of light thick, i.e., about 50 nm. The second is an inherent inability of the human eye accurately to distinguish interference colors, which means that measurement resolution is limited to about ± 10 nm. The limitations of optical interferometry give impetus to further extensions so as to enable it capable of measuring lubricant film thickness falling down to a few nanometers. In 1991, Johnston et al.^[9] used a method of spacer layer in combination with the spectrum analysis of reflective light to expand the optical interference technique, enabling it available to measure thin film thickness down to nano scale. Soon later, in 1999, Hartl and co-workers^[10–12] measured film thickness down to 5 nm and further expressed the film thickness distribution as intuitionistic image by applying colorimetric interferometry technique. Recently, interference method with the help of multi-beam technique also reported the measured film thickness value down to about 1 nm^[13,14].

Luo and co-workers affiliated to Tsinghua University devised NGY-3 film thickness measurement rig (Fig. 2) of TFL, by making use of a technique of relative optical intensity interference^[15] (ROII). The apparatus can provide dynamic measurement of film thickness of the order of nano meter with stable precision. The main qualifications of the system are: measure range: 0–500 nm, vertical resolution: 0.5 nm, horizontal resolution: 1 μm , velocity range: 0.2–1900 mm/s, friction force resolution: 0.1 mN,

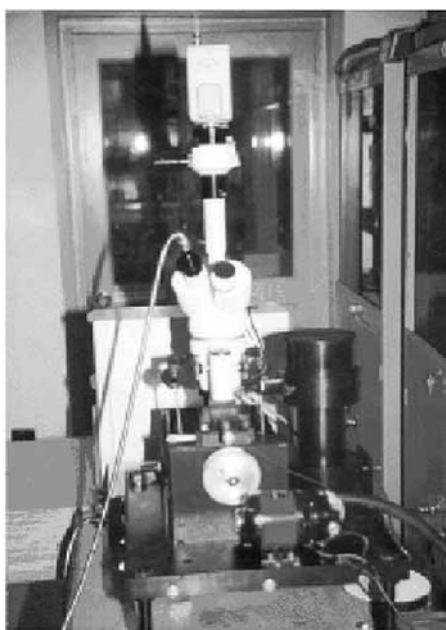


Fig. 2. Measure apparatus for lubrication film thickness at nano scale (NGY-3).

temperature control range: room-temperature $\sim 120^\circ\text{C}$, etc. These successes laid the foundation of investigating properties and lubrication principles of TFL.

These experimental methods and techniques and their utilization have brought out fruitful outcomes, which provide some valuable data of properties and mechanism of TFL, and paved a feasible way for lubrication and tribo-system at nano scale.

2 TFL is a transition state between EHL and BL

Size effects can be seen in TFL and is the deviation of TFL from EHL^[16], i.e., the film thickness vs. velocity, viscosity, pressure etc. relations are no longer linear ones in log-log coordinates. Fig. 3 is a schematic view of the typical test curve from experimental data, wherein the axes are logarithm coordinates. Three regions can be seen from the figure: thick film region (section I), thin film region (section II), and failure region (section III). It can be seen that, the film thickness varies linearly with velocity (or viscosity and pressure) in thick film region. This is the EHL regime, where film thickness variation complies with EHL theory predictions. While in thin film region, with the decreasing of film thickness, film thickness curve levels out progressively, differing from that of EHL predictions. The thinner the film, the more the difference. With further decreasing film thickness, either due to the shear limit of lubricant is arrived, or due to the solid surface cease to sustain an available absorption, the lubricant can no longer hold mobility and bear loads. This heralds a failure region.

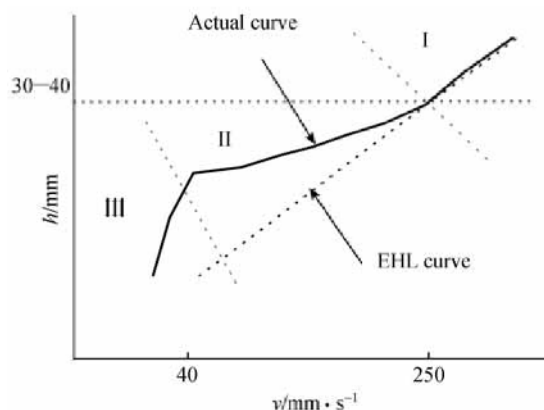


Fig. 3. Schematic view of various regimes of film thickness (Lubricant: $\text{C}_{16}\text{H}_{34}$; Pressure: 185 MPa).

The failure of TFL only means a mobility loss here. In fact, monolayers can stay on solid surfaces to separate the solid surfaces in relative motion, and subsequently sustain a feasible boundary lubrication^[17]. Because film thickness of TFL is on the nano scale or molecular order, from mechanic point of view, in terms of the mobility, TFL can be regarded as the last one of lubrication fields where Reynolds equation is valid.

Boundary lubrication can also evolve into TFL^[18].

Compared to BL, TFL has a thicker film. In the vicinity of solid walls, the liquid molecules take the states of that of the boundary film, the ordered one, and the disordered one, from the wall surfaces to the center of the gap. From a mechanic point of view, the existence of an ordered film makes lubricant film differs strongly from the boundary film, which can form a glassy state or solid-like state^[19]. On the other hand, lubricant of TFL can only sustain some viscosity, expressing mobility to some extent.

3 Study on film-forming of TFL

The property of TFL is strongly associated with the property of solid wall surfaces (Fig. 4). To investigate the influences of solid surface energy, experiments were carried out with four types of materials of various solid surface energy. From the thickness difference among different friction surfaces, it can be seen that the film with TiO₂, which has the highest surface energy, is over than that with any other surfaces. The films with Ti and Cr, whose surface energy are nearly of the same, are nearly of the same thickness. The film with Al, whose surface energy is the least, holds the smallest thickness. It can be deduced that the higher the surface energy is, consequently the higher the surface tension is, the thicker the film and the critical values are^[20,21].

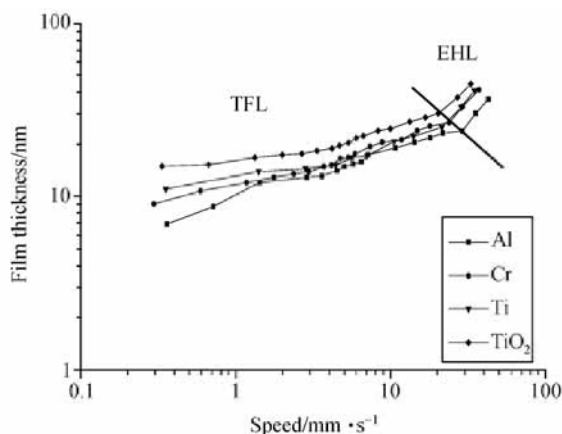


Fig. 4. Film thickness of TFL with solid surface energy^[20]. Lubricant: 13604; Load: 4 N, $T = 25^{\circ}\text{C}$.

The property of TFL is linked closely to the lubricant properties. Fig. 5 shows experimental results of hexadecane with various liquid crystal additives. The abscissa is film thickness while the ordinate is effective viscosity. It tells that the effective viscosities are almost the same while film thickness is high (EHL regime). However, in TFL regime, effective viscosities vary with film thickness, and the variations are overt. This clearly shows that, even though the apparent viscosities (i.e., the bulk viscosity) are almost same in TFL, film forming characteristics are different, indicating a different mechanism of TFL.

The above-mentioned experiments show two main influential factors of TFL: the solid surface effects and the

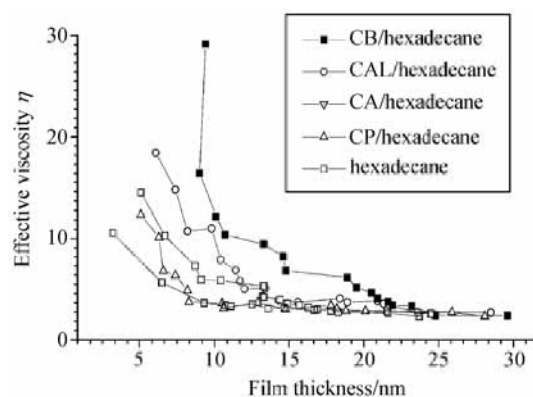


Fig. 5. Effective viscosity vs. film thickness relation^[22].

properties of liquid molecules. As mentioned before, EHL is featured with viscous fluid films. As to a particular lube system, if any other conditions are same, the film characteristics of the lube system will be same for different solid surface and different lubricant (same viscosity, viscosity-pressure relation and viscosity-temperature relation are assumed). In TFL, due to surface adsorption potentials and/or shear induction, the orientation of liquid molecules tends to being ordered from each other, and conclusively resulting in different characteristics. Luo et al.^[23] concluded that TFL is essentially dominated by the ordered film.

4 Ordered model of TFL (analysis on physical model)

In TFL, because the lubricant film is confined in narrow gap separating the solid surfaces in relative motion of tribo system, the molecules of lubricant film tend to be structurally ordered in friction shear process, due to the load and surface energy. Then lubrication film at the sub-micro or nano scales will comprise three types of film with different structures and properties: i.e. the adsorbed film, the ordered film, and the fluid film (Fig. 6). The ordered liquid film bordering between the viscous fluid film and the adsorbed film is thus formed that the liquid molecules undergoes surface forces during friction and shear process. From the viscous fluid film to the adsorbed film, the ordering degree increases gradually. This orientation ordered molecular film is called ordered liquid film, which has a better ordering degree than that of the bulk fluids, and thus subsequently being less mobile. However, the other side of the same coin is, the ordered liquid film has a mobility to some extent, thereby it can bear loads and lead to a decrease of side leakage due to hydrodynamic effects. In TFL, the ordering degree of lubricant molecules decreases from the solid wall to a far distance, which results in different responses to lubrication behavior and size-dependent effects.

Experimental results show that the transition from EHL to TFL is closely linked to the rearrangement structure of liquid molecules and transition conditions are also related

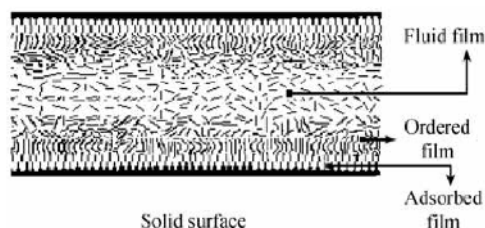


Fig. 6. Ordered liquid film model for TFL^[7].

to the property of the liquid as well as the film thickness. The critical point of film thickness varies with liquid types. TFL is featured with ordered liquid film, wherein the orientation of the molecules are aligned. The thickness of the ordered film is closely associated to the interface adhesive energy and its work range. The relation between the interface adhesive energy and liquid molecular structure, molecular weight, and environment temperature is similar to that of the effective viscosity.

5 Verification of the ordered film model

TFL is essentially a lubrication regime where the ordered film takes a dominant role. Even though it is very difficult to give an in situ measurement of this ordering state and up to date there is hardly report on the observation of it during real TFL process, the ordered property of liquid molecules has drawn more and more broad concerns. Ehara et al.^[24] showed that molecular alignment will take place if the molecules are adsorbed onto a solid surface with an appropriate surface structure. Their investigation shows that the surface with high energy can make the liquid molecules ordered. For low energy surfaces, if it undergoes a macroscopic processing such as mechanical shear brushing, molecular alignment will also take place, which means that the shear induction effects account for the molecular alignment. The lyotropic liquid crystalline can also express this characteristic^[25]. These provide some indirect verifications of the ordered liquid film model.

It should be point out that, the ordered orientation of liquid molecules is prevailing in BL^[17], whilst they usually cease to be mobile. However, the ordered film of TFL has the characteristic of a liquid, and thus sustains mobility.

6 Theoretical analysis on the basis of the ordered model

Theoretical analysis on TFL properties lags far behind experimental investigations, partly because it has close relation to the solid surface adsorption potentials and the property of the lubricants, which obviously contribute to the complexity of the problem. Zhang et al.^[26–30] focused their endeavors on mathematical modeling and theoretical prediction of the lubrication features of TFL.

The orientation of the ordered film has the following characteristics^[26]:

- (1) Molecular weight center does not have long range

order. It lies in surface force field. The surface force usually does not work long away from the solid walls.

- (2) Molecular orientation is ordered, and tends to be parallel to someone axis, which is named “easy axis” and expressed by a unit vector, the director, e .

- (3) The direction of the easy axis in space is arbitrarily distributed, which can be influenced by the solid surface forces.

- (4) The directions e and $-e$ are indiscernible.

- (5) The director is supposedly fixed on solid surface due to the surface potentials.

The ordered molecules are apparently similar to that of nematics. Velocity field of TFL can be analyzed with the help of liquid crystal theories accordingly (Fig. 7).

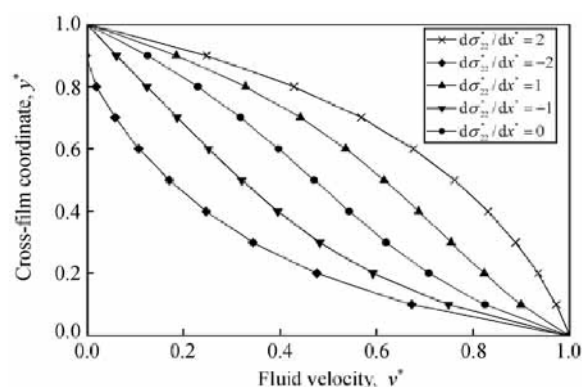


Fig. 7. Velocity distribution under different normal pressure gradient.

Because lubrication film of TFL is very thin, approaching the length scale of the particles of lubricants, the size of the particles will contribute much to lubrication property. The force and moment transition of lubricant layers confined by particular boundary conditions should be considered. And the microstructure and the corresponding deformation should also be taken into account. For instance, three types of deformation can occur in nematics: splay, twist, and bend. By reducing the problem complexity, Zhang et al.^[27,28,31] supposed that the lubricant particles are rigid and randomly ordered in viscous media, and performed numerical simulation of lubrication features of TFL by using micropolar theory. Fig. 8, very similar to Fig. 5, shows the relation between the effective viscosity and the minimum film thickness, taking into account of the micropolar effects (with different characteristic lengths and couple numbers).

These investigations are useful for quantitatively modeling the lubrication features of TFL. The results indicate that the ordered state of liquid molecules have an important influence on the film forming characteristics and lubrication properties. It is a far cry to herald that it is perfect, however. Further exploration on the basis of the ordered film model will lay the foundation for the mechanism of TFL.

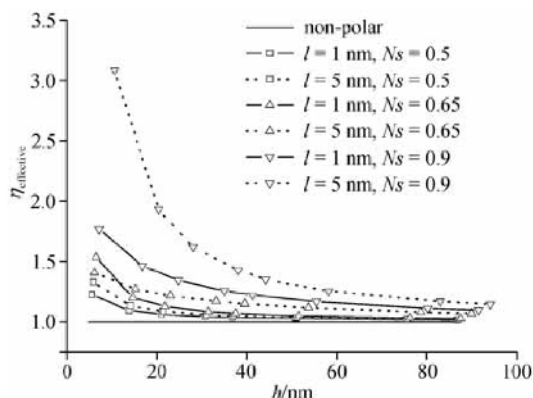


Fig. 8. Effective viscosity vs. minimum film thickness relation (theoretical analysis)^[28].

7 Conclusions

TFL is an important branch of nano tribology. TFL in ultra thin clearance exists extensively in micro/nano components, integrated circuit (IC), micro-electromechanical system (MEMS), and computer hard disks, etc. The impressive developments of these techniques present a rosy future of TFL with an ordered structure at nano scale.

The mechanism of TFL can be summarized to the ordered film lubrication, concluding from preceding researches. It should be noted that the property and mechanism of TFL are not fully understood to date, and there remains a wide blank area to be covered between experiments and theoretical predictions. Some key problems to be addressed are

- 1) measurement technique on lubrication film distribution;
- 2) the relation of the effective viscosity and the property of the lubricant molecules;
- 3) influence factors relating to transition between the liquid phase and the solid-like phase;
- 4) further perfection of the mathematical model based on the ordered film;
- 5) the property of electro-rheology, magneto-rheology, intelligibility and controllability, etc of ordered liquids and so on.

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References

1. Wen, S. Z., From EHL to TFL theory—a new research field of lubrication theory, *Lubrication Engineering* (in Chinese), 1993, 48–55.
2. Luo, J. B., Lu, X. C., Wen, S. Z., Progress and problems of thin film lubrication, *Progress in Natural Science*, 2000, 1057–1065.
3. Tadmor, R., Chen, N. H., Israelachvili, J. N., Thin film rheology and lubricity of hyaluronic acid solutions at a normal physiological concentration, *J. Biomedical Materials Research*, 2002, 61(4): 514–523.
4. Homola, A. M., Israelachvili, J. N., Gee, M. L. et al., Measurements of and relation between the adhesion and friction of two surfaces separated by molecularly thin liquid films, *ASME J. Tribol.*, 1989, 111(3): 675–682.
5. Wen, S. Z., On thin film lubrication, in *Proc. 1st Int. Symp. Tribol.*, Beijing: Tsinghua University Press, 1993, 30–37.
6. Tichy, J. A., Thin film lubrication, in *Proc. 1st Int. Symp. Tribol.*, Beijing: Tsinghua University Press, 1993, 48–57.
7. Luo, J. B., Shen, M. W., Shi, B. et al., Thin film lubrication and lubrication map, *Chinese J. Mechanical Engineering* (in Chinese), 2000, 36(7): 15–21.
8. Zhang, C. H., Research on thin film lubrication: State of the art, *Tribol. Int.*, 2005, 38(4): 443–448.
9. Johnston, G. J., Wayte, R., Spikes, H. A., The measurement and study of very thin lubricant films in concentrated contacts, *Tribol. Trans.*, 1991, 34(2): 187–194.
10. Hartl, M., Krupka, I., Poliscuk, R. et al., An automatic system for real-time evaluation of EHD film thickness and shape based on the colorimetric interferometry, *Tribol. Trans.*, 1999, 42(2): 303–309.
11. Krupka, I., Hartl, M., Poliscuk, R. et al., Experimental study of central and minimum elastohydrodynamic film thickness by colorimetric interferometry technique, *Tribol. Trans.*, 2000, 43(4): 611–618.
12. Hartl, M., Krupka, I., Poliscuk, R. et al., Thin film colorimetric interferometry, *Tribol. Trans.*, 2001, 44(2): 270–276.
13. Guo, F., Wong, P. L., A multi-beam intensity-based approach for lubricant film measurements in non-conformal contacts, *Proc. Inst. Mechanical Engineers Part J-J. Engineering Tribol.*, 2002, 216(J5): 281–291.
14. Glovnea, R. P., Forrest, A. K. et al., Measurement of sub-nanometer lubricant films using ultra-thin film interferometry, *Tribol. Lett.*, 2003, 15: 217–230.
15. Luo, J. B., Wen, S. Z., Huang, P., Thin film lubrication, Part I: The transition between EHL and thin film lubrication, *Wear*, 1996, 194: 107–115.
16. Zhang, C. H., Numerical Analysis on Tribological Performances of Lubricating Film in the Nano Scale (in Chinese), Beijing: Tsinghua University, 2002.
17. Dorinson, A., Ludema, K. C., *Mechanics and Chemistry in Lubrication*, Berlin: Elsevier, 1985.
18. Glovnea, R. P., Spikes, H. A., Elastohydrodynamic film formation at the start-up of the motion, *Proc. Instn. Mech. Engrs. (Part J)*, 2001, 215: 125–138.
19. Homola, A. M., Israelachvili, J. N., Gee, M. L. et al., Measurements of and relation between the adhesion and friction of two surfaces separated by molecularly thin liquid films, *ASME, J. Tribol.*, 1989, 111(3): 675–682.
20. Luo, J. B., Wen, S. Z., Sheng, X. Y. et al., Substrate surface energy effects on liquid lubricant film at nanometer scale, *Lubrication Science*, 1998, 10(11): 23–35.
21. Shen, M. W., Luo, J. B., Wen, S. Z., Influence of friction pair surface's physicochemical properties on nano scale film thickness, *J. Tsinghua University (in Chinese)*, 2000, 40(4): 103–106.
22. Shen, M. W., Luo, J. B., Wen, S. Z. et al., Investigation of the liquid crystal additive's influence on film formation in nano scale, *Lubrication Engineering*, 2002, 58(3): 18–23.
23. Luo, J. B., Huang, P., Wen, S. Z. et al., Characteristics of fluid lubricant films at the nano-scale, *ASME, J. Tribol.*, 1999, 121(4): 872–878.
24. Ehara, T., Hirose, H., Kobayashi, H. et al., Molecular alignment in organic thin films, *Synthetic Metals*, 2000, 109: 43–46.
25. Nesrullajev, A., Tepe, M., Kazanci, N. et al., Surface-induced textures in lyotropic liquid crystalline mesophases, *Materials Chemistry and Physics*, 2000, 65: 125–129.
26. Zhang, C. H., Wen, S. Z., Luo, J. B., Lubrication theory for thin film lubrication accounting for the ordered film model, *Int. J. Nonlinear Sciences and Numerical Simulation*, 2002, 3(3-4): 481–485.
27. Zhang, C. H., Wen, S. Z., Luo, J. B., Simulation of thin film lubrication with micropolar fluids, *Chinese Journal of Mechanical Engineering* (in Chinese), 2001, 37(9): 4–8.
28. Zhang, C. H., Luo, J. B., Wen, S. Z., Exploring micropolar effects in thin film lubrication, *Science in China, Ser. G*, 2004, 47(supp): 65–71.
29. Zhang, C. H., Wen, S. Z., Luo, J. B., On characteristics of lubrication at nano-scale in two-phase fluid system, *Science in China, Ser. B*, 2002, 45(2): 166–172.
30. Zhang, C. H., Wen, S. Z., Luo, J. B., A new postulation of viscosity and its application in computation of film thickness in TFL, *ASME J. Tribol.*, 2002, 124(4): 811–814.
31. Zhang, C. H., Luo, J. B., Wen, S. Z., Effects of nano-scale particles in chemical mechanical polishing process, *Acta Physica Sinica*, 2005, 54(5): 2123–2127.

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