Influence of Adsorption Characteristics of Surfactants Sodium Dodecyl Sulfate and Aerosol-OT on Transition Phase of Water-Based Lubrication

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Abstract

Previous studies of surfactants as water-based lubrication additives have mainly focused on their steady-state behavior, but the “transition phase” between two steady states has been rarely studied, which is more or at least equally important for actual lubrication conditions. In this study, the lubrication behaviors of sodium dodecyl sulfate (SDS) and sodium bis (2-ethylhexyl) sulfosuccinate (Aerosol-OT, AOT) solutions were comparatively investigated under step-changed working conditions. It was found that the duration of the transition phase of AOT solution as lubricant was significantly shorter than that of SDS. During the transition phase, the contact zone experienced a slight running-in and fresh surfaces were created. Compared with SDS, the adsorption rate of AOT was higher, and its adsorption amount was lower due to the larger steric hindrance of the molecular structure. Therefore, the adsorption state of AOT in the contact zone could reach a new dynamic equilibrium faster, so that the friction coefficient would reach a stable value faster. Different from the common opinion that running-in is the cause of the transition phase of lubrication, this study indicates that the dynamic adsorption behavior of surfactant molecules mainly contributes to this process, which should be paid more attention to guide their industrial applications.

1 Introduction

Studies have shown that approximately one-third of the total global primary energy consumption originates from tribological contacts, leading to significant industry demands to reduce friction and wear through proper lubrication [1, 2]. Water-based lubricants are environmentally friendly and have a low cost, and thus warrant studies on appropriate frictional additives [3–5]. In particular, amphiphilic surfactants are typically applied to improve the boundary lubrication performance [1], and their lubrication properties and mechanisms require extensive research.

In previous studies, the effects of surfactants in steady-state lubrication have been widely recognized, wherein one end of amphiphilic surfactant molecule is adsorbed on friction pair surface and the other end is exposed, resulting in the formation of a boundary film to reduce the direct contact of rough peaks [6–8]. In steady-state lubrication, surfactant molecules are in dynamic equilibrium of adsorption, desorption and mechanical disturbance on the solid surface [9, 10]. The antifriction effect is typically determined by the surficial-coverage of the adsorption film, which can be influenced by the adsorption strength and the packing density of the surfactants at the interface [11–14].

Research on the process before the friction system reaches the steady state mainly focuses on the running-in process. In the running-in process, two fresh surfaces that contact with rough peaks start relative sliding and undergo significant plastic deformation and wear until the wear rate or coefficient of friction (COF) reaches a steady state [15]. The microscopic essence involved in this process is mainly the physical and chemical evolution of the surface morphology, composition and third-body distribution of the friction pair [16, 17]. The lubrication states after running-in can be greatly affected by the working conditions of running-in [15, 16, 18].

Different from running-in process, there are relatively few studies on the “transition phase”, that is, on the surface that has finished running-in, the COF changes due to changing working conditions, and then it gradually reaches a stable value due to the molecular behavior of the lubricant. Some studies have noted that for some organic friction modifiers, the COF in the first friction cycle may be higher than that of subsequent cycles because friction stimulates or accelerates the formation of adsorption films [19–22]. Another typical case is the friction-induced extension and orientation of long-chain molecules and liquid crystals, causing the reduction of interfacial viscosity and molecular internal friction. Accordingly, COF is gradually decreased [23–28].

In summary, previous studies have provided a relatively deep understanding of the lubrication mechanism of surfactants, but mainly in the steady-state lubrication situation. For the COF evolution before the steady state, the main focus is on running-in, but research remains lacking on the transition phase, during which the COF gradually decreases to a stable value owing to molecular behavior of lubricants but not running-in of surface. It is important to understand further about this process because it typically occurs under stop–start and variable load conditions, and it has a great influence on the stability and life of the mechanism.

In this study, commonly used anionic surfactants SDS and AOT were selected to assess the characteristics of the transition phase. They have the same number of carbon atoms in the main carbon chains; however, SDS is single-chained while AOT is double-chained, as shown in Figs. 1(a) and (b). The steady-state lubrication properties of SDS have been deeply studied. Zhang studied the relationship between the adsorption structure and COF [29]. Siqing and Liu applied different electrode potential to the friction pair to control the adsorption/desorption of SDS and its tribological properties [30–32]. But AOT has not been investigated as a lubricant additive, probably due to its large-steric-hindrance molecular structure that is hard to form dense adsorption layer. However, AOT has a strong aggregation and transport ability that it can sequentially form micelles, vesicles, and several phase structures as the concentration increases [33–36], and AOT vesicles exhibit a higher diffusion rate and adsorption strength to hydrophobic surfaces than those of SDS micelles [36–38].

Therefore, SDS and AOT aqueous solutions were used as lubricants to comparatively study the transition phase of the lubrication under various working conditions, and the duration of the transition phase was the main focus. The essence of the transition phase was revealed from the evolution of solid surface topography and the establishment of the adsorption state of surfactant molecules. The adsorption characteristics were compared from the aspects of adsorption rate and adsorption amount, and their effects on the transition phase were determined.

2 Experimental Section

2.1 Materials

The two surfactants used in the study were SDS (purity > 99%; Shanghai Macklin Biochemical Technology Co., Ltd.) and AOT (purity > 99%; Tianjin Shinesopod Technology Co., Ltd.). Their aqueous solutions with concentrations of 5 and 10 mM were prepared with deionized water (DW). In addition, aluminum oxide plates (Al_2O_3 ; 15 mm × 6 mm; $R_a \sim 2.7$ nm; Xi'an Laikete Electronic Technology Co., Ltd.), aluminum oxide balls (Al_2O_3 ; ϕ 12.7 mm; $R_a \sim 30$ nm; Dongguan Bluewhale Ceramic Technology Co., Ltd.) and silicon nitride balls (Si_3N_4 ; ϕ 12.7 mm; $R_a \sim 2.5$ nm; Shanghai Nuansheng Electronic Commerce Co., Ltd.) were selected as the friction pairs. They were cleaned in an ultrasonic bath successively with petroleum ether, ethanol, and DW for 5 min each and dried with compressed air.

2.2 Tribology tests

Friction tests were performed with two apparatus (Fig. 1(c)). Firstly, a ball-on-three-plate friction test apparatus (MCR301, Anton Paar Trading Co., Ltd.) was mainly used, and the friction pairs were three Al_2O_3 plates and one Si_3N_4 ball. Secondly, the combined experiment of friction and observation of wear scars was performed on a four-ball test apparatus and its supporting microscope (MS-10JS, Xiamen Tenkey Automation Co., Ltd.), and the friction pairs were three Al_2O_3 balls and one Si_3N_4 ball. The magnification of the microscope was 300X. All friction tests were carried out at 25 °C with DW or surfactant solutions as lubricants. For each newly prepared friction pair, DW was firstly used as the lubricant to run-in for maximum of 1500 s, until the changes in COF over 120 s were within 0.03 (on ball-on-three-plate tester) or 0.08 (on four-ball tester). After running-in, the friction pairs were rinsed four times alternately with DW and ethanol, and was cleaned with Kimwipes tissue, and then the experiments of surfactant solutions as lubricant could be started. In each friction experiment, the normal load used for running-in was the maximum value of the loads in the subsequent friction tests, so that a stable contact state had been established before the friction tests with SDS or AOT solutions, and the wear scars would not expand significantly during friction. The applied normal force and speed were different in different tests, which is further explained in the subsequent sections.

2.3 Adsorption measurement at solid-liquid interface

The adsorption mass of SDS or AOT molecules at different concentrations was measured using a quartz crystal microbalance (QCM; Q-sense E4 system, Biolin Scientific, Sweden) with an Al_2O_3 coated quartz crystal sensor at 25 °C, and the flow rate of the liquids was 100 $\mu\text{L}/\text{min}$. Moreover, the adsorbed film was regarded as rigid [39, 40], and the adsorbed mass (Δm) was calculated using the Sauerbrey equation [40] presented in Eq. (1) when changes in the resonance frequency (Δf) and dissipation (ΔD) of the sensor conform to $\Delta D < 10\Delta f$.

$$\Delta m = -C \frac{\Delta f}{n}$$

1

where C is a constant with a value of $17.7 \text{ ng Hz}^{-1} \text{ cm}^{-2}$ that is related to the properties of the quartz crystal, and n denotes the overtone of the oscillations. In this experiment, Δf and Δm were recorded and

calculated using the seventh overtones.

3 Results

Typical curves of the transition phase with SDS and AOT as lubricants are shown in Fig. 2. The friction tests were performed on the ball-on-three-plate apparatus. On the friction pair that had already been run-in with DW, 5 mM SDS and AOT aqueous solutions were used as lubricants. The applied normal load (F_N) was 1 N and 5 N, respectively, as shown in Figs. 2(a) and 2(b). The angular velocity (ω) of the Si_3N_4 ball was accelerated from zero to 300 rpm within one second. After the onset of friction, the COFs of both AOT and SDS experienced a period of decline, which were typical transition phase curves, and then gradually reached a stable value. Although the stable COF of SDS was lower than that of AOT, the COF of AOT decreased more rapidly during the transition phase, generally reaching stability within 10 s. But the transition phase of SDS required a longer time, approximately 30 s at a load of 1 N, and approximately 80 s when the load increased to 5 N.

On the friction pair that had been run-in with DW under a normal load of 40 N, a step-up normal load was applied as $F_N = 1, 2, 5, 10, 20, 40$ N, and the friction lasted 40 s at each load, with $\omega = 300$ rpm. A similar phenomenon occurred, as shown in Fig. 3. After each load, the COF of SDS and AOT immediately increased, and then experienced a transition phase until reaching a stable value. In comparing AOT and SDS, AOT had a significantly shorter transition phase after an abrupt loading during the four steps from $F_N = 2$ to 20 N, although SDS could achieve a lower final COF value. In comparing SDS solutions of different concentrations, under the same working conditions, higher concentration of SDS had a larger and faster decrease in COF and a shorter transition period. The higher was the F_N , the longer was the transition phase of SDS solutions and the greater was the COF drop. When $F_N = 40$ N, the COF under the lubrication of surfactant solutions oscillated between 0.5 and 0.7, reaching a level similar to that of DW lubrication, indicating the failure of the surfactant molecular adsorption film.

On the friction pair that had been run-in with DW under a normal load of 2 N, a continuous-variable-speed experiment was performed under the load 2 N, as shown in Fig. 4. The friction lasted 3 s at each speed, and the COF was the average over 3 s. For both SDS and AOT, the COF in round 1 was found to be higher than those in the other rounds. This is because the Si_3N_4 ball was lifted while changing lubricants (from DW to the surfactant solution) and was then placed at the measurement position again. Accordingly, the surfaces experienced a short running-in at the start of each experiment. The friction curves of SDS in rounds 2–4 did not coincide well and the COF in the boundary lubrication region decreased significantly among rounds, until the COF of rounds 5–8 were almost coincident. In contrast, the friction curves of AOT in rounds 2–8 all had a high coincidence degree. Thus the transition phase duration of SDS (rounds 1–4) was much longer than that of AOT (round 1).

In summary, friction experiments were conducted under various working conditions, including combinations of various loads and various speeds, using SDS and AOT aqueous solutions as lubricants.

It was found that in the transition phase before the system reached a steady state, the COF of AOT decreased considerably faster than that of SDS.

4 Discussion

There were two possible reasons why the COF decreased with time before the system reached a steady state. Firstly, because the friction pair was temporarily separated when cleaning and adding the surfactant solutions before each experiment, the relative position of the friction pair was changed when contacted again. Therefore, the friction pair would run-in again for a short time. Secondly, at the beginning of the transition phase, the adsorption state of the surfactants in the contact zone had not yet reached a steady state. After a period of friction, the number of molecules adsorbed in the contact zone reached a dynamic equilibrium. The arrangement characteristics, such as the molecular conformation and orientation also reached a balance, and the COF could then reach a stable value. To explore the reason why the time required for the transition phase of SDS and AOT was different, the influencing factors of the above two aspects were respectively verified.

The four-ball friction test apparatus and its supporting microscope were used to conduct friction experiments and observe wear scars, the results for SDS and AOT are respectively shown in Fig. 5 and Fig. 6. The experiment consisted of two stages, stage 1 contained the rising edge of the COF and was stopped after the COF reached its peak value, and stage 2 contained the falling edge of the COF and was stopped when the COF tended to be stable. The purpose of doing this was to compare the wear scars at different moments of the transition phase, to verify whether the transition phase was caused by running-in.

Figure 5(a) shows the COF over time with 5 mM SDS as lubricant. The applied normal load was 5 N, and the angular velocity of the upper ball was 300 rpm. Before starting, the friction pair had been run-in with DW under a normal load of 5 N, and the wear scar of one of the lower ball is shown in Fig. 5(b). Then the SDS solution was added as lubricant to start friction, which was stage 1 in Fig. 5(a). During this period, the COF first increased, and then began to decrease. So the friction was stopped, and the solution was poured out and blow-dried to observe the wear scar of the lower balls, as shown in Fig. 5(c). After that, new SDS solution was added to start the friction again, which was stage 2 in Fig. 5(a). In this stage, the COF increased slightly again, and gradually decreased in the following 30 s until the COF tended to be stable. So the friction was stopped again to enable observation of the wear scar, as shown in Fig. 5(d). Although the COF curve in Fig. 5(a) appears similar to the running-in curve under boundary lubrication [15], changes of COF in Fig. 5(a) is actually different from a pure running-in, because in comparing the size and internal morphology of wear scars in Figs. 5(b)–(d), it could be seen that the size in 5(c) along the friction direction was approximately 0.2% larger than that of 5(b), and the internal grooves were slightly shallower, while the size and grooves of 5(d) showed no obvious changes compared with 5(c). This demonstrates that when the COF of stage 1 increased significantly, a slight running-in occurred in the contact zone, while the change of wear scar in stage 2 was not obvious, and the continuous decline of COF was not only related to running-in.

Similar experiments were also performed with 5 mM AOT solution (Fig. 6), and a similar phenomenon was observed. During each stage, the COF first increased for several seconds, and then decreased. The size and internal grooves of the wear scars changed slightly, similar to Fig. 5. The main difference with Fig. 5 was the duration of the falling edge of COF; thus, the main difference in the duration of the transition phase between AOT and SDS should have been related to molecular behavior, rather than running-in.

Since the transition phase was not completely caused by running-in, the changes in the adsorption state of lubricant molecules could have also been related to the changes in COF. Therefore, the adsorption rates of different concentrations of SDS and AOT solutions were compared, as shown in Fig. 7. Typical curves of the adsorbed number of molecules per unit area are depicted in Fig. 7(a), where the adsorption amount gradually increased and tended to stabilize, and AOT reached saturation faster than SDS. The change in the adsorbed molecular number as a function of time was fitted using the first-order adsorption kinetic equation [41]:

$$\log(q_e - q_t) = \log q_e - \frac{k_{ads}}{2.303} t$$

2

where q_e and q_t denote the maximum value and the value at time t of the adsorbed molecular number, respectively, and k_{ads} represents the rate constant, where a larger k_{ads} indicates a higher adsorption rate. The change in $\log(q_e - q_t)$ as a function of time is depicted in Fig. 7(b), and it can be fitted using Eq. (2). Thus, the value of k_{ads} for different solutions can be calculated, as presented in Table 1. The values of k_{ads} for AOT were larger than those for SDS at all concentrations, indicating that the adsorption of AOT was faster than SDS.

Table 1
Adsorption rate k_{ads} of different solutions.

Solution	5 mM	5 mM	10 mM	10 mM
	SDS	AOT	SDS	AOT
$K_{ads} (s^{-1})$	0.0274	0.0507	0.0343	0.0440

In comparing the final adsorption amount in Fig. 7(a), the final adsorption amount of SDS was greater than that of AOT. This is because SDS with a linear structure can be more densely packed than AOT with a branched structure [6, 42] on solid surface. Moreover, the difference in the molecular structure and adsorption amount of SDS and AOT led to different lubrication effects, as shown in Figs. 2–4. Under steady-state lubrication, the COF of SDS as lubricant was lower than that of AOT in boundary and mixed lubrication regions. There were two possible reasons, firstly, the boundary film of SDS was denser (Fig. 7(a)); secondly, in boundary lubrication regions, some parts of contact zones were very limited in

space, with only one or a few molecular diameters. SDS molecules were able to enter these contact zones due to their smaller steric hindrance, while AOT molecules were difficult to enter with their larger steric hindrance (Fig. 8). Thus, the adsorption amount as well as the surficial-coverage of SDS in the contact zone were gradually increased in the transition phase, and eventually achieved a value greater than that of AOT under steady-state lubrication. The diffusion time of confined molecules could be considerably longer than that of bulk molecules [43], so the duration of transition phase was longer for SDS.

In summary, in the friction tests, the relative position of the friction pair changed slightly before and after adding the surfactant solutions as lubricant; in the step-up-normal-load tests, the area of the contact zone increased after each loading. Thus, the contact zone would experience a slight running-in at the beginning of each transition phase, leading to the generation of fresh surfaces that required rapid adsorption of surfactant molecules. A larger adsorption rate of AOT enabled a faster replenishment into the contact zones, so its COF decreased rapidly; SDS had a lower adsorption rate, but also a larger adsorption amount in confined zones due to its smaller steric hindrance, showing a COF curve that decreased more slowly and continuously to a lower stable value. Therefore, the duration of transition phase of SDS was much longer than that of AOT.

5 Conclusion

In this study, the influence of adsorption characteristics of surfactant SDS and AOT as lubricant additives on the transition phase of the lubrication process was comparatively studied. After a slight running-in, the COF of AOT as lubricant decreased and reached a steady state faster than SDS, which was attributed to the larger adsorption rate and smaller adsorption amount of AOT than that of SDS. Thus, AOT could reach the dynamic equilibrium adsorption state faster, resulting in the shorter duration of the transition phase. This study focused on the unsteady friction process that is often overlooked in research and on the molecular behavior of lubricants during this process. The results may provide in-depth understanding of unsteady lubrication behavior, and important guidance for the selection and application of water-based lubricant additives.

Abbreviations

SDS, sodium dodecyl sulfate, AOT, sodium bis (2-ethylhexyl) sulfosuccinate, COF, coefficient of friction, DW, deionized water, QCM: quartz crystal microbalance.

Declarations

Acknowledgment

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Competing Interests

The authors have no relevant financial or non-financial interests to disclose.

Author Contributions

All authors contributed to the study conception. Jingbo Fang: investigation, experiment, writing. Pengpeng Bai: validation, investigation. Chuke Ouyang: methodology, validation, investigation, editing. Chenxu Liu: validation, investigation, editing. Xiangli Wen: validation, editing. Yuanzhe Li: editing. Yonggang Meng: resources. Liran Ma: resources. Yu Tian: supervision, methodology, resources, editing. All authors read and approved the final manuscript.

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Figures

Figure 1

Schematic of the two molecules and the friction test apparatuses used in this study. (a-b) Structure diagram of SDS and AOT. (c) Illustration of the ball-on-three-plate and four-ball friction test apparatuses and contact region. F and v denotes the load and velocity applied to the contact zone, respectively. For simplicity, only two plates and two lower balls are shown in the figure

Figure 2

COF of the friction tests with fixed load and speed. The normal load was 1 N (a) and 5 N (b). ω was 300 rpm

Figure 3

COF of the friction tests with a fixed speed and step-up load. ω was 300 rpm. The normal load was applied as $F_N = 1, 2, 5, 10, 20, 40$ N

Figure 4

COF evolution in the procedures of the continuous-variable-speed-experiment with 5 mM SDS (a) and AOT (b) as lubricants and $F_N = 2$ N. ω increased from 0.1 to 3000 rpm in round 1, then reduced from 3000 to 0.1 rpm in round 2, and increased again in round 3, and so on, until it decreased to 0.1 rpm in round 8. Each round was continuous, without stopping or changing the direction of rotation

Figure 5

Friction curves with 5 mM SDS solution as lubricant and wear scar at each stage. (a) Two stages of COF curves over time. Before stage 1, the friction pair had been run-in with DW, and the wear scar was in (b). (c–d) The wear scar after stage 1 and stage 2

Figure 6

Friction curves with 5 mM AOT solution as lubricant and wear scar at each stage. (a) Two stages of COF curves over time. The normal load and speed were the same with that of SDS. Before stage 1, the friction pair had been run-in with DW, and the wear scar was as shown in (b). (c–d) The wear scar after stage 1 and stage 2

Figure 7

Adsorption of SDS and AOT solutions measured using QCM. (a) Typical scatter plot of the adsorbed molecular number per unit area varying with the introduction time of the liquid. (b) The adsorption process was fitted to the first-order adsorption kinetic equation, and the data within the time of 10–30 s occurred in accordance with the linear relationship, which could be used to calculate the adsorption rate k_{ads}

Figure 8

Mechanism illustration for the molecular behavior in the transition phase. (a) SDS had small steric hindrance and low adsorption rate, so it could enter the contact zones with very limited-space slowly and continuously. (b) AOT had large steric hindrance, making it difficult to enter the same contact zones as that of SDS, so it reached equilibrium quickly under the high adsorption rate

Supplementary Files

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- [Graphicalabstract.png](#)