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In situ probing of stress-induced nanoparticle dispersion and friction reduction in lubricating grease[☆]

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ABSTRACT

Particulate additives improve the lubricating performance of oils and greases. Effects of nanoparticles on lubrication are not well understood because it is not possible to see them. In this research, an in situ approach is used to visualize the movement of particulate additives in grease under a shear stress. A model grease in the form of petroleum jelly was mixed with 2.3 wt% α-ZrP nanoparticles. A custom rotating concentric tube device was built to shear and monitor the grease using tomography technique. Through the use of dual edge micro x-ray computed tomography (μ-XCT), the distribution of the particulate additives in the Petroleum jelly were visualized. Results showed that prior to shearing, the α-ZrP nanoparticles were randomly aggregated throughout the annular gap between the cylinders. However, after a shear stress was applied to the grease mixture the additives were shown to evenly distribute throughout the annular gap. The visual results indicate that the α-ZrP nanoparticles tend to separate and re-distribute along the shearing direction, which attributes to friction reduction. The shear-stress enabled uniformity of particle distribution indicate potential benefits and effectiveness using nanoparticle as additives. This research opens windows for future investigation in lubrication.

1. Introduction

The effects of friction and wear on everyday mechanical systems lead to large drops in efficiency and overall waste in energy. Automobiles, such as cars and trucks, for instance tend to waste an average of 33% of fuel to overcome frictional losses within the engine, other auxiliary systems [1,2]. Proper lubrication within critical areas will cause a reduction in friction and ultimately lead to higher efficiently for the entire system. The most common types of lubricants used in mechanical systems are liquid lubricants in the form of oils, and semisolids in the form of greases. Lubricating greases typically behave as a non-Newtonian shear thinning fluid when exposed to a mechanical shear stress [3]. These greases can be broken down into three major components: a base oil, a thickening agent; and additives. The incorporation of organic and inorganic additives in lubricants have been shown to contribute to a reduction in friction and wear within different mechanical systems [4–7]. The performance of an additive relies on several factors including the chemical nature, size, shape, and concentration. The mixing process used to incorporate an additive must also be taken into consideration. The consistency in performance of an additive relies heavily on the homogeneity of the entire mixture [8,9]. Decrease in performance can be attributed to an improper mixture, or a mixture that becomes separated over time; de-homogenization [10,11]. The manufacturing procedures for grease is therefore a complex process that requires numerous steps and controlled parameters to ensure a consistent product. In addition, grease mixtures will tend to vary in rheological properties under the mechanical stress of the mixing process, with no direct relationship to its performance post-processing [12]. A new understanding of additive homogenization is needed. Many studies have shown the benefits of incorporating various additives, however there exists a lack of evidence as to how these additive particles interact within the system. In our previous work, we presented a methodology to directly observe additive particles in a grease using X-ray computed tomography (CT) [13]. It was the first time to be able to “see” the particles inside a grease. To further understand how additive particles behavior, the present work will focus on tracking the movement of particle species under shear within a pure base grease.

Computed tomography is a highly versatile technology that is most commonly used for medical purposes to diagnose various ailments

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within a live subject. It is a nondestructive procedure that can generate two dimensional orthogonal cross sections of an object, as well as full three dimensional models. When utilized under controlled conditions, a synchrotron based micro X-ray CT (u-XCT) system can produced high resolution images capable of observing objects at a micron and sub-micron scale [14,15]. Through our past work on grease additives, μ-XCT has been shown to be an effective method for observing particle in grease through the manipulation of the monochromatic X-ray energy [13]. The objective of the present research is to expand on the methodology reported previously and study the movement of particles. A simple base grease will be used, and only one particle additive species will be incorporated. The additives to be observed will be α-ZrP nanoparticles (NP). The weak van der Waals forces between the two dimensional α-ZrP platelets has been attributed to the improvement of friction and wear behavior when incorporated into a lubricant [16–18]. Weak bonding forces between each particle allow for them to be easily exfoliated, thereby providing more effective fluid transport for lubrication. The grease mixture will be observed as received and after a controlled shear stress has been applied. Analyzing a pre and post sheared grease under μ-XCT will allow for a better understanding of the particle behavior and the lubrication mechanism.

2. Methods

2.1. Materials

The α-ZrP NPs were prepared by the hydrothermal method as reported previously [19]. The lubricant used in this study is petroleum jelly, which is a petroleum based semi-solid. Although not normally used for industrial applications, petroleum jelly is a common lubricant used to moisturize human skin [20,21]. It consists primarily of a mineral base oil with various hydrocarbons type thickeners to form a semi-solid, high viscosity fluid. Petroleum jelly exhibits a non-Newtonian shear thinning behavior when under an applied mechanical deformation [22]. Studies have shown petroleum jelly to be a good medium for the analysis of additive particles for friction and wear performance [23,24]. For the purpose of this investigation, it will serve as a good medium for the incorporation and study of nanoparticle behavior under shear.

In order to investigate the behavior of the NP additives mixed in the petroleum jelly, two sample groups were studied using a K-edge tomography method; one mixed as received, and one that underwent controlled sliding (shear). Each sample contained Petroleum jelly mixed with Zirconium phosphate (α-ZrP) additive at 2.3 wt%. The petroleum jelly and NPs were simply mixed by hand inside a test tube until a uniform coloration was achieved. The samples and their rotation status are listed in Table 1. The samples were deposited between the inner and outer cylinders of a rotational device that will described in the next section.

2.2. Methodology

In order to observe the grease/additive mixture under controlled sliding conditions, a special testing rig was built. The rig consists of a rotating inner cylinder and a stationary outer cylinder made from polyamide plastic tubes. Polyamide tubes are necessary as they will appear "transparent" under x-ray and allow for an unobstructed view of the grease mixture. The inner cylinder is connected to the shaft of a 2 phase stepper motor and a rotary encoder. The speed of the motor is electronically controlled, while the encoder measures the rotational speed of the inner cylinder. A closed loop control system was used to supply a sufficient amount of torque to maintain a set motor speed over the test period. A model of the rig design is shown in Fig. 1.

Table 1
List of the two sample groups with their testing conditions.

Group 1	Group 2
Petroleum Jelly+2.3 wt% α-ZrP As received	Petroleum Jelly+2.3 wt% α-ZrP Sheared

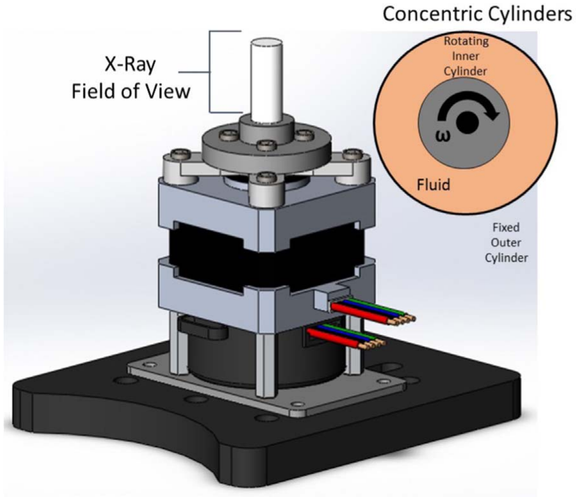

Fig. 1. Diagram of the concentric cylinder testing rig used to shear the grease/nanoparticle mixture.

The entire rig is situated such that the rotational axis of the tubes sit along the central axis of the Beamline sample table as shown in Fig. 2. The outer diameter of the stationary outside tube is 4 mm, which is less than the 5 mm field of vision (FOV) of the microscope used to perform the imaging. This configuration will allow for a uniform scan of the entire annular gap between the cylinders, and a clear image of the grease and NP mixture.

The concentric cylinder device was calibrated prior to placement into the beamline so as to minimize error and simplify the test procedure when placed within the beamline. Based on the specifications of the motor used, a constant torque of 9.5 N-cm was established for the grease sample at a rotational speed of 25 revolutions per min (RPM).

2.3. Micro-tomography

Synchrotron based X-ray micro-tomography was performed at the Advanced Light Source (ALS) facility at Lawrence Berkeley National Laboratory (LBNL). Synchrotron scanning was performed at Beamline 8.3.2 at ALS. Each of the grease samples were scanned at two energy levels that correspond to being higher and lower than the K-edge of zirconium (17.998 keV). This methodology has been discussed in our previous research elsewhere [13,14,25]. The two energies used for the scanning were 17.8 keV and 18.2 keV. A 5x optical lens with 0.00129 mm resolution was used. Other parameter settings and operations were the same as we reported before [13,14].

The distribution of particulate additives in petroleum jelly was visualized using micro x-ray computed tomography. Petroleum jelly was mixed with 2.3 wt% α-ZrP nanoparticles and placed between the concentric cylinders of a rotating device to be sheared. Through the use of dual energy K-edge tomography technique, the zirconium element in the particles was distinguishable from the carbon chains comprising the base petroleum jelly grease. Identifying different species within the petroleum jelly mixture allowed for an image mapping of the particulate movement before and after a shear force was applied. The images of the simply mixed petroleum jelly and NPs showed that the particles aggregated in certain areas, a non-homogenous compound. Under the influence of shear, it was observed that the α-ZrP nanoparticles will tend to separate and re-distribute within the annular gap, becoming homogenized. This behavior lends to the reduction in friction coefficient that was reported for petroleum jelly when α-ZrP nanoparticles