Taiho Kogyo Tribology Research Foundation

Main report

Name:
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Theme:
New Measurement Methods for Molecular Design Theory of Polymer Additives

1. Progress and result of the research

The final goal of this project is to establish a bottom-up molecular design theory for polymer additives. This theory aims to predict the lubrication performance of adsorption films from the molecular structure of additives, utilizing new measurement methods. In this project, we proposed and tried the new methods for measuring (1) intermolecular interaction, (2) molecular state (in oil and adsorption state), and (3) lubrication performance of adsorption film. These methods are essential for establishing the molecular design theory of the polymer additives. We used polyalkylmethacrylate (PAMA), which is gaining attention as both a viscosity index improver and friction modifier for electric vehicle drivetrains (E-axle). The details of the measurement methods are as follows:

(1) Measurement of intermolecular interactions

To quantify the intermolecular interactions between the additive, lubricant, and sliding surface, we introduced the Hansen solubility parameter (HSP) method. In this method, by testing whether the sample dissolves in various test solvents or not, and comparing the properties of the test solvents, the interaction parameter of the sample can be quantified as δ. Hence, the more similar the interaction parameter δ, the better the dissolution. In this project, we acquired the HSPs using theoretical estimation method based on the data obtained so far. We could evaluate the molecular interaction (non-polar, polar, hydrogen-bonding) of the PAMA from the molecular structure by the HSP method.

(2) Measurement of molecular state (in oil and at adsorption sites)

A) Molecular state in oil (molecular radius in oil)

We utilized the dynamic light scattering method to measure the molecular radius of additives in the base oil. This measurement helps to estimate the molecular state of the additives in oil. In this method, laser light is irradiated to the molecule in a liquid and the radius of the molecule is determined from fluctuations in the scattered light intensity caused by Brownian motion of the molecules. Through precise quantification of the fluctuations using a temporal autocorrelation function, the molecular radius can be quantified. Using this method, we could evaluate around 3 nm as the radius of the PAMA. In addition, we successfully obtained the change of the radius caused by the temperature change with a 1-nm resolution. Based on these results, we can estimate the molecular state in oil.

B) Molecular state in adsorption state (adsorption film thickness)

The ellipsometric microscope developed by us was used to measure the film thickness of additive molecules during adsorption in real time. The ellipsometric microscope can convert the distribution of the nano-scale gap into a brightness image based on ellipsometry. Our original optical system has achieved a gap resolution of the order of 0.1 nm and a lateral resolution of the order of 0.1 μm. However, due to the similar refractive indices of base oils and additives, distinguishing between them optically was difficult. To overcome this limitation, a novel method was proposed. In this new approach, after the slider (metal-coated lens) is separated from the sliding surface (glass plate) and the additive is adsorbed on the surface, the slider was pressed against the sliding surface to squeeze out the base oil from the gap. By measuring the resulting gap, the thickness of the adsorbed film was successfully obtained. Using this method, we could evaluate around 5 nm as the adsorption film thickness of the PAMA and successfully obtained the adsorption thickness at different temperatures with a thickness resolution of 1 nm. This enables us to estimate the adsorption state of the PAMA molecules on the sliding surfaces. In addition, we proposed method for estimation of the adsorption energy between the PAMA molecule and sliding surface using this adsorption thickness measurement. The shear strength of the adsorption film can be estimated using the adsorption energy.

(3) Measurement of lubrication performance of adsorption film

By simultaneously measuring the adsorption film thickness and friction properties with the ellipsometric microscope, we quantified the boundary lubrication performance of adsorbed films, whose adsorption process had been clarified above. A sliding mechanism was added to the ellipsometric microscope for simultaneous measurement of friction force, load, and sliding gap. The sliding mechanism consisted of a lens coated with a metallic film as a slider, supported by a vertically deformable plate spring, which was attached to a laterally deformable parallel leaf spring. The glass plate was displaced laterally by a piezo stage, and the adsorption film was slid. The vertical and lateral deformations of the plate springs were measured by optical interferometers to obtain the load and friction force, respectively. We could evaluate around 0.1 as the coefficient of friction (COF) of the adsorption film of the PAMA. Using this COF, we can estimate the friction properties of the adsorption films.

2. Subjects remain to be solved in future/Subjects required further investigation

In this project, we were able to confirm the validity of the proposed methods for measuring (1) intermolecular interaction, (2) molecular state (in oil and adsorption state), and (3) lubrication performance of adsorption film. However, we have not yet established a bottom-up additive design theory using these measurement methods. Moving forward, we aim to organically integrate the results from each measurement method to estimate the molecular states within the oil and the adsorbed film. This will form the basis for developing a design theory capable of predicting friction characteristics. In particular, we will consider incorporating AI techniques, such as machine learning, which has seen significant advancement in recent years, to integrate the results from each measurement method.

3. Plan and past presentation or publication of your research results

Past presentation

1. Kenji Fukuzawa, Measurement of Adsorption and Friction Properties of Additives by Vertical-Objective Type Ellipsometric Microscopy, 49th Leeds Lyon SYMPOSIUM TRIBOLOGY, Lyon France (2024).

2. Masaya Kuroda, Shoya Tanabe, Yuxi Song, Kenji Fukuzawa, Shintaro Itoh, Hedong Zhang, Quantification of Adsorption Energy of Polymer Additive Polyalkylmethacrylate by Direct Measurement of Adsorption Film Thickness, Tribology Conference 2025 Spring, Tokyo Japan (2025).

3. Kenji Fukuzawa, Yuxi Song, Shintaro Itoh, Naoki Azuma, Hedong Zhang, Evaluation of Relationship between Adsorption/Friction Properties and Molecular Properties of Additives by Vertical-Objective Type Ellipsometric Microscopy, 50th Leeds Lyon SYMPOSIUM TRIBOLOGY, Leeds UK (2025).

Publication plan

1. To be submitted to an academic journal such as Tribology International