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Friction and wear characteristics of DLC coatings with different hydrogen content lubricated with several Mo-containing compounds and their related compounds

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ABSTRACT

Although the molybdenum dithiocarbamate (MoDTC) additive results in superior friction-reducing performance for both steel and diamond-like carbon (DLC) surfaces, it sometimes causes wear acceleration of DLC surfaces. This study confirmed the wear acceleration of a DLC surface lubricated with MoDTC-formulated oil. Three types of DLCs with different hydrogen contents were used. The nominal hydrogen contents of DLCs were hydrogen-free (ta-C), 10 at% (a-C:10H) and 30 at% (a-C:30H). The DLC coatings were deposited on a tool steel substrate. It was confirmed that MoDTC accelerated the wear of hydrogenated DLCs, although the formulation of MoDTC showed good friction-reducing performance for every specimen. To confirm the tribological characteristics of the other Mo-containing compounds for DLC lubrication, friction tests were carried out with several types of Mo-containing additives and their related Mo-free organic compounds. When the hydrogenated DLCs were lubricated with Mo-containing additives, the wear of the hydrogenated DLCs was accelerated. However, the hydrogenated DLCs did not show wear acceleration when lubricated with the related Mo-free organic compounds. The DLC surfaces after the friction test with Mo-containing additives were analyzed by X-ray photoelectron spectroscopy, and the MoO_3 formation was confirmed on every specimen. Although the wear behavior was considered in relation to MoO_3 formation on the worn surface to determine whether MoO_3 plays a key role in accelerating the wear of DLC, apparent relationship between the wear acceleration and the MoO_3 formation was not obtained.

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1. Introduction

The amorphous carbon coating known as diamond-like carbon (DLC) has become a leading material as a self-lubricating coating due to its remarkable tribology characteristics, such as low friction and good wear resistance. Among the many fields in which DLC has been applied, the field of automotive engineering has been particularly aided by this coating, which promises to deliver both excellent fuel economy and supreme antiwear performance. In internal combustion engines, DLC coatings have already been applied to several components that are lubricated with engine oil.

In general, the engine oil itself has been developed to show low friction and wear through the formulation of suitable additives. Molybdenum dithiocarbamate (MoDTC) is one of the well-known friction modifiers and yields a very low friction coefficient for steel surfaces. Numerous reports have studied the mechanism by which MoDTC formulations achieve such low boundary frictions [1–8]. Currently, it is widely believed that the very low boundary friction coefficient is brought about by the generation of MoS_2-containing tribofilm on the friction surface. Since MoO_3 is also usually generated on the surface, it is crucial that the amount of MoO_3 formation be reduced – i.e., that the MoS_2 concentration be increased – in order to obtain excellent friction reduction by formulating MoDTC or other Mo-containing additives.

Car manufacturers are expecting dramatic improvements in fuel economy through the synergistic effect of DLC coating and additive formulation. While many reports have concluded that an MoS_2-containing tribofilm is formed on the DLC surface and reveals low friction [9–12], there has also been a report that showed no friction-reducing tribofilm formation due to the chemical inertness of DLC surfaces [13]. It has already been

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established that the combination of glycerol monooleate (GMO) and ta-C coating without MoDTC shows a very low friction coefficient that is equivalent to or lower than that by the combination of steel and MoDTC [14]. Since there are several types of DLCs, the actual tribological performance is not fully understood yet. Moreover, a wear problem has been raised for the combination of some DLCs and some engine oil additives [15–17].

Given these circumstances, the objective of our study was to examine the wear problem of DLC. It is generally accepted that MoDTC accelerates the wear of hydrogenated DLC, and that the formation of MoO_3 on DLC surfaces plays a key role in the increased wear. However, since the wear acceleration by MoO_3 has not been confirmed, the objective of this study was to clarify whether or not the generation of MoO_3 plays a role in the wear acceleration of hydrogenated DLC. Since it is very difficult to obtain direct evidence to explain the mechanism of wear acceleration by MoO_3, we tried to obtain circumstantial evidence in the form of tribo-data with other Mo-containing and related Mo-free organic compounds to understand the relationship between DLC wear and MoO_3 formation.

2. Experimental

2.1. Test specimens

Three types of DLC were used in this study. One was a hydrogen-free DLC (ta-C) and the other two were amorphous hydrogenated DLC(a-C:H) having different hydrogen contents of 10 at% and 30 at%. These two hydrogenated DLCs were named a-C:10H and a-C:30H based on the hydrogen content. All the DLCs were deposited on a steel substrate made of tool steel (SK85) after deposition of interlayer films. The hydrogen-free DLC (ta-C) was deposited with a T-shaped filtered arc deposition system that positively collected the droplets, resulting in a smoother surface compared to the other ta-C made by a conventional method [18]. The a-C:10H DLC was deposited by a multi-cathode unbalanced magnetron sputtering method using solid graphite and methane as a carbon source [19–20], and a-C:30H DLC was deposited by the radio frequency plasma enhanced chemical vapor deposition (RFPECVD) method, using acethylene as a carbon source. Typical characteristics are listed in Table 1.

Table 1
Typical characteristic of DLCs.

	a-C:10H	a-C:30H	ta-C
Type	TYPE IIa	TYPE IIb	TYPE I
Method	UBMS	P-CVD	FAD
Carbon source	H/C and graphite	Hydrocarbon (H/C)	Graphite
Hardness [GPa]	~30	30~40	60~70
Water contact angle	39.2°	41.2°	40.0°
Hydrogen content [at%]	10~15	30	0
Thickness [nm]	1100	1000	250

2.2. Lubricants

Poly alpha olefin (PAO) having viscosities of 16.89 mm^2/s at 40 °C and 3.88 mm^2/s at 100 °C was used as the base oil for all the tested oils used in this study. The names and dose concentrations of the additives were as follows. Zinc dioctyldithiophosphate (ZnDTP; 0.1 mass%P) and molybdenum dioctyldithiocarbamate (MoDTC; 0.025 mass%Mo) were used as additives used for engine oils. As references, three other Mo-containing additives were used: molybdenum dithiophosphate (MoDTP; 0.025 mass%Mo), molybdenum phosphate (MoP; 0.025 mass%Mo), and ditridecyl ammonium molybdate (MoAMN; 0.025 mass%Mo). In addition, related Mo-free additives were used; these were dioctylthionophosphoryldisulfide ($(DTP)_2$; the same molar concentration as ZnDTP), zinc dibutyldithiocarbamate (ZnDTC; the same molar concentration as MoDTC), tricresylphosphate (TCP; 0.1 mass%P) and stearic acid (StA; 2.5 mmol/L).

2.3. Tribometer and tribotest conditions

Fig. 1 shows the three-ball-on-disk tribometer used in this study. A rotating disk specimen was pressed against three stationary steel balls at a prefixed applied load. The balls were made of heat-treated high-carbon chromium-bearing steel (AISI 52100). Before the friction measurement, all the balls were ultrasonically cleaned in a toluene bath, and then cleaned using UV-ozone cleaner after drying toluene [21]. Table 2 summarizes the two test conditions used with the three-ball-on-disk tribometer: a heavy loading condition with 500 N and a light loading condition with 250 N. Under the heavy loading

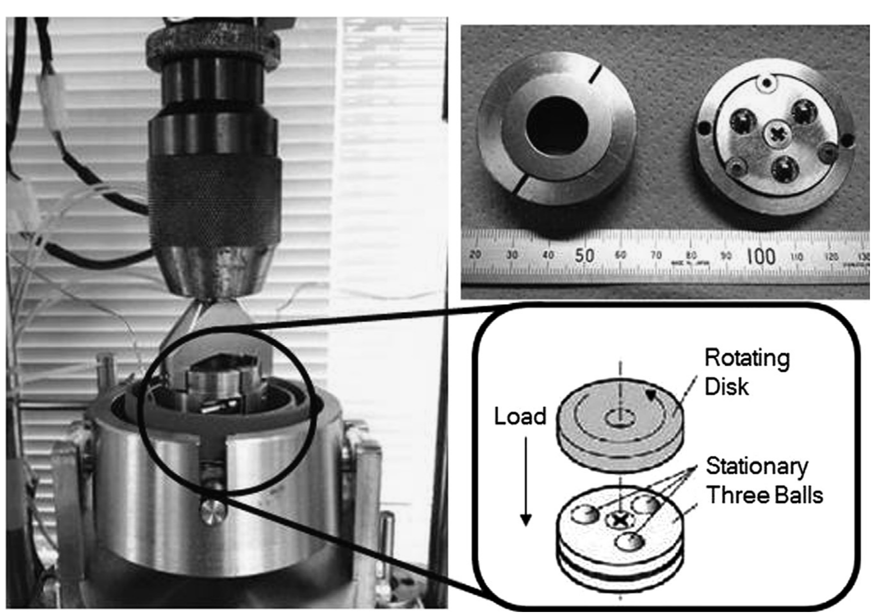

Fig. 1. Three-ball-on-disk tribometer.

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