

Wear properties of a-C:H and Si-DLC in pressurized high-temperature water

M. Takeuchi*, K. Okuno, N. Umehara, M. Murashima

Department of Micro-Nano Mechanical Science and Engineering, Graduate School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan.

*Corresponding e-mail: takeuchi@ume.mech.nagoya-u.ac.jp

Keywords: a-C:H; Si-DLC; wear resistance

ABSTRACT – The wear properties of a-C:H and Si-DLC in pressurized high-temperature water were investigated. We conducted friction tests for DLC pins in pressurized (10 MPa) high-temperature (23, 100, 200, 300 °C) water with two different amounts of dissolved oxygen (0.5, 40 mg/L) to figure out the wear properties of DLC in an oxidative environment. We then clarified that (i) Si-DLC shows great wear resistance even in 300 °C, 10 MPa water with 40 mg/L dissolved oxygen, (ii) while wear scar of a-C:H has soft structure-changed surface, wear scar of Si-DLC has relatively hard surface without much structural change.

1. INTRODUCTION

In order to realize further higher efficiency and longer life of the machine, DLC (Diamond-Like Carbon) coating having excellent characteristics such as low friction coefficient, high hardness, high abrasion resistance and chemical stability has attracted attention [1]. Although it has already been applied to many machines under environments of ambient temperature under atmospheric pressure, the application to equipment operating under a high oxidizing environment is difficult because the friction and wear behavior in a high oxidation environment is unclarified. Zin et al. [2] conducted an abrasion test for a-C:H film in pressurized (10 MPa), high-temperature (300 °C) water environment. As a result, as the amount of dissolved oxygen in water increased, the wear resistance of the a-C:H degraded. It has not been clarified why wear resistance has decreased. Choi et al. [3] conducted experiments of annealing the Si-DLC in a high temperature atmosphere at 500 °C, and clarified the excellent heat resistance of the Si-DLC. However, the wear characteristics of Si-DLC under high oxidation environments are unknown. Therefore, in this study, the friction test using an autoclave was conducted and analyses of the DLC films were carried out in order to evaluate the structural change and aimed to clarify the wear properties of a-C:H and Si-DLC in pressurized high-temperature water.

2. METHODOLOGY

Figure 1 shows a schematic diagram of a friction tester using an autoclave. The specimens were installed in the autoclave, and the pins slid when the autoclave tilted. The autoclave moves like a pendulum by a motor and a crank mechanism. In this study, the abrasion tests were carried out in pressurized (10 MPa) high-temperature (23, 100, 200, 300 °C) water. Amount of dissolved oxygen was controlled by bubbling purified

water with nitrogen gas and oxygen gas (dissolved oxygen $D=0.5 \times 10^{-6}$ mg/mm³, 40×10^{-6} mg/mm³ respectively). On pin specimens, a-C:H or Si-DLC film was formed on the tip of a SCM420 steel pin with a curvature radius of 1 mm. In the test, three pins were attached to one fixing jig, and three points contact the mating material surface. The contact point load of the three DLC pins was 0.056 N/each, and the average surface pressure was 329 MPa, and the sliding distance was 20 m.

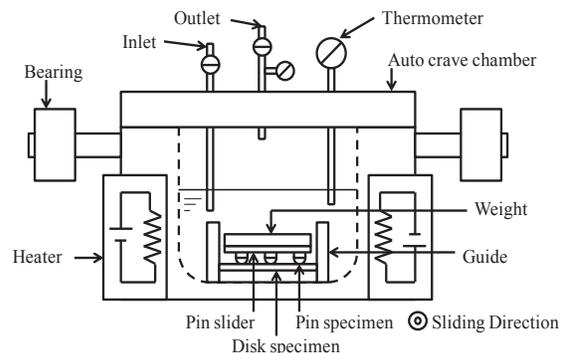


Figure 1 Automotive friction tester.

3. RESULTS AND DISCUSSION

Figure 2 shows the specific wear rate of DLC films in pressurized high-temperature water. As the temperature was changed from 23 °C to 300 °C under $D=40 \times 10^{-6}$ mg/mm³, the specific wear rate of the a-C:H increased about 26.2 times. On the other hand, the increase of Si-DLC was about 2.4 times. In addition, when the temperature was changed from 23 °C to 300 °C under $D=0.5 \times 10^{-6}$ mg/mm³, the specific wear rate of the a-C:H and the Si-DLC remained at 3.7 times and 2.4 times respectively. It was suggested that the wear resistance of the a-C:H is remarkably deteriorated in the high oxidation environment and the Si-DLC maintained wear resistance even in the oxidizing environment. Figure 3 shows the relationship between specific wear rate and inverse of AFM scratch hardness [4] along with the previous study [2], and this shows that wear is governed by the hardness of DLC. Figure 4 shows the results of Raman analysis of DLC film after friction test. As the temperature is changed from 23 °C to 300 °C, as for a-C:H film, the I_D / I_G ratio of the surface increased by about 0.30 where $D=40 \times 10^{-6}$ mg/mm³, and about 0.14 where $D=0.5 \times 10^{-6}$ mg/mm³. It is suggested that the structural change occurred due to an increase in dissolved

oxygen amount. On the other hand, in the Si-DLC film, regardless of the amount of dissolved oxygen, the increase in the I_D/I_G ratio was only about 0.05, and almost no structural change occurred. In other words, it was suggested that the high abrasion resistance of the Si-DLC in the high oxidation environment is caused by the suppression of the structural change of the DLC film surface by the addition of Si. In order to investigate the reasons for these phenomena, Si peak analysis on Si-DLC after friction test was carried out by XPS. As seen in Figure 5, under $D=40 \times 10^{-6} \text{ mg/mm}^3$, as the temperature is changed from 23 °C to 300 °C, the ratio of the Si-C bonds to the entire bonding containing Si decreased from 0.72 to 0.51, whereas the Si-O bond ratio increased from 0.20 to 0.43. From this result, it can be seen that silicon atoms originally bonded to carbon atoms are recombined with oxygen diffused to the film surface in a high oxidizing environment. The characteristic of Si-DLC may decrease carbon atom restructuring and oxidation which remain Si-DLC wear resistance in high oxidation environment.

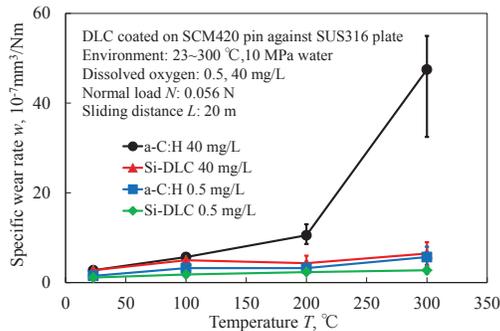


Figure 2 Specific wear rate of DLC films in pressurized high-temperature water.

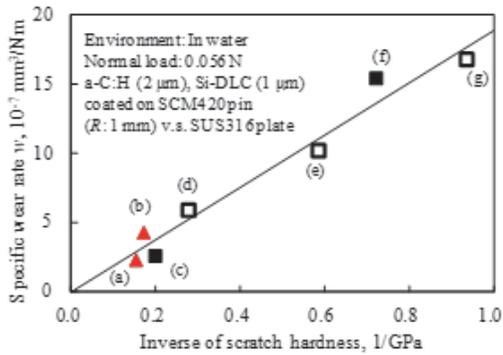


Figure 4 Relationship between specific wear rate and inverse of AFM scratch hardness: (a) Si-DLC, 200 °C, $D=0.5 \times 10^{-6} \text{ mg/mm}^3$, (b) Si-DLC, 200 °C, $D=40 \times 10^{-6} \text{ mg/mm}^3$, (c) a-C:H, 200 °C, $D=0.5 \times 10^{-6} \text{ mg/mm}^3$, (d) a-C:H, 300 °C, $D=0.7 \times 10^{-6} \text{ mg/mm}^3$, (e) a-C:H, 300 °C, $D=16.3 \times 10^{-6} \text{ mg/mm}^3$, (f) a-C:H, 200 °C, $D=40 \times 10^{-6} \text{ mg/mm}^3$, (g) a-C:H, 300 °C, $D=48 \times 10^{-6} \text{ mg/mm}^3$.

4. CONCLUSIONS

(a) As the temperature was changed from 23 °C to 300 °C under $D=40 \times 10^{-6} \text{ mg/mm}^3$, the specific wear rate of the a-C:H increased by 26.2 times, whereas that of Si-DLC was 2.4 times.

(b) As the temperature was changed from 23 °C to 300 °C under $D=40 \times 10^{-6} \text{ mg/mm}^3$ the I_D/I_G ratio of the a-C:H was increased by about 0.30 under $D=40 \times 10^{-6} \text{ mg/mm}^3$, and by 0.14 under $D=0.5 \times 10^{-6} \text{ mg/mm}^3$, whereas that of Si-DLC increased by 0.05 regardless of the oxygen.

(c) As the temperature was changed from 23 °C to 300 °C under $D=40 \times 10^{-6} \text{ mg/mm}^3$, the ratio of Si-C bonds to the entire bonding containing Si decreased from 0.72 to 0.51, whereas the ratio of Si-O bonds was 0.20 to 0.43.

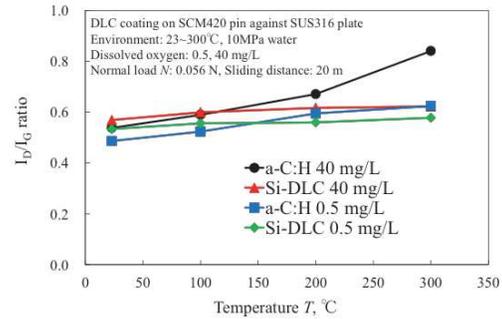


Figure 4 Raman analysis of DLC films after friction.

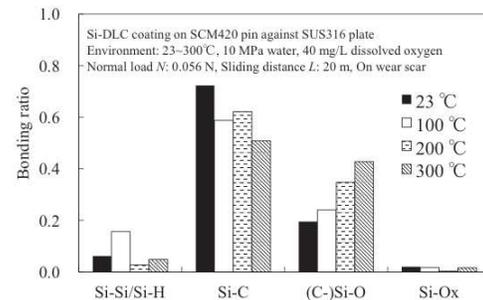


Figure 5 XPS analysis of Si-DLC films after friction.

REFERENCES

[1] Baba, K., & Hatada, R. (2002). Preparation and properties of metal containing diamond-like carbon films by magnetron plasma source ion implantation. *Surface and Coatings Technology*, 158, 373-376.

[2] Zin, M. R. B. M., Yagi, Y., Sasaki, K., Inayoshi, N., Tokoroyama, T., Umehara, N., ... & Kawara, S. (2017). The effects of temperature, pressure and dissolved oxygen concentration (DO) in water on the wear of the hydrogenated diamond-like carbon (HDLC) at high temperature and pressurized water. *Tribology International*, 109, 48-57.

[3] Choi, J., Nakao, S., Miyagawa, S., Ikeyama, M., & Miyagawa, Y. (2007). The effects of Si incorporation on the thermal and tribological properties of DLC films deposited by PBII&D with bipolar pulses. *Surface and Coatings Technology*, 201(19-20), 8357-8361.

[4] Kimura, N., Tsukiyama, Y., Tokoroyama, T., & Umehara, N. (2010). Evaluation of mechanical properties of the superficial layer of CN_x with ultra low friction in N₂ gas. *Transactions of the Japan Society of Mechanical Engineers, Series C*, 76(772), 612-617.