

論文の内容の要旨

論文題目 Structural and Environmental Dependence of Superlubricity
in Ion Vapor Deposited Hydrogenated Amorphous Carbon Films
(イオン化蒸着法により作製した水素化アモルファス炭素膜の超潤滑特性とその構造および環境依存性)

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Superlubricity, a near-frictionless lubrication state, is of high significance for its potential industrial applications since friction and wear is one of the major causes for energy loss and equipment failure during industrial actives. As a solid lubricant, amorphous carbon (a-C) or diamond-like carbon (DLC) film has attracted special attention owing to their superior anti-friction properties. Hydrogenated amorphous carbon (a-C:H) is particularly promising for achieving this superlubric state, for instance, superlow friction in dry inert gas. However, the lubrication mechanism underlying this exceptional phenomenon is not well understood. Moreover, it is still a great challenge to achieve superlubricity in ambient air due to the complex tribo-interactions in the presence of oxygen, water and other gaseous molecules.

The aim of this thesis is to develop a new superlubric-material system based on C-Si-H ternary phase, namely a-C:H and Si-incorporated a-C:H (a-C:H:Si) films. To realize this purpose, the ion vapor deposition (IVD) system was chosen as the synthesis method due to its accurate control of incident ion energy. First, the growth mechanism

for achieving ultrasmooth surface under energetic ions was investigated to serve as the basic knowledge for synthesizing the required films. Special attention was paid to the ion-energy-dependence of structural evolution of a-C:H:Si films. Second, the key factors controlling the superlubric behaviors of hydrogen-rich a-C:H film was systematically studied. Following this, the possibility for suppressing moisture sensitivity by Si-incorporation and the requirements for achieving superlow friction in humid air were investigated. Afterwards, the lubrication effect of hydrogen in a-C:H:Si was highlighted for the purpose to obtain superlow friction in dry N₂ and reactive H₂. Finally, a polymer-like a-C:H:Si film was proposed for its particularly superior anti-friction behaviors in multi-environments. The underlying tribo-chemical lubrication mechanisms were presented. The major achievements in this thesis were summarized as follows:

The IVD system is effective in growing ultrasmooth film surface. The roughness of the as-grown a-C:H:Si films is extremely low (~ 0.1 nm), and the derived roughness exponent and growth exponent by dynamic scaling theory are $\alpha \sim 0.51$ and $\beta \sim 0$, respectively. The extremely small growth exponent during ion dominated deposition requires the presence of energetic ion-impact induced subsurface “polishing” process. Depending on the incident ion energy, the bonding structure of a-C:H:Si films evolves from hydrogen-rich chain-developed polymer-like, to hard cross-linked diamond-like, and finally to hydrogen-deficient sp²-bonded a-C.

The hydrogen-rich (39.3 at.% H) a-C:H film shows high friction and severe wear in humid air when sliding against bare SUJ2 ball, while superlow friction could be achieved in dry N₂. The film structure, counterpart material (self-mated or bare steel ball), atmospheric environment, contact pressure (non-Amontonian behavior) and

sliding velocity are playing paramount roles in determining the friction behavior of a-C:H film. An extremely low friction coefficient of ~ 0.001 can be obtained at normal load of 10 N. Such a near-frictionless rubbing hardly results in any wear and loss of material.

Ultralow and even superlow friction is feasible for a-C:H:Si films in humid air. The surface density of silicon hydroxyl group (Si-OH), humidity level and contact pressure determine the frictional performance of a-C:H:Si films. The dissociative formation of OH-terminated surface and the orientation of molecular structure of adsorbed water is the origin of ultralow friction in humid air. At an appropriate RH level, the formation of a low shear-strength layer-like H₂O film provides effective boundary lubrication.

The hydrogen-induced structural diversity of a-C:H:Si films such as polymer-like, diamond-like or sp²-bonded a-C significantly affects the frictional behaviors. A proper range of hydrogen content in the film is required to achieve stable superlow friction in a distinct gaseous atmosphere, i.e., dry N₂, reactive H₂ or humid air. The polymer-like a-C:H:Si (31.9 at.% H) film can exhibit superlow friction ($\mu \sim 0.001$ –0.01) in various environments including humid air, inert gases (N₂ and Ar) and reactive gas (H₂), and can even maintain ultralow friction ($\mu \sim 0.084$) in a corrosive gas (O₂). The occurrence of superlubricity in a-C:H:Si films is attributed to a synergistic effect of phase transformation, surface passivation and shear localization, for example, the acquisition of an extremely low friction coefficient of ~ 0.001 in dry N₂. The present realization of superlubricity by one material such as the polymer-like a-C:H:Si film in multi-environments may open up a new pathway for designing more efficient lubricating materials in the near future.