Application of Ultrananocrystalline Diamond/Nonhydrogenated Amorphous Carbon Composite Films to Hard Coating on Cemented Carbide

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（超ナノ微結晶ダイヤモンド／水素フリーアモルファスカーボン混相膜の超硬合金へのハードコーティング）

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論 文 内 容 の 要 旨

Thesis Summary

Cemented tungsten carbide (WC-Co) is a sintered composite comprising tungsten carbide and cobalt binder. WC–Co has ever been employed as materials for cutting tools such as drill bits and end mills due to its high hardness and fracture toughness. In order to extend the lifetime of WC-Co cutting tools, they are coated with hard materials such as TiN, TiC, TiCN, TiAlN, diamond like carbon (DLC), and polycrystalline diamond. In particular, diamond and related carbon materials are the most promising coating substance due to their high hardness and excellent mold release.

Nanodiamond is a new candidate as carbon-based hard coating materials for machining nonferrous metals and alloys, owing to the superior properties such as good thermal stability, low friction, and high wear resistance. Ultrananocrystalline diamond (UNCD)/nonhydrogenated amorphous carbon (a-C) composite (UNCD/a-C), which comprises diamond crystallites with diameters of less than 10 nm and an a-C matrix, has received much attention as a new candidate for applications to hard coatings because of their following characteristics: (1) high hardness and Young's modulus, (2) smooth film surface, and (3) higher temperature stability than that of hard amorphous carbon, so-called diamond-like carbon (DLC).

In the case of the deposition of diamond and related carbon films on WC-Co substrates, the removal of Co on the surface or the insertion of buffer layers is required before the film deposition, since Co binder located at WC grain boundaries act as catalysts that facilitates graphitization, which results in a drastic degradation in the hardness and adhesion of the films.

Doping foreign elements such as Si, Cr, and B have been applied to hard a-C films for improving mechanical properties, stabilizing the chemical bonding structures of the films, and relaxing internal stresses in the films. These dopants have been considered to suppress the formation of sp^2 bonds and facilitate the formation of sp^3 bonds.

In this study, UNCD/a-C films were deposited by coaxial arc plasma deposition (CAPD) on WC-Co substrates, and the doping effects of Si, Cr, and B dopants on the film structures, mechanical, and tribological properties were investigated. In addition, the effects of inserting undoped UNCD/a-C buffer layers mainly for the purpose of suppressing from Co diffusion into the films was studied since we noticed the diffusion of Co atoms into the doped films directly deposited on the WC-Co substrates. Briefly, this study divided into six chapters as follow:

Chapter 1 presents an introduction and background of cemented carbide cutting tools, hard coating materials, and carbon-based nanomaterials with highlighting the ultrananocrystalline diamond films, as a core of this study.

Chapter 2 introduces the experimental techniques that have been employed along the study, with the explanation of physical meaning and mechanisms behind each techniques. Started from film preparation methods and ended by the structural, mechanical, and tribological characterization techniques.

Chapter 3 addresses the optimization of deposition conditions including substrate temperature and repetition rate to successfully deposit UNCD/a-C films by CAPD on WC-Co substrates as hard coatings with high hardness and high film thickness. In addition, the films were evaluated mechanically by nanoindentation test, and structurally by using the absorption edge near X-ray absorption fine structure analysis (NEXAFS), soft and hard X-ray photoemission spectroscopy (HAXPES/SXPES), X-ray powder diffraction (XRD), Raman spectroscopy, Scanning electron microscope (SEM), Secondary ion mass spectrometry (SIMS) , and Energy-dispersive X-ray spectroscopy (EDS, EDX, EDXS or XEDS).

In Chapter 4, we focused on the effects of Si and Cr incorporation on the films hardness and Young's modulus, and structure properties of UNCD/a-C hard coating. Si incorporation is evaluated based on the chemical configuration, by X-ray photoemission spectroscopic-survey and EDX measurements. Additionally, reasons of enhancing or degrading the films hardness due to the doping were discussed.

Chapter 5 figures out the effects of B doping on the structure, hardness and Young's modulus, and tribological properties of UNCD/a-C films. Increasing the B concentration in the graphite target resulted in films graphitization and degrading the films hardness and Young's modulus. By inserting an undoped UNCD/a-C buffer layer prior deposition of B-doped films, the films hardness and Young's modulus are enhanced. Reasons of enhancing or degrading the films hardness due to the B doping are presented in details. A comparative study of the friction coefficient and wear resistance of undoped, B-doped films without and with buffer layer is introduced.

Chapter 6 summarizes the results and discussions from chapter 3 to chapter 5 and looks forward to recommendations that enhance the films properties. It is thought that the deposition of UNCD/a-C hard coatings on Ti and CoCr alloy for the medical applications will be a promising research work.