SAGA Light Source

九州シンクロトロン光研究センター 県有ビームライン利用報告書

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（様式第５号）

同軸型アークプラズマ堆積法によりバイアス印加下で堆積されたナノダイヤモンド膜の化学結合構造(II)

Chemical bonding structure of nanodiamond films deposited under negative biases by coaxial arc plasma deposition (II)

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１．概要（注：結論を含めて下さい）

Ultrananocrystaline diamond (UNCD)/amorphous carbon (a-C) composite (UNCD/a-C) films were deposited on negatively biased cemented carbide (WC-Co) substrates by coaxial arc plasma deposition, and the effects of negative bias on the growth and mechanical properties of the films were studied. The negative bias voltage was applied in pulsed process at 10, 50, and 90 kHz. The 62 GPa hardness films were deposited at a thickness of 8.8 μm, which is more than an order of magnitude more than that of comparably hard diamond like carbon films deposited by arc ion plating deposition. In addition, the deposition rate evidently increased under the negative bias. This might be because the negative bias enhances the attraction of positively charged carbon species to the substrates, which results in enhanced film adhesion on the substrates and the release of film internal stresses.

２．背景と目的

Coaxial arc plasma deposition (CAPD) has been recently employed as a promising physical vapor deposition technique to fabricate artificially structured nanomaterials specially nanodiamond composite (NDC) films for hard coating applications [1]. CAPD offers many pros over chemical vapor deposition growth techniques, which has essentially employed for nanodiamond films deposition [2]. Like lower deposition temperature, higher growth rate, and enhanced control of morphology and thickness, as well as its applicability to versatile substrates without surface pretreatments or seeding process, are the main advantages of CAPD to mention. To improve the deposition ability of the CAPD system and enhance the mechanical and structural properties of the deposited films a development of coating systems was driven by controlling the deposition process. Energy of deposited species is a major factor in CAPD, which controlling the adhesion strength and film properties. The energy of the deposited species, which can be controlled by negative substrate bias, is necessary to compensate for the deeper penetration of the arriving species as their energies increase [3]. Investigate the influence of thisese parameter on the deposition process, structural, and mechanical properties of the deposited nanodiamond composite films, are the aim of this study.

３．実験内容（試料、実験方法、解析方法の説明）

UNCD/a-C films were deposited at different substrate bias frequencies using coaxial arc plasma deposition (CAPD) equbied with a graphite target. Tungsten carbide (WC-Co K-type was used as substrates. The deposition was conducted under a substrate bias voltage of −80 V and frequencies of 10, 50, and 90 kHz with a duty cycle of 60%. To investigate the influence of bias frequency on the plasma energy, a triple Langmuir probe diagnostic system was adopted to measure the electron temperature of the generated arc plasma under different bias frequencies. The effects of the substrate bias voltage on the deposition process, mechanical and structural properties of the films were investigated by nanoindentation and Raman spectroscopy, respectively.

４．実験結果と考察

Deposition process and films properties strongly depend on the plasma energy and flux of deposited carbon species. shows the 3D laser recorded surface morphology and the X-ray photoelectron spectra of the chemical surface composition of the UNCD/a-C films deposited at different substrate bias frequencies. The 3D images were discernible that the film surface has a random distribution of particles like clusters. The surface roughness indicated that all deposited films exhibited relatively smooth surfaces. The surface roughness root mean square (RMS) of the film deposited at 10 kHz was around 0.182 μm and this value gradually increase up to 0.207 μm by increasing the substrate bias frequency. Accordingly, accelerating the carbon species toward the substrate at low substrate bias frequency smoothens the surface of the deposited films. This behavior has been reported previously and it is attributed to the decreasing the shadowing effect by substrate biasing [4]. This observed light smoothing of the film surface might be due to the dissipation of the acceleration excessive energy induced by the applied negative bias and enhanced mobility of adatoms which evidently flattened the surface.

The chemical composition of the film surface was examined by X-ray photoelectron spectroscopy as shown in (d), (e) and (f). Intense C 1s and O 1s peaks are observed at binding energies of 284.5 and 532.5 eV, respectively [5]. The C 1s peaks originated from the carbon atoms of the UNCD/a-C films. The O 1s peaks are due to the adsorption of oxygen when the film surface exposed to the

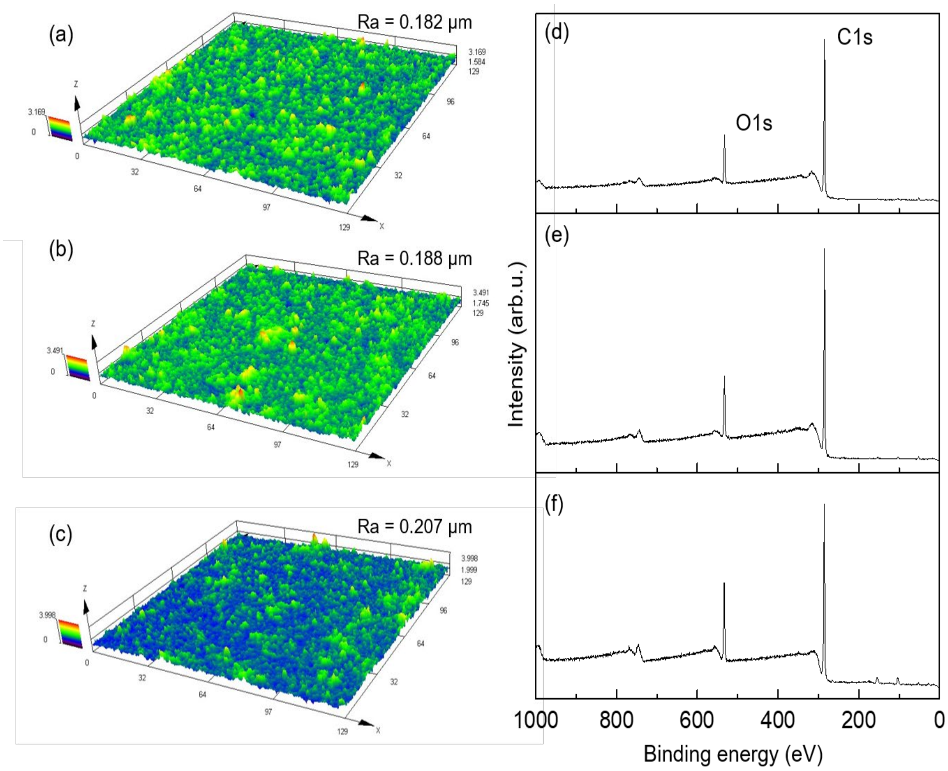

Figure 1 Three-dimensional topographic laser microscopic images of UNCD/a-C films deposited at substrate bias frequency of (a) 10, (b) 30, (c) 90 kHz with a surface elemental composition investigated by X-ray photoelectron spectroscopy in (d), (e), (f), respectively.

atmospheric oxygen after the deposition. The relative intensity of the O 1s peak gradually enhanced with increasing the substrate bias frequency as shown in Figure 1(f). The increased ratio of oxygen can be linked to the high surface roughness of the films deposited at high bias frequency. The high surface roughness means increasing the probability of vacancies and cavities capable of keeping more oxygen atoms.

５．今後の課題

The influences of bias frequency on the properties of ultrananocrystalline diamond/ amorphous carbon (UNCD/a-C) films deposited on WC-Co substrates using coaxial arc plasma deposition has investigated. The negative bias can attract more positive ions to bombard the substrate surface, leaving behind a much denser structure. The surface smoothness and overall oxidation adsorbed on the film surface were improved by applying the negative bias voltage.

６．参考文献

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[5] A. M. Ali *et al*., *Surface & Coatings Technology* **417**, 127185 (2021).

７．論文発表・特許（注：本課題に関連するこれまでの代表的な成果）

- A. M. Ali *et al*., *Diamond and Related Materials* **96**, 67 (2019).
- A. M. Ali *et al*., *Appl. Phys. Express* **13**, 065506 (2020).
- A. M. Ali *et al*., *Surface & Coatings Technology* **417**, 127185 (2021) .

８．キーワード（注：試料及び実験方法を特定する用語を２〜３）

UNCD/a-C films, Negative bias, Coaxial arc plasm deposition.

９．研究成果公開について（注：※２に記載した研究成果の公開について①と②のうち該当しない方を消してください。また、論文（査読付）発表と研究センターへの報告、または研究成果公報への原稿提出時期を記入してください。提出期限は利用年度終了後 2 年以内です。例えば 2018 年度実施課題であれば、2020 年度末（2021 年 3 月 31 日）となります。）

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①　論文（査読付）発表の報告　　　　**（報告時期：2023 年 3 月）**