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Short communication

Fabrication and characterization of HfC/SiC nanomultilayered films with enhanced mechanical properties

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ABSTRACT

A series of HfC/SiC nanomultilayered films with different SiC layer thickness were synthesized by reactive magnetron sputtering. The effect of SiC layer thickness on the microstructure and mechanical properties of HfC/SiC nanomultilayered films were studied. When SiC layer thickness was less than 0.9 nm, originally amorphous SiC layers were forced to crystallize and grow epitaxially with HfC layers, resulting in the improvement of crystallization integrality and enhancement in hardness and elastic modulus. The maximum hardness and elastic modulus could reach 36.2 GPa and 419 GPa, respectively. With further increase of SiC layer thickness, SiC layers changed back to amorphous state and damaged the epitaxial growth structure, leading to deterioration of mechanical properties.

As superhard film material, nanomultilayered films have been widely investigated in the past decades [1,2]. Nanomultilayered films are multi-laminate films formed by depositing two or more kinds of materials alternatively at nanometer-scale thickness, which exhibit not only higher hardness, but also other properties owing to formation laminated composites, such as fracture toughness, thermal stability, wear and corrosion resistance [3,4]. The conception of nanomultilayered film was firstly theoretically proposed by Koehler [5] in 1970, which suggested that solid could be strengthened by forming a laminate structure of thin layers with large shear modulus mismatch. The strengthening effect was firstly experimentally observed in Au–Ni and Cu–Pd superlattice thin films by Yang in 1977 [6], which verified Koehler's theory. Helmersson et al. [7] later reported that the strengthening effect with a maximum hardness of above 50 GPa was obtained in TiN/VN nanomultilayered film, indicating the significant research value and application prospect.

Hafnium carbide (HfC) is one of the most optimal protective coating materials due to the highest melting point (3890 °C) and high hardness [8,9]. Silicon carbide (SiC) possesses excellent wear resistance, high hardness, chemical inertness and high thermal stability [10,11], which is thus a good candidate as a component to be incorporated into the nanomultilayered system. Therefore, the combination of HfC and SiC, namely, HfC/SiC nanomultilayered film should be a promising candidate with excellent mechanical properties. In this work, a series of HfC/SiC nanomultilayered films with different SiC layer thickness were synthesized by reactive magnetron sputtering. The microstructure and mechanical properties of HfC/SiC nanomultilayered film with the controlled thickness of SiC layer were investigated. Particular attention would be paid to the correlation between microstructural evolution and mechanical properties of HfC/SiC nanomultilayered film.

The HfC/SiC nanomultilayered films were fabricated on the silicon substrates by reactive magnetron sputtering system. The HfC layers were deposited from a pure Hf metal target (at.%, 99.99%) by DC mode and the power was set at 150 W. The SiC layer was sputtered from SiC target (99.99%) by RF mode and the power was set at 80 W. When the base pressure was pumped down to 5.0×10^{-4} Pa, Ar (99.99%) and C_2H_2 (99.9% in purity) were separately introduced into the chamber. The Ar and N_2 flow rate were 40 and 1 sccm respectively. The working pressure was 0.4 Pa and substrate was heated up to 300 °C during deposition. The configuration of HfC/SiC nanomultilayered films was designed with HfC

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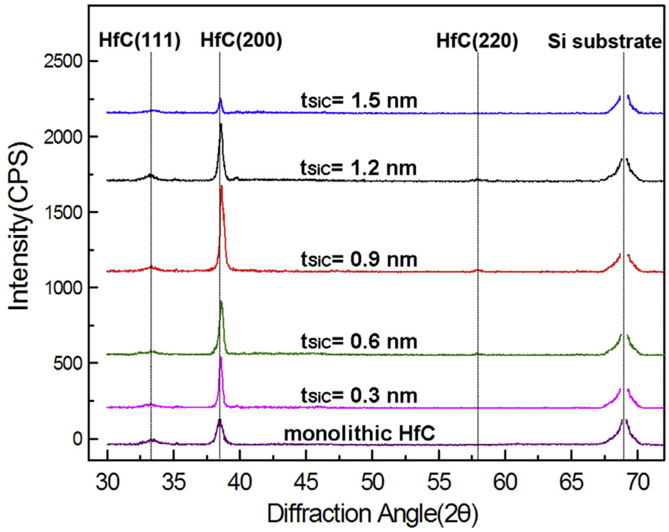

Fig. 1. XRD patterns of HfC/SiC nanomultilayered films with different SiC layer thickness as well as monolithic HfC film.

layers with a fixed thickness by depositing an HfC layer for 15 s along with variable SiC layer thickness by depositing a SiC layer for 3 s, 6 s, 9 s, 12 s and 15 s. The monolithic HfC film was also fabricated for comparison.

The microstructures of HfC/SiC nanomultilayered films were characterized by X-ray diffraction (XRD) using a Bruker D8 Advance with Cu K_a radiation, field emission scanning electron microscopy (FESEM) using a Philips Quanta FEG450, and field emission high-resolution transmission electron microscopy (HRTEM) using a Philips CM200-FEG. The mechanical properties (hardness and elastic modulus) were measured by a MTS G200 nanoindenter by using the Oliver and Pharr method [12]. Each hardness or elastic modulus value was an average of at least 16 measurements.

Fig. 1 shows the XRD patterns of monolithic HfC film and HfC/SiC nanomultilayered films with different SiC layer thickness. It can be seen that all of the films present fcc (face-centered cubic) structured HfC phase with (200) preferred orientation. No other phase except for Si substrate can be detected. According to the deposition time and deposition rate of SiC (about 0.1 nm/s) derived from the SiC film synthesized at the same condition, the thicknesses of SiC nanolayers (t_{SiC}) deposited for 3 s, 6 s, 9 s, 12 s and 15 s at this experimental condition are 0.3 nm, 0.6 nm, 0.9 nm, 1.2 nm and 1.5 nm, respectively, which have been indexed in the corresponding XRD patterns and will be verified by the following HRTEM observations.

With the initial increase of t_{SiC}, the intensity of (200) peak gradually increases, and reaches the maximum value when t_{SiC} is 0.9 nm. With further increase of t_{SiC} however, the peak intensities gradually decrease. Since SiC deposited by magnetron sputtering exhibits an amorphous feature [13], it can be deduced that, when t_{SiC} is below 0.9 nm, amorphous SiC layers are inclined to form the crystalline structure and grow epitaxially with HfC layers, which improves the crystallization integrality of nanomultilayered films. When t_{SiC} exceeds 0.9 nm, however, the improvement effect on crystallization integrality gradually disappears and the intensities of diffraction peaks accordingly decrease. Nevertheless, this deduction should be verified from HRTEM observation.

Fig. 2 shows the cross-sectional HRTEM images of HfC/SiC nanomultilayered films with t_{SiC} of 0.9 nm and 1.5 nm. From the low-magnification images of Fig. 2(a) and (d), it can be seen that the both films have the whole thickness of about 2 μm. By comparison, HfC/SiC nanomultilayered film with t_{SiC} of 0.9 nm has more evident columnar growth structure than that with t_{SiC} of 1.5 nm, which is similar with the monolithic HfC film [14]. In the high-magnification images of Fig. 2(b) and (e), it is clear that both films possess a well-defined layered structure with planar modulation layers. The dark and bright layers correspond to HfC and SiC layers, respectively. The

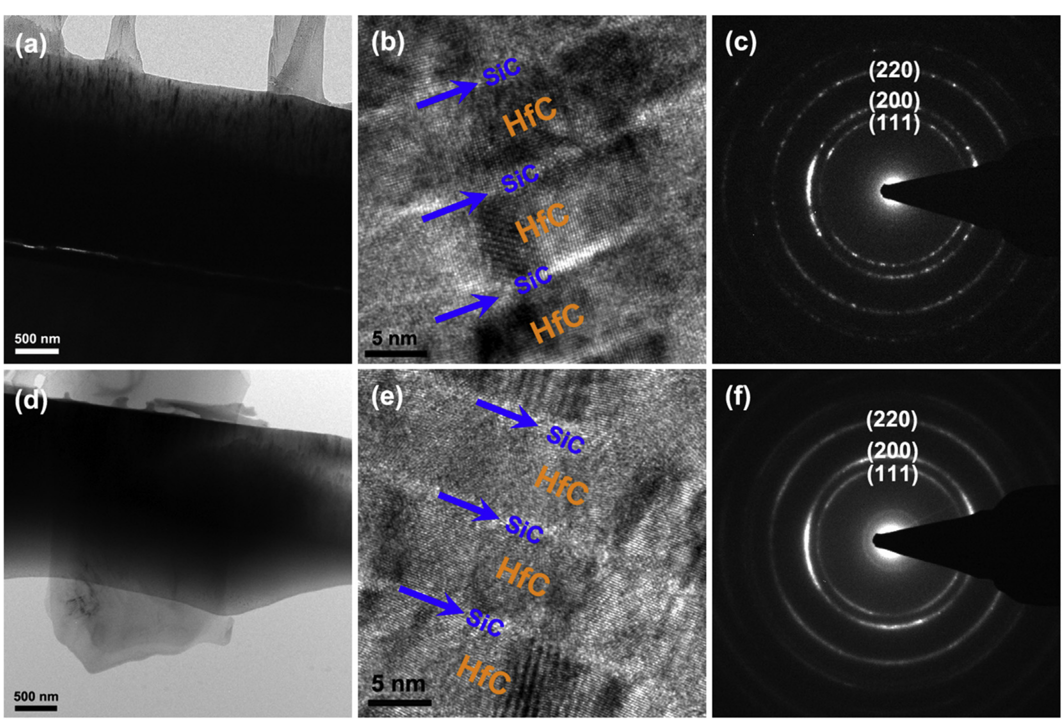

Fig. 2. Cross-sectional HRTEM images of HfC/SiC nanomultilayered films with SiC layer thickness of 0.9 nm ((a)–(c)) and 1.5 nm ((d)–(f)): (a)(d) low-magnification, (b)(e) high-magnification, (c)(f) selected area diffraction patterns.