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Surface roughening transition induced by phase transformation in hafnium nitride films

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ABSTRACT

Although surface roughening of films holds an important role in determining the final surface morphologies and properties, how to control it is not yet well explored. Here, we report that the surface roughening of hafnium nitride films varies significantly as a stoichiometry-driven phase transformation takes place. Through a combination of surface morphological measurements and simulations, we demonstrate that the variation of roughening mechanisms arises from the changes in diffusion behavior during growth. This study shows that the phase transformation can induce surface roughening transition, thus providing a new handle in controlling the morphologies and physical properties of thin films.

1. Introduction

Film growth has been a central focus in surface and materials science [1–4]. Understanding the film growth helps not only predicting the growth conditions of novel structures, but also control of the morphologies and structures for desired physical properties [5–7]. Till now, numerous studies on film growth have been carried out in various metalloid [8,9], metallic [10,11], ceramic [12,13], organic [14,15] and nanocomposite films [16,17]. In recent years, studies have been diversified to include the novel design of nano- and micro-scale structures for optical, photovoltaic, mechanical and magnetic applications [18–21]. A variety of measurement techniques [22–24], theoretical calculations [25] and simulations [9] have been adopted to study the surface evolution and related mechanisms. Many important results on film growth have been reported. For instance, Mata [26] found that the deposition temperature was an important parameter that controlled the surface roughness and scaling exponents. Dotto [27] reported that substrate materials affected significantly the mode of film growth. These studies show that surface evolution behaviors of thin films depend strongly on the deposition conditions and substrate selection.

A number of studies show that phase transformation exists in alloy and compound films when their chemical compositions or deposition conditions change [28–30]. It is believed that the diffusion mode of depositing particles changes with phase transformation because different phase structures possess distinct atomic stacking. This induces variation in surface evolution behavior of the films. Therefore, it is expected that phase structure plays an important role on film growth behaviors, especially on surface roughening process. Although the influence on film growth behaviors has been reported of deposition techniques [9], deposition conditions [26] and substrates [27], that of phase structure is still lacking.

In our previous study [28], we demonstrated that the hafnium nitride (HfN_x) films underwent a three-stage phase transformation from the rock-salt (δ-HfN) phase, through a mixture of rock-salt HfN and cubic Hf_3N_4 (δ-HfN + c-Hf_3N_4), and finally to the cubic Hf_3N_4 (c-Hf_3N_4) phase as the stoichiometry (x) increased from 1.039 to 1.334. The identification and thermodynamic mechanism of the phase transition were conducted by combining the first-principle calculations with five characterizations: Selected Area Electron Diffraction, High Resolution Transmission Electron Microscopy, Raman, Gracing Incident X-ray Diffraction and X-ray Photoelectron Spectra. The driving force of the phase transition is energy minimization. Coupling surface morphological measurements with simulations, we report, in this paper, the

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investigation of the surface roughening of HfN_x films with these three structures. It is shown that the growth of these films undergoes different surface roughening behaviors. To explain the variation in roughening mechanisms, we discuss the relationships among the phase transformation, diffusion behavior and surface morphological evolution. New insights are demonstrated through our experiments and simulations.

2. Experiment and simulation

The HfN_x films were deposited on single-crystal Si (001) wafers by radio-frequency reactive sputtering. The three different phases of the films were realized experimentally through increasing the ratios of flow rate N_2 / (Ar + N_2). The detailed preparation processing and structure characterization were reported in our previous investigation [28, 31]. To observe the surface evolution clearly, the films in same phase structure were deposited for 1, 2, 2.5, 5.0, 10, 20, 40, 80, 120 and 160 min, respectively. The surface morphology of the films was characterized using a field-emission scanning electron microscope (FE-SEM, JSM-7500F, JEOL) and atomic force microscopy (AFM, Dimension Icon, Bruker). The AFM images were scanned over an area of 2 μm × 2 μm. The thickness of the films was determined using a Dektak^3 surface profile measuring system. The thicknesses of δ-HfN films deposited for 10, 20, 40, 160 min are 70, 142, 269, 804 nm,

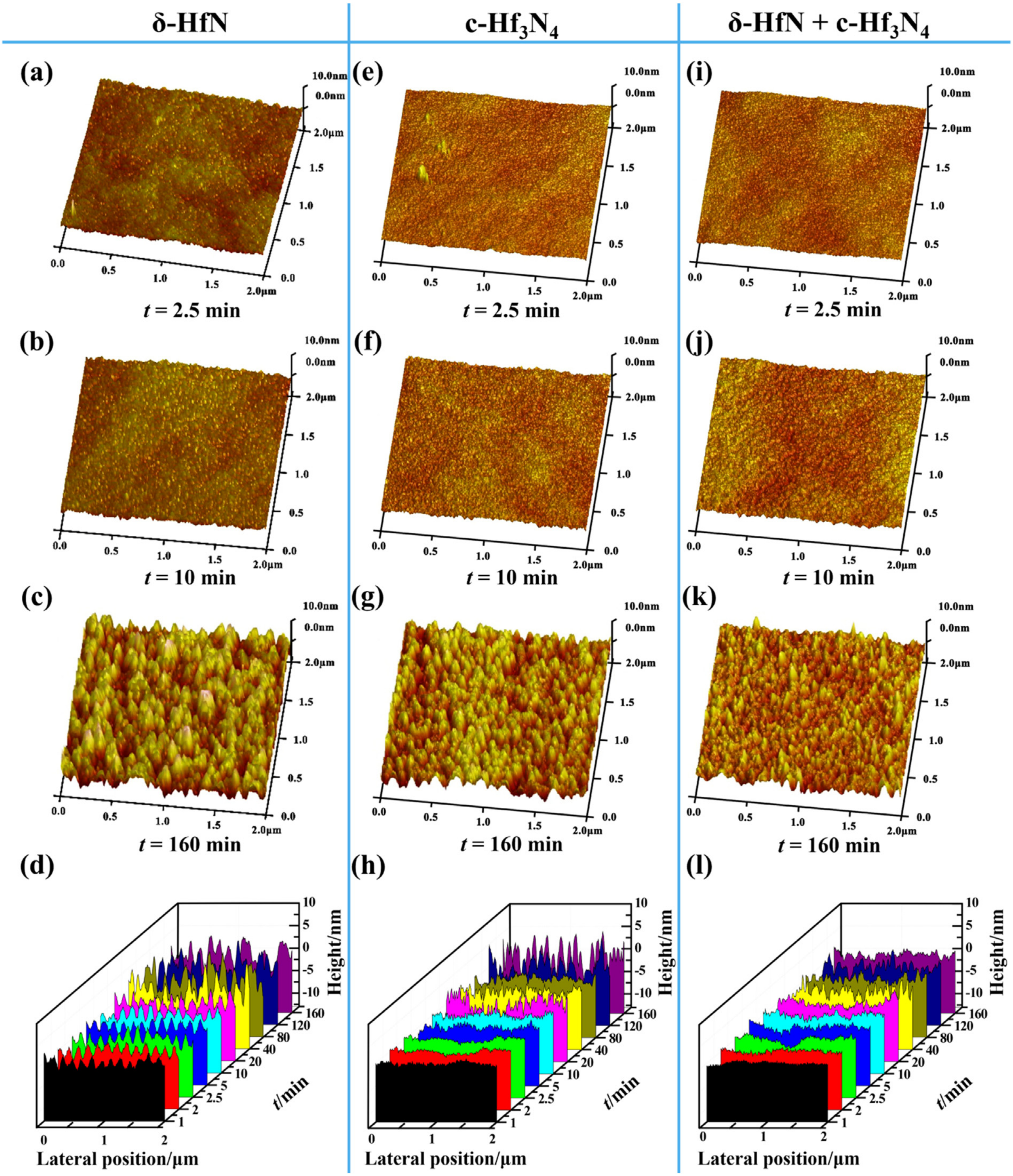

Fig. 1. 3D AFM images and the corresponding cross-sectional surface profiles of (a–d) δ-HfN, (e–h) c-Hf_3N_4 and (i–l) δ-HfN + c-Hf_3N_4 films deposited for different *t*.

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