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Crack-healing behavior induced by oxidation in SiN/SiC nanolaminated films

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ABSTRACT

The crack-healing behavior of SiN/SiC nanolaminated thin films in a high-temperature environment was investigated. Laminated films with a thickness of 1 μm were fabricated on a silicon substrate by ion-beam-assisted deposition. The number of layers was fixed to four, and the bilayer ratio of SiN to SiC was set to either 1 or 3. Cracked samples were heated in an air atmosphere at 600–1200 °C. In the case of the SiN/SiC nanolaminated film, the crack was perfectly filled with the oxide by heating at 1000 °C, whereas the crack of the SiN film was not healed. Moreover, the filled crack length of the SiN/SiC laminated film with a bilayer ratio of 1 was longer than that of the same type of film with a bilayer ratio of 3. These results suggest that inserting SiC layers in SiN films may confer crack-healing ability to SiN thin films. Moreover, the influence of heating on crack-healing was investigated systematically. Crack-healing was improved with increasing heating temperature and time.

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1. Introduction

Ceramic thin films are key materials in micro-electromechanical systems (MEMS) such as sensors and micronozzles. Ceramic thin films such as silicon nitride (Si_3N_4) and alumina (Al_2O_3) are superior to metal films in terms of strength, heat resistance, and corrosion resistance. However, the sensitivity to defects is extremely high in ceramic thin films, and the fracture toughness is low compared to that of metals. Defects formed during deposition and etching processes significantly affect the strength and life of MEMS. For MEMS used in high-temperature environments, ceramic thin films must be made more reliable.

One effective route to improving the strength and reliability of thin films is to provide a self-healing ability to thin-film materials. In self-healing materials, damage itself triggers the healing process. Self-healing materials offer a wide range of possibilities, especially for applications in which long-term reliability is required. Various bulk self-healing materials such as polymers, concrete, composites, and coatings have been developed [1–6]. However, there are very few reports on self-healing micro-materials. If a self-healing ability can be conferred to ceramic thin films, then minor defects can be tolerated and cracks initiated from the defects can be healed before the entire system breaks down while still in service.

To realize self-healing micro-material for high-temperature applications, the self-healing mechanism in bulk ceramic materials must be understood. Some bulk composite ceramics used for high-temperature components exhibit self-crack-healing behavior at high temperature [7–13]. Some composites of this type include Si_3N_4, Al_2O_3 and mullite ($Al_6O_{13}Si_2$), reinforced by silicon carbide (SiC) nanoparticles. When a cracked composite ceramic is heated to a temperature above 800 °C in air, the SiC particles exposed on a crack plane thermochemically react to form silicon dioxide (SiO_2). As this reaction proceeds, the crack is filled with SiO_2 because the molar volume of SiO_2 is larger than that of SiC. Therefore, the additional SiC particles play an important role in healing cracks in composite ceramics reinforced by SiC.

Based on the results of the abovementioned reports, carbide composite thin films may also exhibit a self-crack-healing ability at high temperatures. The self-crack-healing behavior of Si_3N_4/SiC composite ceramics depends on the distribution and size of the SiC particles [12]. However, it is difficult for the distribution and size of the SiC particles to be controlled during thin film deposition. A nanolaminated thin film can overcome this difficulty and has the following advantages: 1) an increase in crack growth resistance due to the need for cracks to penetrate the layer interface and 2) flexibility in tailoring film properties by adjusting the structural parameters of laminates, such as the bilayer ratio and number of layers.

This study aimed to investigate the crack-healing behavior of SiN/SiC nanolaminated thin films. SiN/SiC nanolaminated films with different laminated structures were fabricated by ion-beam-assisted deposition (IBAD). Cracked samples were heated in air under various heating

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conditions. Heated samples were observed using field emission scanning electron microscopy (FE-SEM), a focused-ion-beam system (FIB), and Auger electron spectroscopy (AES) to analyze the crack-healing behavior in detail.

2. Experimental procedures

2.1. Sample fabrication

The SiN/SiC nanolaminated films were deposited on Si(100) substrates by IBAD [14]. Fig. 1 shows a schematic illustration of the IBAD system (Nissin Ion Equipment Co., Ltd.) used in this study. The substrates were cleaned by successive rinsing in ultrasonic bath of acetone. After cleaning, the Si substrates were fixed on a water-cooled holder. After setting the substrates, the chamber was evacuated to a base pressure of 3×10^{-4} Pa. Prior to deposition, the substrates were sputtered using a nitrogen-ion beam at an accelerating voltage of 2 keV for 10 min.

The SiN/SiC nanolaminated films were fabricated with alternating layers of SiN and SiC. The deposition conditions for the layers are shown in Table 1. The SiN layer was obtained by an electron-beam evaporation of silicon (purity: 99.99%) and simultaneous bombardment by a nitrogen-ion beam. The SiC layer was deposited by the electron-beam evaporation of silicon and simultaneous bombardment by an argon-ion beam under an ethylene atmosphere (purity: 99.8%) at a partial pressure of 2.0×10^{-2} Pa [15]. The fabricated nanolaminated films consisted of four layers with the top layer being SiN. The bilayer thickness was fixed to 500 nm, and the bilayer ratio of SiN to SiC was set to either 1 or 3. Table 2 shows the film structures. The label "B" indicates the bilayer ratio of SiN to SiC. That is, the bilayer ratios of the SiN/SiC-B1 and SiN/SiC-B3 films were 1 and 3 respectively. SiN and SiC monolayer films were also fabricated to compare their properties with those of the SiN/SiC nanolaminated films. The total thickness of all the films was 0.9–1.2 μm.

2.2. Observation and analysis

The surface morphology and cross-section of each film were observed by FE-SEM (JEOL Ltd., JSM7001). The crystallographic structure of the coatings was investigated by X-ray diffraction (Rigaku, RINT2000) using Cu Kα radiation (a tube voltage of 40 kV, a tube current of 40 mA). The diffraction was measured by 2θ method (the incident angle of 5°). The chemical composition of the films was analyzed using an X-ray photoelectron spectroscopy (XPS, ULVAC PHI, Inc., PHI5500VersaProbe). The X-ray source used was a monochrome Al Kα with photoelectron energy of 1486.6 eV. The power was 25 W and the voltage was 15 kV. The emitted area was roughly a square of 0.5 × 0.5 mm. The standard take-off angle used was 45°. The multiplex scans for the interesting peaks were made with pass energy of 23.5 eV. The average composition was taken from five locations. An Auger electron spectroscopy (AES, JEOL Ltd., JAMP-9500 F) was used to observe the distribution of the oxides formed by crack healing. SEM and AES elemental map imaging was carried out using a 15 kV primary electron beam at a probe current of 10 nA. The images were obtained at a magnification of 3000. The elemental maps were constructed using the ratio of net peak height to background data of Auger spectra. Before performing XPS and AES, Ar-ion beam sputtering at an accelerating voltage of 3 keV was conducted for 10 s to remove any contamination.

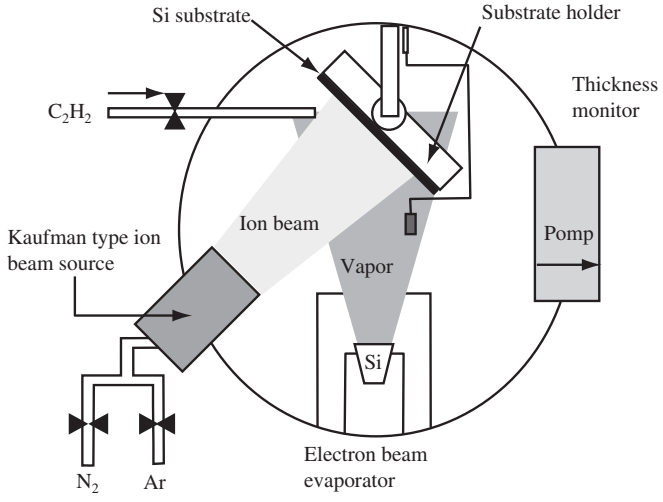

Fig. 1. Schematic illustration of ion beam assisted deposition system.

Table 1
Deposition conditions of SiN and SiC layers.

	SiN	SiC
Arc voltage (V)	80	
Ion beam	Nitrogen	Argon
Gas flow rate (sccm)	4.0	1.5
Acceleration voltage (keV)	2.0	0.3
Deceleration voltage (keV)	0.3	1.0
Acceleration current (mA)	14.0	15.0
Atmosphere gas	–	Acetylene
Vapor rate (nm/s)	0.2	0.1

2.3. Evaluation of mechanical properties

The nanohardness, H, and the elastic modulus, E, were measured using a nano-indentation system (ELIONIX, Inc., ENT-1100a) with a Berkovich diamond indenter at a maximum applied load of 10 mN. The indentation depth was below 100 nm. The tip shape of the indenter was corrected using a method proposed by Oliver and Pharr [16]. The tests were conducted 10 times for each film. A micro-Vickers hardness tester was then used to evaluate the fracture toughness, K_{Ic}, of the coatings based on the following equation [17]:

$$K_{Ic} = \delta \sqrt{\frac{E}{H}} \frac{P}{c^{3/2}} \quad (1)$$

where P is the applied indentation load and δ is an indenter geometry constant equal to 0.016 for a Vickers diamond pyramid indenter. E, H, and c are the elastic modulus, hardness, and radial crack length of the film, respectively. The indentation load was set to 2.0 N. The radial crack length was measured using FE-SEM.

2.4. Crack-healing test

A small artificial crack was introduced using a Vickers micro-indenter at a load of 2.0 N. Fig. 2 shows an example of the indentation and crack observed by FE-SEM. Radial cracks initiated from the corner of the indentation. The cracked films were heated using an electric furnace in an air atmosphere. It has been reported that crack-healing behavior is dependent on various environmental factors such as heating temperature, heating time, and atmosphere [11]. In this study, the crack-healing behavior of the nanolaminated films was systematically

Table 2
Laminated structures of SiN/SiC nano-laminated films.

Film name	Number of layers	Bilayer period (nm)	Bilayer ratio	Layer thickness (nm)		Total thickness (nm)
				SiN	SiC	
SiN	1	–	–	1000	–	1000
SiN/SiC-B1	4	500	1	250	250	1000
SiN/SiC-B3	4	500	3	375	125	1000

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