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Effects of laser power on the growth of polycrystalline AlN films by laser chemical vapor deposition method

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ABSTRACT

Polycrystalline aluminum nitride (AlN) films were prepared by laser chemical vapor deposition method using aluminum acetylacetonate and ammonia as source materials. The effects of deposition conditions on the crystal phase, composition and microstructure were investigated. Polycrystalline AlN films were prepared at a laser power above 100 W and a deposition temperature above 803 K. The microstructure of AlN film changed from aggregated grains to faceted grains to pyramidal grains with increasing laser power and with decreasing total pressure.

1. Introduction

Aluminum nitride (AlN) polycrystalline film is a promising candidate for fabrication of micro-ring optical resonators, surface acoustic wave (SAW) devices, and piezoelectric devices due to its small thermo-optic coefficient, high SAW velocity and excellent piezoelectivity [1–3]. AlN films can also be used as protective coatings for high temperature oxidization [4]. Several characteristics of AlN film such as thermo-optic coefficient, thermal conductivity and oxidation resistance are known to be strongly dependent on the microstructure and the crystal phase [2,4]. Thus, a good controllability of film microstructure and orientation is required for high performance AlN film. To date, AlN films have been prepared by various processes, including physical vapor deposition [5], sputtering [6], and atomic layer deposition [7].

Chemical vapor deposition (CVD) is known to be suitable for preparing thick AlN films with a controlled microstructure [8–11]. Halides, in particular chlorides, have widely been employed in conventional CVD. However, corrosive by-products often corrode the deposition setup, substrates and produced films. Since chloride precursors are rather thermally stable, a high deposition temperature is needed to prepare well-crystallized nitride films. This limits the application of low melting-point substrates. Metal–organic precursor is preferable for low temperature deposition. Aluminum acetylacetonate ($Al(acac)_3$) has been widely used in CVD as an Al precursor for preparing Al_2O_3 films due to its advantages of non-toxicity, low cost and good stability at room temperature [12–14]. To date, $Al(acac)_3$ has never been employed for preparing AlN film on a polycrystalline substrate because inherent aluminum–oxygen bonds are favorable for Al_2O_3 formation.

Laser irradiation facilitates a low temperature film deposition process through accelerating the chemical reactions and promoting the grain growth of deposits in CVD [15], this being termed laser CVD (LCVD). LCVD showed excellent controllability in wide-range of microstructure and orientation in the previous studies [15–17]. In the present study, by examining the effects of laser power, we report on the preparation of AlN film on yttria-stabilized zirconia (YSZ) polycrystalline substrates and investigate the effects of deposition conditions on the crystal phase, composition and microstructure of AlN films.

2. Experimental

AlN films were prepared on YSZ plates (10 mm × 10 mm × 1 mm). $Al(acac)_3$ precursor (99%) and NH_3 gas (99.9995%) were used as source materials. A cold-wall type CVD apparatus was used to prepare AlN films [15]. An InGaAlAs diode laser (wavelength = 808 nm) beam was introduced into the chamber through a quartz-glass window and was slightly expanded by lenses to about 20 mm in diameter at the substrate surface. Table 1 is a summary of the deposition parameters for preparing AlN films. The laser was operated in the continuous mode with power (P_L) ranging from 70 to 160 W. A type K thermocouple was inserted into a slot engraved from the back to a distance of 0.5 mm from the surface of the substrate to measure the deposition temperature (T_{dep}). $Al(acac)_3$ precursor was evaporated by heating at a vaporization temperature (T_{Al}) of 433 K, and its vapor was transported into the chamber using Ar carrier gas (99.99%). NH_3 gas was introduced separately into the chamber through a double-tube nozzle to react with $Al(acac)_3$. We used the same flow rate of 1.65×10^{-6} m^3s^{-1} for both Ar and NH_3 gases, which was measured by mass flow controller. The total pressure (P_{tot}) in the chamber was controlled to be 0.2 to 0.9 kPa. The feed pipes and the nozzle were

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Table 1
Deposition conditions for AlN films on YSZ substrates by LCVD.

Laser power, P_L	70–160 W
Deposition temperature, T_{dep}	589–903 K
Total pressure in chamber, P_{tot}	0.2–0.9 kPa
$Al(acac)_3$ precursor temperature, T_{Al}	433 K
Gas line and nozzle temperature	523 K
Flow rate of $Al(acac)_3$ carrier gas	1.65×10^{-6} m^3s^{-1}
Flow rate of NH_3 gas	1.65×10^{-6} m^3s^{-1}

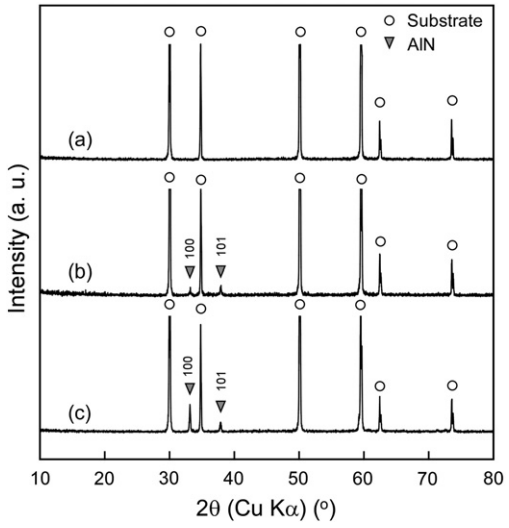

Fig. 1. XRD patterns of AlN films prepared on YSZ substrate at $T_{Al} = 433$ K and $P_{tot} = 0.2$ kPa: (a) $P_L = 70$ W ($T_{dep} = 589$ K), (b) $P_L = 100$ W ($T_{dep} = 803$ K), (c) $P_L = 160$ W ($T_{dep} = 903$ K).

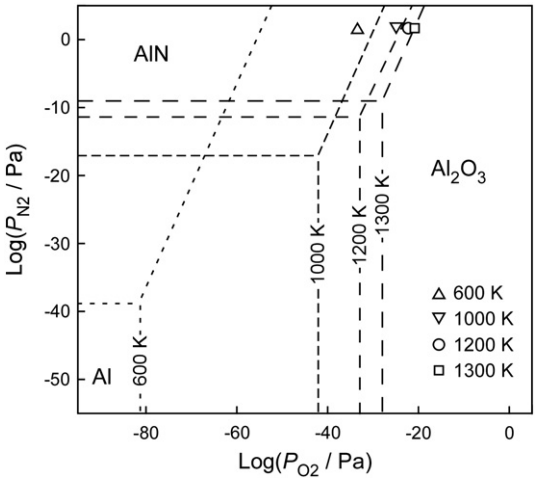

Fig. 3. Thermodynamically calculated N_2–O_2 partial pressures for the preparation of AlN films in the present study at $P_{tot} = 0.2$ kPa and $T_{dep} = 600$ to 1300 K. Potential diagrams of Al–O–N system at 600 to 1300 K are also depicted as dashed lines.

maintained at a temperature of 523 K to prevent condensation of the precursor vapor.

The crystal phases were examined by X-ray diffraction (θ–2θ scan) with Cu Kα radiation (XRD; Rigaku, RAD-2C), and the chemical bonding was analyzed by X-ray photoelectron spectroscopy with Al Kα radiation (XPS; Shimadzu Kratos, Axis-Ultra DLD). The XPS spectra were deconvoluted by Gaussian (70%)–Lorentzian (30%) functions with a Shirley baseline correction using CasaXPS software. The morphology and thickness of AlN films were characterized by using a scanning electron microscope (Hitachi, S-3100H). The deposition rate (R_{dep}) of AlN film was calculated from the thickness of AlN film and deposition time.

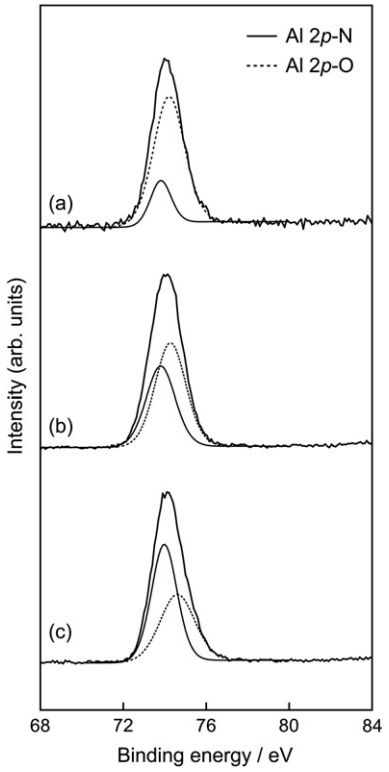

Fig. 2. XPS spectrum of the Al 2*p* photoelectron peaks for AlN films prepared at $P_{tot} = 0.2$ kPa: (a) $P_L = 70$ W ($T_{dep} = 589$ K), (b) $P_L = 100$ W ($T_{dep} = 803$ K), (c) $P_L = 120$ W ($T_{dep} = 875$ K), (d) $P_L = 160$ W ($T_{dep} = 903$ K).

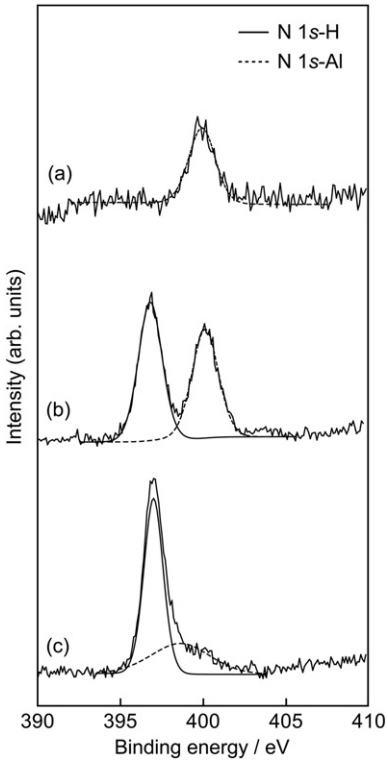

Fig. 4. XPS spectrum of the N 1*s* photoelectron peaks for AlN films prepared at $P_{tot} = 0.2$ kPa: (a) $P_L = 70$ W ($T_{dep} = 589$ K), (b) $P_L = 100$ W ($T_{dep} = 803$ K), (c) $P_L = 120$ W ($T_{dep} = 875$ K), (d) $P_L = 160$ W ($T_{dep} = 903$ K).