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Pulsed direct-current reactive sputtering of high Young's modulus [002] oriented aluminum nitride thin films

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A B S T R A C T

This work reports a process-parameter optimization of [002] oriented high Young's modulus aluminum nitride (AlN) thin films deposited by pulsed direct current (DC) reactive sputtering method. Properties of deposited AlN films are mainly determined by the material's microstructure and morphology, which can be modulated by the process parameters. Commonly explored process parameters of reactive sputtering of AlN are substrate temperature, process pressure, gas composition, substrate type, etc. Process pressure and nitrogen concentration have the most significant influence on the topography and morphology of the film, deposited by the pulsed DC reactive sputtering method. DC pulse parameters such as pulse frequency and pulse duty cycle also influence the properties of deposited AlN films, which have been addressed in this work. The pulse frequency of 100 kHz and a duty cycle of 80% was found to be the most suitable pulse parameters. This paper also reports the voltage and current behavior and deposition rate dependence under varying process parameters and pulsed properties. Optimization of process parameters is commonly done by characterizing the deposited AlN films' crystallinity, surface roughness, and residual stress. This work addresses the influences of process and pulse parameters on Young's modulus besides other characteristic properties of the film. The AlN films deposited with the optimized method and pulse parameters yield Young's modulus of up to 335 GPa while dropping the surface roughness to 1.2 nm.

1. Introduction

Aluminum nitride (AlN) is a group III-V compound semiconductor having a stable wurtzite structure. AlN has a large bandgap (~6.2 eV), good piezoelectric coefficient (d33 = 5.3 pm/V), very high piezoelectric Curie temperature (~1150 °C) [1], high electrical resistivity (10^{11}–10^{14} Ω cm), high thermal conductivity (320 W/m K), high sound acoustic wave velocity (>5500 m/s), and excellent chemical stability [2]. These properties make AlN an attractive material for applications covering deep ultraviolet optoelectronics, piezoelectric material for micro-electromechanical systems (MEMS), resonating components in RF-MEMS oscillators, and hard coating [2–5].

AlN thin films can be grown by various methods, including chemical vapor deposition [6,7], molecular beam epitaxy [8], pulsed laser deposition [9], and magnetron sputtering [10]. Magnetron sputtering is advantageous for being a low-temperature and low-cost process [10]. Magnetron sputtering can be operated by a radio-frequency (RF) or a pulsed direct-current (DC) power supply. Pulsed DC magnetron sputtering offers the advantage of higher deposition rates than RF [11,12]. When sputtering from a metallic aluminum (Al) target with argon and nitrogen as process gas, it is termed reactive sputtering deposition. During reactive sputtering, sputtered Al atoms react with nitrogen gas and form an AlN film on the substrate. To achieve the desired structural and morphological, optimization of deposition parameters are essential. Iqbal et al. summarized most of the work on reactive sputtering of AlN thin films done until 2018, in a review paper [10]. The influence of common process parameters such as substrate temperature, gas pressure, nitrogen concentration, substrate type, and magnetron power on the properties of DC reactive sputtered AlN films have been investigated in various studies [13–23]. Besides these common process parameters, pulse parameters such as pulse frequency and pulse duty cycle can also affect the morphology and structure of a reactively sputtered film [24].

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For instance, Cherng et al. reported a study of the effect of pulse parameters such as reverse voltage, pulse frequency, and pulse duration on the crystalline property of reactively sputtered AlN films through X-ray diffraction (XRD) analysis [24]. An increase in reverse voltage resulted in a detrimental effect on [002] orientation of the AlN film, whereas an increase in pulse frequency within the range of 10 kHz to 50 kHz improved the c-axis orientation of the AlN film [24]. In another study, Kohout et al. reported a reactive low duty cycle magnetron sputtering deposition of [002] oriented AlN films [5]. The authors optimized deposition conditions such as reactive gas flow, working pressure, average target power, substrate temperature, substrate bias, and the level of target erosion. The optimized AlN films had a hardness of 22 GPa and residual stress of around 300 MPa [5].

Despite the extensive research on AlN film processes, there has been an insufficient study on the effects of DC pulse parameters on the structure and properties of pulsed DC reactive sputtered AlN films. The pulse parameters are expected to greatly affect the resulting film structure and properties because they can modulate the average ion kinetic energy. Furthermore, Young's modulus is an important material property of a piezometric film [25]. There has been a breach of study on the influence of the process parameters on the Young's modulus of AlN films. This work reports a comprehensive study on the influence of common process parameters along with DC pulse parameters on the structure and properties as well as the Young's modulus of AlN films. Moreover, this work includes an investigation of the target voltage and discharge current behavior of the pulsed DC reactive sputtering process and their influences on the deposited film property.

2. Experiment

Silicon substrates (25.4 mm × 25.4 mm) were made by cleaving (100) silicon wafers with a diamond tip scribing pen. The substrates were then cleaned ultrasonically in acetone and methanol in sequence for ten minutes in each solvent. Then the substrates were blow-dried using a nitrogen gas gun. A sputtering deposition system (Kurt J. Lesker Company® PRO Line PVD 75) having three Torus magnetron sputtering sources and equipped with a pulsed DC power supply was used for the deposition of AlN films. Table 1 enlists the parameters of the pulsed DC sputtering process. Table 2 shows the parameters for optimizing the pulsed DC reactive sputtering. All the process parameters, i.e., substrate temperature, process gas flow rate, DC power level, DC pulse frequency, and DC pulse duty cycle, were controlled and monitored using an interface software program. DC pulse duty cycle is defined as a ratio of negative pulse duration (t_{on}) to reverse voltage duration (t_{rev}), as shown in Fig. 1, in a pulsed DC reactive sputtering. Though the target voltage is a negative potential relative to the chamber wall, the target voltage has been shown as positive in the following graphs in the results and analysis section for simplicity. Moreover, the "target voltage" will indicate the average value of the absolute voltage over a pulse cycle. The magnitude of the positive "reverse" voltage was set to be 20% of the negative "on" voltage. Resistive-electric heaters were used to heat the substrate and maintain it at the desired temperature. After preliminary temperature studies, the substrate temperature was maintained at 300 °C for all the depositions. Crystalline characterization of the AlN films was carried out using a *Rigaku SmartLab* XRD system with Cu-Kα radiation (λ = 1.5406 Å) in parallel beam - parallel slit analyzer mode. Surface topography and surface roughness (R_q) of the films were characterized using a *Hitachi AFM5100N* scanning probe microscope (SPM) with *μmasch's HQ:NSC14/Al* tips (tip radius: 8 nm, tip height: 12–18 μm, tip cone angle: 40°, resonance frequency: 160 kHz, force constant: 5 N/m) along with *Gwyddion* SPM data analysis software. While evaluating surface roughness (R_q) from the 2 μm × 2 μm SPM image, five scan lines each having lengths of 1.8 μm and a waviness cutoff wavelength of 257 nm were used. The average of those five roughnesses values, for each sample, was plotted with standard deviation error bars. A *LAwave®* (laser acoustic wave) system was used for determining Young's modulus of the AlN films [26]. Three measurements were done for each sample and the average Young's modulus was plotted with standard deviation error bars. Cross-sectional image of 1 μm thick AlN film was taken using an ultra-high resolution JEOL 7500F field emission scanning electron microscope. The unit Torr and mTorr (milliTorr) have been used as units for pressure to maintain consistency with the equipment used for the experiments, where 1 Torr is equal to 133.322 Pa.

Table 1
Parameters of the pulsed DC sputtering process.

Target	Aluminum (99.99% pure), Ø 76.2 mm
Substrate	(100) silicon (25.4 mm × 25.4 mm)
Substrate temperature	300 °C
Sputtering power	250 W (pulsed DC)
Pulse frequency	25 kHz – 300 kHz
Pulse duty cycle	70–90%
Deposition time	45–90 min
Film thickness	200–250 nm
Process gas	Argon (Ar) and Nitrogen (N_2)
Base pressure	2×10^{-7} Torr (2.6664×10^{-5} Pa)
Process pressure	1–5 mTorr (0.133–0.667 Pa)
Target to substrate distance	5.5 inches
Substrate rotation speed	10 revs/min
Temperature ramp up/down	10 K/min
Magnetron ramp up/down	*0.2 Watt/sec*

3. Results and analysis

3.1. Process pressure study

The process pressure was found to significantly influence the discharge voltage and current, as illustrated in Fig. 2, in the pulsed DC reactive sputtering of AlN. The target voltage decreased sharply as the process pressure increased from 1 mTorr to 3 mTorr. As the pressure further increases to above 3 mTorr, the voltage only varied slightly. With the pulsed DC power fixed at 250 W, the discharge current had a tendency opposite to the target voltage; the current increased with the increase in the process pressure. At lower pressure, there were fewer ions, causing lower discharge current, which resulted in a higher target voltage for a fixed power supplied to the magnetron. Fig. 3 shows the variation in deposition rates with process pressure of the pulsed DC reactive sputtering of AlN. With an increase in pressure, the deposition rates went down, which is attributed to background gas scattering.

The process pressure had a significant influence on the topography of the deposited AlN films. Fig. 4 shows the roughness values of AlN films deposited at various process pressures. The roughness of the films continued to decrease with lowering the deposition pressure. AlN films deposited at 1 mTorr had root-mean-square (RMS) roughness of only 1.8 nm. Any further lowering of the chamber pressure was not a viable choice, as the plasma became unstable at pressure levels lower than 1 mTorr while maintaining the pulsed DC power at 250 W. At lower pressure, the sputtered atoms experience fewer collisions before reaching the substrate. Hence they possess higher kinetic energy, enabling better surface diffusion and coalescence, which result in a smoother film surface. Fig. 5 shows the XRD patterns of AlN films deposited by pulsed DC reactive sputtering at various process pressures. The process pressure had a significant influence on the film microstructure. The observed XRD peaks at around 36° are assigned to the (002) wurtzite AlN (card # 01–070–2545) phase. Highly [002] oriented film was achieved when deposited at a pressure of 1 mTorr. At higher process pressures, e.g., 3 mTorr and 5 mTorr, the (002) peak intensity and sharpness were much lower. The full width at half maximum (FWHM) for films deposited at 1 mTorr, 3 mTorr, and 5 mTorr process pressure were 0.40°, 0.47°, and 0.49°, respectively. Increased FWHM values and decreased intensities, with an increase in pressure, were referred to decreased [002] orientation of the film. AlN film deposited at 5 mTorr pressure showed a weak

Table 2
Parameters for process optimization of the pulsed DC reactive sputtering.

Study parameter	Pressure (mTorr)	N_2 gas concentration (%)	Pulse frequency (kHz)	Pulse duty cycle (%)
Process pressure	varying: 1, 3, 5 (0.133, 0.4, 0.667 Pa)	50	50	75
Process gas composition	1	varying: 50, 75, 100	50	75
Pulse frequency	1	100	varying: 25, 50, 100, 200, 300	80
Pulse duty cycle	1	100	100	varying: 70, 80, 90

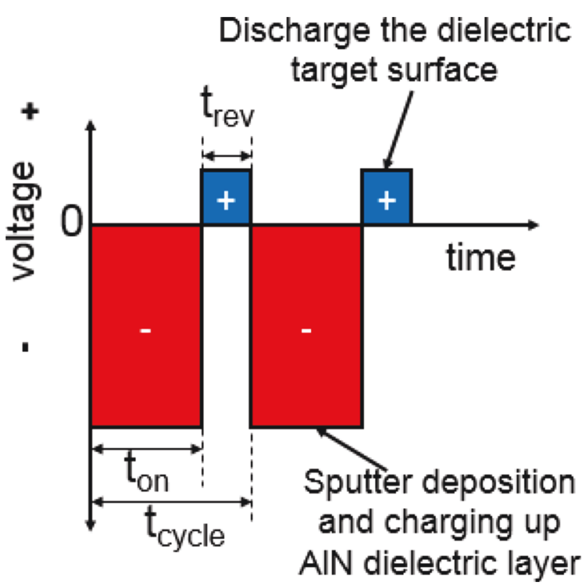

Fig. 1. Asymmetric bipolar voltage waveform used in a typical pulsed DC sputtering method.

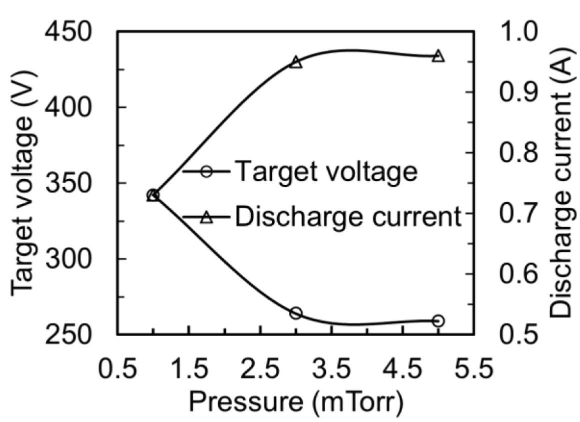

Fig. 2. Variation of target voltage and discharge current with process pressure of the pulsed DC reactive sputtering of AlN.

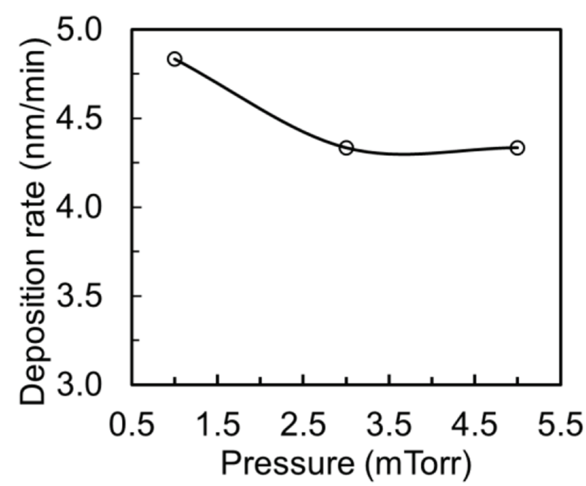

Fig. 3. Variation in deposition rates with process pressure of the pulsed DC reactive sputtering of AlN.

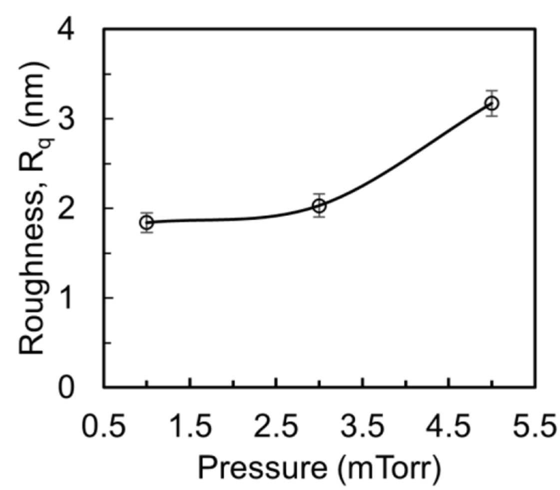

Fig. 4. Roughness with standard deviation errors of AlN films deposited by pulsed DC reactive sputtering at various process pressures.

peak at 37.8°, referring to a (101) peak, according to the AlN phase from the joint committee of powder diffraction, card # 01–070–2545. With the reduction in the process pressure, the (101) peak continued to decrease and vanished at 1 mTorr. At low process pressure, the deposited atoms have high kinetic energy, which could reorder and reassemble on the substrate surface to crystallize in the [002] direction.

The process pressure also influenced Young's modulus of the pulsed DC reactive sputtered AlN films. Fig. 6 shows Young's modulus values of AlN films deposited by DC reactive sputtering at various process pressures. Films deposited at lower process pressure exhibited higher Young's modulus. This result is very much correlative with XRD data. Films deposited at lower pressure showed better crystallinity and [002] orientation, resulting in denser films with higher Young's modulus. AlN film deposited at 1 mTorr had Young's modulus of 312 GPa. Moreover, the discharge voltage had a secondary effect on the film crystallinity, surface roughness, and Young's modulus. Higher target voltage leads to more energetic depositing species, thus denser film with higher Young's modulus, better crystallinity, and lower surface roughness. As a result, the Young's modulus (Fig. 6) and XRD peaks sharpness (Fig. 5) follow the variation in the target voltage (Fig. 2), while the surface roughness (Fig. 4) follows an opposite trend to the target voltage.

3.2. *Process gas composition study*

The Ar-N_2 process gas composition influenced the discharge voltage-current characteristics of the pulsed DC reactive sputtering of AlN. Fig. 7 shows the variation of target voltage and discharge current with the

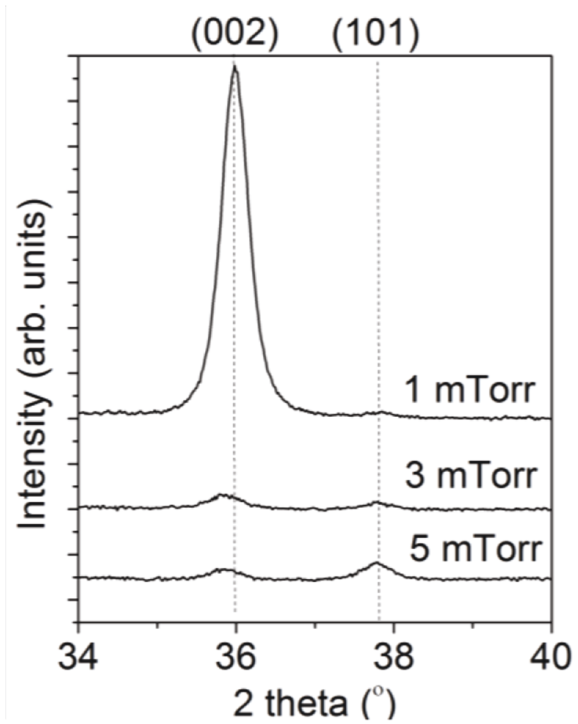

Fig. 5. XRD pattern of AlN films deposited by pulsed DC reactive sputtering at various process pressures. FWHM for films deposited at 1 mTorr, 3 mTorr, and 5 mTorr process pressure were 0.40°, 0.47°, and 0.49°, respectively.

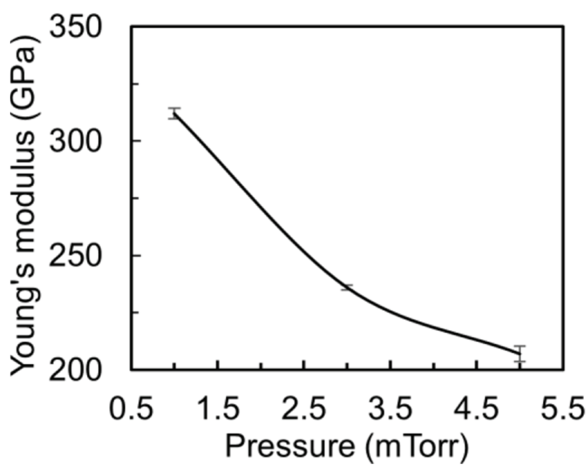

Fig. 6. Young's modulus with standard deviation error of AlN films deposited by DC reactive sputtering at various pressures.

nitrogen concentration of the pulsed DC reactive sputtering of AlN. The discharge current decreased slowly with the increase in nitrogen gas concentration from 50% to 75% but the current decreased rapidly as the nitrogen gas concentration increased from 75% to 100%. As nitrogen ionizes less than argon, the plasma density would be reduced with higher nitrogen concentration, which resulted in a lower discharge current [27,28]. With fixed target power, the target voltage curve varied in an opposite trend of the discharge current. Fig. 8 shows the variation of deposition rate with nitrogen concentration in the pulsed DC reactive sputtering of AlN. Both the variation in target voltage and the presence of lighter N^{+}sputtering species affected the deposition rate. The deposition rate plummeted as the nitrogen concentration was increased from 50% to 75%. As seen in Fig. 7, when the nitrogen concentration varied from 50% to 75%, the target voltage was almost the same. However,

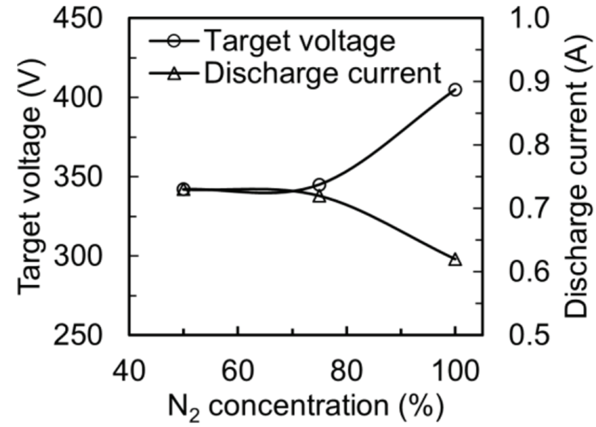

Fig. 7. Variation of target voltage and discharge current with the nitrogen concentration of the pulsed DC reactive sputtering of AlN.

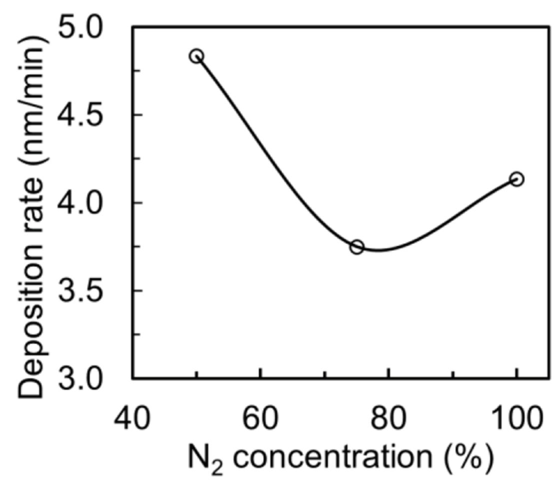

Fig. 8. Variation of deposition rate with the nitrogen concentration of the pulsed DC reactive sputtering of AlN.

with 75% nitrogen concentration, the deposition rate was much lower than that at 50% nitrogen, as nitrogen is much lighter than argon, hence resulting in a much lower sputtering yield and deposition rate. As nitrogen concentration brought from 75% up to 100%, the significant increase in target voltage increased the ions' kinetic energy and higher sputtering yield. The higher sputtering yield could increase the deposition rate slightly at 100% nitrogen concentration.

It is necessary to add nitrogen gas to the process gas to form AlN films from a metallic aluminum target, so sputtered aluminum could react with nitrogen and form aluminum nitride. Preliminary studies on the nitrogen gas concentration revealed that at least 50% nitrogen was needed to deposit stoichiometric films with crystal growth in [002] orientation. Fig. 9 shows the XRD pattern of AlN films deposited by DC reactive sputtering with various nitrogen gas concentrations. FWHM for films deposited with 50%, 75%, and 100% nitrogen gas were 0.4044°, 0.3852°, and 0.3240°, respectively. Continued decrease in FWHM values with the increase in nitrogen gas concentration in process gas mixture refers to better [002] crystal orientation. With an increased nitrogen gas concentration, the intensity of the (002) peak increased but did not appear to influence the stoichiometry anymore. For higher nitrogen gas concentration, collision with lighter nitrogen plasma species in the background would result in less loss in the kinetic energy of the depositing molecules. Also, higher nitrogen gas concentration resulted in higher target voltage, as seen in Fig. 7, which caused a stronger bombardment of sputtering N^{+}on target and ejected energetic AlN

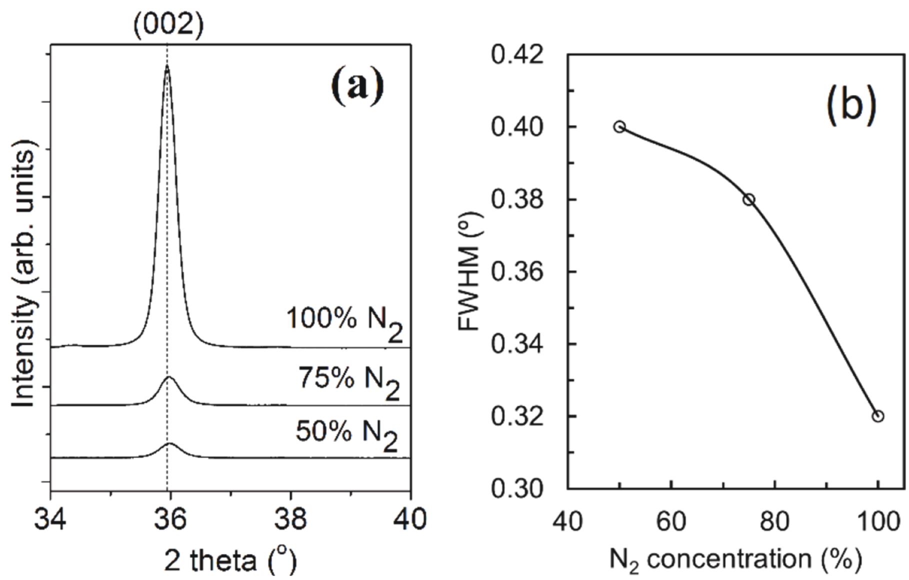

Fig. 9. (a) XRD pattern of AlN films deposited by DC reactive sputtering with various nitrogen gas concentrations. (b) FWHM for films deposited with various nitrogen gas concentrations.

molecules/species. These depositing molecules with higher energy would be favorable for better surface diffusion, thus making larger crystallite clusters and better crystallinity. The influence of N_2 gas concentration on the surface roughness of the deposited AlN films was found to be less significant. Fig. 10 shows the roughness values of the AlN films deposited with pulsed DC reactive sputtering at various nitrogen gas concentrations. The roughness of the film initially increased with the increase in nitrogen concentration from 50% to 75%, and then the roughness declined slightly as the nitrogen concentration was increased to 100%. The first increase in roughness could be attributed to the significant increase in crystallite size, which made the film's surface rougher. But a further increase in nitrogen concentration beyond 75% nitrogen gas concentration reduced the roughness slightly. This could be attributed to the significant increase in target voltage with nitrogen concentration higher than 75%, as seen in Fig. 7, which could favor densification and surface-smoothening of the film. Fig. 11 shows the Young's modulus values of AlN films deposited at various nitrogen gas concentrations. The variation of N_2 gas concentration did not appear to influence the Young's modulus of the deposited AlN films significantly.

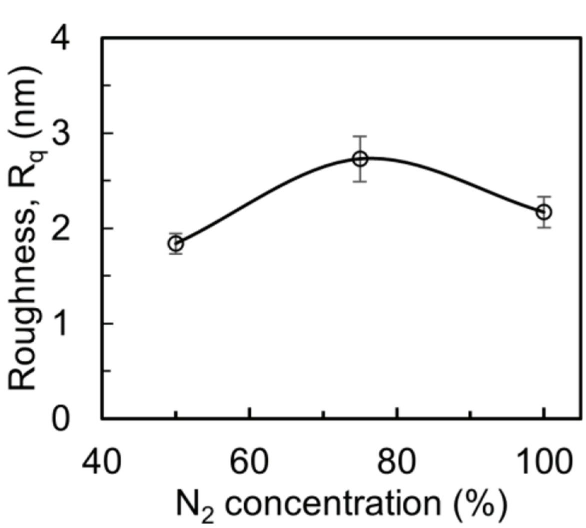

Fig. 10. Roughness values of AlN films deposited by DC reactive sputtering at various nitrogen gas concentrations.

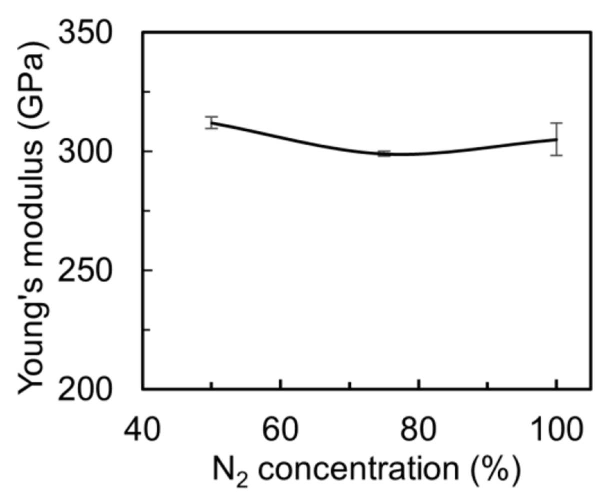

Fig. 11. Young's modulus values of AlN films deposited by DC reactive sputtering at various nitrogen gas concentrations.

The first decrease in Young's modulus from 50% to 75% nitrogen gas concentration could be attributed to the formation of larger crystallites at higher nitrogen concentrations. With larger crystallites, the grain boundaries would be more inconsistent and defective, thus lowering the effective density and Young's modulus of the film. However, with nitrogen concentrations higher than 75%, there was a substantial increase in the target voltage, as seen in Fig. 7. That increased target voltage could favor the film's densification and improve the Young's modulus of the grown film.

3.3. Pulse frequency study

Pulse frequency had little influence on the target voltage and discharge current of the pulsed DC reactive sputtering process. Fig. 12 shows the variation of target voltage and discharge current with the pulse frequency. 100–200 kHz resulted in maximum target voltage and consequently the minimum discharge current. Above 200 kHz, the reverse time (t_{rev}) is squeezed too much to completely discharge the accumulated positive charge on the poisoned or nitrided target surface during the previous "active sputtering time" or "on" time (t_{on}). This

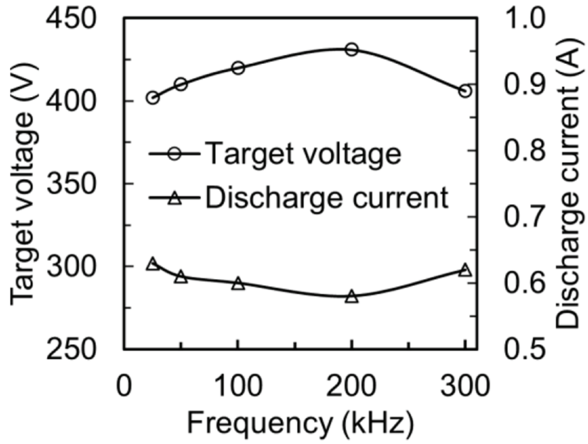

Fig. 12. Variation of target voltage and discharge current with the pulse frequency of the pulsed DC reactive sputtering of AlN.

insufficient discharging of the nitrided target surface resulted in a reduced (negative) potential of the target at a higher pulse frequency at 300 kHz. On the other hand, at a lower frequency below 100 kHz, the "on" time of the pulse is too long that the nitrided target surface accumulates too much positive charge during "on" or "active sputtering time". This higher level of positive charge accumulation causes the effective negative potential of the target to drop at lower frequencies. In the frequency study, the deposition rate is solely dictated by the target voltage. Fig. 13 shows the variation of deposition rate with the pulse frequency of the pulsed DC reactive sputtering of AlN. The deposition rate varied correspondingly with the target voltage as sputtering ion species are fixed at 100% nitrogen. Higher deposition voltage caused higher sputtering yield and vice versa.

Pulse frequency can influence the topographical and morphological properties of the AlN films. Fig. 14 shows the XRD 2-theta curves of AlN films deposited by pulsed DC reactive sputtering with various pulse frequencies. The deposited films have preferred [002] orientation for all frequencies. The FWHMs are 0.46°, 0.45°, 0.42°, 0.43°, and 0.44° for 25 kHz, 50 kHz, 100 kHz, 200 kHz, and 300 kHz, respectively. The film deposited with 100 kHz pulse frequency had the lowest FWHM value indicating the strongest [002] orientation for 100 kHz pulse frequency. However, as the FWHM values of the 2-theta scans are very close for 50 kHz, 100 kHz, and 200 kHz, rocking curve data can confirm the relative differences among films deposited with these pulse frequencies. Fig. 15

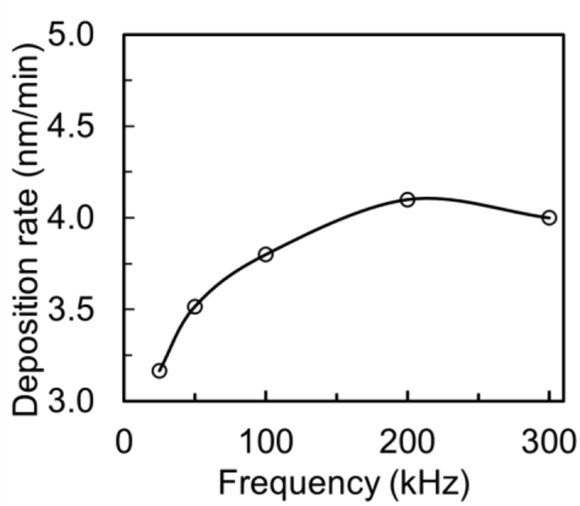

Fig. 13. Variation of deposition rate with the pulse frequency of the pulsed DC reactive sputtering of AlN.

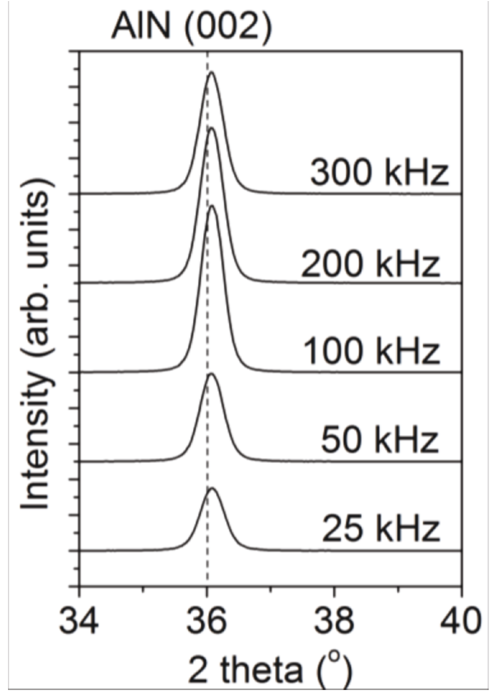

Fig. 14. XRD 2-theta curves of AlN films deposited with various DC pulse frequencies. The FWMs are 0.46°, 0.45°, 0.42°, 0.43°, and 0.44° for 25 kHz, 50 kHz, 100 kHz, 200 kHz, and 300 kHz, respectively.

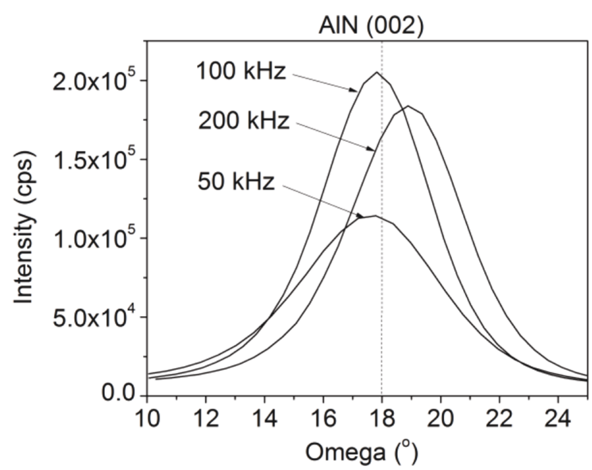

Fig. 15. XRD rocking curve of AlN films deposited with various DC pulse frequencies. The FWHM values are 5.71°, 4.61°, and 4.82° for pulse frequencies of 50 kHz, 100 kHz, and 200 kHz, respectively.

shows the XRD rocking curve of AlN films deposited with various DC pulse frequencies. The FWHM values are 5.71°, 4.61°, and 4.82° for pulse frequencies of 50 kHz, 100 kHz, and 200 kHz, respectively. The rocking curve data reveals the best crystal quality for 100 kHz with no noticeable peak shift and the lowest FWHM values among the films deposited with pulse frequencies of 50–200 kHz.

Fig. 16 shows the roughness values of AlN films deposited by DC reactive sputtering with various pulse frequencies. Films deposited at a pulse frequency of 100 kHz and 200 kHz had the lowest similar minimal roughness. Fig. 17 shows Young's modulus values of AlN films deposited by DC reactive sputtering with various pulse frequencies. Young's modulus data correlates with the roughness data, i.e., the highest Young's modulus was obtained for 100 and 200 kHz with a value around 335 GPa. There are more on-off cycles with 100 kHz than 25 kHz or 50

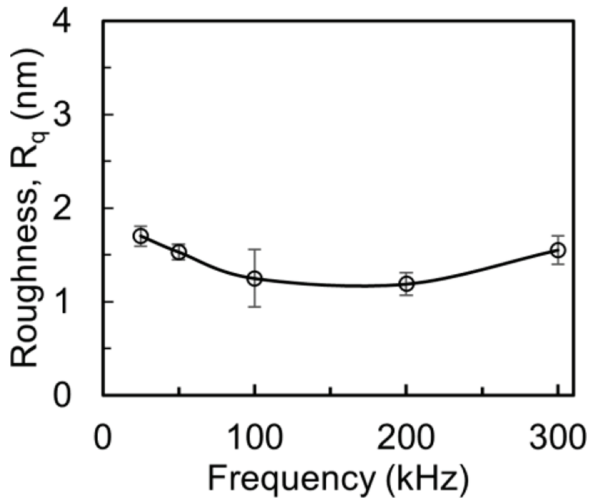

Fig. 16. Roughness values of AlN films deposited by DC reactive sputtering with various pulse frequencies.

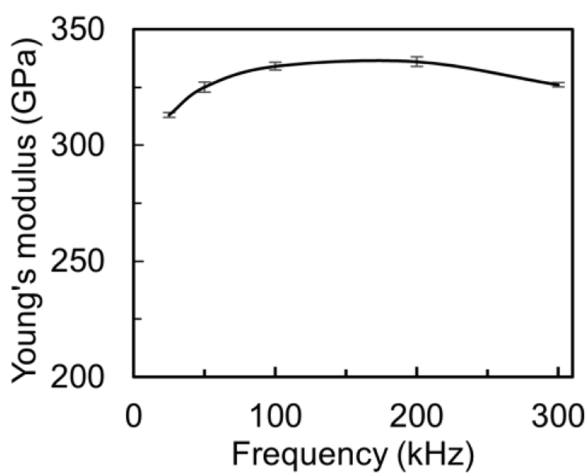

Fig. 17. Young's modulus values of AlN films deposited by DC reactive sputtering with various pulse frequencies.

kHz within a specific time for a fixed duty cycle. That led to more frequent refreshing of the target surface and, additionally, shorter periods of on-time, i.e., the negative potential and active sputtering time, at 100 kHz, which also offers less accumulation of positive $N+$ charge on nitrided target. The result was a homogeneous film with fewer defects. However, at a higher frequency beyond 200 kHz, the discharging reverse time is squeezed to a minimal value, which might not be enough to discharge the nitrided target surface, resulting in a decline in topographical and morphological properties of the deposited films. A higher target voltage favors the densification, crystallization, and surface-smoothening of the depositing film. Therefore, Young's modulus (in Fig. 17) and XRD peak sharpness (Fig. 14) follow the target voltage profile (in Fig. 12), and the surface roughness profile (in Fig. 16) follows an opposite trend to the target voltage profile.

3.4. Pulse duty cycle study

Variation in pulse duty cycle affects target voltage and discharge current quite insignificantly compared to other process parameters. Fig. 18 shows the variation of target voltage and discharge current of the pulsed DC reactive sputtering deposition of AlN. A pulse duty cycle of 80% offered the highest target voltage; thus, the lowest discharge current, which can be attributed to the balanced charge build-up during

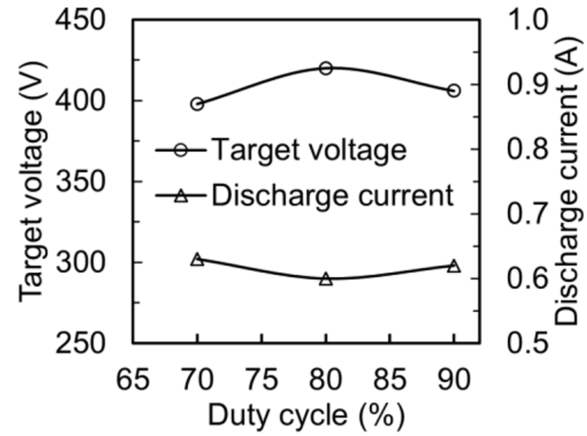

Fig. 18. Variation of target voltage and discharge current of the pulsed DC reactive sputtering deposition of AlN.

"on" time (t_{on}) and discharging during the reverse time (t_{rev}) of the nitrided target. Fig. 19 shows the variation of the deposition rate of the pulsed DC reactive sputtering deposition of AlN. The deposition rate variation follows the variation profile of the target voltage in Fig. 18, which confirms the higher sputtering yield at a higher voltage and vice versa.

The duty cycle of the DC pulse was observed to influence the properties of the AlN films slightly. Fig. 20 shows the XRD pattern of AlN films deposited by pulsed DC reactive sputtering with different duty cycles at a DC pulse frequency of 100 kHz. The FWHMs are 0.44°, 0.42°, and 0.45° for 70%, 80%, and 90% duty cycle, respectively. The lowest FWHM for the (002) AlN peak was found for the sample deposited with a duty cycle of 80%. The occurrence of the lowest FWHM for the 80% duty cycle can be attributed to the highest target voltage, as seen in Fig. 18, which resulted in depositing AlN atoms with the highest energy at that duty cycle. Fig. 21 shows the roughness values of AlN films deposited by pulsed DC reactive sputtering with various pulse duty cycles. The lowest roughness was achieved for the film deposited with the DC pulse duty cycle of 80%. Fig. 22 shows Young's modulus of AlN films deposited by pulsed DC reactive sputtering with various duty cycles. The highest Young's modulus of 335 GPa was achieved for the film deposited with the pulse duty cycle of 80%. For a duty ratio of 80%, which offered the highest target voltage, the film grew efficiently with better crystallinity, higher density, and lower roughness. The same explanation can be applied to Young's modulus. A duty cycle of 80% at a frequency of 100 kHz was found to be the most optimum pulse parameter.

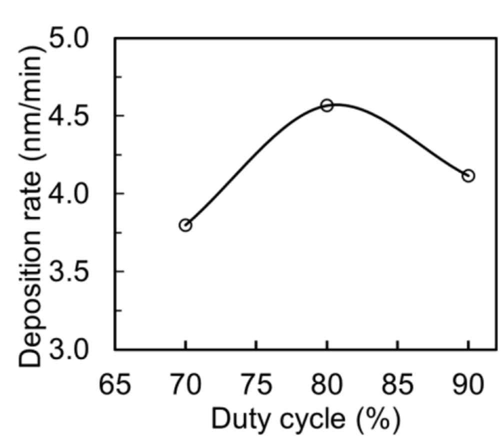

Fig. 19. Variation of the deposition rate of the pulsed DC reactive sputtering deposition of AlN.

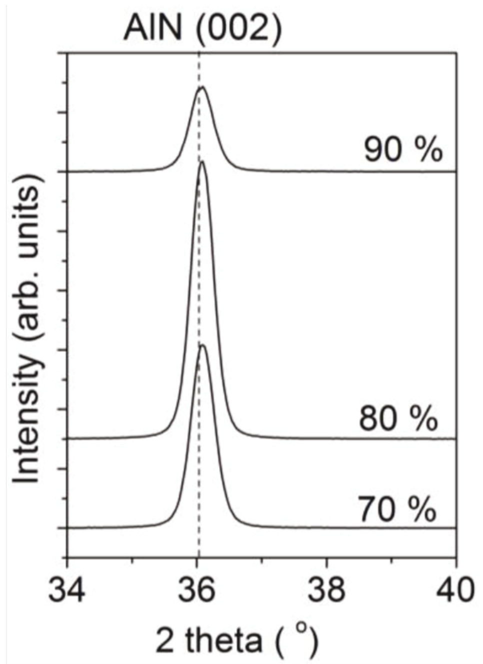

Fig. 20. XRD pattern of AlN films deposited by pulsed DC reactive sputtering with different duty cycles. The FWHMs are 0.44°, 0.42°, and 0.45° for 70%, 80%, and 90% duty cycle, respectively.

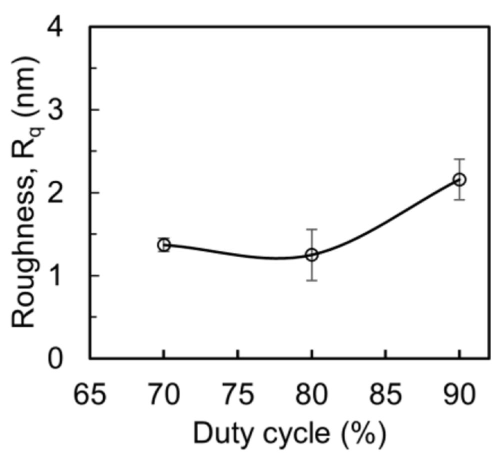

Fig. 21. Roughness values of AlN films deposited by pulsed DC reactive sputtering with various pulse duty cycles.

3.5. *Properties of 1 μm thick ALN film*

A scanning electron microscope (SEM) image of the film's cross-section can provide vital information about the nature of film growth. For understanding the film's growth, a thick film is required. AlN film of 1 μm thickness was deposited with optimized process parameters: 1 mTorr process pressure, 100% N_2 concentration, 100 kHz pulse frequency, and 80% pulse duty cycle. Fig. 23 shows the SEM cross-sectional image of 1 μm thick pulsed DC reactive sputtered AlN film deposited with optimized conditions. Columnar growth of the AlN film can be observed from the SEM cross-sectional image. As the film grew thicker (in the vertical direction), the crystallites became larger. Fig. 24 shows the atomic force microscope (AFM) topography image of the 1 μm thick AlN film deposited with optimized conditions. RMS roughness of the 1 μm thick film was measured to be 3.94 nm, which is higher than the previous samples, deposited for process-parameter optimizations. As the film grew thicker, crystallites became larger, which resulted in a rougher surface.

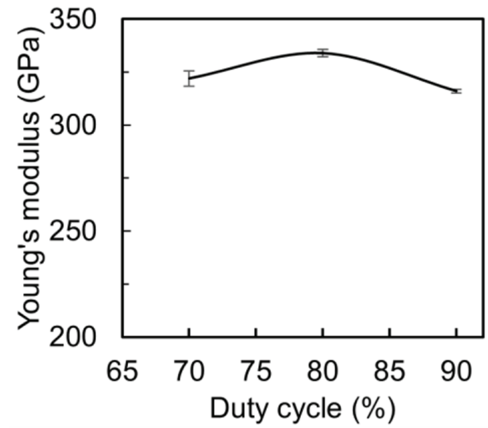

Fig. 22. Young's modulus of AlN films deposited by pulsed DC reactive sputtering with various duty cycles.

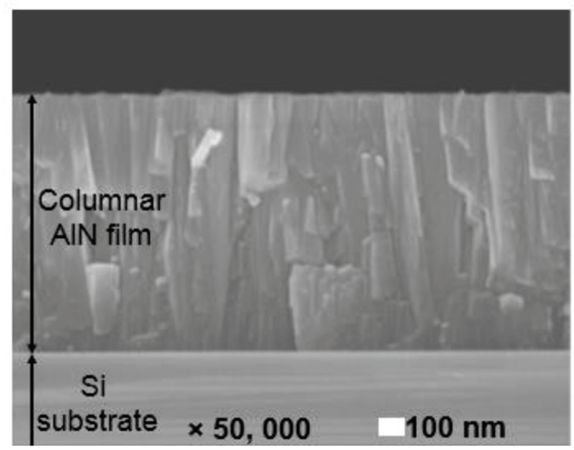

Fig. 23. SEM cross-sectional image of 1 μm thick pulsed DC reactive sputtered AlN film deposited with optimized conditions. The magnification was 50,000 times and the scalebar length was 100 nm.

4. Conclusions

Polycrystalline [002] oriented aluminum nitride films were deposited using the pulsed DC reactive sputtering method. Prior studies revealed that at least 50% nitrogen was needed to deposit stoichiometric films with crystal growth in [002] orientation. With an increasing N_2 gas concentration, the intensity of the (002) peak increased but did not appear to influence the stoichiometry, morphology, or topography. Process pressure had the most significant influence on the film topography and morphology. For AlN films deposited at a process pressure of 1 mTorr, the RMS surface roughness (R_q) was found to be as low as 1.80 nm and Young's modulus reached a value of 312 GPa. These improved properties were due to the formation of a denser film at lower pressure. At lower process pressure, the target voltage was higher and the sputtered atoms experience fewer collisions with the background gas, which led to an increased density and stoichiometric of the film. Increasing nitrogen concentration (e.g., 100%) led to high sputter voltage and energetic sputtered species, which resulted in better crystallinity, smoother surface, and denser films.

Pulse frequency could influence the topographical and morphological of the AlN films. Increasing the frequency from 50 kHz to 200 kHz, the surface roughness (R_q) dropped to as low as 1.20 nm, and Young's

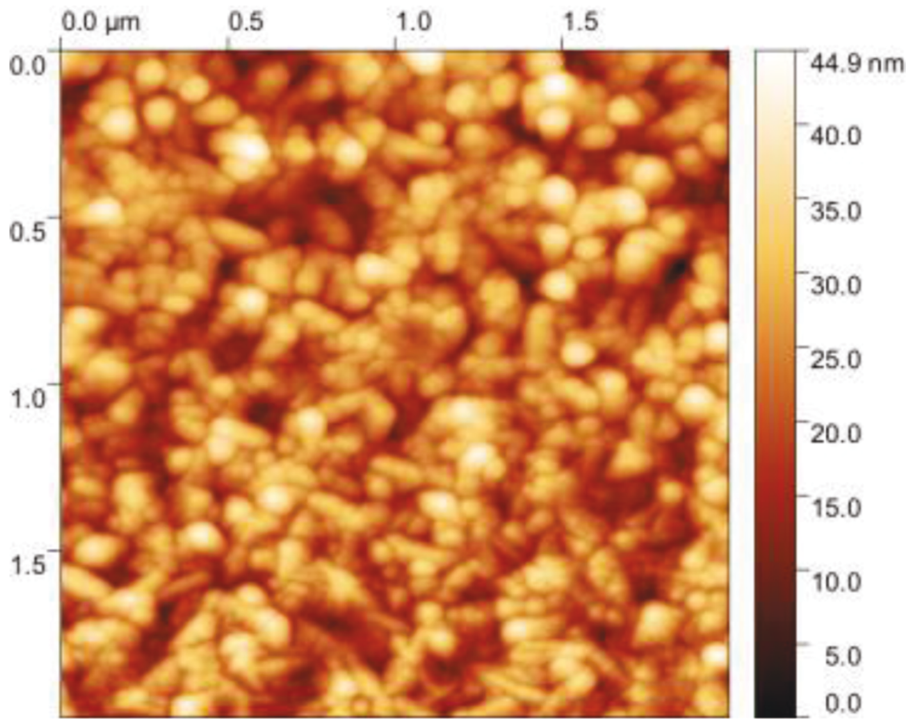

Fig. 24. AFM topography image of the 1 μm thick pulsed DC reactive sputtered AlN film deposited with optimized conditions. RMS roughness was measured to be 3.94 nm.

modulus was increased to 335 GPa, which corresponds to the Young's modulus values for bulk AlN material. The duty ratio did not appear to influence the film properties distinctively. However, at shorter off-times (duty ratio = 0.9), the overall surface roughness increases due to more defects in the growing film because the discharge time was decreased and so the target surface tended to be more contaminated. The same explanation can be applied to Young's modulus. A duty ratio of 0.8 at a frequency of 100 kHz is found to be optimum for growing AlN films with the highest Young's modulus and lowest surface roughness. Films deposited at 300 °C substrate temperature, 100% nitrogen as process gas, 1 mTorr process pressure, 100 kHz pulse frequency, and 80% pulse duty cycle resulted in best-optimized films with the highest Young's modulus of 335 GPa, the lowest surface roughness of 1.2 nm, and best [002] orientation.

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CRediT authorship contribution statement

Al-Ahsan Talukder: Methodology, Investigation, Data curation, Formal analysis, Writing – original draft, Writing – review & editing, Visualization. **Nina Baule:** Data curation, Formal analysis, Validation, Writing – original draft. **Maximilian Steinhorst:** Data curation, Software, Formal analysis. **Maheshwar Shrestha:** Resources, Software. **Qi Hua Fan:** Conceptualization, Supervision, Project administration, Funding acquisition. **Thomas Schuelke:** Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no conflict of interest.

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