© Universiti Tun Hussein Onn Malaysia Publisher's Office

IJIE

Journal homepage: http://penerbit.uthm.edu.my/ojs/index.php/ijie
ISSN : 2229-838X e-ISSN : 2600-7916

The International Journal of Integrated Engineering

Growth of Aluminium Nitride Thin Film Using Pulse-Modulated Rf Magnetron Sputtering Plasma

M. Tahan[1]*, N. Nafarizal[1], M. Z. Sahdan[2], A. S. Bakri[1], N. A. Raship[3], M. K. Ahmad[1], C. F. Soon[1], M. Y. Ahmad[4]

[1]Microelectronics and Nanotechnology-Shamsuddin Research Centre (MiNT-SRC), Institute for Integrated Engineering,
Universiti Tun Hussein Onn Malaysia (UTHM) Batu Pahat, 86400 Johor, MALAYSIA

[2]Preston GeoCEM Sdn. Bhd.,
No.33A Jalan Universiti 4, Taman Universiti, 86400 Parit Raja, Johor, MALAYSIA

[3]Department of Electrical and Electronic Engineering,
National Defense University of Malaysia, 57000 Kem Sungai Besi, Kuala Lumpur, MALAYSIA

[4]Nanorian Technologies Sdn Bhd,
40 & 40, 1, Jln Kajang Perdana 3/2, Taman Kajang Perdana, 43000 Kajang, Selangor, MALAYSIA

*Corresponding Author

DOI: https://doi.org/10.30880/ijie.2020.12.02.016
Received 29 December 2020; Accepted 27 January 2020; Available online 28 February 2020

Abstract: Aluminum nitride (AlN) thin films on silicon (Si) (100) substrates are grown by pulsed rf magnetron sputtering at constant power 200 W, 50 % duty cycle, and substrate at room temperature. The films were characterized using filmetrics, X-ray diffraction, energy dispersive spectroscopy (EDS) and atomic force microscopy (AFM) techniques. The deposited AlN thickness using frequency of 5 Hz for distance of 1-inch and 6-inches were 70.74 nm and 20.40 nm, respectively. The depositions rate is obviously affected by the working distance between the target to the substrates. This is because, at shorter distance between target to substrate, the kinetic energy of bombardment particles become higher. Thus, higher deposition rate and good crystallinity can be obtained as the Al atom and N atom will deposited directly to the substrates. Then, the effect of pulse frequency was investigated. The thickness of AlN thin films using frequency of 5 Hz, 3 Hz and 1 Hz is 70.74 nm 63.72 nm, and 70.23 nm, respectively. The effects of frequency using pulsed rf magnetron was significantly small. The experimental results clearly demonstrate that the energy supplied to the plasma significantly influence the stoichiometric and crystallisation of the thin films.

Keywords: pulse sputtering, AIN thin films, effect of distance, effect of frequency, target to distance

1. Introduction

In the last few years, aluminum nitride (AlN) already attracted much attention for the production of electronic and optoelectronic applications (Abdallah, Chala, Jouan, Besland, & Djouadi, 2007)(Signore et al., 2013)(Kohout et al., 2017). The unique properties of AlN such as wide band gap, high thermal stability and high thermal conductivity enable AlN materials to be used for high power device such as high electron mobility transistor (HEMT), high frequency device such as surface acoustic wave (SAW) devices and deep ultraviolet (UV) optoelectronic devices such as light emitting

---

*Corresponding author: mulianathn15@gmail.com diode (LED) (Strassburg et al., 2004)(Ting, Thao, & Kuo, 2017). The properties of AlN can be controlled by adjusting its bandgap during deposition process.

Several techniques such as hydride vapour phase epitaxy (HVPE) (Bessolov et al., 2017), metal-organic chemical vapour deposition (MOCVD) (Jayasakthi et al., 2016), molecular epitaxy beam (MBE) (Afroz et al., 2015)(Xin, Azurahanim, Abdullah, Zaki, & Yusoff, 2019) and direct current/radio frequency (DC/RF) magnetron sputtering already has been used to grow AlN epitaxial films (Panda et al., 2017). Among these techniques, sputtering is the most promising technique which allows uniform and large scale of AlN thin films to be produced at a low cost (Valcheva, Birch, Persson, Tungasmita, & Hultman, 2006)(Xiao, Suzuki, Miyake, Harada, & Ujihara, 2018).

In this work, DC/RF magnetron sputtering in pulsed rf supply are used to deposit AlN thin films. Pulsed RF sputtering is one of the several functions from DC/RF magnetron sputtering system (SNTEK PSP 5004 (09SN70)) other than pulse DC sputtering, DC sputtering and RF sputtering supply. According to S. Middya et al, pulsing the sputtering power can improved properties of sputtered films while giving a cool effect to the sputtered film via control of the energy of ions impinging on substrates (Middya, Layek, & Ray, 2013)(Kelly & Bradley, 2009). In previous literature, AlN films deposited at close distance between target to substrates are tends to get AlN (100) orientation which is suitable for SAW application (Xu, Wu, Zhang, & Jin, 2001). However, research on pulsed rf sputtering for AlN thin films is still not well studied as no papers have been reported for the deposition of AlN thin film. Therefore, the effects of pulse rf frequency sputtering to AlN deposition will be investigated investigate using XRD, ellipsometry, AFM and EDS.

2. Materials and Methods

The thin film of AlN was deposited using DC/RF magnetron sputtering system (SNTEK PSP 5004 (09SN70)) using rf power supply (Advance Energy). 3mm thick Al 99.99% purity was used as a target and (100) silicon wafer cut into 2x2 cm was used as a substrate. The (100) silicon substrates were dipped in 5% HF solution for 5 minutes (Wang, Lin, Wang, Li, & Luo, 2017) to removes native oxides layers on the surface. The silicon substrates were put into the sputtering chamber and evacuated to 5 x 10^{-6} Torr (base pressure). After the base pressure achieved, the mixture of argon and nitrogen gas was introduced with a constant pressure of 10m Torr. Prior to deposition, pre-sputtering using Ar gas was performed to Al target to removes impurities on the target for 10 minutes. All the depositions in this pulse rf supply had been carried out at 200W power out for 2 hours in different variables as shown in Table 1 and Table 2 without using additional temperature.

Table 1 - Sputtering process parameter for effect of distance

Sputtering process parameters	Value
Power supply	Pulse rf
Target to substrates distance	1-inch and 6-inch
Duty cycle	50 %
Base pressure	5 x10^{-6} mTorr
Working pressure	10 mTorr
Sputtering power	200W
Deposition time	2 hours
Gas flow rate	Ar: 70 sccm, N: 50 sccm
Pulse frequency	5 Hz

Table 2 - Sputtering process parameter for effect of frequency

Sputtering process parameters	Value
Power supply	Pulse rf
Target to substrates distance	1-inch
Duty cycle	50 %
Base pressure	5 x10^{-6} mTorr
Working pressure	10 mTorr
Sputtering power	200W
Deposition time	2 hours
Gas flow rate	Ar: 70 sccm, N: 50 ccm
Pulse frequency	1 Hz, 3 Hz and 1 Hz

The thickness of thin films is measured by using filmetrics (Film measure). The crystallinity of the AlN was determined by X-ray diffraction (XRD) (Panalytical Xpert3 Powder) analysis with radiation Cu K-alpha wavelength of 0.154nm. To evaluate the composition of the thin film, a quantitative analysis of the energy dispersive X-ray spectroscopy

(EDS) spectra was performed using INCA software. The surface morphology was examined by atomic force microscopy (AFM) (Hitachi, XE- Series SPM Controller).

3. Results and Discussion

3.1 Construction of References

The film thickness is very important in every research. It is because the film itself consists of information such as surface structure and physical properties which is very useful in pursuing fundamental materials science studies (Abdallah et al., 2008). In the sputtering process, the AlN thin film is form from the bombardment of aluminum (Al) target and collision with nitrogen (N) atoms by Ar ions. The atom that ejected from Al target and collided from N will be deposited layer by layer onto the silicon (Si) substrates which is opposite from the sputtering target.

As for the results of the effect of distance between substrates to target, the thin films thickness measured by Filmetric using a distance of 6-inch between the target to substrates was 20.40 nm while 1-inch was 70.74 nm. The depositions rate was obviously affected by the difference in working distance. This result has been expected based on the results of previous studies by other types of materials target (Wuhrer & Yeung, 2003)(Yang, He, Tian, Liu, & Fu, 2010) that smaller working distance will increase the deposition rate. In the sputtering process, when the distance between the substrate and target become smaller, the Al and N atoms will be deposited on the substrate with higher kinetic energy as it has fewer collisions with other particles. Thus, a higher deposition rate and good crystallinity can be got. On the other hand, when the distance is larger, the kinetic energy of atoms will be reduced as it having multiple collision with other particles before reaching the substrates which will cause lower deposition rate and poor crystallinity (Iqbal & Mohd-Yasin, 2018).

Fig. 1 reported the XRD patterns obtained from the samples deposited at a distance of 1-inch and 6-inch from target to substrates. Bare Si wafer also scanned to identify the location of Si peak in XRD software. Based on XRD results, no AlN peak is obtained. The XRD pattern in 6-inch working distance thin films shows very sharp and high intensity peak of silicon which was due to the extremely thin AlN atom deposited. However, in the 1-inch working distance the Si peak intensity decreases as a thicker layer of AlN layer had been deposited onto Si substrates. This causes the Si peak intensity decrease as the AlN atom already diffuse into the Si substrate. The reason bare Si wafer had lower XRD intensity compared to Si 6-inch working distance was the wafer already oxidized. In short, all the AlN thin films are in an amorphous structure based on XRD results. The findings of AlN pattern in XRD is amorphous also supported by M. Maqbool et al (Maqbool & Ali, 2009). The amorphous structure is reasonable due to self-sputtering produced by the pulse rf sputtering itself. Berkeley mentioned in his paper that even though with sufficient high voltage is implemented to the sputter target in high power pulsed sputtering, self-sputtering or re-sputtering may be occur as the duration of a self-sputter cycle can be short as 1ms (Berkeley, 2004). Re-sputtering or back sputtering is condition where ejected particles from bombardment of atom during sputtering process are widely distributed and deposited onto the substrates and vacuum chamber is bounced back or re-sputtering back to the target materials and distributed widely. Re-sputtering will affect the crystallization of thin films as it disturbs the arrangement layer of deposited films(Cherng & Chang, 2010)

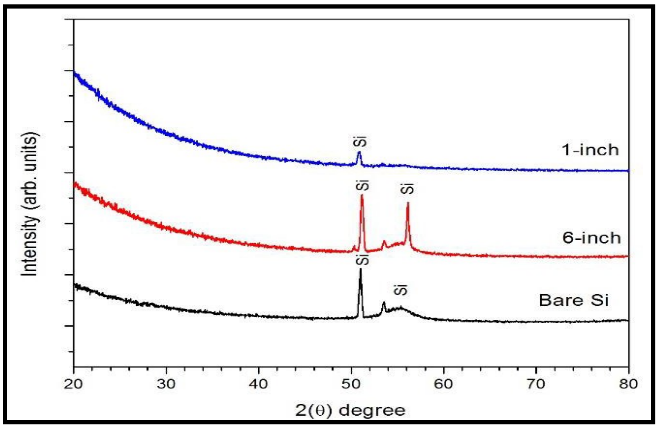

Fig. 1 - XRD patterns of AlN on Si substrates deposited at 200 W for 2 hours using different working distance (6-inch and 1-inch)

EDS analysis enables the researcher to identify the chemical composition and elements that exist inside materials. Table 3 shows the atomic percentages of AlN thin films. The atomic percentages using 1-inch working distance (Al: 51.88%, N: 51.88%) was more balance compared to 6-inch working distance (Al:40.10%, N:59.90%). The reason 6-inch working distance had a lower concentration of Al element may be

caused by less Al atom deposited onto the Si substrates. This was consequences of large working distance which caused Al and N had multiple collision during sputtering process before reaching Si substrates to form AlN layer by layer. It concludes that AlN compound actually exists in the thin films as shown in Fig. 2 EDS spectrum but do not form crystal structure yet.

Table 3 - Atomic percentages, thickness and surface roughness of AlN thin films deposited on different working distance. The data are taken from EDS, ellipsometry and AFM

Working distance inch	Al atomic percentage (%)	N atomic percentage (%)	Film thickness (nm)	Surface roughness (nm)
1	48.12	51.88	70.74	$1.317\text{x}10^{-1}$
6	40.10	59.90	20.40	$1.156\text{x}10^{-1}$

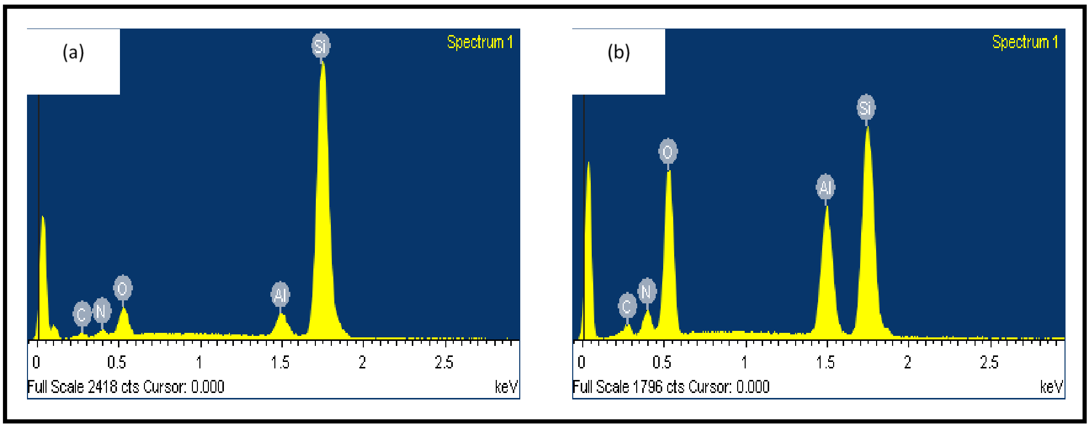

Fig. 2 - EDS spectrum of AlN on Si substrates deposited at (a) 6-inch; and (b) 1-inch

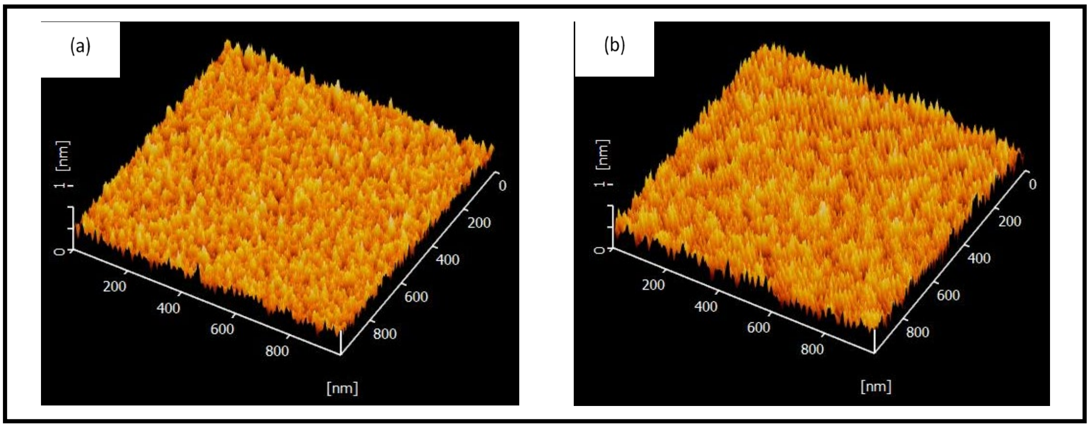

Fig. 3 - AFM morphologies of AlN on Si substrates at (a) 6-inch and (b) 1-inch

Fig. 3 shows three-dimensional AFM images from the surface morphology of a sputtered AlN film. It was found that the surface roughness of AlN thin films in Fig. 3 (a) ($1.156\text{x}10^{-1}$ nm) were smoother compared to Fig. 3 (b) ($1.317\text{x}10^{-1}$). This indicates that decreasing distance between the target to substrates will cause the surface roughness of AlN thin films to become rougher (Yang et al., 2010).

3.2 Effect of Pulse Frequency

Based on the previous experiment using pulse rf magnetron sputtering supply, the 1-inch distance will give a higher deposition rate which may help to produce better AlN crystal structure on Si substrates. Hence, 1-inch target to substrate distance was chosen in this section to investigate the effect of pulse frequency using pulse rf magnetron sputtering supply. Based on paper (Kiyotaka Wasa; Shigeru Hayakawa, 1992), decreasing frequency can increase the deposition rate of the sputtering process. Therefore, the experiments continued to see the effect of pulse frequency to deposition rate, crystal quality and surface morphology. The parameters of this section are shown in Table 2.

The thickness of AlN thin films using a frequency of 5 Hz, 3 Hz and 1 Hz is 70.74 nm 63.72 nm, and 70.23 nm approximately. Based on Table 2, the thickness of all samples was nearly constant and do not shows any trend. The possible reason may be due to a very small gap changed in parameter which caused changing the frequency in this investigation has small effect to the deposition rate. This coincides with P. Samarasekara (Samarasekara, 2003) on his paper as the deposition rates of AlN thin films using rf sputtering method is normally controlled by duty cycle and power.

The XRD pattern of AlN thin film grown on Si substrate at a variable frequency with a 1-inch distance between the target to the substrate is shown in Fig. 4. Again, all the samples of AlN thin films still did not turned into crystalline structure, in other words is amorphous structure. The reason may be caused by lower deposition rates produced by the pulse rf sputtering supply itself even though the frequency has been decreased. A. Iqbal et al. reported that slow deposition rate and poor crystallinity is happened because of weak kinetic energy being supplied to the bombardment ion(Iqbal & Mohd-Yasin, 2018). Therefore, the AlN is assumed to be still extremely thin layer thus resulting in amorphous AlN structure.

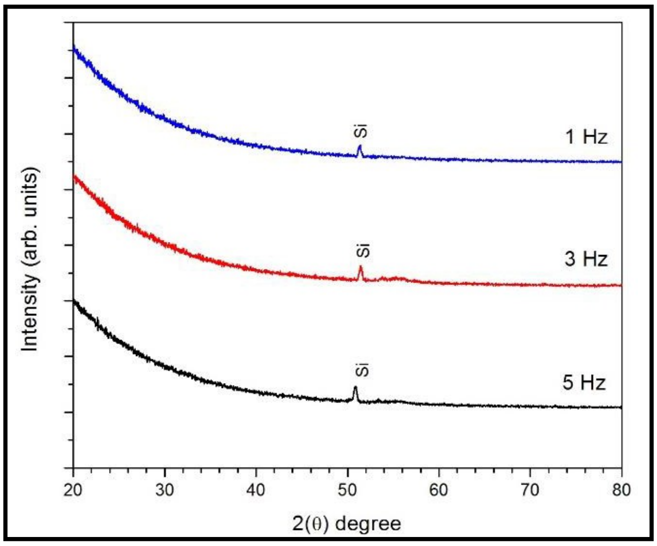

Fig. 4 - XRD patterns of AlN on Si substrates deposited at 200 W for 2 hours using different frequency (5 Hz, 3 Hz and 1 Hz)

To confirm the existence of AlN compound on the Si substrates, EDS analysis has been carried out. The EDS spectrum in Fig. 5 obviously shows the presence of aluminum and nitrogen elements. Table 4 shows the atomic percentages of AlN thin films. It was found the films have good stoichiometric value. As indicates by D. Ma et al (Ma et al., 2019) AlN compound is considered stoichiometric as long as the ratio of stoichiometric value is 1 or plus or minus 0.3. However, percentages of Al atomic percentages were decreased gradually in increasing of pulse frequency meanwhile vice versa to the nitrogen atomic percentages. The deposition rate is the main factor that causes the atomic percentages changed. The frequency can affect the deposition rate of sputtering as it controls the duration time of one cycle in a repeating impulse power. As the frequency decrease from 5 Hz, 3 Hz to 1 Hz, the duration time of power goes on before it goes off will increase from 0.2 s, 0.33 s to 1 s. The duration time is calculated using Equation 1. Therefore, at least frequency, the atomic percentages of Al and N will increase.

$$f = \frac{1}{t} \tag{1}$$

Where *f* is frequency and *t* is time.

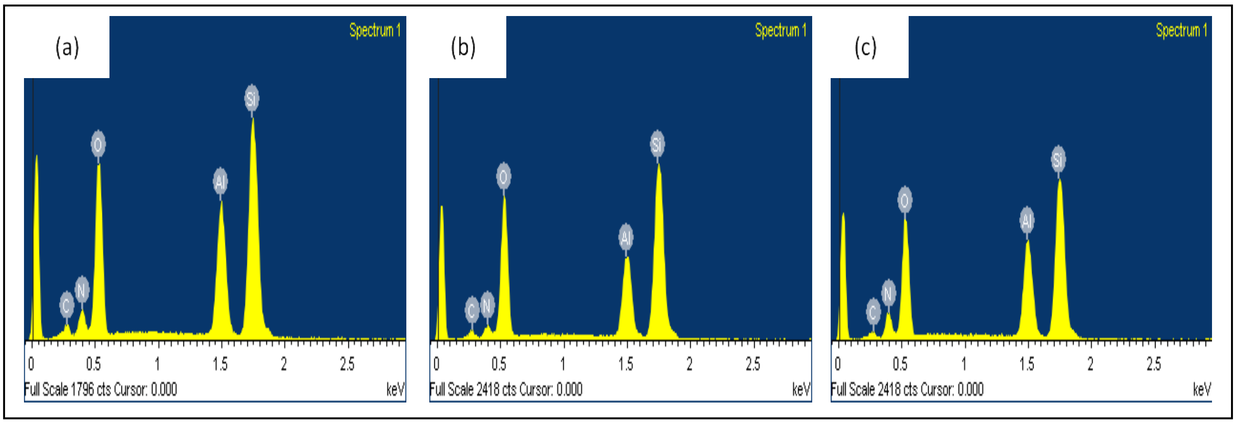

Fig. 5 - EDS spectrum of AlN on Si substrates deposited at frequency (a) 5 Hz, (b) 3 Hz and (c) 1 Hz

Table 4 - Atomic percentages, thickness and surface roughness of AlN thin films deposited on different frequency. The data are taken from EDS, ellipsometry and AFM

Frequency (Hz)	Al atomic percentage (%)	N atomic percentage (%)	Film thickness (nm)	Surface roughness (nm)
1	52.50	47.50	70.23	$1.040x10^{-1}$
3	50.10	49.90	63.72	$1.299x10^{-1}$
5	48.12	51.88	70.74	$1.317x10^{-1}$

AFM surface morphologies to study the effect of frequency in AlN films has been shown in Fig. 6. The roughness value is increased from $1.040x10^{-1}$ nm, $1.299x10^{-1}$ nm and $1.317x10^{-1}$ nm, as the pulse frequency increase from 1 Hz, 3 Hz to 5 Hz. The surface morphology in 1 Hz is the smoothest because the kinetic energy received by ion from 1 Hz frequency is the highest compared to 3 Hz and 5 Hz frequency. The reason 1 Hz received higher kinetic energy compared to others because the duration time for power to on are longer. Therefore, smoother AlN thin films were produced using 1 Hz frequency.

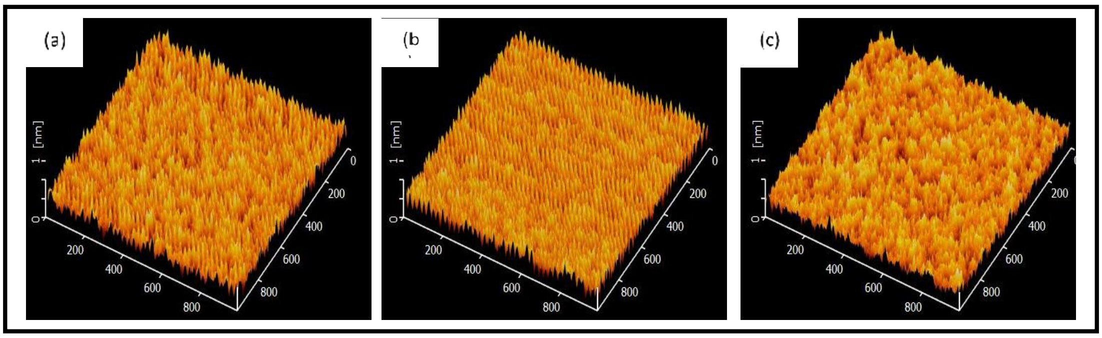

Fig. 6 - AFM morphologies of AlN on Si substrates at frequency (a) 5 Hz, (b) 3 Hz and (c) 1 Hz

4. Conclusion

In this study, the pulsed rf magnetron sputtering method is adopted to deposit AlN on Si (100) to see the effect of working distance between the target to substrates. The thickness of thin films for 1-inch is 70.74 nm while 6-inch is 20.40 nm. Experimental results revealed that smaller distance between the target to substrates gives higher deposition rate to the sputtering process and caused surface roughness to become rougher. However, the AlN still did not form crystalline structure in other words is amorphous. Therefore, other experiments were set up by decreasing the frequency of pulsed rf magnetron sputtering supply to see the effect of frequency. The thickness of AlN thin films using a frequency of 5 Hz, 3 Hz and 1 Hz is 70.74 nm 63.72 nm, and 70.23 nm approximately. As for the atomic percentages, for 5 Hz, 3 Hz and 1 Hz are (Al: 48.12, N: 51.88), (Al: 50.10, N: 49.90) and (Al: 52.50, N: 47.50) meanwhile the surface roughness is $1.317x10^{-1}$ nm, $1.299x10^{-1}$ nm and $1.040x10^{-1}$ nm. Despite of that, the AlN film structure is still amorphous. These results concluded that the effect of decreasing frequency to the thickness, atomic percentages, surface roughness and crystalline structure is very small. This may be caused by a very small gap changed in the parameter which caused changing the frequency of sputtering yielded very small effect to the deposition rate. Nevertheless, further research about the effect of pulsed rf sputtering on AlN thin films should be continued. In future studies, the usage of mid-frequency with bigger parameter gap, high power and smaller distance can be applied so that the effect of frequency will be crystal clear.

Acknowledgement

The author would like to express her thanks to the sponsor by CREST Malaysia through P28C1-17. We would like to express our gratitude towards Microelectronic and Nanotechnology Shamsudin Research Centre for their contribution to this work. This study is partially supported by Ministry of Education Malaysia.

References

[1] Abdallah, B., Chala, A., Jouan, P., Besland, M. P., & Djouadi, M. A. (2007). Deposition of AlN films by reactive sputtering : Effect of radio frequency substrate bias. *Thin Solid Films*, *515*, 7105–7108.

[2] Abdallah, B., Duquenne, C., Besland, M. P., Gautron, E., Jouan, P. Y., Tessier, P. Y., … Cordier, Y. (2008). Thickness and substrate effects on AlN thin film growth. *The European Physical Journal Applied Physics*, *43*, 309–313.

[3] Afroz, F., Nomoto, K., Hu, Z., Rouvimov, S., Xing, H. G., & Jena, D. (2015). Low temperature AlN growth by MBE and its application in HEMTs. *Journal of Crystal Growth*, *425*, 133–137.

[4] Berkeley, L. (2004). Fundamentals of pulsed plasmas for materials processing. *Surface & Coatins Technology*, *183*, 301–311.

[5] Bessolov, V., Kalmykov, A., Konenkova, E., Kukushkin, S., Myasoedov, A., Poletaev, N., & Rodin, S. (2017). Semipolar AlN and GaN on Si(100): HVPE technology and layer properties. *Journal of Crystal Growth*, *457*, 202–206.

[6] Cherng, J. S., & Chang, D. S. (2010). Effects of pulse parameters on the pulsed-DC reactive sputtering of AlN thin films. *Vaccum*, *84*(5), 653–656.

[7] Iqbal, A., & Mohd-Yasin, F. (2018). Reactive sputtering of aluminum nitride (002) thin films for piezoelectric applications: A review. *Sensors (Switzerland)*, *18*(6), 1–21.

[8] Jayasakthi, M., Juillaguet, S., Peyre, H., Konczewicz, L., Baskar, K., & Contreras, S. (2016). Superlattices and Microstructures In fl uence of AlN thickness on AlGaN epilayer grown by MOCVD. *Superlattices and Microstructures*, *98*, 515–521.

[9] Kelly, P. J., & Bradley, J. W. (2009). Pulsed magnetron sputtering – process overview and applications. *Journal of Optoelectronic and Advanced Materials*, *11*(9), 1101–1107.

[10] Kiyotaka Wasa; Shigeru Hayakawa. (1992). *Handbook of Sputter Deposition Technology: Principles, Technology, and Applications*. *Handbook of Sputter Deposition Technology*.

[11] Kohout, J., Qian, J., Schmitt, T., Vernhes, R., Zabeida, O., Klemberg-Sapieha, J., & Martinu, L. (2017). Hard AlN films prepared by low duty cycle magnetron sputtering and by other deposition techniques. *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films*, *35*(6), 061505.

[12] Ma, D. L., Liu, H. Y., Deng, Q. Y., Yang, W. M., Silins, K., Huang, N., & Leng, Y. X. (2019). Optimal target sputtering mode for aluminum nitride thin fi lm deposition by high power pulsed magnetron sputtering. *Vacuum*, *160*, 410–417.

[13] Maqbool, M., & Ali, Æ. T. (2009). Intense red catho- and photoluminescence from 200 nm thick samarium doped amorphous AlN thin films. *Nanoscale Research Letters*, *4*, 748–752.

[14] Middya, S., Layek, A., & Ray, P. P. (2013). Possibility to Use low temperature pulsed RF sputtered indium tin oxide for the fabrication of organic solar cell. *Conference Paper in Energy*, *2013*, 1–5.

[15] Panda, P., Ramaseshan, R., Ravi, N., Mangamma, G., Jose, F., Dash, S.,Suematsu, H. (2017). Reduction of residual stress in AlN thin films synthesized by magnetron sputtering technique. *Materials Chemistry and Physics*, *200*, 78–84.

[16] Samarasekara, P. (2003). A pulsed RF sputtering method for obtaining higher deposition Rates. *Chinese Journal of Physics*, *41*(1), 70–74.

[17] Signore, M. A., Bellini, E., Taurino, A., Catalano, M., Martucci, M. C., Cretì, P., … Quaranta, F. (2013). Structural and morphological evolution of aluminum nitride thin films: Influence of additional energy to the sputtering process. *Journal of Physics and Chemistry of Solids*, *74*(10), 1444–1451.

[18] Strassburg, M., Senawiratne, J., Dietz, N., Haboeck, U., Hoffmann, A., Noveski, V., … Sitar, Z. (2004). The growth and optical properties of large, high-quality AlN single crystals. *Journal of Applied Physics*, *96*(10), 5870–5876.

[19] Ting, C. W., Thao, C. P., & Kuo, D. –H. (2017). Electrical and structural characteristics of tin-doped GaN thin films and its hetero-junction diode made all by RF reactive sputtering. *Materials Science in Semiconductor Processing*, *59*(September 2017), 50–55.

[20] Valcheva, E., Birch, J., Persson, P. O. Å., Tungasmita, S., & Hultman, L. (2006). Epitaxial growth and orientation of AlN thin films on Si(001) substrates deposited by reactive magnetron sputtering. *Journal of Applied Physics*, *100*(12), 0–6.

[21] Wang, H., Lin, Z., Wang, W., Li, G., & Luo, J. (2017). Growth mechanisms of GaN epitaxial films grown on ex situ low-temperature AlN templates on Si substrates by the combination methods of PLD and MOCVD. *Journal of Alloys and Compounds*, *718*, 28–35.

[22] Wuhrer, R., & Yeung, W. Y. (2003). Effect of target-substrate working distance on magnetron sputter deposition of nanostructured titanium aluminium nitride coatings. *Scripta Materialia*, *49*(3), 199–205.

[23] Xiao, S., Suzuki, R., Miyake, H., Harada, S., & Ujihara, T. (2018). Improvement mechanism of sputtered AlN fi lms by high-temperature annealing. *Journal of Crystal Growth*, *502*(May), 41–44.

[24] Xin, H., Azurahanim, C., Abdullah, C., Zaki, M., & Yusoff, M. (2019). Structural and optical properties of AlN / GaN and AlN / AlGaN / GaN thin films on silicon substrate prepared by plasma assisted molecular beam epitaxy(MBE). *Results in Physics*, *12*(December 2018), 1177–1181.

[25] Xu, X., Wu, H., Zhang, C., & Jin, Z. (2001). Morphological properties of AlN piezoelectric thin films deposited by DC reactive magnetron sputtering. *Thin Solid Films*, *388*, 62–67.

[26] Yang, H., He, J., Tian, C., Liu, C., & Fu, D. (2010). Influence of target-substrate distance and sputtering power on chromium oxide films prepared by medium-frequency magnetron sputtering. *Wuhan University Journal of Natural Sciences*, *15*(5), 440–443.