[논문] 열처리공학회지, 제23권 제2호(2010)
J. of the Korean Society for Heat Treatment, *Vol. 23, No. 2, (2010) pp. 69~74*

A Study on the Fabrication and Structural Evaluation of AlN Thin Films

Seung-Oh Han[*†], **Chang-Suk Han**[**]
[*]Institute of Fusion Technology, Hoseo University
[**]Dept. of Defense Science & Technology, Hoseo University,
165 Sechul-Ri, Baebang-Myun, Asan City, Chungnam 336-795, Korea

Abstract AlN thin films were deposited by using a two-facing-targets type sputtering system (TFTS), and their deposition characteristics, microstructure and texture were investigated. Total gas pressure was kept constant at 0.4 Pa and the partial pressures of nitrogen, PN_2 ((N_2 pressure)/(Ar+N_2 pressure)) varied from 0 to 0.4 Pa. The texture of the film cross-sections and surface morphology were observed by field emission scanning electron microscope (FE-SEM). The crystallographic orientation of the films were analyzed by X-ray diffraction (XRD). Deposition of AlN film depends on N_2 partial pressure. The best preferred oriented AlN thin films can be deposited at a nitrogen partial pressure of PN_2 = 0.52. As-deposited AlN films show preferred orientation and columnar structure, and the grain size of AlN films increases with increasing sputtering current.

(Received January 4, 2010; Revised January 29, 2010; Accepted February 17, 2010)

***Key words:** Two-facing-targets type sputtering system (TFTS), AlN thin films, Preferred orientation, Columnar structure*

1. Introduction

Aluminum nitride (AlN) is a large energy gap III-V compound that has been widely studied owing to its optical, electrical, chemical inertness and acoustic properties[1-4]. The chemical and thermal stabilities as well as high resistivity of AlN was expected to be used for insulating and passivating layers in semiconductor devices[5,6], and AlN coated films were also found to be used as high temperature oxidation resistant[7,8]. Though the AlN films can be produced in different ways such as RF sputtering[9], molecular beam epitaxy[10,11], chemical vapor deposition[12] and reactive sputtering[13-17]; the most popular method is reactive sputtering owing to the merits of inexpensive process, low temperature deposition, and good crystallinity. Since the purpose of this study is to synthesize AlN thin films containing Co and Fe particles and to examine the microstructure, magnetic and electrical properties, it is necessary to study first the formation processing of AlN thin films. In this paper, fabrication of AlN thin films and their microstructure will be discussed briefly.

2. Experimental procedure

In this study, the distance between the target and shield was kept constant at 0.7 cm. Two Al disks of 10 cm diameter and 0.5 cm thickness were used as targets. Ar and N_2 gases were inserted into the chamber through needle valve, which was designed as shown Fig. 1. After the chamber was evacuated below 2×10^{-4} Pa, the sputtering gas Ar was introduced and the pressure of the Ar was kept constant at 0.4 Pa. After pre-sputtering for 1.8 ks, the chamber was again evacuated below 2×10^{-4} Pa, and then reactive sputtering started by introducing Ar and N_2 gases and after 1 ks the shutter was removed for deposition. Total gas pressure was kept constant at 0.4 Pa and the partial pressures of nitrogen, PN_2 ((N_2 pressure)/(Ar+N_2 pressure))

[†] E-mail : sohan@hoseo.edu

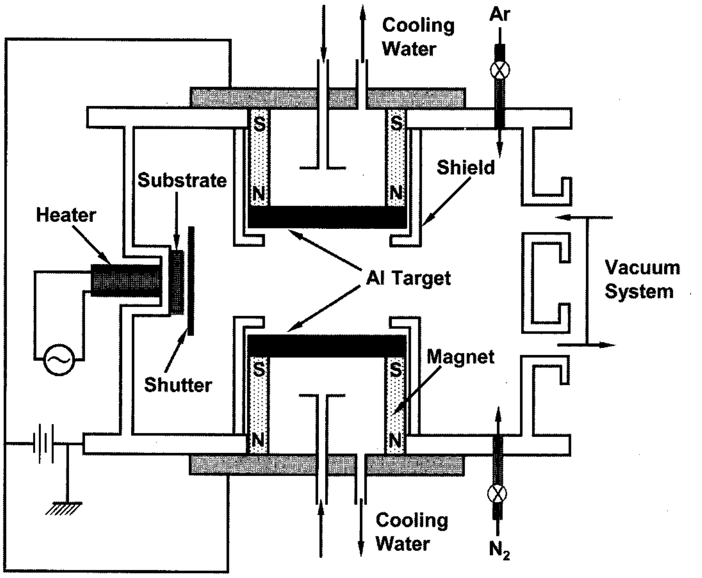

Fig. 1. Schematic drawing of a Two-Facing-Target-Type DC sputtering (TFTS) apparatus.

Table 1. Deposition conditions

Target	Al (99.99)
Substrate	Glass & NaCl
Substrate temperature	< 323 K
Target voltage	DC –300 V to –500 V
Sputtering current	300 mA, 400 mA, 500 mA
Composite gas	(Ar + N_2) of various N_2 partial gas pressure
Initial pressure	$< 2 \times 10^{-4}$ Pa
Total gas pressure	0.4 Pa

varied from 0 to 0.4. Several films were prepared for different partial pressures of N_2 on glass substrate for deposition time of 14.4 ks. Some films were also deposited on NaCl substrates. The deposition conditions are listed in Table 1.

A taly step profilometer was used to measure the thickness of prepared films, and the deposition rate was calculated from the thickness of deposited film and deposition time. In order to study the growing process, the films deposited on NaCl substrates were pilled out by solving NaCl in a mixture solution of water and ethanol and subjected to transmission electron microscopy (TEM) observations, including electron diffraction and the observation of bright field images. The texture of the film cross-sections and surface morphology were also observed by field emission scanning electron microscope (FE-SEM). The crystallographic orientation of the films and the relative peak intensity were analyzed by X-ray diffraction (XRD).

3. Experimental results

3.1. Deposition characteristics

Fig. 2 shows the dependence of applied voltage on the N_2 partial pressure. As shown in the figure, the voltage is the highest for $PN_2 = 0$ partial pressure and decreases to the lowest -295 V for the N_2 partial pressure of 0.44 and then again increases slowly with increasing N_2 partial pressure.

Fig. 3 shows the effect of the partial pressure of N_2 on deposition rate. The deposition rate is almost constant at 0.14 nm/s up to the partial pressure of $PN_2 = 0.19$. The deposition rate

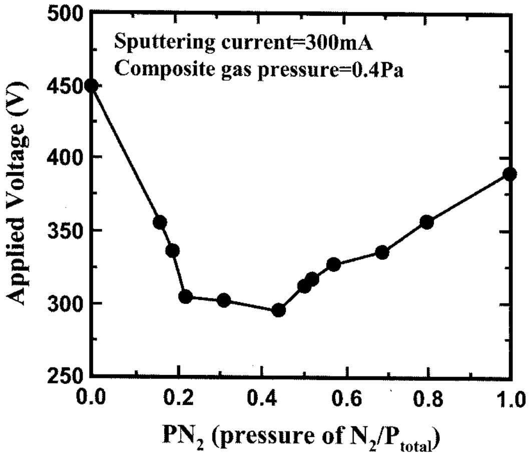

Fig. 2. Dependence of the applied voltage on the N_2 partial pressures PN_2.

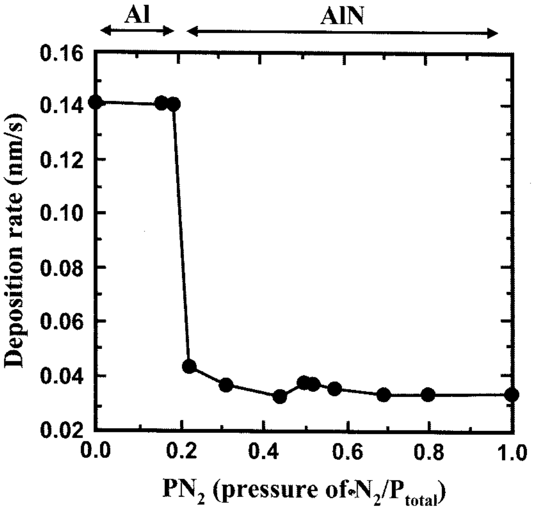

Fig. 3. Dependence of the deposition rate on nitrogen partial pressure (PN_2).

abruptly decreases to about 0.043 nm/s at the partial pressure of $PN_2 = 0.22$ and the deposition rate is almost unchanged for higher N_2 partial pressures.

3.2. Crystallographic orientation

Fig. 4 shows some typical XRD patterns of thin films deposited at different partial gas pressures of N_2 on glass substrate, with a sputtering current of 300 mA. The prepared film deposited below the nitrogen partial pressure of $PN_2 = 0.19$ shows the peaks for only pure Al. And the films deposited above $PN_2 = 0.22$, were found to be ascribed to AlN phase, no peak for Al was observed. In AlN film only the 0002 peak was observed, indicating that the AlN film has the c-plane parallel to the substrate as a preferred orientation.

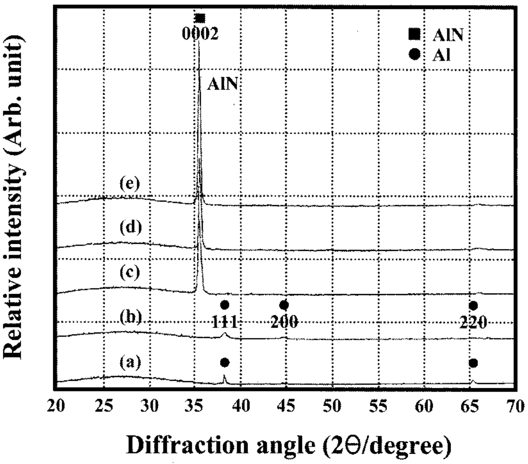

Fig. 4. Typical XRD profiles of Al and AlN films deposited at different N_2 partial pressures with a total pressure of 0.4 Pa; (a) $PN_2 = 0$, (b) $PN_2 = 0.19$, (c) $PN_2 = 1.0$, (d) $PN_2 = 0.5$, (e) $PN_2 = 0.52$.

Fig. 5 shows the relative peak intensity of AlN 0002 peak as a function of the partial pressure of N_2 (PN_2/P_{total}). The highest intensity of the peak was observed for the film deposited at a partial pressure of $PN_2 = 0.52$. Beyond this, the 0002 peak intensity is much lower. Therefore, it can be thought that good crystalline AlN is deposited at a partial pressure $PN_2 = 0.52$.

The results of X-ray observations were also confirmed with TEM microscopy. Fig. 6 and 7 show TEM observations of Al and AlN thin films deposited on NaCl substrate respectively. Both the films were deposited with a total pressure of 0.4 Pa, sputtering current 300 mA. The film deposited with a N_2 partial pressure of 0 is ascribed to pure Al film, as the corresponding ED patterns show that all the rings are

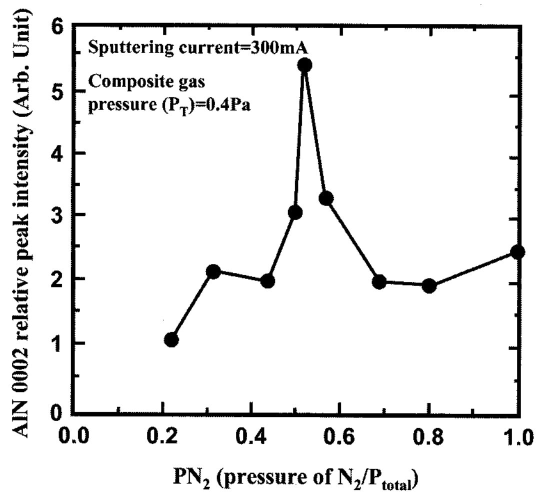

Fig. 5. Effect of N_2 partial pressure on 0002 preferred orientation of deposited AlN films.

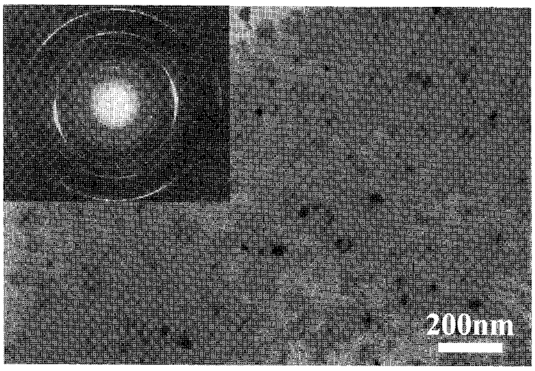

Fig. 6. TEM bright field image and corresponding ED pattern of a pure Al film prepared with $PN_2 = 0$.

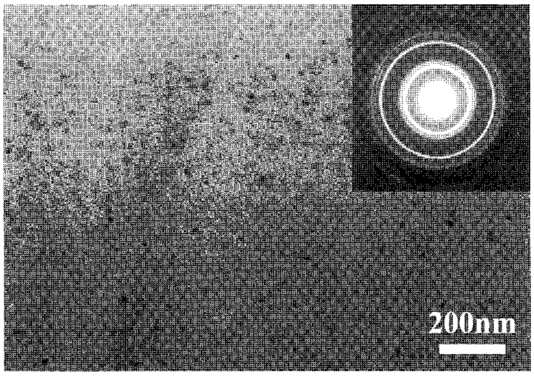

Fig. 7. TEM bright field image and corresponding ED pattern of AlN film prepared with a nitrogen partial pressure of $PN_2 = 0.052$.

ascribed to pure Al. On the other hand, for the film deposited with a N_2 partial pressure of 0.52 is attributed to be a pure AlN film. Corresponding ED patterns also show that all the rings are ascribed to the hcp AlN phase. It is also clear from the figures that the grain size of AlN crystal is much smaller and more homogeneous than that of Al crystal. Crystal structure obtained from electron diffraction patterns are tabulated in Table 2.

3.3. Microstructure and morphology

Fig. 8 shows some FE-SEM micrographs of surface and cross-section of AlN films deposited

Table 2. Crystal structure information obtained from electron diffraction results

Nitrogen partial pressure (Pa)	State of the film	Ring No.	Ring Pattern	Plane spacing (Å)		Crystal structure	hkl
				Measured	JCPDS		
0	as-deposited	1	Sharp, spotted	2.336	2.338	Al fcc	111
		2	Sharp, spotted	2.014	2.024	Al fcc	200
		3	Sharp	1.426	1.431	Al fcc	220
		4	Sharp	1.122	1.221	Al fcc	311
		5	Sharp	0.923	0.929	Al fcc	331
		6	Weak	0.818	0.827	Al fcc	422
0.52	as-deposited	1	Strong, board	2.688	2.695	AlN hcp	100
		2	Strong, sharp	2.362	2.371	AlN hcp	101
		3	Weak	1.875	1.829	AlN hcp	102
		4	Strong, sharp	1.545	1.556	AlN hcp	110
		5	Weak, sharp	1.409	1.413	AlN hcp	103
		6	Broad	1.320	1.319	AlN hcp	112

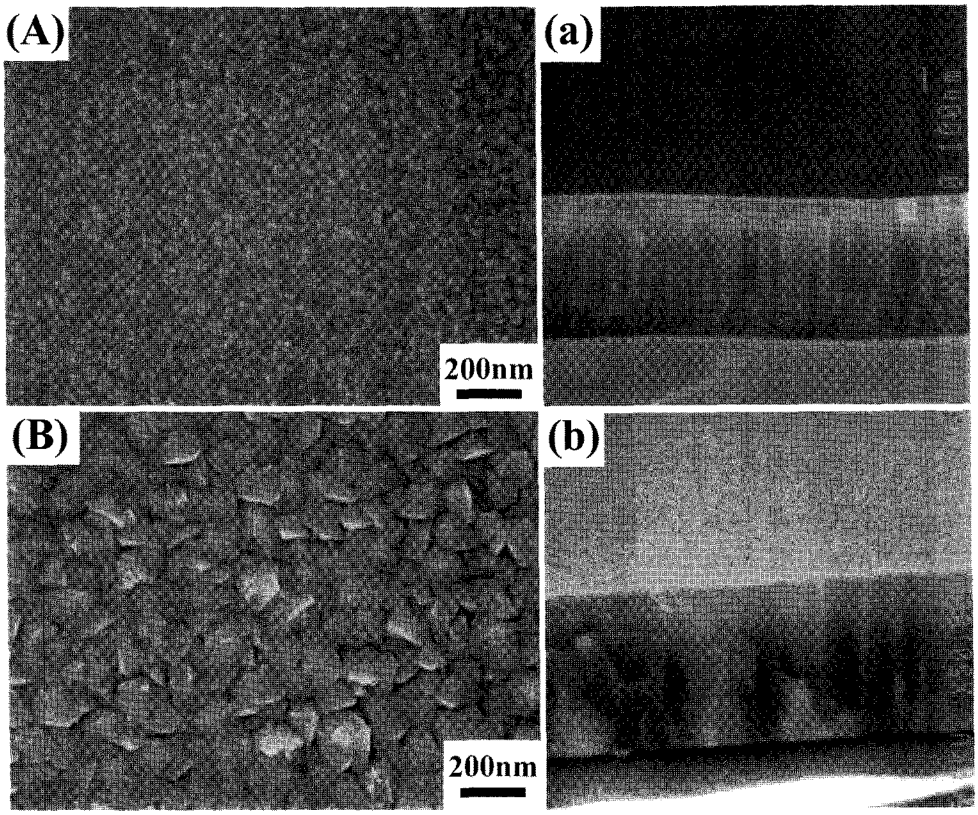

Fig. 8. FE-SEM images of surface and cross-section of AlN films deposited on glass substrate at a nitrogen partial pressure $PN_2 = 0.052$; (A) for 300 mA and (B) for 500 mA, (a) and (b) are their cross-sectional view, respectively.

on glass substrate at a N_2 partial pressure of 0.52 and a total pressure of 0.4 Pa for 14.4 ks. (A), (B) are the surface morphology of films for sputtering currents of 300 mA and 500 mA respectively and (a), (b) are their corresponding cross sectional views. Hence, it is clear from the figures that the grain size of AlN films increases with increasing sputtering current; AlN films show columnar structure and also the column diameter increases with increasing sputtering current.

The hardness of prepared AlN films: The hardness of AlN thin films prepared with a N_2 partial pressure of 0.52, was measured for several films by a micro-Vickers hardness tester (Rigaku). The Vickers hardness of the film was in the range of 1,050~1,120 Hv.

4. Discussion

From the experimental results, it was found that the voltage is the highest at $PN_2 = 0$ then decreases to the lowest value and again increases slowly with increasing N_2 partial pressure. The origin may be explained by considering that some nitrogen atom ionized and takes part in the sputtering process, and as a result, current increases and the voltage decreases at the constant current. However, the plasma behavior at higher N_2 partial gas pressures is not clear.

From the results, it was found that AlN did not deposit below the partial pressure $PN_2 =$ 0.22 and also that the deposition rate abruptly falled down from 0.14 nm/s to 0.043 nm/s in the partial pressure range of N_2 0.19 to 0.22. Therefore, it could be said that the critical nitrogen partial pressure for the formation of AlN compound is between 0.19~0.22. The abrupt fall of deposition rate was attributed to the compound formation of AlN on the target surface, resulting in reduction of the sputtering yield.

The relative peak intensity of preferred-oriented 0002 plane of AlN is the highest at the partial pressure of $PN_2 = 0.52$. It may be related to the best suitable ratio of Ar and N_2 gas mixture to form best crystalline AlN with this specific setting of this apparatus. The XRD patterns and TEM observations are consistent with the above results.

The growth rate of nuclei of AlN compound may be higher for higher sputtering currents. As a result, the grain size and the diameter of the columns as well increase with increasing sputtering current.

5. Conclusion

Pure AlN thin films were fabricated by using a specially designed sputtering apparatus, and their deposition characteristics, microstructure and texture were investigated. The main results are summarized as follows:

1. Deposition of AlN film depends mainly on N_2 partial pressure. In this system AlN films does not deposit below $PN_2 = 0.22$ and also the deposition rate abruptly falls down from 0.14 nm/s to 0.043 nm/s in the range of $PN_2 =$ 0.19~0.22, which is the critical partial pressure for the formation of AlN.

2. The best preferred-oriented AlN thin films can be deposited at a nitrogen partial pressure of $PN_2 = 0.52$ due to the best suitable ratio of Ar and N_2 gas mixture.

3. As-deposited AlN films show preferred orientation and columnar structure; large columns are observed for higher sputtering currents. The grain size of AlN films increases with increasing sputtering current, probably ascribed to the higher growth rate of AlN nuclei at higher sputtering current.

Acknowledgment

"This research was supported by the Academic Research fund of Hoseo University in 2008" (2009-0036).

References

1. A. Mahmood, N. Rakov and M. Xiao : Mater. Lett., **57** (2003) 1925.
2. Z. An, C. Men and Z. Xu : Surf. & Coat. Tech., **196** (2005) 130.
3. C. L. Aardahl, H. K. Yun and Y. Ono : Thin Solid Films, **346** (1999) 174.
4. K. S. Kao, C. C. Cheng and Y. C. Chen : Appl. Ferroelect., Proc. 13th IEEE Int.- Symp. (2002) 403.
5. L. Zhang, E. F. McCullen and M. H. Rahman : Sensors & Actuators. B, **113** (2006) 843.
6. E. A. Chowdhury, J. Kolodzey and J. O. Olowolafe : Appl. Phys. Lett., **70** (1997) 2732.
7. X. D. Wang, K. W. Hipps and J. T. Dickinson : J. Mater. Res., **9** (1994) 1449.
8. Y. Hara, N. Kitazawa and Y. Watanabe : Ceramic Soc. of Japan, (1999) 464.
9. X. Li, X. Xu and H. Wu : J. Inorg. Mater., **20** (2005) 1481.
10. X. Q. Shen, Y. Tanizu and T. Ide : Phys. Status Solidi. C, Conf. & Critical Rev., **7** (2003) 2511.
11. T. Kitahara, N. Ichikawa and Y. Nomoto : Vacuum, **82** (2007) 109.
12. K. Hara, A. Sato and K. Azumada : Phys. Status Solidi. C, Conf. & Critical Rev., **7** (2003) 2274.
13. H. E. Cheng, T. C. Lin and W. C. Chen : Thin Solid Films, **425** (2003) 85.
14. N. Rakov, A. Mahmood and M. Xiao : Scripta Mater., **50** (2004) 589.
15. A. Mahmood, N. Rakov and M. Xiao : Mater. Lett., **57** (2003) 1925.
16. D. Manova, V. Dimitrova and W. Fukarek : Surf. & Coat. Tech., **106** (1998) 205.
17. J. A. Ruffner, P. G. Clem and B. A. Tuttle : Thin Solid Films, **354** (1999) 256.