AlN thin film growth using electron cyclotron resonance reactive sputtering

N. H. Hung, H. Oguchi, H. Kuwano
Department of Nanomechanics, Tohoku University, Japan
E-mail: hung@nanosys.mech.tohoku.ac.jp

Abstract. In this report, we investigated conditions to deposit stoichiometric aluminium nitride (AlN) thin films grown on (100)-oriented Si substrates under various Ar/N_2 gas flow rates at a wide range of temperature from room temperature (RT) to 350°C using electron cyclotron resonance (ECR) reactive sputtering. This study revealed that stoichiometric of thin film can be controlled by N_2/Ar flow rate and that stoichiometric N/Al = 1 was archived at N_2/Ar = 2. This study also revealed that crystallinity can be controlled by substrate temperature. From RT to 200°C, thin films were amorphous or poly-crystal, at 350°C however, thin film was mainly [110] and [100] AlN. Obtained thin films are densely packed and have very low root mean square (RMS) roughness of 0.41 nm which is much less than other sputtering methods.

1. Introduction
AlN is one of the III-nitride semiconductors having a large direct band gap of about 5.9–6.02 eV [1], high hardness, and good thermal conductivity [2]. In recent years, much attention has been paid on fabricating AlN thin films for applications in a micro-electronics field, especially as an important component of energy harvesters [3] and surface acoustic wave (SAW) [4] or bulk acoustic wave (BAW) [5] devices. It has been the most standard lead-free piezoelectric material for such application because of the high piezoelectric properties.

For AlN powder production, there are two major processes that were used [6]: direct nitridation of aluminium in presence of N_2 or NH_3 [7] and carbon thermal reduction of alumina [8]. For thin films, however, only the former approach likely to be available. For example, metal organic chemical vapour deposition [9], plasma-assisted molecular beam epitaxy [10], and chemical vapour deposition [11] have been used so far. In addition to these techniques, RF and DC reactive sputtering were often chosen since those techniques are suitable to prepare high quality AlN thin films [12]. Furthermore, these techniques allow us to control film stoichiometry and crystallinity by adjusting flow rate of gases during deposition.

ECR sputtering is another type of sputtering methods. This method can improve crystallinity [13] more effectively than other sputtering methods even at lower growth temperature due to activated plasma formed by reactive gases (in our case, nitrogen). It also improves density and flatness of the thin films that is favourable for micro device fabrication [3]. So far, however, ECR sputtering has been seldom reported for AlN films [13], [14].

In this study, we investigate the composition of film deposited by ECR sputtering by changing N_2/Ar gases flow rate, and crystallinity of thin film by changing substrate temperature. Furthermore, we evaluate density and flatness of the film

2. Experiments

Figure 1 shows an illustration of the ECR ion beam sputtering system used for AlN deposition. The system has two ion-guns that generate plasma to sputter targets or to N_2 gas to facilitate gas-film reaction. Nitrogen gas is introduced into the chamber directly or through the ion gun. The deposition rate of films grown on the (100) silicon substrates is 0.9 nm per minute averagely. Typical sputtering parameters were shown in Table 1. Prior to AlN film deposition, silicon substrates were cleaned by two standard cleaning processes of Radio Corporation of America (RCA1 and RCA2) in order. The Al-target was pre-sputtered before each deposition for 30 minutes in Ar and then 10 minutes in the mixture of Ar and N_2. Thickness of the films was measured by a surface profiler. Field emission scanning electron microscopy (FE-SEM, Hitachi SU-70) was used to perform morphological analysis. The composition was confirmed by using the EDX spectroscopy and the crystal structure of the samples was measured using an X-ray diffraction (XRD, Bruker D8). An atomic force microscopy (AFM, Nikon TE2000-U) was used to examine thin film's surface roughness.

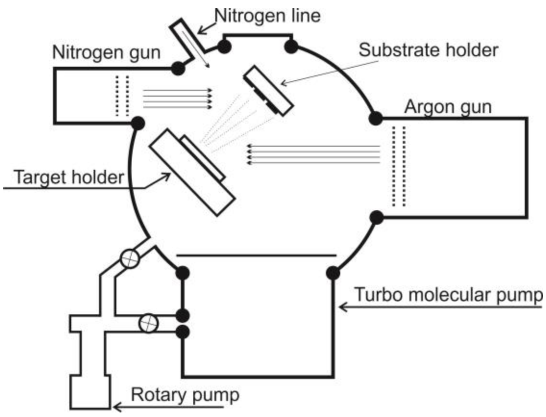

Figure 1. A schematic of the ECR apparatus

Table 1. Deposition conditions

Substrate temperature	RT – 350°C
Ar gas flow rate	0.6 sccm
N_2 gas flow rate	0 – 1.2 sccm
Ar ion beam current	12 mA/cm^2
N_2 ion beam current	0 and 4 mA/cm^2
ECR microwave power	100 W (both guns)
Ar accelerating voltage	2000 V
N_2 accelerating voltage	20 V
Back pressure	5x10^{-3} Pascal
Depositing pressure	0.33 Pascal

3. Results and discussions

To optimize the growth condition of Al nitridation, we investigated compositions of the thin films deposited at RT by changing N_2/Ar gas flow rate (Figure 2). Composition of the films increases almost linearly with N_2/Ar flow rate, and reaches to almost stoichiometric value (N : Al ~ 1 : 1) when the ratio of the N_2/Ar = 2.

Figure 3 shows XRD 2θ-θ patterns of the stoichiometric films deposited at different temperatures. Thin films did not show any peak of AlN for growth temperature of up to 200°C, regardless of normal and activated nitrogen. However, at 350°C, a peak of AlN (110) and (100) diffraction planes appeared, indicating that crystallinity of the film was improved by increasing substrate temperature. Judging from the intensity ratio, a primary AlN domain has [110] orientation normal to the substrate.

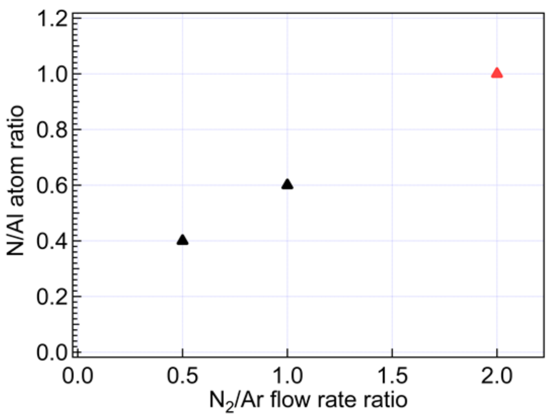

Figure 2. Dependence of Al and N ratio in the thin film deposited at room temperature as a function of Ar and N_2 flow rate.

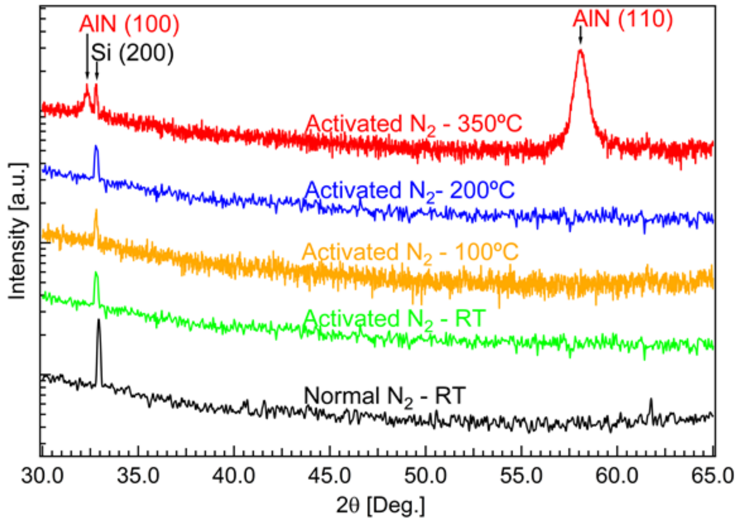

Figure 3. X-ray 2θ–θ scan of AlN deposited on Si at various substrate temperatures.

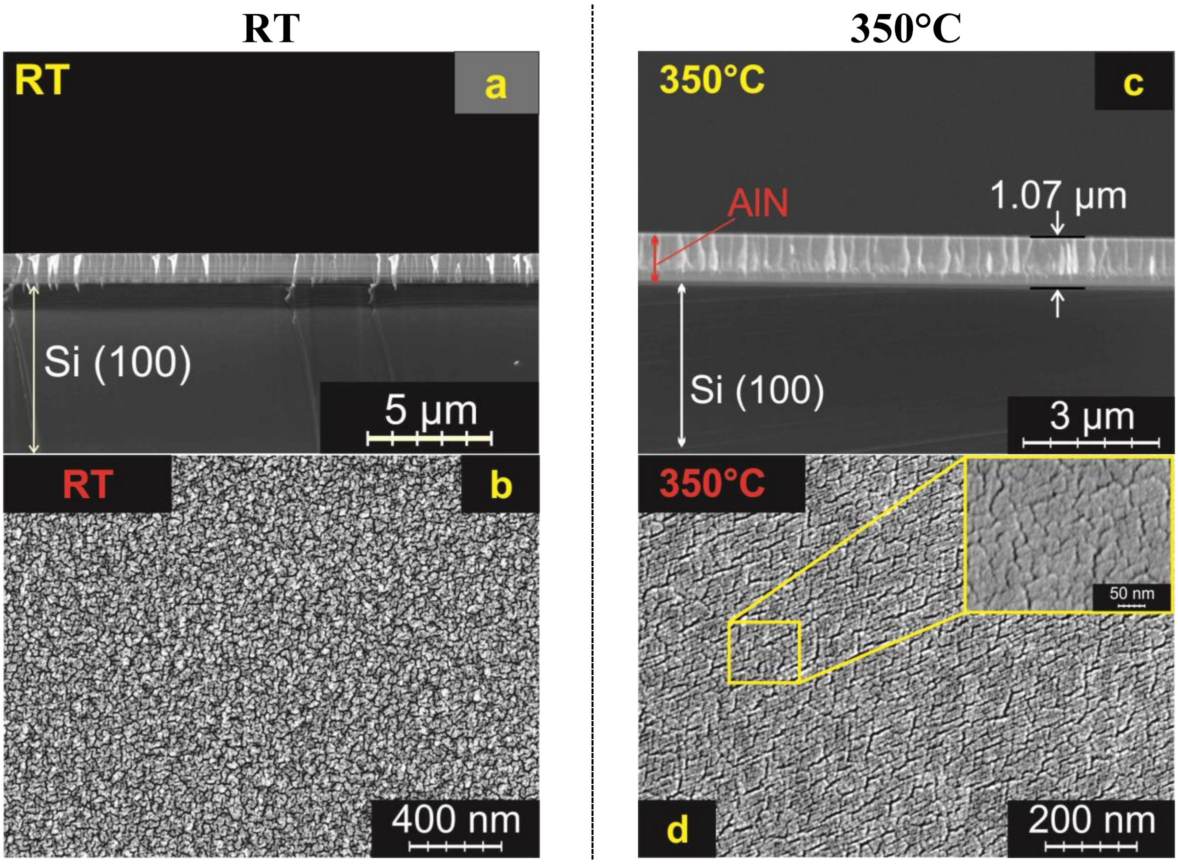

Figure 4. Cross-sectional FE-SEM and plane view images of AlN thin film deposited at RT and 350°C.

Typical cross-sectional and plane view FE-SEM images of the thin film deposited at RT and 350°C were shown in Figure 4. As shown in the figure 4(a) and 4(c), the cross-sectional images of the thin films deposited at RT and 350°C, respectively reveal that the both films are densely packed with high homogeneity and surface flatness. Surface morphologies of the films were shown in the figure 4(b) (deposited at RT) and figure 4(d) (deposited at 350 °C). One could be seen from the images, the surface of thin films kept almost identical for low and high temperature deposition. Cracks which were observed on the surface of thin films have widths in range of 7 – 10 nm. The cross-sectional image revealed that the cracks did not go through the thin film to the substrate (vertical lines on the images were created when sample had been broken). This will help avoiding short between top and bottom electrodes that is often a problem suffered by researchers using films grown by normal sputtering in micro device fabrications.

Figure 5 shows an AFM image (1 μm x 1 μm) of the thin film surface. Closely packed pebble-like nanostructure was observed. The measured RMS roughness and peak to valley value of the typical films are 0.41 nm and 3.8 nm, respectively. These values are clearly less than those of the films deposited by RF sputtering [15]-[17]. The flatness of our films is preferable in, for instance, SAW devices [15] which favour as low roughness as possible. It should be noted that roughness of thin films that was deposited by other sputtering methods often increase with substrate temperature while AlN prefers high temperature.

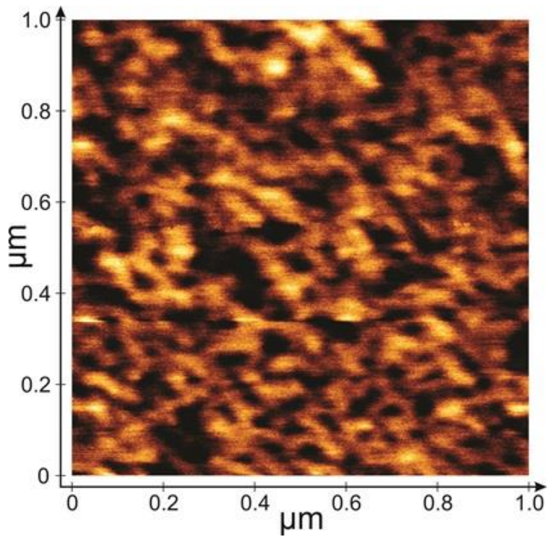

Figure 5. An AFM surface image of AlN thin film deposited at 350°C.

4. Conclusion

We investigated the optimum growth conditions to obtain stoichiometry and high crystallinity in the AlN thin films growth by ECR sputtering. The composition was controlled by changing N_2/Ar gas flow ratio during the film growth, and realized stoichiometry at N_2/Ar = 2. Crystallinity of the AlN was poor for the films deposited at the temperature range between RT and 200°C, but improved at 350°C. The AlN film deposited at 350°C are closely packed and consisted mainly of [110]-oriented domains. Its thickness is homogeneous and surface is flat as proved by RMS roughness of 0.41 nm. These results indicated that the AlN films grown by the ECR sputtering is suitable to fabricate functional micro-electromechanical devices.

References

[1] Shah A and Mahmood A 2012 *Phys. B Cond. Matter.* **407** 3987.
[2] Ait Aissa K, Achour A, Camus J, Le Brizoual L, Jouan P Y and Djouadi M A 2014 *Thin Sol. Fil.* **550** 264.
[3] Zhang J, Cao Z, and Kuwano H 2011 Jpn. J. Appl. Phys. **50** 09ND18.

[4] Chen S W, Lin H F, Sung T T, Wu J D, Kao H L and Chen J S 2003 Elec. Lett. **39** 1691.
[5] Hara M. and Kuwano H. 2012 Jpn. J. Appl. Phys. **51** 07GC11
[6] Sakurai T, Yamada O and Miyamoto Y 2006 *Mater. Sc. Eng.* A **415** 40.
[7] Weimar A W, Cochran G A, Eisman G A, Henley J P, Hook B D, Mills L K, Guiton T A, Knudsen A K, Nicholas N R, Volmering J E and Moor W G J. 1994 *Am. Ceram. Soc.* **77** 3.
[8] Lefort P and Billy M 1993 *J. Am. Ceram. Soc.* **76** 2295.
[9] Chen Y, Song H, Lia D, Suna X, Jianga H, Lia Z, Miaoa G, Zhanga Z and Zhoub Y 2014 *Matter. Let.* **114** 26.
[10] Yusoff M Z M, Mahyuddinc A, Hassanb Z, Yusofb Y, Ahmadb M A, Chinb C W, Hassanb H A and Abdullahb M J 2013 *Supper. Mic.* **60** 500.
[11] You Y, Ito A, Tu R and Goto T 2013 *J. Crys. Grow.* **365** 1.
[12] Schneider J M, Rohde S, Sproul W D and Matthews A 2000 *J. Phys. D. Appl. Phys.* **33** R173.
[13] Okano H, Tanaka T, Shibata K and Nakano S 1992 *Jpn. J. Appl. Phys.* **31** 3017.
[14] Ryunosuke H, Takeshi M, Masayoshi E and Shuji T 2010 *IEEJ Trans. Sens. Micr.* **30** 523.
[15] Kuang X, Zang H, Wang G, Cui L, Zhu C, Jin L, Sun R and Han J 2012 *App. Surf. Sc.* **263** 62.
[16] Vashaei Z, Aikawa T, Ohtsuka M, Kobatake H, Fukuyama H, Ikeda S and Takada K 2009 *J. Crys. Grow.* **311** 459.
[17] Kar J P, Boss G and Tuli S 2005 *Mater. Sc. Sem. Proc.* **8** 646.