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Reduction of Defects on Microstructure Aluminium Nitride Using High Temperature Annealing Heat Treatment

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Abstract. Aluminium Nitride (AlN) is a ceramic 111-nitride material that is used widely as components in functional devices. Besides good thermal conductivity, it also has a high band gap in emitting light which is 6 eV. AlN thin film is grown on the sapphire substrate (0001). However, lattice mismatch between both materials has caused defects to exist along the microstructure of AlN thin films. The defects have affected the properties of Aluminium Nitride. Annealing heat treatment has been proved by the previous researcher to be the best method to improve the microstructure of Aluminium Nitride thin films. Hence, this method is applied at four different temperatures for two hour. The changes of Aluminium Nitride microstructures before and after annealing is observed using Transmission Electron Microscope. It is observed that inversion domains start to occur at temperature of 1500 ℃. Convergent Beam Electron Diffraction pattern simulation has confirmed the defects as inversion domain. Therefore, this paper is about to extract the matters occurred during the process of producing high quality Aluminium Nitride thin films and the ways to overcome this problem.

1. Introduction

AlN was discovered in 1862 by F. Briegler and A. Geuther. However, the potential of this material for functional devices component only been applied after 1980 [1]. Since then, the demand for this 111-nitride material gained high attention among the industries due to costing, easy to produce and easy to machine compared to other ceramic materials. Most recently, AlN has been used widely in electronic components such as Light Emitting Diode (LED), Laser Diode (LD) and photo detector for ultraviolet region [2]. Until today, more than 20 companies worldwide are developing AlN technology.

AlN is a 111-nitride ceramic material produced from the covalent bonding between Aluminium (Al) and Nitrogen (N). The arrangement of the atomic positions in AlN microstructure placed non-tetrahedral or non-centro symmetric positions. The positions called Wurtzite crystal structure. Due to that, the polarity exhibits the (0001) axis give AlN the piezoelectric properties [3]. AlN also good in emitting lights with shorter wavelength of 210 nm and high band gap up to 6 eV.

Other promising properties by this material are high thermal conductivity, good dielectric, low thermal expansion coefficient and non-reactive to normal semiconductor process chemical and gaseous [4]. Thermal conductivity of AlN is $140 W/mK$ to $180\ W/mK$. However, previous researcher reported that the thermal

conductivity of polycrystalline AlN at room temperature is in the range of $100\ W/mK$ to $260\ W/mK$ and for single crystal can reach until $300\ W/mK$ [4]. The different of thermal conductivity due to the density of oxygen impurities presence along the microstructure of AlN. In other words, thermal conductivity level of material is inversely proportional to the presence of defects in their microstructure [5].
The growth process of AlN thin films on sapphire substrates simultaneous to the existence of defects such as inversion domains and dislocations. This conditions becoming the major problems among the manufacturers. J. Jasinski et al identified the formed of Al polarity inversion domains on AlN layer growth on sapphire substrate [6]. The columnar inversion domain formed at boundaries that deviated $2\ \pm 0.5°$ from c-axis in V-like shaped. Figure 1 and Figure 1.1 shows the dark field TEM images of AlN layer recorded with a g-vector perpendicular and parallel to c-axis.

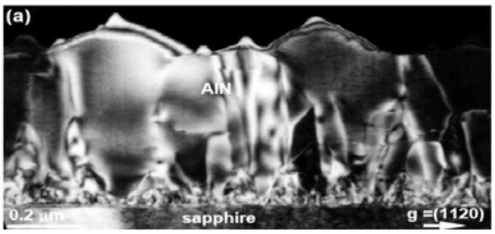

Figure 1. Dark-field TEM images of the AlN layer recorded with a g-vector perpendicular to c-axis [6]

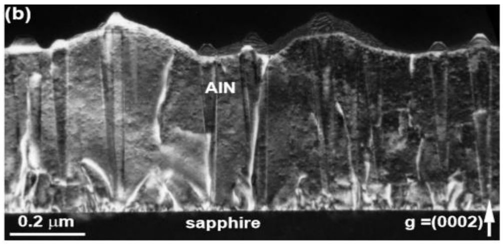

Figure 1.1 Dark-field TEM images of the AlN layer recorded with a g-vector parallel to c-axis [6]

From the images the boundaries of inversion domain along the AlN boundary shows the strong strain contrast. This is indicated that there is a displacement associated with these boundaries along the c-axis. Before this, the treatment using annealing already done to overcome this problem at the temperature less than $1500\ ℃$. However, the defects exist in the AlN microstructure after annealing process still considers as high percentage. Therefore, this paper will be focused on annealing temperature of $1500\ ℃, 1550\ ℃, 1600\ ℃$ and $1650\ ℃$. The temperature set is still below the melting point of AlN (T_{AlN} $= 2200\ ℃$) and this material is very stable at the high temperature [6].

2. Experimental Procedure

2.1 Growing process of AlN on the sapphire substrate

Before going further with annealing heat treatment process, AlN thin films must be growth on (0001) plane of sapphire substrate. On the (0001) plane of sapphire, the c-axis of AlN is aligned normal to the substrate surface, while on sapphire (0112), the c-axis of AlN is inclined at about $28\ ℃$ to the film normal. This situation called to be the different in lattice mismatch between both AlN thin films and sapphire that lead to the existence of defects on the microstructure of AlN thin films. The existence of defects on the AlN microstructure affected the effectiveness of this films when applied in the functional devices. Growing of AlN thin films on sapphire substrates done in the Metal Organic Vapor Phase Epitaxy Reactor (MOVPE) machine. Basically, this machine process based on the chemical deposition principle. Trimethylaluminium (TMAl) were supplied in the machine and after 10 seconds Ammonia (NH_3) introduced. During the reaction take places, Hydrogen gas (H_2) were flowed as a gas carrier under a low-pressure of 13.3 kPa. Along the process, the concentration of these substances controlled to get the desired alloy decomposition over the whole range with velocity $100\ ms^{-1}$. Lastly, AlN thin film with thickness 4.5 μm produced. However, this thickness is beyond the limitation to be observed under the Transmission Electron Microscope (TEM).

2.2 Focused Ion Beam (FIB)

To comply the TEM requirement the material must be thicken less than $200\ nm$. Focused Ion Beam (FIB) HITACHI ML-4000L was introduced for cross sectioning the AlN thin film. Firstly, AlN thin film on sapphire substrate is attached on the thicker glass by using a wax. Sample then cut using cutter into $2\ mm\ \times 4\ mm$ and $2\ mm\ \times 3\ mm$ respectively. Next, the samples coated with carbon for 20 times in 10 s until the coating thickness reach $20\ mm$ to $30\ mm$. Noted that the carbon used was already heated for $30\ s$ at $5\ V$. Before furthering the process, FIB were setup then the samples loaded using rod. After that, the process of sloping, etching and deposition on the samples taken place. Lastly, the samples picked up by using W needle. During this process, precaution must be concern to avoid needles clashes with the stages. Noted that the needle must be at their eucentric positions when picked up the samples.

2.3 Annealing heat treatment process

Annealing heat treatment is a process to heat the microstructure of AlN thin film. Previous research has proved that annealing heat treatment has ability in improving the microstructure of AlN thin film besides increasing the mechanical and electrical properties [9]. The process involved is to heated AlN thin films until reaches it critical temperature but lower than it melting point, $T_{AlN} = 2200$ ℃. AlN thin films on sapphire substrates annealed t temperatures of 1500 ℃, 1550 ℃, 1600 ℃ and 1650 ℃ for two hours.

2.4 Transmission Electron Microscope (TEM)

The final stage for this research is observing and analyzing the conditions on the microstructure of AlN thin films after applying the annealing heat treatment. As mentioned above, the thickness of AlN thin films must be less than $200\ nm$ in order to get the better imaging quality. So, an electron beam can easily passes through the cross sections of AlN thin film. The thickness of specimens set to be $50\ nm$ and $100\ nm$. An electron beam shoot with the high velocity at the accelerating voltage of $300\ kV$.

3. Results and Discussion

3.1 $T_{annealing} = 1500$ ℃

Figure 2 is bright field image TEM results of AlN thin film microstructure after facing the annealing heat treatment at temperature of 1500 ℃. The thickness of specimen is $50\ nm$. From the observation, cone shaped like of inversion domain was formed along the $[\bar{1}2\bar{1}0]$ zone axis. The line of defect also can be seen clearly in the image. At this stage the density of inversion domain boundaries is high due to the lattice mismatch between AlN thin film and sapphire substrate (0001) during the growth process.

Figure 2. Bright field image TEM of AlN thin film microstructure after annealed at temperature of 1500 ℃.

Other than inversion domain, dislocation defect type also found along the boundary of AlN microstructure. Convergent Beam Electron Diffraction (CBED) pattern has confirmed the defects through the inverted polarity region between region A and region B as shown in Figure 2.1. The CBED pattern analyzed the AlN microstructure along $[2\bar{1}\bar{1}0]$ zone axis. Both region also shows different polarity. Region A shows more on Al polarity while region B is N polarity. The quality of AlN thin film at this condition is very poor.

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This is due to the low thermal conductivity that affected by the inversion domain and not suitable for functional devices.

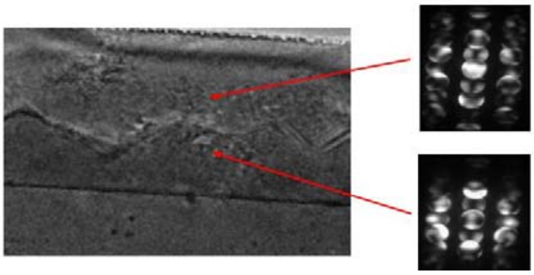

Figure 2.1. CBED pattern of AlN microstructure along $[2\overline{1}\overline{1}0]$ zone axis.

3.2 $T_{annealing} = 1550$ ℃

Figure 3 is bright field image TEM of AlN thin films microstructure after annealed at 1550 ℃. The specimen thickness is 100 nm. The formation of inversion domain on the microstructure of AlN thin film shows some changed compared to the annealed at 1500 ℃. The cone shaped of inversion domain is reduced into the wavy shaped. Other than that, the surface boundary also seen to be sharper. The formation of wavy shaped boundary due to the incline in X and Y axis. The sharp boundary is because it is parallel with the incident beam. At this stage the density of inversion domain boundary still consider as high percentage for efficient function. Besides that, the polarity mechanisms from CBED pattern analysis shows only small changes between both region A and region B.

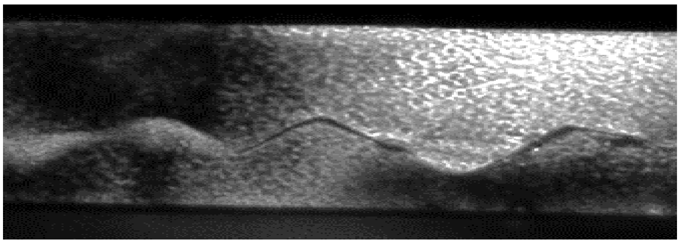

Figure 3. Bright field image TEM of AlN thin film microstructure after annealed at temperature of 1550 ℃.

3.3 $T_{annealing} = 1600$ ℃

Figure 4 is the bright field image TEM of AlN thin film microstructure after annealed at temperature of 1600 ℃. The thickness set for this specimen is 100 nm. At this level of temperature, the microstructure of AlN become flatter and smoother compared to before. The length of wavy shaped is increased compare to before and the sharp boundary also less visible. That condition shows the decreasing for density of inversion domain boundaries along the AlN microstructure.

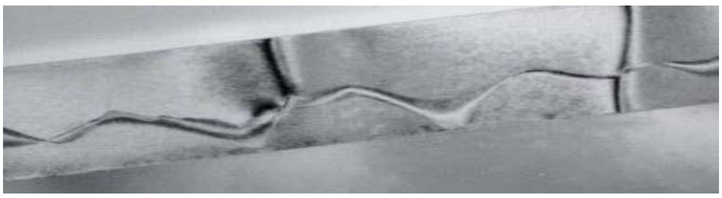

Figure 4. Bright field image TEM of AlN thin film microstructure after annealed at temperature of 1600 ℃.

3.4 $T_{annealing} = 1650$ ℃

Lastly, AlN thin film annealed at the temperature of 1650 ℃. From the observation of TEM image as shown in Figure 5, the surface of AlN thin film microstructure become fine and the grain boundary seem to be smoother. At this stage the density of inversion domain boundary is stable for functional devices.

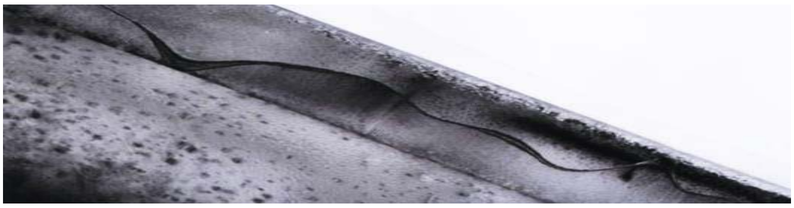

Figure 5. Bright field image TEM of AlN thin film microstructure after annealed at temperature of 1650 ℃.

4. Conclusion

The existence of defects inside the AlN microstructure was the major effect in producing the high quality of thin films. The poor thermal conductivity of AlN thin film is due to the high density of inversion domain boundaries found along it microstructure. The defects started to occurred during growth process of AlN thin films on the sapphire substrate (0001) due to the lattice mismatch. Annealing heat treatment at different temperature (1500 ℃, 1550 ℃, 1600 ℃ *and* 1650 ℃) can reduced the defects inside AlN microstructure. The defect mechanism formation has shown through the TEM imaging techniques. At temperature of 1500 ℃, inversion domain can be seen clearly and become less visible until 1650 ℃. The inverted polarity from CBED pattern between Al and N along the AlN microstructure has confirmed the defect type is inversion domains. Therefore, annealing heat treatment is effective in improving the AlN microstructure. Hence, this method contributed in developing the high quality of AlN thin films then producing the high efficiency functional devices for customer satisfaction.

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