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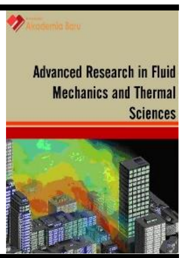

Effect of High-Temperature Annealing Heat Treatment to Microstructure of Aluminium Nitride on Sapphire Substrate

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ABSTRACT

Aluminium Nitride (AlN) is classified in the 3rd nitride ceramic materials and able to emit the lights at shorter wavelength of 210 nm with high band gap up to 6 eV. In addition, with thermal conductivity up to 140 W/mK has made this material in demand for electronic devices applications such as light emitting diode, laser diode and photodetector for the ultraviolet region. However, previous researchers have confirmed the existence of defects in the microstructure of AlN that affected the properties of this material during the growth process. Hence, high-temperature annealing heat treatment is utilized as one of the methods to cure the problems of AlN thin film. Therefore, this paper is about to identify the effects of high-temperature annealing heat treatment on the microstructure of AlN. The changes on microstructure were observed through Transmission Electron Microscopy after the annealing process. The results showed an increase of the annealing temperature contributed to the reduction of defects in the microstructure of AlN.

Keywords:
Aluminium Nitride, AlN, Annealing Heat Treatment, Inversion Domain, Microstructure

1. Introduction

Aluminium nitride (AlN) is a semiconductor ceramic material that made up from covalent bonding between aluminium (Al) and nitrogen gas (N_2). Although F. Briegler and A. Geuther discovered this material in 1862, its potential for electronic devices only been noticed after the year 1980 [1]. Since then, this material was introduced commercially and gained high demands among the industries. This is due to its good properties for electronic devices such as high thermal conductivity, low thermal expansion coefficient, high electrical resistivity, high thermal stability in inert atmosphere and non-reactive to normal semiconductor process chemical and gaseous [2, 3]. The wurtzite crystal structure as shown in Figure 1 was found in AlN materials which contributed to piezoelectric properties as needed in the electric and electronic components [4].

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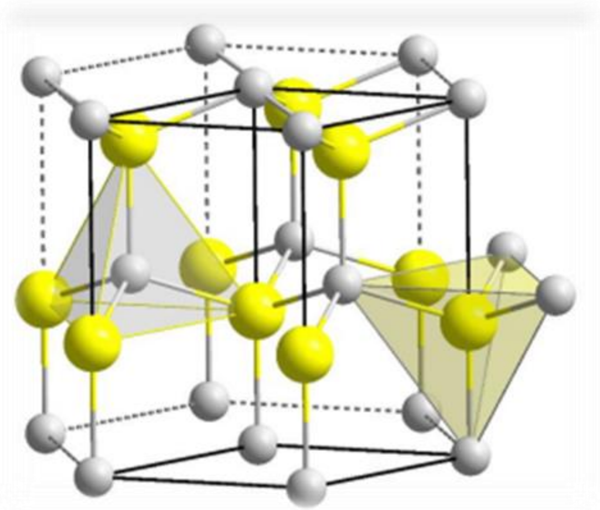

Fig. 1. Wurtzite structure of aluminium nitride [4]

Furthermore, to equip the requirement of electronic components the properties of thermal conductivity is a must to enhance its capability. Therefore, AlN is one of the suitable materials that have this advantage. For example, AlN thermal conductivity values are changing depending on three conditions. In normal conditions, the thermal conductivity range is between 140 W/mK to 180 W/mK, for polycrystalline aluminium nitride the range is between 100 W/mK to 260 W/mK and for a single crystal can reach up to 300 W/mK [5]. These three conditions are influenced by the level of oxygen impurities density which is presence in the microstructure of AlN [6]. Theoretically, the higher the thermal conductivity of the materials will reduce the presence of defects inside its microstructure [7].

At present, the prior usage of AlN thin film material is regards to producing the lights. This is because the material can emit lights at shorter wavelength of 210 nm with high band gap up to 6 eV [8], [9]. The examples of devices utilizing this AlN material including light emitting diode (LED) shown in Figure 2, laser diode (LD) and photodetector for the ultraviolet region [8]. The demand from miniaturization of consumer electronics, AlN has been selected as one of suitable carrier for electronic components especially in automotive, telecommunications, appliances and many more [10, 11].

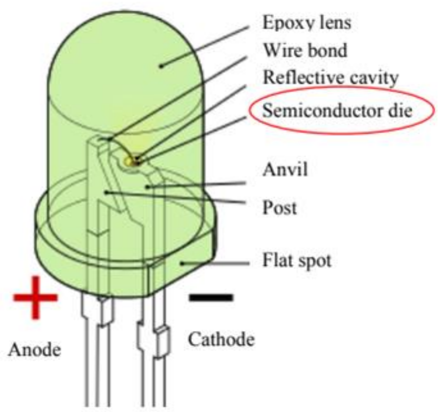

Fig. 2. LED components [8]

Therefore, growing process of AlN thin film has been studied throughout the decade. Until today, almost more than 20 companies worldwide involved in developing this product [12]. Generally, there are several established methods to synthesize AlN, they are known as nitridation of aluminium under nitrogen and ammonia gas flow, carbothermal reduction nitridation, self-sustain high-temperature synthesis and gas-reduction nitridation [13]. In this research, the method chosen is an AlN growth through nitridation of aluminium under nitrogen and ammonia gas. Previous researchers have tried to growth an AlN thin film on a sapphire substrate in laboratories [14–19]. However, the result found unsatisfactory due to the presence of defects such as inversion domain and dislocation in the microstructure of this material. Other than that, a large lattice mismatch and high difference in thermal expansion between the AlN layer and the sapphire substrate were identified during the growth process. The relationship between the lattice defect structure and properties of AlN has also been studied in [20, 21], however very few annealing heat treatment methods has been introduced to overcome these problems.

Annealing heat treatment is found to be one of the methods to curing the microstructure and surface of semiconductor materials. The advantages of this treatment will increase the mechanical and electrical properties of the materials [22, 23]. The standard processes are followed where firstly, heating the materials until reaches its critical temperature. Next, the temperature is maintained for few hours and lastly its cooled at a suitable temperature. Annealing heat treatment on AlN was already done by many researchers but at the temperatures below 1500℃ [24, 25]. These due to the existences of defects inside the microstructure of this material where they were considered unstable to be implemented for functional devices. Therefore, this paper is aimed to investigate the effect of high annealing temperature from 1500℃ to 1650℃ on the microstructure of the aluminium nitride thin film.

2. Methodology

The quality of AlN thin film plays an important role in producing a high-efficiency functional device. Thus, careful procedures are essential during performing the growing process of this film. This is because the AlN film is tiny and breakable. Figure 3 shown the stages of AlN thin film production.

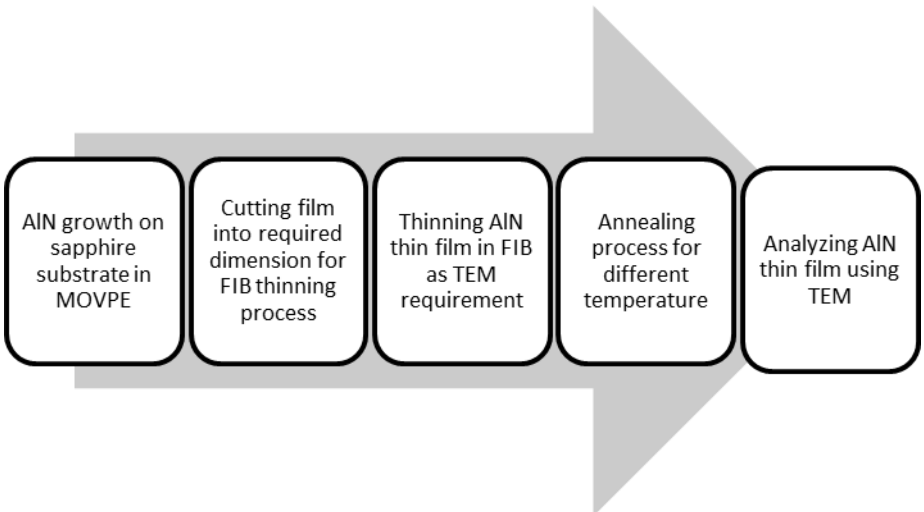

Fig. 3. The Stages of Aluminium Nitride Thin Film Production

2.1 Metal Organic Vapor Phase Epitaxy Reactor (MOVPE)

AlN thin film is grown on sapphire substrate. It begins with a Trimethylaluminium (TMAl) supplied into the MOVPE machine. Then, as 10 seconds passed an ammonia (NH_3) are supplied into the machine and mixed with Trimethylaluminium. As the reactions take places, hydrogen (H_2) flowed into the mixture at low pressure of $13.3\ kPa$. Hydrogen gas acted as a gas carrier. Along the process, the concentrations of this mixture are controlled with the velocity of $100\ ms^{-1}$,and this step is very important to get the desired alloy decomposition over the whole range. Finally, an AlN thin film with a thickness of $4.5\ \mu m$ is produced.

2.2 Focused Ion Beam (FIB)

FIB functioned as a milling machine to thinning and cutting materials into the size of nanometer scale. For this experiment, AlN thin film is milled using FIB HITACHI ML-4000L model. This process is needed before preparing AlN film analyzed under the Transmission Electron Microscopy (TEM). The AlN thin film must be less than $200\ nm$ of its thickness so that its able to be observed under TEM.

The preparation of samples under the FIB process are following several steps. Firstly, the sample is attached on the thicker glass by using wax. Secondly, the sample is cut into two segments which are $2\ mm\ \times 4\ mm$ and $2\ mm\ \times 3\ mm$. After that, these samples are coated for $400\ times$ in 200 seconds periods; $20\ times\ per\ 10\ seconds$, until its thickness reached between $20\ mm$ and $30\ mm$. The coating material used is carbon which was already heated at $5\ V$ for $30\ seconds$. As the cutting process is done, the samples is next loaded carefully into the sub-chamber by using a rod. Later, the processes of sloping, etching and deposition of samples are continued to be done. After all processes are well monitored, using W-needle the samples are carefully placed inside the FIB machine. Care must be given during the inserting process, where the W-needle shall be in a right distance to ensure no clashes with the stages and finally to maintain its eucentric positions during the picking up samples. The thin films samples of AIN are finally ready under the range of $50\ nm$ to $100\ nm$.

In Figure 4 shown the AIN sample has deposited into the TEM mesh. TEM mesh is functioned to hold the samples during the thinning process. During this process, carbon coating on the sample must not overload as it may result in poor adhesion and easy to fall when deposited into the mesh.

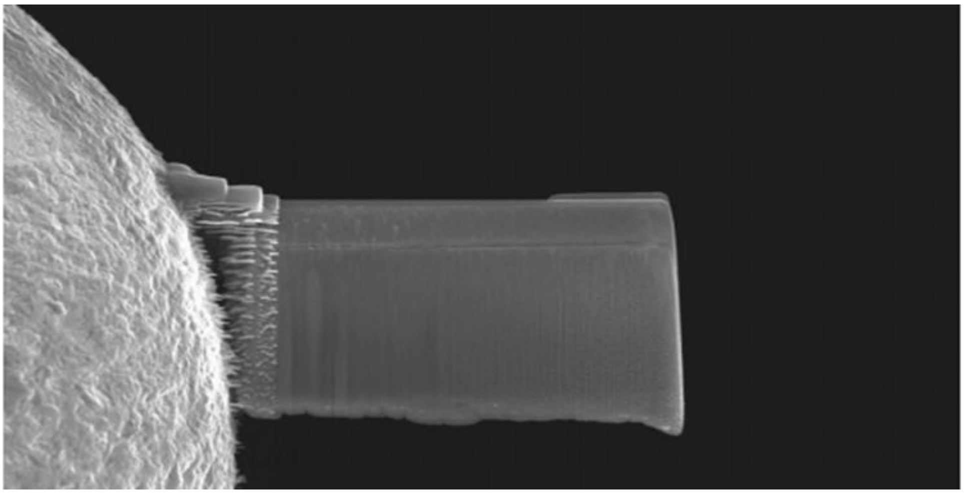

Fig. 4. AlN on sapphire substrate deposited into TEM mesh

Finally, the samples are thinned into three stages which are mid beam, fine beam and u-fine beam conditions. The results of these processes are shown in Figure 5, Figure 6 and Figure 7 respectively.

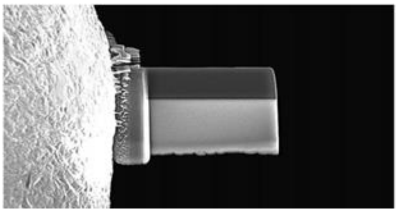

Fig. 5. AIN thinning process in FIB machine in beam condition

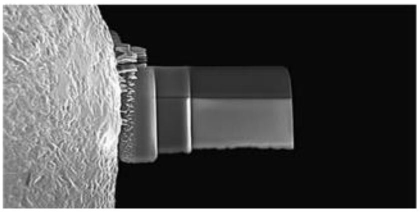

Fig. 6. AIN thinning process in FIB machine in fine beam condition

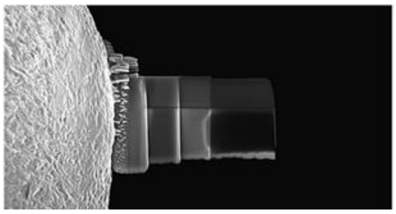

Fig. 7. AIN thinning process in FIB machine in U-fine beam condition

In mid beam stage, a high ion beam energy is applied to the thin film and gradually it removed all of rough surfaces on the sample. Next, the sample is thinning further by a fine beam as in Figure 6. At this stage, the sample is clear from all the remaining roughness on its surface. Figure 7 is the last sample thinning using U-fine beam and during this process, the surface of the sample is polished until its looked smoother and flat. All Above figures shown are captured through the Scanning Electron Microscope (SEM) at $10\ kV$ of scale $5\ \mu$.

2.3 Annealing Heat Treatment Process

The most important step for this experiment is the annealing heat treatment process of AlN thin film. During this process, the temperatures used are 1500 ℃, 1550 ℃, 1600 ℃ and 1650 ℃. The samples are placed into the annealing chamber and heat them at set temperature until it reached its critical temperature for two hours. Lastly, the samples cooled at suitable temperature. It must be noted that the heating process shall be lower than AlN melting temperature of 2200 ℃. After the annealing heat treatment is completed, the samples are then analysed under the Transmission Electron Microscopy.

2.4 Transmission Electron Microscopy (TEM)

TEM is the second important steps where under TEM, the samples can be observed and analysed to characterise the changes on the microstructure of AlN after the annealing heat treatment process. TEM JEM-21000 is the model used to characterize the microstructure of AlN on a sapphire substrate. The samples with thickness of $50\ nm$ and $100\ nm$ were examined. However, must aware that TEM microscope must be setup according to the AIN sample's thickness. The setting of TEM microscope are provided as in Table 1.

Table 1
TEM microscope setting for AlN thin film

No	TEM setting	Value
1	Ion pump	$5 \times 10^{-5}\ Pa$
2	Objective lens	$2.63\ V$
3	Filament image	Symmetric
4	Image resolution sharpen	DEF/STIG
5	Spot knob	1 and 5
6	Current of optimum magnification (Bright Field and Dark Field)	$2.63\ V$
7	Camera length for diffraction mode	$80\ cm$
8	Accelerating voltage	$300\ kV$

3. Results and Discussions

The discussion is based on the difference of annealing heat treatment temperatures.

3.1 Annealing Temperature at 1500℃

Figure 8 is TEM image of AlN microstructure after its annealed at temperature of 1500 °C. The cone liked shape inversion domain can be seen clearly along the AlN microstructure. As previously discussed, this defect formed due to the lattice mismatch between AlN and sapphire substrate. Unfortunately, at this stage the density of inversion domain boundaries is found to be very high, therefore caused the material thermal conductivity is at low level. Thus, with this condition the thin film is unstable to be applied for functional devices.

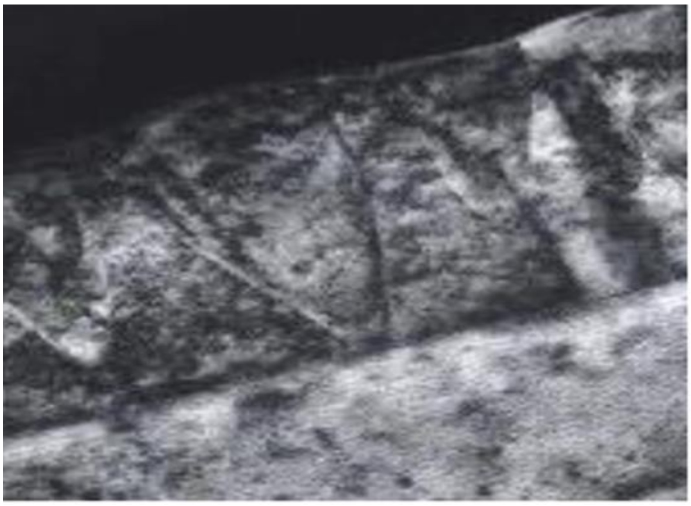

Fig. 8. TEM image of AlN microstructure when annealed at temperature of 1500 °C

3.2 Annealing Temperature at 1550℃

When the sample is annealed at temperature of 1550 °C, the microstructure of AlN found to gain little improvement compared to the previous annealing temperature as shown in Figure 9. One can clearly have observed the cone-like shape is reduced to a wavy shaped which is due to the incline of X and Y axes between both materials. Furthermore, the surface boundary of the microstructure now turned to be a bit sharper and parallel to the incident beam. However, the density of inversion domain boundary is still considered high and need for further treatment.

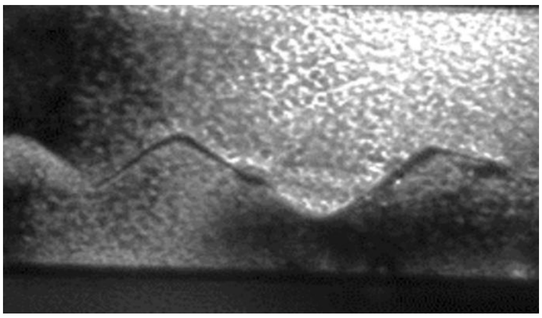

Fig. 9. TEM image of AlN microstructure when annealed at temperature of 1550 °C

3.3 Annealing Temperature at 1600℃

Figure 10 is a TEM image of AlN microstructure after its annealed at the temperature of 1600 °C. Based on the image, the pattern of boundary microstructure of AlN is found rather smooth and flatter. At the same time too, the length of the wavy shape boundary is found to be improved compared to previous treatment. Thus, these conditions indicated the decreasing on density of inversion domain along the AlN microstructure.

Fig. 10. TEM image of AlN microstructure when annealed at temperature of 1600 °C.

3.4 Annealing Temperature at 1650℃

Finally, as AlN thin film is annealed at temperature of 1650 °C. Its grain microstructure shown an utmost improvement compared to previous annealing temperature discussed. This time, the boundary microstructure of AlN had finer growth and the wavy-shaped had improved as well as shown in Figure 11. This signifies the density of inversion domain boundary is at stable condition and improved its thermal conductivity. Therefore, at this stage the film is concluded to be the best condition and suitable to be utilised for efficient functional devices.

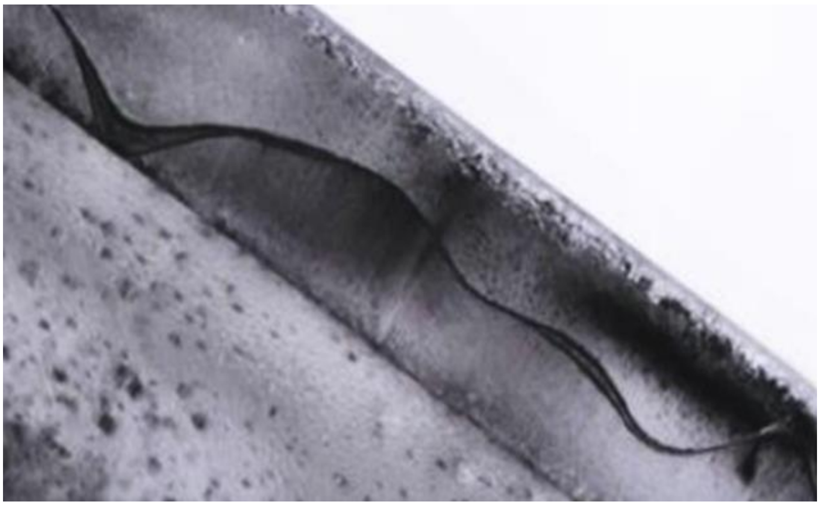

Fig. 11. TEM image of AlN microstructure when annealed at temperature of 1650 °C.

4. Conclusions

In conclusion, annealing heat treatment has affected the grain microstructure of aluminium nitride. In addition, this treatment also reduced the formation of inversion domain defect therefore, increased its thermal conductivity. TEM images have shown the defect in the microstructure of AlN reduced as the annealing temperature is increased. It is proved that at the annealing temperature of 1500 ℃, the inversion domain cone-like shaped is visualized clearly thus this material is considered as unstable. As the annealing temperature is increased to 1650 ℃, the grain microstructure of AlN become smoother and this indicates the inversion domain is reduced significantly. Hence, this signified the material is stable and safe to be applied for functional devices. In nutshell, the high-temperature annealing heat treatment method is proven to be very effective in improving the microstructure of aluminium nitride.

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