

The Electrical Properties of $Al_xGa_{1-x}N$ Thin Films Deposited on Si(111) Substrate by Chemical Solution Deposition Method

¹Heri Sutanto, ¹Iis Nurhasanah, ²Istadi and ²Hadiyanto,

¹Department of Physics, Diponegoro University, Semarang

²Department of Chemical Engineering, Diponegoro University, Semarang

Submitted: Jan 28, 2014; **Accepted:** Apr 18, 2014; **Published:** May 2, 2014

Abstract: $Al_xGa_{1-x}N$ thin films with the GaN buffer layer was deposited on Si(111) substrate by chemical solution deposition (CSD) method. The objective of this research was to evaluate the electrical properties of $Al_xGa_{1-x}N$ with variation of Al mole fraction. The source for Ga and Al were gallium oxide (Ga_2O_3) and aluminum oxide (Al_2O_3), respectively as group III precursors and radical nitrogen resulted by high temperature decomposition of nitrogen gas. The mole fractions of solutions were varied between 15 to 35%. The formed white crystal was dissolved in ethylene diamine to form gel with molarities of 3-6 M. Therefore, the gel was superimposed on substrates Si with spin-coater at speed of 1100 rpm. The formed layer was then sintered at temperature of 900°C such that it decomposed at N_2 gas environment to form $Al_xGa_{1-x}N$. The composition of films was measured by energy dispersive of x-ray (EDX) and the measured Al concentrations were in the range of 9.52% to 24.19%. The electrical transport of the films was measured by means of Hall effect measurement at room temperature. The Hall mobility decreases with the increase of aluminum (Al) concentration and decrease carbon concentration. The resistivity increases with the increase of Al concentration. From the Hall effects measurement the $Al_xGa_{1-x}N$ films have n-type conduction originated from nitrogen vacancy.

Key words: $Al_xGa_{1-x}N$ thin films • Al concentration • Hall mobility • MOCVD

INTRODUCTION

Materials semiconductor based III–V nitride have recently attracted intensive experimental and theoretical interests due to their physical properties, such as a wide and direct band gap, low compressibility and high thermal conductivity, which make them strong candidates for short-wavelength optical devices and high-temperature/high power diodes and transistors[1]. Semiconductor nitrides such as aluminum nitride (AlN), gallium nitride (GaN) and indium nitride (InN) are very promising materials for optoelectronic devices (both emitters and detectors) and high power/temperature electronic devices as have been treated in length and reviewer recently. These materials and their ternary and quaternary alloys cover an energy band gap range of 1.9 to 6.2 eV, which is suitable for band to band light generation with colors ranging from red to ultraviolet (UV) wavelengths[1].

Gallium Nitride (GaN) thin film and its alloy growth are generally carried out by modern methods such as Metal Organic Chemical Vapor Deposition (MOCVD), Molecular beam epitaxy (MBE), Pulsed Laser Deposition(PLD) and Metalorganic vapour phase epitaxy (MOVPE) on sapphire and SiC substrates with a high operational cost. GaN and AlGaIn/GaN thin film growths on silicon substrates (Si) is most expected by the electronics industries for application of very large-scale integrated circuits (VLSI). Besides of its large sizes,, Si substrate has good electrical properties and thermal conductivity and low cost[8]. In addition, growth of GaN semiconductors with low cost is desirable, one of these methods is sol-gel spin coating technique. Sol-gel method is deposition systems which is not complex and has cheaper operational costs. Moreover. It has the ability to produce a thin layer of solid material with good quality. A thin layer of solid material is varied such as material for semiconductor I, magnetic, ceramic, superconductor and polymer [9,10,11].

Recently, high electron mobility transistors (HEMTs) and heterostructures field effect transistors (HFETs) based on AlGa_xN/GaN heterostructures are attracting much attentions due to their potential for making electric and optoelectronic devices based on the formation of a two-dimensional electron gas (2DEG) at the AlGa_xN/GaN interface [2,3,4]. GaN based electronic and optoelectronic devices on Si substrates are thus highly desirable and have potential to be integrated with well-developed silicon microelectronic circuit [5,6]. Nevertheless, it is still difficult to deposit high quality GaN on Si substrates due to a large lattice mismatch of 17% and a thermal expansion coefficient incompatibility of 56%. Therefore, in order to obtain high quality GaN and its alloy deposited on Si substrate, buffer layer growth method can be applied [7,8,9]. One of the method is by using chemical Solution deposition method (CSD).

The aim of this paper was to evaluate the influence of Al and impurities concentrations on the electrical properties of AlGa_xN thin films deposited on Si(111) at 900°C using CSD/sol-gel method.

Experimental Procedures: All samples AlGa_xN thin films were deposited on Si(111) substrates by sol-gel method of spin-coating technique using a crystal aluminum-gallium-citrate-amine as precursors of Ga and Al. The UHP (ultra high purity) N₂ gas of 99.99% as a precursor of N was used which has high reactivity through heating at high temperatures. Aluminum-gallium-citrate-white amines crystal, produced by process of gel preparation of the solution containing citric acid and Ga⁺³, Al³⁺ ions.

Mechanism of crystal preparation process of Al-Ga-citrate-amine (15% mole fraction of Al) was conducted as: 2.16 g of Ga₂O₃ powder and 0.15 g of Al₂O₃ powder were dissolved in a mixture of HCl and HNO₃ (volume ratio of 1:1). Furthermore, the solution was added by 1.1 g citric acid (CA) (the molar ratio of Ga:CA was approximately 1:1) and it was stirred at a temperature of 80°C for 2 hours, to obtain white crystals. Crystal powder was then dissolved in a solution of ethylenediamine to obtain clear gel solutions. The gel was used for deposition of GaN thin films with spin-coating technique on single crystal substrate Si(111).

Before its use for deposition, the substrate Si(111) was rinsed with acetone and methanol for 10 minutes in an ultrasonic bath to remove organic impurities substrate surface. The substrate was soaked with a HF (20%)

solution for 10 seconds to remove the oxide layer that may occur due to the substrate surface oxidation process. After that, the substrate was rinsed with deionized water (DI-water) and dried with N₂ gas. Substrate placed on a spin coater. About 1-2 drops of gel was placed on the substrate and rotated with a speed of 1100 rpm for 1 minute. The next layer was placed in a programmable furnace at temperatures of 900°C with nitrogen gas flowing for 120 sccm for 2 hours and then cooled to the room temperature by the cooling rate of 10°C/minute.

The composition of AlGa_xN films was measured by energy dispersive of X-ray (EDX, JEOL JSM-6360 LA). The electrical transport of the films was measured by means of Hall effect measurement at room temperature.

RESULT AND DISCUSSION

We have studied the influence of the Al and impurities concentration on the electrical properties of Al_xGa_{1-x}N thin films. Table 1 shows the results of EDX analysis of Al_xGa_{1-x}N thin films deposited on Si(111) substrate with the CSD method. The results of Al_xGa_{1-x}N compositions still has low nitrogen concentration (<50%). In this case the Al_xGa_{1-x}N thin films have nitrogen vacation. The lower of nitrogen concentration in the film caused the decrease of Al-N-Ga bonding and natural defect act as residual doping. This case generally occurs in the material deposition semiconductor III-N groups (such as GaN, AlN or InN). In addition, there are still dominant carbons (C) impurities, which derived from organic sources solvent used during process synthesis.

The electrical properties of the films such as mobility, carrier concentration and resistivity were measured by using Hall effect method. Figure 1 showed Hall mobility effect due to variation of Al concentration in Al_xGa_{1-x}N thin films. The result showed that Al_xGa_{1-x}N thin films have n-type semiconductor with electrons as the majority carrier charge. The value of Hall mobility of films decreases by increasing Al concentration.

Table 1: EDX analysis of Al_xGa_{1-x}N thin films with variation of Al concentration.

Sample	Al (%)	Ga (%)	N (%)	C (%)
15%	14,60	28,30	24,54	32,56
20%	9,52	8,69	15,53	53,77
25%	12,27	11,29	26,70	49,73
30%	13,30	5,17	21,91	45,68
35%	24,19	18,97	17,79	39,54

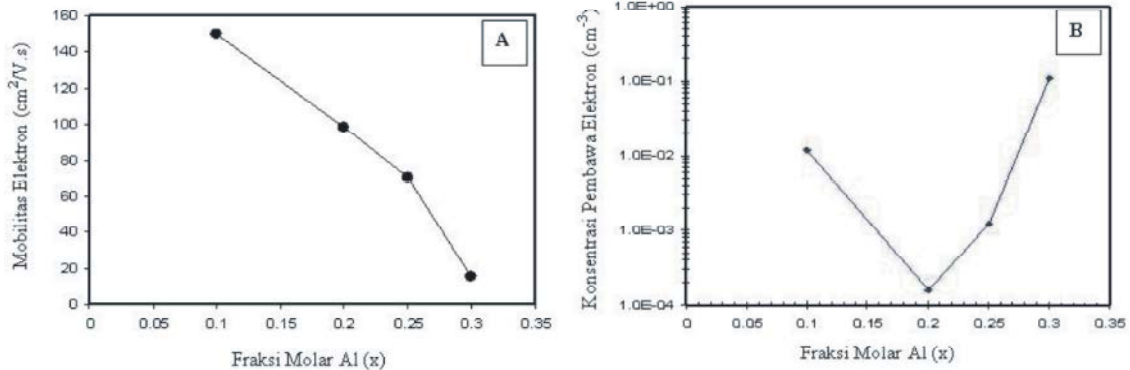


Fig. 1: (a) Hall mobility and (b) carrier concentration versus molar fraction Al concentration (x) of $Al_xGa_{1-x}N$ thin films.

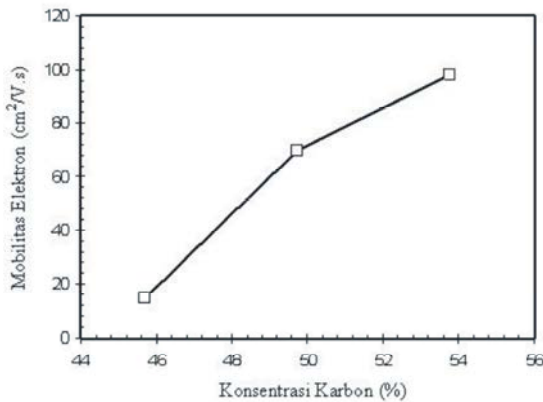


Fig. 2: Effect of carbon concentration on Hall mobility in $Al_xGa_{1-x}N$ thin films.

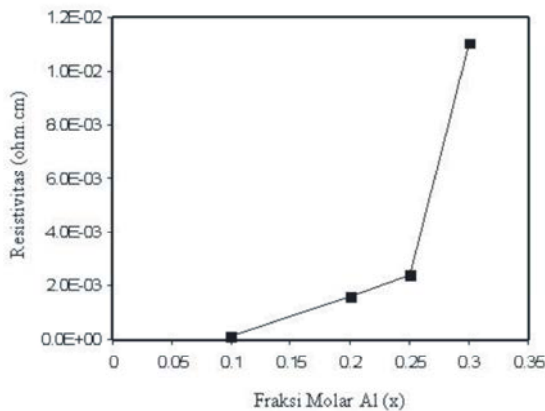


Fig. 3: Resistivity versus molar fraction Al concentration (x) of $Al_xGa_{1-x}N$ thin films.

The increase of Al concentration causes the increase of effective mass (m^*) of AlGa_n charge and the mobility of electrons in the film reduce.

Figure 2 depicts the value of Hall mobility versus oxygen impurity. We observed that the Hall mobility increases by increasing carbon concentration in the films.

Increasing amount of carbon atoms will affect of decreasing the barrier potential in the films.

Figure 3 shows the value of resistivity versus Al concentration of $Al_xGa_{1-x}N$ thin films. We found that by increasing molar fraction of Al could increase the resistivity due to higher energy band gap of the films (Al-N semiconductor with $E_g = 6.2$ eV). This property of AlN semiconductor is generally in the category of insulator material.

CONCLUSION

$Al_xGa_{1-x}N$ thin films have been deposited on Si(111) substrate by sol-gel method of spin-coating technique. The composition of films was measured by energy dispersive of x-ray (EDX) and the measured Al concentrations were in the range of 9.52% to 24.19% with low nitrogen concentration. From the Hall effects measurement, the $Al_xGa_{1-x}N$ films have n-type conduction. The Hall mobility decreases with the increase of aluminum (Al) concentration and decrease the carbon concentration in the film. The resistivity increases with the increase of Al concentration. The highest value of mobility of $Al_xGa_{1-x}N$ films is $150\text{ cm}^2/V.s$.

ACKNOWLEDGMENTS

The research was supported by Ministry of Research and Technology of Indonesian Republic through the project of “Riset Insentif Dasar” with the contract No. UNDIP-INDONESIA, No.: 47/RD/D.PSIPTN/ Insentif/ PPK/I/2010.

REFERENCES

1. Gil, B., *Low dimensional nitride semiconductors*, Oxford University Press, New York, 2002.

2. Wells, A.M., M.J. Uren, R.S. Balmer, K.P. Hilton, T. Martin and M. Missous, 2005. *Direct demonstration of the gate mechanism for current collapse in AlGaIn/GaN HFETs*, Solid State Electronics, 49: 279-282.
3. Zhang, Q., D. Wang, X. Wei, T. Xie, Z. Li, Y. Lin and M. Yang, 2005. *A study of interface and related electronic properties in n-Al_{0.35}Ga_{0.65}N/GaN heterostructure*, Thin Solid Films, 491: 242-248.
4. Balmer, R.S., K.P. Hilton, K.J. Nash, M.J. Uren, D.J. Wallis, D. Lee, A. Wells, M. Missous and T. Martin, 2004. *Analysis of thin film AlN carrier exclusion layers in AlGaIn/GaN microwave heterojunction field effect transistors*, Semicond. Sci. Tech., 19: L65-L67.
5. Kato, T., Y. Honda, M. Yamaguchi and N. Sawak 2002. *Fabrication of GaN/AlGaIn on a (111) Si substrate by selective MOVPE*, Journal of Crystal Growth, 237(239): 1099-1103.
6. Bernat, J., P. Javorika, A. Fox, M. Marso, H. Luth, and P. Kordos, 2003. *Effect of surface pasivation on performance of AlGaIn/GaN/Si HEMTs*, Solid State Electronics, 47: 2097-2103.
7. Lu, Y., X. Liu, X Wang, D.C. Lu, D. Li, X. Han, G. Cong and Z. Wang, 2004. *Influence of the growth temperature of the high temperature AlN buffer on the properties of GaN grown on Si(111) substrate*, Journal of Crystal Growth, 263: 4-11
8. Hasan, Z., Y.C. Lee, F.K. Yam, K. Ibrahim, M.E. Kordesch, W. Halven and P.C. Colter, 2005. *Characteristics of low temperature grown GaN films on Si(111)*, Solid State Communications, 133: 283-287.
9. Wang, D., Y. Dikme, S. Jia, K.J. Chen, K.M. Lau, van P. Gemmern, Y.C. Lin, H. Kalisch, R.H. Jansen and M. Heuken, 2005. *Characterization of GaN grown on patterned Si(111) substrates*, Journal of Crystal Growth, 272: 489-495.