Solid State Science and Technology, Vol. 25, No 2 (2017) 1-7
ISSN 0128-7389 | *http://journal.masshp.net*

GROWTH AND CHARACTERIZATION OF AlGaN THIN FILMS VIA SOL-GEL SPIN COATING METHOD

Nurul Atikah Mohd Isa[1,2], Sha Shiong Ng[1] and Zainuriah Hassan[1]

[1]*Institute of Nano Optoelectronics Research and Technology (INOR), Universiti Sains Malaysia, 11800, Minden, Penang, Malaysia*

[2]*School of Physics, Universiti Sains Malaysia, 11800 Penang, Malaysia.*

Corresponding author: atikah_nurul19@yahoo.com

ABSTRACT

In this study, the growth and characterization of aluminum gallium nitride (AlGaN) thin film grown on aluminum nitride on silicon (111) template via sol-gel spin coating method were reported. The structural, morphological and optical properties of $Al_{0.1}Ga_{0.9}N$ thin film were compared with GaN thin film which prepared using the same method. The structural and surface morphological properties of the deposited films were characterized using X-ray diffraction (XRD), field-emission scanning electron microscope (FESEM) and atomic force microscopy (AFM). XRD results revealed that all the deposited GaN and AlGaN thin films have wurtzite structure and with preferred growth orientation of (002). The FESEM and AFM revealed that the deposited GaN and AlGaN thin films have uniform and smooth surface. The optical characteristics of the GaN and AlGaN thin films were excessed by using Fourier transform infrared (IR) spectrometer. It was observed from the IR results that the E_1(TO) peak was shifted to higher wavenumber for the AlGaN sample.

Keywords : Aluminum gallium nitride; sol-gel, spin coating; structural properties; optical properties

INTRODUCTION

Group-III nitride ternary alloys, in particular aluminum gallium nitride (AlGaN) semiconductors have been recognized to have the potential applications in semiconductor light emitting devices and solar-blind ultra-violet detectors [1-3] as well as sensor devices, particularly for operating under harsh environment conditions. These are strongly driven by their superior physical properties such as excellent thermal, mechanical, and chemical stability, as well as unique optical properties like direct and tune able band gap energy (ranging from 3.4 to 6.2 eV).

Traditionally, metal organic chemical vapor deposition, molecular beam epitaxy and hydride vapor phase epitaxy methods were employed as the preferred manufacturing technology for growing the epitaxial AlGaN thin films [4-6]. However, these methods involved complicated set up and high in production cost. Apart from that, some of these method also involved with toxic chemicals. Therefore, technique such as sol-gel method which is simple and cost effective is highly desirable [7-9].

Synthesizing of GaN and AlGaN using sol-gel method have been reported by Kuppulingam et al. (2014) using ethylene diamine tetra acetic acid complex route [10]. However, in this work, we reported for the first time the growth of the AlGaN thin film using the sol-gel spin coating method where a simple ethanol-based precursor with the addition of diethanolamine (DEA) was used. The structural, morphological and optical properties of the deposited thin films were characterized with various characterization tools. Finally, the obtained results were compared with those obtained from the sol-gel spin coating growth of the GaN thin film.

EXPERIMENTAL

In this work, the GaN and $Al_{0.1}Ga_{0.9}N$ thin films were grown on aluminum nitride on silicon (111) [AlN/Si (111)] template via sol-gel spin coating method. The precursor material for the GaN and AlGaN are gallium nitrate hydrate ($Ga(NO_3)_3.\times H_2O$) and aluminum nitrate nonahydrate ($Al(NO_3)_3.\times H_2O$), respectively. Ethanol (99.7%), distilled water and DEA were used as the solvent and surfactant, respectively. Firstly, the $Ga(NO_3)_3.\times H_2O$ powder and $Al(NO_3)_3.\times H_2O$ powder were dissolved in ethanol and distilled water. Then, DEA was slowly added into the solution. The resulting precursor is stirred to obtain a clear and homogeneous solution. Next, the spin coating process was done by dropping the prepared precursor onto the AlN/Si (111) substrate and spun in ambient condition. The spin process was repeated a few times to obtain the desired thickness, roughly ~300 nm. The thin films were annealed at $950^{o}C$ with the flow of nitrogen gas. Finally, the GaN and AlGaN coated films were nitridated under ammonia gas ambient at $950^{o}C$ for 75 min.

The structural characteristics of the deposited thin films were studied by using high resolution X-ray diffraction (XRD) phase analysis using a PANalytical X'Pert system with Cu*K*α (λ = 1.5406 Å) X-ray source. The surface morphologies of thin films were evaluated by field-emission scanning electron microscopy (FESEM, FEI Nova NanoSEM 450) and atomic force microscopy (AFM, Dimension EDGE, Bruker). Moreover, the optical properties were accessed by means of Fourier-transform infrared (FTIR) spectroscopy (Spectrum GX FT-IR, Perkin-Elmer).

RESULTS AND DISCUSSION

Figure 1 shows the XRD diffraction patterns of GaN and $Al_{0.1}Ga_{0.9}N$ thin films prepared using sol-gel spin coating method. The XRD pattern of the GaN and $Al_{0.1}Ga_{0.9}N$ thin films on AlN/Si(111) were measured under the 2theta-omega (2θ-ω) scan mode. For GaN ($Al_{0.}1Ga_{0.9}N$) thin film, two predominent diffraction peaks correspond to the GaN(002) [$Al_{0.1}Ga_{0.9}N$(002)] and GaN(004) [$Al_{0.1}Ga_{0.9}N$(004)] can be observed at 34.55° (34.65°) and 72.95° (73.05°), respectively. Apart from that, diffraction peaks correspond to the AlN(002) and Si(111) can be clearly observed in Figure 1.

The XRD results indicate that the deposited GaN and $Al_{0.1}Ga_{0.9}N$ thin films have wurtzite structure with (002) preferential orientation. Due to the incorporation of Al contents in the GaN crystal lattice, it was found the diffraction that peaks of $Al_{0.1}Ga_{0.9}N$(002) and $Al_{0.1}Ga_{0.9}N$(004) were shifted towards higher angle by 0.10°. In fact, the incorporation of Al will causes the lattice structure of GaN changes, subsequently affect its lattice constant. From Figure 1, it can be seen that the intensity of the (002) diffraction peak for the GaN is about 4 times higher than that of the $Al_{0.1}Ga_{0.9}N$ sample. This indicates that GaN thin film has better crystalline quality as compared to that of the AlGaN thin film. Similar trend has been reported by Kuppulingam, et al. [10] where it is quite difficult to grow high quality of AlGaN thin films as compared with GaN due to its intrinsic properties [11]. The intrinsic properties of AlGaN has caused the crystalline quality of AlGaN deteriorates as the Al composition increases.

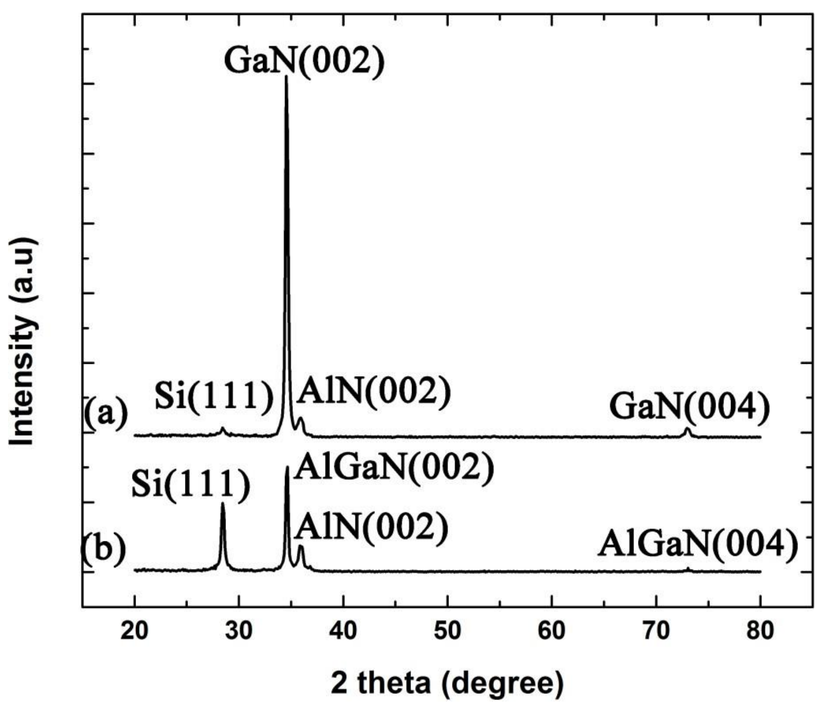

Figure 1: XRD diffraction patterns of (a) GaN and (b) $Al_{0.1}Ga_{0.9}N$ thin films on AlN/Si(111) substrate prepared using sol-gel spin coating method

Figure 2 shows the FESEM images of the GaN and $Al_{0.1}Ga_{0.9}N$ thin films deposited on AlN/Si(111) substrate. Both samples exhibit smooth and uniform surface without any significant crack. In addition, densely packed grains is formed in GaN thin film sample and it is in good agreement with the XRD result, where higher intensity of XRD diffraction peak is observed for GaN (002) peak as compared with $Al_{0.1}Ga_{0.9}N$ sample. However, it is found that the formation of the AlGaN grains size is larger than the GaN samples with the present of some pinholes [12]. According to Fong et al. [7], the present of some pinholes was mainly due to the grains not fully packed. Moreover, some grains are agglomerated to form grains with rose-like structure which is observed in the inset of Fig 2 (b). The formation of rose-like structure and larger AlGaN grain size is likely due to the incorporation of Al into the lattice structure of GaN.

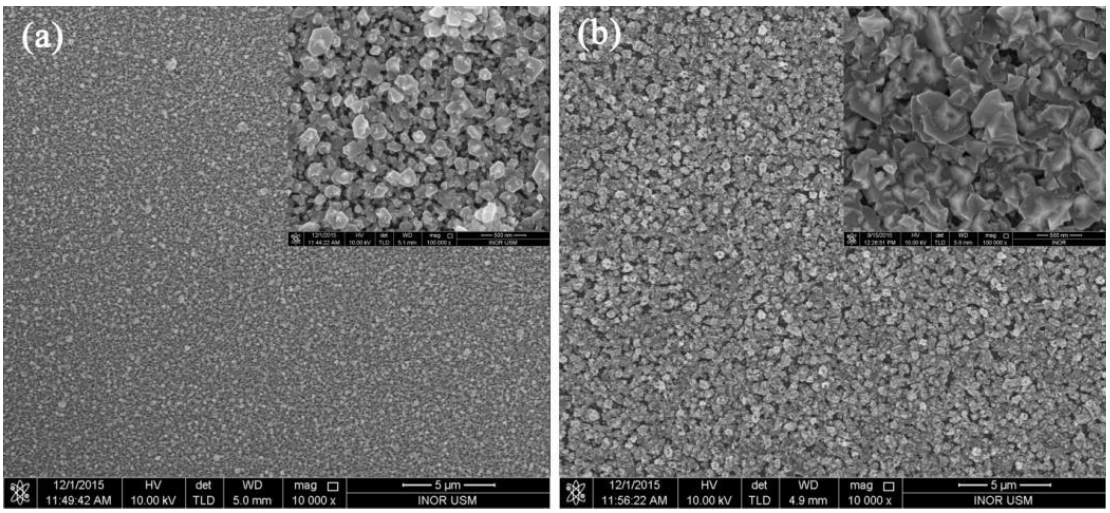

Figure 2: Lower magnification (×10K) of FESEM images of the deposited (a) GaN and (b) $Al_{0.1}Ga_{0.9}N$ thin films deposited on AlN/Si (111) substrate. Inset are the higher-magnification (×100K) of FESEM images

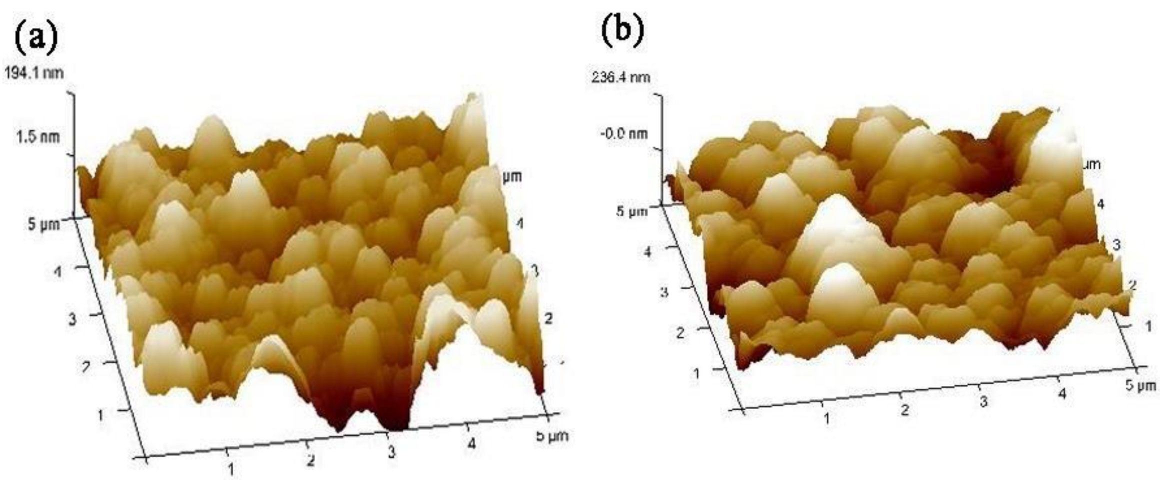

Figure 3: AFM surface morphologies and roughness analysis (under 5 µm × 5 µm scans area) of the (a) GaN and (b) $Al_{0.1}Ga_{0.9}N$ thin films deposited on AlN/Si (111) substrate

Solid State Science and Technology, Vol. 25, No 2 (2017) 1-7
ISSN 0128-7389 | *http://journal.masshp.net*

For further study of surface morphology, the AFM measurements are conducted. Figure 3 shows the three-dimensional (3D) AFM images of GaN and $Al_{0.1}Ga_{0.9}N$ thin films deposited on AlN/Si (111) substrate under 5 µm × 5µm scan area. The AFM images reveal that the deposited AlGaN thin film has larger grain and higher root-means-square (rms) surface roughness as compared to that of the GaN thin film. The rms surface roughness for GaN and $Al_{0.1}Ga_{0.9}N$ thin films are 68 nm and 73 nm, respectively. The obtained results are consistent with the FESEM results as discussed previously.

Figure 4 shows the *p*-polarized IR reflectance spectra of the GaN and $Al_{0.1}Ga_{0.9}N$ thin films deposited on AlN/Si (111) substrate over the spectral range of 400 cm^{-1} to 1000 cm^{-1}. A strong restsrahlen peak corresponds to E_1 transverse-optical (TO) phonon modes of wurtzite GaN and $Al_{0.1}Ga_{0.9}N$ can be clearly seen at 559 cm^{-1} and 563 cm^{-1}, respectively. In addition, a weak pronounced dip corresponds to A_1 longitudinal-optical (LO) phonon modes of wurtzite GaN and AlGaN can be detected at around 737 cm^{-1} and 751 cm^{-1}, respectively. The detected E_1(TO) and A_1(LO) phonon modes of wurtzite GaN are in reasonable agreement with the reported results [9, 13]. Apart from that, a strong reflectance peak located at 670 cm^{-1} and a weak dip located around 885 cm^{-1} can be observed. The origin of these features is corresponds to E_1(TO) and A_1(LO) optical phonon mode of the AlN buffer layer.

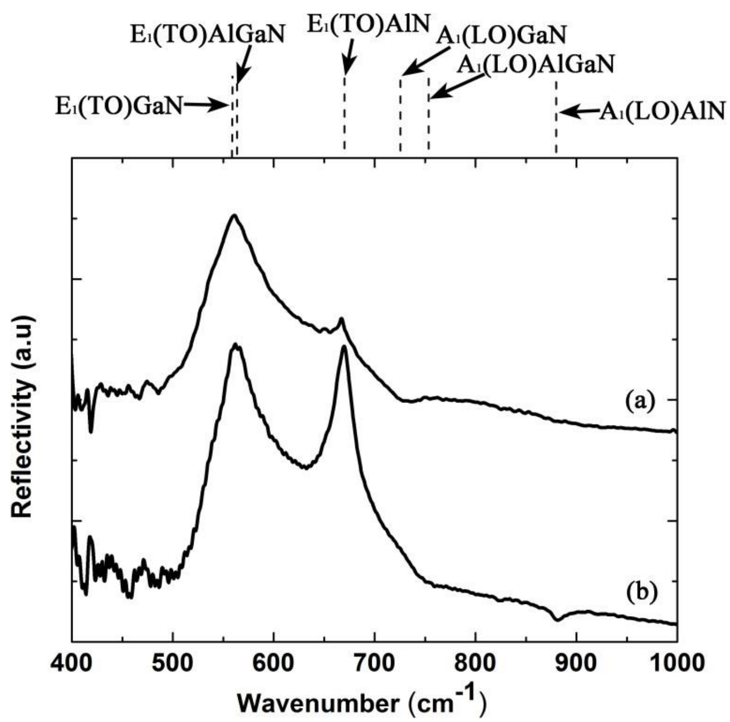

Figure 4: *P*-polarized IR reflectance spectra of the (a) GaN and (b) $Al_{0.1}Ga_{0.9}N$ thin films deposited on AlN/Si (111) substrate

CONCLUSION

In this work, GaN and $Al_{0.1}Ga_{0.9}N$ thin films were grown on AlN/Si (111) substrate via sol-gel spin coating method. The obtained results of XRD revealed that the wurtzite GaN and $Al_{0.1}Ga_{0.9}N$ thin films has preferred growth orientation of (002). The deposited GaN and AlGaN thin films exhibit smooth and uniform surface. From *p*-polarized IR reflectance measurements, all the predominant features of AlGaN are detected. Finally, the obtained results demonstrated that AlGaN thin film can be grown by using sol-gel spin coating method, i.e., a rather simple and cheap method.

ACKNOWLEDGEMENTS

This work is supported by a Science Fund grant from the Ministry of Science, Technology, and Innovation (MOSTI), Malaysia under project number 03-01-05-SF-0750 (Account No.: 305/CINOR/613329). The author would like to acknowledge the staffs of Institute of Nano- Optoelectronics Research and Technology (INOR) and School of Physics, Universiti Sains Malaysia for the technical support.The author also acknowledge the Ministry of Higher Education Malaysia for MyMaster scholarship scheme.

REFERENCES

[1] N. Biyikli, O. Aytur, I. Kimukin, T. Tut, and E. Ozbay, *Applied Physics Letters.* **81** 3272-3274 (2002)

[2] B. Liu, R. Zhang, Z. L. Xie, Q. J. Liu, Z. Zhang, Y. Li, X. Q. Xiu, J. Yao, Q. Mei, H. Zhao, P. Han, H. Lu, P. Chen, S. L. Gu, Y. Shi, Y. D. Zheng, W. Y. Cheung, N. Ke, and J. B. Xu, *Journal of Crystal Growth.* **310** 4499-4502 (2008)

[3] S. Sawyer, S. L. Rumyantsev, and M. S. Shur, *Solid-State Electronics.* **52** 968-972 (2008)

[4] M. Z. Mohd Yusoff, Z. Hassan, H. Abu Hassan, Y. Yusoff, and M. J. Abdullah, *Optoelectronics and Advance Materials - Rapid Comminications.* **4** 1289-1292 (2010)

[5] J. Li, M. Zhao, and X. F. Wang, *Physica B: Condensed Matter*. **405** 996-998 (2010)

[6] X. Wang, X. Wang, G. Hu, B. Wang, Z. Ma, H. Xiao, C. Wang, J. Ran, and J. Li, *Microelectronics Journal*. **38** 838-841 (2007)

[7] C. Y. Fong, S. S. Ng, F. K. Yam, H. Abu Hassan, and Z. Hassan, *Journal of Sol-Gel Science and Technology.* **68** 95-101 (2013)

[8] C. Y. Fong, S. S. Ng, F. K. Yam, H. Abu Hassan, and Z. Hassan, *Journal of Sol-Gel Science and Technology.* **71** 329-332 (2014)

[9] C. Y. Fong, S. S. Ng, F. K. Yam, H. A. Hassan, and Z. Hassan, *Materials Science in Semiconductor Processing.* **17** 63-66 (2014)

[10] B. Kuppulingam, S. Singh, and K. Baskar, AIP Conference Proceedings. **1591** 1437- 1439 (2014)

[11] Y. Jianchang, W. Junxi, L. Naixin, L. Zhe, R. Jun, and L. Jinmin, *Journal of*

Semiconductors. **30** 103001 (2009)

[12] S. S. Ng. Structural and optical studies of wide band-gap $Al_xGa_{1-x}N$ ($0 \leq x \leq 1$) semiconductors Universiti Sains Malaysia (2007).

[13] C. Y. Fong, S. S. Ng, F. K. Yam, H. A. Hassan, and Z. Hassan, *Journal of Crystal Growth.* **413** 1-4 (2015)