

Growth and structural properties of β -Ga₂O₃ thin films on GaN substrates by an oxygen plasma treatment

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β -Ga₂O₃ thin films were successfully fabricated on the surface of GaN substrates by an oxygen plasma treatment. The thickness of the films was measured by scanning electron microscopy (SEM). The monoclinic structure and chemical composition of the β -Ga₂O₃ crystals were confirmed by X-ray diffraction (XRD) and X-ray photoelectron spectroscopy (XPS). In addition, the surface morphology of the films was investigated by atomic force microscopy (AFM). The results obtained for the β -Ga₂O₃ thin films indicated the potential use of GaN crystals.

Key words: Oxidation, β -Ga₂O₃ thin film, GaN substrate, Plasma treatment

Introduction

Gallium nitride (GaN)-based materials are used in a wide variety of applications, including the fabrication of blue/green/ultra-violet emitters (light emitting diodes (LED) and lasers) and high-temperature, high power electrical devices [1-3]. In particular, growth of good quality oxide on GaN is required for the fabrication of metal-oxide-semiconductor (MOS) devices [4]. The structural and electrical characteristics of oxide prepared by dry thermal oxidation of GaN have been reported [5-7]. The method reveals a slow oxidation rate with the possibility of surface degradation because of the high temperature treatment [8]. And the photoenhanced wet oxidation of GaN has also been studied [9, 10]. However, direct plasma oxidation of GaN has not been explored in detail as yet.

In the present study, β -Ga₂O₃ thin films were grown on GaN substrates by an oxygen plasma treatment. The cross-section of β -Ga₂O₃ thin films was observed using environmental scanning electron microscopy (ESEM). The chemical and structural characterizations of β -Ga₂O₃ thin films were confirmed by X-ray photoelectron spectroscopy (XPS) and high-resolution X-ray diffraction (HR-XRD). The surface roughness of the films was measured by atomic force microscopy (AFM). The main purpose of the present study is to report the structural characteristics of β -Ga₂O₃ thin films in an oxygen plasma with an increase in the oxidation time.

Experimental

The β -Ga₂O₃ thin films were grown on GaN substrates using an oxygen plasma treatment. A schematic of the oxygen plasma treatment system is shown in Fig. 1. The substrates were transferred to the reaction chamber through a load lock. The reaction system was a parallel planar discharge system that used a rectangular rf electrode (lower) and a substrate electrode (upper). The upper electrode, which supports the substrate, was connected to a 13.56 MHz rf bias power supply. The substrate was placed on a tray with the surface to be coated facing downward in order to minimize the deposition of dust particles and flakes.

Before the growth of the β -Ga₂O₃ thin films, a short pre-cleaning of the samples was performed in situ to improve film adhesion to the substrate using a H₂ discharge with an rf power of 60 W. The samples were oxidized in oxygen plasma with a flow rate of 100 sccm for 30, 60, 90 and 120 minutes. The working pressure (133.32 μ Pa) and the substrate temperature of the samples were held constant at 300°C.

After the main growth, the oxidized GaN substrates were cooled naturally to room temperature (RT) and analyzed by high-resolution X-ray diffraction (HR-XRD, Bruker D8 Discover system) with Cu K α radiation. The cross-section of the β -Ga₂O₃ thin films was observed using environmental scanning electron microscopy (ESEM, XL30 ESEM-FEG). The chemical change of the oxidized GaN substrate was confirmed by X-ray photoelectron spectroscopy (XPS, ECSA 2000). The surface morphological properties of all of the samples were characterized by atomic force microscopy (AFM, Thermo-Microscopes CP Research).

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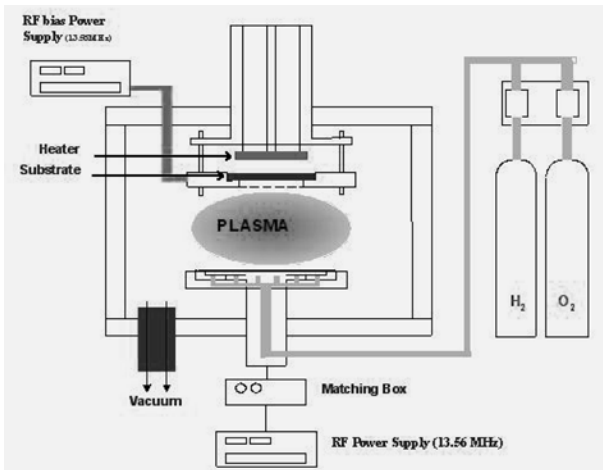


Fig. 1. Schematic of the oxygen plasma treatment system.

Results and discussion

Fig. 2 (a), (b), (c) and (d) show cross-sectional SEM images of β -Ga₂O₃ thin films on GaN substrates for 30, 60, 90 and 120 minutes oxidation time, respectively. In addition, EDX analysis was carried out to identify the oxidation region. Fig. 2(e) shows the EDX spectra taken from the cross-section of the α -Ga₂O₃ film and GaN substrate. The oxygen peak increased remarkably,

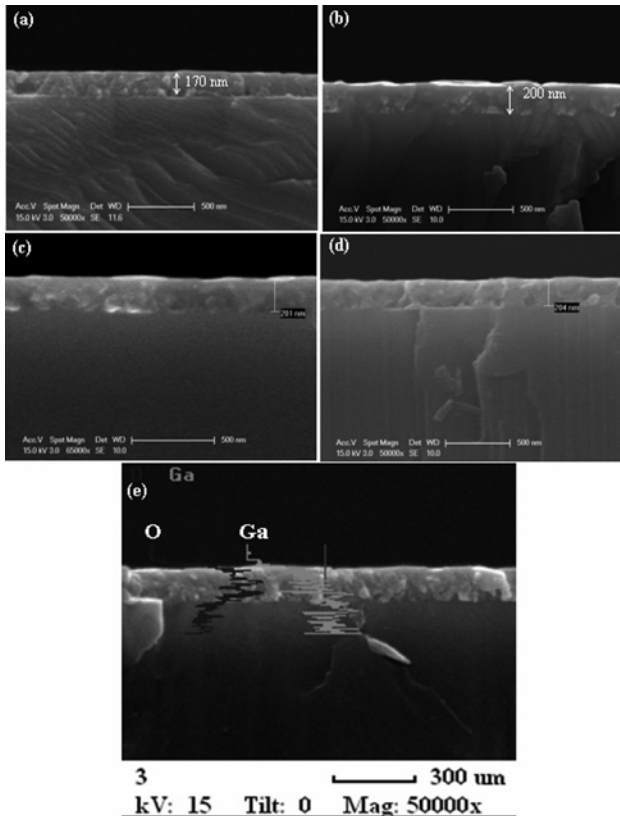


Fig. 2. The cross-sectional SEM micrographs of the β -Ga₂O₃ films: (a) 30 minutes oxidation; (b) 60 minutes oxidation; (c) 90 minutes oxidation; (d) 120 minutes oxidation; (e) EDX spectra.

while the gallium peak decreased slightly due to the stoichiometric atomic ratio Ga : O = 2 : 3. The thicknesses of the β -Ga₂O₃ films increased from 170 to 204 nm with increasing oxidation time. Fig. 3 shows the thickness change of β -Ga₂O₃ films with increasing oxidation time. First, the thickness of films increased to 200 nm up to 60 minutes oxidation time. Thereafter, the thickness of films was not observed to change beyond 60 minutes oxidation time. This was because of the fixed oxygen plasma density.

Fig. 4(a) shows the XRD spectra (θ -2 θ) of a GaN substrate. As shown Fig. 4(a), the pattern of the sample contained a strong diffraction peak with a (002) orientation originating from GaN crystalline substrate. However, no crystalline oxide phase was detected in the β -Ga₂O₃ films using a θ -2 θ scan because the β -Ga₂O₃ films were too thin. So, the XRD patterns of the β -Ga₂O₃ films were obtained using a fixed $\theta = 2.5^\circ$ -2 θ scan. The reflection peaks at $2\theta = 69.8^\circ$ (Fig. 4b) and 66.3° (Fig. 4c) correspond to the (421) and (004) planes of the monoclinic structure of β -Ga₂O₃, respectively, and were well matched with the JCPDS data (41-1103). Thus, the β -Ga₂O₃ films obtained were determined to have a monoclinic structure with diffraction planes of (4 2 1) and (004).

Fig. 5 shows the XPS spectra from the O 1s core level (Fig. 5a) and N 1s (Fig. 5b) of the β -Ga₂O₃ film surface and GaN substrate. The intensity of the O 1s peaks at 533.3 eV (Fig. 5a) was observed to increase significantly after plasma oxidation, while the N 1s peaks at 396.7 eV (Fig. 5b) diminished after the plasma oxidation of the GaN surface. This indicates that N atoms on top of the surface have been substituted by O atoms. These results clearly reveal a more gradual change in composition in the near surface region of the GaN substrate, as a function of the oxidation time at 300°C.

The surface morphology of the GaN substrate and β -Ga₂O₃ films was observed by AFM in the contact mode at room temperature. Fig. 6 shows the AFM images ($20 \times 20 \mu\text{m}^2$) with the root-mean-square (rms) surface roughness. Fig. 6(a) shows the surface morphology of

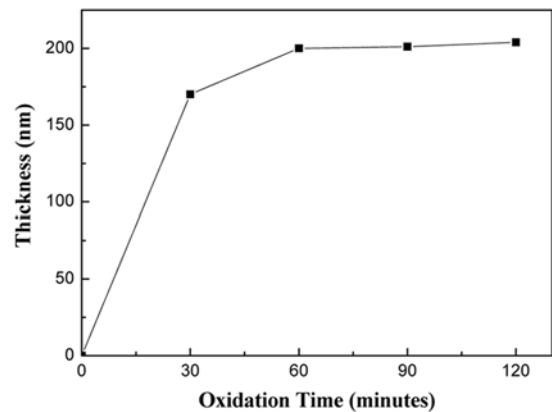


Fig. 3. The thickness of β -Ga₂O₃ films with increasing oxidation time.

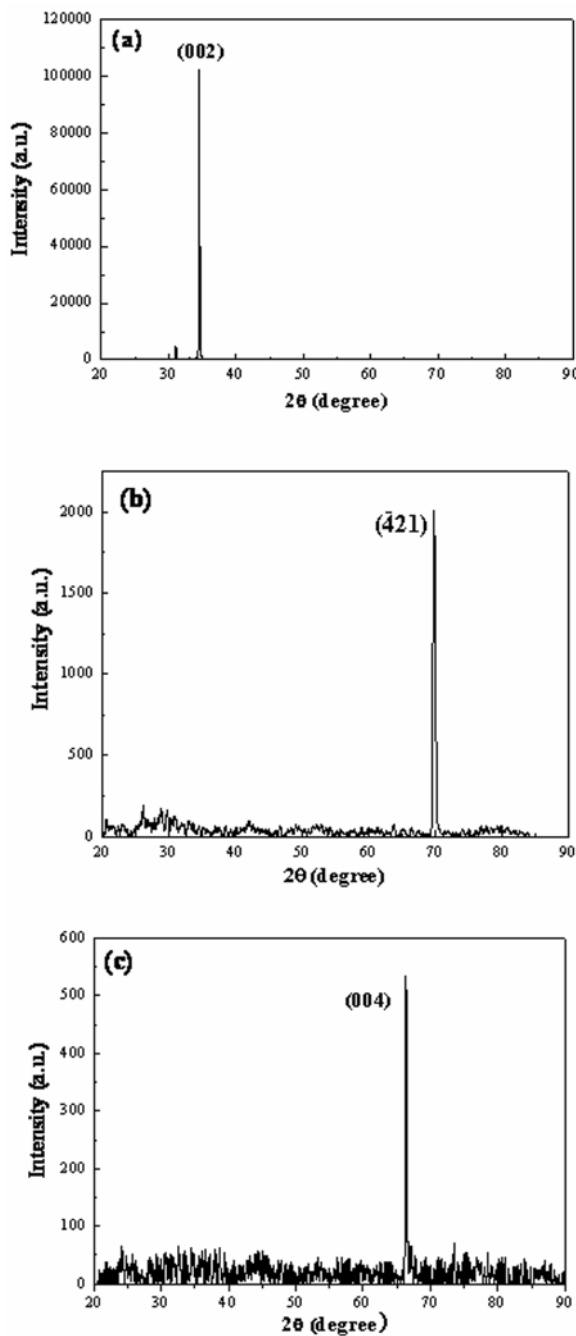


Fig. 4. The XRD patterns of (a) GaN substrate; (b) and (c) β -Ga₂O₃ films after 30 and 60 minutes oxidation.

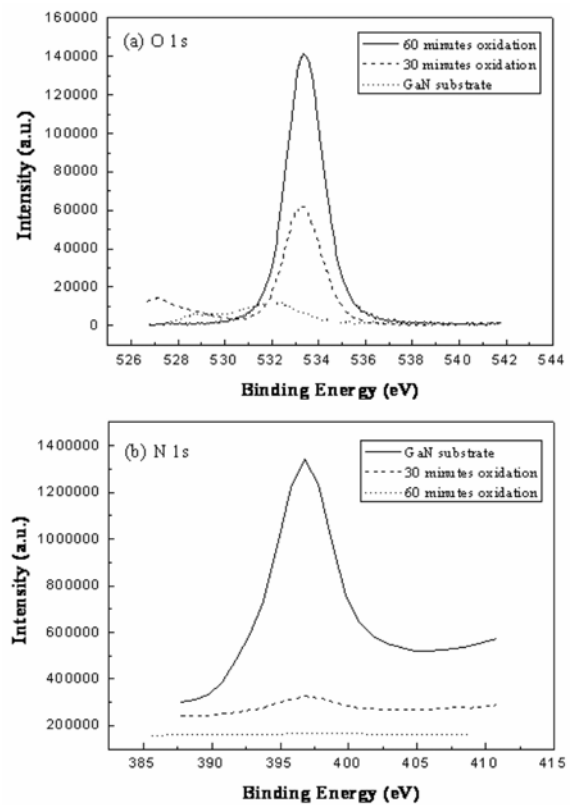


Fig. 5. The XPS spectra: (a) O 1s core level; (b) N 1s core level.

the GaN substrate before the growth of β -Ga₂O₃ films, and Fig. 7(b) and 7(c) show the surface morphology of β -Ga₂O₃ films with increasing plasma treatment time. The rms roughness of the GaN substrate was 2.87 nm. In contrast, the rms roughness of the β -Ga₂O₃ films exposed to the oxygen plasma for 30 and 60 minutes were 1.98 nm and 1.96 nm, respectively.

Conclusions

It was demonstrated that monoclinic β -Ga₂O₃ has been successfully grown on the surface of a GaN substrate by an oxygen plasma treatment. The thicknesses of films measured approximately from 170 to 204 nm by SEM. While the thickness of the films increased from 30 to 60 minutes oxidation time, there was no

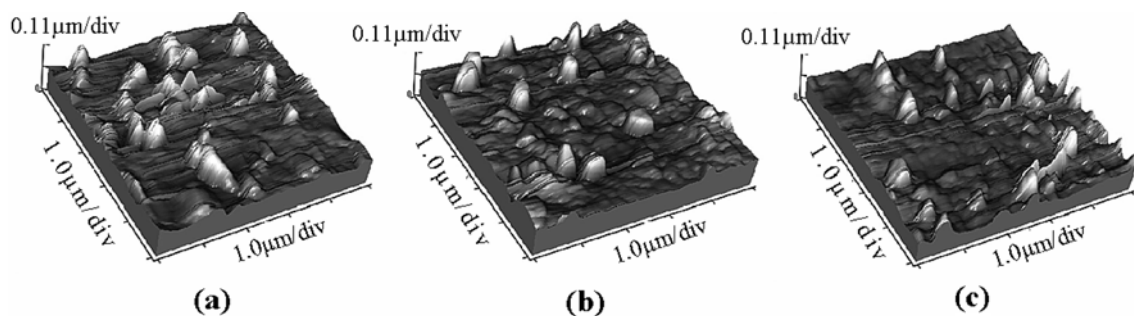


Fig. 6. The AFM images: (a) GaN substrate; (b) and (c) β -Ga₂O₃ films after 30 and 60 minutes oxidation.

change of thickness beyond 60 minutes oxidation time because of the fixed oxygen plasma density. XRD results revealed that the synthesized β -Ga₂O₃ films were identified as having the monoclinic structure with diffraction planes of (4 2 1) and (004). The XPS data confirm the chemical composition of β -Ga₂O₃ thin films after plasma treatment. The intensity of the O 1s peaks are observed to increase significantly after the oxygen plasma treatment, while the N 1s peaks are almost undetectable upon plasma oxidation of GaN surface. The rms roughness of the GaN substrate was 2.87 nm and that of the β -Ga₂O₃ films were 1.98 nm and 1.96 nm. After the oxygen plasma treatment, the surface morphology of β -Ga₂O₃ films were more advanced than that of the GaN substrate.

Acknowledgements

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References

1. S. Nakamura, T. Mukai, M. Senoh, Appl. Phys. Lett. 64 (1994) 1687-1689.
2. S. J. Pearton, F. Ren, A. P. Zhang, K. P. Lee, Mater. Sci. Eng. Rep. R30, (2000) 55-212.
3. T. Miyajima, T. Tojyo, T. Asano, K. Yanashima, S. Kijima, T. Hino, M. Takeya, S. Uchida, K. Funato, T. Asatsuma, T. Kobayashi, M. Ikeda, J. Phys. Cond. Matter 13, (2001) 7099-7114.
4. S. Pal, R. Mahapatra, S. K. Ray, B. R. Chakraborty, S. M. Shivaprasad, S. K. Lahiri, D. N. Bose, Thin Solid Films, 425, (2003) 20-23.
5. S. D. Wolter, B. P. Luther, D. L. Waltemyer, C. Onnby, S. E. Mohny, R. J. Molnar, Appl. Phys. Lett. 70, (1997) 2156-2158.
6. H. Kim, S.-J. Park, H. Hwang, "Thermally oxidized GaN film for use as gate insulators", J. Vac. Sci. Technol. B 19, (2001) 579-581.
7. N. J. Watkins, G. W. Wicks, Y. Gao, Appl. Phys. Lett. 75, (1999) 2602-2604.
8. C. B. Vartuli, S. J. Pearton, C. R. Abernathy, J. D. Mackenzie, E. S. Lambers, J. C. Zolper, J. Vac. Sci. Technol. B 14, (1996) 3523-3531.
9. E. D. Readinger, S. D. Wolter, D. L. Waltemyer, J. M. DeLucca, S. E. Mohny, B. I. Prenitzer, L. A. Giannuzzi, R. J. Molnar, J. Electron. Mater. 28, (1999) 257-260.
10. L.-H. Peng, C.-H. Liao, Y.-C. Hsu, C.-S. Jong, C.-N. Huang, J.-K. Hu, C.-C. Chiu, C.-Y. Chen, Appl. Phys. Lett. 76, (2000) 511-513.