Research Article

Investigation of GaN/InGaN thin film growth on ITO substrate by thermionic vacuum arc (TVA)

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Abstract

Group-III nitride semiconductors covering infrared, visible and ultraviolet spectral range have direct bandgaps changing from 0.7 eV (InN) to 3.4 eV (GaN). With this feature, optoelectronic devices such as light emitting diodes, laser diodes and ultraviolet (visible rays–UV) photodetectors are made. It is possible to grow high-quality InGaN epitaxial layers by modern crystal growth techniques such as molecular beam epitaxy, radio frequency sputtering method and metal organic chemical vapor deposition. Compared with these methods, the Thermionic Vacuum Arc, which is promising thin film growth technique, is relatively inexpensive and quite effective approach for preparing InGaN thin films. The purpose of this research is to investigate the physical properties of the film. The XRD patterns of the InGaN thin films deposited on the ITO substrate exhibited polycrystal structure. The larger crystallite size and smaller FWHM indicate better crystallization. Microstrain values also exhibit good crystallite films respect to the low dislocation density. This film has a potential for photovoltaic devices based on the absorbance graph. It was observed that the compound is homogeneously dispersed on the surface and that there is a nanoporous structure.

Keywords GaN/InGaN · Thin film growth · Thermionic vacuum arc (TVA) · Physical properties

1 Introduction

III-V Nitride semiconductors have many applications. Each energy bandgap (0.7–6.2 eV) between III-Nitride semiconductors forms a large series of triple alloys [2, 8, 15, 22]. With this feature, optoelectronic devices such as light emitting diodes (LEDs), laser diodes and ultraviolet (visible rays–UV) photodetectors are made [12, 21, 29]. III-Nitride systems (InN, GaN, and AlN) are known as wide bandgap semiconductors. Due to their direct bandgap, they have high absorption constants and a sharper cut-off wavelength. By modifying the molar fraction of the triple alloys, the wavelength cutoff can be adjusted and the capacity of the multi-joint devices can be increased with wide bandgap engineering [18]. With all these advantages, III-Nitride structures are the most suitable materials for fabrication of optoelectronic devices in blue and ultraviolet (UV) spectral regions. It is possible to grow high-quality InGaN epitaxial layers by modern crystal growth techniques such as molecular beam epitaxy (MBE) [7, 9, 10, 13, 19, 20, 23, 30], radio frequency sputtering technique (RFSM) [11, 14, 16, 37–39] and metal organic chemical vapor deposition (MOCVD) [3–5, 17, 26, 36, 40]. The method of epitaxial crystal growth with a molecular beam involves the reaction of a thermal beam of atoms or molecules with a crystal surface in a very high vacuum environment. It is also a highly advanced single crystal growth technique in which the film thickness can be controlled very precisely. The fact that the speed of the crystal growth process is too slow is also not suitable for mass production and the

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installation and operation of the devices are very costly [31]. Sputtering deposition is a phenomenon in which the surface atoms of a target material are dislodged by ionized gas atoms and then deposited onto the desired substrate where the ejected atoms are coated with a thin layer. Especially in sputtering technique, it is absolutely necessary to use a buffer gas. This buffer causes the formation of undesirable impurities in the produced films during the analysis of the materials covered by the gas [1]. Therefore, the Thermionic Vacuum Arc (TVA) technique has an important place in terms of impurity. The chemical vapor deposition method is to coat the surface of the heated material in an average closed vessel with a solid material resulting from a chemical reaction of a carrier gas in the vapor state. However, the reactive gases used in the coating are usually dangerous and expensive gases and some unintended components that form as a result of the reaction can affect the coating base [27]. Compared with these methods TVA, which is promising thin film growth technique, is relatively inexpensive and quite effective approach for preparing InGaN thin films. In addition, this method is good for depositing films with high crystallinity and is advantageous for the preparation of films with a short production time. With TVA technique it is possible to work in a high vacuum or very high vacuum conditions. Due to these vacuum conditions, the quality of coated film increases and the effects that may occur on the coated film due to oxidation or some gases are minimized [41].

In this research, InGaN thin films have been deposited on GaN and indium tin oxide (ITO) substrate by TVA. The purpose of this research is to investigate some physical properties of InGaN thin films. XRD was used to analyze the crystal structure of the deposited films. The optical properties of the films have been studied via absorption measurements with ultraviolet–visible spectroscopy (UV/VIS) in the wavelength 300–800 nm. Surface morphology properties of the glass/GaN/InGaN film structure were investigated by an Atomic Force Microscopy (AFM). The structure and surface morphology of deposited InGaN thin film was determined by scanning electron microscopy (SEM). Compositional analysis was done by energy dispersive X-ray spectroscopy (EDAX).

2 Experimental

GaN and InGaN films on ITO substrate were grown by TVA with very short production time being 40 s for GaN and 90 s for InGaN. The film was deposited at 2×10^{-4} Torr working pressure, 18.5 A filament current. Plasma was produced at 200 V for GaN and 500 V for InGaN, at 0.5 A plasma current. ITO has been preferred as a substrate material over types of the metal oxides. Because ITO is a well-known transparent semiconducting oxide thin film. Transparent and conductive layers on substrates are an important component of today's optoelectronic technology. ITO substrate have been bought from Sigma-Aldrich Co. With 70-100 ohms/sq surface resistivity. TVA is a new and different technique from other techniques that produce plasma in anode metal vapors for the regulation of electrodes. The TVA takes place among the anode where the material is placed and a thermionic cathode which is heated directly [24, 28, 34]. Since there is no high temperature during coating process, there is no thermal expansion problem in the coating of metals [25]. Structure of the thin films was studied by XRD with Empyrean, PANalytical with Cu K_{α} radiation. The measurements were performed at grazing incidence angle of 0.5°. One of the most commonly used methods for determining absorption coefficient and energy bandgap of semiconductors is the absorption measurement method. Absorbance measurements were taken using a double-beam UV–vis Spectrometer (300–800 nm) with a Shimadzu UV-3600 Plus model. SEM images and EDX spectrum measurements were taken with a Sigma 300 Model Zeiss Gemini FEG-SEM device. The morphologies of the produced thin films were obtained with Hitachi AFM 5000 II Model device using AFM (dynamic force mode measurement) device. All characterization measurements have been taken at East Anatolian High Technology Application and Research Center (DAYTAM).

3 Results and discussion

The thickness of deposited InGaN film was measured using an interferometer and the thickness measurement system Filmetrics F20. The average thickness of the thin film was found to be about 250 nm. XRD in the 2θ range of 10°–80° was used for crystallographic data of InGaN thin film deposited on ITO substrate at room temperature. X-ray diffraction patterns of deposited thin films are shown in Fig. 1.

The XRD patterns of the InGaN thin films deposited on the ITO substrate exhibited polycrystal structure with three diffraction peaks at 30.30° corresponding to the (100) plane of the InGaN, 33.00° and 36.30° corresponding to the (0002) and (10–11) planes of the GaN, respectively. Wang et al. [39] found (002), (100) and (101) peaks of $In_xGa_{1-x}N$ films deposited on silicon for different In compositions. They reported that deposited films have hexagonal crystal system. (The obtained results for parameters are given in the supplementary material file). For ITO substrate 50.60° and 54.50° are observed with d values of 1.81 Å and 1.69 Å, respectively.

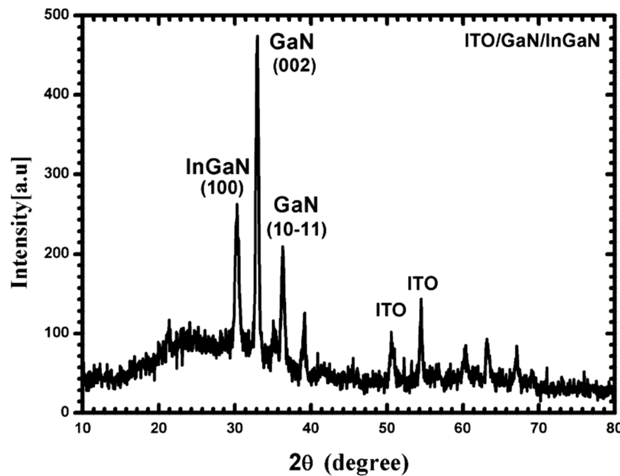

Fig. 1 XRD diffraction patterns of InGaN thin films grown on ITO substrate by TVA method

Some specific properties of the grown films are determined by utilizing the XRD pattern. The particle size of the deposited films was calculated using the Debye–Scherrer formula given at;

$$L = \frac{K\lambda}{\beta \cos \theta} \tag{1}$$

where K is shape constant, which is 0.94, λ is the X-ray wavelength, θ is the Bragg diffraction angle, and β is the full width at half-maximum (FWHM) of the XRD peak appearing at the diffraction angle. In crystals, point defects such as the vacant atomic sites in a structure or the extra atoms positioned between atomic sites may occur. This leads to a change in the conductivities, transmissivities and many properties of the films. Dislocations are linear defects in the crystal structure. Dislocations play an important role in the growth mechanism of films. Dislocation density (δ) is defined as the dislocation line length in the unit volume and is a measure of the number of defects in the crystal. The dislocation density of the films is calculated by the following equality:

$$\delta = \frac{1}{L^2} \tag{2}$$

We used the following expression to determine the microstrain:

$$\epsilon = \frac{\beta \cos \theta}{4} \tag{3}$$

where β is the width at half-maximum (FWHM) and θ is the diffraction Bragg angle. Additionally, crystal number (N) of produced InGaN thin films calculated using the following equation:

$$N = \frac{t}{L^3} \tag{4}$$

where t is the thickness of the produced film. (The obtained results for parameters are given in the supplementary material file). It is seen that the predicted dislocation density and microstrain values are inversely proportional to the crystallite size values. The larger crystallite size, smaller FWHM and indicate better crystallization. Microstrain values also exhibit good crystallite films respect to the low dislocation density. Similar studies have been reported for wide band gap semiconductor material ZnO with different doping situations. Highest peak value of XRD, the narrowest value of FWHM, large crystallite size and low dislocation density are the major parameters that show the better crystal structures [32, 35].

One of the most commonly used methods for determining optical absorption and energy band range of semiconductors is the absorption measurement method. Absorption measurements of thin films on the ITO substrate were taken at room temperature with the aid of a Shimadzu UV-3600 Plus spectrometer. The spectrometric measurement range used is 300–800 nm. In the measurements made, information about the absorption and bandgap of the material was obtained. As can be seen in Fig. 2a, below 450 nm there is a strong absorbance in the film. On the other hand, absorbance values are almost same, after the value of 450 nm, which results in low absorption losses in the visible range. This film indicates a good optical quality and has a potential for photovoltaic devices based on the absorbance graph.

The absorption of light by an optical medium is quantified by its absorption coefficient (α), which defined as the fraction of the power absorbed in a unit length of the medium given by

$$\alpha = 2.303 \frac{Abs}{t} \tag{5}$$

where t is the thickness of the produced film. In Fig. 2b, the value of the optical band gap is calculated as 2.53 eV with the fit plot drawn on the energy graph of $(\alpha h\upsilon)^2$ $(cm^{-1}eV)^2$ of the GaN/InGaN thin film grown on the ITO by RFMS method. This result also supports absorbance values against wavelength. The value of optical band gap of ITO susbtrate is calculated as 4.14 eV, which is shown in Fig. 2b. The optical band gap for ITO as a thin film was found to be in the range of 4.01–4.03 eV [6].

It is very important to investigate surface morphology of the films. For studying surface morphology, Sigma 300 Model Zeiss Gemini FEG-SEM device was used. SEM images are shown in Fig. 3. In Fig. 3, the GaN/InGaN compound grown at 1 µm and 200 nm magnification at 3.8 mm working distance with the in-lens detector. It was observed that

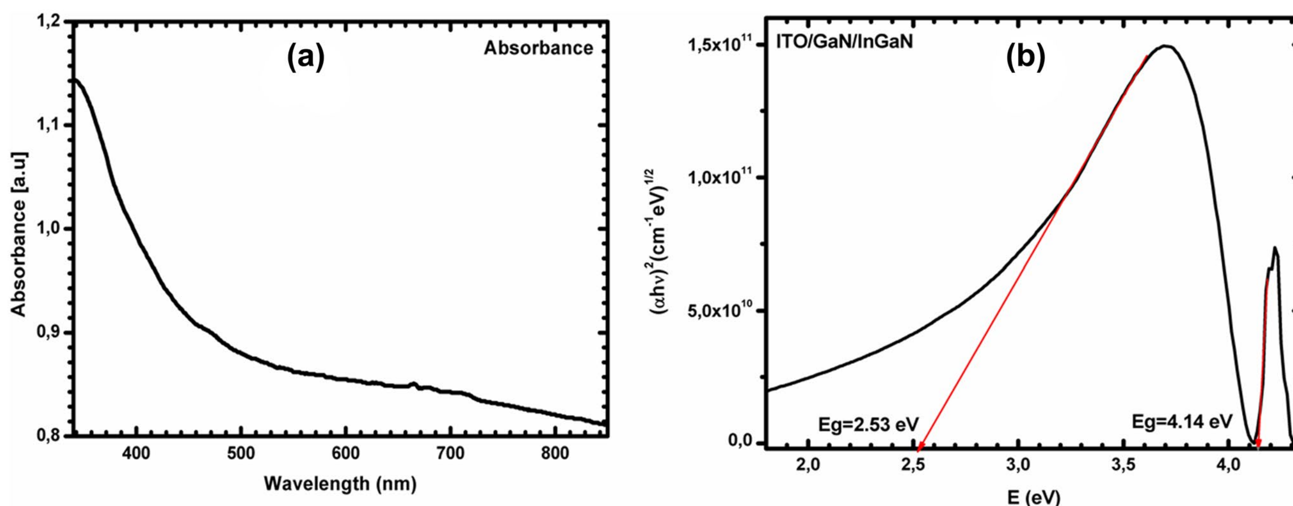

Fig. 2 **a** Absorption graph against wavelength and **b** $(\alpha h\upsilon)^2$ $(cm^{-1}eV)^2$ graph against energy of GaN/InGaN thin film

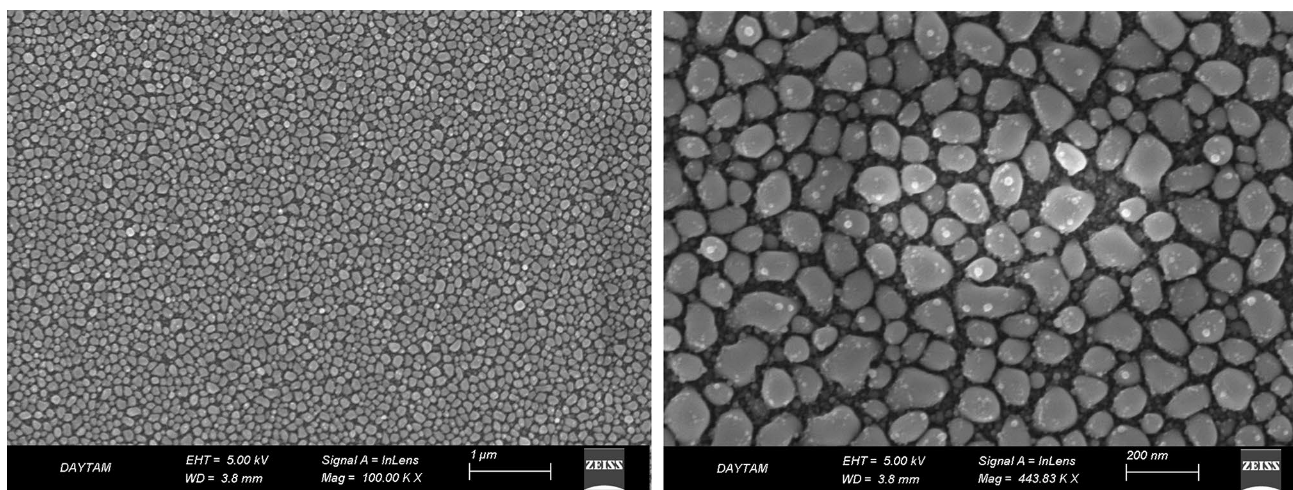

Fig. 3 Different magnitudes of FEG-SEM images of GaN/InGaN thin film grown on ITO by TVA method

the compound is homogeneously dispersed on the surface and that there is a nanoporous structure. In addition, as seen in the image, thin film has a granular structure. The deposited film is dense, smooth, and continue without any holes and cracks.

We see that the EDX results reveal that nitrogen, oxygen, gallium, silicon and indium are the main elements present within the inspection field, with oxygen being the most abundant. The tabulated results provide a semi-quantitative view of the elemental composition in the inspection field in units of both weight percent and atomic percent. The elemental analysis results showed that In composition is 12.37% in the InGaN alloy. (EDX spectrum and elemental analysis results are given in the supplementary material file).

The 5 µm × 5 µm scale atomic force microscope (AFM) images of the top surface were performed in dynamic force mode by using Hitachi AFM 5000 II Model device using AFM device. Surface morphology images of the film are showed in Fig. 4. The linear roughness value is 6.86 nm and the average roughness value is 5.41 nm, which is consistent with each other. Height of the highest peak above the mean line in the profile (Rp) was 23.23 nm and depth of the deepest valley below the mean line in the profile (Rv) was 13.09 nm. Hence, total roughness (Rt), which is the sum of the (Rp) and (Rv) and was calculated as 36.32 nm. (The obtained results for parameters are given in the supplementary material file). Saleh et al. [33] synthesized $In_{0.08}Ga_{0.92}N$/AlN/Si thin films with a photoelectrochemical etching technique. They have investigated the

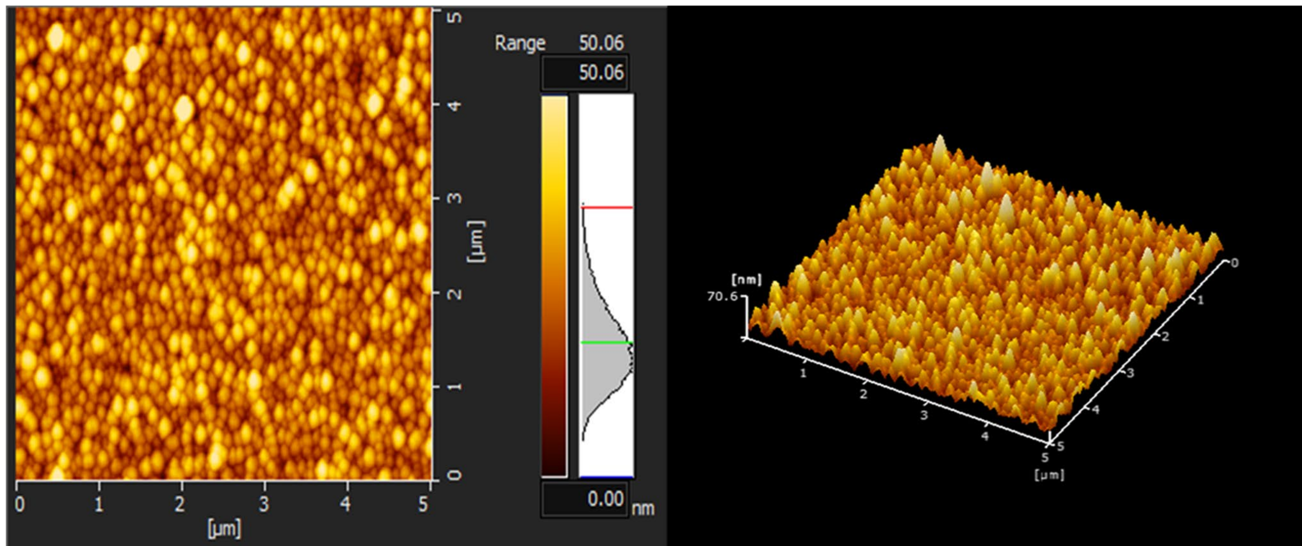

Fig. 4 The 2D and 3D AFM images of the produced film

effects of etching durations on films morphology. Root mean square (RMS) roughness of that film was found to be 2.2 nm.

4 Conclusion

GaN/InGaN thin film was deposited on ITO substrate to analyze its physical properties. We successfully achieved hexagonal GaN/InGaN thin film on ITO substrate using TVA method. The XRD patterns of the InGaN thin films deposited on the ITO substrate exhibited polycrystal structure with three diffraction peaks at 30.30°, 33.00° and 36.30° corresponding to the (100), (0002) and (10–11) planes of the InGaN, respectively. Below 450 nm there is a strong absorbance in the film. On the other hand, absorbance values are almost same, after the value of 450 nm, which results in low absorption losses in the visible range. The value of the energy bandgap is calculated as 2.53 eV with the fit plot drawn on the energy graph of $(\alpha h\upsilon)^2$ $(cm^{-1}eV)^2$ of the GaN/InGaN thin film grown on the ITO by RFMS method. This result also supports absorbance values against wavelength. SEM images are shown that the films are crystalline with a uniform dimension of crystals. Surface morphology images of the film indicated that the linear roughness value is 6.86 nm and the average roughness value is 5.41 nm, which is consistent with each other. Elemental analysis results confirmed the presence of the Si, Ga, In, N and O in the film. Finally, TVA is a fast growth method for GaN/InGaN thin film.

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Compliance with ethical standards

Conflict of ınterest The authors declare that they have no conflict of interest.

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