Comparison of Optical, Electrical, and Surface Characteristics of InGaN Thin Films at Non-Flow and Small Nitrogen Flow Cases

Asim Mantarci (✉ asimmantarci@gmail.com)

Mus Alparslan Universitesi <https://orcid.org/0000-0001-8369-3559>

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Comparison of optical, electrical, and surface characteristics of InGaN thin films at non-flow and small nitrogen flow cases

Asim Mantarci^{1,&}

¹ Muş Alparslan University, Physics Department, Güzeltepe/Muş, Turkey

& a.mantarci@alparslan.edu.tr; asimmantarci@gmail.com;

& <https://orcid.org/0000-0001-8369-3559>

Abstract

InGaN films in the non-flow and a small flow of nitrogen cases were fabricated by the RFMS (Radio Frequency Magnetron Sputter) method to compare crucial physical characteristics of its material. From the XRD analysis, application of small nitrogen flow in InGaN thin film growth has been observed to result in changes in the crystal size, texture coefficient, and crystal structure parameters of the film. AFM results showed both films obtained have tightly packed granular, and almost homogeneous, and Nano-structural properties, but they are different in roughness, as increased by applying small nitrogen flow. Optical conductance peaks of the material in non-flow and small flow case were 1.3957×10^{10} and 1.1496×10^{10} (S/m), showed a decrement in optical conductance by small nitrogen flow. In the same manner, electrical conductance peaks of the material in non-flow and small flow case were 5.2512×10^{12} and 5.2236×10^{12} (S), showed a decrement in electrical conductance by small nitrogen flow. In addition, the electrical conductivity of the InGaN material has been obtained at higher than the optical conductivity value of the InGaN material in both cases. Also, it was noticed that direct allowed optical band gap energy non-flow and small flow cases were 2.4701 and 2.5225 eV, displayed increased by applied small nitrogen flow. Essentially, many noteworthy physical properties such as crystalline

size, texture coefficient, optical/electrical conductivity, the surface roughness of the films have been compared and studied for the non-flow and a small flow of nitrogen cases.

Keywords: non-flow, InGaN, texture coefficient, electrical conductivity, nano-structure

1. Introduction

InGaN (Indium Gallium Nitride) material, which is included in 3 nitride groups, has been used in device production (micro LEDs, solar water splitting, ultrasensitive strain sensor, etc.) that require high technology and has emitted from the ultraviolet to the green region (Dalapati et al. 2020) (Smith et al. 2020) (Alizadeh et al. 2020) (Hu et al. 2020) (Iyer et al. 2020) (Chen et al. 2020b) (Pasayat et al. 2020). Theoretically speaking, $\text{In}_x\text{Ga}_{1-x}\text{N}$ has a capacity to absorb all photons at region energies covering the entire spectrum from 0.7 eV to 3.4 eV, proved from (Moses et al. 2011). Moreover, the material has high mobility (Yarar 2008) and high absorption coefficient (Schenk et al. 2000) properties, which makes it a potential candidate for manufacturing multi-joint solar cells (approx.> 50 % efficiency). Materials used in space applications are required to be resistant to high radiation, and the InGaN material has been proven in this research (Zhang et al. 2013) that its high radiation resistance can be used in space applications. Nowadays, many important InGaN-based devices such as field emitters (Chen et al. 2020a), multicolor display (Evropeitsev et al. 2020), and intermediate band solar cell (Cheriton et al. 2020) are produced. Studies (Yang et al. 2017) in the literature have shown us that the electrical, optical and structural parameters of the InGaN material limit the efficiency of the material for many key parameters such as electron mobility (Gökden et al. 2010) and photon absorption (Liu et al. 2017). Furthermore, when the material is used in an optoelectronic device, the mentioned properties will affect the efficiency of the device. For these reasons, investigating the changes in the structural, electrical and optical properties of the InGaN for different

conditions will shed light on the studies to be carried out. In this research, the changes in the electrical, optical and structural properties of the films produced in non-flow and small nitrogen flow cases were compared. The original aspect of this work is to understand and compare the effects of different conditions designed while producing the film on the key properties of the film: electrical conductivity, optical conductivity, optical band gap energy, crystal-structure parameters.

2. Material and Methods

2.1. The growth of the material

InGaN thin films in the non-flow and a small flow of nitrogen case were coated onto GaN/n-Si (100) substrate, which was previously published by our research team (Mantarçı,Kundakçı 2019). Films with the best properties among the films produced before were used as substrates. The target used in the study (the InGaN target (99.99 % (4n) purity, 3.18×50 mm)) was obtained from the ACI Alloys Comp. In the research center called DAYTAM, the films were produced by the sputtering system (VAKSIS, 3M- RF sputter) in the Cleanroom. In preparation for the film growth stage, the substrates were cleaned by HF: DI (1:1) processing, the detailed procedure of which is given in the reference (Smith et al. 1996). The coating process was as follows respectively. First, the cleaned substrates were placed in the holder in the system. The system was put into the initial vacuum. When the initial vacuum is reached, the working gas (argon gas) was given to the system. The system was powered up and the plasma was ignited. The surface was pre-sputtered for about 10 minutes before the growth was initiated. More details about the growth process can be found in the reference in another study (Mantarçı,Kundakçı 2020). The following are the parameters applied in the coating. The initial pressure is $\sim 5 \times 10^{-3}$ mTorr, argon as a working gas (50 sccm), target to substrate angle $\sim 31.0^\circ$, target to substrate distance 12.3 cm,

substrate temperature 500 °C, substrate rotation 3 rpm, the sputtering power is 100 W. During growth, the coating parameters of the films were kept constant except that the small nitrogen flow (1 sccm) was applied to one of the films. To do this, the change of coating parameters was monitored instantly by an automatic control panel.

2.2. Characterization of the material

Surface topography properties of materials were determined using Hitachi 5100N –AFM from the $5\mu\text{m} \times 5\mu\text{m}$ area using the tapping mode in which the tip was hit on the surface. The absorption properties of the materials of non-flow and small flow cases were investigated using a Shimadzu UV-3600 Plus spectrophotometer. This system has Pbs and InGaAs detectors and an R-928 model photomultiplier. The main monochromator is of high-performance holographic grating type in aberration-corrected Czerny-Turner mounting. XRD technique (D-8, PANalytical Empyrean) was used to understand the crystal-structure properties of films. Grazing incidence mode (GI-XRD) was preferred while taking measurements. Here are our XRD operate parameters; generator voltage = 45 kV, omega=0.5, psi=0, Phi=0, z=9.329, time per step=2.64 s, tube current = 40 mA, the anode material = Cu, K-Alpha1 wavelength = $1.5405980 \text{ \AA}$, K-Alpha2 wavelength = $1.5444260 \text{ \AA}$. The thickness measurements of the obtained films were made with the KLa Tencor Stylus Profiler P7. The resolution in the vertical axis was $1 \text{ \AA}$ and the repeatability of step thickness measurement in the Z-axis was 0.5 nm. The thickness of the films obtained was measured to be 93 nm within error $\pm 2 \text{ nm}$.

3. Results and Discussion

3.1 Structure-Crystal and Surface Properties

XRD diffraction properties of thin films (with non-flow and with the small nitrogen flow) are shown in **Figure 1(a, b)**. According to the PDF card no: 98-018-1363 (supplementary file), films

with a polycrystalline structure ((002) XRD phase and (011) XRD phase) were obtained. From Scherrer theorem, crystallite sizes of the films for (002) phase were obtained to be 29.83 (± 0.6) and 22.77 (± 0.3) nm at non-flow and small nitrogen flow case as a decreased trend. Thanks to Bragg's law, inter-planar spacing (d) was calculated to be 2.7120 and 2.7080 Å, and lattice parameters (a, c) were calculated to be 3.3214, 5.4240 and 3.3165, 5.4160 Å, as can be seen in **Table 1**, they are close to the theoretical values. Vegard's law, representing by $x = (c_o(\text{InGa}) - c_o(\text{Ga})) / (c_o(\text{In}) - c_o(\text{Ga}))$, allows calculating the indium ratio (x) in $\text{In}_x\text{Ga}_{(1-x)}\text{N}$ structure. As a comparison, the non-flow film has an $\text{In}_{0.4846}\text{Ga}_{0.5154}\text{N}$ film structure; the small nitrogen flow film has an $\text{In}_{0.4690}\text{Ga}_{0.5310}\text{N}$ film structure. This result may be mainly due to a decrement in $c_o(\text{InGa})$ value. A statistical entity of crystalline orientations in the polycrystalline is defined as the crystallographic texture of the material. To put it another way; it expresses deviations from the statistical randomness of preferred orientations. With the Harris texture theorem stated in the past study (Harris 1952), the texture coefficient ($C_{(h)}$) of our films can be determined. On the one hand; texture coefficients for (002) orientation, which is a preferred orientation, were 0.857 and 0.821 at non-flow and small nitrogen flow cases, respectively, showing a decreasing trend. A decrease in preferred orientation is associated with a decreased number of crystallites along that plane. On the other hand, texture coefficients for (011) orientation were 0.142 and 0.178 at non-flow and small nitrogen flow cases, respectively, showing an increasing trend. An increase in preferred orientation is associated with an increased number of crystallites along that plane. In a polycrystalline film, both surface and interface energy minimization support development of texture via preferred growth of crystallite by specific orientation (Thompson 1990). Final texture depends on growth conditions (including total pressure, power supplied to the target, substrate and its temperature) and growth techniques.

Briefly, application of small nitrogen flow in InGaN thin film growth has been observed to result in changes in the crystal size, texture coefficient, and crystal structure parameters of the film.

Figure 1 (a,b)

Table 1

RMS (Root Mean Square) roughness of films at non-flow and small flow case were 38.02, 45.67 nm, respectively, as increased with applying small nitrogen flow. This increase in surface roughness can be attributed to the increase in surface atom mobility due to increased nitrogen flow. As can be seen in **Figure 2 (a, b)**, both films obtained have tightly packed, granular, and almost homogeneous and Nano-structural properties. Film at small flow case has more white areas than in non-flow case may be due to white rust and oxide on the surface. Other surface topographic properties of films (surface maximum valley depth, surface absolute slope, etc.) are given in **Table S1** as supplemental information.

Figure 2 (a, b)

3.2 Optical Band Gap Energy and Optical/Electrical Conductivity Properties

Figure 3 (a, b) gives us the measurements of the absorption behavior of films non-flow and small flow cases. Using Tauc theory from $\alpha^2 E^2 - E$ plot (intersect value in the y-axis at zero), the optical band gap energies of films can be determined. By doing the following operations respectively, analysis>Fitting>Fit Linear in the OriginPro 8 program, direct allowed optical band gap energy non-flow and small flow cases were 2.4701 ± 0.019 and 2.5225 ± 0.025 eV, displayed increased by applied small nitrogen flow. This increase in optical bandgap energy may be due to the difference in indium ratio in films. In the past published article (Özen et al. 2016), the direct - allowed optical bandgap energy of the InGaN was found to be 2.75 eV and this value is bigger

than the value we found. The refractive index values of films at non-flow and small flow cases can be calculated with some theorems (Moss (Moss 1985), Ravindra (Ravindra et al. 2007), Herve-Vandamme (Hervé,Vandamme 1994), and Kumar and Singh (Kumar,Singh 2010)) using the optical bandgap energy. The refractive indices of films display a decreasing behavior when applied small nitrogen flow. The obtained refractive index values are given in the supplementary document (**Table S2**).

Figure 3 (a, b)

Studying the electrical (Qashou et al. 2019) and optical conductivity (Roknuzzaman et al. 2017) behavior of materials plays a key role in optoelectronic device studies; can be described with the following formulas;

$$\sigma_{(\text{electr.})} = (2\lambda). (\sigma_{(\text{opt.})}). (\alpha)^{-1} \quad (1)$$

$$\sigma_{(\text{opt.})} = (\alpha nc). (4\pi)^{-1} \quad (2)$$

Where n, the refractive index for Moss model. The optical and electrical conductivity behavior of the InGaN material in non-flow and small flow case is shown in **Figure 4 (a, b)**. In the energy range of 2.1 to 3.9 eV, optical conductance peaks of the material in non-flow and small flow case were 1.3957×10^{10} and 1.1496×10^{10} (Siemens/m), showed a decrement in optical conductance by small nitrogen flow. In a work (Berrah et al. 2009) published in the literature, the optical conductivity of the material was obtained a little higher than our value. In the energy range of $\sim 2.4 - 3.5$ eV, electrical conductance peaks of the material in non-flow and small flow case were 5.2512×10^{12} and 5.2236×10^{12} (Siemens), showed a decrement in electrical conductance by small nitrogen flow. In a past published article (Hu et al. 2010), the electrical conductivity of the material was found to be higher than our value. The electrical conductivity of

the InGa_N material has been obtained at higher than the optical conductivity value of the InGa_N material in both non-flow and small flow cases. In summary, applying small nitrogen flow to the film causes a reducing change in electrical and optical conductivity values.

Figure 4 (a, b)

4. Conclusions

InGa_N films in the non-flow and a small flow of nitrogen cases were fabricated by the RFMS method to compare noteworthy physical characteristics of its material. XRD proved that films with a polycrystalline structure ((002) XRD phase and (011) XRD phase) were obtained. Crystallite sizes of the films for (002) phase were obtained to be 29.83 (± 0.6) and 22.77 (± 0.3) nm at non-flow and small nitrogen flow case as a decreased trend. As a comparison, the non-flow film has an In_{0.4846}Ga_{0.5154}N film structure; the small nitrogen flow film has an In_{0.4690}Ga_{0.5310}N film structure. This result may be mainly due to a decrement in c_0 (InGa_N) value. On the one hand; texture coefficients for (002) orientation, which is a preferred orientation, were 0.857 and 0.821 at non-flow and small nitrogen flow cases, respectively, showing a decreasing trend. A decrease in preferred orientation is associated with a decreased number of crystallites along that plane. On the other hand, texture coefficients for (011) orientation were 0.142 and 0.178 at non-flow and small nitrogen flow cases, respectively, showing an increasing trend. An increase in preferred orientation is associated with an increased number of crystallites along that plane. RMS (Root Mean Square) roughness of films at non-flow and small flow case were 38.02, 45.67 nm, respectively, as increased with applying small nitrogen flow. This increase in surface roughness can be attributed to the increase in surface atom mobility due to increased nitrogen flow. Film at small flow case has more white areas than in non-flow case may be due to white rust and oxide

on the surface. Direct allowed optical band gap energy non-flow and small flow cases were 2.4701 ± 0.019 and 2.5225 ± 0.025 eV, displayed increased by applied small nitrogen flow. This increase in optical bandgap energy may be due to the difference in indium ratio in films. Optical conductance peaks of the material in non-flow and small flow case were 1.3957×10^{10} and 1.1496×10^{10} (S/m), showed a decrement in optical conductance by small nitrogen flow. In the same manner, electrical conductance peaks of the material in non-flow and small flow case were 5.2512×10^{12} and 5.2236×10^{12} (S), showed a decrement in electrical conductance by small nitrogen flow. Shortly, application of small nitrogen flow in InGaN thin film growth has been observed to result in changes in the crystal size, texture coefficient, and optical/electrical conductivity and surface roughness of the film, which will be able to shed light on InGaN-based studies.

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TABLES

Table 1 Comparison of crystal-structure properties of films under condition of non-flow and small Nitrogen flow (**E**: experimental value **T**: theory value)

Nitrogen flow condition	FWHM (°) (002)	Crystallite size (Scherrer) (nm) (002)	Lattice constants ^E (002) (Å)	Lattice constants ^T (002) (Å)	d-value ^E (Å)	d-value ^T (Å)	2θ ^E (°)	2θ ^T (°)
			a c	a c				
Non-flow	0.29±0.09	29.83±0.6	3.3214±0.01 5.4240±0.01	3.3300 5.3990	2.7120±0.01	2.6995	33.00	33.15
Nitrogen flow of 1 sccm	0.38±0.07	22.77±0.3	3.3165±0.01 5.4160±0.01	3.3300 5.3990	2.7080±0.01	2.6995	33.05	33.15

FIGURES CAPTIONS

Figure 1 a) XRD graph of the material **b)** Texture coefficient and crystal size versus nitrogen flow condition

Figure 2 a) AFM images for non-flow case **b)** for small flow case

Figure 3 a) Absorbance pattern of the material for non-flow case **b)** for small flow case

Figure 4 a) Optical Conductivity **b)** Electrical Conductivity of the material

Figures

Figure 1

a) XRD graph of the material b) Texture coefficient and crystal size versus nitrogen flow condition

Figure 2

a) AFM images for non-flow case b) for small flow case

Figure 3

a) Absorbance pattern of the material for non-flow case b) for small flow case

Figure 4

a) Optical Conductivity b) Electrical Conductivity of the material

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