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Original Research Article

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ENHANCEMENT OF POWER CONVERSION EFFICIENCY OF DYE-SENSITIZED SOLAR CELLS VIA INCORPORATION OF GAN SEMICONDUCTOR MATERIAL SYNTHESIZED IN HOT-WALL CHEMICAL VAPOR DEPOSITION FURNACE

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Abstract. *This study discusses the results of plasma enhanced chemical vapor deposition synthesis of GaN on sapphire and silicon substrates using specific parameters: a forward output voltage of 150 watts, a N_2 gas flow rate of 60 standard cubic centimeters per minute, a chamber pressure of 2.48 mmHg, and a synthesis time of 2hours. Characterization by scanning electron microscope, Raman and energy dispersive X-ray revealed the non-stoichiometric formation of GaN, with Ga clearly predominating in the composition. scanning electron microscope analysis of the substrate surface morphology revealed the presence of small islands, which are considered to be the first step in the chemical vapor deposition process. The research also examined the effects of incorporating GaN into the photoanode of dye-sensitized solar cells. The study investigated the optimal amount of GaN powder in the TiO_2 matrix. The initial experiments used commercial GaN powder to determine the optimal weight percentage. Four different weight percentages (wt%) 10 wt%, 20 wt%, 30wt % and 40 wt% GaN were selected for the study. Among them, the 20 wt% GaN had the highest power conversion efficiency of 0.75%. The fill factor values showed a tendency to decrease as the weight fraction of GaN increased.*

Keywords: power conversion, semiconductor, dye, TiO_2, GaN.

1. Introduction

The special physical and chemical properties of semiconductor nanostructures inspire their use as functional devices and basic components for electronic and optoelectronic nanodevices [1]. Gallium-based semiconductor materials are currently of great practical importance and are widely used in the production of high-efficiency optical information storage devices, displays, field-grade lasers, environmental detectors and other applications [2, 3]. The production of semiconductor materials, particularly gallium nitride, for solid-state lighting and high-performance electronic devices represents a promising and cost-effective technology

for producing light-emitting devices. The goal of this advancement is to provide consumers with a viable alternative to highly efficient and less reliable lighting technologies [4].

The strategic design and modification of structures based on gallium compounds with the aim of reducing costs in the end product is based on their environmentally friendly and chemically inert nature. Therefore, there is an urgent need to further develop technologies that can improve living standards by reducing energy consumption and thereby minimizing environmental impact. III-nitride semiconductor materials, including aluminum nitride (AlN), gallium nitride (GaN), and indium nitride (InN), similar to silicon, have the potential to serve as the basis for semiconductor devices with novel functionalities and the ability to upgrade existing technologies [5].

To achieve high-efficiency LEDs with high brightness, the development of bulk or free-standing GaN structures with GaN as a substrate is required. Established crystal growth techniques such as high-pressure nitrogen solution (HPNS), the sodium (Na) flow method, ammon thermal growth and hydride vapor phase epitaxy (HVPE) are used to obtain gallium nitride (GaN) crystals in large quantities. In recent years, Nobel Prize-winning physicist Prof. Shuji Nakamura introduced two-component Metal-Organic Chemical Vapor Deposition (MOCVD) technology, a breakthrough for growing high-quality GaN [6].

Dye solar cells represent a low-cost, easy-to-produce third-generation solar cell technology with promising applications in certain markets, including wearable electronics and indoor and outdoor applications such as smart labels, posters and decorative tiles for building facades. However, the widespread adoption of dye-sensitized solar cells faces two key performance challenges: limited lifetime and low energy conversion efficiency. This project focuses on maximizing the power conversion efficiency of dye solar cells [7].

Despite the discovery of dye-sensitized solar cells several decades ago, improvement in power conversion efficiency has been slower compared to other solar cell technologies. Various methods have been introduced to improve the power conversion efficiency of dye-sensitized solar cells (DSSCs). The approach taken in this study involves the development of composite GaN-TiO_2 photoanode material. The hypothesis is that the distinctive physical and chemical properties of GaN and TiO_2 will synergistically contribute to the overall improved performance of dye-sensitized solar cells.

One of the predominant materials in DSSCi s titanium dioxide (TiO_2). TiO_2 is used as a photoelectrode in solar cells due to its resistance to UV-A radiation. TiO_2 has a large band gap on the order of 3–3.6 eV [8]. To improve the properties of TiO_2, structures are modified by adding impurities and various organic dyes on the surface to accelerate the charge transfer process. The electronic charge transfer properties in DSSC problems are influenced by crystal defects and structural porosity [9] material thickness [10], the efficiency of power conversion depends on charge transfer and recombination processes. There is an optimal film thickness parameter at which these two processes are most balanced. In the choice of the optimal thickness of TiO_2 to balance the processes of the electronic transition layer is given. The GaN material has a band gap of 3.4 eV and UV-A resistance. As we can see, they are in the same range as TiO_2 [11]. This suggests the possibility of sharing these materials [12].

Use of the GaN/TiO_2 structure in literature reviews indicates excellent performance as UV-activated oxygen sensors at room temperatures, but there is no information on use as a DSSC. In this regard, we assume that physical and chemical properties of GaN and TiO_2 will synergistically contribute to the overall improved performance of dye-sensitized solar cells.

2. Experimental process

2.1 Synthesis of GaN films

Plasma enhanced chemical vapor deposition (PECVD) system with comprising a 500 Watts R.F. plasma source, a 2” split tube furnace, a 4-channel precision mass flow meter with a gas mixer tank, an oil-less Pfeiffer High-Speed (226 L/min) vacuum pump, and a dual-zone furnace were used. The R.F. source's output power is adjustable within the range of 5-500 W with a stability of ±1. The plasma source operates at an RF frequency of 13.56 MHz with ±0.005% stability, and the reflection power can reach up to 200 Watts.

In the synthesis procedure, metallic Ga and powdered ammonium chloride served as the sources of Ga and N atoms, positioned at the entrance of a 20 mm diameter quartz tube with respect to gas flow. This tube was centrally aligned within the main furnace tube to enhance activation by the plasma. Pure nitrogen, with a flow rate of 20 ml/h, was employed as the carrier gas. A vacuum of $5x10^{-2}$ Torr was achieved inside the main tube using the furnace's vacuum pump. Substrates, consisting of sapphire and quartz glass, were positioned at varying distances from the plasma source after the source reagents in the high-temperature zone.

The oven followed a three-stage temperature gradient program, initially heating to 320 °C to prevent boiling of the ammonium chloride but to ensure its uniform evaporation. The oven was then gradually heated up for the actual deposition process and kept at 500 °C. When the set temperature was reached, the plasma source was activated at 120 W and the process was maintained for 2 hours. After the deposition process, the oven was gradually cooled and the substrates were subjected to further examination to determine the presence of heterostructures. Our methods are not complicated compared with this study [13].

2.2 Paste Formulation

The pastes for our dye-sensitized solar cells were made according to the recipe from Hee-Je Kim et al [14] formulated. In this phase of the project, we prepared four different pastes with different weight percentages of GaN in the TiO_2 matrix, in line with our research objectives. Additionally, a reference TiO_2 paste was created. The amounts of chemicals used in the formulation of TiO_2 and 10–40 wt% GaN/TiO_2 pastes are listed in Table 1, following established procedures in the literature [15].

The preparation process adhered to standard protocols. Initially, the measured GaN was ground on an alumina mortar for approximately 10 minutes, with the addition of 400 uL of ethanol during grinding. Grinding continued until a uniformly sized powder was achieved. Following GaN grinding, TiO_2 and polyvinylpyrrolidone (PVP) were incorporated as per the formulation, and the mixture underwent an additional 10 minutes of grinding. Acetyl acetone and ethanol were gradually introduced in specified quantities to prevent aggregation. Subsequently, acetic acid (99.9%) was added, followed by Triton X-100 as a dispersing agent and ethanol. If necessary, the prepared solution could be heated at 90 ºC for 10 minutes to enhance the viscosity of the resulting pastes.

Table 1. Composition of TiO_2, GaN and composite pastes for DSSCs.

Materials	10wt%GaN	20wt%GaN	30wt%GaN	40wt%GaN
Poly(vinylpyrrolidone) (g)	0.08	0.0432	0.0432	0.04
TiO_2 nanoparticles (g)	0.09	0.0432	0.0378	0.03
GaN (g)	0.01	0.0108	0.0162	0.02
Triton X-100 (mL)	0.2	0.108	0.108	0.1
Acetyl acetone (mL)	0.07	0.0324 (0.07)	0.0324 (0.07)	0.03 (0.07)
Acetic acid (mL)	0.07	0.0216 (0.07)	0.0216 (0.07)	0.02 (0.07)
Ethanol (mL)	1	0.54 (1)	0.54 (1)	1

2.3 DSSC construction

To fabricate dye-sensitized solar cells, fluorine-doped tin oxide (FTO) and Pt-coated glass substrates were subjected to a thorough cleaning process. This included a 10-minute ultrasonic bath in deionized water, followed by another 10-minute ultrasonic bath in ethanol. Subsequently, the FTO substrate with Pt coating was determined as a counter electrode after the cleaning process was completed. A Pt layer was then electrochemically deposited on this substrate at a potential of -0.5 V using an aqueous solution containing H_2PtCl_6 (10 mM) and KCl (10 mM) at room temperature. The deposition process took 5 minutes. The cleaned glasses were then air-dried for 5 minutes at ambient temperature.

Next, four different pastes were applied to the FTO-coated side of the glass plates using the squeegee method. A polymer film, specifically Meltonix 1170–25 (Solaronix, Switzerland), served as a spacer between the electrodes in the DSSC cell. To improve the contacts when measring the efficiency of DSSC cells, a tin layer was applied to the edges using the ultrasonic soldering station CS55–X151 (CHEERSONIC) with a frequency of 55 kHz and ECOSOLDER RMA98 SUPER as soldering material. This film effectively prevented short circuits between the electrodes.

After application and drying, the samples were sintered step by step in a muffle furnace (8.21100, Snol) at specific temperatures and durations: 325 °C – 5 minutes; 375 °C – 5 minutes; 450 °C – 15 minutes; 500 °C – 15 minutes. The sintered glass plates were then immersed in a 0.25 mM solution of the commercial ruthenium-based dye N719 for 24 hours. After this period of time, the dye-sensitized photoanodes were rinsed with ethanol and allowed to dry. After drying, 2 μL of an electrolyte consisting of 0.1 M LiI, 1.0 M 1,2-dimethyl-3-propylimidazolium iodide (DMPII), 0.12 M I2, and 0.5 M 4-TBP were applied to the surface of the sensitized films. The sequential steps to construct the dye-sensitized solar cell are shown in Figure 1.

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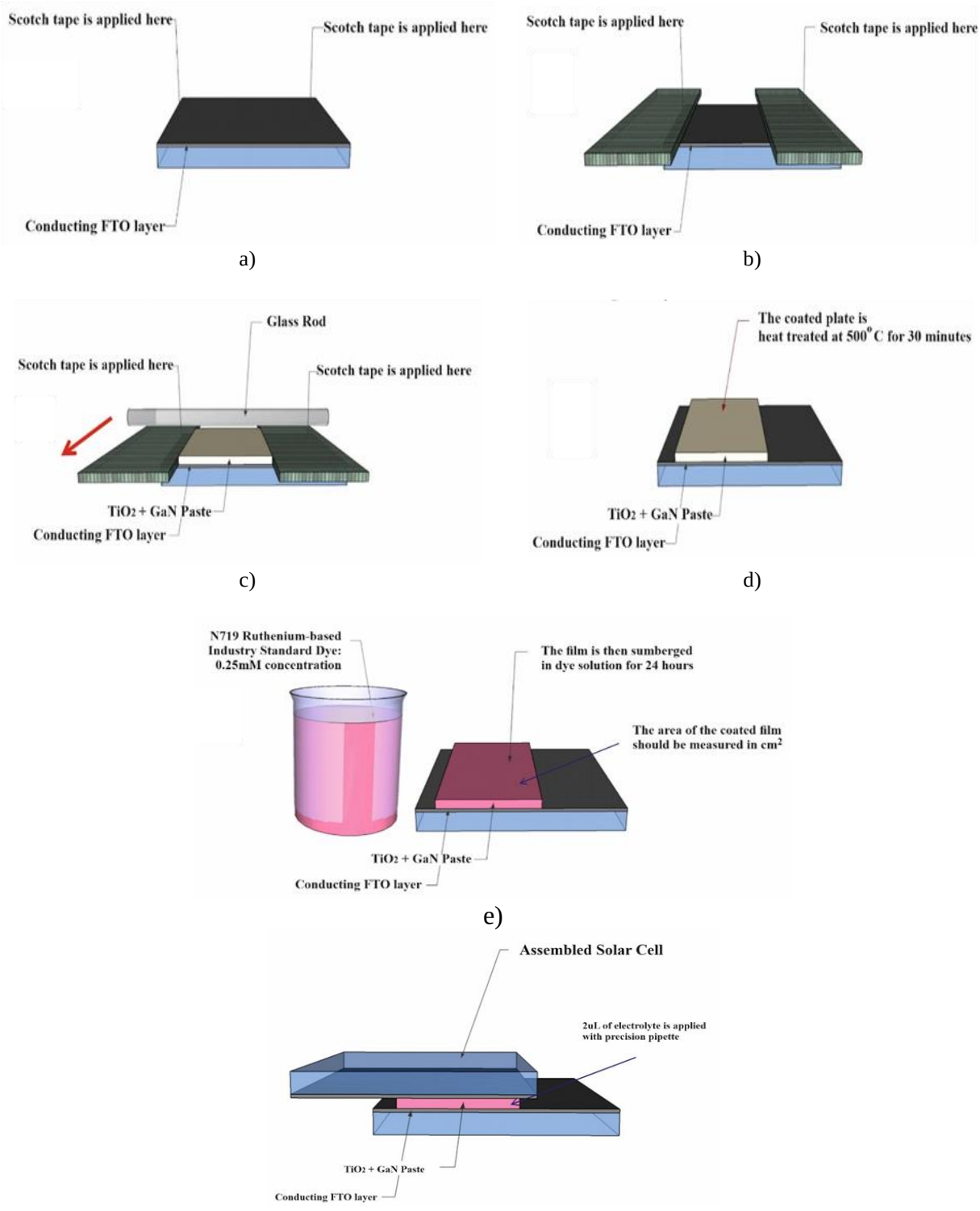


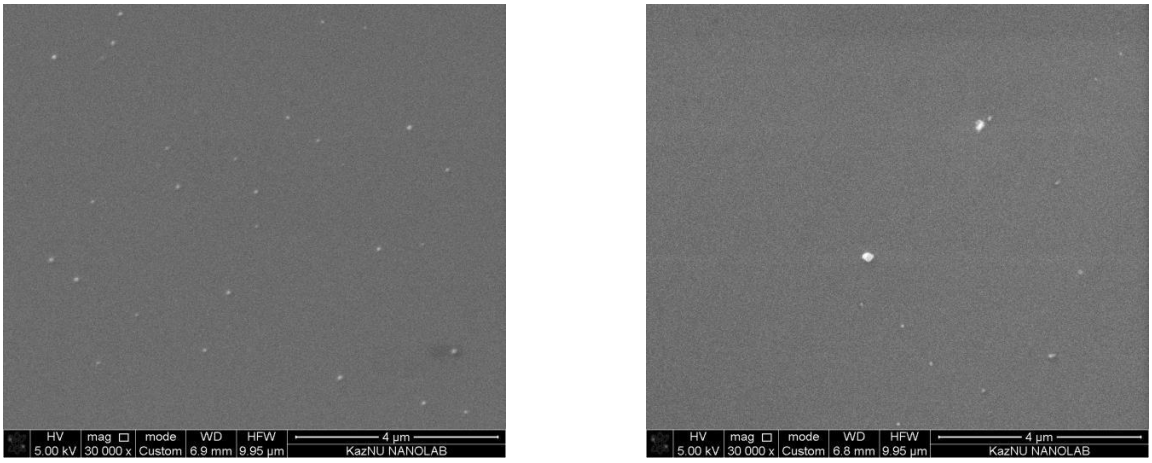
Fig.1. The graphical depiction of the sequential steps in assembling DSSCs.

3. Discussion of results

Surface analyzes of the sapphire and quartz substrates used in PECVD synthesis included scanning electron microscopy, Raman spectroscopy, and X-ray elemental analysis (EDX). According to the SEM results, GaN crystal structures in the form of different islands were observed on both types of substrates (Fig.2). These structures then expanded on the substrate surface and formed a continuous thin polycrystalline film, which is characteristic of Chemical Vapor Deposition **(**CVD) processes. The results suggest that the deposition

technology used, operating at relatively low temperatures with plasma enhancement, is effective in producing polycrystalline GaN structures[4]. The relatively low deposition rate is due to the selected temperature of 500 ºC and can be increased by increasing the temperature if necessary [16]. While the PECVD oven allows temperature increases of up to 1100 ºC, achieving such high temperatures requires additional adjustments to parameters such as gas flow rate, vacuum level and plasma source power [17]. Observations from SEM images indicate that a uniform thin film was not formed on the surfaces of the sapphire and quartz substrates, except for small scattered islands that islands were observed in a previous report [18, 19], as shown in Figure 2.

To confirm the absence of a uniform thin film, Raman spectroscopy analysis was performed (see Fig.3). The absence of GaN-specific peaks corresponding to the zone boundary phonons E1 (high) and A1 (LO) in the spectra confirms the absence of a uniform thin film on the surfaces of sapphire and quartz substrates.



a) b)

Fig.2. The SEM images of GaN islands deposited on different substrates, where (a) Al_2O_3 and (b) SiO_2 substrates

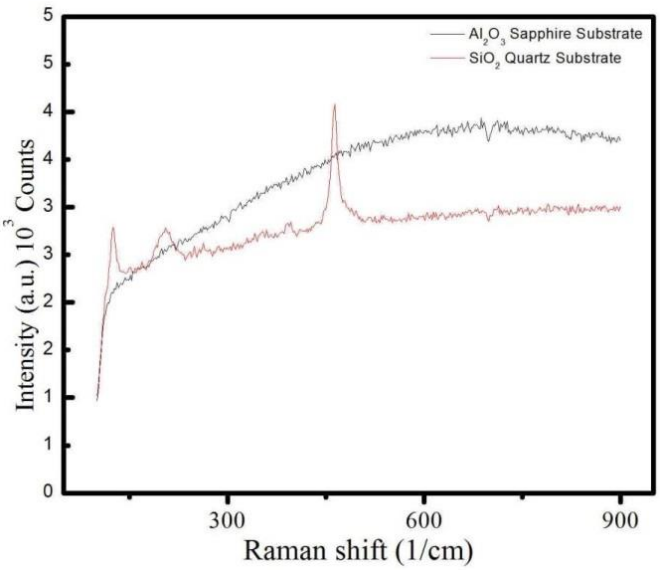


Fig.3. Raman spectra of Al_2O_3 sapphire and SiO_2 quartz substrate samples obtained from a PECVD reactor.

EDX spectroscopy was used to study the surface composition of the substrates, as shown in Figure 4. The chemical composition data of the substrate surfaces show that the ratio of Ga to N exceeds the stoichiometric ratio of GaN. This discrepancy arises from the condensation of Ga from chemical vapors at low temperature and low gas flow rate. The optimal conditions for higher purity GaN films include higher temperatures, a slower cooling rate, and an increased gas flow rate during the post-deposition phase.

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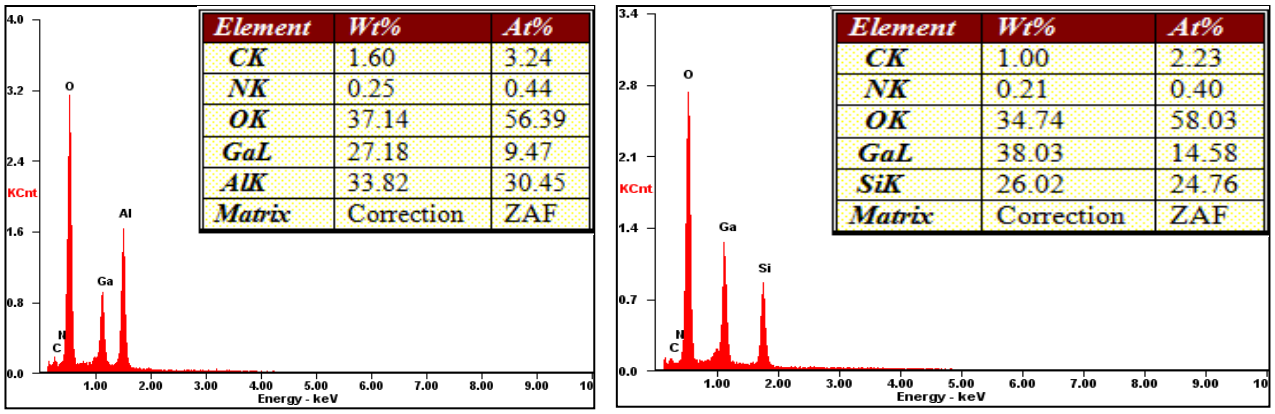

a) b)

Fig.4. EDX data of GaN samples deposited on (a) sapphire-Al_2O_3 and (b) quartz-SiO_2 substrates.

In addition to conducting characterization studies on samples prepared using the plasma-enhanced chemical vapor deposition method, we have expanded our research to investigate the influence of GaN powder in dye-sensitized solar cells. This extension of our work specifically focuses on studying how different amounts of GaN affect photovoltaic performance in mixed structure composite photoanodes. The results of the photovoltaic characterization of solar cells are listed in Table 2. The volt-ampere characteristics and efficiency were then adjusted using a solar simulator, the ST150AAA (PET PHOTO Emission TECH.), under AM 1.5 conditions with the power of the source determined at 100 mW/cm².

Table 2. The photovoltaic parameters of the DSSCs.

Samples	V_{oc} (mV)	I_{sc} (mA)	Fill Factor	Eficiency (%)	Active Area (cm^2)
TiO_2, Reference DSSC	598.60	1.853	0.63	1.76	0.40
10 wt% GaN/TiO_2, mixed DSSC	561.21	0.459	0.65	0.50	0.34
20 wt% GaN/TiO_2, mixed DSSC	656.96	1.049	0.62	0.75	0.58
30wt%GaN/TiO_2, mixed DSSC	591.51	1.148	0.59	0.72	0.60
40wt%GaN/TiO_2, mixed DSSC	491.87	0.480	0.57	0.25	0.55

Due to the time-consuming synthesis process and the limited availability of laboratory-synthesized GaN powder, we initially decided to use commercially available GaN powder. The aim was to determine the optimal weight fraction of GaN in the TiO_2 matrix, which could later be reproduced using GaN powders synthesized in the laboratory. When examining the fill factor (FF) values of the solar cells, it becomes clear that in a mixed structure the FF values decrease with increasing weight proportion of GaN in the TiO_2 matrix. FF values provide information about the level of internal resistance within the solar cell, suggesting that higher amounts of GaN lead to increased parasitic resistance [20].

Conversely, the behavior of the next photovoltaic parameter, power conversion efficiency, differs from FF. Power conversion efficiency starts to increase from 0.50 % for the 10 wt% GaN sample and reaches a maximum of 0.75% for the 20 wt% GaN sample, after which the value starts to decrease that confirmed in a previous report [21, 22]. Therefore, the results of the current experiments suggest that the optimal amount of GaN in the composite photoanode is 20 wt% and our results are consistent with this work [23].

The commercial GaN was analyzed using Raman spectroscopy, as shown in Figure 5. Raman spectroscopy analysis of commercial GaN shows the same three distinct peaks of phonon modes such as zone boundary, E2 (high, 572.81) and A1 (LO) observed in our laboratory synthesized GaN flakes. More E_2 (high) peaks were also found in this report as well [24, 25].

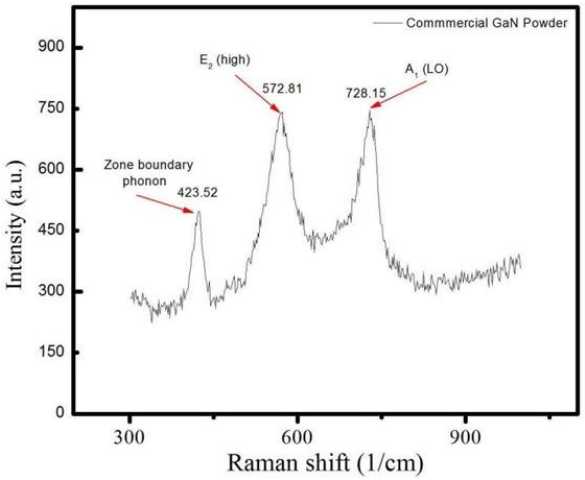

Fig. 5. Raman spectra of commercial GaN powder.

4. Conclusion

In summary, characterization studies were performed on GaN samples cultured on sapphire and quartz substrates. Elemental analysis revealed an excess of Ga on the substrate surfaces, indicating nonstoichiometric formation of GaN with abundant evaporation of liquid Ga at the synthesis temperature of 500 °C. To mitigate this, it is recommended to lower the deposition temperature and/or increase the RF power to improve the delivery of nitrogen-active species to the surface reaction sites and obtain a stoichiometric GaN thin film.

Results were demonstrated for the optimal weight fraction of GaN powder in a mixed photoanode structure. Different GaN weight fractions were incorporated into the TiO_2 matrix to determine the optimal power conversion efficiency of the mixed powder photoanode film. The most favorable result in terms of power conversion efficiency was observed with a GaN composition of 20 wt%, reaching a value of 0.75%. Conversely, the highest fill factor value was achieved with a GaN composition of 10 wt%. Through parameter adjustments, we identified the optimal conditions that resulted in high power conversion efficiencies and fill factor values.

Conflict of interest statement

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

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Toktarbaiuly O.: Conceptualization, Methodology, Supervision; **Baisariyev M.:** Investigation, Writing; **Kaisha A.:** Investigation, Formal Analysis; **Duisebayev T.S.:** Writing - original draft; **Ibrayev N.Kh. and Serikov T.M.:** Review and Editing; **Ibraimov M.K. and Khaniyev B.A.:** Software and Analysis; **Tezekbay Y.Zh. and Zhambyl A. N.:** Investigation; **Nuraje N. and Sugurbekova** G.: Writing-Reviewing and Editing and Supervision.
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