

Study of Optical and Electrical Properties of Carbon Nanotubes (CNTs) Thin Film Deposited by Spray Pyrolysis Method for Optoelectronics Application

Oluwasusi T.V.^{1,2}; Gurku U.M.²; Alayande S.O.³; Babalola V.A.²; Fasiku T.B.⁴; Eneye J.¹,
Sarki M.U.²; Idris M.M.²; Mundi A. A.²; Aribisala A.⁵; Adeleye M.O.¹

¹Department of Physics,
Bingham University,
Karu, Nasarawa State,
Nigeria.

²Department of Physics,
Nasarawa State University,
Keffi, Nasarawa State,
Nigeria.

³First Technical University,
Ibadan, Oyo State,
Nigeria.

Department of Physics,
Ajayi Crowther University,
Oyo, Oyo State,
Nigeria.

Federal Capital Territory Secondary Education Board,
FCT, Abuja.
Nigeria.

Email: oluwasusi.taye@binghamuni.edu.ng

Abstract

Carbon nanotubes (CNTs) are novel material with exceptional optoelectronics properties. CNTs thin films have been used in many different applications. Despite its vast potential, particularly as a promising alternative to indium tin oxide (ITO) and silicon in transparent conducting films, research on CNT thin film remains surprisingly limited. This study creates CNT thin films at 0.1 M and 0.2 M concentration using the spray pyrolysis technique. This technique was used due to its low cost and its ease of use. The optical and electrical characteristics of CNTs thin films created on the sodalime glass substrates at 350 °C were studied in relation to precursor concentration. Ultraviolet Spectrophotometer (UV) and Four-Point Probe Technique characterized the thin films. With rising precursor concentration, CNTs thin films' optical band gap energy decrease. With rise in precursor concentration, the films' transmittance and electrical conductivity grow. The result of the study concludes that concentration has effect on the optical and electrical properties of CNTs thin films and that the high electrical conductivity, high transmittance, low absorbance and low optical band gap energy of the films makes it useful in optoelectronics devices.

Keywords Thin Film; Spray Pyrolysis; Carbon Nanotubes (CNTs); Indium tin oxide (ITO); Optoelectronics.

*Author for Correspondence

INTRODUCTION

The market for transparent conductors is anticipated to increase steadily as electronic gadgets, including displays, solar cells, and touch screens, become increasingly interactive and ubiquitous. Nevertheless, widely utilized transparent conductivity materials like Indium tin oxide (ITO) are limited in availability, costly, and prone to heat deterioration. Therefore, there is a demand for cost-effective materials that exhibit superior performance. Carbon nanotube thin films possess significant potential as high-performance transparent conductors.

Carbon nanotubes (CNTs) are a well-known material with a molecular structure that can be viewed as a sequence of graphene sheets rolled up in specific orientations specified by pairs of integers. They are a type of nanomaterial with exceptional optoelectronic (Yaya *et al.*, 2017; Dabera *et al.*) and mechanical properties (John *et al.*, 2021). CNTs are of two types based on outer layer or wall thickness: single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs) (Sneh *et al.*, 2025). SWCNTs are single graphene rolls with diameters of 0.8 to a few nanometers while MWCNTs are concentric graphene tubes that can have diameters from a few to more than a hundred nanometers. SWCNTs possess high mobility, high transparency, and good flexibility simultaneously.

These attractive properties satisfy the requirements of thin film making CNTs the most promising candidates for optoelectronics applications. The hollow structure, large specific surface area, good electrical, thermal, and mechanical properties, have also drawn a lot of attention to CNTs (Weng *et al.*, 2014). Additionally, CNTs are frequently utilized as thin film materials. CNTs thin films have prospective applications as conductive or semiconductive layers in various electrical, optoelectronic, and sensor systems. The optoelectronic industry requires novel and innovative materials that are lightweight and have outstanding mechanical, thermal, optical and electrical properties. Thanks to their unique properties, nanostructured CNT thin films represent a potential opportunity for advanced applications in the optoelectronics sector. CNT films are guaranteed to have superior performance as well as a lower mass, a critical aspect in optoelectronics applications. CNTs films are also thought to be the best substitute for silicon in future devices due to their high mobilities and ballistic transport characteristics (Appenzeller, 2008).

The production of thin films has been accomplished by a variety of techniques, including the spray pyrolysis method, Pulsed laser deposition (PLD), sputtering, molecular beam epitaxy (MBE), atomic layer deposition (ALD), metal organic chemical vapour deposition (MOCVD), and the sol-gel technique. For the purpose of this study, the spray pyrolysis method was utilized to produced CNTs film. Using this approach is advantageous for a number of reasons, including its low cost, its ease of use, the absence of a high vacuum need, its safety, the reduction of waste production, and its capacity to coat huge surfaces.

Despite the attractive properties of CNT film, few studies have been done on it. Arthur *et al.* (2020) fabricated films of carbon nanotubes (CNTs) using chemical vapor deposition (CVD) technique, optical and electrical characteristics of the film was studied, it was noticed that increased thin layer thickness results in enhanced conductivity. This contrasts with this study in terms of method, as they employed the chemical vapor deposition (CVD) technique to generate CNTs films.

Jinquan *et al.* (2007) directly utilized double-walled carbon nanotubes as energy conversion materials to construct thin-film solar cells, with the nanotubes functioning as both photogeneration sites and a charge carrier collection/transport layer. The solar cells comprise

a semitransparent thin film of nanotubes conformally deposited on an n-type crystalline silicon substrate, forming high density p-n heterojunctions that facilitate charge separation and enable the extraction of electrons via n-Si and holes through nanotubes. Preliminary studies indicate a power conversion efficiency over 1%, demonstrating that DWNTs-on-Si may be a viable structure for solar cell production.

The devices differ from previously documented organic solar cells that utilize mixtures of polymers and nanomaterials, wherein conjugated polymers produce excitons while nanotubes just function as conduits for transport. This contrasts with this study as they generated CNT films from double-walled carbon nanotubes.

This study focuses on production of SWCNTs thin films for advanced applications in the optoelectronics most importantly thin film solar cell. The study investigated the effect of molar concentration on the optical and electrical properties of carbon nanotubes thin film by a spray pyrolysis method. The results from the optical and electrical study gives insight on the applicability of the film in optoelectronics application.

MATERIALS AND METHODS

Materials

Short single-wall carbon nanotubes (SWCNTs) and Dimethyl sulfoxide (DMSO) of purities 99.5 % and 98 % respectively were the reagents used and microscopy glass slides. For the deposition of the thin films specific substrates were employed. The chemical reagents involved in the process were all sourced commercially from Sigma-Aldrich and were utilized directly without any additional purification steps.

Preparation of Precursor Solutions

CNTs precursor solution was prepared at concentrations of 0.1 M and 0.2 M. 1.44 g of 0.1 M CNTs and 2.88 g of 0.2 M of CNTs were dissolved in 20 ml of Dimethyl sulfoxide (DMSO). The solutions were agitated for eight hours. Carbon nanotubes (CNTs) are hydrophobic; hence, the solution does not achieve total dissolution but rather disperses to create a colloidal black solution. The distributed solution was collected for deposition. The glass substrate was first pre-cleaned by sequentially rinsing it with dilute hydrochloric acid (HCl), ethanol, and distilled water. After cleaning, it was positioned on the substrate heater using the substrate holder for the deposition process.

Thin-film Deposition

The thin film deposition method used for this work is spray pyrolysis method. The spray pyrolysis setup is primarily comprised of the following components: a spraying unit, a precursor feeding unit, a temperature control unit, and air blast atomizers, which are all located within the spraying unit. The substrate temperature for the CNTs deposition was kept at a consistent temperature of 350 °C throughout the spray process by maintaining a steady temperature. Spraying the prepared solution directly onto glass substrates that were put on a hot plate stove that was set to 350 °C was accomplished by using an infusion syringe pump to pump the solution at a constant flow rate of 2 milliliters per minute. By passing the spray solution through the nozzle that was positioned 15 centimeters directly above the substrate, a stream of compressed air was utilized to atomize the spray solution. This causes the solvent to evaporate, the solute to deposit into the substrate, and the hydrated salts to undergo pyrolysis, which results in the formation of CNTs thin films. This process occurs when droplets of hydrated salts solutions are sprayed over heated substrates.

Characterization Procedure for the Deposited Thin Films of CNT

The electrical properties of the films were examined using a four-point probe technique, while their optical characteristics were analyzed with a UV-visible spectrophotometer.

Optical Characterization

UV-Visible spectrophotometer was used to examine the transmittance at a wavelength range of 300-900 nm. The value of the transmittance enables other parameters such as absorbance, the absorption coefficient α , the bandgap E_g , of the CNTs thin films to be determined. The relation between absorbance and transmittance is given by the equation below:

$$A = \log \left(\frac{1}{T} \right) \quad (1)$$

Where A is the absorbance and T is transmittance.

Optical bandgap energy is an important parameter in optical and opto-electronics properties of the film. This can be determined by first determining the absorption coefficient α .

Absorption coefficient (α) is an important factor for optoelectronic devices. A characteristic of a material, the absorption coefficient defines how much light it absorbs. The average distance a photon travels before absorption is the inverse of the absorption coefficient. According to the absorption intensity of these materials, this aids in categorizing them as opaque, translucent, and transparent.

The absorption coefficient (α) of the CNTs thin films was determined from the transmittance measurements using Swanepoel's method (Rafia *et al.*, 2017).

$$\alpha = \frac{1}{t} \ln \left(\frac{1}{T} \right) \quad (2)$$

Where t, is the estimated thickness of the film and T is the optical transmittance values.

The estimated films thickness (t) was determined by Equation 3 according to Akinwunmi *et al.* (2022).

$$t = \frac{1}{2n \left(\frac{1}{\lambda_2} - \frac{1}{\lambda_1} \right)} \quad (3)$$

Where n is the index of refraction of the substances, λ_2 and λ_1 are the two associated transmittance highest and lowest values, respectively.

The energy bandgap of the CNTs films was found by analyzing the relationship between the absorption coefficient and photon energy. Tauc's Equation (Rafia *et al.*, 2017) illustrates the association between the intake coefficient (α) and the incident photon energy ($h\nu$):

$$(\alpha h\nu) = A(h\nu - E_g)^n \quad (4)$$

In this context, h represents Planck's constant, ν denotes frequency, $h\nu$ signifies the energy of the input photon, α indicates the absorption coefficient, A refers to the band tail parameter, E_g is the band gap of the material, and the exponent n is contingent upon the nature of the transition.

For direct and permitted transitions, $n = 1/2$; for indirect transitions, $n = 2$; and for direct forbidden transitions, $n = 3/2$ (Owoeye *et al.*, 2023). The calculation of the direct band gap value is represented as of $(\alpha h\nu)^2$ versus $h\nu$. Tauc's plots, depicting of $(\alpha h\nu)^2$ against $h\nu$, were evaluated for the linear extrapolation to the x-axis intercepts, indicating that the deposited CNTs thin films exhibit direct allowed transition characteristics.

Electrical Characterization

Apart from optical characteristics of thin films, the electrical property is also a crucial factor to acquire the performance of thin films. Using a four-point probe (FPP), the electrical characteristics of the films were investigated in order to obtain low resistivity measurements with great degree of precision.

Key electrical characterization factors including resistivity, ρ , and conductivity, σ can be computed from the I-V properties. The conductivity was done inversely from the resistive value computed. The resistivity can be easily calculated from the below equation using the sheet resistance value.

$$R_s = \rho t \quad (5)$$

Where: R_s = sheet resistance, ρ = resistivity, t = Thickness

The four-point probe comprises distinct pairs of electrodes for current delivery and voltage measurement, providing enhanced precision compared to resistivity measurement techniques that utilize just two probes (Islam & Podder, (2009). This method involves the voltage value obtained from the voltmeter and the current supplied by the DC power source. The formula defines the sheet resistance. Islam & Podder (2009).

$$R_s = 4.53 \frac{V}{I} \quad (6)$$

V represent the voltage that was measured linking the two inner probes and I represents the current that was passed by the outer probes. The resistivity was calculated from the Equation 7.

$$\rho = R_s \times t \quad (7)$$

t represents the conducting layer film thickness, R_s represent the sheet resistance and ρ equals the resistivity. Equation 8 enables the conductivity (σ) to be determined as.

$$\sigma = \frac{1}{\rho} \quad (8)$$

RESULTS AND DISCUSSION

Optical Transmittance, Absorbance, and Energy Band Gap of Carbon Nanotube (CNTs) Film

The optical properties of spray-deposited carbon nanotube (CNTs) thin films on glass substrates were analysed using transmittance and absorbance spectra as a function of wavelength (λ) in the ultraviolet-visible (UV-Vis) spectral range (300 - 900 nm). The corresponding spectra are illustrated in Figures 1 and 2. Figure 1 illustrates the transmittance spectra of films produced with varying concentrations of CNTs precursor solutions. The film produced with a 0.1 M CNTs concentration exhibited a high transmittance of approximately 75%, indicating low absorption and minimal light attenuation. Moreover, elevating the precursor concentration to 0.2 M resulted in an augmented transmittance of around 96% across the visible wavelength spectrum.

These results demonstrate a direct correlation between precursor concentration and optical transmittance, whereby transmittance increases with higher CNTs precursor concentrations in the spray deposition process. It can be concluded that the higher the concentration, the more light transmits through the sample. A high transmittance is an important property for photovoltaic and optoelectronics applications.

The optical absorption spectra of the deposited 0.1 M and 0.2 M CNTs films are depicted in Figure 2 where the result obtained was opposed with the transmittance. The films exhibit low

absorbance values; however, the absorbance value of 0.2 M CNTs film is lower than 0.1 M CNTs film.

According to the illustration, the films have high absorbance close to the basic absorption edge, which is around 360 nm. In the visible and near-infrared spectrums, the films show very little absorbance. The reason for this is that most light passes through because photons with longer wavelengths lack the energy necessary to interact strongly with the atoms in the film. Absorbance rises as the wavelength decreases because the interaction between the incident photon and the material becomes more intense. (Hassan,2014). The generated CNTs films' low absorption properties in the visible wavelength range suggest that they might be used as a window layer in thin films solar cell (Adewinbi *et al.*, 2021).

Tauc's approach, as indicated in Equation 4, was used to estimate the optical band gaps of the spray-deposited CNTs thin films. The band gap energy findings for CNTs films produced with 0.2 M and 0.1 M precursor concentrations, respectively, are shown in Figures 3 and 4. The band gap values were determined to be 3.3 eV for the 0.2 M film and 4.23 eV for the 0.1 M film. The film with the higher concentration (0.2 M) has a smaller band gap than the 0.1 M film. A smaller band gap means that electrons can more easily move across the gap. The higher transmittance combined with the lower band gap makes these CNTs films suitable for use as transparent layers in thin-film solar cells and other electronic devices.

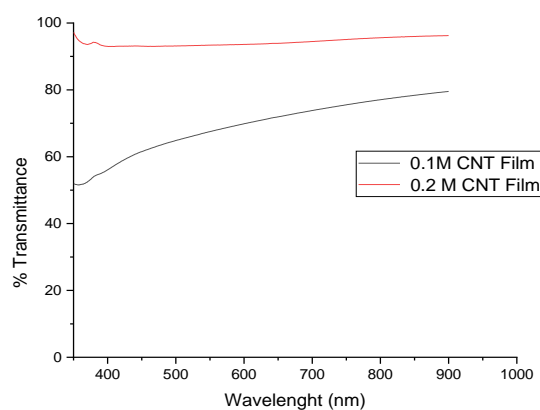


Figure 1
Optical Transmittance Spectra of CNTs Films

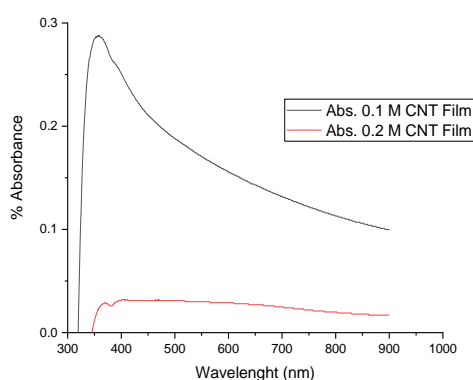


Figure 2
Optical Absorbance of CNTs Film

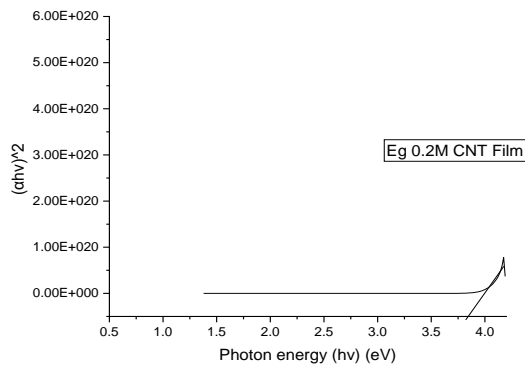


Figure 3: Optical Band Gap of 0.2 M CNTs Film

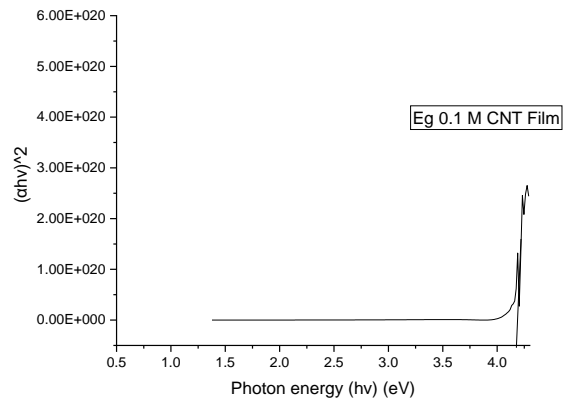


Figure 4: Optical BandGap of 0.1M CNT Film

Electrical Properties of the CNTs Films from Four-point Probe Analysis

Table 1 shows the resistivity and conductivity of the CNTs films deposited at concentrations of 0.1 M and 0.2 M. The electrical analysis indicates that the resistivity of the films decreased from $8.6 \times 10^{-3} \Omega \cdot \text{cm}$ at 0.1 M to $7.87 \times 10^{-3} \Omega \cdot \text{cm}$ at 0.2 M. Meanwhile, the conductivity increased to $12.7 \times 10^1 \Omega^{-1} \cdot \text{cm}^{-1}$ at 0.2 M. This confirms that the single-walled carbon nanotubes (SWCNTs) were most uniform at the higher concentration of 0.2 M. The higher conductivity of the CNTs films makes them suitable as hole-transport materials (HTMs) in light emitting devices (LEDs) and solar cells.

Table 1: Electrical Characterization Parameters of Deposited Films

S/n	Samples	Resistivity ρ ($\Omega \cdot \text{cm}$)	Conductivity σ ($\Omega^{-1} \cdot \text{cm}^{-1}$)
1	0.1M CNTs film	8.6×10^{-3}	11.64×10^1
2	0.2 M CNTs film	7.87×10^{-3}	12.7×10^1

CONCLUSION

The spray pyrolysis technique is a method of thin film deposition that has gained wide spread acceptance owing to its unique properties, especially its simplicity to operate and low cost. The deposited thin films of CNTs have gained a widespread-attention owing to its direct bandgap energy, high transmittance and high electrical conductivity value. The CNTs thin film properties investigated include electrical conductivity, bandgap, transmittance and absorbance. The transmittance value of the CNTs film is between 75% and 96% in the visible region, its low band gap energy and high conductivity values makes it a candidate for applications in the field of optoelectronics.

REFERENCES

- Adewinbi, S.A. Busari, R.A. Adewumi, O.E. & Taleatu, B.A. (2021). Effective photoabsorption of two-way spin-coated metal oxides interfacial layers: surface microstructural and optical studies, *Surface. Interfac.* 23 (2021), 101029.
- Akinwunmi, O.O., Adelabu, O.P., Famojuro, A.T., Akinwumi, O.A., Olaopa, P.O., Olafisan, K.F. & Ajayi, E.O.B. (2022). Characterisation of Zinc Oxynitride Thin Films Prepared Using Zinc (II) Complex of Hexamethylenetetramine as the Precursor. *Materials Sciences and Applications*, 13, 479-489. <https://doi.org/10.4236/msa.2022.138029>

- Appenzeller J. (2008). Carbon Nanotubes for High-Performance Electronics – Progress and Prospect. Proceedings of the IEEE, 96(2), Issue: 2, 201 – 211. DOI: 10.1109/JPROC.2007.911051.
- Arthur, E., Obed O. and Blessing U. O. (2020). Deposition and Characterization of doped carbon nanotube thin film on glass substrate, *European Journal of Material Science* Vol.7, No.1, pp.1-14.
- Hassan Ahmed J. (2014), Study of Optical and Electrical Properties of Nickel Oxide (NiO) Thin Films Deposited by Using a Spray Pyrolysis Technique (2014), *Journal of Modern Physics*,, 5, 2184-2191 <http://www.scirp.org/journal/jmp> <http://dx.doi.org/10.4236/jmp.2014.518212>
- Islam, M. R. & Podder, J. (2009). Optical Properties of ZnO Nano fibre Thin Films grown by Spray Pyrolysis of Zinc Acetate Precursor. *Crystal Research Technology*, 44 (3): 286- 292.
- John J C, Sebastian T, Cyriac J, Paul A, Jose A, Shaji S & Augustine S 2021 Optoelectronic Characteristics of In₂S₃-CNT Nanocomposite Thin Films for Photodetector Application *J. Electron. Mater.* 50 2800–12
- Owoeye V.A., Adewinbi S.A., Salau A.O., Ayodele O.N., Adeoye A.E. & Akindadelo A.T. (2023). Effect of precursor concentration on stoichiometry and optical properties of spray pyrolyzed nanostructured NiO thin films *Heliyon* 9 e13023. <https://doi.org/10.1016/j.heliyon.2023.e13023>.
- Rafia, B. (2017). Effect of Precursor Concentration on Structural Optical and Electrical Properties of NiO Thin Films Prepared by Spray Pyrolysis, *Journal of Nanomaterials* Volume 2017 <https://doi.org/10.1155/2017/5204639>.
- Sneh P.B. William S.W., Priyanka K. & Milad T. (2025). A review of advancements, properties, and challenges of carbon nanotubes in food packaging, *Journal of Food Measurement and Characterization* (2025) 19:2172–2194 <https://doi.org/10.1007/s11694-025-03127-7>
- Theraja, B. L. (2007). A Textbook of Electrical Technology; S. Chand Publishing, 2014 - Technology & Engineering - 2784 pages.
- Weng, B., Yang, M. & Xu, Y. (2014). Toward the enhanced photoactivity and photostability of ZnO nanospheres via intimate surface coating with reduced graphene oxide. *Journal of Materials Chemistry A*, 2(23), 9380-9389.
- Yahya I., Theng L.L., Mustaza S.M., Abdullah H & Amin N 2017 Characterization of transparent conducting carbon nanotube thin films prepared via different methods *Sains Malaysiana* 46 1103–9