



Enhancing the Optical and Solid-State Properties of Spray-Deposited Polyaniline Thin Films via Green Doping with *Moringa oleifera* Leaf Extract for Advanced Light-Harvesting Applications

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Abstract: The pursuit of sustainable materials for optoelectronics has intensified research into conducting polymers doped with eco-friendly agents. This study presents a comprehensive investigation into the synthesis and characterization of polyaniline (PANI) thin films doped with a natural dye extracted from *Moringa oleifera* leaves using a cost-effective spray pyrolysis technique. The primary objective was to evaluate the impact of this green dopant on the structural, optical, and solid-state properties of PANI. The optical properties of undoped and dye-doped PANI films (with dye volumes of 5, 10, 15, and 20 mL) were systematically analyzed using UV-Vis-NIR spectroscopy. The results demonstrate that *Moringa oleifera* doping profoundly enhances the films' photophysical characteristics. A significant increase in absorbance within the ultraviolet and visible spectra was observed, with the 15 mL doped sample exhibiting optimal performance. Crucially, the optical band gap narrowed from 1.88 eV for undoped PANI to 0.89 eV for the 15 mL doped film, indicating enhanced electronic conductivity. Furthermore, the doped films showed substantially reduced reflectance and transmittance, alongside modified dielectric constants and refractive indices. These alterations are attributed to the introduction of new charge carriers and electronic states by the phytochemicals in the moringa extract. The findings conclusively establish *Moringa oleifera* as an effective, sustainable dopant for engineering PANI's properties, positioning these novel thin films as highly promising, eco-friendly materials for application as absorber layers in photovoltaic cells and anti-reflective coatings.

1. Introduction

The escalating global energy demand, coupled with the environmental degradation caused by fossil fuels, has necessitated an urgent transition to renewable energy sources. Solar energy, being abundant and clean, is at the forefront of this transition. Photovoltaic technology, which converts sunlight directly into electricity, is a critical pillar of solar energy harnessing. While silicon-based solar cells dominate the market, their production is energy-intensive and costly, driving the search for alternative, cost-effective materials (Kolhar *et al.*, 2023; Patni *et al.*, 2020; Jordon *et al.*, 2020; Paul-Nwokocho *et al.*, 2018).

Conducting polymers have emerged as a revolutionary class of materials in this context. Among them, polyaniline (PANI) has garnered significant attention due to its unique combination of properties, including excellent environmental stability, straightforward synthesis, tunable electrical conductivity,

and low cost (Nandi *et al.*, 2015; Alzoubi *et al.*, 2025; Shnkar *et al.*, 2020; Chander *et al.*, 2025; Mansour *et al.*, 2015; Jarad, 2018). The electrical and optical properties of PANI are highly dependent on its oxidation state and the degree of protonation (doping) (Reda *et al.*, 2012; Sharma *et al.*, 2013; Vadiraj *et al.*, 2015; Jeroh *et al.*, 2012; Kumar *et al.*, 2024; Abouri *et al.*, 2025). PANI can exist in several distinct oxidation states: the fully reduced leucoemeraldine base, the partially oxidized emeraldine base (insulating), and the fully oxidized pernigraniline base. The emeraldine salt, obtained by protonating the emeraldine base, is the highly conductive form of the polymer (Aziz *et al.*, 2024; Bensedira *et al.*, 2022). As an absorber layers, PANI thin films primarily function as light-absorbing components in optoelectronic and electromagnetic applications rather than as primary photovoltaic absorbers (where inorganic semiconductors like perovskites or CIGS dominate). Their absorption arises from π - π^* transitions and polaron/bipolaron bands in doped states, typically showing peaks around 370–800 nm depending on doping level and morphology (Al-Azzawi *et al.*, 2024; Medhin *et al.*, 2024).

The potential applications of PANI are vast, spanning from sensors and batteries to corrosion protection and electromagnetic shielding (Conroy *et al.*, 2003). In the realm of photovoltaics, PANI has been explored as a hole-transport layer, a counter electrode in dye-sensitized solar cells (DSSCs), and as a sensitizer itself due to its π -conjugated electron system (Saoudi *et al.*, 2021; Pousali *et al.*, 2016; Bouziani *et al.*, 2023). However, the performance of intrinsic PANI in light-harvesting applications is often limited by its relatively wide band gap and less-than-optimal charge transport properties. These limitations can be overcome through strategic doping, which tailors its electronic band structure and enhances its optical absorption (Baker, 2019; Patni *et al.*, 2020; Roy *et al.*, 2019).

2. Methodology

2.1 Materials

The chemicals used in the preparation of PANI are Ammonium peroxide sulphate (APS) [(NH₄)₂S₂O₈], Hydrochloric acid (HCl), and Aniline [C₆H₅NH₂] as the reactants, *Moringa oleifera* (MO) leaves, glass substrates, electrical oven, distilled water, Veeco Dektak 150 Surface Profiler, UV-Visible spectrophotometer Shimadzu UV-1601 and Four-point probe with a signatone quad pro resistance mapping V3.7 machine.

2.2 Sourcing of materials

Commercial glass slides were supplied by CONRAWS Chemical Company, Enugu State, Nigeria was used as substrates for the deposition. While *Moringa oleifera* (MO) as dye was sourced locally from Presco Campus, Ebonyi State University, Abakaliki, Ebonyi State, and was identified by taxonomist in the Department of Applied Biology, Ebonyi State University, Abakaliki, Nigeria. The ethanol, Ammonium peroxide sulphate (APS) [(NH₄)₂S₂O₈], Hydrochloric acid (HCl), and Aniline [C₆H₅NH₂] were purchased from CONRAWS Enugu State, Nigeria.

2.3 Preparation of Polyaniline (PANI)

0.3M of aniline, 0.5M of HCl and 0.5M of Ammonium persulphate were used in the synthesis. 20mL of each of them was used. Polyaniline (PANI) was synthesized from aniline through oxidative polymerization, typically using ammonium persulphate as an oxidizing agent in an acidic solution. 20mL of the prepared aniline was first dissolved in 20mL HCl acid and stirred at room temperature for 4hr before 20mL of Ammonium persulphate was added drop wise until a dark brown coloured solution formed and was stirred further for another 1hr to ensure homogeneity of the solution. The solution was

kept at room temperature for 24hr before deposition. Spray pyrolysis method was used to deposit PANI on glass substrate. Concentration of PANI was kept constant at 20mL for every deposition while the concentration of the green dye extract was varied as 5mL, 10mL, 15mL and 20mL for every deposition.

2.3 Optical and Solid State Characterization of Polyaniline (PANI) Thin Film

The optical and solid state properties of the synthesized PANI thin films were recorded over a wavelength range of 300 to 1000 nm, within an ultraviolet (UV) region, visible light (VIS) region and near infrared (NIR) region. A UV-Visible Spectrophotometer (Shimadzu UV-1601) was used for the optical measurements.

3. Results and Discussion

3.1 UV Spectroscopy analysis

The UV-VIS results for un-doped and doped PANI are shown in Figures 1-6.

Absorbance

The plots of absorbance against wavelength for un-doped and doped PANI thin films with 5 mL, 10 mL, 15 mL and 20 mL volumes of *moringa oleifera* dye extracts are shown in Figure 1. Figure 1 indicates the plots of absorbance versus wavelength for un-doped and doped PANI thin films with 5 mL 10 mL, 15 mL and 20 mL volumes of *moringa oleifera* dye extracts. Generally, the absorbance decreased with increasing wavelength. It was also observed that all the films indicated maximum absorbance in the ultra-violet region, and from there the absorbance decreased as the wavelength increased towards the NIR region.

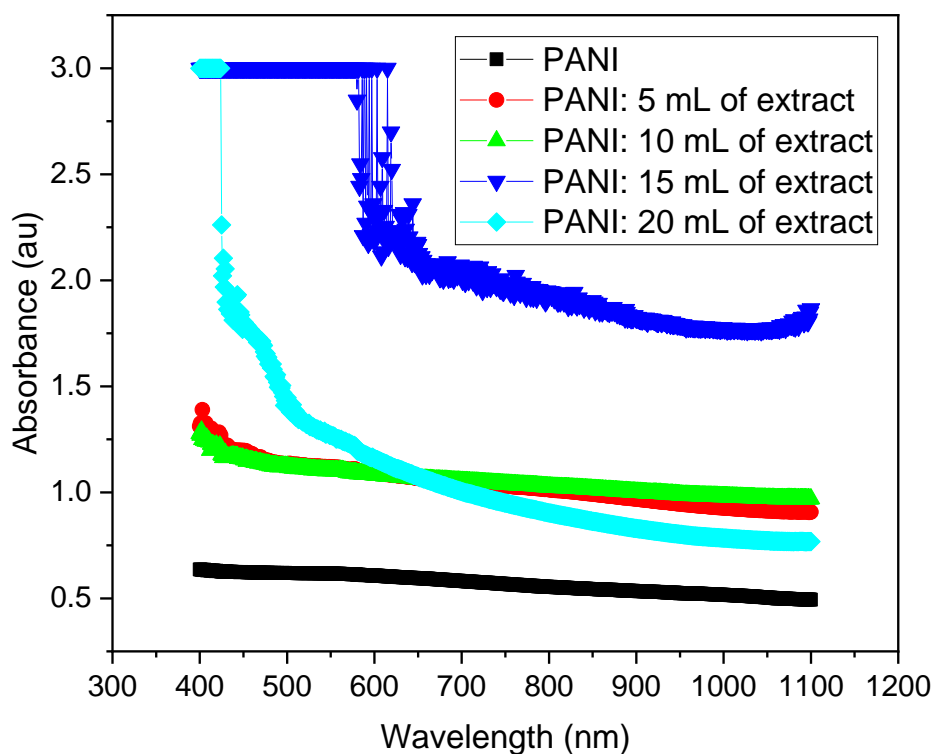


Figure 1. Plots of absorbance against wavelength for un-doped and doped PANI thin films at various volumes of moringa dye extracts

For PANI thin films doped with 20 mL volume of *Moringa oleifera* dye extracts, absorbance decreased from 2.2 at 400 nm to 0.75 at 1100 nm. For PANI thin films doped with 15 mL volume of *moringa oleifera* dye extracts, absorbance decreased from 3.6 at 400 nm to 1.7 at 1100 nm. For PANI thin films doped with 10 mL volume of *Moringa oleifera* dye extracts, absorbance decreased from 1.25 at 400 nm to 1.0 at 1100nm. For PANI thin films doped with 5 mL volume of *Moringa oleifera* dye extracts, absorbance decreased from 1.3 at 400 nm to 0.9 at 1100 nm. For un-doped thin films, absorbance was approximately constant across the entire electromagnetic spectrum. Again, it was also observed that the obtained range of absorbance in this work was above the range reported by Paul-Nwokocho and Ozuomba (2015).

Moreover, it was also observed that the 15 mL of *Moringa oleifera* doped PANI exhibited the best optical absorbance in the UV, visible and NIR regions among the five samples. Furthermore, it was also observed that the introduction of dye extracts of moringa into the spray solution, altered the absorbance spectra. The absorbance of the doped samples was higher than that of the undoped sample. The high absorption of light by the doped samples could be attributed to the pigment (photoreceptor) in the moringa extracts that trap or absorb the ultraviolet light. Strong absorptions were observed at wavelength of 400 nm. Thus, the films have potential application in the fabrication of solar cells.

Transmittance

A graph of the film's transmittance plotted versus wavelength is indicated in Figure 2. The transmittance increased with an increase in wavelength, exhibiting a minimum in the ultraviolet region and a maximum in the near infrared region of the electromagnetic spectrum. The poor transmission of solar radiation exhibited by films in the ultraviolet region was a result of high absorbance of solar radiation in the ultraviolet region, as shown in Figure 2.

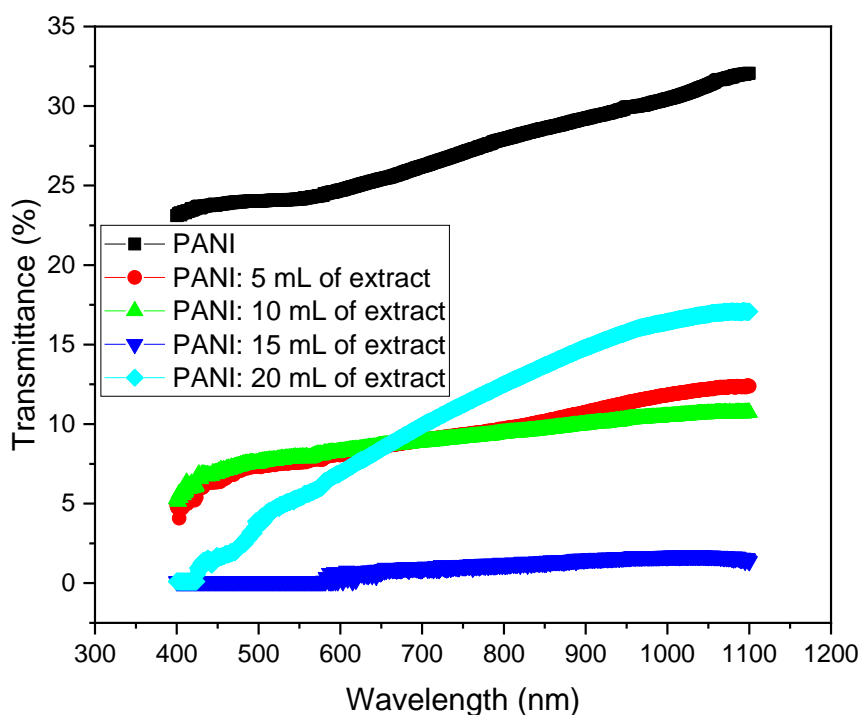


Figure 2. Plots of transmittance against wavelength for un-doped and doped PANI thin films at various volumes of moringa dye extracts.

For PANI thin films doped with 20ml of *Moringa oleifera* dye extracts, transmittance increased from 0% at 400 nm to 17% at 1100nm. For PANI thin films doped with 15ml volume of *moringa oleifera* dye extracts, transmittance increased from 0% at 400 nm to 2% at 1100nm. For PANI thin films doped with 10ml volume of *moringa oleifera* dye extracts, transmittance increased from 5% at 400nm to 11% at 1100nm. For PANI thin films doped with 5ml volume of *moringa oleifera* dye extracts, transmittance increased from 4% at 400nm to 12% at 1100nm. For un-doped thin films, transmittance increased from 22.5% at 400nm to 32.5% at 1100nm. It was also observed that the un-doped sample indicated higher transmittance than the doped samples.

Moreso, it was observed that the doping of PANI with the moringa extracts modified the transmittance spectra. The transmittance of the un-doped sample was higher than the doped sample. The higher transmission of light by the un-doped sample may be due to the absence of the pigment in the PANI. As earlier explained that the doped samples contain pigment that absorb or trap the light.

Reflectance

The plots of reflectance against wavelength for un-doped and doped PANI thin films with 5 mL, 10 mL, 15 mL and 20 mL volumes of *moringa oleifera* dye extracts are shown in Figure 3. As observed in the reflectance spectra in Figure 3, the peak reflectance is about 98 %. Generally, the reflectances of the films were high, suggesting that most of the light was strongly reflected. Put in another way, the reflectances of the films were high, suggesting that most of the light were neither absorbed nor transmitted. It was also observed that the reflectance of the un-doped films was lower than the doped films. These films exhibited high reflectance. High reflectance shown by the films indicated that the films are good reflector of light and they could be employed in high-reflector coatings such as in the coating of mirrors.

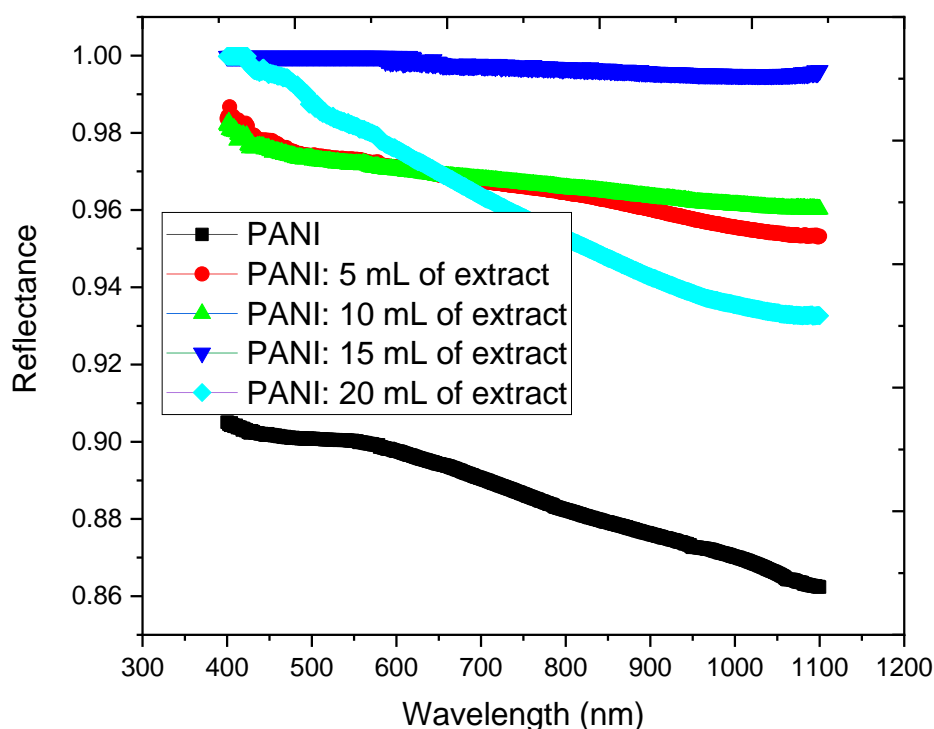


Figure 3. Plots of reflectance against wavelength for undoped and doped PANI thin films at various volumes of moringa dye extracts

Jeroh and Okoli (2012) stated that materials that exhibit high reflectance, especially in the infrared region of the electromagnetic spectrum, could be useful in the manufacturing highly reflectance mirrors commonly found in desktop scanners, photocopy machines, astronomical telescopes, car head lamps, and halogen lamps. The spray-deposited films exhibited high reflectance in the infrared regions, and therefore, they could be used in the applications listed above by Jeroh and Okoli (2012).

Refractive index (n)

The plots of refractive index (n) against photon energy for undoped and doped PANI thin films with 5 mL, 10 mL, 15 mL and 20 mL volumes of *moringa oleifera* dye extracts are shown in Figure 4. Generally, the refractive index of the films decreased as the photon energy increased, as shown in Figure 4. For PANI thin films doped with 20 mL volume of *moringa oleifera* dye extracts, refractive index decreased from 1.25 at 1.0 eV to 0.0 at 3.0 eV. For PANI thin films doped with 15 mL volume of *moringa oleifera* dye extracts, refractive index remained nearly constant across the entire electromagnetic spectrum. For PANI thin films doped with 10 mL volume of *moringa oleifera* dye extracts, refractive index decreased from 0.75 at 1.0 eV to 0.25 at 3.0 eV. For PANI thin films doped with 5 mL volume of *moringa oleifera* dye extracts, refractive index decreased from 0.8 at 1.0 eV to 0.2 at 3.0 eV. For un-doped thin films, refractive index decreased from 2.1 at 1.0 eV to 1.75 at 3.0 eV. The obtained refractive indices of the doped PANI in this work were below the refractive indices of the doped PANI obtained by Baker (2019). The variation in refractive index may be attributed to the doping conditions, dopants type and the methods used in growing the films.

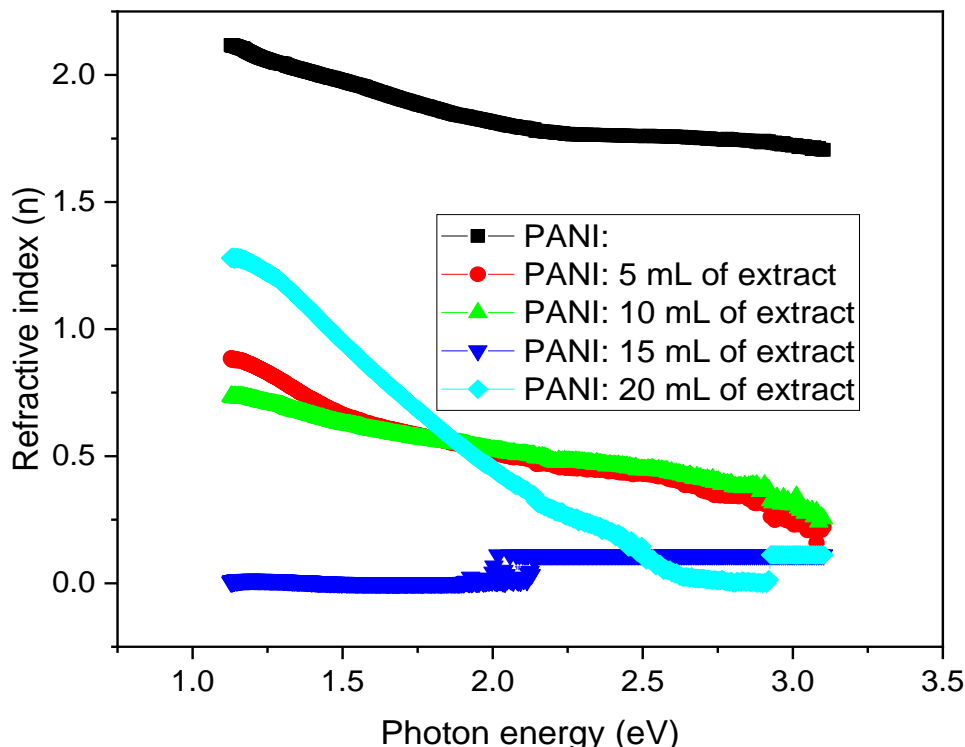


Figure 4. Plots of refractive index (n) against photon energy for un-doped and doped PANI thin films at various volumes of moringa dye extracts

Absorption coefficient (α)

The plots of Absorption coefficient (α) against photon energy for un-doped and doped PANI thin films with 5 mL, 10 mL, 15 mL and 20 mL volumes of *moringa oleifera* dye extracts are shown in Figure 5. As can be seen in Figure 5, the absorption coefficient of both the un-doped and doped samples increased as photon energy increased. It was also noted that the absorption coefficient of the un-doped sample was lower than the doped samples. For PANI thin films doped with 20 mL volume of *moringa oleifera* dye extracts, absorption coefficient increased from 1.7 at 1.25eV to 4.1 at 2.7eV. For PANI thin films doped with 15ml volume of *moringa oleifera* dye extracts, absorption coefficient increased from 4 at 1.25 eV to 7 at 2.0 eV.

For PANI thin films doped with 10ml volume of *moringa oleifera* dye extracts, absorption coefficient increased from 2.1 at 1.25eV to 2.2 at 3.0eV. For PANI thin films doped with 5ml volume of *moringa oleifera* dye extracts, absorption coefficient increased from 2 at 1.25 eV to 2.3 at 3.0 eV. For un-doped thin films, absorption coefficient remained approximately constant across the photon energy axis. The obtained values of absorption coefficients of the films indicate that they could be used as perfect absorber in photovoltaic conversions in energy industries.

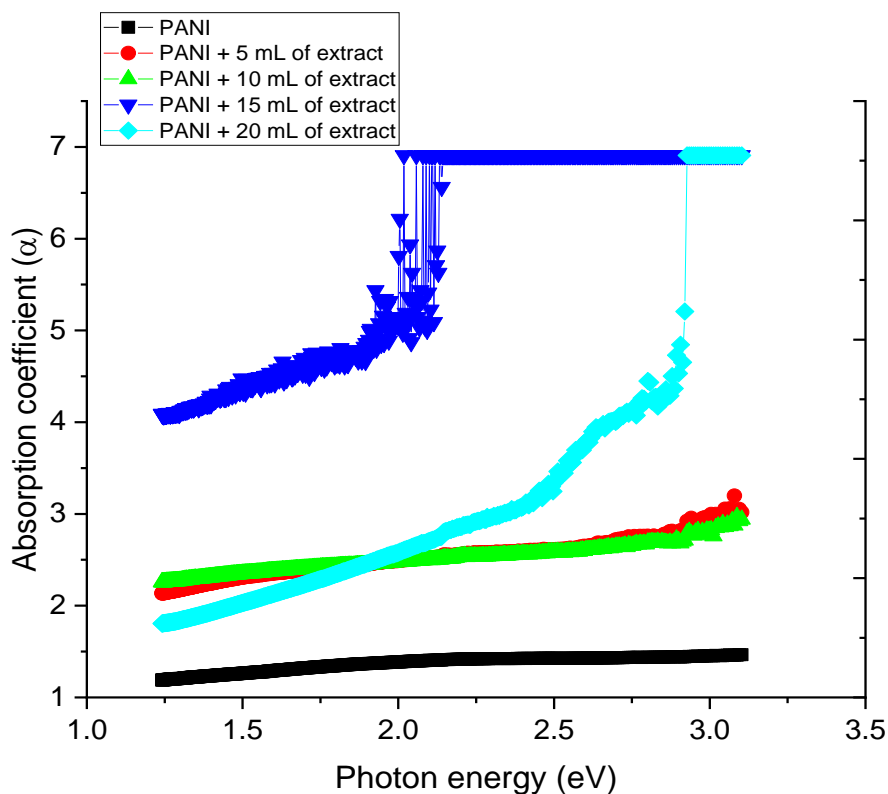


Figure 5. Plots of absorption coefficient against photon energy for undoped and doped PANI thin films at various volumes of moringa dye extracts

Optical band gap ($\alpha h\nu$)^{1/2}

The Tauc plots describe the dependence of the absorption coefficient and the photon energy for indirect interband transitions (Conroy *et al.*, 2003) as presented in the Eqn. 1

$$(\alpha h\nu)^{1/2} = A(h\nu - E_g) \quad \text{Eqn. 1}$$

where A is a constant, α is absorption coefficient, h is a Plank constant and ν is the frequency of the radiation and E_g is the band gap energy. As indicated in Figure 6, the optical band gap, E_g , of the films were determined from extrapolation of the straight portion to the photon energy axis at $(\alpha h\nu)^{1/2} = 0$.

They were found to be 2.2eV, 1.62eV, 0.89eV, 1.26eV and 1.38eV for the un-doped, doped with 20 mL, 15mL, 10 mL and 5 mL, respectively. The obtained value of optical band gap was found to decrease from 2.2eV to 0.89eV after doping with various volumes of *moringa oleifera* extracts implying an increase in conductivity.

The decrease in the optical band gap of the PANI by doping with various volumes of moringa dye extracts from 1.62eV to 0.89eV may be attributed to reduction in the disorder of the system [Reda and Al-Ghannam, \(2012\)](#) and modification of the polymer structure ([Mansour *et al.*, 2015](#)). Similar observation in the variation of the optical band gap from 2.62eV to 2.42eV for pure and doped PANI with HCl and H₂SO₄ acids was reported by Baker (2019). Again, the same observation in the variation of the optical band gap from 2.75eV to 2.4eV for pure PANI, citric acid-doped PANI and H₂SO₄-doped PANI was reported by [Paul-Nwokocha and Ozuomba \(2015\)](#).

However, the optical band gaps obtained in this work were smaller than the optical band gaps reported by [Paul-Nwokocha and Ozuomba \(2015\)](#). This variation in the optical band gap could be due to the type of dopants, deposition conditions, and deposition techniques used to deposit the films. According to [Asogwa *et al.* \(2009\)](#), thin films with optical band gap smaller than 1.90eV are used as absorber materials in solar cell architecture, while those with a larger optical band gap could be used as window layers. Since the obtained values in this research work are below this value 1.90eV. For these reasons, the films could be used as ideal absorbers in the fabrication of solar cells.

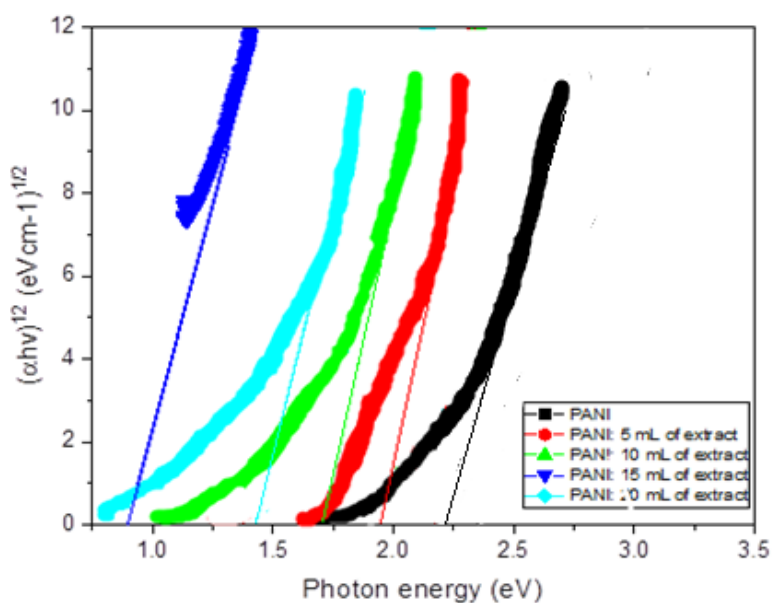


Figure 6. Plots of $(\alpha h\nu)^{1/2}$ against photon energy for un-doped and doped PANI thin films with various volumes of moringa dye extracts

Conclusion

PANI and PANI/*Moringa oleifera* dye nanocomposites have been successfully synthesized by the spray pyrolysis deposition method and characterized using VU-Visible absorption spectroscopy. The results confirmed that doping PANI thin films with moringa dye extracts significantly enhances their optical properties, including light absorption, reflectance, refractive index, and optical band gap.

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Compliance with Ethical Standards: This article does not contain any studies involving human or animal subjects.

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