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ENHANCED CONDUCTIVITY AND APPLICATION POTENTIAL OF POLYANILINE NANOCELLULOSE COMPOSITE FILMS IN FLEXIBLE ELECTRONICS

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ABSTRACT
This work presents the study of polymer-based nanocomposite films, consisting of polyaniline doped with nanocellulose. The difficulty in doping PANI with semiconductor composites, and low solubility in most of the available solvents constrains the commercial applications of PANI, especially in flexible electronic devices. Hence, the need to explore new and promising organic conducting materials for high-temperature applications and optoelectrical applications cannot be overstated. The charge transport abilities of the films were studied as a function of the concentration of PANI. The syntheses were carried out using ammonium persulfate as an oxidant with different molar ratios of oxidant to monomer. Deposition of the material was done to facilitate the Profilometry results which showed the thickness of each Polyaniline nanocellulose composite films. Spectroscopic techniques, such as UV-Visible spectroscopy, was used to carefully study the optical characteristics of the composite films. The electrical properties of the PANI nanocomposite films were investigated using the Hall Effect measurement system which showed the carrier density, electrical conductivity, resistivity, sheet resistance and mobility of polyaniline nanocellulose composite films. The results show that the composite films have excellent resistance to temperature, and with a charge mobility of approximately 4.002E-01 – 1.893E+10 (cm^2/Vs). Additionally, the effects of PANI concentration on the charge transport were investigated. It was found that increasing the concentration of PANI up to 0.5 wt% results in an increase in the charge transport coefficient, while further increases in concentration caused a decrease in the charge transport coefficient. The synthesized films exhibited enhanced electrical conductivity, improved mechanical flexibility, and demonstrated potential applications in flexible electronics, sensors, and actuators. This advancement in composite materials shows significant promise for future flexible electronic devices. Overall, the results of this study demonstrate that PANI nanocomposite films when doped with nanocellulose possess excellent charge transport properties and are capable of reducing the electrical leakage current.

KEYWORD: Polyaniline, Nanocellulose, Conductive Polymers, Flexible Electronics, Composite Films, Optical properties, electrical properties.

INTRODUCTION
The advent of conducting polymers marked the arrival of a novel class of materials that have captured the interest of both academic research and industrial development. These materials uniquely marry processability and solubility with semiconducting or metallic electronic behaviour, rendering them valuable in devices such as light-emitting diodes and photovoltaic cells. The interface between the polymer and the electrode plays a critical role in device function, as the choice of electrode significantly influences charge injection and overall performance (Ravindranadh & Rao, 2013). Traditional models often treat electrode polymer junctions as abrupt, with charge transfer barriers derived from bulk properties of the two materials. In practice, though, interfacial regions can undergo compositional and electronic reorganizations that deviate from this simplified picture. Before the discovery of conducting (conjugated) polymers, polymers were largely regarded as electrical insulators. However, conjugated polymers possess extended π-electron systems that allow for delocalization of charge imparting them with electrical and optical properties akin to inorganic semiconductors (Anas et al., 2020). In these systems, alternating single and double bonds foster

highly delocalized, polarized π-bond frameworks that underpin their conductive and optoelectronic behaviour.

Despite decades of study, a complete mechanistic understanding of charge transport in conducting polymers remains elusive, largely because of the significant structural disorder often present. The electrochemical properties of conductive polymers have long attracted attention due to their intriguing physics, versatility of structure, and wide-ranging applications (Ferloni et al., 2000). As organic electronics continues to evolve, conducting polymers have risen to prominence as candidate materials for many devices (Inzelt, 2011). Although ultimate applications generally demand solid-state performance, electrochemical and Spectro electrochemical investigations still provide essential insights into the electronic behaviour of these materials (Lyons, 2013).

While broad mechanisms for charge conduction in conductive polymers are known, rigorous, universal quantitative relations for charge-transfer parameters are lacking. Conductivity in such systems is not a fixed property but a complex function of charge carrier density and mobility (Dyer et al., 2007). Real polymer systems may host multiple types of mobile carriers, each with field-dependent density and mobility. In disordered, non-ideal semiconducting systems, constructing predictive models thus becomes highly challenging. To unravel conductivity behaviour, it is essential to distinguish between DC (direct current) and AC (alternating current) conductivity phenomena (Lyons, 2013). AC conductivity reflects the ability of mobile charge species to oscillate under a time-varying field, whereas DC conduction involves steady-state charge flow subject to saturation at contacts. A system may show significant AC conductivity without supporting appreciable DC conduction especially if ionic conduction dominates rather than electronic conduction (Dyer et al., 2007).

Among conductive polymers, polyaniline is one of the earliest discovered and remains a widely studied system. It continues to serve as a model material for testing characterization methods and exploring structural property relationships. In this work, we focus on the synthesis and in-depth characterization of nanocellulose-doped polyaniline composite films. Our goal is to enhance both electrical conductivity and explore its potentials in flexible electronic applications.

MATERIALS AND METHODS

Nanocellulose served as the catalyst for the in-situ polymerization of aniline in this study. Ammonium persulfate was used as the oxidant in the various syntheses that were carried out with varying molar ratios of monomer to oxidant.

Preparation of Nanocellulose: Nanocellulose was prepared from medical-grade cotton wool by partial acid hydrolysis with 50% sulfuric acid at 40 °C for 10 minutes (Wulandari et al., 2016). The sample was purified by centrifugation at 4000rpm and washing with water to remove excess acid and soluble sugars. Final concentration 21mg/ml.

Preparation of PANI By Self-Stabilized Dispersion Polymerization as Discussed by (Lee et al., 2005)

(1) Ammonium persulfate (APS) 0.1M/L Dissolved in 1M HCL (chill to < 0^{o}C)
(2) Aniline 0.44mol/L in 1M HCL (chill to < 0^{o}C)
(3) Nanocellulose 21mg/ml (chill to < 0^{o}C)

Solvent 1:1:2 mixtures of propanol, water, and Chloroform

20ml of (2) is mixed with (3) (volume ratios of 1:0; 1:1; 1:2; 1:3; 1:4; 1:5; 1:6; 1:7; 1;8 designated as samples 0 to 8) finally, 20ml of (1) is added dropwise throughout 40min with stirring and simultaneously cooling.

Subsequently, the reaction mixture was maintained at < 0^{o}C for 8 hours to complete the propagation reaction.

The product was purified by centrifugation and washing with distilled water, and the process was repeated six times.

Sample Preparation

i. Substrate cleaning
ii. Sonicate soda lime glass substrates with 1% sodium lauryl sulfate aqueous solution/5min.
iii. Rinse with distilled water.
iv. Acid etch by sonicating with 0.5M HCL
v. Water rinse
vi. Dry by spinning at 3000RPM for 20 seconds

The samples were nine and were labelled 0 to 8, they were prepared by spin coating the polyaniline Nanocellulose composite on cleaned and dried glass substrates at 1000 rpm (static mode).

Characterization Techniques

Thickness Measurement

A profilometer (Dektak 150 from Veco instrument TMC U.S. A) was used in determining the profilometry (thickness) of the samples.

Optical Characterization

The optical investigation of the samples was carried out using a UV-VIS-NI spectrophotometer (UV 752), measuring wavelengths from 230 nm to 1100 nm.

Electrical characterization

The electrical properties of the polyaniline nanocomposite films were obtained using the Hall Effect measurement system (HMS-3000, Ecopia).

RESULTS

Table 1: Results of thickness measurement of the sample

SAMPLE NAME	THICKNESS IN ANGSTROM (Å)	THICKNESS IN NANOMETER (nm)
0	2000	200
1	8000	800
2	58000	5800
3	67000	6700
4	30000	3000
5	21000	2100
6	45000	4500
7	48000	4800
8	40000	4000

Results of Optical Characterization

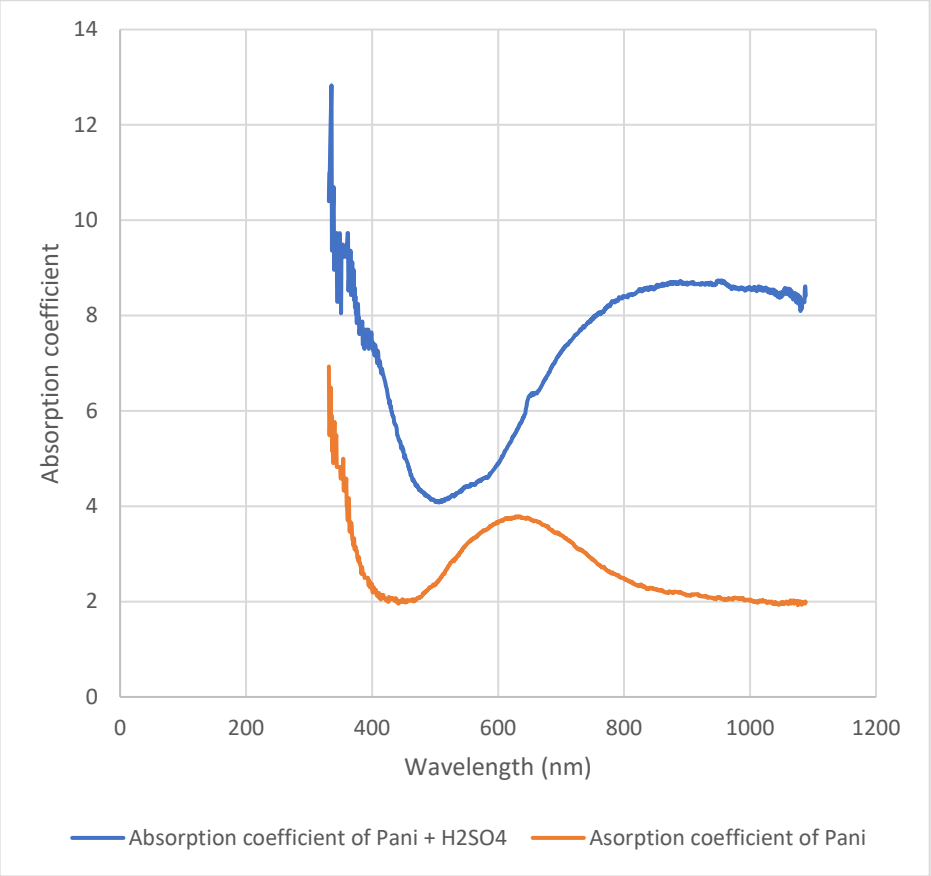

Figure 1: Uv-vis- Absorption coefficient spectra of the polyaniline nanocellulose and polyaniline nanocellulose treated with sulfuric acid (H_2SO_4).

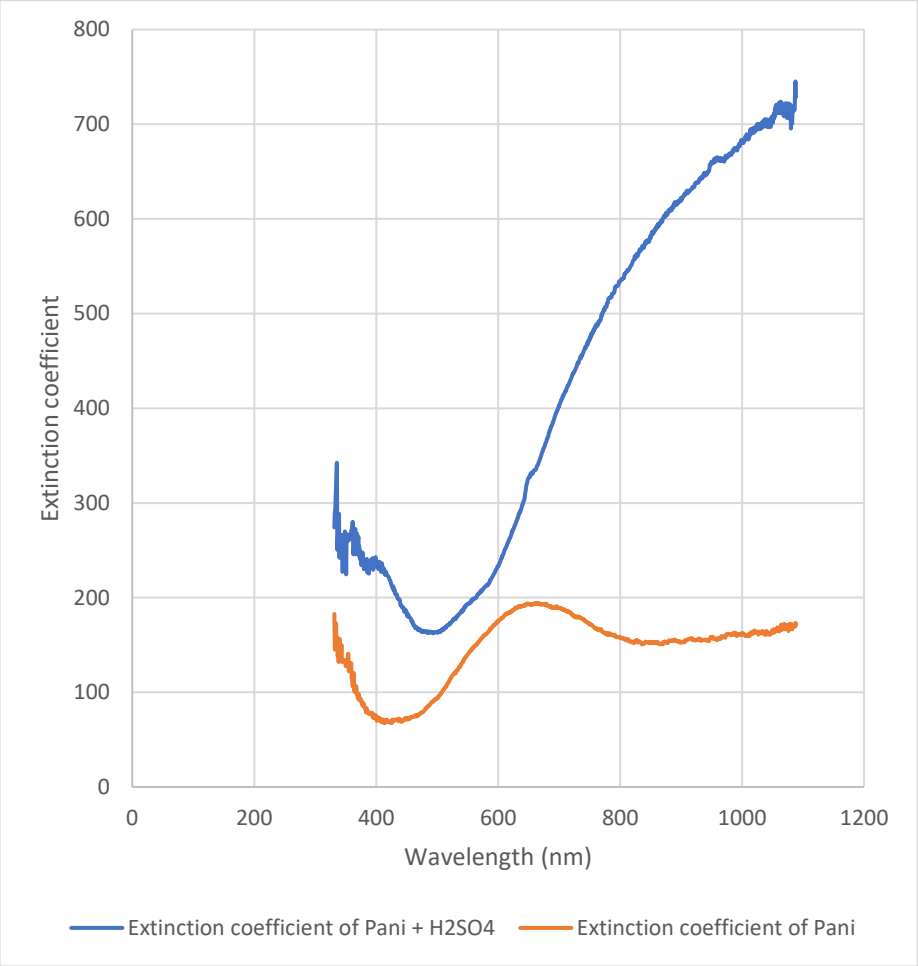

Figure 2: Uv-vis- Extinction coefficient spectra of the polyaniline nanocellulose and polyaniline nanocellulose treated with sulfuric acid (H_2SO_4).

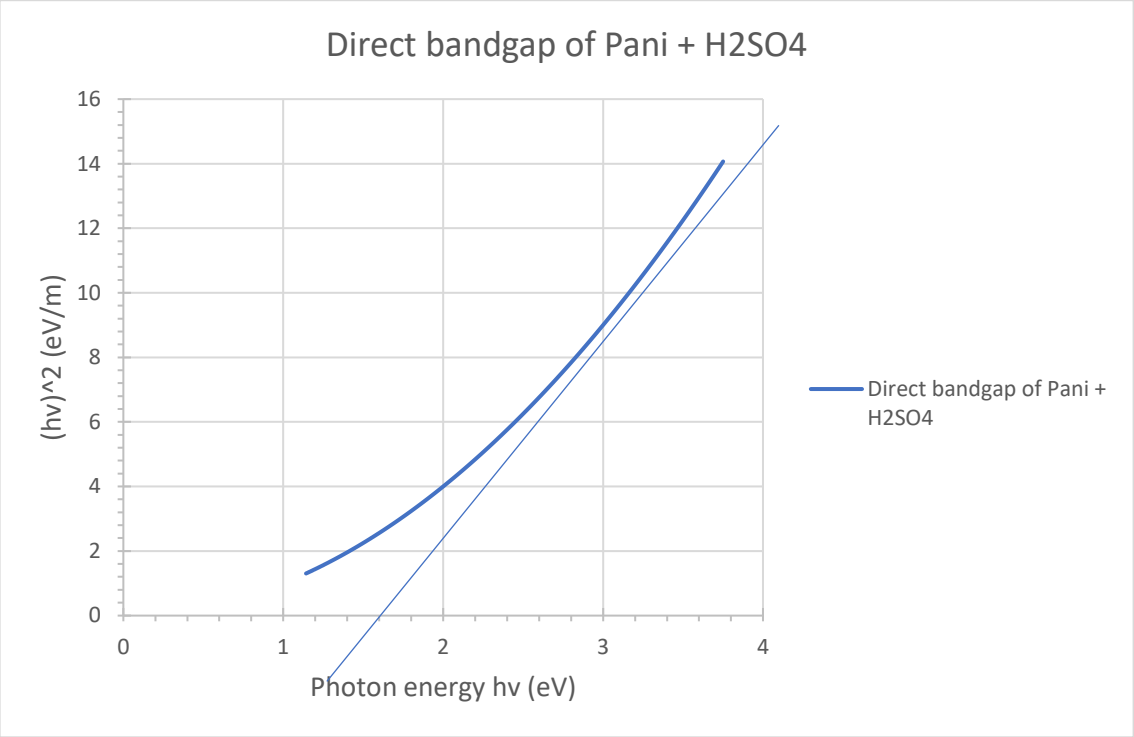

Figure 3: Uv-vis- optical bandgap of the polyaniline nanocellulose film treated with sulfuric acid (H_2SO_4).

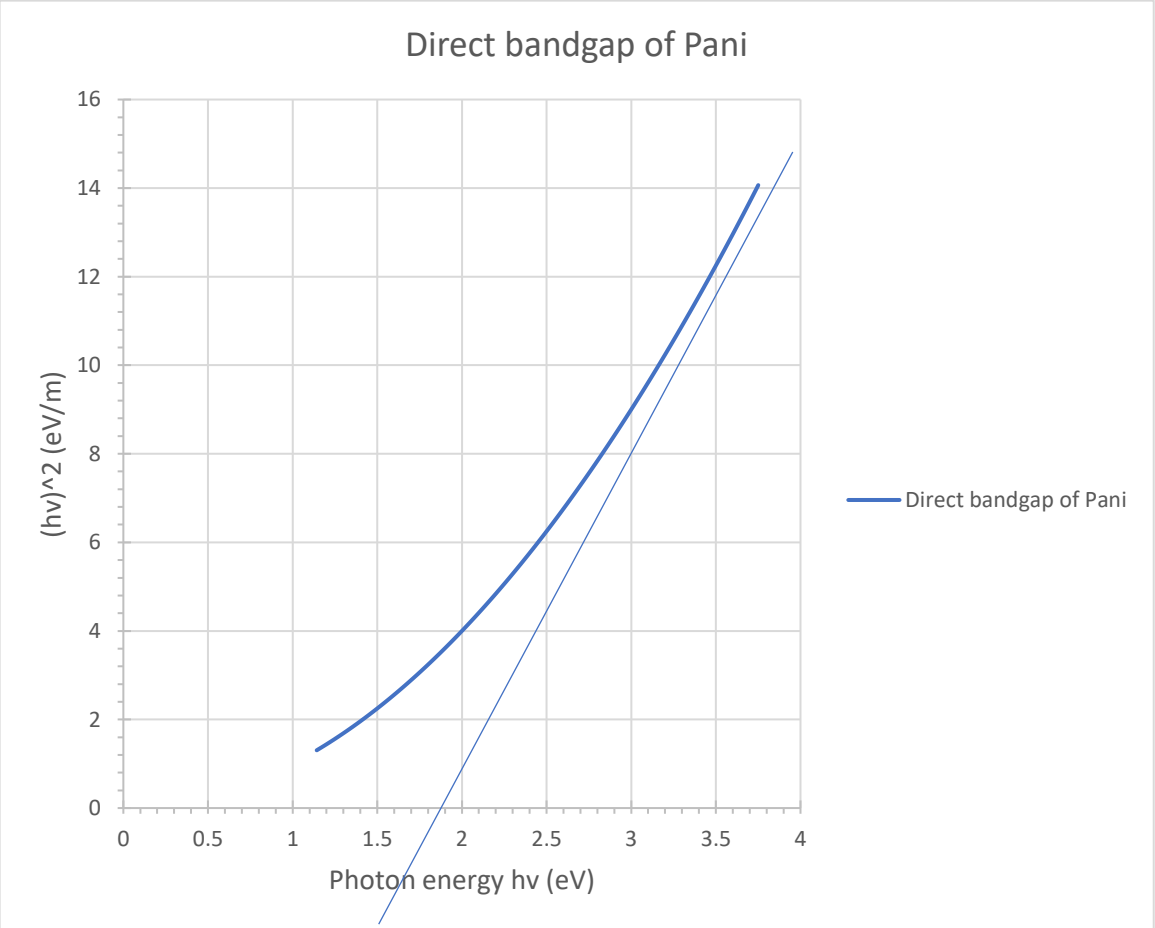

Figure 4: Uv-vis- optical bandgap of the polyaniline nanocellulose film

Results of the Electrical Characterization

Table 2: Result of the hall effect measurement

Sample name	Bulk concentration (/cm^3)	Mobility (cm^2/V$_5$)	Sheet Resistance (Ω)	Resistivity (Ωcm)	Sheet concentration (/cm^2)	Conductivity (1/ Ωcm)	Average Hall coefficient (cm^3/C)	B-D cross hall coefficient (cm^3/C)	Ratio of vertical/horizontal
Sample 0	-7.257E+11	2.045E+02	7.011E+08	4.207E+04	-4.354E+07	2.377E-05	-8.601E+06	2.260E+07	-1.153E-01
Sample 1	1.348E+17	3.369E+00	3.27E+04	1.375E+01	5.660E+13	7.273E-02	4.632E+01	4.848E+01	1.539E-03
Sample 2	7.217E+17	1.423E+01	1.013E+04	6.081E+00	4.330E+14	1.645E-01	8.650E+00	1.654E+0	9.254E-02
Sample 3	3.493E+17	6.573E+00	2.000E+04	3.200E+00	-7.107E+13	3.125E-01	-1.405E+01	-4.322E+01	4.355E-01
Sample 4	-4.442E+17	4.392E+00	2.000E+04	3.200E+00	-7.107E+13	3.125E-01	-1.405E+01	-4.322E+01	4.355E-01
Sample 5	3.117E+17	1.893E+10	2.402E+00	8.889E+00	1.373E+14	1.125E-01	1.682E+01	3.541E+01	2.340E-01
Sample 6	2.394E+15	2.538E+02	2.388E+04	1.027E+01	1.030E+12	9.737E-02	2.607E+3	5.331E+1	-2.841E+00
Sample 7	-8.272E+17	4.002E-01	4.962E+04	1.886E+01	-3.143E+14	5.303E-02	-7.546E+00	-2.202E+02	3.025E-01
Sample 8	-2.238E+17	1.120E+00	8.305E+04	2.492E+01	-6.713E+13	4.013E-02	-2.790E+01	-2.790E+01	3.646E-01

DISCUSSION

Discussion of the Profilometry (sample thickness) results.

The thickness of the polyaniline nanocellulose composite films was determined using a proilometer (Dektak 150 from veco instrument U.S.A). The surface thickness was measured in Angstrom and were converted to nanometer (nm) by dividing the length value by 10. Thus, the converted surface thickness values are as follows: sample 0 is 200nm, sample 1 is 800nm, sample 2 is 5800nm, sample 3 is 6700nm, sample 4 is 3000nm, sample 5 is 2100nm, sample 6 is 4500nm, sample 7 is 4800nm, and sample 8 is 4000nm. The obtained thickness values fall within the range reported by Funabiki et al. (2018) for samples 0 and 1, but are higher for the other samples. This indicates that as the film thickness increases, the carrier concentration tends to decrease. Moreover, the results suggest that films with optimized layering exhibit improved optical transparency, possibly due to enhanced crystallinity and reduced surface defects. These findings demonstrated that when film thickness grows, optical transmittance falls as a result of an increase in crystal size that thickens the layer. The distinct layers altered scattering cross-sections leading to a reduction in carrier concentration.

Discussion of the Optical Characterization Results

A UV-VIS-NIR spectrophotometer (UV 752), which covers a wavelength range of 230 nm to 1100 nm, was used to analyse the UV-VIS spectra of the polyaniline nanocellulose composite films. Several optical characteristics, such as absorbance coefficient, extinction coefficient, and optical band gap, are well covered by the acquired data, as shown in Figures 1 to 4. Films made of polyaniline nanocellulose and films treated with sulfuric acid were tested for these characteristics. The results showed that the polyaniline nanocellulose composite sheets greatly absorbed the visible range (380–700 nm). There was a minor decrease in absorption in the ultraviolet range and a slight increase in absorption in the infrared region. The composite films' light sensitivity in the visible and ultraviolet ranges and potential application as transparent conducting materials are indicated by these results, which is encouraging. In comparison to untreated films, polyaniline nanocellulose composite films performed better after being treated with sulphuric acid in terms of absorbance, transmittance, reflectance, refractive index, absorption coefficient, and extinction coefficient. The films treated with sulphuric acid exhibited extremely high absorbance and absorption coefficient values. A higher absorbance level indicates that the film is more effective at absorbing light, meaning it can capture more incident light. There was a 1.4eV and 1.7eV extrapolated direct optical band gap for the treated films, respectively. The findings demonstrate that the polyaniline nanocellulose composite films have favourable optical properties, such as elevated absorbance and absorption coefficient, especially following sulphuric acid treatment. Transparent conducting materials and other fields requiring efficient light absorption are possible application areas that could profit from this. Further applications in optoelectronics and other domains are made possible by the outstanding photo response in both the visible and ultraviolet spectrums.

Results of the Electrical Characterization

From the hall effect results in Table 2, it is observed that the electrical data of the polyaniline nano-cellulose composite films labeled sample 0 to sample 8 with different thicknesses measured at room temperature are summarized. All films were n-type semiconducting. As the film thickness was increased, the carrier concentration decreased and the mobility increased. The 6700 nm thick film had a resistivity of ~3.200 Ωcm, a carrier concentration of $\sim 3.493 \times 10^{17}$ cm^{-3}, and a mobility of ~6.573 cm^2 V^{-1} s^{-1}. According to the literature, PANI can be either n-type (with a carrier concentration of $\sim 1 \times 10^{14}$ to $\sim 7 \times 10^{19}$ cm^{-3} and a mobility of 2.78–310 cm^2 V^{-1} s^{-1}) or p-type (Daideche et al., 2020). The results are similar to those Lee et al., (2014) reported. This result roughly agrees with the fact that the energy difference between conductivity increases as the irradiance increases. It is also observed that the drift mobility of the samples increased with increasing thickness of the films.

CONCLUSION

In conclusion, this study successfully polyaniline nanocellulose composite films, demonstrating their promise for electronic applications. Nanocrystalline cellulose–polyaniline films were effectively prepared and analyzed using profilometry to examine surface morphology, UV–visible spectrophotometry to assess optical behavior, and Hall effect measurements to evaluate electrical properties. The results revealed key relationships between film structure, optical response, and electrical performance. Collectively, these findings enhance understanding of the composite's transport mechanisms and support its potential use in flexible and sustainable electronic devices, while providing a foundation for further optimization toward improved functionality and efficiency in advanced material applications.

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