Next Nanotechnology 9 (2026) 100414

Contents lists available at ScienceDirect

Next Nanotechnology

journal homepage: www.sciencedirect.com/journal/next-nanotechnology

Research article

Unveiling the Hall effect and electrical transport properties of chlorophyll-polypyrrole films deposited on ITO substrate for optoelectronic applications

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ARTICLE INFO

Keywords:
Hall effect
Conductivity
Chlorophyll
Polypyrrole
Magnetic flux density

ABSTRACT

This work presents, for the first time, the comprehensive characterization of the electrical properties and Hall effect response of a chlorophyll–polypyrrole composite thin film deposited on an ITO substrate. A markedly higher conductivity is achieved for the composite thin film compared with the pure ITO substrate, which demonstrates the significant role of the chlorophyll–polypyrrole matrix in enhancing charge carrier transport. The conductivity is observed to decrease systematically with increasing thickness of the deposited layer, a trend commonly attributed to enhanced scattering, increased defect density, and reduced carrier mobility in thicker films. The current–voltage (I–V) characteristic curve exhibits a linear electrical response, reflecting the intrinsic ohmic nature of the charge transport within the film, and thereby highlighting the strong potential of this hybrid material as a candidate for capacitor-based solar energy storage as well as for integration into optoelectronic devices. Moreover, both the Hall voltage and the charge carrier density exhibit a pronounced dependence on the applied magnetics flux density, underlining the sensitivity of the composite to external electromagnetic perturbations. Finally, the consistently observed negative polarity of the Hall voltage provides unambiguous evidence for electrons being the dominant charge carriers, thereby confirming the n-type semiconducting behavior of the composite thin film

1. Introduction

The Hall effect refers to the potential difference (Hall voltage) that develops across the opposite sides of an electrical conductor carrying an electric current when subjected to a magnetic field applied perpendicular to the current. This phenomenon arises from the deflection of charge carriers under the Lorentz force, which causes their accumulation on one side of the conductor, thereby generating a measurable voltage. In addition to the Hall voltage, Hall effect measurements are also widely employed to determine not only the sheet carrier density but also the mobility and type of charge carriers present in a conductor or semiconductor, providing crucial insights into the fundamental electrical transport properties of materials. Hall measurements are particularly versatile, as they enable the probing of charge transport phenomena of a composite. Moreover, Hall effect devices find extensive applications in motion sensing, current detection, and electrochemical switches, offering high sensitivity, long-term stability, and enhanced reliability even under extreme environmental conditions, such as high temperatures, strong magnetic fields, and harsh chemical environments [1–9]. Despite the extensive Hall measurements in inorganic semiconductors, reported Hall studies on organic–conducting polymer hybrid composite thin films, particularly those involving natural chromophores, remain scarce, representing a notable gap in current transport studies.

Polypyrrole is one of the most extensively studied conducting polymers since the monomer pyrrole is easily oxidized, water soluble, commercially available, and possesses environmental stability, good mechanical properties, and high electrical conductivity. Due to its good intrinsic properties, polypyrrole appears promising for use in batteries, supercapacitors, electrochemical biosensors, conductive textiles and fabrics, mechanical actuators, electromagnetic interference shielding, antistatic coatings, and drug delivery systems. Moreover, compared to

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https://doi.org/10.1016/j.nxnano.2026.100414
Received 21 October 2025; Received in revised form 9 February 2026; Accepted 27 February 2026

conventional metals, polypyrrole films provide distinct benefits such as corrosion resistance, flexibility, and low weight, rendering them highly suitable for advanced technological applications. Consequently, polypyrrole has been the focus of extensive research due to its exceptional stability and superior electrical conductivity relative to other conductive organic polymers, highlighting its versatility as a functional material for next-generation electronic and energy-related devices [10–13]. However, most reported studies on polypyrrole focus either on bulk films or electrochemically deposited layers, while its Hall transport behavior in composite thin films with other organic materials remains greatly unexplored.

Chlorophyll is structurally a substituted tetrapyrrole with a centrally coordinated magnesium atom. The primary role of chlorophyll molecules is to absorb incident light and efficiently transfer the energy via resonance to a specific chlorophyll pair located in the reaction center of the photosystem. Previous studies have demonstrated that chlorophyll pigments possess a key property essential for high-performance photovoltaic applications, namely strong absorption within the visible spectral region and an activation energy between 1.0 and 2.0 eV. This energy range corresponds to conditions under which the theoretical efficiencies of photovoltaic devices are maximized, emphasizing the suitability of chlorophyll as a natural and light-harvesting biomaterial for solar energy conversion and optoelectronic devices [14–18]. Despite these optoelectronic properties, chlorophyll has rarely been investigated in conjunction with other conducting polymers for electrical transport and Hall effect studies, particularly in composite thin films where charge transfer may dominate the carrier dynamics. When incorporated into composite thin films, chlorophyll molecules might actively participate in charge generation and transfer throughout the surface, influencing the electronic properties. In addition, the inherent photostability and biocompatibility of chlorophyll further enhance its potential for integration into environmentally friendly and sustainable device architectures.

Thin films are material layers with thicknesses ranging from fractions of a nanometer to several micrometers. Electronic semiconductor devices and optical coatings represent primary applications that benefit from such thin constructions. Furthermore, thin film deposition is a well-established technique for fabricating uniform layers on substrates, providing precise control over thickness and material properties. Composite thin films, formed by combining two or more distinct materials, often exhibit enhanced functionalities compared to single-component films, rendering them highly attractive for a wide range of technological applications, including optical, electronic, and energy storage devices. In a composite system, surface and interface interactions play a decisive role in governing charge transport, carrier scattering, and overall electrical performance [19–29]. Nevertheless, a clear understanding of how film thickness influences Hall parameters, carrier type, and mobility in bio-organic composite thin films is still lacking, particularly in the ultrathin regime relevant to practical devices.

In this study, we report, for the first time, the electrical properties and Hall effect of a chlorophyll–polypyrrole composite thin film deposited on ITO substrates. The chlorophyll–polypyrrole blend was chosen to integrate the strong optical absorption capability of chlorophyll with the high conductivity of polypyrrole, thereby forming a synergistic hybrid composite for charge carrier transport studies that have not been clearly addressed before in previous works. The novelty of this work lies not only in the Hall effect measurements on the hybrid films, but also in the systematic control of film thickness within the targeted regime. Two desired thicknesses, corresponding to 5-layer and 15-layer coating films, were selected to study the transition from relatively thin to thicker films, where the bulk Hall effect becomes dominant. Furthermore, this work offers new perspectives into charge transport, Hall effect, carrier type contribution, and Hall mobility arising from the synergistic coupling between chromophores and conducting polypyrrole, which introduces for the first time the relation between electronic transport and Hall parameters governed by a chlorophyll–polypyrrole hybrid system, with implications for future optoelectronic and solar energy storage applications.

2. Materials and method

2.1. Sample preparation

The indium tin oxide (ITO)-coated glass substrates (7.5 cm × 2.5 cm) were subjected to ultrasonic cleaning with acetone and ethanol, followed by rinsing with deionized water to ensure the complete removal of residual contaminants and organic impurities from the surface. This multistep cleaning process is crucial for enhancing the surface energy of the substrate, thereby promoting better adhesion of the deposited film. After cleaning, the substrates were dried under a stream of compressed air and subsequently stored in a desiccator prior to deposition to prevent unwanted surface contamination. The desiccation step is particularly important to eliminate any residual moisture that could interfere with the uniform coating and alter the film's electrical properties.

A chlorophyll solution (3 ml, 1.56×10^{-4} g/L) was then prepared and mixed with 20 ml of polypyrrole solution in an ultrasonic bath to achieve uniform blending. The use of ultrasonic agitation ensures proper molecular dispersion of chlorophyll within the polypyrrole matrix, preventing aggregation and ensuring a homogeneous composite solution. This uniformity is essential for achieving consistent film morphology and reliable electrical behavior in the final deposited layers. The deposition of the chlorophyll–polypyrrole thin films was carried out using a spin-coating unit (model WS-400B-6NPP/LITE), with the gas flow pressure maintained at 1500 atm. The spin-coating technique allows precise control of film thickness, surface uniformity, and deposition rate by adjusting the rotational speed and gas pressure parameters.

To investigate the influence of film thickness, two distinct deposition conditions were employed, corresponding to 5 and 15 layers of the blend film on the ITO substrates. Each layer was deposited sequentially, followed by a short drying step to remove residual solvent and ensure proper adhesion between successive coatings. This layer-by-layer approach provides better control over the film's structural buildup and ensures reproducibility of thickness across all samples. By varying the number of layers, the effect of thickness on charge transport, conductivity, and surface morphology could be systematically analyzed, thereby establishing a direct correlation between deposition parameters and the resulting electronic performance of the chlorophyll–polypyrrole composite films.

2.2. Methods of characterization

The electrical conductivity of the deposited films was measured using a four-point probe system (model Changmin Tech CMT-SR2000N), ensuring high precision in resistance and conductivity evaluation. Film thicknesses were determined through scanning electron microscopy (SEM, model JEOL JSM-6360LA), which provided accurate morphological and cross-sectional measurements. Furthermore, Hall effect characterization was performed using a Hall effect measurement system (7600 series, Lake Shore Inc.) to evaluate both the magnetic flux density and the carrier mobility of the thin films. This technique provides comprehensive information on charge carrier type, density, and transport efficiency under controlled magnetic field conditions. Data acquisition and analysis were conducted using the Leois-JSF software, enabling reliable extraction of electronic transport parameters and ensuring comprehensive assessment of the charge carrier dynamics within the chlorophyll–polypyrrole composite thin films. The integration of these advanced measurement systems ensures high accuracy, reproducibility, and detailed understanding of the electrical and electronic behavior of the samples, making it possible to correlate the microstructural characteristics with their transport properties for further optimization in electronic and optoelectronic applications.

3. Results and discussion

3.1. Electrical conductivity

The values of electrical conductivity and average film thickness for different numbers of layers of polypyrrole and chlorophyll composite blend thin films deposited on ITO substrates are presented in Table 1. From the results, it is worth noting that higher conductivity is obtained for the composite thin films deposited on the ITO substrate compared to the bare ITO. This enhancement is attributed to the intrinsic conducting properties of polypyrrole, which effectively improve the overall electrical conductivity of the substrate. However, the conductivity exhibits a slight decrease with an increasing number of deposited layers on the ITO glass substrate. This characteristic can be explained by the fact that a higher proportion of chlorophyll conjugated with polypyrrole introduces a comparatively insulating effect, which reduces the charge carrier transport efficiency across the sample surface. Moreover, as the number of layers increases, the films become thicker, which may lead to enhanced interfacial scattering at polymer–chlorophyll interfaces and reduced charge carrier mobility, thereby further suppressing the measured conductivity. The aggregation of chlorophyll molecules within the composite may also disrupt the conjugated pathways of polypyrrole, further hindering electron delocalization and charge transfer. Consequently, the optimal electrical performance is obtained at a lower number of layers, where the balance between conductivity and the uniformity of the composite structure is well maintained. The present study involves the deposition of an organic–conductive polymer hybrid composite layer on the ITO substrate, where charge transport is predominantly governed by the composite thin film rather than the ITO substrate. The reduced conductivity as the deposited layer increases can also be attributed to the intrinsically lower carrier mobility of conductive polymer and organic components compared to inorganic oxide films. Furthermore, since four-point probe measurements primarily probe lateral charge transport within the composite film, the presence of localized states and interfacial barriers might impede carrier motion, leading to lower measured conductivity values. From the literature, temperature-dependent I–V characteristics of chlorophyll–polypyrrole films on ITO are expected to exhibit thermally activated charge transport, with ohmic behavior at low bias and trap-assisted or space-charge-limited conduction at higher bias. The presence of chlorophyll introduces localized states that influence carrier hopping and interfacial transport, as widely reported for conjugated polymer–based hybrid films (references).

Furthermore, SEM analysis confirms that the film thickness increases proportionally with the number of layers, demonstrating the consistency and accuracy of the deposition procedure, as illustrated in Fig. 1. The uniform growth pattern observed across the film surface also indicates a good adhesion between the successive layers, which means that the blending of polypyrrole and chlorophyll results in a stable and continuous film structure. This uniform deposition ensures minimal surface defects and promotes efficient charge transport pathways within the composite layer. Fig. 2 illustrates the current-voltage (I-V) characteristic curves of the samples. From the figure, the voltage shows a linear increase with increasing current, indicating a sensitive electrical response and good ohmic contact of the composite films. The linearity of the I-V curves further confirms that the charge carriers experience minimal potential barriers at the film–electrode interfaces, thereby ensuring efficient charge injection and extraction processes. These results suggest that the present materials are well-suited for applications in capacitors and inductor-based solar devices, where stable and predictable electrical behavior is essential. Additionally, the observed electrical stability and the repeatable responses denote that the polypyrrole–chlorophyll composite thin films have excellent interfacial compatibility and structural integrity, which are important factors for long-term device performance and reliability in energy storage and photoconversion systems.

Table 1
Values of Hall conductivity and average film thickness for different number of layers of polypyrrole and chlorophyll blend thin film deposited on ITO substrate.

Number of Layer	Conductivity $(\Omega m)^{-1}$	Average film thickness (μm)
0	0.150	0
5	0.158	14
15	0.155	98

3.2. Hall effect

Fig. 3 shows the Hall voltage as a function of the magnetic flux density for the samples deposited with 5 layers and 15 layers of the polypyrrole-chlorophyll blend film. The Hall voltage values obtained range from -1.51×10^{-3} to -16.9×10^{-3} mV. From the figure, it can be clearly observed that the Hall voltage increases linearly with the magnetic flux density when a constant current is applied during the measurement, demonstrating a stable and predictable Hall response. This linear relationship indicates that the charge carriers within the composite film move in a uniform and controlled manner under the influence of the applied magnetic field, which means a strong indication of good electrical homogeneity and material quality. Furthermore, a negligible intercept at zero magnetic field confirms that offset voltages arising from contact misalignment or thermoelectric effects are minimal. This indicates that the Hall measurements were performed steadily under a constant current configuration, and the Hall voltage was able to be extracted from the linear dependence of the transverse voltage on the applied magnetic flux density. Similarly, it can be inferred that the Hall voltage is directly proportional to the applied current at a fixed magnetic flux density, which reflects the intrinsic charge transport behaviour of the films. This linear proportionality also confirms that the conduction mechanism in the composite is primarily dominated by free charge carriers rather than trap-assisted hopping, which implies an efficient transport network formed through the polypyrrole matrix.

In addition, the Hall voltage of the 5-layer deposited film is higher than that of the 15-layer deposited film, indicating that the thinner film possesses lower resistivity (higher conductivity) and a larger sheet carrier density. This observation suggests that as the film thickness increases, the charge carrier pathways become more tortuous, and additional interfaces are introduced, which could scatter or trap carriers, thereby decreasing the effective mobility. Furthermore, the increased presence of chlorophyll molecules in thicker films may contribute to localized states that act as recombination centers, further reducing the overall charge carrier density. The corresponding values for sheet carrier density are summarized in Table 2, which provide quantitative insight into the effect of film thickness on the electronic properties of the composite films and their potential implications for electronic and optoelectronic applications. These results collectively demonstrate that optimizing the number of deposited layers is crucial for achieving desirable electrical characteristics, particularly in applications where higher carrier mobility and conductivity are essential for device performance and energy efficiency.

In addition, the type of charge carrier can also be determined from the Hall voltage polarity. For instance, a positive Hall voltage polarity indicates that the majority charge carriers are positive holes, classifying the material as a p-type semiconductor. In the present study, however, the Hall voltage for all samples exhibits a negative polarity, thus confirming that electrons are the majority charge carriers. Therefore, the materials contain a higher concentration of electrons than holes and thus can be classified as n-type semiconductors. This behaviour can be due to the dominance of the polypyrrole component that facilitates electron conduction through its conjugated π-electron system. Conducting polymers such as polypyrrole are known to exhibit high apparent carrier densities under Hall measurements due to the presence of doped states, which contribute to charge transport through hopping

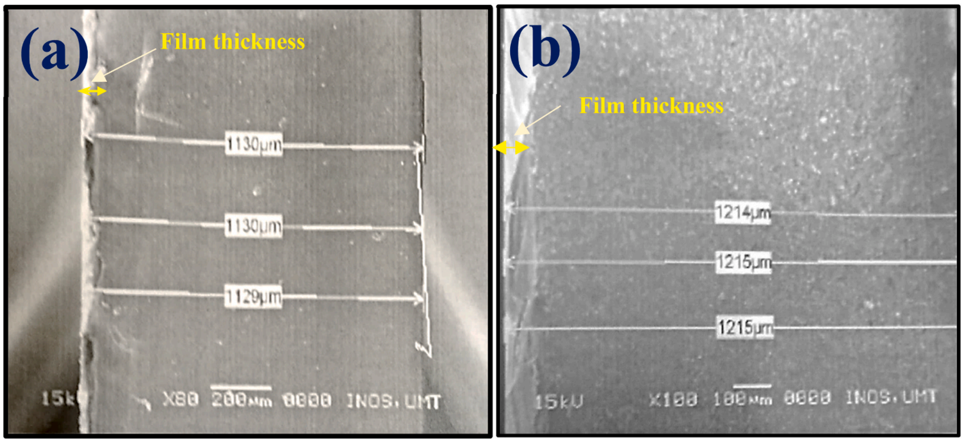

Fig. 1. SEM images for the film thickness of (a) 5-layer (b) 15-layer deposition.

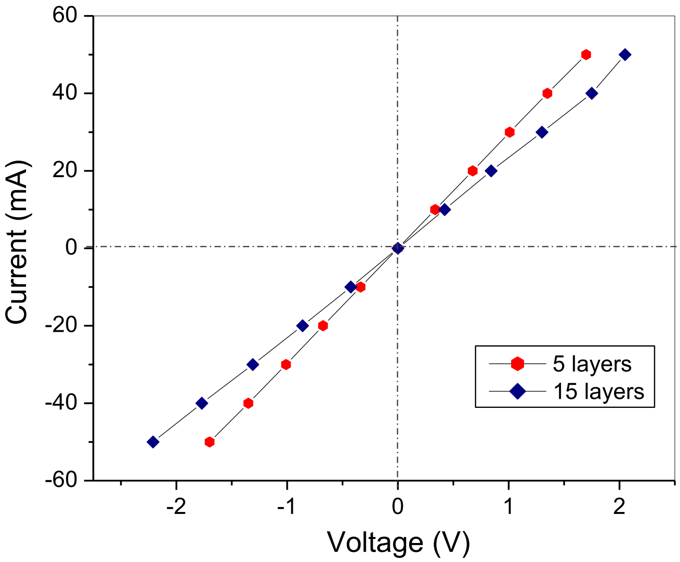

Fig. 2. I-V characteristics for the samples deposited with 5 layers and 15 layers of polypyrrole and chlorophyll blend film.

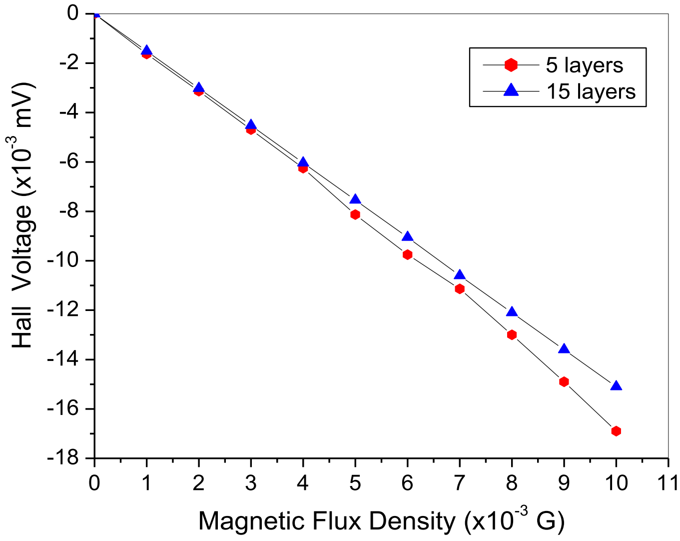

Fig. 3. Hall voltage versus the magnetic flux density for the samples deposited with 5 layers and 15 layers of polypyrrole and chlorophyll blend film.

Table 2
Values of sheet carrier density for different number of layers of polypyrrole and chlorophyll blend thin film deposited on ITO substrate.

Magnetic flux density (G)	Sheet carrier density ($\times 10^{16}$ cm^{-2})	
	5 layers	15 layers
0.001	2.0622	1.9391
0.002	2.0671	1.9951
0.003	2.0679	2.0048
0.004	2.0685	1.9885
0.005	2.0689	1.9552
0.006	2.0690	1.9854
0.007	2.0688	1.9839
0.008	2.0693	1.9288
0.009	2.0690	1.9079
0.010	2.0709	1.8786

and delocalized states rather than conventional band conduction. These charge carriers are often confined to interconnected conductive pathways, leading to elevated sheet carrier densities without implying the actual bulk carrier densities comparable to those of an inorganic semiconductor [3–5]. The presence of chlorophyll in the composite may contribute to electron delocalization through its porphyrin ring structure, thereby enhancing the n-type character by allowing efficient electron transfer across the polymeric network. Moreover, the formation of interfacial junctions between the conducting polypyrrole chains and the chlorophyll molecules might be creating potential wells that facilitate electron accumulation, further reinforcing the n-type behaviour observed in all the studied samples. Particularly, the incorporation of chlorophyll and the interaction with the polypyrrole and ITO substrate can induce charge carrier transfer that favors electron transport. Such interfacial charge redistribution has been previously reported in conductive polymer–natural organic composite thin films deposited on conductive metallic oxide substrates [3,4,12]. Therefore, the observed n-type Hall response reflects effective charge transport in the composite film as a whole integrated system, rather than the intrinsic conduction of individual constituents. Henceforward, the proposed mechanism for carrier dynamics of the present samples is as follows: Upon Hall-electrical biasing, electrons are transported within the polypyrrole matrix through interconnected π-conjugated pathways and hopping between doped states. Chlorophyll molecules embedded within the polymer act as electron delocalization and trapping centers via their porphyrin ring structures, which enable charge redistribution and localized electron accumulation at polypyrrole–chlorophyll interfaces. This interfacial region facilitates the formation of energetically favorable electron transport pathways, resulting in a continuous electron percolation transfer across the composite film. Consequently, electron-dominated transport prevails and gives rise to the measured negative Hall voltage along with the observed n-type Hall response [29–33].

Hall mobility refers to the ability of electrons to move across a semiconductor material under a constant applied field [2]. The Hall mobility (μ_H) is given as follows:

$$\mu_H = \sigma/ne \quad (1)$$

where σ is the electrical conductivity extracted from the I–V characteristics and n is the carrier concentration obtained from Hall measurements. This relation highlights that Hall mobility depends on the ratio of conductivity to carrier density rather than on their absolute values. The variation of Hall mobility with magnetic flux density is presented in Fig. 4. Interestingly, in the present case, all samples exhibit a nearly constant Hall mobility value of ~3.75×10^{-3} $m^2 V^{-1} s^{-1}$ over the investigated magnetic flux density range, with only marginal variation across measurements, regardless of film thickness. This apparent constancy occurs due to the measurements were performed in the linear Hall measurement regime, where the applied magnetic field does not significantly perturb carrier trajectories and hopping pathways in disordered organic systems [5–7]. Within this regime, the extracted Hall mobility corresponds to an effective mobility averaged over the dominant percolative conduction pathways, for which the influence of disorder or morphological variations is expected to be minimal. Even though the 5-layer and 15-layer films differ in thickness, the increase in carrier concentration with layer number is accompanied by a compensating reduction in conductivity due to enhanced interlayer resistance. As a result, the ratio σ/n remains approximately constant, leading to close apparent Hall mobility values for both films. Thickness-dependent effects are therefore more prominently manifested in carrier concentration and electrical conductivity rather than in Hall mobility. Hence, it is worth to note that the observed field independence of Hall mobility does not imply the absence of disorder-induced scattering or localization effects. Rather, it reflects the specific magnetic-field conditions employed in this study. Furthermore, because the Hall measurement probes lateral charge transport, the conduction pathways and contact geometry are comparable for both samples, further contributing to the observed similarity in effective Hall mobility. Accordingly, the reported values should be interpreted as effective Hall mobilities measured under linear Hall conditions, confirming stable transport behaviour trough the composite film [32–35].

4. Conclusion

In summary, the electrical properties and Hall effect of the polypyrrole and chlorophyll composite blend thin film deposited on ITO substrates were systematically characterized in this study. The electrical conductivity was observed to range from 0.150 to 0.155 $(\Omega m)^{-1}$ and decreased with increasing film thickness, highlighting the influence of layer deposition on charge transport. Higher Hall voltage values were recorded for thinner films, indicating enhanced conductivity and greater sheet carrier density, consistent with the Hall measurement analysis. The linear behavior observed in the I-V characteristics further underscores the ohmic nature of the films and points to their potential utility in solar energy storage systems and advanced optoelectronic devices. Overall, the results provide critical perspective into the relation between Hall effect, charge carrier dynamics, and electrical transport performance, reinforcing the promise of chlorophyll-polypyrrole composite films for next-generation electronic and energy conversion applications. Future work will focus on optimizing film thickness and chlorophyll content to improve charge transport and electrical stability. Integration of the composite films into prototype photovoltaic, superconductor and energy storage devices, along with long term stability evaluation, will be pursued for practical applications.

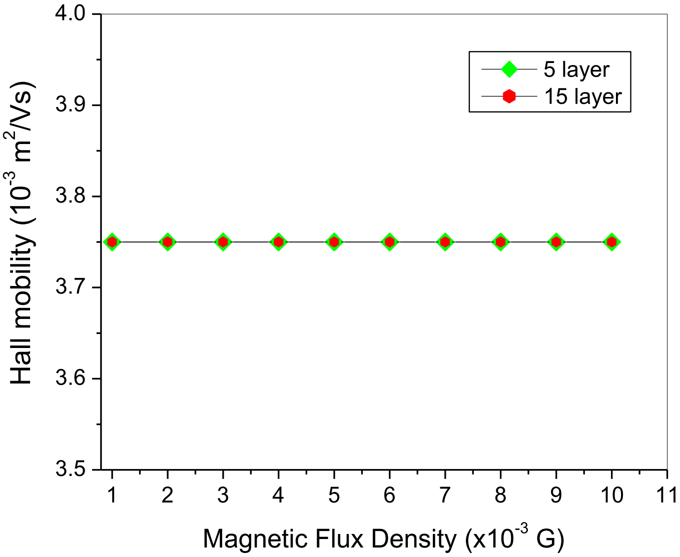

Fig. 4. Hall mobility versus magnetic flux density for the samples deposited with 5 layers and 15 layers of polypyrrole and chlorophyll blend film.

CRediT authorship contribution statement

Chan Kok Sheng: Writing – review & editing, Visualization, Validation, Supervision, Software, Formal analysis, Data curation. **Yap Lai Yoon:** Writing – original draft, Resources, Project administration, Methodology, Investigation, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors would like to thank Universiti Malaysia Terengganu for providing the research grant trough (UMT/SRG-IPHD/2025/55610) and Universiti Putra Malaysia for the technical assistance in this work.

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