

## PREPARATION OF CONDUCTIVE MEMBRANES BY CHEMICAL *IN SITU* POLYMERIZATION OF POLYPYRROLE ON CELLULOSE FIBERS

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**Abstract.** The development of flexible electronic devices, particularly flexible and bendable energy storage devices, has catalyzed significant interest in the research of flexible composite materials. In this study, conductive paper membranes were synthesized by polymerizing polypyrrole (PPy) on the surface of cellulose fibers. The cellulose material, derived from cardboard, underscores an eco-friendly approach to waste reduction and environmental protection. Characterization of the cellulose/PPy conductive membranes using Fourier-transform infrared spectroscopy (FT-IR), field-emission scanning electron microscopy (FE-SEM), and differential scanning calorimetry (DSC) confirmed the formation and uniform coverage of PPy on the cellulose surface. To enhance the uniformity of PPy polymerization on cellulose relative to previous studies, this work focuses on elucidating the formation and deposition of PPy particles on cellulose fibers, leading to the development of a homogeneous membrane. The membrane exhibited a peak electrical conductivity of 18.04 mS/cm at 0.1 mA, with conductivity increasing alongside PPy concentration, albeit at the expense of mechanical properties. Additionally, the membrane demonstrated charge storage capability, with specific capacitance values ranging from 22.5 to 50 pF/cm<sup>2</sup> at a frequency of 1 kHz. The uniformity of PPy coverage on the cellulose surface was a crucial factor influencing the electrical properties of the composite membrane. This research highlights the significant potential of conductive membranes for application in flexible and bendable energy storage devices.

**Keywords:** cellulose fiber, flexible, polymerization, polypyrrole.

### 1. Introduction

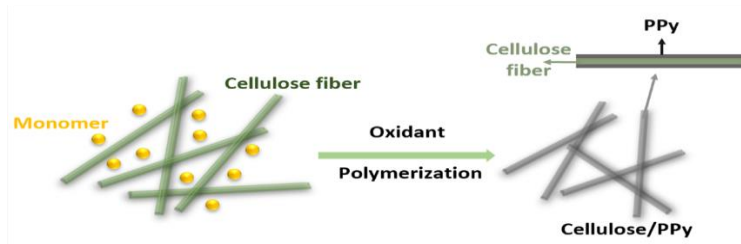
In recent years, significant research has focused on green materials, emphasizing the use of biodegradable and environmentally friendly polymers for energy storage devices [1]-[3]. These polymers are sustainable, recyclable, flexible, lightweight, and capable of easily interacting with other materials to form composites with exceptional properties [4], [5].

Among these materials, cellulose is particularly valued for its suitability as a matrix material in applications involving conductive polymers. Cellulose, a natural organic polymer, is abundant in nature, primarily found in plant cell walls, and can be extracted from various sources [6]-[8]. It possesses unique characteristics, such as low weight, low density, and a high elastic modulus, and contains surface hydroxyl groups that allow for diverse functionalization to achieve different surface properties, facilitating good interaction with other materials [9].

The structure of cellulose encompasses fibrous, crystalline, and amorphous forms. Cellulose fibers (CF) create a porous, multi-channel structure ideal for absorbing and transporting water and essential ions across both the outer and inner surfaces of the fibers. [10]. Due to its film-forming ability and fibrous properties, CF can enhance the mechanical stability of composite materials [11]. This makes cellulose an attractive component in the development of advanced materials for energy storage applications, combining environmental sustainability with functional versatility.

Cellulose fibers (CF) are inherently non-conductive. For electronic applications, they must be doped with metals or semiconductors or combined with conductive polymers to form composite materials. The combination of cellulose with polymers is considered highly compatible, with polypyrrole (PPy) being a prototypical conductive polymer due to its large surface area, good hydrophobic properties, excellent oxidation resistance, and notable electrical conductivity [12], alongside a simple and rapid synthesis process [13]. Dispersing cellulose in PPy is challenging because both materials are insoluble in water and common solvents. Therefore, *in situ* polymerization is the most prevalent method for depositing PPy onto cellulose fibers to synthesize cellulose/PPy composite.

The polymerization process employs ferric chloride ( $\text{FeCl}_3$ ) as a catalyst, which oxidizes pyrrole into pyrrole radical cations. These cations subsequently combine through double bonds to form PPy chains. During their formation, the PPy chains directly coat the surface of the cellulose fibers, resulting in a composite material with enhanced conductive properties (Figure 1) [14].



**Figure 1. The mechanism of PPy polymerization on cellulose fibers [14]**

Composite materials exhibit various morphologies that significantly influence their properties, including flexibility, elasticity, and electrical conductivity. One key factor affecting the capacitance of supercapacitors is the thickness of the electrodes and the electrolyte layer [15]. Therefore, utilizing a membrane fabrication method may be an appropriate approach to create supercapacitors with high energy storage capacity, durability, and flexibility. In this study, we synthesized a conductive polymer membrane

material based on cellulose fibers recovered from cardboard, combined with a conductive polymer, using polymerization and blending methods.

Previous studies have investigated the synthesis of PPy conductive polymer and cellulose composites via polymerization reactions, yielding materials with good electrical conductivity [14], [16], [17]. However, these studies have not demonstrated the morphology of PPy coverage on the cellulose surface, which is crucial for the material's conductive properties. Furthermore, the use of cardboard-derived cellulose in this study is of environmental significance, as it utilizes readily available materials and addresses the need for recycling and waste reduction.

The resulting composite membrane is electrically conductive, has a smooth surface, high tensile strength, and notably, the ability to be flexible and bend without compromising its properties. The properties of the membrane were characterized using various techniques to investigate its morphological characteristics, material properties, and particularly the uniformity of PPy particle coverage on the cellulose fiber surface.

## 2. Content

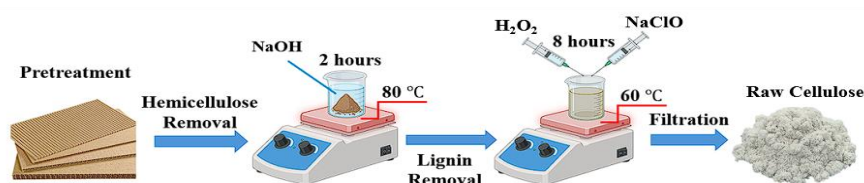
### 2.1. Experimental methods

#### 2.1.1. Materials

Cellulose was extracted from commercially utilized cardboard, commonly used in daily life. The composition of the cardboard includes approximately 60-70% cellulose, 15-25% hemicellulose, and 10-15% lignin. Sodium hydroxide (NaOH, 98%), Sodium hypochlorite (NaClO, 8%), Hydrogen peroxide ( $H_2O_2$ , 30%), Sulfuric acid ( $H_2SO_4$ , 98%), Iron(III) chloride hexahydrate ( $FeCl_3 \cdot 6H_2O$ ) were sourced from Xilong Chemical Co. Ltd, China. Pyrrole ( $C_4H_5N$ , 99%) was sourced from Shanghai Macklin Co. Ltd, China. DI water was produced using an FST-UV water distillation system at the University of Engineering and Technology, Vietnam National University, Hanoi.

#### 2.1.2. Methods

##### \* Extraction of raw cellulose from waste papers

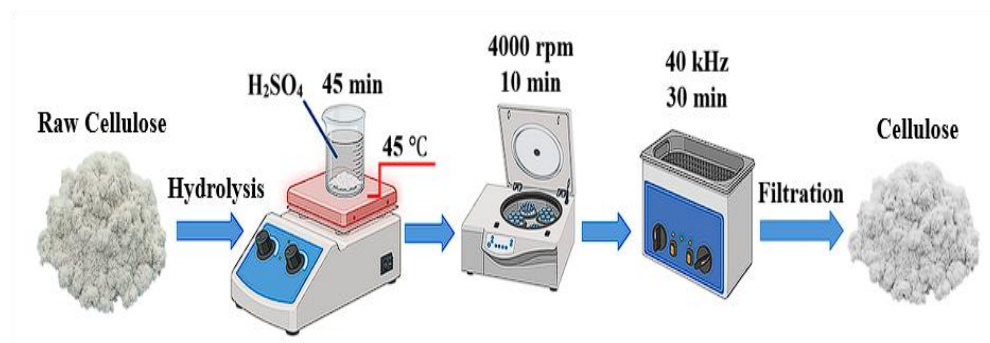


**Figure 2. Cellulose synthesis process from waste paper**

The experimental process for cellulose extraction was based on the methods outlined by Patchiya Phanthong *et al.* [9]. Various types of cardboard were used as raw materials. During the pretreatment process, the cardboard was cut into small pieces, and a 2% wt NaOH alkaline solution was employed to remove hemicellulose at 80 °C for 2 hours. The alkaline process involved the hydrolysis of cellulose to enhance impurity removal and improve the cellulose structure. The bleaching process used a mixture of NaClO and  $H_2O_2$  to remove residual lignin, wax, and lipids at 60 °C for 8 hours. The final mixture was filtered through a vacuum filtration funnel and dried to obtain raw cellulose (Figure 2).

**\* Processing cellulose from raw cellulose**

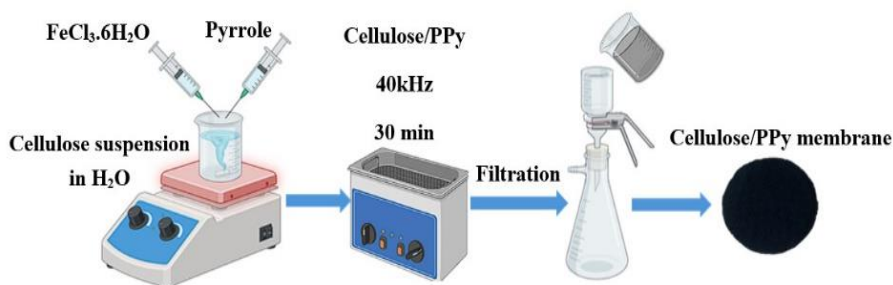
Following the pretreatment process, the extracted raw cellulose material had most of the hemicellulose and lignin removed from the cardboard; however, the sample still contained several impurities, and the fiber structure remained coarse. The hydrolysis of cellulose using a strong acid serves to reduce the size of the cellulose fibers, remove residual impurities, and improve the fibrous structure post-extraction. Accordingly, cellulose was hydrolyzed with an acid at a ratio of 1 g of raw cellulose powder to 9 ml of  $\text{H}_2\text{SO}_4$  solution (60%) for 45 minutes at 45 °C. The hydrolysis process was terminated by adding distilled water to the mixture and centrifuging for 10 minutes at 4000 rpm to remove surface water. Subsequently, the cellulose was ultrasonicated using an ultrasonic vibrator (DR-MS40, frequency 40 kHz) for 30 minutes to evenly disperse the cellulose fibers. A NaOH (2 M) solution was employed to neutralize the mixture. The cellulose suspension was then filtered through a vacuum filtration funnel and dried to obtain pure cellulose fiber powder (Figure 3).



**Figure 3. Cellulose treatment process employing acid hydrolysis**

**\* Preparation of conductive membrane based on cellulose/PPy composite**

The synthesis process of the conductive cellulose/PPy membrane is depicted in the schematic diagram shown in Figure 4. Initially, the hydrolyzed cellulose powder was added to 200 ml of distilled water and stirred thoroughly. An aqueous stock solution of  $\text{FeCl}_3$  was prepared at a concentration of 3.24 mM. To investigate the effect of PPy on the properties of the conductive polymer membranes, the polymerization process was performed by simultaneously adding a mixture of pyrrole and pyrrole/ $\text{FeCl}_3$  solutions to the cellulose suspension. The volume ratios of pyrrole to  $\text{FeCl}_3$  aqueous solution were varied (0.1/20, 0.2/20, 0.3/20) to assess the impact of different concentrations. This mixture was continuously stirred at 1000 rpm for 90 minutes at room temperature to ensure thorough incorporation and polymerization of the pyrrole onto the cellulose fibers. The mixture was then subjected to ultrasonication for 30 minutes to achieve uniform dispersion of the cellulose/PPy in the solution. Subsequently, the mixture was subjected to multiple filtration cycles using a mixed cellulose ester membrane with a pore size of 0.22  $\mu\text{m}$ . This process, conducted with distilled water and employing a vacuum filtration funnel, was performed to ensure the complete removal of inorganic components. Upon drying, the conductive cellulose/PPy membrane was obtained.



**Figure 4.** Polymerization process for synthesizing conductive cellulose/PPy membrane

### 2.1.3. Characterization methods

#### \* Sheet resistance measurements

The sheet resistance of the conductive cellulose/PPy membrane samples was systematically examined with varying PPy concentrations. The samples were analyzed using the Jandel CYL-RM300 Four-Point Probe Bridge, with measurements conducted at currents ranging from 0.1 to 0.9 mA.

#### \* Membrane thickness

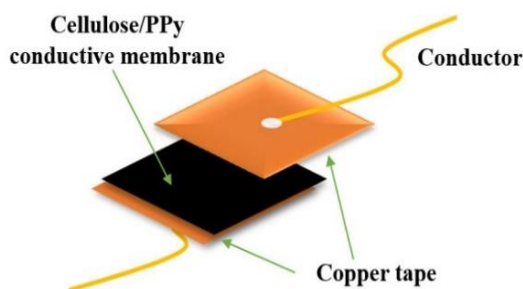
The thickness of the cellulose/PPy film samples was measured using a Mitutoyo High-Accuracy Series 293-831-30 electronic micrometer. For each film, thickness measurements were taken at 10 different points, and the average thickness was determined based on these measurements.

#### \* Tensile strength measurement

The tensile strength of cellulose and cellulose/PPy film samples was evaluated using a Tensile Technologies B instrument at a pulling speed of 20 mm/min. The length, width, and thickness of the films were recorded for analysis. Three samples from each film were tested, and the average values were used to determine the tensile strength of the films.

#### \* Capacitance measurements

The capacitance of the material samples with varying PPy concentrations was measured using an LCR Meter Model 3550 across a frequency range from 1 kHz to 10 kHz. The cellulose/PPy membrane samples were positioned between two pieces of copper tape, which were connected to the device for measurement (Figure 5). Detailed specifications and performance metrics for each sample will be discussed in the Results and Discussion section (Table 1).



**Figure 5.** Capacitance measurement method of conductive cellulose/PPy membranes

**\* Fourier Transform Infrared Spectroscopy (FT-IR)**

The cellulose/PPy powder samples, obtained after the drying and water evaporation processes, along with individual cellulose and PPy samples synthesized similarly for comparison with the composite samples, were analyzed using a Nicolet™ iS50 FT-IR Spectrometer. The spectra were recorded in the wavenumber range from 4000  $\text{cm}^{-1}$  to 500  $\text{cm}^{-1}$ , encompassing 16 scans per sample at a resolution of 4  $\text{cm}^{-1}$ .

**\* Field Emission Scanning Electron Microscopy (FE-SEM)**

The surface morphology of the synthesized cellulose and cellulose/PPy membranes was examined using a Hitachi S-4800 FE-SEM at an operating voltage of 3.00 kV.

**\* Particle size distribution measurements**

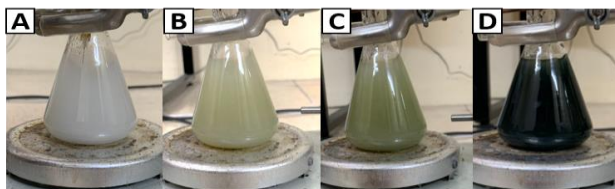
Particle size measurements were conducted using dynamic light scattering (DLS) with a Horiba LB-550 DLS Nanoparticle Size Analyzer. PPy particles, synthesized through polymerization under conditions and ratios similar to those used for the cellulose composite samples, were dispersed in distilled water for subsequent analysis. The measurements were conducted using a quartz cuvette to ensure precision and accuracy in the evaluation of particle characteristics.

**\* Differential Scanning Calorimetry (DSC)**

The thermal properties of cellulose films and conductive cellulose/PPy membranes were investigated using a Flash Differential Scanning Calorimetry DSC2 instrument. The analysis was performed over a temperature range from 25 °C to 500 °C, with a heating rate of 10 °C per minute.

## 2.2. Results and discussion

### 2.2.1. Results of material synthesis

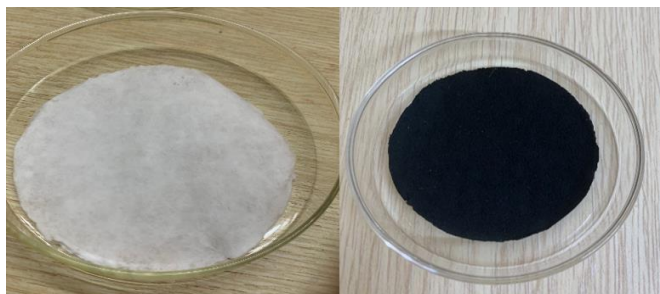


**Figure 6. Polypyrrole/cellulose fibers (cellulose/PPy) polymerization process (A) Cellulose; PPy polymerization reaction after (B) 30 minutes, (C) 60 minutes, and (D) 90 minutes**

Following the extraction process from cardboard, the resulting cellulose sample is white, indicative of the successful removal of impurities such as lignin and hemicellulose (Figure 6A). During the polymerization of PPy from pyrrole and  $\text{FeCl}_3$  solution, the cellulose suspension gradually transitions from white to dark blue, reflecting the formation of PPy within the solution (Figure 6). The cellulose fibers interlace to form a cellulose membrane, which appears black due to the incorporation of polypyrrole (Figure 7).

As outlined in the previous section, the composite cellulose/PPy membrane samples, distinguished by varying PPy content, are labeled as Sample 1 (0.1 ml pyrrole), Sample 2 (0.2 ml pyrrole), and Sample 3 (0.3 ml pyrrole). Detailed specifications for each sample are provided in Table 1.



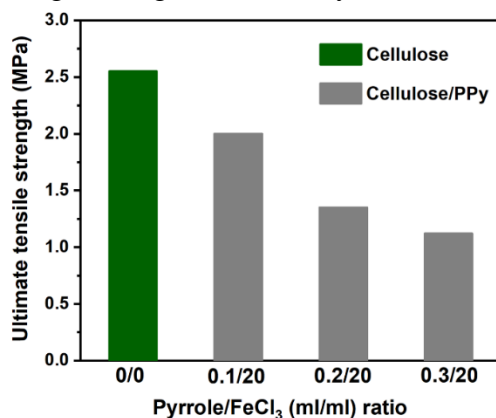


**Figure 7. Cellulose membrane (left) and cellulose/PPy membrane (right)**

**Table 1. Specifications of cellulose/PPy membrane samples**

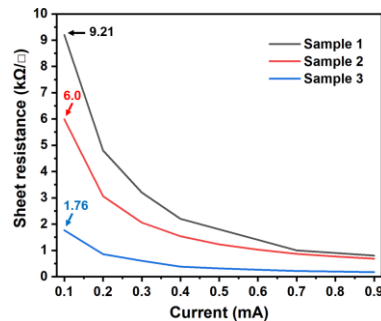
Sample	Pyrrole/FeCl <sub>3</sub> (3.24 mM aqueous solution) volume ratio	Thickness (mm)	Mass (g)	Sheet resistance at 0.1 mA (kΩ/□)	Conductivity at 0.1 mA (mS/cm)	Capacitance at 1000 Hz (pF/cm <sup>2</sup> )
Sample 1	0.1/20	0.319	0.017	9.21	3.40	22.5
Sample 2	0.2/20	0.366	0.020	6	4.55	38.5
Sample 3	0.3/20	0.315	0.016	1.76	18.04	50

In the cellulose/PPy films, cellulose fibers serve as reinforcing agents, enhancing the mechanical properties of the material. The tensile strength of the film samples is attributed to the ordered structure of cellulose and its complex hydrogen bonding network. As the PPy content increases, the tensile strength decreases significantly (Figure 8). The pure cellulose film exhibits high tensile strength, reaching up to 2.6 MPa, while the cellulose/PPy composite films with varying ratios show a decreasing trend in strength, ranging from 2.1 MPa to 1.2 MPa. The PPy coating on the cellulose surface markedly reduces the hydrogen bonding interactions between cellulose molecules. When the Pyrrole/FeCl<sub>3</sub> ratio exceeds 0.3/20, the material becomes brittle, easily breaking, and fails to form a film. Within the range of cellulose/PPy film ratios investigated, the obtained films display high tensile strength and good flexibility without material breakage.



**Figure 8. Tensile strength of cellulose and cellulose/PPy film samples**

### 2.2.2. Electrical conductivity of composite materials



**Figure 9.** Sheet resistance versus current curves for the three cellulose/PPy composite samples obtained by the four-point probe resistance measurement technique

Transforming cellulose from an insulating polymer into a conductive material presents substantial challenges. The integration of cellulose with PPy through polymerization reactions demonstrates promising potential for enhancing the conductivity of the composite material. In this composite, PPy serves as the primary conductor, while cellulose contributes to mechanical strength, and flexibility, and serves as the structural framework of the membrane. To assess the conductivity of the composite material, samples with varying PPy concentrations were evaluated (see Table 1). The sheet resistance of the material was observed to increase proportionally with the amount of conducting polymer incorporated. Sample 3 exhibited the lowest sheet resistance, measuring 1.76 kΩ/□ at 0.1 mA (conductivity of 18 mS/cm), highlighting the significant impact of PPy content on the stability of the material's conductivity (Figure 9). The electrical conductivity of the composite membrane is primarily influenced by the quality of the PPy conducting polymer (which depends on the oxidation/monomer ratio) and the cellulose content. While reducing cellulose content and increasing PPy concentration can enhance conductivity, cellulose remains crucial for reinforcing and improving the mechanical properties of the material [18]. In this study, coating cellulose fibers with PPy through polymerization has endowed the composite material with conductive properties comparable to those in previous research (see Table 2). Given the resource advantages and cost-effectiveness, producing cellulose/PPy conductive films from cardboard is a promising approach. To identify the optimal ratio for energy storage applications, it is crucial to investigate the capacitance values of these conductive films.

**Table 2.** Comparison of conductivity level of the cellulose/PPy and previously reported conductive cellulose composite

Materials	Conductivity level (mS/cm)	References
Cellulose nanofiber/polypyrrole/polyvinylpyrrolidone	100–6230	[19]
Polypyrrole/SnCl <sub>2</sub> -modified bacterial cellulose composite	500	[20]
Cellulose and polypyrrole composite	8400	[21]
Bacterial cellulose/polypyrrole membrane	3390	[22]
Softwood pulp and polypyrrole	52	[23]
Cellulose from bamboo and polypyrrole	9×10 <sup>-6</sup> –3.33×10 <sup>-2</sup>	[24]
Cellulose/polypyrrole composite	18	This work

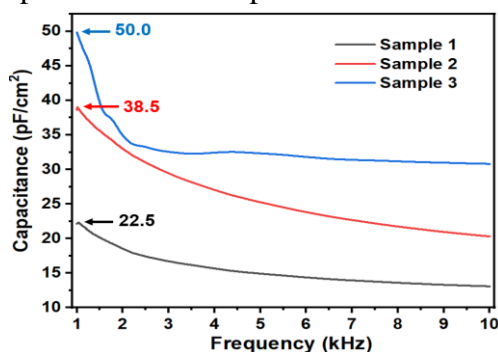


### 2.2.3. Capacitive characteristics of composite materials

The capacitance of samples listed in Table 1 was evaluated over a frequency range of 1–10 kHz, as illustrated in Figure 10. The capacitance values at 1000 Hz exhibited a progressive increase with higher PPy content: Sample 1 showed 22.5 pF/cm<sup>2</sup> (1.324 pF/g), Sample 2 exhibited 38.5 pF/cm<sup>2</sup> (1.925 pF/g), and Sample 3 reached 50 pF/cm<sup>2</sup> (3.125 pF/g).

The dielectric constant of CF is approximately 5–8 at 1000 Hz [25]–[27], whereas that of PPy is nearly 1000, significantly exceeding that of CF by more than two orders of magnitude [28]. Consequently, an increase in PPy content in the composite material results in higher capacitance values. This finding aligns with previous reports indicating that the incorporation of polar materials, such as PPy, enhances dipolar polarization and thus increases the dielectric constant of the composite [29].

Figure 10 reveals that Sample 3 exhibits capacitance instability, particularly at frequencies below 2 kHz when compared to Samples 1 and 2. This instability is attributed to excessive PPy content, which causes a non-uniform distribution of the conducting polymer and leads to variable capacitance values across different frequencies. Moreover, an excessive amount of PPy can compromise the mechanical properties of the material, complicating the control of the polymerization process. In comparison with previous research, such as Agarwal *et al.*, who achieved a capacitance of 16 pF/in<sup>2</sup> (2.48 pF/cm<sup>2</sup>) for conductive paper membranes [30], the cellulose/PPy composite membrane in this study demonstrates comparable, if not superior, capacitance values [31]. This suggests that the process of coating PPy onto the cellulose surface significantly influences the material's properties and potential applications. Given its superior stability, charge storage capability, and high conductivity, Sample 2 will be further examined to investigate the formation and distribution of PPy on the cellulose membrane and its impact on the thermal properties of the composite.

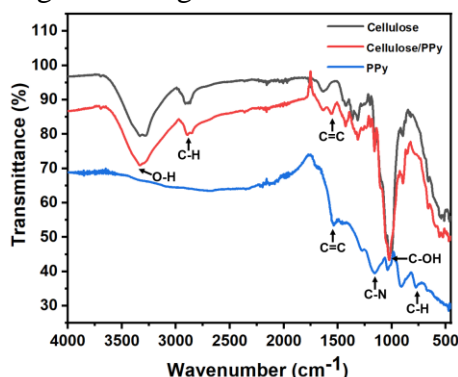


**Figure 10. Capacitance versus frequency curves of different cellulose/PPy composite samples**

### 2.2.4. Fourier transform infrared spectroscopy (FT-IR)

Cellulose is distinguished by hydrogen bonding interactions among hydroxyl groups on its linear chains, leading to various fibrous structures and morphologies. As illustrated in Figure 11, the FT-IR spectrum of cellulose reveals characteristic peaks associated with its structural components. The peak at 1028 cm<sup>-1</sup> corresponds to the –C–O–C pyranose ring skeletal vibration [32]. The peak at 2890 cm<sup>-1</sup> is attributed to the stretching vibrations

of C–H bonds, while the broad peak observed between 3200 and 3400  $\text{cm}^{-1}$  is indicative of O–H stretching and hydrogen bonding within the  $\text{CH}_2$  and  $\text{CH}_3$  groups of cellulose [33].

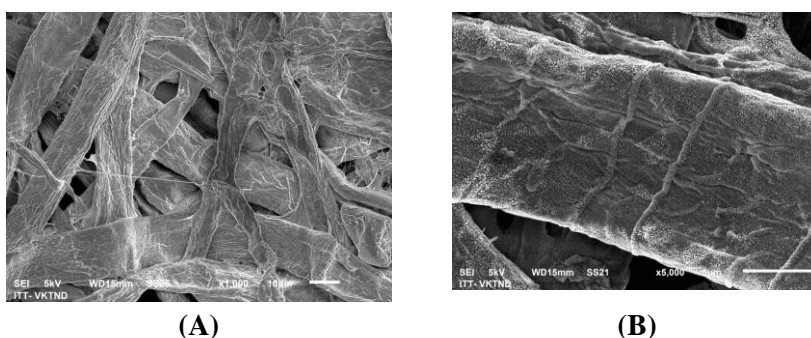


**Figure 11.** FT-IR spectra of cellulose, PPy, and cellulose/PPy composite

In contrast, the FT-IR spectrum of PPy exhibits distinct features. Out-of-plane C–H bending vibrations within the PPy ring structure are observed in the 700–900  $\text{cm}^{-1}$  range. The prominent peak at 1160  $\text{cm}^{-1}$  is associated with the C–N bonds in pyrrole, while the peak at 1550  $\text{cm}^{-1}$  corresponds to the stretching vibrations of C=C double bonds, characteristic of the benzene rings in PPy. This peak reflects the conjugation length of the PPy chains [34].

In the case of cellulose/PPy composite material, the FT-IR spectrum retains the characteristic peaks of cellulose, with an additional peak at 1550  $\text{cm}^{-1}$  corresponding to the C=C vibrations of PPy. The observed reduction in transmittance intensity in this sample signifies the successful formation of PPy on cellulose, confirming the efficacy of the polymerization reaction. The morphology and distribution of the components within the composite material will be further investigated using Field Emission Scanning Electron Microscopy (FE-SEM).

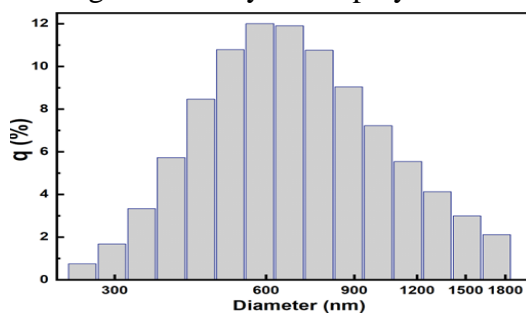
### 2.2.5. Surface morphology



**Figure 12.** FE-SEM images of (A) Cellulose, (B) cellulose/PPy (Sample 2)

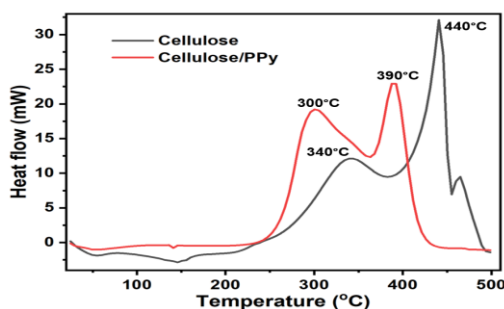
The cellulose material extracted from cardboard displays distinct fibrous properties, forming a biologically structured membrane, as illustrated in Figure 12A. In contrast, the composite material reveals a uniform distribution of PPy particles on the cellulose surface, indicating thorough coverage across the fiber structure, as shown in Figure 12B. To assess particle size, PPy was synthesized using a chemical polymerization method

with a pyrrole/ $\text{FeCl}_3$  (3.24 mM aqueous solution) volume ratio of 0.2/20 (Sample 2), under conditions analogous to those applied for the composite samples. Analysis using Field Emission Scanning Electron Microscopy (FE-SEM) and Dynamic Light Scattering (DLS) revealed that the PPy particles exhibit a granular morphology, with sizes ranging from 250 to 1800 nm, predominantly clustered in the 600 to 700 nm range (Figure 13). The uniform deposition of PPy on the cellulose fibers significantly modifies the surface properties and structural characteristics of the composite material, enhancing its electrical conductivity and demonstrating the efficacy of the polymerization process.



**Figure 13.** The particle size distribution graph of polypyrrole

### 2.2.6. Differential Scanning Calorimetry (DSC)



**Figure 14.** DSC plots of cellulose and cellulose/PPy composite

Differential scanning calorimetry (DSC) analysis of cellulose and cellulose/PPy composite samples demonstrates a notable influence of PPy on the thermal properties of these materials (Figure 14). The DSC curve for pure cellulose reveals a minor endothermic peak at approximately 150 °C, attributed to cellulose dehydration. As the temperature increases, cellulose begins to decompose at around 340 °C, with complete degradation occurring at approximately 440 °C. In contrast, the cellulose/PPy composite (Sample 2) exhibits enhanced thermal stability below 250 °C, with a small endothermic peak corresponding to water evaporation from the sample. At higher temperatures, between 250 and 350 °C, a significant exothermic peak is observed at 300 °C, indicating mass loss due to the thermal decomposition of PPy. A further exothermic peak at 390 °C, within the 375–420 °C range, is attributed to the combined decomposition of cellulose and residual PPy within the composite membrane structure. While the inclusion of PPy enhances the low-temperature stability of the cellulose/PPy membrane (below 200 °C), it simultaneously decreases the material's resistance to high temperatures compared to conventional cellulose membranes.

### 3. Conclusions

Cellulose fibers derived from waste paper were synthesized through a series of chemical treatments, including alkalization, bleaching, and acid hydrolysis, and subsequently employed as substrates in conductive polymer membranes. The *in situ* polymerization of pyrrole on the cellulose surface proved highly effective in fabricating cellulose/PPy conductive membranes. The resulting cellulose/PPy composite membrane demonstrated thermal stability across a temperature range of 25–250 °C. The analysis confirmed the presence of C–H, C–N, and C=C chemical bonds, indicative of successful PPy formation on the cellulose fibers. The size and uniformity of the PPy particles are critical factors influencing the membrane's properties and potential applications. These properties can be modulated during the polymerization process by adjusting the pyrrole concentration. The composite membrane demonstrated high electrical conductivity, reaching up to 18.04 mS/cm at 0.1 mA. Additionally, it exhibited excellent charge storage capacity, with the highest recorded capacitance of 50 pF/cm<sup>2</sup> (3.125 nF/g) at 1000 Hz. These properties render it a highly promising candidate for applications in electronics and energy storage.

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