# Synthesis and Characterization of Polypyrrole/Graphene Oxide/Natural Polymer Bio composites with Enhanced Electrical Conductivity and Biodegradability

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## Abstract

This work reports the successful synthesis of novel polypyrrole (Ppy) based biocomposites with enhanced electrical conductivity and biodegradability. These materials were synthesized through chemical in situ polymerization, incorporating graphene oxide and various natural fillers; cellulose, starch and sodium alginate into the polypyrrole matrix. The composites were comprehensively characterized using FTIR, SEM, UV-DRS, four probe electrical conductivity measurements and biodegradability evaluation through weight loss method. FTIR studies confirmed the integration of all three the fillers into the polypyrrole matrix. SEM micrographs revealed that sodium alginate containing composites exhibited increased porosity, which can be correlated with enhanced biodegradability. UV-DRS results showed that the composites exhibited lower band gaps compared to pure polypyrrole, indicating improved electrical conductivity. The biodegradability tests showed significant improvement in the decomposition rates for the biocomposites relative to pure polypyrrole, especially Ppy/GO/Sodium alginate, exhibited better biodegradation properties. Comparative analysis of all the synthesized materials revealed that all the three PPy based biocomposites demonstrated superior performance with respect to both conductivity and biodegradability than pure Ppy, with Ppy/GO/Sodium alginate composite evolving as a potential candidate for environmentally sustainable applications.

## Introduction

The remarkable integration of electrical conductivity with polymer properties has positioned conducting polymers at the forefront of innovations in materials science and technology development(Balint et al., 2014; Kumar et al., 2025; Wang et al., 2019). Among the conducting polymers, Polypyrrole has attracted special interest due to its high electrical conductivity, ease of synthesis and high environmental stability, good mechanical properties(Nakata et al., 1992) along with its heterocyclic structure (Meher et al., 2025; Wang et al., 2015)and non-toxicity (Wang et al., 2015). Ppy can be easily prepared by either an oxidative, chemical or electro chemical polymerization of Pyrrole. Ppy can be often used as biosensors, gas sensors, wires, microactuators, anti-electro static coatings, solid electrolytic capacitor, electro chromic windows and displays, and packaging polymeric batteries, electronic devices and functional membranes, etc.(Ateh et al., 2006; Bhattacharya and De, 1996; Khuyen et al., 2022). However, conventional conducting polymers including Ppy lack biodegradability, which present significant environmental concerns as electronic waste continues to accumulate across the globe(Lei et al., 2017). To overcome this limitation, recent research have focused on the synthesis of biocomposites, by incorporating natural polymers into conducting polymer matrix, towards developing materials that combine electrical functionality with environmental sustainability(Ahmad et al., 2017; Han et al., 2022; Lyu et al., 2016).

An additional strategy to improve electrical conductivity and overall performance of Ppy can be achieved through integration of graphene oxide into Ppy matrix. Graphene oxide (GO), a highly functionalized form of graphene, with its two dimensional structure and exceptional thermal, electrical and mechanical properties serves as an ideal reinforcing material for enhancing polymer composite performance(Zhang, 2025). It has gained significant attention due to its high surface area and strong interaction with conducting polymers(Anjali et al., 2025). When integrated into conducting polymer matrices GO can create synergistic effects that can improve electrical conductivity. Even though, there are number of studies on the combinations of Ppy and GO, there is limited research(Ahmad et al., 2017; Kenry and Liu, 2018) that integrates natural biopolymers alongside GO into Ppy matrix to achieve improved conductivity and enhanced biodegradability(Han et al., 2022; Haseen et al., 2020).

The present study, addresses these limitations by investigating the synthesis of novel polypyrrole based biocomposites, using a combination of graphene oxide and natural biopolymers (cellulose, starch and sodium alginate). The main aim of this research is to focus on the interplay between conducting properties of polypyrrole, the electrical enhancement provided by GO and the biodegradability imparted by the natural polymeric fillers. The desired biocomposites will be prepared by in situ polymerization and thus synthesized composites will be characterized reaction by various spectroscopic techniques; structural information by FTIR, surface morphology by scanning electron microscopy (SEM), electrical conductivity by using Four-Probe method, band gap analysis using the UV visible diffuse reflectance spectra. Biodegradability is assessed by monitoring the weight loss of polymer composite pellets.

## MATERIALS AND METHODS

### Experimental Section

### 2.1 Materials:

Graphite powder, $KMnO_4$, Pyrrole, $FeCl_3$, Starch, Cellulose and Sodium alginate were of AR grade and sourced from Sigma Aldrich.

### 2.1 Synthesis of Polypyrrole

Polypyrrole was chemically synthesized through chemical oxidative polymerization method in the aqueous medium. A 1 M pyrrole solution was prepared by dissolving pyrrole (43mM) in 50 ml and cooled in an ice bath to maintain the reaction temperature between 5-7°C. A separate oxidizing solution of $FeCl_3$ (100mM) was prepared in distilled water, maintaining a monomer to oxidant ratio 1:2.3. The $FeCl_3$ was added slowly, to the pyrrole solution with constant stirring for 30 minutes, by maintaining the reaction temperature low throughout. Then the polymerization was conducted for 4 hours under constant stirring. The black Ppy precipitate obtained was kept unagitated for 24 hours to allow it to settle down. The Polypyrrole powder was filtered out under vacuum and washed with distilled water several times to remove any impurities present. The polypyrrole was dried for 2 days at room temperature.

### Synthesis of Graphene oxide

Graphene oxide(GO) was prepared by the well-known Hummers method (Anegbe et al., 2024; Yusoff et al., 2022). In a typical synthesis, 15g of Graphite powder was mixed with 7.5g of $_3$ and 360ml of $H_2SO_4$.The mixture was then stirred in an ice bath for 10 min followed by the slow addition of $KMnO_4$ (45g) by maintaining the temperature below 5°C. The suspension was then allowed to react for 2 hours in an ice bath and stirred for 2 hours at room temperature. The temperature of the reaction mixture was then kept constant at 98°C for 1 hour and deionized water was added continuously to make up the volume to 400 ml. At the end, 45ml of $H_2O_2$ was poured into the reaction mixture in order to oxidize the graphene. The final product was washed with deionized water and 5% HCl, several times followed by filtration and centrifugation. Finally the product was dried at 60°C.

### 2.2 Synthesis of Polypyrrole Biocomposites

Using the above formulation for the preparation of Polypyrrole, 0.4g each of natural polymer (Starch,Cellulose and Sodium alginate) seperately and GO were added to prepare the desired bio composites. The black precipitate of GO containing conducting polymer was filtered, washed thoroughly with plenty of water and dried at 50°C in an oven.

### Characterization Techniques

### 2.3 Fourier Transform Spectroscopy (FTIR)

The structural information of the prepared conducting polymer composites was done by using FTIR spectrophotometer - Thermofisher Scientific Nicolet IS50FTIR. Solid sample was mixed with solid KBr (transparent in the mid IR region), then ground and pressed in a special dye to yield a transparent window.

### 2.4 UV-DRS Spectroscopy

Band gap energy of the prepared conducting polymer composites were measured using UV – DRS spectrophotometer -Cary 5000 with spectrophotometric performance in the 175- 3300 nm range. 50 mg of sample is made into aqueous solution for easy analysis.

### 2.5 Scanning Electron Microscopy (SEM)

Surface morphology of the prepared polymer composites were analyzed by SEM -Carl Zeiss EVO 18 Secondary Electron microscope with EDS. Samples were thoroughly degreased and dried to eliminate any outgassing from organic contamination and water. Samples can be cleaned using acetone or alcohol. After cleaning the sample, it is dried and compressed into small disks for sample mounting.

## 2.6 Conductivity Measurement by Four Probe Method

Electronic conductivity of the samples were measured by using the four-probe method (INCOXIN). The sample for the analysis were made into pellets of 1mm diameter to get finer results.

## 2.7 Biodegradability Test

Biodegradability studies are very important to ensure the bio accumulative potential of the material under investigation and to measure the extent of damage the material may cause to the environment if it is non-biodegradable. Thus, the main objective behind this test was to substantiate the claim that the as prepared polymer samples were non-toxic and biodegradable. This test was done by dipping, the pellets of samples in soil and take the weight in each five days. The percentage weight loss is directly proportional to the biodegradability of the samples and calculated by the equation,

% weight loss =**100 x** (initial weight – final weight)/initial weight

## 3. RESULTS AND DISCUSSION

### FTIR Analysis

Incorporation of various fillers into the matrix were determined with the help of a Fourier transform infra-red spectrometer (FTIR).

### 3.11 Polypyrrole/Graphene oxide/Cellulose (Ppy/GO/Cellulose)

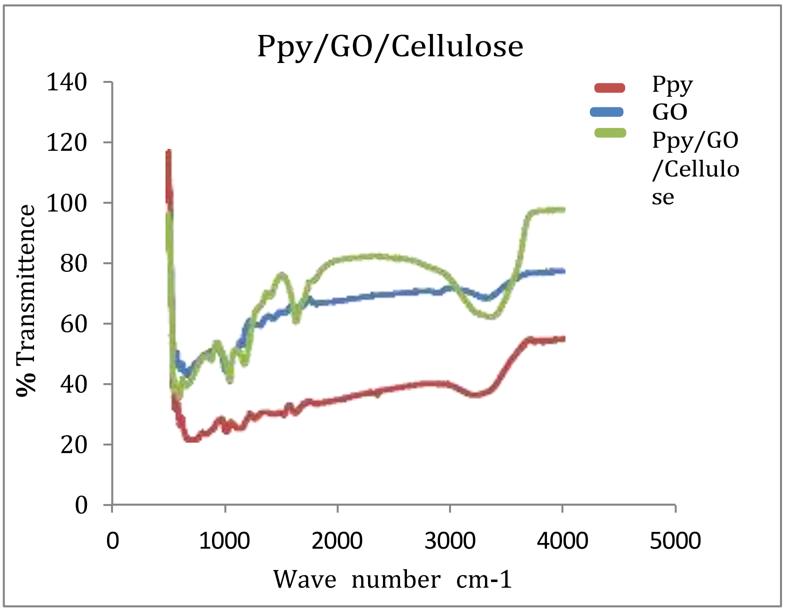


**FIG: 3.1**

From the fig; 3.1, it gives the FTIR spectra of Ppy / GO / Cellulose polymer composite. In Ppy/GO/Cellulose composite, the broad peak at 3400-3500Cm-1 in the IR spectra of Ppy/GO/Cellulose corresponds to O-H stretching vibration while the peak at around 2900-300Cm-1 is ascribed to the C-H stretching mode in the Pyranoid ring. The absorption band at 1100-1200Cm-1 is due to asymmetric stretching of a Glycosidic C-O-C bridge in the cellulose chains. The absorption band at near 1000Cm-1 is attributed to a stretching vibration of C-O. Moreover, the peaks at near 3400Cm-1 and 1400-1600cm$^{-1}$ can be assigned to the stretching vibrations of N=H and C=C bonds, respectively(Han and Lu, 2007). The peaks at 1400-1600Cm-1 and near 1300 Cm-1 correspond to C-N and C-H stretching vibrations with aromatic conjugation. It can be seen from the FTIR spectra that the polymer composite possesses the characteristic bands of both cellulose and Ppy. The characteristic broad peaks at 3300-3400Cm-1 and near 3100Cm-1 can be assigned to O-H and N-H stretching vibration of cellulose and Ppy, respectively(Konwer et al., 2011). The peaks at 2800- 3000cm-1 correspond to C-H stretching modes of the Pyranoid ring, methyl and methylene groups of a dodecyl substituent, which confirms the presence of the grapheme molecules. The absorption bands at 1100- 1200 cm$^{-1}$ are due to asymmetric stretching of the C-O-C bridges and stretching vibration of C-O on cellulose(Ding et al., 2010). The observed red shift of the O-H and N-H stretching bands 3100-3400 cm$^{-1}$ may be caused by intermolecular hydrogen bonding between the N-H of the Ppy ring and the O-H group of cellulose in the bio- composite. The appearance of the C-C and C-N vibration bands 9400-1600 cm-1 of the composites confirms that the Ppy units are introduced into the matrix of the composite, and the results suggested that a Ppy/GO/Cellulose bio-composite, was successfully prepared.

## 3.12 olypyrrole/Graphene oxide/Starch (Ppy/GO/Starch)

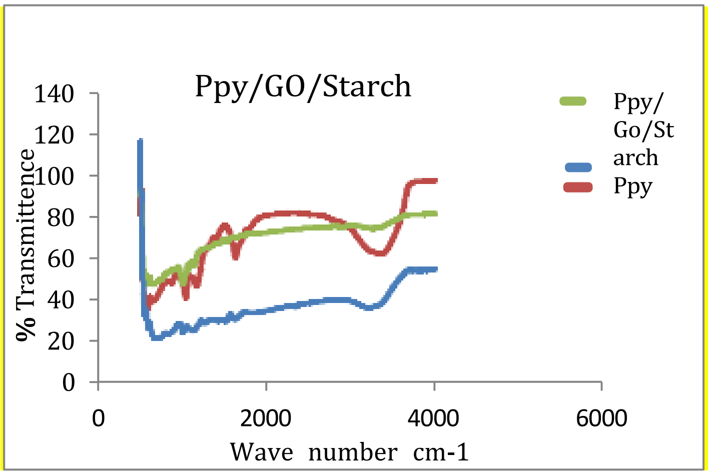
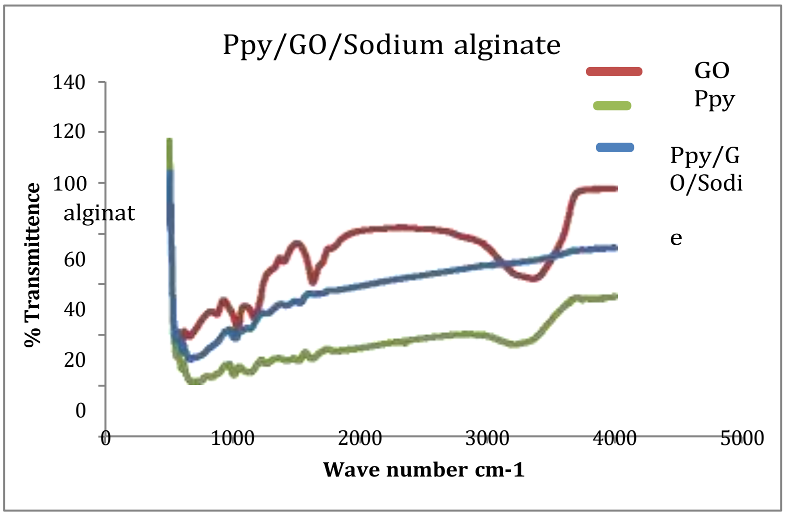


Fig 3.2

From the fig;3.2, it shows the FTIR spectra of Ppy / GO / Starch polymer composites. In Ppy/GO/Starch polymer composite, there is a small peak near 1600 cm$^{-1}$ which is the characteristic peak of pyrrole. The peak near 1000 cm-1 indicates the N-H deformation of pyrrole. The small peaks appeared in the region between 1000-1600 cm-1 indicates the skeletal vibrations and ring stretching of pyrrole ring, C-N, C-H stretching vibrations with aromatic conjugation of Pyrrole ring, benzene ring and carboxylic group of grapheme oxide, COOH stretching of GO and C-O stretching of GO, C=C stretching vibrations of pyrrole ring and C=N stretching vibrations of pyrrole ring. The peaks near 700 $cm^{-1}$ indicates that the Polypyrrole is polymerized in through α position(Bose et al., 2010). The peak region below 800 $cm^{-1}$ exhibited complex vibrational modes due to the skeletal vibrations of pyranose ring in the glucose unit. The peaks appear in the region 1000- 1200 $cm^{-1}$ also indicates the anhydrous glucose ring O-C stretching, C-O, C-C and C-O- H stretching vibrations and C-O-H bending vibrations of starch(Chen et al., 2006). The region below 900 $cm^{-1}$ indicates the absorption by excess water.

## 3.13 Polypyrrole/GO/Sodium alginate (Ppy/GO/Sodium alginate)



Fig; 3.3

From the fig; 3.3, it is evident that, In Ppy/GO/Sodium alginate polymer composite, the peak near 1600 $cm^{-1}$ is the characteristic peak of Pyrrole the peak near 1000 $cm^{-1}$ indicates the N-H deformation of pyrrole, the small peaks near 1000-1600 $cm^{-1}$ provided the following information, also indicate the COOH stretching of GO. Skeletal vibrations and ring stretching of pyrrole ring, C-N,C-H stretching with aromatic conjugation of pyrrole ring, benzene ring and carboxylic group of grapheme oxide, C=C and C=N stretching of pyrrole ring. The weak broad peaks in between 1000-1500 $cm^{-1}$ indicate that symmetric stretching of COO in sodium alginate, C-H in plane deformation of sodium alginate, C-O, C-C, and C-O-C stretching vibrations in sodium alginate, O-H in plane deformation in sodium alginate(Verma et al., 2023). The peaks below 1000Cm-1 indicates the CCH deformation and CO deformation.

### SEM Analysis

Surface morphology of the prepared bio composites were analyzed by SEM. From the SEM micrographs it may be helpful to get information about the biodegradable nature of the prepared bio composites.

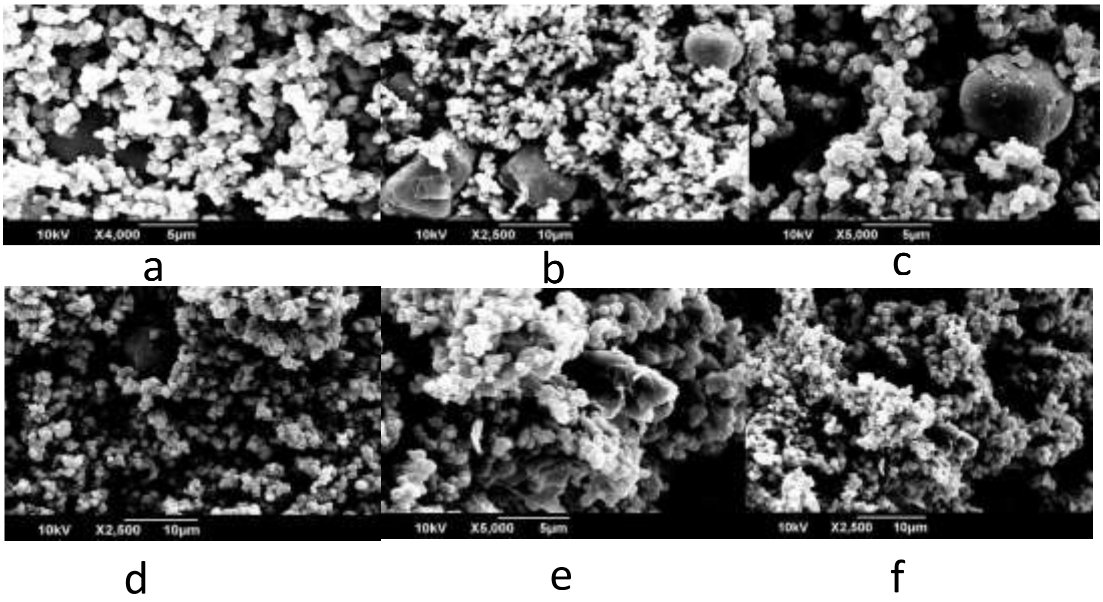


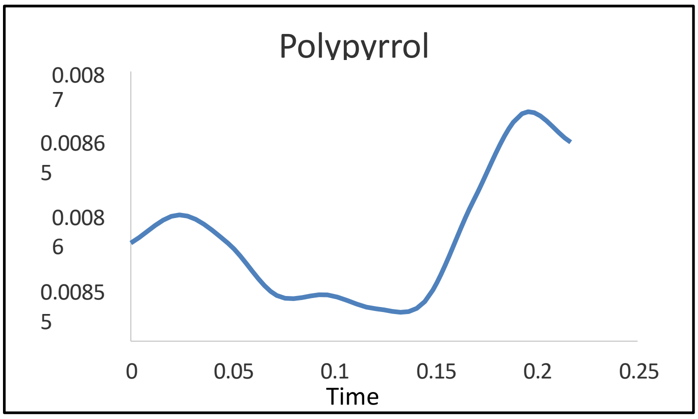
**Fig. 4 SEM images of (a-b) Ppy/GO/Cellulose, (c-d) Ppy/GO/ Starch (e-f) Ppy/GO/ SodiumAlginate**

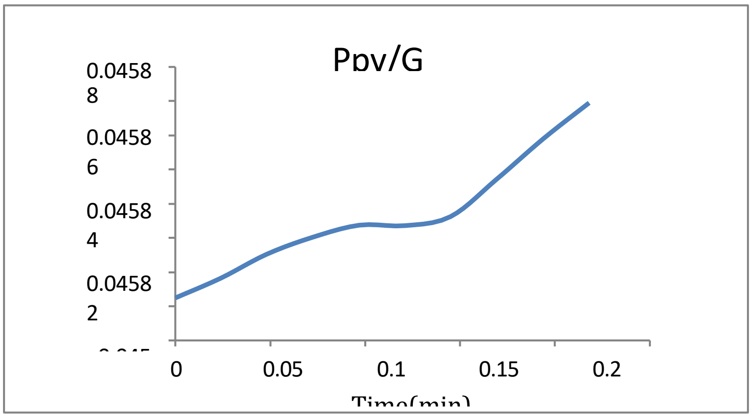
The surface morphology of polypyrrole and Graphene oxide composites with cellulose, starch and sodium alginate as natural fillers was studied using SEM analysis. The SEM micrographs of Ppy/GO/Cellulose, Ppy/GO/ Starch and Ppy/GO/ Alginate are presented in Fig.4, which depicts the characteristic features of a biodegradable polymer and the as- prepared biocomposite (Ppy/GO/ Cellulose). All three biocomposites exhibit a highly porous with granular particle distribution and densely packed structured aggregates. The particles are irregularly arranged with Graphene oxide particles embedded uniformly in the polymer matrix. The Fig. 4 further illustrates that the materials have appreciable porosity, indicating high surface area and structural uniformity with spherical granular structures(Wang et al., 2013). The light coloured cellulose was uniformly mixed with dark Ppy chains showing a cauliflower- like structure(Ding et al., 2010). The micro structure of very fine GO mono dispersed into the biodegradable polymers is also distinct.

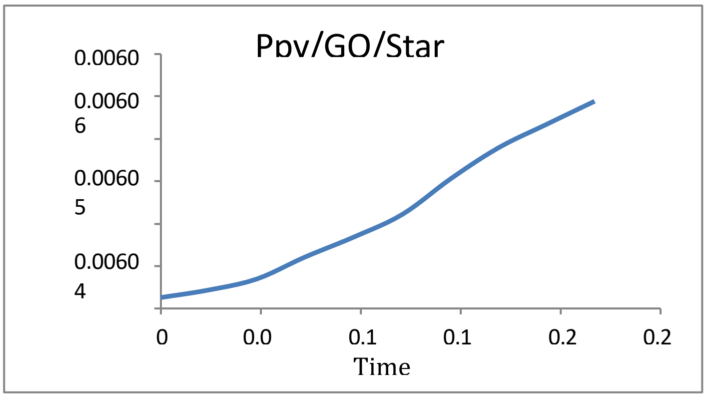
Among the three polymer composites, it has been observed that, ppy /GO/ Sodium alginate composite displays the uniform particle distribution, appreciable porosity with interconnected morphology, which is expected to enhance the conductivity and bio decomposition of the polymer composite.

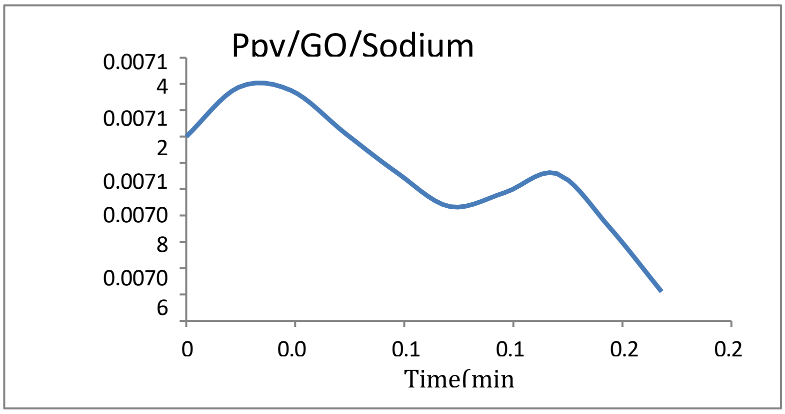
**Conductivity**

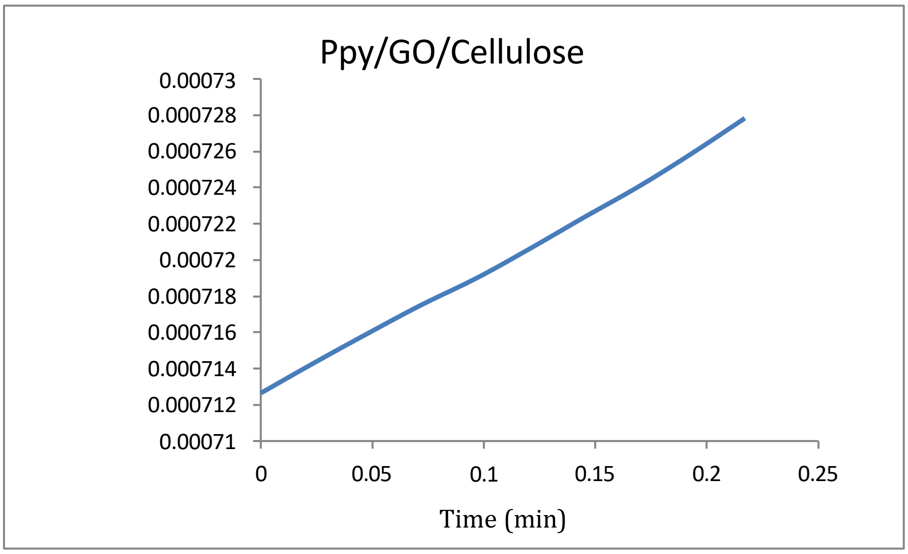
Conductivity of the samples was evaluated using the four probe method.











Conductance was measured at half-minute intervals for all samples, and the results are presented in Figures 5.

Fig. 5 displays the conductance-time profile of pure polypyrrole (Ppy), which suggests a non-linear, wave-like behaviour pattern. The conductance initially increasd, then decreased slightly at 0.5 minutes, followed by another increase peaking at 1 minute. This is succeeded by a decrease at 1.5 minutes and a sharp rise at 2 minutes. Such fluctuation points towards unstable charge transport characteristics in pure Ppy.

In contrast, the Ppy/GO composite displays a more stable and increasing trend in conductance, with a notable rise at 1.5 minutes. The incorporation of graphene oxide enhances the conductivity by facilitating improved charge mobility and network formation.

The Ppy/GO/Cellulose composite exhibits a nearly linear increase in conductance, indicating consistent and progressive enhancement in electrical behavior. This stability may be attributed to the structural support provided by cellulose, enabling uniform dispersion of the conductive phase.

Similarly, the Ppy/GO/Starch composite shows a smooth, steadily increasing conductance profile, further confirming that natural polymers such as starch contribute to better integration and conductivity within the composite matrix.

The Ppy/GO/Sodium Alginate composite presents a wave-like pattern, with conductance peaking at 0.5 and 1.5 minutes and declining thereafter. These periodic variations suggest dynamic interactions between the conductive polymer and the sodium alginate matrix, potentially due to changes in internal structural alignment over time.

All Ppy-based composites exhibited higher conductivity than pure Ppy(Basavaraja et al., 2011; Zhong et al., 2015). The incorporation of GO significantly enhanced charge transport, while the addition of biopolymers such as cellulose, starch, and sodium alginate further modifies the conductivity behavior—either by stabilizing it or introducing time-dependent modulation. These findings highlight the potential of such composites for use in applications requiring tailored electrical performance, such as flexible electronics and responsive sensors.

## Biodegradability

Biodegradability studies are very important to ensure the bio accumulative potential of the material under investigation and to measure the extent of damage the material may cause to the environment if it is non-biodegradable. Thus, the primary objective behind this test was to substantiate the claim that the as prepared polymer samples were biodegradable. This test was done by dipping, the pellets of samples in soil(Sareena et al., 2014) and take the weight in each five days. Sample was properly washed and dried followed by making small pellets for dipping in soil to measure % weight loss.

Table 1. **Biodegradability profile of Ppy-GO composites**

| Sample | Initial weight loss% (Zero day) | Wtloss% after 5 days | Wt loss % after 10 days | Wt loss% after 15 days |
|---|---|---|---|---|
| Ppy | 0 | 0.17 | 0.56 | 0.68 |
| Ppy/Go/Cellulose | 0 | 3.11 | 3.48 | 4.58 |
| Ppy/Go/Starch | 0 | 1.32 | 2.22 | 2.68 |
| Ppy/Go/Na alginate | 0 | 9.16 | 14 | 17.40 |

**Table no: 1**

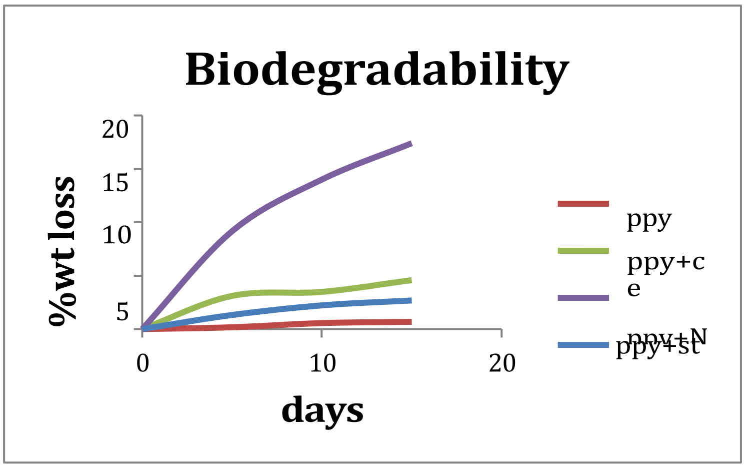


**Fig6. Biodegradability profile of Ppy-GO composites**

By knowing the Biodegradability of the prepared composite, % of weight loss of the samples in regular intervals of time(5 days) was calculated by using the equation,

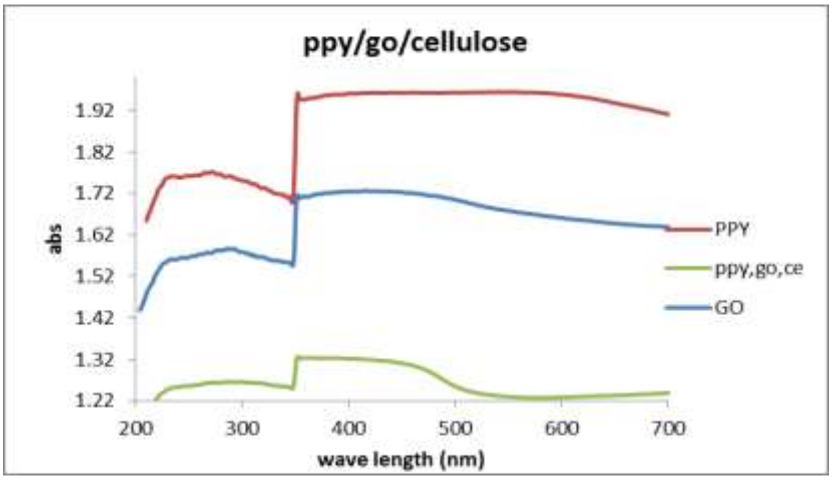
$$\textbf{\% of weight loss} = \frac{\textit{initial weight} - \textit{final weight}}{\textit{initial weight}} \times 100$$

**Fig 6** and Table 1, illustrates the biodegradability trend of each sample over the 15-day period. The data reveal that **pure polypyrrole (Ppy)** exhibits minimal weight loss, indicating very limited biodegradation. In contrast, all composite samples show significant increases in weight loss over time, suggesting improved biodegradability due to the incorporation of biodegradable components(Piccioni et al., 2024). Among the composites, **Ppy/GO/Sodium Alginate** demonstrates the highest biodegradation rate, with a 17.40% weight loss after 15 days. This is followed by **Ppy/GO/Cellulose** (4.58%) and **Ppy/GO/Starch** (2.68%). These results confirm that the inclusion of natural biopolymers such as sodium alginate, cellulose, and starch enhances the environmental degradability of the composites, making them more eco-friendly alternatives to pure Ppy(Ferreira and Andrade, 2021).

## UV - DRS

UV-DRS spectra are very useful for calculating the band gap energy of the as prepared bio composites. In the present study, UV-DRS spectra have been generated by plotting absorbance vs band gap energy for prepared composites(Luo et al., 2011). The band gap energy is inversely proportional to the conductivity of the materials, that is, the lower the band gap energy, higher will be the conductivity of the bio composites.

**Ppy/GO/Cellulose composite**



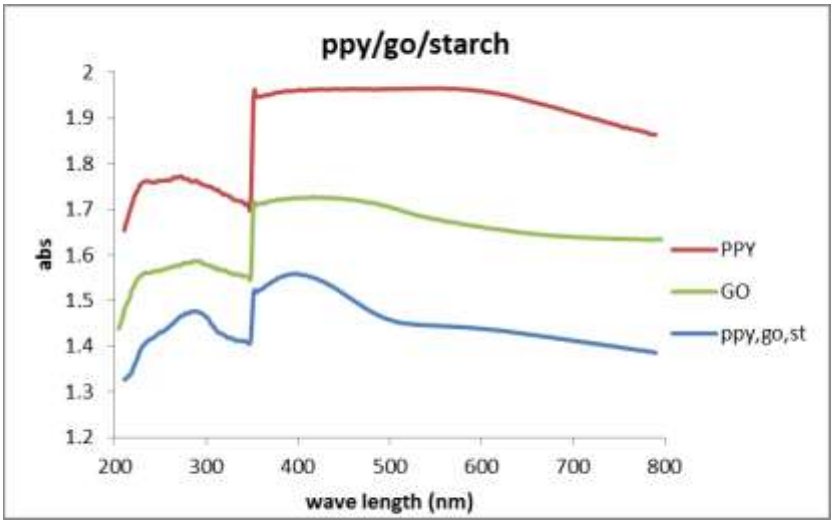
Fig; 7

From the fig; 7, we can calculate the band gap of Ppy / GO / Cellulose polymer composite using the equation Eg ≈1240/ λ

**Band gap of GO = 3.12 ev Band gap of Ppy = 3.50 eV**

**Band gap of Ppy/GO/Cellulose = 3.47 eV**

## 8. Ppy/GO/Starch composite



Fig; 8

From the Fig; 8, we can calculate the band gap of Ppy / GO / Starch polymer composite as,

**Band gap of go = 3.12 eV Band gap of Ppy = 3.50 eV**

**Band gap of Ppy/GO/Starch = 2.65**

**Ppy/GO/Sodium alginate composite**

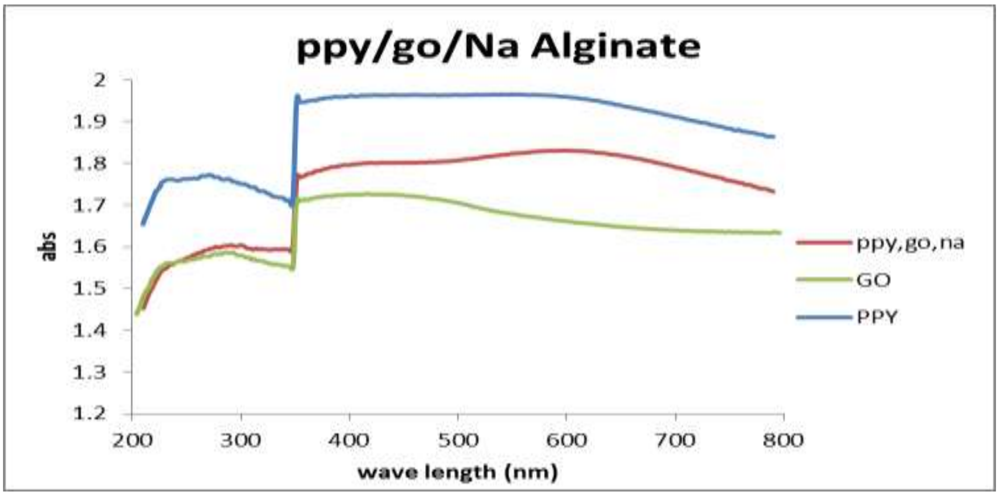


**Fig9**

From the Fig9, we can calculate the band gap of Ppy / GO / Sodium alginate polymer composite as,

**Band gap of GO = 3.12eV Band gap of Ppy = 3.50 eV**

**Band gap of Ppy/GO/Sodium alginate = 2.05 eV**

Band gap energy of Go is 3.12eV, the maximum absorption takes place at 398nm. Polypyrrole has the maximum absorption at 354nm, it band gap is 3.50eV its maximum absorption takes, place at 357nm. Ppy/Go/Starch has maximum absorption at 467nm, its band gap is 2.65eV. The band gap of Ppy/Go/Sodium alginate is 2.05eV and its maximum absorption takes place at 605nm.

From the above results it can be concluded that, the band gap energy of Ppy/GO/Sodium alginate composite is lower. So it is highly conductive than Ppy/GO and other composites. The conductivity of Ppy is much lower because its band gap energy is high. So it indicates that Polypyrrole composites are more conductive than pure Ppy.

## CONCLUSION

In this work, polypyrrole based novel bio-composites materials were successfully prepared by chemical in situ polymerization with excellent biodegradability and electrical conductivity. The preparation of polymer composites was carried out by using polypyrrole as matrix, with graphene oxide, cellulose, starch and sodium alginate incorporated as natural fillers. The materials were characterized by FTIR, SEM, UV-DRS, Four probe method and biodegradability analysis using weight loss method. The comprehensive characterization confirms that these composites not only retain enhanced electrical conductivity but also exhibit significantly improved biodegradability compared to pure polypyrrole. Among the three composites prepared, the Ppy/GO/Sodium alginate exhibited the good combination of conductive performance and environment compatibility. These findings point out the suitability of utilizing these biocomposites into sustainable electronics and ecological applications, contributing to the advancement of eco-friendly materials science.

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