Composite electrospun membranes based on PET-PAN modified with LDH-hybrid as promising adsorbent for pollutants removal from wastewater

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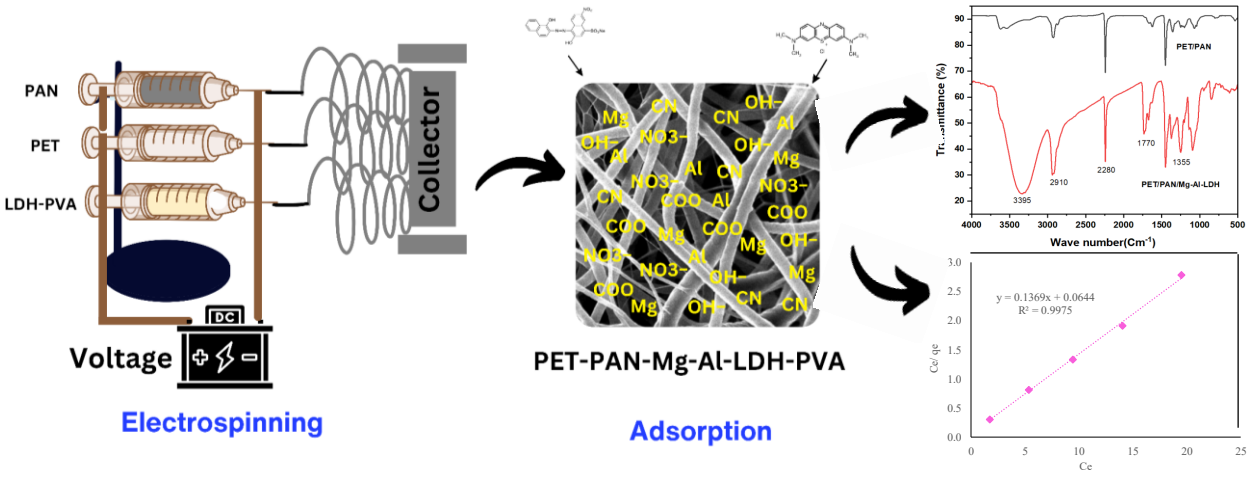
Graphical abstract



Abstract

The current work reveals the fabrication of a novel nanofiber composite membrane of PET-PAN modified with Mg-Al-LDH-PVA through electrospinning process. The nanocomposite membranes characterization was conducted with different techniques i.e. SEM, EDS, FTIR, XRD, and water contact angle to evaluate the structure and surface morphology. The optimized nanomembrane was utilized as a useful adsorbent for removal of toxic anionic dye Eriochrome Black T (EBT) and cationic dye Methylene blue (MB) from wastewater. Experimental results identified that $PPLP_3$ membrane has a potential for the removal of EBT (83%) and MB (52%) at pH 3 and 7, respectively, from aqueous solution. The optimum adsorption capacity of $PPLP_3$ nanocomposite membrane was identified and calculated as 7.3 $mg.g^{-1}$ followed by the pseudo 2^{nd} order kinetics and Langmuir adsorption isotherm fit well with R^2 values of 0.964 and 0.997, respectively. The synthesized nanocomposite membrane could be utilized for effective adsorption of contaminations from different wastewaters.

Keywords: Electrospun membranes; Nanocomposite polymers; Layered double hydroxides; adsorption; dye pollutants.

1. Introduction

Water is regarded as the most important natural resource that is found on this planet. For the survival of any living thing, water is the most required thing. Owing to having a high level of solubility, water is regarded as the universal solvent (Ahmad et al., 2015; Zhang et al., 2016). The key environmental challenge experienced by the people of the world is to maintain the quality of water. As the current level of available reserves of fresh water is insufficient and getting more and more polluted by the passage of time. Such water reserve depletion is causing an inability due to the excess consumption by the increased level of population growth, urbanization,

industrialization, agricultural and other activities (Ayandiran, Fawole, & Dahunsi, 2018; da Silva Alves, Healy, Pinto, Cadaval Jr, & Breslin, 2021; Jeong et al., 2019).

Water contamination is regarded as serious problem worldwide, which has significantly affected the health of people and the life present in the ecosystem (Anandh Babu, Hemavathi, Kousalyadevi, & Shanmuga Priya, 2023; Preethi & Jeyanthi, 2023). Polluted water becomes the main cause of various diseases in the human being which also posed challenges in the field of sciences (Picón, Vergara-Rubio, Estevez-Areco, Cerveny, & Goyanes, 2022). The various industries such as cosmetics , plastics, food, textiles, rubber, paper, and printing have released a very large quantity of wastewater which contains the significant amount of toxic indispensable dyes (Khalid, Zubair, & Ihsanullah, 2018; Radoor, Karayil, Parameswaranpillai, & Siengchin, 2020). The untreated wastewater discharge of these dyes into the water bodies give serious threats to aquatic life and human being (Manzar, Waheed, Qazi, Blaisi, & Ullah, 2019). Therefore, introducing new effective methods and materials for removing the pollutants and dyes from wastewater is drastically required (Akram et al., 2023; Bano et al., 2024; Inam et al., 2021; Kishore et al., 2023).

At present, the methods for treating dye wastewater comprise of different categories which include chemical, physical and biological processes. These methods contain several disadvantages, for example they take the high amount of energy along with higher level of costs and produce highly toxic and hazardous byproducts. Therefore, the process of adsorption is regarded as one of the most promising approach because it is easy to operate, less costly and can be recycled with ease and efficient one in comparison with the other conventional techniques (Cheng et al., 2020; Z.-P. Hu, Gao, Liu, & Yuan, 2018).

Electrospinning nanofiber membranes are considered for having capability of dealing with the industrial wastewater, because their structure is highly adjustable along with higher efficiency, porosity, and higher surface area. These attractive characteristics make adsorbents to be prepared using the electrospinning membranes (Dai, Wu, Zhang, Fu, & Li, 2018; Farooqi, Akram, Begum, Wu, & Irfan, 2021; Zhao et al., 2017; Zhu, Zheng, Zhang, & Dai, 2021). At present, many studies are bringing the usage of different homopolymers such as polyethylene terephthalate (PET) and polyacrylonitrile (PAN). In order to prepare the composite membrane modified with the nanoparticles such as layer double hydroxides (LDH) and hybrids of polyvinyl alcohol (PVA) with the purpose of increasing the adsorption removal efficiency for pollutants (Ebrahimi, Nabavi, & Omrani, 2022; Khorram, Mousavi, & Mehranbod, 2017; Mittal, 2021; Sajid, Jillani, Baig, & Alhooshani, 2022).

Previously, many researchers have used different types of polymer combination for fabricating the electrospun nanofibers membrane in order to treat the wastewater. For example, synthesis of different LDHs including MgAl, NiFe and CoAl through co-precipitation technique for the elimination of Eriochrome Black T (EBT) dye and shows adsorption of 540.91 $mg.g^{-1}$, 132.49 $mg.g^{-1}$ and 419.87 $mg.g^{-1}$, respectively (Zubair et al., 2017). PVA-Starch modified ZSM-5 zeolite nanomembrane displayed an adsorption for EBT about 2.17 $mg.g^{-1}$ (Radoor et al., 2020). The layered Ag/PDA/PS nanomembrane was fabricated and showed a complete removal of EBT pollutant from effluent (Baig et al., 2022).

The study worked on the fabrication of Fe-SCD-Mg-Al-LDH through ion exchange reaction process and by the method of co precipitation to prepare composite and it display the capability of about 83.40 $mg.g^{-1}$ for color pollutant methylene blue (MB) adsorption (W. Hu, Wu, Jiao, Yang, & Zhou, 2016). PVA-CS modified with nanoparticles $CeAlO_3$ showed the adsorption for MB about 817.81 $mg.g^{-1}$ (Shahverdi, Barati, & Bayat, 2022). The nanocomposite was fabricated with the help of LDH and activated carbon (AC) through the hydrothermal process. The prepared nanofiber mat showed exceptional attraction for the MB dyes and displayed 250.2 $mg.g^{-1}$ at PH 9. The prepared nanocomposite membrane shown higher efficiency at the room temperature about 816.0 $mg.g^{-1}$ (Aldawsari et al., 2021). PET NF-MWCNTs were prepared and estimated the adsorption for MB about 7.047 $mg.g^{-1}$ (Essa et al., 2022).

Therefore, in this study a new efficient PET-PAN modified using Mg-Al-LDH-PVA was synthesized with electrospun methodology for the removal of EBT and MB color pollutants from effluent. The objective of this research is the utilization of the developed novel membrane as an adsorbent material for the removal of the cationic and anionic dyes i.e. MB and EBT from the effluent. The effect of different operating parameters i.e., pH of solution, adsorption time, pollutant concentration, and membrane dosage were also checked. Adsorption isotherm and kinetics were performed to check the membrane efficiency. The comparison of the current study with other related studies were also performed. Additionally, the proposed adsorption mechanism for pollutant removal was suggested.

2. Methodology

2.1 Chemicals and Materials

In this research, post-consumer waste bottles of PET were used. Likewise, other polymers and chemicals were used in this study, i.e. PVA with an average molecular weight (Mw) of 9,000–10,000 (80% hydrolyzed), white powder form PAN with Mw of 150,000, ($Mg(NO_3)_2.6H_2O$) magnesium nitrate hexahydrate of 99%), ($Al(NO_3)_3.9H_2O$) aluminum nitrate nonahydrate $\geq$ 98%), sodium carbonate powder ($Na_2CO_3 \geq 99.5\%$), hydrochloric acid (HCI, 37%), sodium hydroxide pellets (NaOH), and sulfuric acid (H_2SO_4), were received from Sigma-Aldrich, Burlington, USA. Also, various other solvents and dyes namely, N, N-Dimethyl formamide (DMF), trifluoro acetic acid (TFA), dichloro methane (DCM), EBT dye, and MB dye were got from Dae-Jung, Busan, South Korea. In addition, the distilled water and analytical research grade type of chemicals were utilized for the research experiments.

2.2 Fabrication of nanoparticles (LDH)

Nanoparticles (Mg-A1-LDH) were synthesized using the coprecipitation method (Alnaqbi, Samson, & Greish, 2020; Qin et al., 2012). During nanoparticle synthesis, as precursors $Al(NO_3)_3.9H_2O$ and $Mg(NO_3)_2.6H_2O$ were used in a proportion of 1:3 (Mg:Al) and under constant pH conditions. 0.025 M of $Al(NO_3)_3.9H_2O$ and 0.075 M of $Mg(NO_3)_2.6H_2O$ of 50 mL mixed aqueous solution and then was added with 50 mL of 0.05 M of Na_2CO_3 and 1 M of NaOH with vigorous and continuous stirring (hotplate magnetic stirrer MS-H280-Pro, OniLab, CA USA). NaOH of 1 M solution was utilized to maintain the solution pH in 9.0-9.5 range. The slurry was aged for 12 h at the set temperature of 65°C. The obtained solid was then processed for centrifuge (80-2 Electronic centrifuge, Atlas medical italiano, China) and cleaned with deionized water several times and kept for drying for 12 h period in an oven (DHG-9202, SANFA, China) at a fixed temperature of 70°C.

2.3 Preparation of electrospinning polymer solution

Polymer solution of PET was prepared by cutting the post-consumer waste PET bottles into square shaped pieces of size 1x1cm^2 followed by the cleaning and rinsing with deionized water for the electrospinning process. The PET bottle's pieces were then heated in ethanol solution for a duration of 30 min at 40°C to remove the contamination. 5 wt.% homogeneous solution was prepared by dissolving the PET bottle pieces in a DCM-TFA mixture having a ratio of 3:1, followed by mixing at duration of 4 h at room temperature using magnetic stirrer.

Similarly, PAN (8 wt.%) solution in DMF was prepared with continuous stirring for 12 h at ambient temperature. PVA (8 wt.%) solution was prepared in distilled (double) water with continuous stirring for a duration of 5 h at 80ºC. Mg-Al-LDH nanoparticles were mixed in PVA (10 gm) solution at different loadings (0.8, 1.2, and 1.6%) for making the Mg-Al-LDH-hybrid solution for $PPLP_1$, $PPLP_2$, and $PPLP_3$ membranes, respectively. Later, the resulting spinnable solution was homogenized by sonicating (Ultrasonic cleaner 2L, China) for 1 h at ambient temperature.

2.4 Fabrication of PET-PAN-LDH-Hybrid membrane

As reported in our previous study, PP and PPLP nanocomposite membranes were synthesized by using electrospinning technique (Pirzada, Ali, Mallah, & Maitlo, 2023). The ready electrospun polymer solutions were transferred into syringes of 10 mL capacity with nozzle diameter of 0.5 mm fitted on top of syringe holders, as illustrated in Figure 1. PP and PPLP nanocomposite membranes were fabricated by co- electrospinning of the polymer solutions at flow rate (0.5 mL.h$^-$

[1]) and speed of drum (30 rpm), DC voltage (19 kV), and 10 cm tip distance. The aluminum covered collecting drum was used for collecting the prepared membranes. The collected membranes were subsequently kept for drying for 24 h at ambient temperature. The prepared nanocomposite membranes were peeled off by employing tweezers for subsequent use in adsorption experiments.

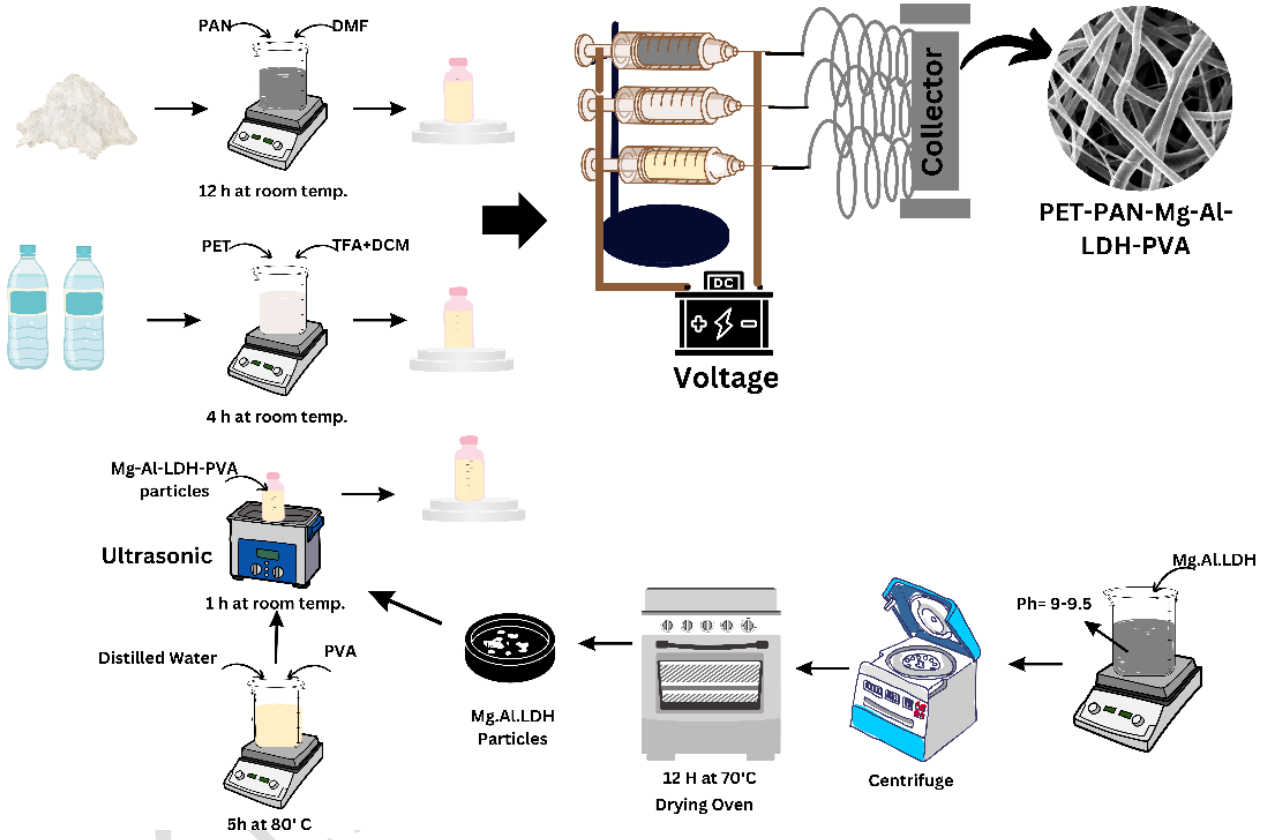


Figure 1. Schematic representation of electrospinning setup for synthesis of PET-PAN-Mg-Al-LDH-PVA nanocomposite membrane.

2.5 Characterization techniques

The physico-chemical properties of the membranes were verified with a variety of techniques. Scanning electron microscopy (SEM, JSM-IT-100, JEOL, Tokyo, Japan) analysis was applied for studying the morphology of the as prepared nanocomposite membrane. To find the average fiber

diameter of electrospun membrane ImageJ software version 1.54-d was utilized for the measurement of 50 fibers diameter distribution. The phase structure and crystal orientation were identified by using Xray diffraction (XRD, D8 Advance, Bruker, Mannheim, Germany). Energy dispersive Xray spectroscopy (EDS, JSM-IT-100, JEOL, Tokyo, Japan) was utilized to analyze membrane's elemental composition. Likewise, infrared spectroscopic analysis of chemical structure and functional groups of membrane was achieved utilizing Fourier Transform Infrared Spectroscopy (FTIR, Perkin Elmer, Waltham, USA) technique. Hydrophilic behavior of the synthesized membrane was determined by water contact angle test.

2.6 Adsorption experiments of EBT and MB

Developed membranes adsorption properties were assessed through basic experiments and were conducted in a 20 mL volume beaker containing EBT and MB aqueous solutions for each set of experiments. Optimization for the assessment of EBT, several parameters were varied one by one like adsorption time 30 ~ 120 min, pollutant concentration 10 ~ 30 $mg.L^{-1}$, membrane dosage 10 ~ 50 mg and pH 1 ~ 9. A similar method was adopted for the assessment of MB at the optimum parameters, contain pH (7.0), pollutant concentration (10 $mg.L^{-1}$), membrane dosage (30 mg) and adsorption time (120 min). The solutions pH was maintained with the help of 0.01 M HCL and 0.01 M NaOH. The membranes were taken out from the beakers after the completion of the batch experiments. EBT and MB solutions concentration were estimated by means of UV-Vis spectrophotometer (L7 dual beam, BioBase, Jinan, China) at absorbance wavelength of 530 nm and 665 nm, respectively.

The equations (1 and 2) given below were utilized for measuring the percent of adsorption capacity and removal efficiency of EBT and MB (da Silva et al., 2021; Mansor, Ali, & Abdel-Karim, 2020; Manzar et al., 2019).

$$q_e = \frac{(C_o - C_f)}{m}\ x\ V \qquad (1)$$

$$Dye\ Removal(\%) = \frac{(C_o - C_f)}{C_o}\ x\ 100 \qquad (2)$$

Here "C_o" stands for the pollutant initial concentration of EBT and MB in $mg.L^{-1}$, while "C_f " is the dye final concentration of EBT and MB in $mg.L^{-1}$, respectively. "qe" represents the quantity of EBT and MB adsorbed on the surface of membrane in ($mg.g^{-1}$). While "m" represents weight dosage of the adsorbent in (g), and 'V' denotes the volume in (mL) of pollutant solution.

2.7 Isotherms and Kinetic models

2.7.1 Adsorption Isotherms

Adsorption isotherm models i.e. Langmuir and Freundlich were applied to examine the isotherm of adsorption for the pollutants. As depicted in the model (Langmuir isotherm), the optimum adsorption level is determined by the monolayer adsorption. The surface equilibrium point shows the maximum adsorption in Langmuir isotherm model, and it presented in equation 3 as below (Guo et al., 2021; Habiba, Lee, Joo, Ang, & Afifi, 2019).

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{K_L q_m} \qquad (3)$$

In this, "C_e" indicates the mg.L^{-1} of pollutant concentration equilibrium, "q_e" is the mg.g^{-1} of adsorption capacity equilibrium. "K_L" represents the solution affinity, whereas "q_m" is the dye maximum adsorption.

The multilayer process of adsorption for a heterogenous system can be defined by the Freundlich isotherm model. This model can be written as following equation 4 (Pathirana, Dissanayake, Wanasekara, Mahltig, & Nandasiri, 2023).

$$\ln q_e = \ln(k_f) + \ln\frac{C_e}{n} \quad (4)$$

Here, "$1/n$" indicates the adsorption intensity, and "k_f" is a constant value.

2.7.2 Adsorption Kinetics

The process of adsorption kinetics can calculate the adsorbent's rate of pollutant adsorption. To know the mechanism of adsorption of the membrane, Pseudo-1st- order and 2nd- order kinetics models were utilized. Pseudo-1st- order kinetic adsorption model applies to physical adsorption process, whereas, chemical adsorption relates to pseudo-2nd- order kinetic model (He et al., 2019). The below equations 5 and 6 were used for the kinetic calculations in relation to the pseudo 1st- order and 2nd- order kinetic adsorption models, respectively (Radoor, Karayil, Jayakumar, Parameswaranpillai, & Siengchin, 2021).

$$log(q_e - q_t) = Logq - \frac{k_1}{2.303}t \quad (5)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad \text{_____(6)}$$

Whereas "q_t" represents the adsorption capacity in mg/g at time "t", "q_e" is the mg/g of the adsorbate quantity adsorbed at equilibrium. Whereas "k_1" and "k_2" are utilized for the 1st order and 2nd order adsorption rate constant.

3. Results and discussion

3.1 Membrane characterization results

3.1.1 Surface morphology results

The morphology of PP, $PPLP_1$, $PPLP_2$, and $PPLP_3$ membranes surface with different loading of Mg-Al-LDH were investigated by SEM. The morphology image of PP membrane displays the very smooth distribution of nanofibers as shown in Figure 2(a). Similarly, modified nanocomposite membranes $PPLP_1$, $PPLP_2$, and $PPLP_3$ with Mg-Al-LDH-Hybrid revealed the rough and porous structure as compared to the unmodified PP nanomembrane as depicted in Figures 2(b-d), respectively. The synthesized nanoparticles Mg-Al-LDH represent the irregular cubicles and dispersed non uniform porous agglomerates (Abdollahi, Heidari, Mohammadi, Asadi, & Tofighy, 2021; Alnaqbi et al., 2020; Qin et al., 2012). The modified membranes fiber diameter $PPLP_1$ (623 nm), $PPLP_2$ (655 nm), and $PPLP_3$(671 nm) were achieved to be greater, as compared to the PP (488 nm) as depicted in supplementary information Figure S1(a-d). The increase in fiber diameter of the modified membrane was due to the modification with LDH hybrid. The major factor that influences the enhancement in nanofiber diameter is solution viscosity. The increase in nanofiber diameter with increase in viscosity is discussed in previous studies (Bakhsh, Ahmed, Mahar, &

Khatri, 2021; Habiba, Afifi, Salleh, & Ang, 2017; Nasouri, Shoushtari, & Mojtahedi, 2015; S. Wu, Li, Shi, & Cai, 2022).

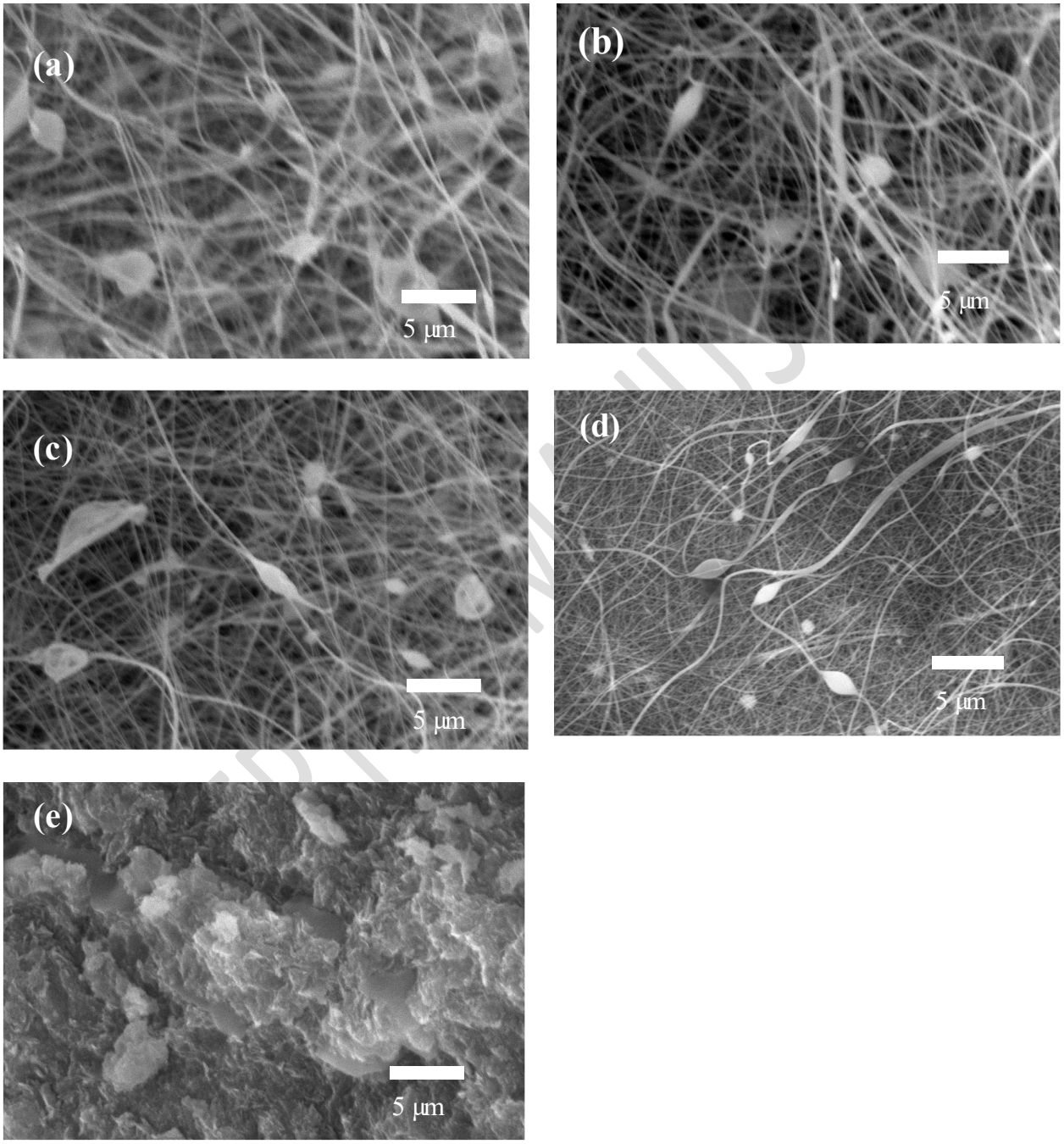


Figure 2. The surface morphology images of: (a) PP, (b) $PPLP_1$, (c) $PPLP_2$, (d) $PPLP_3$ and (e) Mg-Al-LDH nanoparticles.

3.1.2 EDS analysis results

The elemental composition of the modified ($PPLP_1$, $PPLP_2$, and $PPLP_3$) and nonmodified (PP) electrospun membranes and Mg-Al-LDH nanoparticles were identified by using the EDS technique, as depicted in Table 1 and supplementary information Figure S2. The nonmodified PP membrane contains only C, N, and O elements. While modified PPLP membranes contain Mg and Al atoms along with C, N, and O elements. According to Table 1, the elements (Mg and Al) atomic % values were higher (0.25 and 0.63) in $PPLP_3$ membrane with increasing LDH loading as compared with other membranes. The EDS spectrum displays the occurrence of elements (Mg, Al, O, C, and N) that confirms the successfully fabrication of modified membrane.

Table 1. EDS elemental composition (atomic %) of PP, $PPLP_1$, $PPLP_2$, $PPLP_3$, and Mg-Al-LDH.

Sr #	Type	Mg	Al	C	N	O
1	LDH	15.11	5.30	11.93	-	67.40
2	PP	-	-	89.49	5.14	5.37
3	$PPLP_1$	0.21	0.19	62.46	5..81	31.33
4	$PPLP_2$	0.37	0.24	55..79	5.37	38.23
5	$PPLP_3$	0.63	0.25	60.02	6.45	32.65

3.1.3 FTIR results

Figure 3(a) represents the functional groups characterization of modified ($PPLP_3$) and unmodified (PP) membranes were analysis by FTIR technique. PAN nanofibers spectra presented the characteristics peaks due to stretching vibration of –CH2– occurred at 1455 cm^{-1} and 1073 cm^{-1}.

The stretching bands of nitrile group −C≡N of PAN nanofibers revealed the characteristics peak at 2280 cm^{-1} (Hartati et al., 2022; Shakiba, Abdouss, Mazinani, & Kalaee, 2023; Xu et al., 2022). The PET peaks were occurred at 2910 cm^{-1} and 1770 cm^{-1} representing to be formed by methylene groups (-CH2-) longitudinal and oscillation of carbonyl (C = O) group, respectively (Khorram et al., 2017). The PVA spectrum, resonance band were appeared at 3395 cm^{-1} due to the presence of the –OH group and characteristics band of C-H group was observed at 2909 cm^{-1} (S. Wu et al., 2022). The developed Mg-Al-LDH nanoparticles spectrum band at 3395 cm^{-1}, were due to the existence of hydroxyl (OH) vibration stretching. Peaks at 1355 cm^{-1} and 1670 cm^{-1} occurred by the presence of NO_3^- (Novillo et al., 2014). The characteristics bands confirm the fabrication of modified and unmodified membranes.

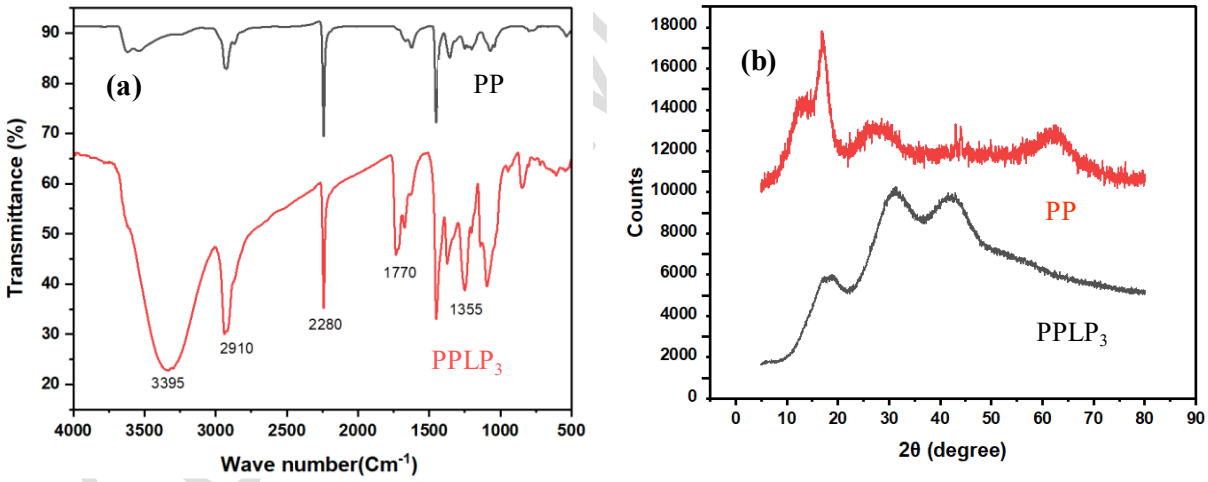


Figure 3. PP and $PPLP_3$ membranes: (a) FTIR spectrum and (b) XRD analysis.

3.1.4 XRD results

Figure 3(b) displayed the XRD analysis of PP and $PPLP_3$ membranes. The XRD pattern observed for PET displayed peak at 2θ = 17° (Aziz et al., 2021; Q. Wang, Geng, Lu, & Zhang, 2015; Yasin,

Sharaf Zeebaree, Sharaf Zeebaree, Haji Zebari, & Saeed, 2021). Whereas three sharp diffraction peaks of PAN were identified at 2θ = 18º, 22 º, and 26 º that represent to the crystalline structure (Chiu, Lin, Cheng, & Chou, 2011; Makarov et al., 2022; Ullah et al., 2019). Furthermore, major sharp peaks of Mg-Al-LDH nanoparticles were occurred at 2θ = 31º and 44^0 (Alnaqbi et al., 2020; Swain, Barik, Pradhan, & Behera, 2018). The XRD pattern for PVA was observed at peak 2θ = 19.5º (Qin et al., 2012). The result of XRD analysis of PET, PAN, LDH and PVA showed the successful synthesis of PPLP membrane.

3.1.5 Hydrophilicity measurement

The wettability performance of PP and $PPLP_3$ nanocomposite membranes was measured by water contact angle technique. As shown in Figure 4, the water contact angle of $PPLP_3$ membrane (19º) is lower than the PP membrane (81º). The reduction in contact angle values of $PPLP_3$ electrospun membrane showed better hydrophilicity properties as compared to the PP membrane. Increased in membrane hydrophilicity may be due to the cause of surface roughness (Jiang, Zhao, & Zhai, 2004; X. Li, Ding, Lin, Yu, & Sun, 2009; Skornyakov & Komar, 1998; N. Wang, Zhao, & Jiang, 2008). The wettability of the $PPLP_3$ membrane increased may also be due to the addition of LDH-hybrid (Jia, Liang, & Yang, 2021; G. Li, Zhao, Lv, Shi, & Cao, 2013). The decrease in water contact angle will help to enhance the adsorption of pollutant removal from wastewater.

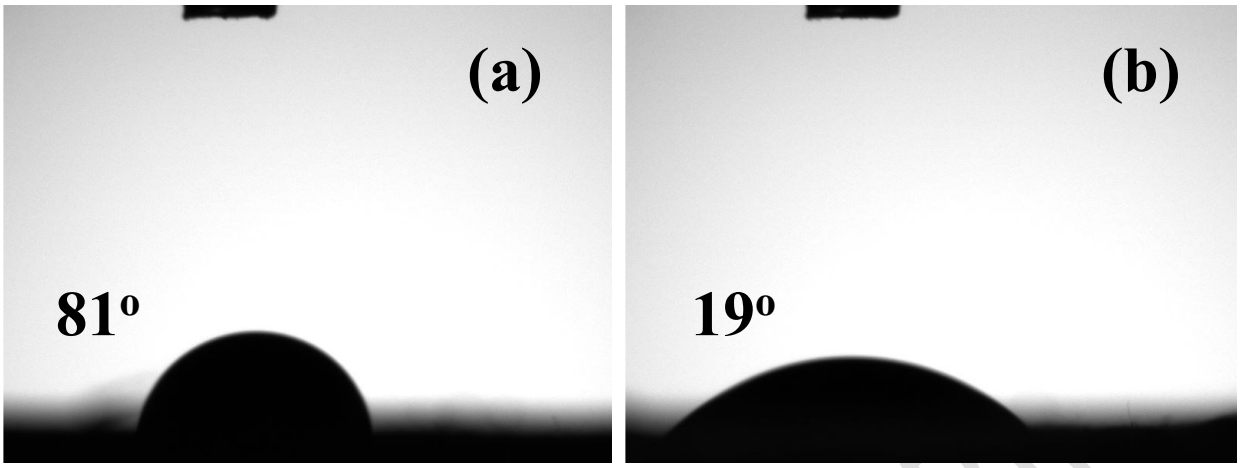


Figure 4. Hydrophilicity analysis using water contact angle test: (a) PP and (b) $PPLP_3$ membranes.

3.2 Initial adsorption experiment results

Firstly, basic experiments were achieved to examine the adsorption efficiency of the modified $PPLP_1$, $PPLP_2$, and $PPLP_3$ membranes and nonmodified PP membranes for EBT and MB dyes. $PPLP_3$ membrane removal efficiency for pollutants EBT (83%) and MB (52%) is higher than that of other membranes as shown in supplementary information Figure S3. The increased removal efficiency of pollutants occurred through the surface phenomena, wettability, and the existence of the functional groups on the surface of membrane (C–N, C–O, –OH, etc.). The $PPLP_3$ membrane was used to perform additional experiment trials for kinetic and adsorption isotherm models. The parameters to check the adsorption efficiency of MB and EBT are shown in supplementary information Table S1.

3.3 Optimization of adsorption experiment

3.3.1 pH

A solution's pH significantly affects the adsorption process as it influences both efficiency and behavior of adsorption method. The minor variation in the solution's initial pH could affect the

removal efficiency, while it also influencing the solution's properties and the membrane surface charge (Anah & Astrini, 2017). The experimental design for the assessment of EBT adsorption on the surface of membrane was performed with the variation in pH 1 ~ 9 keeping initial pollutant concentration (10 mg.L^{-1}), adsorption time (90 min), and membrane dose (30 mg). It was observed that at pH 3 the EBT adsorption shows maximum removal efficiency 83% as presented in Figure 5(a). From the experimental results it was identified that when the solution pH was increased, the removal efficiency of EBT decreases. At pH 3, maximum adsorption performance obtained may be associated to the surface characteristics of the electrospun membrane (Radoor et al., 2021; Xu et al., 2022).

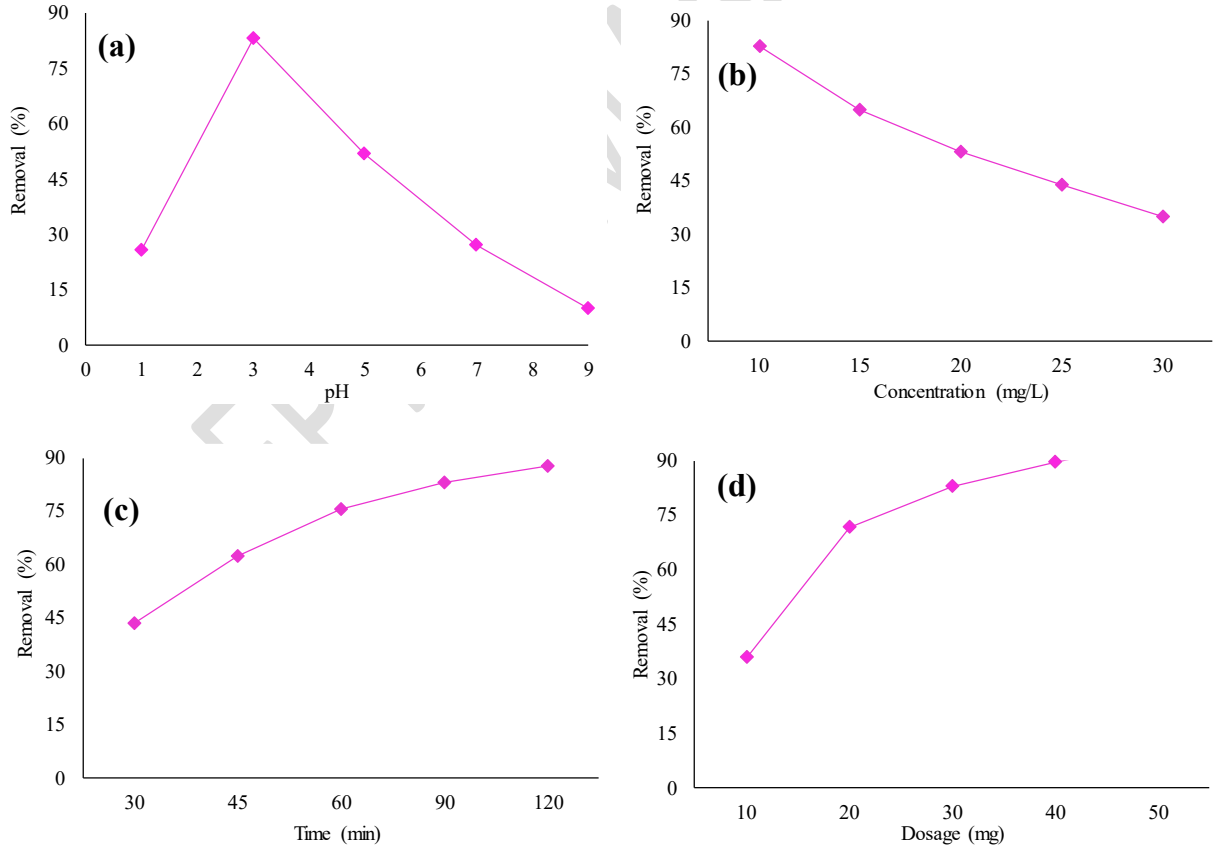


Figure 5. Effect of: (a) solution pH, (b) pollutant concentration, (c) adsorption time, and (d) membrane dosage on EBT removal using $PPLP_3$ membrane.

3.3.2 Initial dye concentration

In order to examine impact on the adsorption of EBT, the pollutant concentration was adjusted in between 10 to 30 $mg.L^{-1}$. With the gradual rise in the pollutant concentration, there was a drop in the efficacy of EBT adsorption; Figure 5(b) shows that the optimum removal performance of EBT was at 10 $mg.L^{-1}$. Higher pollutant concentration leads to decrease in percentage of dye elimination at the same adsorption time (Khan et al., 2021; Y. Wu et al., 2013).

3.3.3 Contact time

Several tests were carried out to obtain maximum equilibrium time that tends to offer the optimum EBT adsorption. The adsorption efficiency of EBT at varying contact times (30 ~ 120 min) are shown in Figure 5(c). Because of the existence of adsorption functional groups and active sites on the $PPLP_3$ membrane surface, the adsorption efficiency values for EBT increased initially. Later, a gradual trend was noticed till it achieves equilibrium condition which was attained after 90 min. It has been noted that the active sites on the membrane surface of every polymer has a separate equilibrium duration due to the active functional groups (Chaukura, Murimba, & Gwenzi, 2017; Y. Wu et al., 2013).

3.3.4 Membrane dosage

The dosage of the adsorbent is also one of the significant factors that influence the membrane materials adsorption efficiency. Varying in the adsorbent dosage of $PPLP_3$ from 10 ~ 50 mg the

efficiency of the removal of EBT was analyzed. It was observed that the increase in the membrane dosage results in an improved pollutant adsorption by the membrane (Bensalah, Younssi, Ouammou, Gurlo, & Bekheet, 2020). Therefore, the result shows the enhanced adsorption process is due to the increase of active adsorption sites on the $PPLP_3$ membrane surface, which is because of the increase in membrane dosage (Eltaweil, Mohamed, Abd El-Monaem, & El-Subruiti, 2020). As shown in Figure 5(d), the maximum 95.4% of EBT removal was achieved with a $PPLP_3$ dosage of 50 mg. For further experiments the optimum $PPLP_3$ dosage was selected as 30 mg.

3.4 Adsorption Isotherms

The isotherms models i.e. Freundlich and Langmuir were utilized to explore the $PPLP_3$ membrane's adsorption capacity. An adsorption isotherm analysis was achieved to identify the functions of the membrane quantity attached to the membrane's surface (Al-Sou'od, 2012). Figure 6(a-b) and Table 2 present a relationship of the adsorption parameters derived from the Freundlich and Langmuir isotherms model for the adsorption of EBT on $PPLP_3$ nanomembrane surface. For dye adsorption on $PPLP_3$ the Langmuir isotherm was observed suitable with R^2 value of 0.997. According to Table 2, the Langmuir isotherm model was observed to be sufficiently applied to the membrane adsorption process and is suitable to $PPLP_3$ membrane having an optimum EBT adsorption capacity of 7.3 $mg.g^{-1}$.

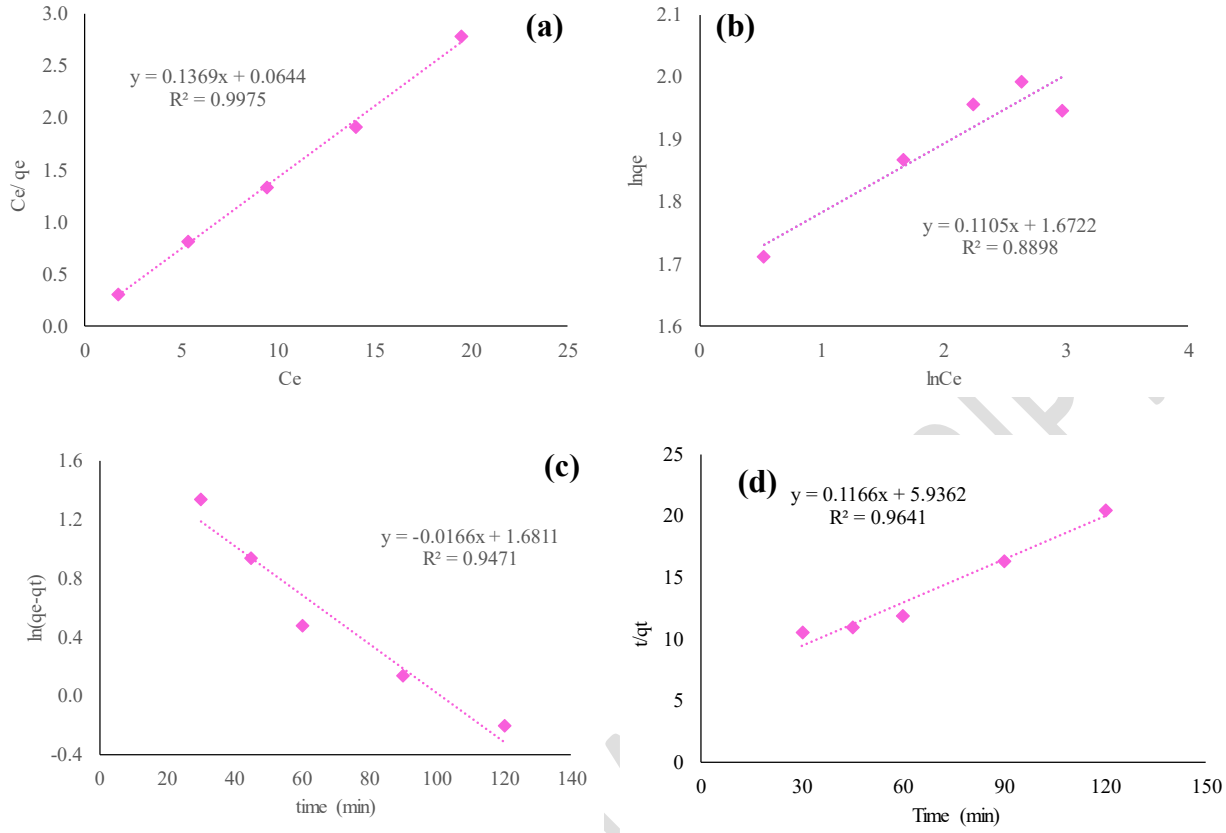


Figure 6. Adsorption isotherm models for EBT: (a) Langmuir and (b) Freundlich. Adsorption kinetic curves of EBT: (c) pseudo-1st order and (d) pseudo-2nd order.

Table 2. Parameters for Langmuir and Freundlich adsorption models, pseudo-1st order and pseudo-2nd order kinetic models.

Langmuir Isotherm			Freundlich isotherm		
q_{max} ($mg.g^{-1}$)	KL ($L.mg^{-1}$)	R^2	KF ($mg.g^{-1}$)($L.mg^{-1}$)1/n	1/n	R^2
7.3	2.126	0.997	5.3	0.1	0.889
Pseudo-1st order			Pseudo-2nd order		

q_e (mg.g^{-1})	k_1 (1/h)	R^2	q_e (mg.g^{-1})	k_2 (g.mg^{-1}.h^{-1})	R^2
5.37	0.017	0.947	8.58	0.002	0.964

3.5 Adsorption Kinetics

The obtained experimental data were fitted into various kinetic models to examine the adsorption mechanism and adsorption rate constant. As shown in Table 2 and Fig. 6(c-d), the pseudo-1st and pseudo-2nd order model parameters were identified, and the corresponding R^2 values were found to be 0.947 and 0.964, respectively. The pseudo-2nd order model gives a well explanation of the EBT adsorption mechanism as compared to the pseudo-1st order model. The pseudo-2nd order model is fit well in the current research, as it is effective at managing the EBT adsorption onto the $PPLP_3$ membrane surface. This also provided that the adsorption mechanism is done through the process of chemisorption process (Konicki, Aleksandrzak, Moszyński, & Mijowska, 2017; L. Li, Luo, Li, Duan, & Wang, 2014).

3.6 Proposed PPLP adsorption mechanism

The adsorption mechanism is primarily caused by ions or liquid molecules adhering on the PPLP membrane surface. It happens because of the reason of force of attraction in between the adsorbent and adsorbate (Konicki et al., 2017; L. Li et al., 2014). Figure 7 discusses the suggested mechanism process of adsorption in between EBT, MB and the PPLP membrane. It was evident from characterization results that there are different functional groups present in the PPLP membrane. Due to electrostatic force of attraction or hydrogen bond, the dye EBT having N=O functional group is adsorbed on the PPLP membrane (Dhar et al., 2022; Zubair et al., 2017). Similarly, hydroxyl ions on the PPLP membrane is most likely to participate in MB adsorption through

electrostatic force of attraction (Dhar et al., 2021; Pan et al., 2020). Table 3 displays the comparison performance of $PPLH_3$ membrane with other related studies, which shows that this membrane can remove pollutants efficiently.

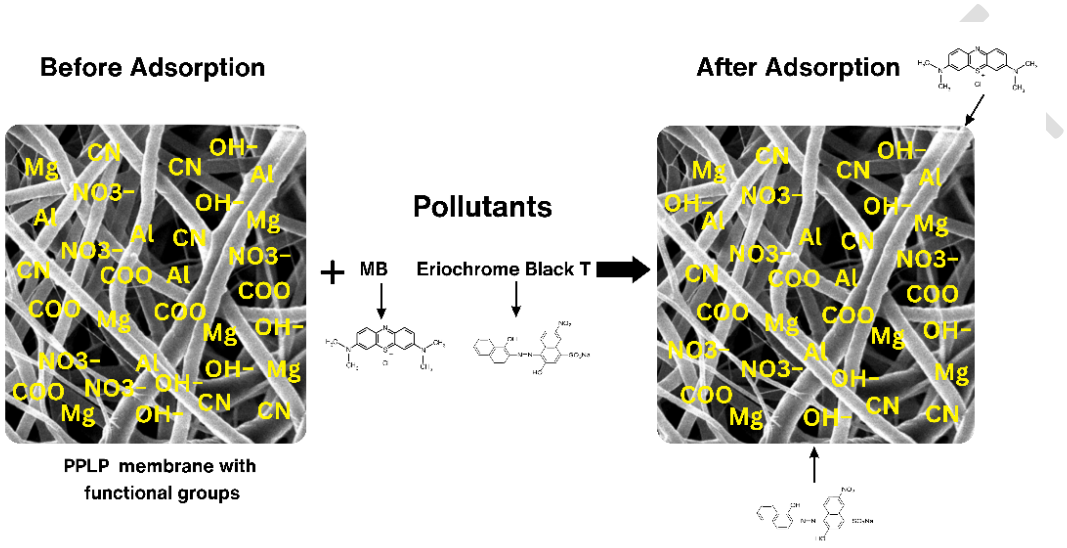


Figure 7. Proposed adsorption mechanism of MB and EBT dyes for PPLP membrane.

Table 3. Summary of comparative studies of the current research with others related research.

Sr No.	Adsorbent type	Target pollutant	Solution pH	Adsorption time (min)	q_{max} ($mg.g^{-1}$)	Reference
1	PET-PAN-Mg-AI-LDH-PVA	EBT	3	90	7.30	This study
2	Polyvinyl Alcohol/Starch/ZSM-5 Zeolite	EBT	3	180	2.17	(Radoor et al., 2020)

3	CTAB-PA	EBT	4	--	89.93	(Ben Arfi, Karoui, Mougin, & Ghorbal, 2019)
4	PET NF-MWCNTs	MB	8	120	7.05	(Essa et al., 2022)
5	OMWCNT-K-Carrageenan-Fe_3O_4	MB	--	360	1.24	(Duman, Tunç, Polat, & Bozoğlan, 2016)

4. Conclusions

The nanocomposite membrane was successfully synthesized using PET-PAN modified with Mg-AI-LDH hybrid with the electrospinning technique, and was confirmed by SEM, EDS, FTIR, and XRD results. From SEM results it was observed that the membrane diameter was increased with addition of LDH hybrid. The addition of different concentration of Mg-Al-LDH, the PPLP membrane's hydrophilicity was improved with notable adsorption efficiency of 83% for EBT and 52% for MB, respectively. Adsorption of MB and EBT on the surface of $PPLP_3$ membrane is pH dependent. The adsorption of PPLP membrane's follows the pseudo-2nd-order kinetics model and the Langmuir isotherm adsorption model, with R^2 values of 0.964 and 0.997, respectively. The adsorption mechanism suggested that the pollutants were removed due to the presence of functional groups on PPLP membrane surface. Results show that $PPLP_3$ has good adsorptive properties for a variety of pollutants from wastewater, therefore, it could be used as a potential adsorbent in the future.

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Conflicts of Interest: There are no conflicts of interest to declare.

Abbreviation	Details
PET	Polyethylene terephthalate
PAN	Polyacrylonitrile
LDH	Layer double hydroxides
PVA	Polyvinyl alcohol
EBT	Eriochrome Black T
MB	Methylene blue
Mw	Molecular weight
$Mg(NO_3)_2.6H_2O$	Magnesium nitrate hexahydrate
$Al(NO_3)_3.9H_2O$	Aluminum nitrate nonahydrate
Na_2CO_3	Sodium carbonate
HCI	Hydrochloric acid
NaOH	Sodium hydroxide pellets
H_2SO_4	Sulfuric acid
DMF	N, N-Dimethyl formamide
TFA	Trifluoro acetic acid
DCM	Dichloro methane
SEM	Scanning electron microscopy
XRD	Xray diffraction
EDS	Energy dispersive Xray spectroscopy
FTIR	Fourier Transform Infrared Spectroscopy
PP	PET-PAN
PPLP	PET-PAN-Mg-Al-LDH-PVA

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Supplementary information

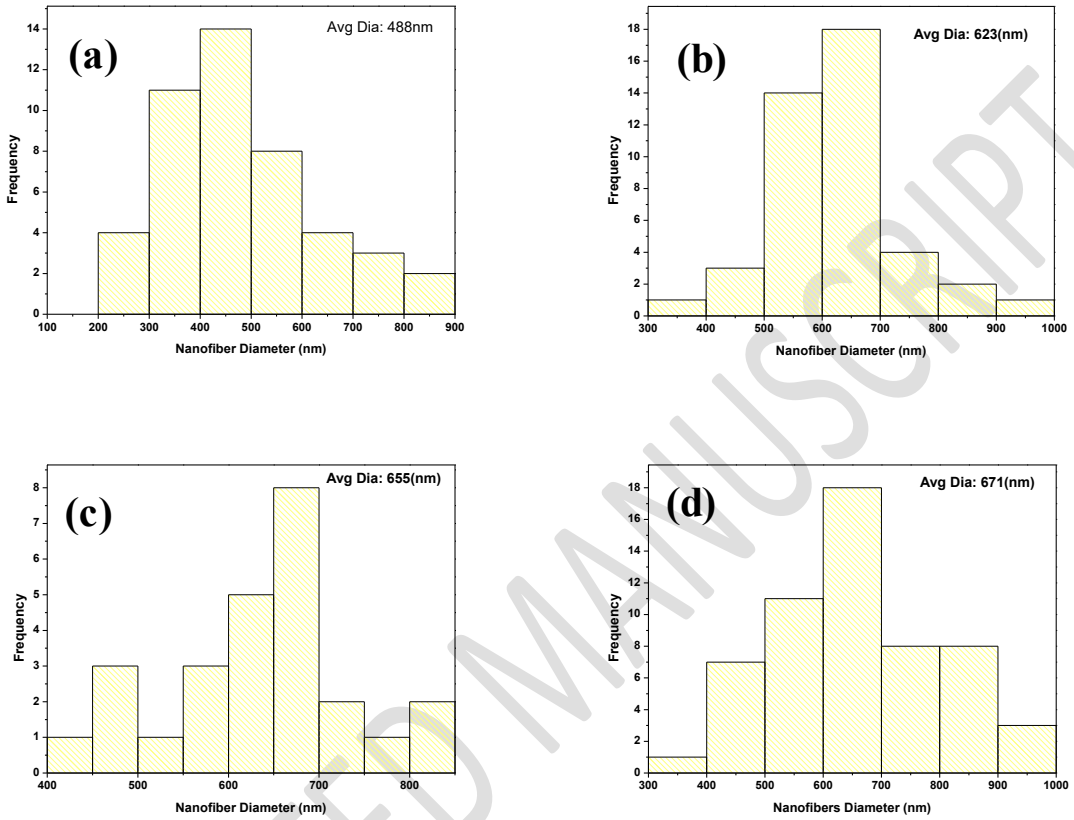

Figure S1. The nanofiber diameter histograms of: (a) PP, (b) $PPLP_1$, (c) $PPLP_2$, and (d) $PPLP_3$ membranes.

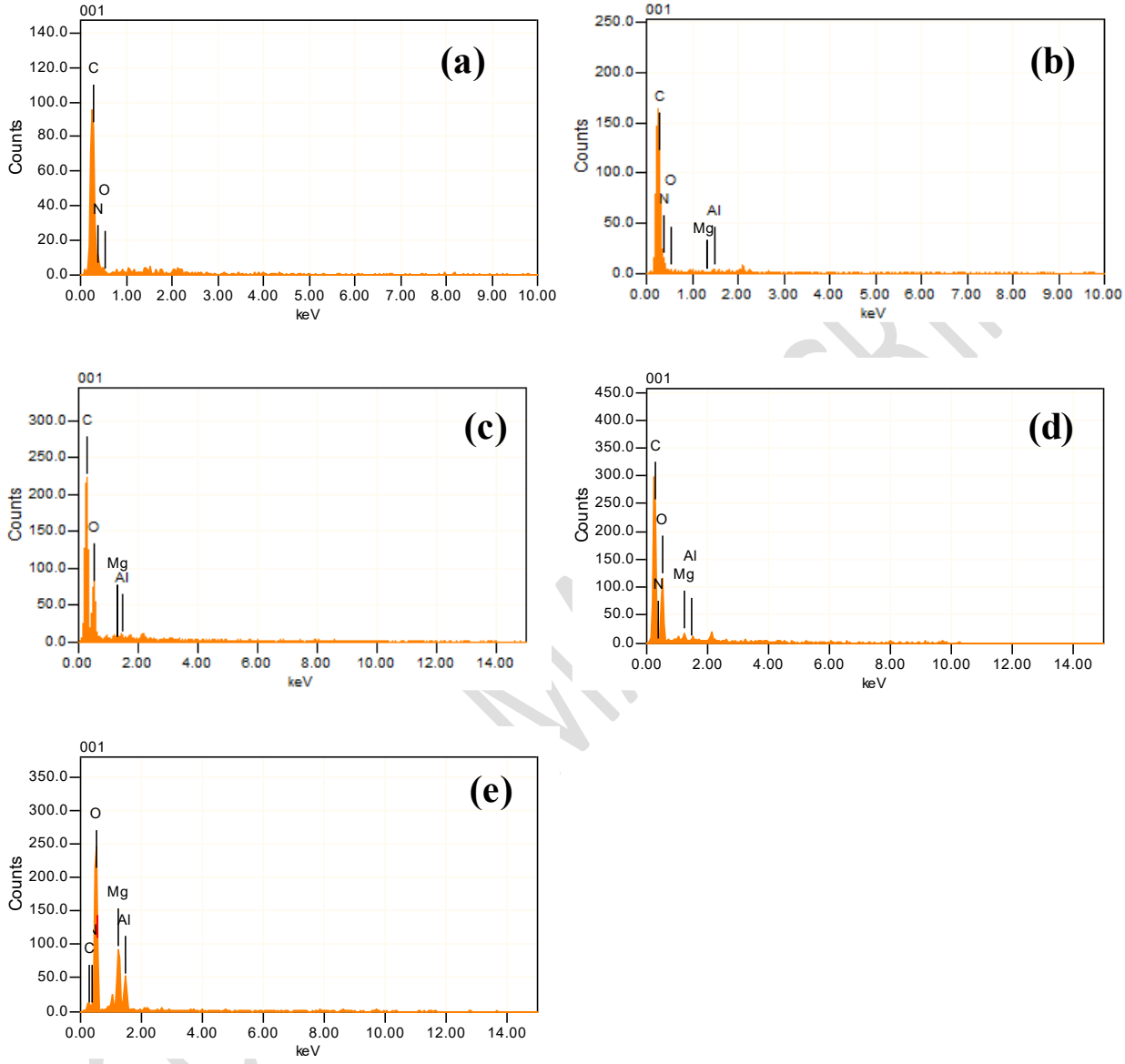

Figure S2. EDS spectrum of: (a) PP, (b) $PPLP_1$, (c) $PPLP_2$, (d) $PPLP_3$, and (e) Mg-Al-LDH.

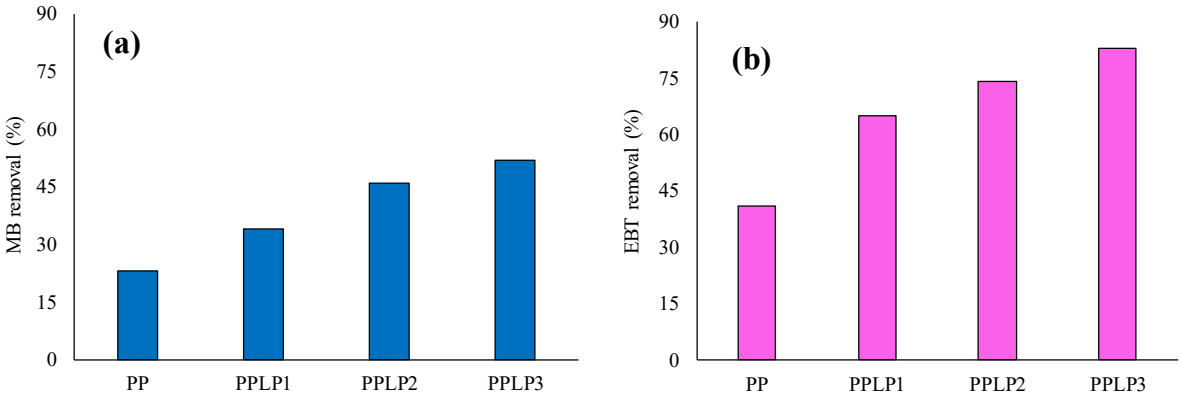

Figure S3. Basic adsorption experiment results for removal of: (a) MB and (b) EBT dyes.

Table S1. Basic adsorption experimental conditions for MB and EBT using PP and PPLP membranes.

Membrane type	Pollutant type	Solution pH	Adsorption time (min)	Concentration ($mg.L^{-1}$)	Membrane dosage (mg)	Solution volume (mL)
PP $PPLP_1$ $PPLP_2$ $PPLP_3$	MB	7	120	10	30	20
PP $PPLP_1$ $PPLP_2$ $PPLP_3$	EBT	3	90	10	30	20