

# Green Innovation in Environmental Remediation: J10-PET Thin Films for Efficient Removal of Methylene Blue and Methyl Orange Contaminants

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## Abstract

This study introduces an innovative approach employing N-(3-benzylureido)(methyl)-2-(6-methoxynaphthalen-2-yl)propanamide (J10) as an additive for recycled polyethylene terephthalate (PET) to produce thin film (J10-PET thin film), with a focus on their application in the removal of methylene blue (MB) and methyl orange (MO) from aqueous solutions. The study is primarily focused on unraveling the kinetics and equilibrium behaviors governing the removal of MB and MO. The investigation includes the determination of the equilibrium adsorption capacities ( $Q_e$ ) of MB and MO at different temperatures (308, 323, and 333 K) and concentrations (5, 10, and 15 mg/g). Remarkably, the pseudo-second-order model is found to best elucidate the adsorption kinetics for both MB and MO. Notably, the J10-PET thin film exhibits promising results with an activation energy of 14.42 kJ/mol for MB and 36.08 kJ/mol for MO, indicating its potential for effective pollutant removal. This research contributes to a comprehensive understanding of adsorption processes and highlights the J10-PET thin film as a promising solution for addressing MB and MO pollutants in aqueous environments.

**Keywords:** Adsorption, Expired pharmaceuticals, Kinetics, Methylene blue (MB), Methyl orange (MO), PET solid waste, Thin film, Wastewater treatment.

## Introduction

Water contamination is a pressing global issue exacerbated by industrialization and population growth, driven by the increasing quantity of wastewater released into water systems<sup>1</sup>. Synthetic dyes, extensively used in industries such as textiles, food, printing, and pharmaceuticals, are among the leading contributors to water pollution<sup>2</sup>.

Dyes are organic chemicals used in various industries, including textiles, paper, food, printing, plastics, beverages, leather, and pharmaceuticals<sup>3,4</sup>. Approximately 0.8 million tonnes of organic dyes, including over 10,000 different types, are produced

annually<sup>5</sup>, with alarming statistics indicating that approximately 15% of these dyes are lost during various processing activities, releasing toxic organic substances into water systems<sup>2,6,7</sup>. A significant portion of these dyes consists of anionic (acidic) and cationic (basic) dyes<sup>8</sup>, with azo dyes, containing nitrogen-nitrogen double bonds<sup>9</sup>, being the most comprehensive<sup>10</sup>, and a major environmental concern due to their carcinogenic metabolites.<sup>11-13</sup>

MB is a common type of cationic thiazine dye used for dyeing wood, cotton, silk, and leather; also used in medicine, as an antiseptic, and in the sciences

(chemistry, biology) laboratories<sup>14</sup>. Because MB is so poisonous, its presence in water can do much damage to the human body, producing conditions such as nausea, anemia, dizziness, jaundice, and chest pain<sup>15</sup>, Long-lasting harm to the eyes and skin can occur, along with the possibility of causing negative impacts on living organisms. These can manifest as symptoms like confusion, profuse sweating when inhaled through water, digestive issues such as diarrhea<sup>16</sup>, and vomiting, as well as discomfort in the mouth, throat, and stomach leading to feelings of nausea.<sup>8, 17</sup>

Similarly, MO an azo dye that can dissolve in water, is often found in the wastewater discharged by various industries such as textiles, food processing, pharmaceuticals, printing, and paper manufacturing,<sup>18,19</sup> this dye is among the most commonly encountered pollutants in water systems due to the toxicity and persistence the discharges can cause severe threats to the physicochemical properties of fresh water and aquatic life. Thus, removing MB and MO from aqueous solutions is essential<sup>20</sup>. Removing MO from wastewater poses challenges due to its complex aromatic ring structure, resistance to degradation, xenobiotic properties, high visibility under strong light, stability, and resistance to oxidation.<sup>21</sup>

In recent years, researchers have dedicated significant efforts to test and advance various techniques for eliminating dyes from wastewater. These methods include coagulation, chemical oxidation, biodegradation, ultrasonic degradation, photodegradation, membrane separation, and adsorption.<sup>1,22</sup>

However, many of these methods come with drawbacks, such as high costs and the generation of secondary waste products. Among these techniques, adsorption stands out as the most suitable method<sup>23</sup> due to its cost-effectiveness, flexibility, and ability to efficiently remove a wide range of pollutants without producing hazardous chemicals.<sup>24,25</sup>

Plastic items have become vital to our daily lives in recent decades. PET is the most widely used polymer in food, medicine, and soft drink bottle and container manufacturing due to its lightweight, low cost, transparency, sound insulation, Outstanding tensile properties, resistance to chemicals, ease of use, and flexibility;<sup>26,27</sup> Consequently, there has been a notable rise in the generation of PET solid waste every year, resulting in significant ecological

concerns<sup>28</sup>. Due to its non-biodegradability, inappropriate mechanical qualities, thermal stability, and reduced electrical conductivity, PET is the most abundant municipal and industrial waste but has no beneficial purpose<sup>29</sup>. Thus, PET waste disposal is a global issue, especially in developing nations. Incineration and landfilling are prominent PET waste management methods<sup>30</sup>; however, these technologies may harm the environment by emitting heat, volatile chemicals, and landfill contamination. The chemical recycling of PET has many processes. The main obstacles to PET chemical recycling are scale-up, cost, and organic solvent handling.<sup>31</sup>

Many researchers are interested in developing creative, affordable, and ecologically acceptable adsorbent compounds from PET bottle waste, including activated carbon,<sup>32,33</sup> Recycled PET, Fibres Modified with Graphene Oxide<sup>34</sup> modified (phenol-PET)<sup>35</sup> and PET Nanofibrous membranes<sup>36</sup> for the removal of pollutants from wastewater.

The increase in the production and consumption of drugs has led to an exponential rise in the improper disposal of drugs.<sup>37,38</sup> Expiring medications are complicated chemicals that can pollute water, soil, and animals. Additionally, outdated drugs may be hazardous or ineffective<sup>39</sup>.

Based on a study by Chinese researchers, 20 tonnes of expired pharmaceuticals and 724.5 tonnes of disinfection products were destroyed. Drug expiration impacts health issues and hurts the government due to the loss of funds allotted for the health sector. In a study conducted in Africa, particularly in East Africa, the amount of money lost due to expired medications was unexpected.<sup>40</sup>

Recent studies investigated ways to recycle active pharmaceutical ingredients (API) from pharmaceuticals. For instance, Pratama et al. used solid-liquid extraction to recover API from expired paracetamol, tetracycline, and ibuprofen. The study found that the optimal solvent composition can retain highly pure API with recovery yields of 58.7%, 73.1%, and 67.6% for paracetamol, tetracycline, and ibuprofen when removing the most excipients<sup>41</sup>

The objective of this study is to investigate the adsorption capabilities of MB and MO on J10- PET thin film. The J10 compound was synthesized using expired naproxen drug and PET from discarded water bottles. The thin film was created by mixing and dissolving a mixture of compound J10 with PET

and casting it into the desired thin film shapes that were used as an effective adsorbent to remove methyl blue and methyl orange from wastewater.

## Materials and Methods

### Chemicals

N-(isocyanatomethyl)-2-(6-methoxynaphthalen-2-yl)propanamide; Bilim Pharmaceutical; benzylamine from (Sigma-Aldrich); Toluene from (Uni-Chem); diethyl ether from (Biosolve B.V); The polymer (PET) waste material is sourced from a (local water packaging company), initially undergoing washing and removal of non-PET components, followed by a drying process. All chemicals utilized in the study are of high-quality analytical reagent grade. Trifluoroacetic acid (TFA) is acquired from (ROTH), dichloromethane (DCM) from (Uni-Chem), while methyl orange (MO) and methylene blue (MB) are obtained from (Sigma-Aldrich). sodium hydroxide (NaOH) and hydrochloric acid (HCl) were obtained from Scharlau, ensuring the use of high-purity chemicals from a reputable supplier for all experimental procedures.

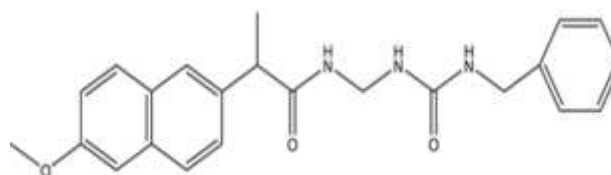
### Experimental

#### Preparation and characterization of N-((3-benzylureido)methyl)-2-(6-methoxynaphthalen-2-yl)propanamide (J10)

A mixture containing N-(isocyanatomethyl)-2-(6-methoxynaphthalen-2-yl) propanamide (2.84g, 0.01mole) and benzylamine (1.07g, 0.01mole) was exposed to reflux in dry toluene (25 ml) for 72 hours on a sand bath. After the reflux period, the surplus toluene solvent was eliminated under reduced pressure, yielding a residue, and diethyl ether was used to wash the residue. The resulting solid product was subjected to a recrystallization procedure. The recrystallization was executed using a solvent mixture composed of ethanol and water. Fig. 1 shows the structure of J10.

Yield: 91%, mp: 98-100 °C; brown solid; eluent:(hexane: ethyl acetate 1:1) R<sub>f</sub>=0.21; <sup>1</sup>H NMR (CHCl<sub>3</sub>-d) δ (ppm): 1.50-1.56 (d, J=8 Hz, 3H) CHCH<sub>3</sub>; 3.66-3.75 (q, J=8 Hz, 1H) CHCH<sub>3</sub>; 3.85 (s, 2H) NHCH<sub>2</sub>Ar; 3.90 (s, 3H) OCH<sub>3</sub>; 4.30 (s, 2H) NHCH<sub>2</sub>NH; 7.09 -7.83 (m, 11H) Ar-H; 6.60, 6.89, 8.40 (b, 3H) 3NH; <sup>13</sup>C-NMR (CHCl<sub>3</sub>-d) δ (ppm): 18.27 CHCH<sub>3</sub>; 43.36 CHCH<sub>3</sub>; 46.61 NHCH<sub>2</sub>NH; 55.30 OCH<sub>3</sub>; 66.04 NHCH<sub>2</sub>Ar; 105.58, 119.19, 125.93, 126.99, 127.43, 127.57, 127.97, 128.60,

128.90, 129.22, 133.74, 137.75, 139.21, 157.70 (16C) Ar-C; 168.81, 175.11 (2C=O); IR (KBr), ν (cm<sup>-1</sup>): 3306, 3200, 1630, 1605, 1264.



**Figure 1. Structure of N-((3-benzylureido)methyl)-2-(6-methoxynaphthalen-2-yl)propanamide (J10)**

#### Preparation of polymer Solution

A PET polymer solution is created by dissolving 0.28g of PET and 0.06g of J10 in a solvent mixture composed of DCM and TFA (in a ratio of 3:1). Initially, PET is ground using a common household grinder and then sifted through a test sieve (with a mesh size of 250 μm). The PET solution is stirred using a magnetic stirrer at room temperature for the duration of four hours to enhance the polyethylene's solubility<sup>42</sup>.

#### Preparation of J10- PET thin film

The polymer solution is transformed into a thin film through a casting process. The prepared polymer solution is initially spread onto a substrate, forming a layer. The solvent is subsequently eliminated through evaporation, forming a solid film adhering to the substrate. This cast layer is then separated from the substrate, yielding an independent thin film. This procedure successfully produces a thin film with a thickness ranging from 27 to 36 micrometers. To facilitate subsequent utilization, the thin film is divided into pieces weighing 0.01 grams.

The morphologies between the PET thin film and the J10-PET thin film are compared using a field emission scanning electron microscope (TESCAN), Fourier transforms infrared spectra (FTIR) (Shimadzu), and the measurements of the thermal gravimetric analysis (DSC-TGA) are attested with (Q600).

#### Preparation of Solutions

To prepare the MB and MO stock solutions at a concentration of 1000 mg/L, 0.01 g of MB and MO were individually dissolved in 100 ml of deionized water, resulting in 100 mg/L stock solutions. Workable solutions with concentrations of 3, 5, 7, 9, 11, and 13 mg/L for both MB and MO were obtained by dilution with water.

For the creation of a 0.1 mol/L HCl stock solution, 2.1 ml of concentrated HCl (12.07 mol/L) was diluted with 250 ml of distilled water, yielding the desired concentration. Working solutions were subsequently prepared through further dilutions with water.

In a 250 ml volumetric flask, a 0.4 g quantity of NaOH was dissolved in distilled water to produce a stock solution with a concentration of 1.0 mol/L NaOH.

### Batch Adsorption Studies of MB and MO

The adsorption of MB and MO is conducted in a batch process by employing different doses of J10-PET thin film adsorptive, variable concentrations of MB and MO, and adjusting the pH and temperature.

A measured quantity of the dehydrated J10-PET thin film is combined with a 10 ml solution of either MB or MO, with concentrations that vary. The solution is agitated in a temperature-controlled water bath for 30 minutes. The concentrations of MB and MO are determined using a spectrophotometer of the Janeway 7315 type after the adsorption process. These concentrations are obtained by referencing a calibration curve with a contrast measurement.

### Impact of Contact Time and Temperature on Adsorption

## Results and Discussion

Fig. 2 depicts scanning electron microscopy (SEM) images showcasing the morphological features of both the PET thin film and the J10-PET thin film. In Fig. 2A, the PET thin film exhibits a uniform and smooth surface. In contrast, Fig. 2B illustrates the J10-PET thin film, which displays agglomerates comprising grains of varying sizes. This data compellingly demonstrates the interaction between the J10 chemical and the PET particles, leading to the noticeable formation of particle clusters.

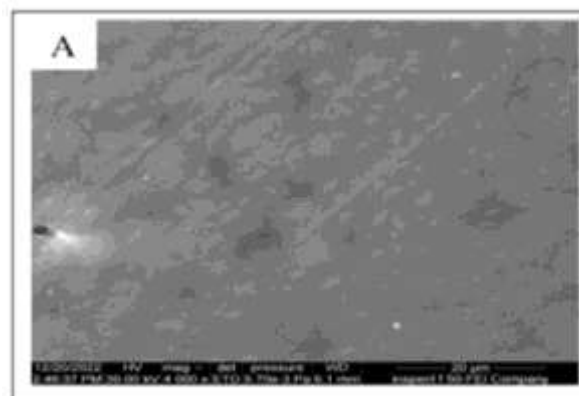
The influence of temperature on the adsorption process of MB and MO onto the J10-PET thin film was explored. This was achieved by mixing 5 ml of MB or MO solution (with concentrations of 5 mg/L at pH 12 for MB and pH 3 for MO) with 0.01g of the J10-PET thin film. The experiments were conducted at various temperatures (308, 323, and 333 K) while agitating the mixture at 150 rpm for varying durations (ranging from 5 to 140 minutes).

### Impact of Contact Time and (MB and MO) Concentrations on Adsorption

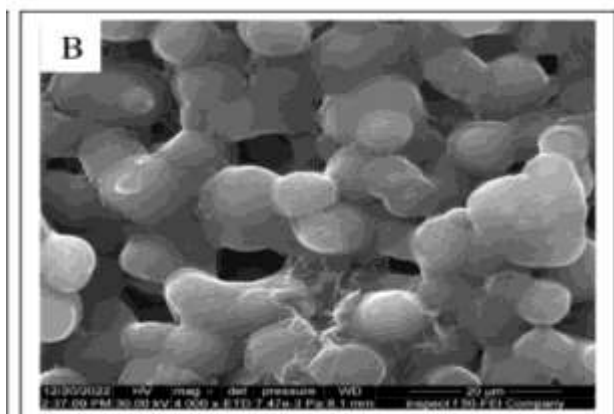
The impact of initial concentration on the adsorption of MB and MO onto the J10-PET thin film was assessed. This involved combining 5 ml of solutions with different initial MB or MO concentrations (5, 10, and 15 mg/L) at pH 12 for MB and pH 3 for MO, with 0.01 g of the J10-PET thin film. The experiments were carried out at a temperature of 308 K, while agitating the mixture at 150 rpm for durations ranging from 5 to 140 minutes.

### Investigation of Adsorption Kinetics

Analyzing changes in adsorption capacity over time offers valuable insights into adsorption kinetics. It is crucial to comprehend the specific adsorption mechanism in a given system. To characterize the kinetics of MB and MO adsorption onto the J10-PET thin film, four kinetic models were employed: pseudo-first-order, pseudo-second-order, Elovich, and intraparticle diffusion. These models aid in shedding light on the adsorption process behavior as it unfolds over time.<sup>43,44</sup>







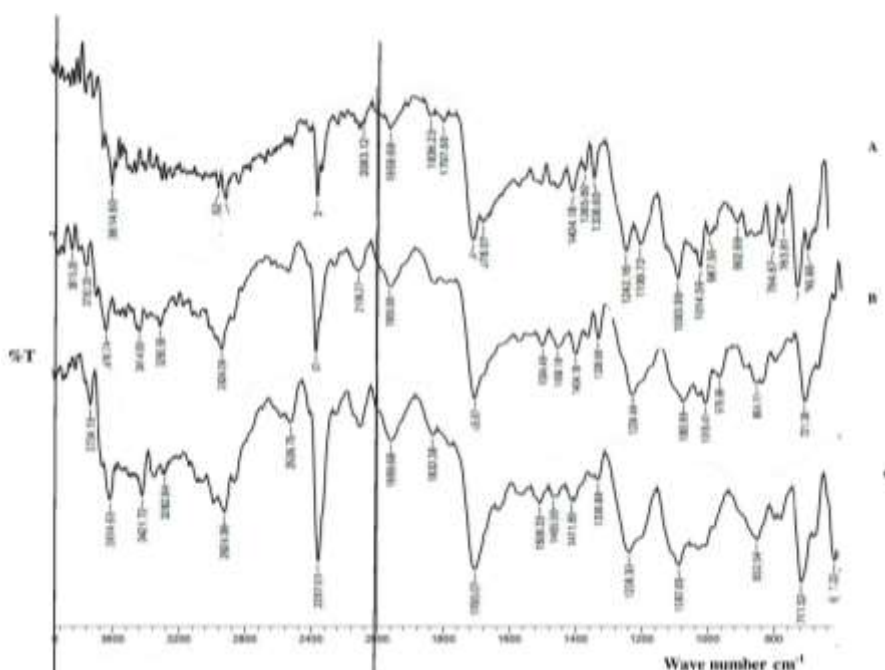
**Figure 2. SEM Micrographs of A) PET thin film B) J10- PET thin film**

The specific peaks detected in the FTIR spectra, including those in the amide range typically spanning  $1600\text{-}1800\text{ cm}^{-1}$  and  $1250\text{-}1350\text{ cm}^{-1}$ , are indicative of characteristic infrared signatures associated with the J10 compound. Additionally, the presence of peaks at  $1836$ ,  $1797$ , and  $1708\text{ cm}^{-1}$  aligns with carbonyl ( $\text{C}=\text{O}$ ) stretching vibrations, a well-recognized feature of amides. The absorption band at  $1678\text{ cm}^{-1}$ , which corresponds to  $\text{C}=\text{O}$  stretching vibrations, further substantiates the presence of

amide groups within the J10 compound. The bands at  $1242$  and  $1199\text{ cm}^{-1}$  are in agreement with  $\text{C-N}$  stretching vibrations, consistent with the chemical structure of the J10 compound. Furthermore, the absorption band at  $717\text{ cm}^{-1}$ , attributed to out-of-plane bending vibrations of aromatic  $\text{C-H}$  bonds, aligns with the anticipated characteristics of aromatic compounds found in the J10 compound Fig. 3A. These FTIR results provide strong confidence in the presence and characteristics of the J10 compound on the surface of the thin film.

The presence of active functional groups on the surface of the J10-PET thin film serves as a crucial factor in its role as an adsorbent for MB and MO. To discern any changes in the positions of these active functional groups after MB and MO adsorption, FTIR spectra of the J10-PET thin film post-adsorption were collected using an FTIR spectrometer Fig. 3B, 3C.

Notably, the redshift observed in the peaks at positions  $1678$  and  $1242\text{ cm}^{-1}$  for MO and MB after adsorption provides compelling evidence of the interaction between the adsorbent and the adsorbate, further elucidating the adsorption mechanism.<sup>45, 46</sup>



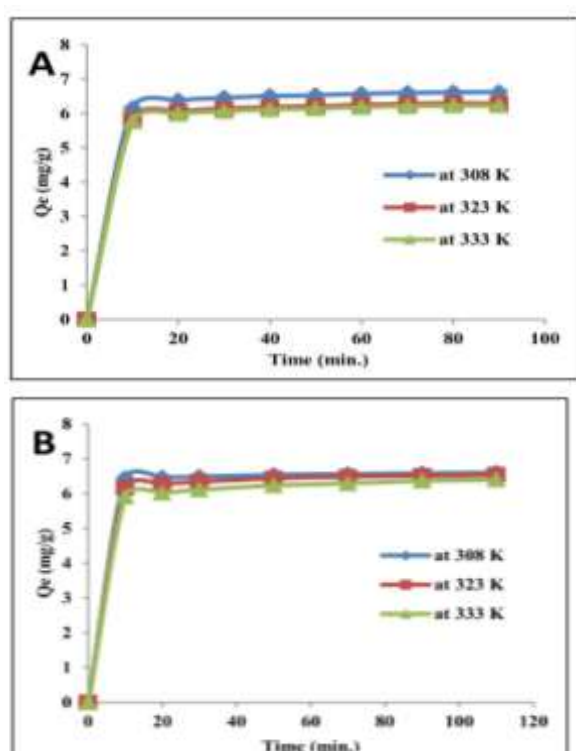
**Figure 3. FTIR Spectra of A) J10- PET thin film B) J10- PET thin film after adsorption of MO C) J10- PET thin film after adsorption of MB**

### Impact of Contact Time and Temperature on Adsorption

Fig. 4A, B depict plots showing the adsorbed quantities of MB and MO, denoted as  $Q_e$  (mg/g),

about the contact time at different temperatures ( $308$ ,  $323$ , and  $333\text{ K}$ ). Notably, there is a conspicuous trend wherein the uptake of both MB and MO, represented by  $Q_t$  (mg/g), exhibits a rapid surge

within the initial 10 minutes across all temperature levels, indicative of film diffusion, where the adsorbate molecules on the surface of the adsorbent quickly establish a thin layer, or film," due to the high concentration gradient between the bulk solution and the surface. A swift adsorption rate characterizes this process during the early stages of contact. Beyond this period, the rise in adsorbed quantities becomes more gradual, suggesting the involvement of pore diffusion, eventually culminating in equilibrium around the 100-minute mark.<sup>47</sup>

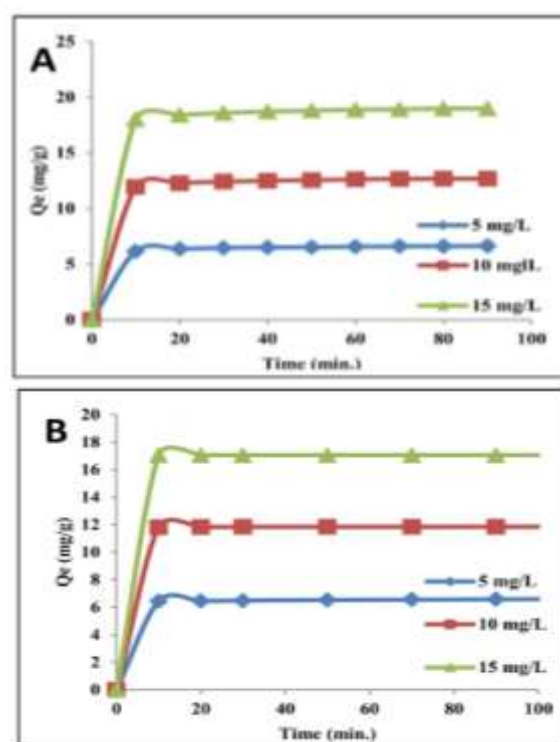


**Figure 4. Impact of contact time and temperature on the adsorption of A) MB B)MO on J10- PET thin film**

#### **Impact of Contact Time and Dyes Concentration on Adsorption**

The initial concentrations of both MB and MO are crucial factors in facilitating the effective transfer of these molecules between the aqueous and solid phases, especially when dealing with potential resistance. The adsorption behavior of MB and MO onto the J10-PET thin film was examined in relation to contact time and varying initial concentrations, as shown in Figs. 5A, B. The results revealed a common trend across all concentrations. Initially, during the first ten minutes of contact, the adsorption process was rapid, and it progressively increased as the contact time extended until it reached equilibrium.

Furthermore, it was observed that increasing the initial concentrations of MB and MO led to a corresponding increase in  $Q_t$  within the initial ten minutes. This behavior can be attributed to the higher initial concentrations creating greater momentum within the pores, thereby enhancing the adsorption capacity<sup>48</sup>. These findings closely parallel previous research on the adsorption of methylene blue dye using a carbonaceous hydro char adsorbent.<sup>49</sup>



**Figure 5. Impact of contact time and MB concentration on the adsorption of A) MB B) MO on J10- PET thin film**

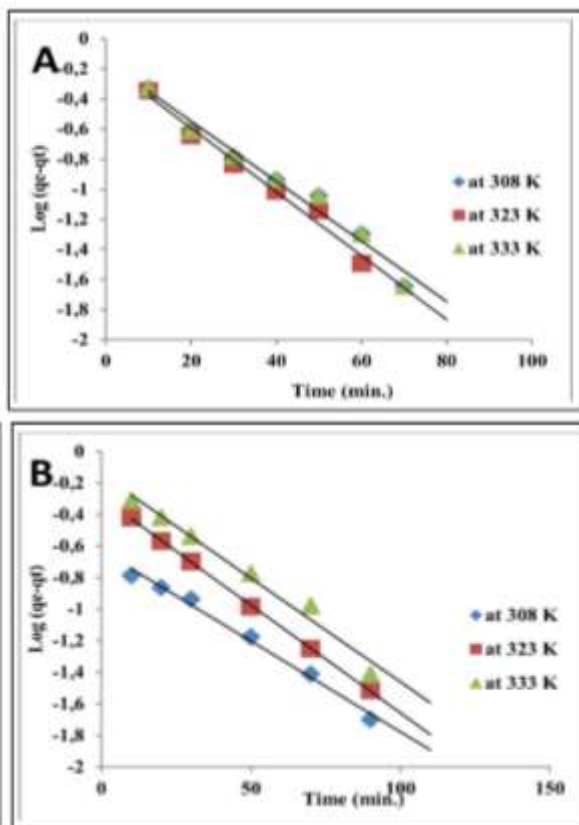
#### **Investigation of Adsorption Kinetics**

Estimating sorption mechanisms and rate-control processes is crucial in determining the optimal operating parameters for the complete batch process. To do this, four kinetic models are employed. The adsorption capability of MB and MO onto a thin film of J10-PET is seen at various concentrations and temperatures. The monitoring is done over time until a steady state is achieved, indicating a constant amount of MB and MO adsorbed. The adsorption process entails the attainment of balance. The equilibrium time is constant at 100 minutes across all concentrations and temperatures examined. The  $Q_e$  of MB and MO onto J10-PET thin film is directly related to the initial concentrations of MB and MO and the temperatures at which the adsorption process

happens. The tables provided in this study reflect the equilibrium adsorption capacity values achieved across various experimental settings, as outlined in Tables 1,2.

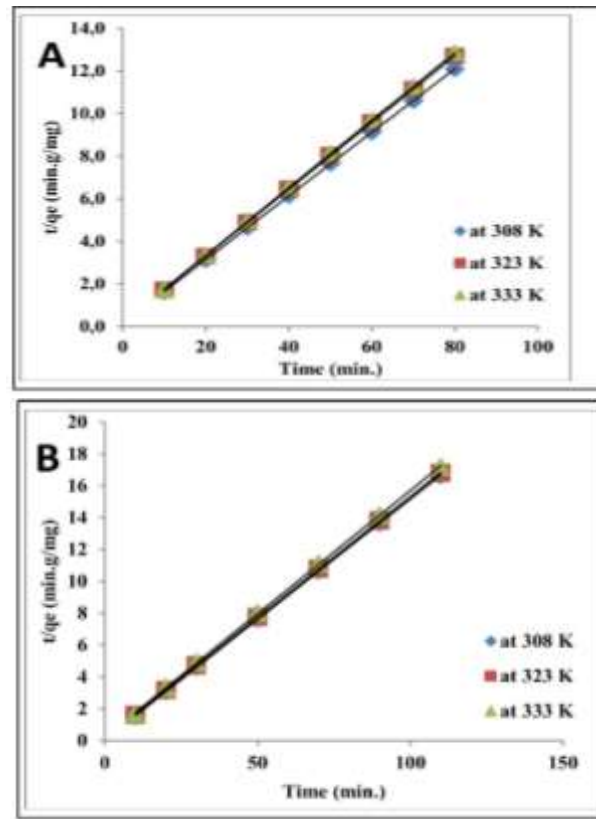
### Impact of Temperature on Adsorption Kinetics

The impact of temperature on the pseudo-first order adsorption kinetics of MB and MO onto a J10-PET thin film is illustrated in Figs. 6A, B.



**Figure 6. Pseudo-first order adsorption kinetics of the adsorption of A) MB B) MO on J10- PET thin film**

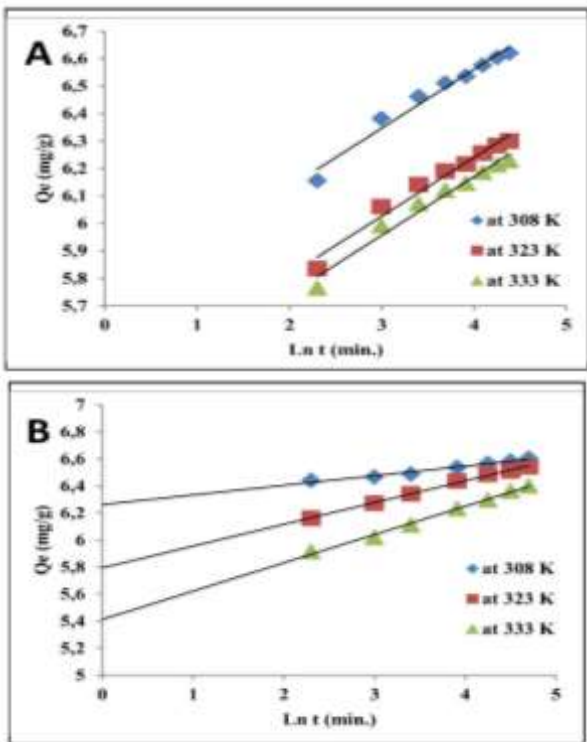
Even though the experimental data gave correlation coefficients ( $R^2$ ) that were quite large, exceeding 0.958, it is clear from Table .1 that the calculated equilibrium sorption capacities  $Q_t$  do not match the experimental values. This finding suggests the pseudo-first-order model is unsuitable for accurately predicting MB and MO's adsorption behavior onto a J10-PET thin film.



**Figure 7. Pseudo-second order adsorption kinetics of the adsorption of A) MB B) MO on J10- PET thin film**

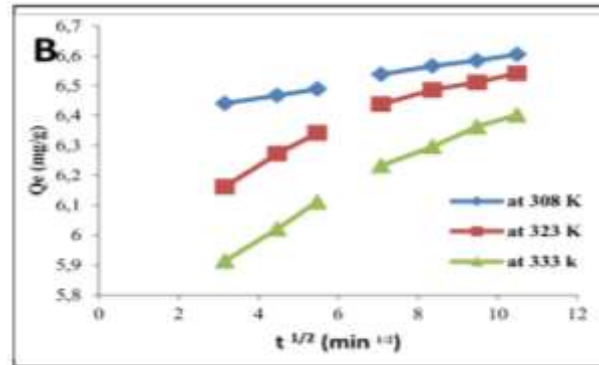
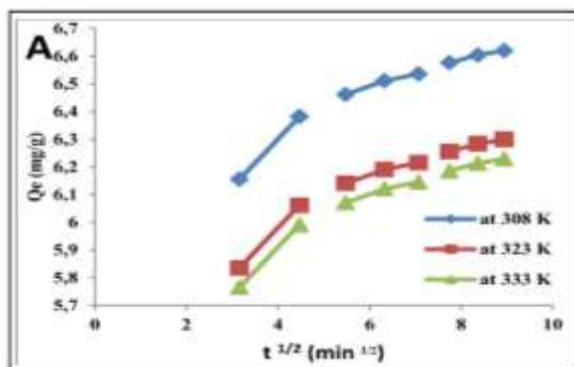
Fig. 7 depicts the kinetics of pseudo-second-order adsorption for MB and MO dyes on a J10-PET thin film under varying temperature conditions (308, 323, and 333 K). The figure illustrates the relationship between time and the degree of adsorption for both dyes. A notable finding is the consistent linear correlation observed across varying temperature conditions. This strong linear relationship indicates the suitability of the pseudo-second-order kinetic model for describing the adsorption process.

The Elovich equation is a rate equation that is derived from the consideration of adsorption capacity. Fig. 8 displays a plot of  $qt$  against  $\ln(t)$ . If the adsorption of MB and MO on the J10-PET thin film is consistent with the Elovich model, then a plot of  $q_t$  versus  $\ln(t)$  reveals a linear relationship with high  $R^2$ .



**Figure 8. Elovich kinetic for adsorption kinetics of the adsorption of A) MB B) MO on J10- PET thin film**

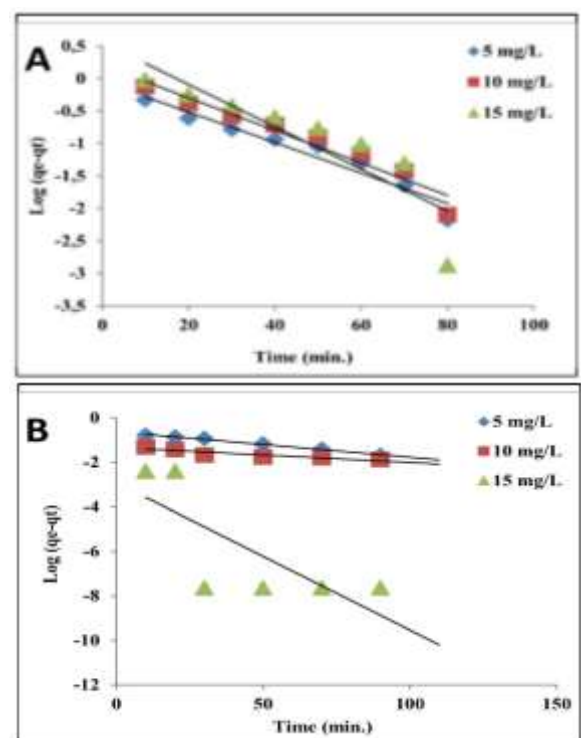
Intraparticle diffusion kinetics is a concept used to describe the rate at which solute molecules or ions diffuse into the interior of solid adsorbent material during an adsorption process. The intraparticle diffusion model is often used to describe the diffusion-controlled step of the adsorption process. The model assumes that the adsorption rate is influenced by the diffusion rate of the solute within the adsorbent particles. Fig. 9 illustrates the impact of temperature on the intraparticle diffusion kinetics of MB and MO.



**Figure 9. Intraparticle diffusion for adsorption kinetics of the adsorption of A) MB B) MO on J10- PET thin film**

### Impact of Initial Dyes Concentration on Adsorption Kinetics

Fig. 10 illustrates the pseudo-first-order sorption kinetics observed during the adsorption process of MB and MO onto the J10-PET thin film. The experiment was conducted at a temperature of 308 K, with initial dye concentrations of 5, 10, and 15 mg/L. The pseudo-first-order model exhibits an increase in the rate of constant values as the initial concentrations of the dye gradually grow.



**Figure 10. Pseudo-first order adsorption kinetics of the adsorption of A) MB B) MO on J10- PET thin film.**



Fig. 11 illustrates the kinetics of pseudo-second-order adsorption for the dyes MB and MO onto a J10-PET thin film. The initial dye concentrations used in the experiment were 5, 10, and 15 mg/L, and the temperature was maintained at 308 K. A linear relationship was seen for all initial dye concentrations, with  $R^2$  values exceeding those of pseudo first-order kinetics.

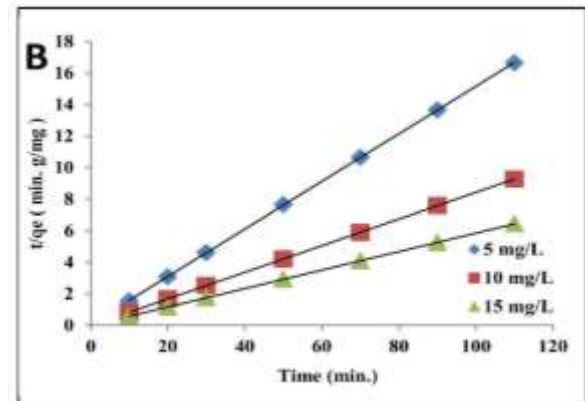
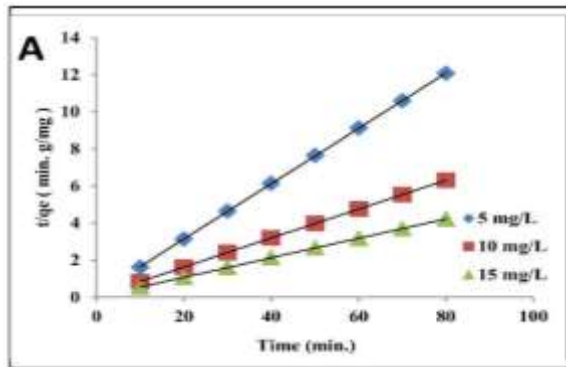
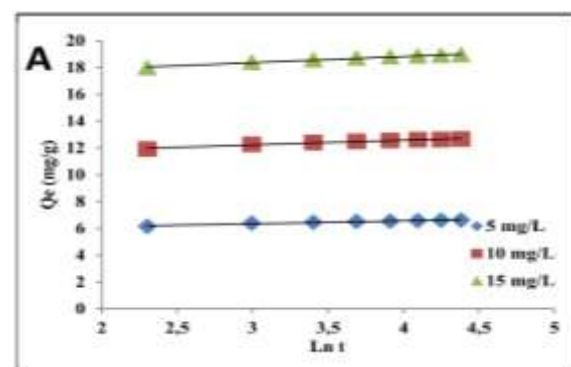


Figure 11. Pseudo-second order adsorption kinetics of the adsorption of A) MB B) MO on J10- PET thin film.

Table .1 Pseudo-first-order and Pseudo -second-order kinetic parameters of adsorption of MB and MO onto a J10-PET thin film at different temperatures and different initial MB and MO concentrations.

Dye	Temp./Conc.	$Q_e$ (exp.)	Pseudo – first order reaction			Pseudo – second - order reaction			
			$R^2$	$K_1$	$Q_e$ (calc.)	$R^2$	$h$	$K_2$	$Q_e$ (calc.)
MB	308 K	6.619	0.974	0.0458	1.414	1	10	0.225	6.666
	323 K	6.299	0.981	0.0488	1.478	0.999	8	0.194	6.410
	333 K	6.229	0.974	0.0458	1.414	0.999	5.714	0.144	6.289
	5 mg/L	6.619	0.944	0.0541	1.104	1	6.6	0.15	6.69
	10 mg/L	12.666	0.942	0.0575	1.059	1	14.7	0.09	12.79
	15 mg/L	18.965	0.781	0.0748	3.62	1	24.3	0.07	19.12
MO	308 K	6.604	0.991	0.0264	4.246	1	13.071	0.298	6.622
	323 K	6.542	0.999	0.0315	1.957	1	6.622	0.152	6.587
	333 K	6.402	0.979	0.0301	1.401	0.999	4.334	0.103	6.464
	5 mg/L	6.604	0.991	0.0265	4.25	1	13.1	0.298	6.623
	10 mg/L	11.862	0.834	0.0154	21.59	1	181.8	1.292	11.862
	15 mg/L	17.059	0.565	0.152	819.22	1	5000.0	17.170	17.065

The values representing the Elovich adsorption kinetics and rates of intra-particle diffusion can be determined by analyzing the slopes and intercepts of the plots depicted in Figs. 12,13.



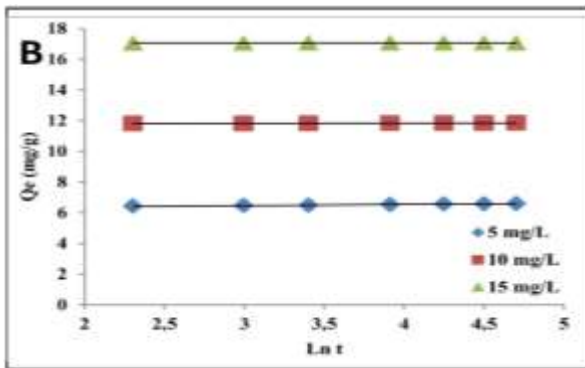


Figure 12. Elovich adsorption kinetics of the adsorption of A) MB B) MO on J10- PET thin film.

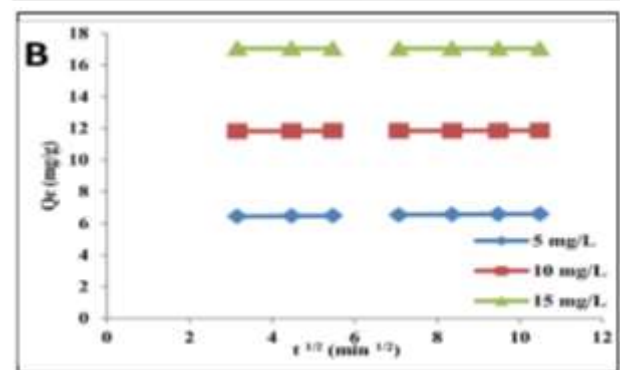
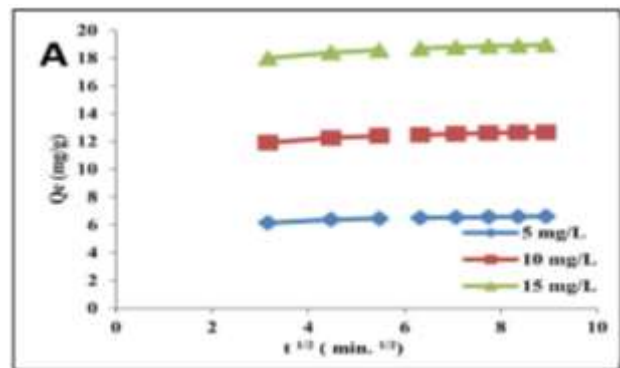


Figure 13. Intraparticle diffusion adsorption kinetics of the adsorption of A) MB B) MO on J10- PET thin film.

Table 2 Elovich and Intraparticle diffusion adsorption kinetics parameters of adsorption of MB and MO onto a J10-PET thin film at different temperatures and different initial MB and MO concentrations.

Dye	Temp./Conc.	Elovich kinetic equation			Intraparticle diffusion		
		R <sup>2</sup>	$\alpha$	$\beta$	R <sub>2</sub>	K <sub>diff</sub>	B <sub>L</sub>
MB	308 K	0.971	25.113	4.672	0.904	0.0731	6.0095
	323 K	0.971	23.618	4.672	0.904	0.0731	5.6895
	333 K	0.971	23.291	4.672	0.904	0.0731	5.6195
	5 mg/L	0.97	25.11	4.67	0.90	0.07	6.01
	10 mg/L	0.98	30.70	2.84	0.91	0.12	11.67
	15 mg/L	0.99	36.32	2.18	0.94	0.16	17.64
MO	308 K	0.9745	85.436	14.0647	0.993	0.0231	6.3671
	323 K	0.9965	34.210	6.218	0.956	0.0507	6.0421
	333 K	0.9947	24.283	4.775	0.985	0.0671	5.7267
	5 mg/L	0.975	85.44	14.06	0.99	0.02	6.37
	10 mg/L	0.928	635.79	54.35	0.89	0.01	11.80
	15 mg/L	0.714	8967.94	526.32	0.62	0.00	17.06

The kinetics of MB and MO adsorption onto J10-PET thin film are assessed using four kinetic models: pseudo-first-order, pseudo-second-order, Elovich, and intraparticle diffusion. A higher correlation coefficient signifies a firmer fit, with a value of 1 denoting a perfect fit. The results presented in Table .1 reveal that correlation coefficients ranging from 0.999 to 1 across various temperatures (308, 323, and 333 K ) and various concentrations (5, 10, and 15

mg/L indicate an exceptionally robust positive correlation between experimental data and predictions from the pseudo-second-order kinetic model.<sup>50</sup>

The calculated equilibrium sorption capacities,  $Q_e$  and  $Q_0$ , align closely with experimental findings. This agreement underscores the model's effectiveness in accurately representing the data,

suggesting a strong resemblance between observed and predicted adsorption behaviors.<sup>49</sup>

Consequently, the pseudo-second-order kinetic model emerges as a fitting portrayal of the adsorption process of MB and MO onto J10-PET thin film.

The findings suggest that the observed pseudo-second-order kinetics can be attributed to chemisorption, wherein the MB and MO dyes form chemical bonds with the J10-PET thin film. The pseudo-second-order kinetics model indicates that the adsorption process is predominantly governed by the interaction between the solute and the active sites of the adsorbent, resulting in the establishment of chemical bonds.<sup>51</sup>

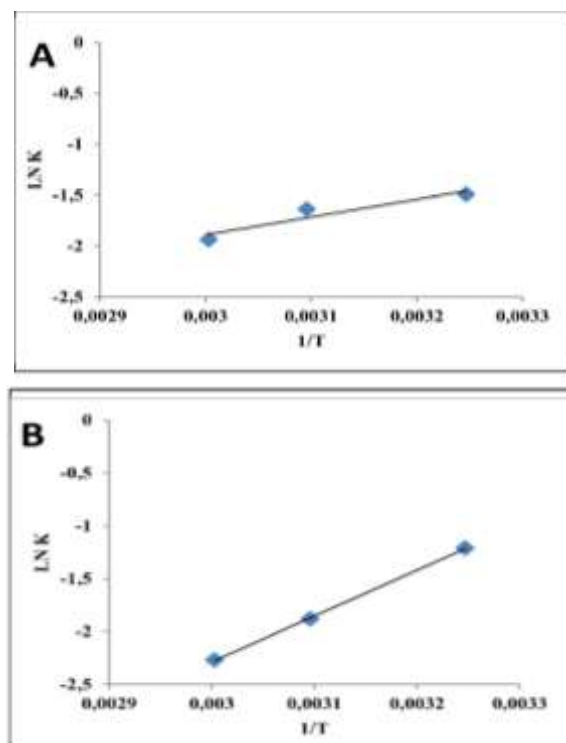
As shown in Table.2, the Elovich equation of MB and MO adsorption onto J10-PET thin film also has a respectable  $R^2$  (0.971-0.99) across various temperatures (308, 323, and 333 K) and various concentrations (5, 10, and 15 mg/L) suggesting a good fit indicates a strong positive correlation between the experimental data and the predictions made by the Elovich equation. However, this correlation is slightly weaker compared to the pseudo-second-order model. While the Elovich equation provides a good fit for data, it might not capture the data's behavior as closely as the pseudo-second-order model.

The rates of intra-particle diffusion can be estimated by analyzing the slopes of the plots illustrating  $qt$  against  $t^{1/2}$ , as depicted in Figs. 9,13. The linear trends observed in both figures emphasize the significant role of intra-particle diffusion in the adsorption process of MB and MO onto the J10-PET thin film. This observation confirms that the adsorption process encompasses multiple sequential steps within the material. When these steps occur independently, the plot typically exhibits intersecting lines, each representing a distinct mechanism—surface adsorption and intra-particle diffusion.<sup>52,53</sup>

Furthermore, the intercept  $B_L$  provides insights into the escalating influence of the boundary layer as the intercept value rises. A higher  $B_L$  intercept value signifies a greater abundance of solute adsorption within the boundary layer. The values of  $B_L$ , derived from the intercepts of linear  $qt$  vs.  $t^{1/2}$  plots across all studied conditions, have been compiled in Table 2.

### Activation Energy

The Arrhenius equation establishes a relationship between the rate constant and the temperature, which is utilized to estimate the activation energy<sup>47</sup>. Fig. 14 shows the relationship between the rate constant in Table 1 and the corresponding temperature to estimate the activation energy for MB and MO adsorption on a J10-PET thin film.



**Figure 14.** A plot of  $\ln K_2$  versus  $1/T$  of the adsorption of A) MB B) MO on J10- PET thin film.

The activation energy in adsorption signifies the energy needed to overcome the interactions between the adsorbate (dye molecules) and the adsorbent surface. The value of activation energy for the adsorption of MB and MO onto J10-PET thin film calculated from the slope of the plot is 14.42 and 36.08 kJ/mol for MB and MO, respectively, representing the energy required to initiate the adsorption process for MB and MO onto J10-PET thin film.

Activation energy values provide insights into the energetics and mechanisms of MB and MO adsorption onto the J10-PET thin film. The fact that activation energy values are relatively low suggests that the adsorption process is favorable and relatively easy to initiate, which can be important for designing energy-efficient adsorption processes.

## Mechanism of Adsorption and Attraction between J10, MB, and MO

The adsorption process is driven by several factors, including surface charge and the presence of compatible functional groups. An examination of the FTIR spectra reveals the mechanisms of attraction between molecule J10 and the dye molecules MB and MO. The adsorption of J10 onto MB and MO is driven by a combination of dipole-dipole interactions, Van der Waals forces, n- $\pi$  interactions, and, in the case of MB, hydrogen bonding Fig. 15. These complex mechanisms of attraction contribute to the adsorption of J10 onto the dye molecules, providing a comprehensive understanding of the adsorption process.

### 1- Dipole-Dipole and Van der Waals Interactions:

One of the primary driving forces of attraction involves dipole-dipole interactions and Van der Waals forces:

The carbonyl group (C=O) in J10 plays a central role in these interactions. It has a polar bond, with oxygen being more electronegative, resulting in a partial negative charge on the oxygen atom and a partial positive charge on the carbon atom. MB and MO also contain regions of electron density and electronegative nitrogen and sulfur atoms. These electronegative atoms can interact with the partially positively charged carbon atom or the partially negatively charged oxygen atom in J10, leading to dipole-dipole interactions.

2- Van der Waals forces, specifically London dispersion forces, exist in all molecules due to temporary fluctuations in electron distribution. J10, MB, and MO have nonpolar segments within their structures that can interact through Van der Waals forces. Additionally, the presence of aromatic rings in all three molecules can lead to pi-stacking

## Conclusions

In this study, we introduced an innovative approach utilizing N-(3-benzylureido)(methyl)-2-(6-methoxynaphthalen-2-yl)propanamide (J10) as an additive for recycled PET to create J10-PET thin films. These thin films demonstrated impressive adsorption capabilities, with equilibrium adsorption capacities ( $Q_e$ ) of 19.12 mg/g for MB and 17.065 mg/g for MO. The research emphasized the effectiveness of the J10-PET thin film for the removal of MB and MO from aqueous solutions. The

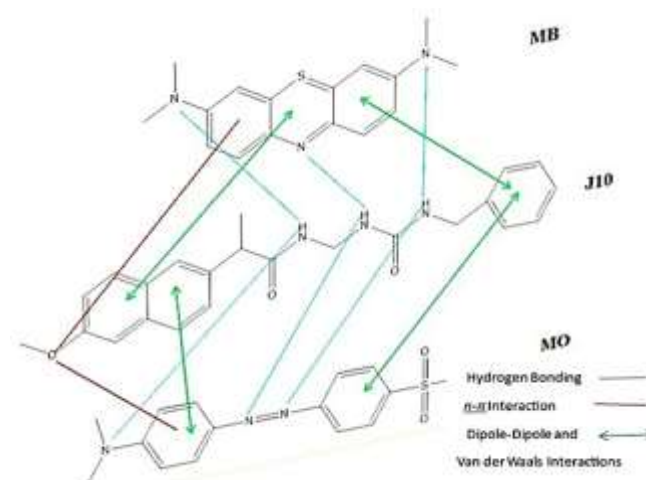
interactions, where the  $\pi$ -electron clouds of these rings engage in Van der Waals interactions, further enhancing mutual attraction.<sup>54</sup>

### 3- n- $\pi$ Interaction:

An additional mechanism contributing to the attraction is the n- $\pi$  interaction. This arises from the delocalization of the lone pair of electrons on the oxygen atom in J10 into the  $\pi$  orbital of the aromatic rings in the dye molecules. This interaction serves to strengthen the bonding between J10 and MB or MO.<sup>55</sup>

### 4- Hydrogen Bonding:

In the case of MB and MO, both the nitrogen atom in MB, MO, and the hydrogen atom within the NH group of J10 have the potential to establish hydrogen bonds. This interaction arises from the nitrogen atom in MB and MO functioning as a hydrogen bond acceptor, utilizing its available lone pairs to engage in hydrogen bonding with the hydrogen atom in the NH group of J10.<sup>56</sup>



**Figure 15. Mechanism of Adsorption and Attraction between J10, MB, and MO**

pseudo-second-order model was found to aptly describe the kinetics of adsorption, highlighting the efficiency of the J10-PET thin film. Moreover, the activation energy values of 14.42 kJ/mol for MB and 36.08 kJ/mol for MO underscored the potential of this adsorbent for the effective removal of these pollutants. Our study contributes to a comprehensive understanding of adsorption processes, presenting the J10-PET thin film as a promising and sustainable solution for addressing the removal of MB and MO



pollutants in aqueous environments. Future work may focus on further optimization of the J10-PET thin film production, exploration of additional pollutants, and the practical applicability of this adsorbent in real-world environmental scenarios. These results offer promising possibilities for eco-

friendly solutions in water purification and pollution control, emphasizing the potential of J10-PET thin films as a sustainable and efficient adsorption technology.

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### Authors' Declaration

- Conflicts of Interest: None.
- We hereby confirm that all the Figures and Tables in the manuscript are ours. Furthermore, any Figures and images, that are not ours, have been included with the necessary permission for re-publication, which is attached to the manuscript.

- Ethical Clearance: The project was approved by the local ethical committee at University of Duhok.
- No animal studies are present in the manuscript.
- No human studies are present in the manuscript.
- No potentially identified images or data are present in the manuscript

### Authors' Contributions Statement

J. A., P. H., and H. A. played vital roles in formulating and executing the research, analyzing the outcomes, and drafting the manuscript.

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## الابتكار الأخضر في المعالجة البيئية: الأغشية الرقيقة J10-PET لإزالة ملوثات الميثيلين الأزرق والميثيل البرتقالي بكفاءة

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### الخلاصة

تتناول هذه الدراسة التحديات البيئية الحرجة المتعلقة بمعالجة مياه الصرف، وتصريف الأدوية المنتهية الصلاحية، والتخلص من النفايات الصلبة مثل البولي إيثيلين تيريفثاليت PET. يركز البحث على نهج مبتكر باستخدام N-(3-benzylureido)(methyl)-2-(6-methoxynaphthalen-2-yl)propanamide (J10) كمادة مضافة لبولي إيثيلين تيريفثاليت (PET) المعاد تدويره من زجاجات المياه المستهلكة، وإنشاء فيلم رقيق سهل التحضير (J10-PET). تشمل الدراسة تحليلاً مقارناً بين الأفلام الرقيقة المصنعة من PET النقية والأفلام الرقيقة المصنعة من PET المعدلة بواسطة J10، باستخدام تقنيات متقدمة مثل FTIR و SEM للحصول على رؤى حول الخصائص الهيكلية والمورفولوجية لها. وللإشارة، تظهر الأفلام الرقيقة المطورة فعالية ملحوظة في استخراج الميثيلين الأزرق (MB) والبرتقالي الميثيلين (MO) من الحلول المائية. تركز الدراسة بشكل أساسي على كشف النقاب عن السلوكيات الحركية والتوازن التي تحكم إزالة الميثيلين الأزرق (MB) والبرتقالي الميثيلين (MO). تتضمن الدراسة تحديد ساعات الامتصاص في التوازن (qe) للميثيلين الأزرق (MB) والبرتقالي الميثيلين (MO) عند درجات حرارة مختلفة (35، 50، و 60 درجة مئوية) وتركيز مختلفة (5، 10، و 15 ملغم/جم). وبشكل ملحوظ، يتبين أن النموذج الزائف من الدرجة الثانية هو الأنسب لشرح السيناريو الحركي لكل من الميثيلين الأزرق (MB) والبرتقالي الميثيلين (MO). وعلاوة على ذلك، وجد أن الطاقة النشطة (Ea) المرتبطة بامتصاص الميثيلين الأزرق (MB) والبرتقالي الميثيلين (MO) على الفيلم الرقيق J10-PET بقيمة 14.42 كيلوجول/مول و 36.08 كيلوجول/مول على التوالي. تسهم هذه النتائج بشكل جماعي في فهم شامل لعملية الامتصاص، مما يلقي الضوء على إمكانية استخدام فيلم PET الرقيق J10 كحل فعال لمعالجة إزالة الملوثات الزرقاء الميثيلين (MB) والبرتقالية الميثيلين (MO) من البيئات المائية.

**الكلمات المفتاحية:** الأمتزاز، الأدوية المنتهية الصلاحية، الحركية، صبغة الميثيلين الأزرق، صبغة الميثيل البرتقالي، النفايات الصلبة من البولي أثلين تيريفثاليت، الأغشية الرقيقة، معالجة مياه الصرف الصحي.