Research Article

Experimental Design Analysis of Murexide Dye Removal by Carbon Produced from Waste Biomass Material

Rimene Dhahri 1,2, Mokhtar Guizani 1, Murat Yılmaz 4, Lassaad Mechi 5, Abdulmohsen Khalaf Dhahi Alsukaibi, 5 Fathi Alimi, 5 Ridha ben Salem 6, and Younes Moussaoui 2,6

1Laboratory for the Application of Materials to the Environment, Water and Energy (LR21ES15), Faculty of Sciences of Gafsa, University of Gafsa, Gafsa, Tunisia
2Faculty of Sciences of Gafsa, University of Gafsa, Gafsa, Tunisia
3Division of Environmental Engineering, Faculty of Engineering, Hokkaido University, Sapporo, Japan
4Department of Chemical Engineering, Faculty of Engineering, Osmaniye Korkut Ata University, Osmaniye 80000, Turkey
5Chemistry Department, Faculty of Science, Ha’il University, P.O. Box 2440, Ha’il 81451, Saudi Arabia
6Organic Chemistry Laboratory (LR17ES08), Faculty of Sciences of Sfax, University of Sfax, Sfax, Tunisia

Correspondence should be addressed to Younes Moussaoui; y.moussaoui2@gmx.fr

Received 30 October 2021; Revised 6 December 2021; Accepted 4 January 2022; Published 8 March 2022

Academic Editor: Liviu Mitu

Copyright © 2022 Rimene Dhahri et al. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

The aim of this work is to investigate the adsorption of an anionic dye, the Murexide (MX) present in aqueous solution, on activated carbon, derived from prickly pear seed cake biomass after bio-oil extraction. The obtained adsorbent used was characterized by Bohem titration, pH of point of zero charge (pH_{PZC}), FTIR spectroscopy, Brunauer–Emmett–Teller surface area (S_BET), and scanning electron microscopy (SEM). The different experimental parameters of the adsorption process, such as temperature, contact time, initial dye concentration, and adsorbent dose, were studied. For the optimization of the process, the effects of these parameters were investigated using the full factorial experimental design methodology. Design Expert 11.1.2.0 Trial software was used for generating the statistical experimental design and analysing the observed data. Langmuir and Freundlich’s adsorption models were employed to provide a description of the equilibrium isotherm. The adsorption process was found to obey Langmuir, which indicates that the Murexide had formed a monolayer onto activated carbon. Furthermore, according to the regression coefficients, it was observed that the kinetic adsorption data can fit better by the pseudo-second-order model compared to the first-order Lagergren’s model. The thermodynamic studies indicated that the adsorption of Murexide occurs in a spontaneous and exothermic process. The regeneration process of the exhausted adsorbent was studied to assess the economic and operational feasibility. According to the obtained findings, it is proposed that the activated carbon prepared from prickly pear seed cake retains a high potential for Murexide removal and is suitable for repetitive usage.

1. Introduction

In recent years, synthetic dyes have been excessively used around the world and have become very common contributors to wastewater effluents from a variety of sectors [1], including discharges from various industries, such as the textile industry [2]. The majority of these dyes represent serious environmental problems through carcinogenic substance accumulation and discharge of toxic elements [3]. In addition, trace amounts of dyes in water may be able to induce allergic dermatitis and cutaneous irritation [4, 5]. There are three types of dyes: anionic, cationic, and nonionic. Anionic dyes are classified as direct, acid, and reactive dyes, whereas
cationic dyes can be considered basic dyes [6]. Recently, significant interest has centred on the elimination of dyes from wastewater due to their resistance to biodegradation and their hazardous nature [4]. The high toxicity and solubility of these pollutants require the use of various processes for their removal, such as electrochemical treatment [7], chemical oxidation [8], photocatalysis [9], and adsorption [10]. Adsorption is considered an excellent promising technology simultaneously appropriate and economical for wastewater treatment [11]. Often high adsorption capacity is needed for the best performance for the removal of contaminants [12]. Activated carbon is extensively used as an adsorbent applied for the removal of pollutants due to its large specific surface area and high adsorption capacity.

On the other side, commercially activated carbons are perceived as costly materials because of the use of nonrenewable and relatively costly raw materials, which are inappropriate for use in pollution prevention applications [13]. Therefore, there is a need for alternative low-cost materials that do not necessitate additional costly treatment, allowing the adsorption process to be economical and feasible. An efficient adsorption process requires not only the adsorption capacity of the adsorbents but also a consistent availability of the adsorbent precursor for the process. Therefore, it is recommended to use low-cost precursors (industrial waste, mineral, or agricultural waste).

Among these precursors, different agricultural biomasses have been used for the production of activated carbon, such as coconut shells [14], orange peels [15], Posidonia oceanica [16], date palm petiole [12], maize tassel [17], and coffee waste [18]. Motivated by the search for a new precursor, prickly pear seed cake has been considered in this study. Prickly pear, the fruit of cactus which grows in abundance in the Mediterranean region, is a popular fruit in north Africa, and it is being extensively used as a food and for cosmetic oils production [19]. Prickly pear seed cake was produced in large quantities as a by-product of the prickly pear seed oil extraction industry. This by-product cake has low economic value and has not been valorised yet, and concerns are growing about its dumping as waste. Consequently, preparing activated carbons with this raw material is an interesting approach in order to valorise this raw material and appropriately manage it. For this purpose, there are currently two activation techniques applied: chemical and physical activation [20, 21]. In the present study, a chemical activation method for the production of activated carbon was investigated. Also, the obtained activated carbon was used as an adsorbent for Murexide removal from aqueous solutions. Murexide (C₈H₈N₆O₆) is an anionic dye that has been widely applied in several applications [22, 23]. Also, this dye is named ammonium purpurate, and it is the ammonium salt of purpuric acid [24]. It is available by heating alloxantine in ammonia gas at 100°C and also by boiling uramil using Mercury oxide. In the dry state, Murexide has the aspect of a red-violet powder that is soluble slightly in water [25]. It is applied as an indicator for complexometric titrations. Due to various uses of Murexide, when used, a huge quantity of Murexide flows into rivers and oceans. As a result, removing the dyes is an essential issue in wastewater treatment [23, 26].

Adsorption was performed according to different parameters: pH solution, adsorbent dosage, temperature, contact time, and initial concentration. Adsorption kinetics data were tested by pseudo-first-order and pseudo-second-order kinetic models. The equilibrium data were analysed using Langmuir and Freundlich models. The surface properties of activated carbon were characterized by scanning electron microscopy, FTIR spectroscopy, Brunauer–Emmett–Teller surface area ($S_{BET}$), Boehm titration, and pH\textsubscript{PZC}.

2. Experimental Part

2.1. Reagents and Raw Materials. Sodium carbonate (Na₂CO₃, ≥99%) was supplied by Fluka. Sodium hydroxide (BioXtra, ≥98%) and hydrochloric acid (HCl, ACS reagent, 37%) were provided by Sigma-Aldrich. Phosphoric acid (H₃PO₄, ≥98%), sodium bicarbonate (NaHCO₃, ≥99%), and Murexide dye (NH₄C₈H₄N₅O₆) (Table 1) were supplied by Scharlau.

Prickly pear seed cake was considered as a raw material for the development of activated carbon based on its availability and physicochemical properties. The prickly pear seed cake employed in this study was furnished by Omega Tunisia Ltd., a company operating in Sidi Bouzid, Tunisia. Prickly pear cake is a by-product of the extraction of oil from prickly pear seeds.

2.2. Preparation and Characterization of Activated Carbon. Prickly pear seed cake was crushed into small size pieces (≤2 mm). The prepared powder was carbonized at 600 for 2 h under N₂ flow rate (100 cm³/min). The activation of prepared carbon was insured by impregnation in H₃PO₄ with a ratio of 1:1 (g H₃PO₄/g carbon). Subsequently, the impregnated carbon was thermally activated at 500°C during 1 h. The resulting activated carbon was dried at 105°C and stored in a desiccator. To quantify acidic and basic functional groups on the obtained activated carbons, Boehm titration was applied [12]. The pH of point zero charge (pH\textsubscript{PZC}) of the prepared adsorbent was determined using Newcomb’s method [27]. To achieve this purpose, 50 mL of 0.01 mol/L NaCl solution was poured into six beakers, and for each beaker, the pH was adjusted to 2, 4, 6, 8, 10, or 12 by adding HCl and NaOH (0.1 mol/L), where necessary. An amount of activated carbon (about 0.2 g) was added to each beaker. After that, all the beakers were stirred for 24 h at room temperature (25 ± 1°C), and the final pH (pH₟) of the solution was measured and plotted against pHₚ (pHₚ = pH₟). The pH\textsubscript{PZC} value was deduced from the intersection of the plot of pHₚ = f (pH₟) with that of the bisector.

The FTIR spectroscopy was recorded over a range from 400 to 4000 cm⁻¹ using KBr pellets (10% solid) in a Shimadzu 8400–S spectrometer with the aim of collecting information about functional groups of the newly prepared activated carbon.
The Brunauer–Emmett–Teller surface area (S\text{BET}) and pore structure parameters [28] of the adsorbent were acquired from nitrogen adsorption-desorption measurements at 77 K using a Micromeritics ASAP 2020 instrument. Before the measurement, activated carbon (40 mg) was degassed at 300°C for 8 h. The SEM photos were obtained with a JEOL JSM-IT100 type device.

2.3. Bath Adsorption Experiments. The adsorption was carried out in batch mode. Murexide solutions were prepared by dissolving the required amount of the dye in deionized water. The effects of different process parameters such as pH of solution, adsorbent dose, time, initial concentration, and temperature were evaluated. The adsorption of Murexide was followed up by UV/Vis liquid spectrophotometer at a maximum wavelength of 523 nm. The equilibrium adsorption capacity \( Q\) (mg/g) was determined according to [29]

\[
Q = \frac{(C_0 - C_e) \times V}{m},
\]

where \( C_0 \) is the initial concentration (mg/L), \( C_e \) is the equilibrium concentration (mg/L), \( V \) is the volume of solution (L), and \( m \) is the weight of the adsorbent (g).

2.3.1. Effect of pH and Adsorbent Dose. In the adsorption process, pH has a significant role [30]. Many investigations have established that pH is a critical parameter that affects the adsorption capacity of organic compounds [31]. Generally, acidic substances are adsorbed more efficiently at lower pH values, while basic substances are more strongly adsorbed at high pH values [5]. To assess the effect of the initial pH of the solution, the pH of the solution was adjusted over the range 2–10.

The adsorbent dose is among the parameters that affect the adsorption capacity directly since it influences the adsorption capacity and enables us to predict the cost of activated carbon per unit of a solution to be treated [32]. In practical terms, 10 mL solution of a given concentration of MX was put in contact with doses of adsorbent ranging from 0.2 to 2 g/L for a predetermined time (180 minutes). Once filtered, the solution was analysed to determine its concentration, which was used to measure the adsorption rate of MX.

2.3.2. Effect of Time and Kinetic Study. In order to establish the contact time required to reach the equilibrium adsorption of a pollutant on activated carbon, it is necessary to follow the adsorption kinetics of MX in an aqueous solution on activated carbon. The contact time was varied between 0 and 720 min.

Adsorption equilibrium was established for different dyes concentration between 20 and 200 mg/L. The conventional kinetic models, including pseudo-first-order [33] and pseudo-second-order [34], were applied to explain the adsorption mechanism.

The pseudo-first-order model shown in equation (2) and the pseudo-second-order model shown in equation (3) [29] were used for the kinetics study:

\[
\ln(Q_{\text{ads}} - Q_t) = \ln Q_{\text{ads}} - K_1 t,
\]

\[
t = \frac{1}{Q_t} \left( \frac{1}{K_2} + \frac{1}{Q_e} \right) Q_t.
\]

where \( Q_e \) and \( Q_{\text{ads}} \) (mg/g) are the adsorption capacity at equilibrium and at time "t," respectively, and \( K_1 \) (min\(^{-1}\)) and \( K_2 \) (g/(mg·min)) are the rate constants for pseudo-first-order and pseudo-second-order adsorption, respectively.

2.3.3. Adsorption Equilibrium Study. Adsorption isotherm models are used to describe the behavior of the adsorbent/adsorbate system and to provide comprehensive explanations on the nature of the interaction between them. In the present study, two theoretical models have been adopted to explain the current phenomenon (Langmuir and Freundlich).

Langmuir isotherm model is one of the most frequently used isotherms for the adsorption by activated carbons in order to evaluate the maximum adsorption capacity of an adsorbate on an adsorbent. It requires that adsorption occurs on monolayer coverage of adsorbate over a homogeneous adsorbent surface. Also, this model assumes that adsorption performs on specific homogeneous and energetically identical sites [35].

\[
\frac{1}{Q_e} = \frac{1}{Q_m} + \frac{1}{Q_m K_L C_e},
\]

where \( Q_m \) (mg/g) is the maximum monolayer adsorption capacity, \( K_L \) (L/mg) is the Langmuir equilibrium constant related to the adsorption affinity, and \( C_e \) is the equilibrium concentration.

The dimensionless equilibrium constant for the Langmuir isotherm can be expressed as [36]

\[
R_L = \frac{1}{1 + K_L C_0}.
\]

The Freundlich model assumes heterogeneous surface energy. This model is widely used to describe multilayer adsorption [12]. The nonlinear form of the Freundlich can be expressed as follows:

\[
\log Q_e = \log K_f + \frac{1}{n} \log C_e,
\]
where $K_f$ is the Freundlich constant (heterogeneity factor), $1/n$ is the Freundlich coefficient, $n$ is related to sorption affinity, and $K_f$ is related to the adsorption capacity.

The materials and methods section should contain sufficient detail so that all procedures can be repeated. It may be divided into headed subsections if several methods are described.

2.3.4. Effect of Temperature and Thermodynamic Parameters. In order to investigate the influence of temperature variation on the adsorption of MX, an amount of adsorbent was mixed with the anionic dye solution until it reached equilibrium, under temperatures of 15, 30, and 40°C, and then filtered and with the anionic dyesolution until it reached equilibrium, the adsorption of MX, an amount of adsorbent was mixed on the adsorbent.

The thermodynamic parameters, standard enthalpy variation ($\Delta H^0$), the standard Gibbs free energy variation ($\Delta G^0$), and the standard entropy variation ($\Delta S^0$) were evaluated to confirm the adsorption nature of Murexide dye onto the activated carbon. The thermodynamic constants related to the adsorption will be calculated using the following equations [3]:

$$\Delta G^0 = -RT \ln K_L,$$  \hspace{1cm} (7)

$$\ln K_L = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT},$$  \hspace{1cm} (8)

where $T$ is the temperature (K), $K_L$ is the Langmuir constant (L/mol), and $R$ is the universal gas constant (8.314 J/mol K).

All adsorption experiments were performed at least twice with the same operating conditions to guarantee the reproducibility of the findings.

2.3.5. Desorption Experiment and Regeneration of Adsorbents. The regeneration of the exhausted adsorbent was studied to assess the reusability of the adsorbent and thus increase its potential as an economically attractive adsorbent [37]. The chemical regeneration method has the widest application in the separation of the absorbed molecules from the sites of the adsorbent, as it is comparatively rapid and occurs without affecting the pore structure of the activated carbon [38]. A 0.1 M NaOH solution was used in order to regenerate the exhausted adsorbent.

0.1 g of saturated activated carbon with ion dye was treated with 50 mL of 0.1 mol·L⁻¹ NaOH solution for 12 h. After separation by centrifugation, the treated adsorbent was subsequently washed several times with distilled water and then reused to adsorb 200 mg·L⁻¹ of Murexide in 50 mL of solution. The adsorption/desorption procedure is repeated five times using the same procedure, and the concentrations of MX in the supernatant after centrifugation are measured.

2.4. Full Factorial Design of Experiments. Full factorial experimental design considers all factors simultaneously and has been used largely as an alternative to the conventional experimental design, which treats each parameter separately and changes it one by one. The optimal experimental conditions are established by varying the factors at the same time and by applying several levels of the variables. Factorial designs are commonly used in experiments that consider multiple factors and for which it is necessary to investigate the effect of interaction between factors on the response [2]. However, the factorial design of experiments $2^n$ requires a reduced number of experiments for multiple factors; as a result, both materials and time used are reduced [39, 40]. The factorial design illustrates the impact and influence of each factor on the other factors at two levels [41]. Factors including pH, adsorbent dosage, and initial concentration of dye were considered in this study (Table 2). These variables with their respective domains were chosen from the literature as well as preliminary experiments.

Design Expert 11.1.2.0 software was used to generate the experimental design and analyse the observed data. A first-order model with all possible interactions was chosen to fit the experimental data (equation (9)). The goodness of fit of the model was checked by the coefficient of determination $R^2$ and $R^2_{adj}$; the $R^2$ value is a statistical measure of how well a model fits the real data points. $R^2$ values range from 0 to 1, where 1 represents the ideal model.

$$Y = b_0 + b_1A + b_2B + b_3C + b_{12}AB + b_{13}AC + b_{23}BC + b_{123}ABC,$$  \hspace{1cm} (9)

where $A$, $B$, and $C$ refer to the factors of the study and $Y$ is the responses of interest: pH, adsorbent dosage, and initial concentration of dye.

Response surface methodology was applied to optimize the combination effect on the adsorption efficiency (Ads%) [32, 42].

3. Results and Discussion

3.1. Activated Carbon Characterization. Surface functional groups have a considerable influence on the process of adsorption. Such functional groups are classified principally into acidic and basic groups [43], affecting both the surface charge of the adsorbent and consequently the adsorption performance [44].

According to the obtained results shown in Table 3, Bohem’s titration confirms the presence of acidic groups due to the existence of carboxylic, lactonic, and phenolic groups, as well as the presence of a high number of basic groups. The pH\textsubscript{PZC} of the adsorbent was 6.8, indicating a relatively acidic nature. This is in accordance with the Bohem titration result, which indicates a predominance of acidic groups on the surface of the adsorbent.

These findings were validated by the FTIR spectrum (Figure 1). The absorption bands in the range of 3415 cm⁻¹ represent the stretching vibration of the intermolecular and intramolecular O-H band. The absorption band at 1600 cm⁻¹ was attributed to carboxyl groups.

Additionally, the peak of the stretching vibrations of the C-O group is detected at 1030 cm⁻¹.

Nitrogen (N\textsubscript{2}) gas adsorption-desorption isotherm of activated carbon was performed to investigate the porous characteristics. The obtained activated carbon has a large specific surface area $S_{BET}$ equal to 551.14 m²/g, an average
pore diameter in the range of 4.71 (nm), and a total pore volume of 0.159 cm$^3$/g. According to the International Union of Pure and Applied Chemistry (IUPAC) isotherm classification standard \[45\], the obtained activated carbon has a mesoporous structure. This mesoporosity provides a favorable structural characteristic of the adsorbent used in this study to eliminate large molecules such as Murexide.

Scanning electron micrograph of the activated carbon is given in Figure 2. It can be seen clearly that the surface of the adsorbent was covered with cavities and pores of different sizes and shapes. The pores walls are filled with many subpores that connect the pores to the inside of the activated carbon prepared. Overall, the development of pores in the obtained activated carbon is a consequence of the decomposition of the sample induced by the activation process with H$_3$PO$_4$ as confirmed by Han et al. \[46\]. From these observations, it can be considered that the texture of the obtained adsorbent is suitable for the removal of dyes from aqueous solutions.

### 3.2. Batch Adsorption Results

#### 3.2.1. Effect of pH.

The adsorption of Murexide dye was established at different pH (2, 4, 6, 8, and 10). The variation of the adsorbed amount of Murexide dye as a function of pH is represented in Figure 3(a). The activated carbon adsorbed high quantities of Murexide at low (acidic) pH 2, and its adsorption was gradually decreased with increasing pH. The obtained results prove that the removal efficiency of MX was maximized for acidic pH. This is explicable on the basis of the zero charge point. It was noted that the acid dye is dissolved in the aqueous medium initially \[24\], followed by dissociation and conversion to anionic dye ions:

$$\text{HXONH}_4^+ \rightarrow H_2O \text{MXO}^- + \text{NH}_4^+. \quad (10)$$

The pHPCZ of the obtained activated carbon is 6.8. For lower pH ranges, the surface charge of the adsorbent is predominantly positive \[47\], which leads to an increase in electrostatic attraction towards anionic dyes and, consequently, an increase in adsorption efficiency \[48\]. The protonated groups of activated carbon are principally carboxylic (–CO–OH$^+$) and phenolic (–OH$^+$) groups (equations (11a) and (11b)) \[49\]. For a pH above pHPCZ, the charge of the surface of the adsorbent becomes negative. The deprotonated groups of activated carbon are mainly carboxylic (–CO–O$^-$) and phenolic (–O$^-$) groups \[49\].

$$\text{ACSurface} - \text{COOH} \rightarrow \text{H}^+ + \text{ACSurface} - \text{COOH}_2^-, \quad (11a)$$

$$\text{ACSurface} - \text{OH} \rightarrow \text{H}^+ + \text{ACSurface} - \text{OH}_2^+, \quad (11b)$$

$$\text{ACSurface} - \text{COOH} + \text{OH}^- \rightarrow \text{ACSurface} - \text{COO}^- + H_2O. \quad (12a)$$

$$\text{ACSurface} - \text{OH} + \text{OH}^- \rightarrow \text{ACSurface} - \text{O}^- + H_2O. \quad (12b)$$

The large reduction in dye adsorption at highly basic conditions can be attributed to electrostatic repulsion between the negatively charged activated carbon and the deprotonated dye. These results are in coincidence with these observed by Mall et al. \[50\].
3.2.2. Effect of Adsorbent Dose. The influence of the adsorbent dose on the adsorption of MX dye (200 mg/L) was investigated by ranging the adsorbent mass from 0.2 g to 2 g for 1000 mL of volume samples. Figure 3(b) reveals that the adsorption capacity increases with an increased dose of activated carbon. It can be seen that the MX removal efficiency increases from 126.3 to 197.7 mg/g, while the adsorbent dose increases from 0.2 to 2 g/L, the greatest amount adsorbed is 1 g/L for MX. This increase in adsorption with the increase in adsorbent dose was due to the availability of more adsorption sites [51]. This result is consistent with the findings reported [52].

3.2.3. Adsorption Kinetic Studies. Figure 3(c) shows that the amount of adsorbed Murexide increases progressively with time (probably this is explained by the high ratio of vacant surface sites to adsorbate molecules). The equilibrium of the system was achieved in about 180 minutes, which is appropriate for the equilibrium state between the adsorbate and the adsorbent. The investigation of the adsorption of the dye on the activated carbon as a function of time allows us to examine the influence of the contact time on its retention and to describe the mechanism that occurs. For this purpose, two models have been employed to outline the kinetic mechanism of adsorption of Murexide on the adsorbent of interest: the pseudo-first-order (equation (2)) and the pseudo-second-order (equation (3)).

The results exposed in Table 4 show that the regression coefficients for the pseudo-second-order model were very close to unity, indicating that the adsorption of MX follows the pseudo-second-order model kinetics. For this purpose, the predicted adsorbed quantities obtained with this model are consistent with the experimental values.

3.2.4. Isotherms Study. The experimental results were investigated using the Langmuir and Freundlich models. The calculated constants with the linear forms (equations (4), (5), and (6)) are reported in Table 5. Based on the results of Table 5, the Langmuir isotherm model seemed to be much more adjusted than the Freundlich one. The fitness of the Langmuir model to the adsorption process points that MX was adsorbed on a specific monolayer onto activated carbon [53]. The \( R_L \) value was found to be in the range from 0 to 1, suggesting the favorable adsorption of MX onto activated carbon [54]. However, \( n \) from the Freundlich model was greater than unity, which denotes the favorable sorption of MX onto the activated carbon [55].

Furthermore, the maximal capacities of absorption calculated with the Langmuir isotherm model were 196 mg/
g. The highest capacity of AC for MX is relatively high with respect to the knowledge in the literature, as reported in Table 6.

3.2.5. Adsorption Thermodynamics. The effect of temperature was studied. The adsorption was carried under different temperatures: 15, 30, and 40°C. The results were graphically presented in Figure 3(d). These findings reveal that the minimum equilibrium concentration of the MX dye corresponds to 15°C. Therefore, it can be deduced that heat excitation of the adsorption reaction decreased the adsorption capacities for MX. It is worth mentioning that similar observations were reported by other researchers where the maximum removal of Murexide dye on activated carbon was achieved at a lower temperature [25].

\[ \Delta H^0 \text{ and } \Delta S^0 \text{ are calculated from the linear plot } \ln(K_L) \text{ versus } (1/T) \] (Figure 4).

The obtained results illustrated in Table 7 indicate that the adsorption process on activated carbon occurs in a spontaneous and favorable process (\( \Delta G^0 < 0 \)) [57]. For the adsorption of MX, \( \Delta H^0 \) values are negative, proving that it is an exothermic process [58]. The positive value of entropy indicates that, during the reaction of MX on AC, the irregularity of MX molecules and adsorbent in the solution increases [59].

3.3. Full Factorial Design of Experiments. The removal of dyes using an adsorbent in a batch test generally relies on multiple factors. Thereafter, the simple and common method to study the effect of each factor and look for optimums is to use the one factor at a time (OFAT) approach, also called the univariate method. In OFAT, one single factor is changed at a time, and the rest are fixed. The OFAT method requires more tests and cannot allow interactions estimation between all factors [60]. However, the full factorial method, where more than one variable is changed at a time, is more precise and can detect interactions between variables with less experiment. Using full factorial experiments design in this study, optimum adsorption conditions were obtained as follows pH = 2, \( m = 2 \), \( C_0 = 20 \text{ mg/L} \), and \( T = 15^\circ \text{C} \) (Table 8). The highest adsorption percentage of Murexide dye was 100%.

The normal probability plot of the residuals is shown in Figure 5. The normality of the data can be checked by

| Table 4: Kinetics constants for the adsorption of Murexide. |
|-----------------|-----------------|-----------------|-----------------|
| \( C_0 \) (mg/L) | \( Q_{exp} \) (mg/g) | \( Q_1 \) (mg/g) | \( K_1 \) (min\(^{-1}\)) | \( R^2 \) | \( Q_2 \) (mg/g) | \( K_2 \) (g/mg min) | \( R^2 \) |
|-----------------|-----------------|-----------------|-----------------|-------|-----------------|-----------------|-------|
| 50              | 50              | 37.45           | -0.0064         | 0.773 | 49.2            | 0.00030         | 0.989 |
| 100             | 100             | 64.47           | -0.0059         | 0.954 | 98.1            | 0.00018         | 0.969 |
| 150             | 114.4           | 106.9           | -0.0061         | 0.893 | 114.0           | 0.00045         | 0.999 |
| 200             | 170.7           | 149.8           | 0.004           | 0.877 | 169.9           | 0.0015          | 0.999 |

| Table 5: Isotherm model constants for batch adsorption of Murexide. |
|-----------------|-------|-------|
| Temperature (°C) | 15    | 30    | 40    |
| Langmuir        |       |       |       |
| \( Q_m \) (mg/g) | 196.0 | 192.3 | 188.6 |
| \( K_L \) (L/g)  | 2.428 | 1.677 | 1.261 |
| \( R_L \)       | 0.0020| 0.0029| 0.0039|
| \( R^2 \)       | 0.9755| 0.987 | 0.984 |
| Freundlich      |       |       |       |
| \( K_f \) (L/g)  | 9.1705| 7.658 | 6.654 |
| \( R^2 \)       | 0.920 | 0.865 | 0.915 |

| Table 6: Maximum capacities for MX on various adsorbents. |
|-----------------|-----------------|-----------------|
| Precursor       | \( Q_{max} \) (mg/g) | Reference |
| SnO₂-NP-AC      | 67.1            | [23]           |
| Calcined eggshell | 1.0            | [56]           |
| Treated pomegranate bark | 1.7 | [24] |
| Untreated pomegranate bark | 0.7 | [24] |
| Rice husk       | 15.0            | [25]           |
| AC from prickly pear seed cake | 196.0 | This study |

The findings confirm that AC prepared from prickly pear seed cake can be favored as an efficient adsorbent for MX elimination.

Figure 4: ln \( K_L \) versus \( 1/T \).

Figure 5: The normal probability plot of the residuals.
plotting a normal probability of residues. It is obvious that, for the adsorption percentage of Murexide, the data points were quite close to the straight line, and this indicates that the experiments originated from a normally distributed population. In addition, a regression analysis was performed to adapt the response function to the experimental data. The effect of one factor is the variation in response that is induced by varying the level of the factor [61].

According to Table 9, pH, $C_0$, and $T$ are the most influential factors on the adsorption efficiency (Ads%), with $b_1 = -22.16$, $b_3 = -6.67$, and $b_4 = -7.13$, for pH, $C_0$, and $T$, respectively.

It can also be noted that adsorbent mass "m" has a positive effect on Ads% ($b_2 = 5.29$). A significant interaction was found between the pH and $C_0$ for the Ads% with a negative effect ($b_{13} = -1.75$).

**Table 7: Thermodynamic parameters for the adsorption of Murexide onto activated carbon.**

| Temperature (K) | $\Delta G^0$ (kJ/mol) | $\Delta H^0$ (kJ/mol) | $\Delta S^0$ (J/molK) |
|-----------------|------------------------|------------------------|------------------------|
| 288             | $-32.191$              |                        |                        |
| 303             | $-32.936$              | $-19.471$              | 44.242                 |
| 313             | $-33.281$              |                        |                        |

**Table 8: Factorial experimental design matrix.**

| Std. | pH | m | $C_0$ | T | pH | m | $C_0$ | T | Ads% |
|------|----|---|-------|---|----|---|-------|---|------|
| 1    | −1 | −1 | −1    | −1| 2  | 0.2| 20    | 15| 97.4 |
| 2    | 1  | −1 | −1    | −1| 12 | 0.2| 20    | 15| 49.6 |
| 3    | −1 | 1  | −1    | −1| 2  | 2  | 20    | 15| 100  |
| 4    | 1  | 1  | −1    | −1| 12 | 2  | 20    | 15| 68.3 |
| 5    | −1 | −1 | 1     | −1| 2  | 0.2| 200   | 15| 83.4 |
| 6    | 1  | −1 | 1     | −1| 12 | 0.2| 200   | 15| 39.7 |
| 7    | −1 | 1  | 1     | −1| 2  | 2  | 200   | 15| 92.5 |
| 8    | 1  | 1  | 1     | −1| 12 | 2  | 200   | 15| 48.5 |
| 9    | −1 | −1 | −1    | 1 | 2  | 0.2| 20    | 40| 82.6 |
| 10   | 1  | −1 | −1    | 1 | 12 | 0.2| 20    | 40| 34.9 |
| 11   | −1 | 1  | −1    | 1 | 2  | 2  | 20    | 40| 89.5 |
| 12   | 1  | 1  | −1    | 1 | 12 | 2  | 20    | 40| 53.4 |
| 13   | −1 | −1 | 1     | 1 | 2  | 0.2| 200   | 40| 71.3 |
| 14   | 1  | −1 | 1     | 1 | 12 | 0.2| 200   | 40| 21.1 |
| 15   | −1 | 1  | 1     | 1 | 2  | 2  | 200   | 40| 82.9 |
| 16   | 1  | 1  | 1     | 1 | 12 | 2  | 200   | 40| 29.5 |

**Table 9: Values of model coefficients of the responses.**

| Coefficient | Values |
|-------------|--------|
| $b_0$       | 65.287 |
| $b_1$       | -22.162|
| $b_2$       | 5.28   |
| $b_3$       | -6.675 |
| $b_4$       | -7.137 |
| $b_{12}$    | 0.512  |
| $b_{13}$    | -1.75  |
| $b_{14}$    | -0.262 |
| $b_{23}$    | -0.55  |
| $b_{24}$    | -0.387 |
| $b_{34}$    | -0.275 |
| $b_{123}$   | 0.195  |
| $b_{124}$   | -0.046 |
| $b_{134}$   | -0.725 |
| $b_{234}$   | -0.125 |
| $b_{1234}$  | -0.1   |

**Figure 5: Normal probability plots of residuals for the adsorption capacity of MX.**
The analysis of variance (ANOVA) was employed to assess the significance of the curvature in the responses at a confidence level of 95%. The interactions 2nd and 3rd orders between the different process parameters were considered insignificant. Next, the model, associated with the factors level and adsorption capacity, was generated by replacing the regression coefficients into leveld and adsorption capacity, was generated by replacing the regression coefficients into 

\[
\text{Ads\%} = +65.29 - 22.16 \times \text{pH} + 5.29 \times m - 6.68 \\
\times C_0 - 7.14 \times T - 1.75 \times (\text{pH} \times C_0).
\]  

The ANOVA results revealed that the equation sufficiently describes the actual relationship between the response and the factors with a linear regression coefficient \(R^2 = 0.9857\) and adjusted coefficient determination \(R^2_{\text{adj}} = 0.978\) (Table 10). The predicted determination coefficient \(R^2_{\text{pred}}\) of 0.963 is in reasonable agreement with \(R^2_{\text{adj}}\) (the difference is less than 0.2). Value of \(p\)-value \(> F\) was less than 0.05, indicating that the model terms are significant. In addition, large values of the \(F\)-value and reduced values of \(p\)-value \(> F\) confirm the significant effect of the corresponding variables [62, 63].

The relatively low value of the standard deviation (Std. Dev. = 3.79) demonstrates the small difference between the experimental and predicted values and also confirms the validity of the acquired model [64]. The mathematical model for the determination of the percentage of adsorption removal was applied to build response surfaces and to optimize the conditions of the adsorption process. Figure 6 illustrates the 3D response surface plots for the most significant interaction between the parameters of the process.

For Ads%, the best significant interaction was pH-initial concentration (pH-C0). The Ads% increased with decreasing pH and C0 (Figure 6). Then, a greater Ads% appeared at pH = 2 and C0 = 20 mg/L. The m was fixed at 2 g/L and the temperature at 15°C.

A desirability function approach has been frequently applied in multiresponse optimization due to its simplicity (Figure 7). All variables were considered equally important, and thus the values for the response variables were set to one. The maximum value of the desirability function (1.000) was obtained at pH = 10.82, m = 0.99 g, C0 = 96.01 mg/L, and T = 20°C. Moreover, under these conditions, the predicted response for the Ads% was 52.72%.

To find optimal conditions for the adsorption process of MX onto the activated carbon, model predictions values were compared to experimental data. The correlations between predictions and experimental responses indicated correspondence between the mathematical model and the experimental data.

Therefore, it can be deduced that pH has the most significant effect on the adsorption process. Increasing the pH leads to a reduction in the adsorption capacity of MX on the activated carbon. The coefficient was the strongest negative for the model equation (8). The second significant factor affecting the performance of the batch adsorption process was the adsorbent dose. Its positive coefficient indicates that the adsorption efficiency of AC was favored over an adsorbent dosage of 2 g/L. A higher adsorbent dose produced an increase in the percent of the elimination of the MX dye, as the increased adsorbent dose furnished an adequate surface area for adsorption. The third significant parameter was the initial concentration of MX. The surface and the active sites of the adsorbent may become saturated at increased concentration; thus, the adsorption capacity (%) is reduced with increasing initial MX concentration.

The interaction between pH and C0 was the fourth significant factor. This interaction had a negative coefficient; consequently, a reduction in the pH value of the solution combined with a reduced dose of the adsorbent induced an
Figure 7: Prediction profile plot with adjusted desirability and factor values.

Figure 8: Regeneration cycles of the activated carbon.

Figure 9: Proposed mechanism of adsorption/desorption of Murexide on activated carbon.
increase in the removal efficiency (%). In the same way, for the fourth factor, the adsorption capacity decreases as temperature increases; this is also an appropriate factor in the regeneration of activated carbon.

3.4. Regeneration Study. In this section, we discuss the results of the regeneration experiment. The results illustrate that, after each regeneration cycle, the adsorption capacity of MX was reduced, as presented in Figure 8. During the first cycle of regeneration, the percentage of MX removal (Ads%) was 99.6%, but after five cycles of regeneration, the efficiency dropped to 42.4%, suggesting that the regenerated adsorbent retains a high potential for MX removal and is suitable for repetitive usage. As a result, it will be economically productive, and it is very important for commercial applications to reduce secondary pollution in wastewater processing.

The decrease in performance may be explained by the residual of MX molecules inside the pores of the adsorbent [65, 66]. In the basic medium, which is the addition of NaOH in the above situation, the surface of the adsorbent is deprotonated [67]. Thus, the number of negatively charged sites increases, and the number of positively charged sites decreases, leading to a repulsive force between the anionic dye and the adsorbent (Figure 9), as confirmed previously by the study of the effect of pH on the adsorption process of MX. The presence of an excess of OH ions destabilizes the anionic dye MX and favors the desorption phenomenon, consequently the possibility of reuse of the regenerated adsorbent [68, 69].

4. Conclusion
Activated carbon produced by chemical activation of prickly pear seed cake with H₃PO₄ was able to remove Murexide dye molecules from aqueous solutions. Maximum adsorption of MX occurred at low values of pH, and adsorption capacity (%) also increased with increasing adsorbent dosage. Adsorption kinetics follows the pseudo-second-order model and fits the Langmuir model suggesting that the adsorption is a monolayer phenomenon, which probably occurs on energetically homogeneous sites. Also, the influence of pH (2–10), adsorbent dose (0.2–2 g/L), initial dye concentration (20–200 mg/L), and temperature (15–40°C) on adsorption efficiency (Ads%) was designed by using experimental methodology. It was discussed based on analysis of variance (ANOVA). The results of the desorption studies show that the adsorbent exhibits excellent regeneration performance and high adsorption capacity after five cycles.

According to the obtained findings, activated carbon, produced by the chemical activation of prickly pear seed cake, could be used as an effective and low-cost adsorbent to decrease the concentrations of dyes in wastewater. The results of this research highlight the importance of recycling agricultural waste to decrease environmental contamination and conserve natural resources. This encourages researchers to conduct further investigations in this area.

Data Availability
The data used to support this study are included within the article.

Conflicts of Interest
The authors declare that they have no conflicts of interest.

Acknowledgments
The authors would like to express their deep gratitude to Professor Mohamed Naceur Belgacem for his valuable and constructive suggestions during the development of this research work. The study was supported by the Deanship of Scientific Research at the University of Ha’il, Saudi Arabia, through Project no. RG-191251.

References
[1] H. Patel and R. T. Vashi, "Adsorption of crystal violet dye onto tamarind seed powder," E-Journal of Chemistry, vol. 7, no. 3, pp. 975–984, 2010.
[2] N. Özbay, A. F. Yargić, R. Z. Yarbay-Sahin, and E. Önal, "Full factorial experimental design analysis of reactive dye removal by carbon adsorption," Journal of Chemistry, vol. 2013, Article ID 234904, 13 pages, 2013.
[3] D. Lin, F. Wu, Y. Hu et al., "Adsorption of dye by waste black tea powder: parameters, kinetic, equilibrium, and thermodynamic studies," Journal of Chemistry, vol. 2020, Article ID 5431046, 13 pages, 2020.
[4] M. R. Al-Mamun, M. N. Karim, N. A. Nitun, S. Kader, S. Islam, and Z. H. Khan, "Photocatalytic performance assessment of GO and Ag co-synthesized TiO2nanocomposite for the removal of methyl orange dye under solar irradiation," Environmental Technology & Innovation, vol. 22, Article ID 101537, 2021.
[5] F. Taleb, M. Ammar, M. Ben Mosbah, R. ben Salem, and Y. Moussaiou, "Chemical modification of lignin derived from spent coffee grounds for methylene blue adsorption," Scientific Reports, vol. 10, Article ID 11048, 2020.
[6] A. M. Božecka, M. O. Naturalna, and M. Kopec, "Methods of dyes removal from aqueous environment," Journal of Ecological Engineering, vol. 22, no. 9, pp. 111–118, 2018.
[7] J. M. Peralta-Hernández, M. Méndez-Tovar, R. Guerra-Sánchez, C. A. Martínez-Huitie, and J. L. Nava, "A brief review on environmental application of boron doped diamond electrodes as a new way for electrochemical incineration of synthetic dyes," International Journal of Electrochemistry, vol. 2012, Article ID 153416, 18 pages, 2012.
[8] M. M. Sari, "Removal of acidic indigo carmine textile dye from aqueous solutions using radiation induced cationic hydrogels," Water Science and Technology, vol. 61, no. 8, pp. 2097–2104, 2010.
[9] M. R. Al-Mamun, M. N. Karim, and Z. H. Khan, "Photocatalytic performance assessment of GO and Ag co-synthesized TiO2nanocomposite for the removal of methyl orange dye under solar irradiation," International Journal of Electrochemistry, vol. 2012, Article ID 153416, 18 pages, 2012.
[10] X. Yu, A. Qin, L. Liao et al., "Removal of organic dyes by nanostructure ZnO-bamboo charcoal composites with photocatalysis function," Advances in Materials Science and Engineering, vol. 2015, Article ID 252951, 6 pages, 2015.
[11] P. Li, B. Gao, A. Li, and H. Yang, "Evaluation of the selective adsorption of silica-sand/anionized-starch composite for removal of dyes and Copper(II) from their aqueous mixtures," International Journal of Biological Macromolecules, vol. 149, no. 3, pp. 1285–1293, 2020.
[11] R. Dhahri, A. Bouzidi, and Y. Moussaoui, "Activated carbon from prickly pear seeds: optimization of preparation conditions and cadmium removal using experimental design approach," *Recent Advances in Environmental Science from the Euro-Mediterranean and Surrounding Region*, pp. 1457–1461, Springer, New York, NY, USA, 2nd edition, 2021.

[12] N. Khadiri, M. K. Saad, M. ben Mosbah, and Y. Moussaoui, "Batch and continuous column adsorption of indigo carmine onto activated carbon derived from date palm peels," *Journal of Environmental Chemical Engineering*, vol. 7, Article ID 102775, 2019.

[13] V. Karthik, P. Selvakumar, N. Sivarajasekar et al., "Comparative and equilibrium studies on anionic and cationic dyes removal by nano-alumina-doped catechol formaldehyde composite," *Journal of Chemistry*, vol. 2020, Article ID 7617989, 15 pages, 2020.

[14] P. H. Huang, H. H. Cheng, and S. H. Lin, "Adsorption of carbon dioxide onto activated carbon prepared from coconut shells," *Journal of Chemistry*, vol. 2015, Article ID 106590, 10 pages, 2015.

[15] J. C. M. Pirajlan and L. Giraldo, "Heavy metal ions adsorption from wastewater using activated carbon from orange peel," *Journal of Chemistry*, vol. 9, Article ID 383742, 12 pages, 2012.

[16] M. El Khames Saad, R. Khiairi, E. Elaloui, and Y. Moussaoui, "Adsorption of anthracene using activated carbon and Posidonia oceanica," *Arabian Journal of Chemistry*, vol. 7, no. 1, pp. 109–113, 2014.

[17] M. Moyo, L. Chikazaza, B. C. Nyamunda, and U. Guyo, "Adsorption batch studies on the removal of Pb(II) Using Maize tassel based activated carbon," *Journal of Chemistry*, vol. 2013, Article ID 508934, 8 pages, 2013.

[18] K. K. Naganathan, A. N. M. Faizal, M. A. A. Zaini, and A. Ali, "Adsorptive removal of Bisphenol a from aqueous solution using activated carbon from coffee residue," *Materials Today: Proceedings*, vol. 47, pp. 1307–1312, 2021.

[19] C. El-Guezzane, H. El-Moudden, H. Harhar, N. Chaboun, M. Tabyaoui, and A. Zarrouk, "A comparative study of the antioxidant activity of two Moroccan prickly pear cultivars collected in different regions," *Chemical Data Collections*, vol. 31, Article ID 100637, 2021.

[20] Y. Gao, Q. Yue, B. Gao, and A. Li, "Insight into activated carbon from different kinds of chemical activating agents: a review," *The Science of the Total Environment*, vol. 746, Article ID 141094, 2020.

[21] A. Melliti, V. Srivastava, J. Kheriji, M. Sillanpää, and B. Hamrouni, "Date Palm Fiber as a novel precursor for porous activated carbon: optimization, characterization and its application as Tylosin antibiotic scavenger from aqueous solution," *Surfaces and Interfaces*, vol. 24, Article ID 101047, 2021.

[22] N. Kannam, M. Acar, M. Yilmazoglu, and J. Hizal, "Rhodamine B and murexide retention onto sulfonated poly (ether ether ketone) (sPEEK)," *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, vol. 605, Article ID 125341, 2020.

[23] S. Davoodi, F. Marahel, M. Ghaedi, M. Roosta, and A. H. Jah, "Titan oxide nanoparticles loaded on activated carbon as adsorbent for removal of Murexide," *Desalination Water Treatment*, vol. 52, pp. 37–41, 2015.

[24] M. Ishaaq, K. Saeed, M. Shakirullah, I. Ahmad, and S. Sultan, "Removal of murexide from aqueous solution using pomegranate bark as adsorbent," *Journal of the Chemical Society of Pakistan*, vol. 34, pp. 1498–1501, 2012.

[25] R. Rehman, J. Anwar, T. Mahmud, and M. Salman, "Removal of murexide (dye) from aqueous media using rice husk as an adsorbent," *Journal of the Chemical Society of Pakistan*, vol. 33, pp. 598–603, 2011.

[26] A. Shokrollahi, M. Ghaedi, M. Ranjbar, and A. Alizadeh, "Kinetic and thermodynamic studies of the removal of murexide from aqueous solutions on to activated carbon," *Journal of the Iranian Chemical Society*, vol. 3, pp. 219–235, 2010.

[27] G. Newcombe, R. Hayes, and M. Drikas, "Granular activated carbon: importance of surface properties in the adsorption of naturally occurring organic contaminants," *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, vol. 78, pp. 65–71, 1993.

[28] Q. Ji and H. Li, "High surface area activated carbon derived from chitin for efficient adsorption of Crystal Violet," *Diamond and Related Materials*, vol. 118, Article ID 108516, 2021.

[29] F. Taleb, M. ben Mosbah, E. Elaloui, and Y. Moussaoui, "Adsorption of ibuprofen sodium salt onto Amberlite resin IRN-78: kinetics, isotherm and thermodynamic investigations," *Korean Journal of Chemical Engineering*, vol. 34, no. 4, pp. 1141–1148, 2017.

[30] J. Qian, M. Shen, P. Wang et al., "Perfluorooctane sulfonate adsorption on powder activated carbon: effect of phosphate (P) competition, pH, and temperature," *Chemosphere*, vol. 182, pp. 215–222, 2017.

[31] M. E. K. Saad, N. Mnasri, M. Mhamdi, T. Chaﬁk, E. Elaloui, and Y. Moussaoui, "Removal of methylene blue on activated carbon derived from Pistacia kahni," *Desalination and Water Treatment*, vol. 56, no. 10, pp. 2773–2780, 2015.

[32] J. M. Salman, "Preparation of mesoporous-activated carbon from branches of pomegranate trees: optimization on removal of methylene blue using response surface methodology," *Journal of Chemistry*, vol. 2013, Article ID 489670, 6 pages, 2013.

[33] M. Ghaedi, A. M. Ghaedi, F. Abdi, M. Roosta, A. Vafaei, and A. Asghari, "Principal component analysis- adaptive neuro-fuzzy inference system modeling and genetic algorithm optimization of adsorption of methylene blue by activated carbon derived from Pistacia kahni," *Ecotoxicology and Environmental Safety*, vol. 96, pp. 110–117, 2013.

[34] Y. S. Ho, G. McKay, D. A. J. Wase, and C. F. Forster, "Study of the sorption of divalent metal ions on to peat," *Adsorption Science and Technology*, vol. 18, no. 7, pp. 639–650, 2000.

[35] X. Guo and J. Wang, "Comparison of linearization methods for modeling the Langmuir adsorption isotherm," *Journal of Molecular Liquids*, vol. 296, Article ID 111850, 2019.

[36] M. Araissi, I. Ayed, E. Elaloui, and Y. Moussaoui, "Removal of barium and strontium from aqueous solution using zeolite 4A," *Water Science and Technology*, vol. 73, no. 7, pp. 1628–1636, 2016.

[37] H. R. Pouretedal and N. Sadegh, "Effective removal of Amoxicillin, Cephalexin, Tetracycline and Penicillin G from aqueous solutions using activated carbon nanoparticles prepared from pine wood," *Journal of Water Process Engineering*, vol. 1, pp. 64–73, 2014.

[38] P.-J. Lu, H.-C. Lin, W.-T. Yu, and J.-M. Chern, "Chemical regeneration of activated carbon used for dye adsorption," *Journal of the Taiwan Institute of Chemical Engineers*, vol. 42, no. 2, pp. 305–311, 2011.

[39] L. R. Bonetto, J. S. Crespo, R. Guégan, V. I. Esteves, and M. Giovanela, "Removal of methylene blue from aqueous solutions using a solid residue of the apple juice industry: full factorial design, equilibrium, thermodynamics and kinetics
aspects,” *Journal of Molecular Structure*, vol. 1224, Article ID 129296, 2021.
[40] G. Jariz, C. P. Silva, J. A. B. P. Oliveira et al., “Production of highly efficient activated carbons from industrial wastes for the removal of pharmaceuticals from water-A full factorial design,” *Journal of Hazardous Materials*, vol. 370, pp. 212–218, 2019.
[41] S. A. Kordkandi and M. Forouzesh, “Application of full factorial design for methylene blue dye removal using heat-activated persulfate oxidation,” *Journal of the Taiwan Institute of Chemical Engineers*, vol. 45, no. 5, pp. 2597–2604, 2014.
[42] S. Bhownik, V. Chakraborty, and P. Das, “Batch adsorption of indigo carmine on activated carbon prepared from sawdust: a comparative study and optimization of operating conditions using Response Surface Methodology,” *Results in Surfaces and Interfaces*, vol. 3, Article ID 100011, 2021.
[43] L. Tsechansky and E. R. Graber, “Methodological limitations to determining acidic groups at biochar surfaces via the Boehm titration,” *Carbon*, vol. 66, pp. 730–733, 2014.
[44] I. I. Salame and T. J. Bandoz, “Surface Chemistry of activated carbons: combining the results of temperature-programmed desorption, Boehm, and potentiometric titrations,” *Journal of Colloid and Interface Science*, vol. 240, no. 1, pp. 252–258, 2001.
[45] E. Misran, S. Magid, M. Irvan, S. F. Dina, S. A. A. Harahap, and A. Nizar, “Activated carbon preparation from bagasse and banana stem at various impregnation ratio,” *Journal of Physics: Conference Series*, vol. 1542, Article ID 012068, 2020.
[46] Q. Han, J. Wang, B. A. Goodman, J. Xie, and Z. Liu, “High adsorption of methylene blue by activated carbon prepared from phosphoric acid treated eucalyptus residue,” *Powder Technology*, vol. 366, pp. 239–248, 2020.
[47] D. Mihayo, M. R. Vegi, and S. A. H. Vuai, “Defluoridation of aqueous solution using thermally activated biosorbents prepared from *Adansonia digitata* fruit pericarp,” *Adsorption Science and Technology*, vol. 2021, Article ID 5574900, 16 pages, 2021.
[48] G. Revathi, S. Ramalingam, P. Subramaniam, and A. Ganapathi, “Removal of direct yellow-12 dye from water by adsorption on activated carbon prepared from *Ficus racemosa* L.,” *Journal of Chemistry*, vol. 8, Article ID 902421, 10 pages, 2011.
[49] K. Mahmoudi, K. Hosni, N. Hamdi, and E. Srasra, “Kinetics and equilibrium studies on removal of methylene blue and methyl orange by adsorption onto activated carbon prepared from date pits-A comparative study,” *Korean Journal of Chemical Engineering*, vol. 32, no. 2, pp. 274–283, 2015.
[50] I. D. Mall, V. C. Srivastava, G. V. A. Kumar, and I. M. Mishra, “Characterization and utilization of mesoporous fertilizer plant waste carbon for adsorptive removal of dyes from aqueous solution,” *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, vol. 278, Article ID 175, 2006.
[51] A. Akil, S. Mohd-Setapar, C. Chuo Sing, A. Khatoon, and W. A. Wani, R. Kumar and M. Rafatullah, Recent advances in new generation dye removal technologies: novel search for approaches to reprocess wastewater,” *RSC Advances*, vol. 5, no. 39, pp. 30801–30818, 2015.
[52] M. Niyaz and M. Adonis, “Kinetics and isotherm of cationic dye removal from multicomponent system using the synthesized silica nanoparticle,” *Desalination and Water Treatment*, vol. 54, no. 2, pp. 562–571, 2015.
[53] Y. El Maguana, N. Elhadiri, M. Benchanaa, and R. Chikri, “Activated carbon for dyes removal: modeling and understanding the adsorption process,” *Journal of Chemistry*, vol. 2020, Article ID 2096834, 9 pages, 2020.
[54] M. A. Ahmad, S. G. Herawan, and A. A. Yusof, “Equilibrium, kinetics, and thermodynamics of remazol brilliant blue R dye adsorption onto activated carbon prepared from pinang frond,” *International Scholarly Research Notices*, vol. 2014, Article ID 184265, 7 pages, 2014.
[55] I. Leng, X. Yuan, G. Zeng et al., “Surface characterization of rice husk bio-char produced by liquefaction and application for cationic dye (Malachite green) adsorption,” *Fuel*, vol. 155, pp. 77–85, 2015.
[56] A. V. Borhade and A. S. Kale, “Calcinied eggshell as a cost effective material for removal of dyes from aqueous solution,” *Applied Water Science*, vol. 7, no. 8, pp. 4255–4268, 2017.
[57] S. Salvestrini, V. Leone, P. Iovino, S. Canzano, and S. Capasso, “Considerations about the correct evaluation of sorption thermodynamic parameters from equilibrium isotherms,” *The Journal of Chemical Thermodynamics*, vol. 68, pp. 310–316, 2014.
[58] A. M. Elgarahy, K. Z. Elwakeel, G. A. Elshoubaky, and S. H. Mohammad, “Microwave-accelerated sorption of cationic dyes onto green marine algal biomass,” *Environmental Science and Pollution Research*, vol. 26, no. 22, pp. 22704–22722, 2019.
[59] D. Mitrogiannis, G. Markou, A. Çelekli, and H. Bozkurt, “Biosorption of methylene blue onto Arthospira platensis biomass: kinetic, equilibrium and thermodynamic studies,” *Journal of Environmental Chemical Engineering*, vol. 3, no. 2, pp. 670–680, 2015.
[60] H. A. Ahsaine, Z. Anfar, M. Zbair, M. Ezahri, and N. El Alem, “Adsorptive removal of methylene blue and crystal violet onto micro-mesoporous ZrO/activated carbon composite: a joint experimental and statistical modeling considerations,” *Journal of Chemistry*, vol. 2018, Article ID 6982014, 14 pages, 2018.
[61] S. Teixeira, C. Delerue-Matos, and L. Santos, “Application of experimental design methodology to optimize antibiotics removal by walnut shell based activated carbon,” *The Science of the Total Environment*, vol. 646, pp. 168–176, 2019.
[62] M. R. Parsaean, S. Dadfarinia, A. M. Haji Shabani, and R. Hafezi Moghaddam, “Green synthesis of a high capacity magnetic polymer nanocomposite sorbent based on the natural products for removal of Reactive Black 5,” *International Journal of Environmental Analytical Chemistry*, vol. 100, pp. 1–15, 2020.
[63] M. K. Satapathy and P. Das, “Optimization of crystal violet dye removal using novel soil-silver nanocomposite as nanoadsorbent using response surface methodology,” *Journal of Environmental Chemical Engineering*, vol. 2, no. 1, pp. 708–714, 2014.
[64] H. Mazaheri, M. Ghaedi, A. Asfaram, and S. Hajati, “Performance of CuS nanoparticle loaded on activated carbon in the adsorption of methylene blue and bromophenol blue dyes in binary aqueous solutions: using ultrasound power and optimization by central composite design,” *Journal of Molecular Liquids*, vol. 219, pp. 667–676, 2016.
[65] A. Naghizadeh, F. Momeni, and H. Kamani, “Study of ultrasonic regeneration and adsorption of humic acid on activated carbon,” *Health scope*, vol. 7, Article ID 80338, 2018.
[66] J. Wang, X. Han, H. Ma, Y. Ji, and L. Bi, “Adsorptive removal of humic acid from aqueous solution on polyaniline/attapulgite composite,” *Chemical Engineering Journal*, vol. 173, no. 1, pp. 171–177, 2011.
[67] A. Khasri, M. R. M. Jamir, A. A. Ahmad, and M. A. Ahmad, “Adsorption of remazol brilliant violet 5R dye from aqueous
solution onto melunak and rubberwood sawdust based activated carbon: interaction mechanism, isotherm, kinetic and thermodynamic properties,” Desalination and Water Treatment, vol. 216, pp. 401–411, 2021.

[68] A. Kanwal, H. N. Bhatti, M. Iqbal, and S. Noreen, “Basic dye adsorption onto clay/MnFe2O4 composite: a mechanistic study,” Water Environment Research, vol. 89, 2017.

[69] R. Eszter and S. Tonk, “Factors affecting synthetic dye adsorption; desorption studies: a review of results from the last five years (2017–2021),” Molecules, vol. 26, Article ID 5419, 2021.