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Study of the conditions of activated carbon preparation from an agriculture by-product for 4BA elimination in aqueous solution using full factorial design

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ABSTRACT

An activated carbon prepared from an agricultural by-product (apricot stones) is used as an adsorbent for 4-bromoaniline (4BA) elimination. Several activation chemical agents are tested; the hydrochloric acid is found to be the best choice with 79% of elimination yield. The main and interactive effects of four experimentally controlled elaboration activated carbon factors, which are the carbonization temperature, the carbonization time, the concentration of activating agent (% HCl), and the activation time, are investigated through model equations designed with a two-level full factorial design by using a batch experimental method. Statistical design of experiments for the manufacture of activated carbon from apricot stones for 4BA adsorption is an efficient and rapid technique to quantify the effect of variable parameters compared with the conventional optimization. The characteristics of the optimal adsorbent are studied using scanning electron microscope, energy dispersive X-ray analysis, and Brunauer–Emmett–Teller nitrogen adsorption technique. The equilibrium adsorption isotherms have been analyzed with the Langmuir and Freundlich models at 28°C.

Keywords: Adsorption; Apricot stone carbon; 4-Bromoaniline; Factorial design

1. Introduction

Aniline and its derivatives are organic intermediaries that considered as potential pollutants. Their presence in water at very low concentrations is harmful to aquatic life [1]. Various processes of water treatment have been developed for removing these pollutants from aqueous effluents. Adsorption is one of the purification and separation techniques used in this area. Algeria produces yearly 80,000 tons of apricot; therefore, a large amount of apricot stones is generated, and the conversion of this waste into adsorbent allows the valorization of agricultural residue and the preparation of activated carbon to clean water. Two steps are required to convert an apricot stones to an activated carbon: (i) the activation and (ii) the carbonization. The process of activated carbons preparation depends upon the nature of the raw materials, the nature of the activating reagent, and the conditions of carbonization. A large number of low-cost adsorbents such as skim almond [2], olives

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stone [3], almond hulls [4], peach stone [5], apricot stone [6] date stone [7,8], cherry stones [9] anaerobic granular biomass [10], tea waste [11], spent-grain [12], walnut, hazelnut, almond, pistachio shell, and apricot stone [13], modification of activated carbon using sodium acetate [14], spent activated clay [15], wood sawdust [16], nonliving lichen biomass of *Cladonia rangiformis* Hoffm. [17], seaweeds [18], etc. have been reported in the literature for the removal of organic and inorganic molecules.

Many researches are devoted to using the apricot stones as raw materials to prepare an activated carbon. In these studies, the effect and the optimization of various parameters on preparation conditions are examined using the classical method. The process variables studied, using this method, are the carbonization temperature and activating agent nature [19], activating agent nature and activation time [2]. One of disadvantage of this approach is that it does not take into account the interaction effect between factors in the optimization procedure. To overcome this, the statistical methods were used successfully. Response surface methodology (RSM) has been found to be a useful tool to study the interactions of two or more variables [20]; Tan et al. optimized the manufacturing conditions for activated carbon using RSM, the variables studied are activation temperature, activation time, and KOH impregnation ratio [21]. Optimization of experimental conditions using RSM is widely applied in various processes, and the previous studies found in applying RSM in preparation of activated carbons are using precursors, such as biochar [22], olive-waste cakes [23], and Luscar char [24]. Optimization of experimental conditions using a two-level full factorial design is widely applied in various processes [25,26], so a limited number of studies that are in related to the optimization of activated carbon preparation conditions using this method [27,28]; however, its application in stone apricot activated carbon production is very rare.

The main purpose of this present work is to describe an application of statistical method for optimization the conditions of the activated carbon preparation from agricultural by-product (apricot stone), locally available and its application for the water treatment, polluted by 4-bromoaniline (4BA). The most important independent variables that have a strong influence on the activated carbon preparation were determined and evaluated by Full Factorial Design; the simulated conditions of the best adsorption yield have been identified from iso-response curves.

2. Experimental procedure

All chemicals used are of analytical grade purity and without further purification.

2.1. Materials and reagents

4BA (98.0%, Aldrich) is used as adsorbate. HCl (37%, Biochem) is used as activating agent. Distilled water (0.7 M Ω) is used to prepare all solutions.

2.2. Preparation and characterization of activated carbon

The apricot stones (Family: Rosacée) are thoroughly washed with water and dried in air oven at 105˚C for 24 h. The precursor is treated with hydrochloric acid during a time (t) in an agitated reactor (capacity 500 mL), equipped with a cooling system to maintain the acid concentration constant. The activated precursor is carbonized in a thermally controlled muffle furnace with a linear rise of 5˚C $\min^{-1}$ under a current of nitrogen (150 $cm^3 \min^{-1}$). The treated sample is repeatedly washed with water until getting neutral pH and dried at 105˚C for 24 h, after which they is ground in an agate mortar and sieved. The grains with sizes less than 320 μm are stored in bottles.

The Brunauer-Emmett-Teller (BET) method is used for specific surface area measurement, such as for the sorption phenomenon of nitrogen gas on the adsorbent surface at the temperature of liquid nitrogen (77 K). The morphological analysis and the energy dispersive X-ray analysis (EDX) of the studied sorbent are performed via scanning electron microscopy (SEM) using a Philips XL 20 Welton Joanne, 1950.

The point of zero charge (pH_{PZC}), commonly known as isoelectric point, is the pH value at which either the sorbent surface has a net neutral charge or the difference between cation-exchange capacity and anion-exchange capacity is zero [29]. When the pH is lower than pH_{PZC}, the adsorbent surface is positively charged (thus attracting anions). Conversely, above pH_{PZC}, the surface is negatively charged (attracting cations/repelling anions). In order to understand what kind of interactions exist between carbon surface and charged soluble species, we determine pH_{PZC} using the following method. Distilled water is used to prepare a 0.1 M solution of $NaNO_3$. From which ten different standards with pH ranging from 2 to 11 are made. The pH is adjusted using either 0.1 M NaOH or 0.1 M HCl. Activated carbon (0.1 g) is mixed with 50 mL of each solution during 24 h in sealed vials. The final pH is measured and plotted vs. the initial solution pH. The pH at the point of zero charge, pH_{PZC}, is determined as the pH of the $NaNO_3$ solution that does not change after contact with the carbon [30].

The amount of oxygen containing functional groups with acidic character on the carbon surface is determined applying the Boehm method [31]. The basicity is determined by introducing 0.2 g of sample in a well-sealed flask together with $25\,cm^3$ of 0.02 M HCl solution. The mixture is then shaken for 48 h at ambient temperature. The resulting suspension is filtered, and the residual HCl is titrated against a 0.02 M NaOH solution. Acidity is determined in a similar way using a solution of NaOH (0.02 M) instead of 0.02 M HCl. The solution is mixed with the activated carbon and titrated against solution was 0.02 M HCl [31].

2.3. Sorption tests

All adsorption tests are carried out in batch mode according to the same experimental protocol. A 100 mL aqueous solution of 4BA ($100\,mg\,L^{-1}$, pH 6 = 6.8) is agitated (300 rpm) with activated carbon ($1\,g\,L^{-1}$) for 60 min whose temperature is regulated at 28 °C. The aliquots are withdrawn at regular times and subjected to vigorous centrifugation (Nahita—Centrifuge model 2698). The residual 4BA concentration is analyzed by a double-beam spectrophotometer (UV–Visible, SAFAS, EASYPEC II 320 D) at 240 nm. The adsorption yield (Y %) is calculated via (Eq. (1)).

$$Y(\%) = \frac{(C_0 - C_t)}{C_t} \times 100 \tag{1}$$

where C_0 is the initial concentration of the solution ($mg\,L^{-1}$) and C_t is the concentration of the solution at time t ($mg\,L^{-1}$).

2.4. Design of experiments

The full factorial design 2^4 is applied to optimize the effective parameters and to analyze their interaction. It is a collection of mathematical model in the form of a first-degree polynomial, and statistical technique useful for modeling and analyzing the problems in which a response of interest is influenced by several variables [32]. The studied variables are (i) X_1, Carbonization temperature; (ii) X_2, carbonization time; (iii) X_3, concentration of activating agent, and (iv) X_4 activation time. These variables together with their respective ranges are chosen on the basis of the literature data and preliminary experiments. They affect the characteristics of the activated carbon production [33,34]. The effectiveness and the performance of the activated carbon are tested for their capacities to eliminate 4BA. The experimental results are presented by the mathematical model:

$$\begin{aligned} Y = A_o &+ \sum_{i=1}^{n} A_i X_i + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} A_{ij} X_i X_j \\ &+ \sum_{i=1}^{n-2} \sum_{j=i+1}^{n-1} \sum_{k=j+1}^{n} A_{ijk} X_i X_j X_k \\ &+ \sum_{i=1}^{n-3} \sum_{j=i+1}^{n-2} \sum_{k=j+1}^{n-1} \sum_{l=k+1}^{n} A_{ijkl} X_i X_j X_k X_l \end{aligned} \tag{2}$$

where Y is the predicted response (4BA adsorption yield), A_o a constant coefficient; A_i the linear coefficients; A_{i-1} the interaction coefficients; X_i, X_j, X_k, and X_l are the coded values of the activated carbon preparation variables.

3. Results and discussion

3.1. Simulation of optimal conditions

Generally, four main parameters are studied for the activated carbon prepared from lignocellulosic derivatives [35,36]. The investigation is carried out according to the full factorial experiment at two levels. The experimental data have the coded values (−1) and (+1), respectively, for lower and higher levels. The levels along with the study ranges are given in Table 1.

3.1.1. Planning of adsorption tests

An experimental design with four parameters requires 2^4 (=16) tests, and these experiments are

Table 1
Factor levels used in the experiments

Parameters	Designation	High level (+1)	Low level (−1)
Carbonization temperature T (°C)	X_1	700	800
Carbonization time t_{car} (h)	X_2	1	2
Concentration of activating agent	X_3	10	20
Activation time t_{act} (h)	X_4	2	3

Table 2
Experimental design and adsorption yields

Experiment	T (℃)	t_{car} (h)	C_{HCl} (%)	t_{act} (h)	X_1	X_2	X_3	X_4	Y
1	700	1	10	2	−	−	−	−	26.96
2	800	1	10	2	+	−	−	−	52.81
3	700	2	10	2	−	+	−	−	43.44
4	800	2	10	2	+	+	−	−	49.95
5	700	1	20	2	−	−	+	−	50.11
6	800	1	20	2	+	−	+	−	71.99
7	700	2	20	2	−	+	+	−	60.97
8	800	2	20	2	+	+	+	−	79.92
9	700	1	10	3	−	−	−	+	29.59
10	800	1	10	3	+	−	−	+	48.44
11	700	2	10	3	−	+	−	+	46.81
12	800	2	10	3	+	+	−	+	47.22
13	700	1	20	3	−	−	+	+	54.24
14	800	1	20	3	+	−	+	+	73.94
15	700	2	20	3	−	+	+	+	67.72
16	800	2	20	3	+	+	+	+	77.70

conducted according to the matrix of experience (Table 2), which is a combination between the four studied parameters; this requires the preparation of 16 activated carbons. The yield of the 4BA adsorption is given in the last column.

3.1.2. Development of regression model

To study the influence of the main variables and their interactions, we have used the Yates method which yields a system of 16 equations and 16 unknown variables [37,38]. Such system is presented in the form of matrixes product and the data are statistically treated by using the MODDE 6.0 software. The mathematical equation is expressed as follows:

$$\begin{aligned} Y = {} & 55.11 + 7.63X_1 + 4.10X_2 + 11.96X_3 + 0.59X_4 \\ & - 3.15X_1X_2 + 1.18X_1X_3 - 1.52X_1X_4 \\ & + 0.40X_2X_3 + 0.05X_2X_4 + 0.73X_3X_4 \end{aligned} \tag{3}$$

The regression equation (Eq. (3)) shows that each of the carbonization temperature, the carbonization time, the concentration of activating agent (HCl), and the activation time has an individual effect on 4BA adsorption yield. Statistical design of experiments is an efficient technique to quantify the effect of various parameters. The model fitness quality is evaluated via analysis of variance (ANOVA). The regression coefficient R^2 quantitatively evaluates the correlation between the experimental data and the predicted response. In our study, the R^2 value is 0.988. The obtained R^2 value suggests a good fit to the experimental data and indicates that 98.8% of the variability in the response may be explained by the model. If the curve R^2 value is larger than 0.90, the system may be considered linear [39]. The coefficient of determination R^2 which measures the quality of the model is defined by the following expression [40].

$$R^2 = \frac{\left(\sum_{i=0}^{16}(Y_{i\,\mathrm{exp}} - \overline{Y_{\mathrm{exp}}})(Y_{i\mathrm{th}} - \overline{Y_{\mathrm{th}}})\right)^2}{\sum_{i=0}^{16}(Y_{i\,\mathrm{exp}} - \overline{Y_{\mathrm{exp}}})^2 \sum_{i=0}^{16}(Y_{i\mathrm{th}} - \overline{Y_{\mathrm{th}}})^2} \tag{4}$$

According to Eq. (4), R^2 is equal to 0.99 and is close to 1. Therefore, the model enables one to find the measured values. Adjusted R^2 (Adj R^2) is also a measure of goodness of fit, but it is more suitable for comparing models with different numbers of independent variables. Adjusted R^2 may be visibly smaller than R^2 [41]. The adjusted R^2 value (0.963) is very close to the corresponding R^2 value (0.988). The value of Adj R^2 (0.963) being close to 1 indicates a high degree of correlation between the observed and predicted values. The ANOVA, which tests the significance and the adequacy of the regression model, is given in Table 3.

ANOVA subdivides the total variation of the results into two components: variation associated with the model and variation associated with the experimental error. This shows whether the variation from the model is significant or not when compared with the variations associated with the residual error [41].

Table 3
Analyze variances for the rate of adsorption (Table ANOVA)

Sources of variation	Degrees of freedom	Sum of squares	Mean square	F-ratio	Prob > F
Total	16	52371.20	3273.20	40.03	0.00001
Constant	1	48599.30	48599.30		
Total corrected	15	3771.89	251.46		
Regressions	10	3725.35	372.54		
Residual	5	46.54	9.31		

This comparison is performed via the F-value, which is the ratio between the mean square of the model and the residual error. If the model is a good predictor of the experimental results, the F-value should be larger than the tabulated value [41]. The F-value obtained for 4BA adsorption is 40.0257. Clearly this value is larger than the tabulated F value (2.5 at 95% significance), confirming the adequacy of the model. When Prob > F all the variations in the results are due to the random error [41]. Thus, the very low value obtained for 4BA (<0.00001) indicates that the result is not random and the terms in the model have a significant effect on the response [41]. The P-values are used as a tool to check the coefficient significance and indicate the importance of the interaction between the variables.

The significance of each coefficient is determined via either the t-value or the P-value which are listed in Table 4. The smaller the magnitude of the P-value, the more significant is the corresponding coefficient [42].

Fig. 1 shows that the first-order main effects of various parameters (A_1, A_2, A_3) are more significant than their collaborate effect (A_{12}, A_{13}, A_{23}) interaction. The effect of activating agent concentration (C_{HCl}), x_3, is very important ($p < 0.00001$). This suggests that carbonization temperature, duration of carbonization, and activating agent concentration have a direct relationship with the 4BA elimination. In addition, the collaborate effect A_{12} ($p < 0.009$) is more significant than ones. The 4BA elimination yield for various variable values can also be predicted via Eq. (5):

$$Y = 55.11 + 7.63X_1 + 4.10X_2 + 11.96X_3 - 3.15X_1X_2 \quad (5)$$

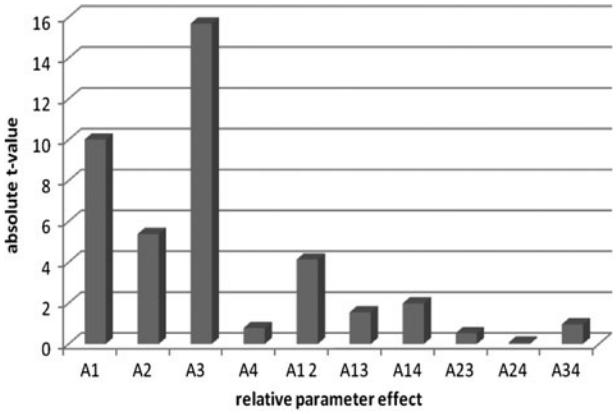

Fig. 1. Comparison of relative parameter effects.

Table 4
Estimated effects, standard errors, and student's t-test for 4BA yield of removal (%) using 2^4 full factorial design

Model term	Coefficient estimate	t-value	Std. err.	$P < \mid t \mid$
Constant intercept	55.11	72.26	0.762	0.0000
X_1	7.63	10.01	0.762	0.00017
X_2	4.10	5.38	0.762	0.00299
X_3	11.96	15,68	0.762	0.00001
X_4	0,59	0.78	0.762	0.47106
X_1X_2	−3.15	−4.13	0.762	0.00906
X_1X_3	1.18	1.54	0.762	0.18231
X_1X_4	−1.51	−1.99	0.762	0.10362
X_2X_3	0.40	0.52	0.762	0.62185
X_2X_4	0.052	0.068	0.762	0.94841
X_3X_4	0.73	0.96	0.762	0.38133

The 4BA elimination yield can also be predicted by the plots (Figs. 2(a–c)) for several variable values. Each curve represents an infinite number of combinations of two test variables with the third variable held constant. The effect of both activating agent concentration and carbonization temperature on the elimination yield is shown in Fig. 2(a) while the effect of carbonization temperature and carbonization time is displayed in Fig. 2(b).

In both cases, the elimination yield increases with increasing parameter values. Fig. 2(c) shows that when both carbonization temperature and carbonization time are held constant, the elimination of 4BA increases with the concentration of activating agent, while when both carbonization temperature and concentration of activating agent are held constant, the carbonization time does not affect much the elimination yield.

The mathematical model (Eq. (5)) can be used to simulate the optimal conditions for the preparation of activated carbon (carbonization temperature, carbonization time, concentration of activating agent and activation time).

In the parameter variation range, the best yield of 4BA elimination (79%) is obtained for the following conditions: $X_1 = +1$ ($T = 800^\circ C$), $X_2 = +1$ ($t_{car} = 2\,h$), $X_3 = +1$ ($C_{HCl} = 20\%$) and $X_4 = -1$ ($t_{act} = 2\,h$), these results correspond to the experiment N°8 (Table 2).

3.2. Characterization of optimal activated carbon

The activated carbon obtained under the optimal conditions has been characterized. The main physical and chemical properties are evaluated via surface area, IR spectroscopy, SEM, EDX analysis, and isoelectric point. The results are given in Table 5.

In order to know the surface structure of untreated and treated adsorbents, morphological analysis is performed by SEM. Fig. 3 shows the SEM micrographs of untreated and treated samples. It can be seen that the chemically treated adsorbent surface exhibits a heterogeneous type pores. It is revealed that chemically treated adsorbent is porous as compared to untreated adsorbent. The surface area of untreated apricot stones determined by BET method is $45\,m^2\,g^{-1}$ while the surface area of the treated adsorbent is

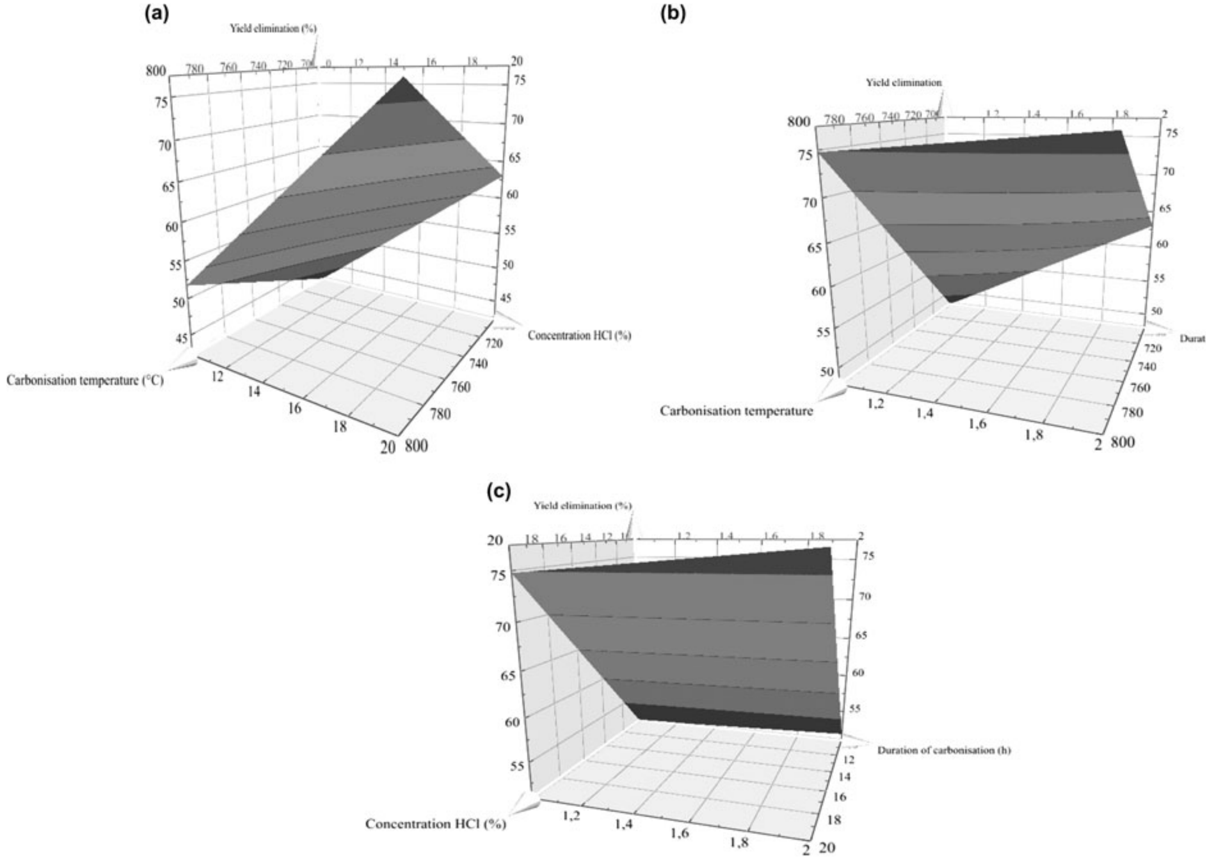

Fig. 2. Isoresponse curves for (a) duration of carbonization 2 h and duration of activation 2 h, (b) HCl concentration 20% and duration of activation 2 h, (c) carbonization temperature 800°C and duration of activation 2 h.

Table 5
Characteristics of coal treated with hydrochloric acid

Average diameter of particles (μm)	150
pH_{pzc}	6.20
Apparent density ($g\,cm^{-3}$)	0.57
Moisture (%)	3
Ash (%)	4.10
Total basicity ($mol\,g^{-1}$)	0.008
Total acidity ($mol\,g^{-1}$)	0.048
S_{BET} (m^2/g)	399
S_{mic} (m^2/g)	375

$399\,m^2\,g^{-1}$. This analysis shows that hydrochloric acid is effective in creating pores on the surface of the precursor, and similar result has been reported by Mui et al. [43].

As seen in Table 6, the EDX microanalysis shows that the treated adsorbent has higher contents of carbon and oxygen than the untreated one, suggesting that the former possesses more surface oxygen-containing functional groups [44]. This is also exhibited by Boehm titration, for which the treated adsorbent has higher contents of acidic functional groups than the untreated adsorbent. The treated adsorbent has $0.0481\,mol\,g^{-1}$of acidic surface groups, which is more than the untreated adsorbent ($0.0035\,mol\,g^{-1}$). This is further confirmed by the lower pH_{PZC} value (6.20) of the treated adsorbent (7.34 for the untreated adsorbent). The activation with hydrochloric acid leads the considerable increase in the amount of acid groups, which is in a good agreement with the pH value. For the treated adsorbent, the content of chlorine increases due to the treatment with hydrochloric acid. It should be noted that the treated adsorbent has relatively low P, K, Mg, and Zn contents and low sulfur content, which makes it an appropriate material for purification processes

4. Adsorption isotherm

The adsorption isotherm is important to describe how solutes interact with adsorbents and is critical in optimizing the use of adsorbents. The evolution of the adsorbed quantity according to the residual concentration at equilibrium (Fig. 4) shows that, for the considered adsorbent, the isotherm obtained is of the type I. Several isotherms are available for such analysis. Most often, two models are applied for the equilibrium description of the systems organic compounds adsorption behavior.

4.1. Langmuir model

The Langmuir model is the simplest theoretical model for monolayer adsorption onto a surface with a finite number of identical sites. This model predicts the maximum monolayer adsorption capacity of the adsorbent and also determines if the adsorption is favorable or not. It is used in several forms, in particular, in the form of a hyperbolic function:

$$Q_e = \frac{b \times Q_o \times C_e}{1 + b \times C_e} \tag{6}$$

when linearized Eq. (6) becomes

$$\frac{1}{Q_e} = \frac{1}{Q_o} + \frac{1}{b} \times \frac{1}{Q_o C_e} \tag{7}$$

where C_e is the adsorbate concentration in the bulk solution at equilibrium ($mg\,L^{-1}$), Q_e is the amount of solute adsorbed per unit weight of adsorbent at

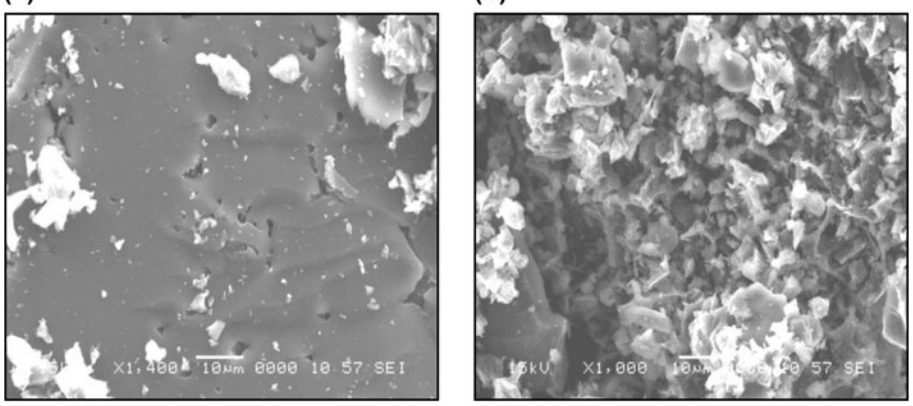

Fig. 3. The SEM photograph of untreated (a) and treated apricot stones (b).

Table 6
Elemental composition of untreated and treated adsorbent

Element	C	O	Mg	P	Cl	K	Ca	S	Zn
Untreated adsorbent (%)	89.96	2.87	0.86	0.50	0.05	1.36	0.10	2.85	1.45
Treated adsorbent (%)	91.37	5.80	0.28	0.40	0.14	0.58	0.52	0.57	0.35

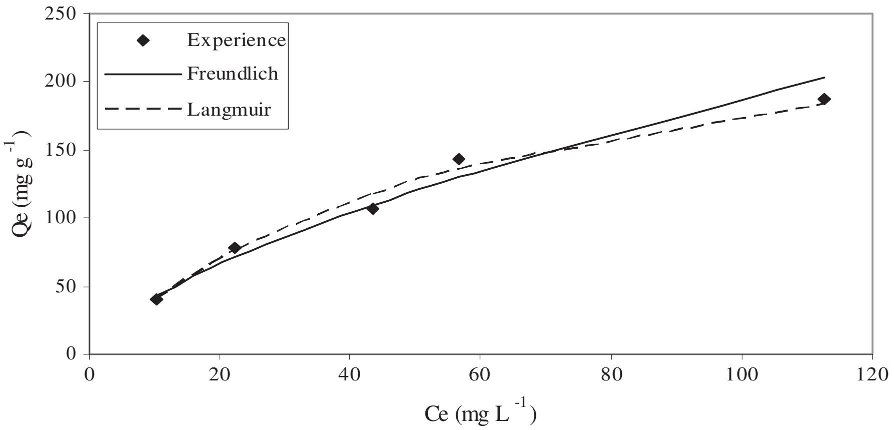

Fig. 4. Adsorption isotherms for 4BA on optimal activated carbon (pH = 6.8, [m] = 1 g L^{-1}, contact time = 60 min, T = 28°C).

Table 7
Values of Langmuir adsorption capacity (Q_0)

Adsorbate	Q_0 (mg g^{-1})	References
Astrazon Blue FGRL	181.50	[46]
Astrazon Yellow 7GL	134.59	[47]
Hg(II)	153.00	[48]
4BA	285.71	This work

equilibrium (mg g^{-1}), Q_o and b are the Langmuir constants related to adsorption capacity and rate of adsorption, respectively.

The constants can be evaluated from the intercept and the slope of the linear plots of $1/Q_e$ vs. $1/C_e$ [45]. The Langmuir isotherm model is found to fit the data reasonably with a correlation coefficient of $R^2 = 0.996$. The values of Q_o and b from this graph are found to be 285.71 mg g^{-1} and 0.0159 L mg^{-1}. Table 7 summarizes the values of Q_o obtained in other works using apricot stone activated carbon [46–48].

4.2. Freundlich model

The Freundlich Eq. (8) is an empirical equation employed to describe heterogeneous systems characterized by the heterogeneity factor $1/n$. The empirical equation can be written as follows[49]:

$$Q_e = K_f Ce^{\frac{1}{n}} \tag{8}$$

where C_e is the equilibrium concentration of the solute (mg L^{-1}), Q_e is the amount of solute adsorbed per unit mass of adsorbent (mg g^{-1}), K_f and n are Freundlich constants with n giving an indication of how favorable the adsorption process is and K_f (mg g^{-1}(mg $L^{-1})^{-1/n}$) is the adsorption capacity of the adsorbent. K_f can be defined as the adsorption or distribution coefficient and represents the quantity of 4BA adsorbed onto activated carbon for a unit equilibrium concentration. A linear form of the Freundlich expression can be obtained by taking the logarithm on both sides of Eq. (9):

$$\ln Q_e = \ln K_{\mathrm{f}} + \frac{1}{n} \ln Ce \tag{9}$$

Therefore, a plot of ln Q_e vs. ln C_e enables the constant K_f and exponent $1/n$ to be determined. The Freundlich isotherm describes reversible adsorption and is not restricted to the formation of the monolayer. It predicts that the 4BA concentration on the adsorbent will increase so long as there is an increase in the 4BA concentration in the liquid. The slope of $1/n$ ranging between 0 and 1 is a measure of adsorption intensity or surface heterogeneity, becoming more heterogeneous as its value gets closer to zero [45,50]. The value of the correlation coefficient, R^2 is 0.979. The value of $1/n$ is found to be 0.648 and indicates a favorable adsorption, whereas the value of K_f is found to be 9.468 ($\mathrm{mg\,g^{-1}(mg\,L^{-1})^{-1/n}}$). Gonzalez Pradas reported that the Freundlich adsorption constants (K_f and n) for 4BA on the calcareous soil were found to be 5.79 $\mathrm{mg\,g^{-1}(mg\,L^{-1})^{-1/n}}$ and 0.60, respectively [1]. Fig. 4 shows the fitted equilibrium (at 28˚C) data with the Freundlich and the Langmuir expressions. It can be seen that the Langmuir curve fits the data better than the Freundlich model. This is also confirmed by the high value of R^2 for Langmuir (0.996) in contrast to 0.979 for Freundlich (0.979). But in all cases, high-regression correlation coefficients (>0.97) are obtained for 4BA adsorption.

5. Conclusion

The present study confirms that the prepared activated carbon from vegetable origin (apricot stones) has an effective adsorbent for 4BA removal from an aqueous solution. Its potential usage as adsorbent is an encouraging factor because an activated carbon prepared from apricot stones is low-cost, easily available, and may be particularly appropriate for the treatment of wastewater containing 4BA. The optimal conditions of activated carbon preparation have been studied via the method of experimental design. A first-degree model has been established. Statistical design of experiments for the manufacture of activated carbon from apricot stones for 4BA adsorption is an efficient and rapid technique to quantify the effect of variable parameters compared with the conventional optimization. Activating agent concentration, carbonization temperature, and duration of carbonization are the most significant parameters affecting 4BA adsorption. The carbonization time does not affect the elimination yield significantly.

The adsorption capacity of activated carbon for 4BA considerably increases with an acid solution. The Langmuir equation shows good agreement with the adsorption isotherm data for the 4BA on activated carbons at 28˚C. Energy adsorption shows that the adsorption of this pollutant is a physical sorption. The n value for the Freundlich isotherm shows that 4BA is favorably adsorbed.

List of symbols

C_o — initial concentration of the solution ($\mathrm{mg\,L^{-1}}$)

C_t — concentration of the solution at time t ($\mathrm{mg\,L^{-1}}$)

Y — 4BA adsorption yield

$X_1, X_2, \ldots, X_k$ — input variables

A_0 — value of fitted response at the center point of design

A_i — linear effects

$A_{ij}; A_{ijk}; A_{ijkl}$ — interaction effects

References

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