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A new linear form analysis of Redlich–Peterson isotherm equation for the adsorptions of dyes

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ABSTRACT

Many papers report that accuracy of the Redlich–Peterson (R–P) isotherm equation (three parameters) is higher than those of Langmuir and Freundlich isotherm equations (two parameters). This paper first explains why the accuracy of the R–P isotherm equation is equal to or higher than that of two parameters isotherm equations. This study suggests a new exponential linear equation with exponent variable α, and solves the most suitable α value with the aid of Microsoft Excel and Sigma Plot 9.0. This exponential method is quick, simple, and accurate for fitting the R–P isotherm equation to experimental data sets of adsorption systems compared with a logarithmical linear form frequently used previously. This investigation prepares four kinds of pistachio shell activated carbons with various doses of NaOH for studying isotherm equilibrium adsorptions of three dyes. The R–P isotherm equation best fit adsorption system data sets studied in this work.

1. Introduction

Redlich and Peterson suggested a three parameter adsorption isotherm equation [1] in 1959, unanimously called the Redlich–Peterson (R–P) isotherm equation (or model). The equation amends inaccuracies of two parameter Langmuir and Freundlich isotherm equations in some adsorption systems. In the literature survey, thirty papers reported the R–P equation was more accurate than the Langmuir and Freundlich equation in describing adsorption systems [2–31]; twelve papers reported that both R–P and Langmuir isotherm equations had equally high accuracy [32–43] because when α value in the R–P equation equals 1, its form is the same as the Langmuir equation. Three papers additionally reported that both R–P and Freundlich isotherm equations had equally high accuracy [44–46] because when the constant value of the R–P equation is large enough, its form is the same as the Freundlich equation (proved later in this text); but the same papers reported Langmuir isotherm equation accuracy was higher than the R–P isotherm equation (not shown). These results are doubtful because when α value is adjusted to equal 1, both equations are the same and both accuracies should be the same. Another papers reported Freundlich isotherm equation accuracy was higher than the R–P isotherm equation (not shown). This cannot be correct either.

The poorer accuracy of the R–P isotherm equation reported in much of the literature is due to poor fitting methods. Therefore, this study suggests a simple, accurate operating method of the Redlich–Peterson isotherm equation. The adsorption of three dyes (BB1, MB, and AB74) used activated carbon prepared from pistachio shells with NaOH activation was carried out in this investigation. The results show the superiority of the Redlich–Peterson isotherm equation for describing adsorption systems by comparing R–P, Langmuir, and Freundlich isotherm equations and a suitable α value range. The characteristic curve of the dimensionless R–P equation form explains the wide application of this equation.

2. Principle

2.1. Linear form of R–P isotherm equation

The R–P isotherm equation is expressed as

$$q_e = \frac{q'_{mon} b_{RP} C_e}{1 + b_{RP} C_e^{\alpha}} \quad (1)$$

where q'_{mon} and b_{RP} are parameters of the R–P isotherm equation. Two linear forms of Eq. (1) can be obtained by transformation as

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and

$$\ln\left(b_{RP}q'_{mon}\frac{C_e}{q_e}-1\right)=\ln b_{RP}+\alpha\ln C_e \tag{2}$$

and

$$\frac{C_e}{q_e}=\frac{1}{b_{RP}q'_{mon}}+\left(\frac{1}{q'_{mon}}\right)C_e^{\alpha} \tag{3}$$

The logarithmic linear form of Eq. (2) was adopted by many researchers [6,8,11,13,19,24,25,27,28,30,32,36–38,43]. For fitting Eq. (2) to the experimental data to obtain a linear plot of $\ln(b_{RP}q'_{mon}(C_e/q_e)-1)$ vs. $\ln C_e$, various constant ($b_{RP}q'_{mon}$) values must be tried before the optimum line is obtained. After obtaining the optimum line, calculate the constants of Eq. (2). The range of $b_{RP}q'_{mon}$ values is wide, from 0.01 to several hundred, so it is not easy to obtain the correct value. Eq. (3) is the exponential linear form obtained by plotting C_e/q_e vs. C_e^{α}. By trial and error, this study adopted an α value for the optimum line. In the specific range, α values are limited and it is easy to obtain the correct value (to be proved later). Another similar form of Eq. (3) has appeared in a few papers [17,47–49].

2.2. Special conditions of R–P isotherm equation

When $\alpha = 1$, it is the same as the Langmuir isotherm equation.

$$\frac{C_e}{q_e}=\frac{1}{K_L q_{mon}}+\left(\frac{1}{q_{mon}}\right)C_e \tag{4}$$

where b_{RP} of Eq. (3) equals K_L of Eq. (4), and q'_{mon} equals q_{mon}. When $1/b_{RP}q'_{mon}=0$, it is the same as the Freundlich isotherm equation, that is

$$q_e=K_F C_e^{1/n} \tag{5}$$

where $1/q'_{mon}$ of Eq. (3) equals K_F of Eq. (5), and α equals $1/n$. Usually, $1/b_{RP}q'_{mon}$ is not equal to 0, and q'_{mon} is constant, so only when b_{RP} is large enough, is the result of the R–P equation close to the Freundlich equation. When $\alpha = 0$, Eq. (1) can be written as

$$q_e=\frac{q'_{mon}b_{RP}C_e}{1+b_{RP}} \tag{6}$$

Eq. (6) is the same as Henry's law equation.

2.3. Characteristic curves of R–P isotherm equation

Eq. (3) can be rewritten as:

$$\frac{C_{ref}}{q_{ref}}=\frac{1}{b_{RP}q'_{mon}}+\left(\frac{1}{q'_{mon}}\right)C_{ref}^{\alpha} \tag{7}$$

where C_{ref} is the highest equilibrium concentration of the adsorption system, and q_{ref} is the equilibrium adsorption amount at C_{ref}. When Eq. (7) is divided by Eq. (3), it yields

$$\frac{q_e}{q_{ref}}=\left(\frac{C_e}{C_{ref}}\right)\frac{(1/b_{RP}C_{ref}^{\alpha})+1}{(1/b_{RP}C_{ref}^{\alpha})+(C_e/C_{ref})^{\alpha}} \tag{8}$$

Eq. (8) is the dimensionless form of the R–P isotherm equation. Fig. 1(a) and (b) depict plots of q_e/q_{ref} vs. C_e/C_{ref} with α as a parameter for $b_{RP}C_{ref}^{\alpha}=2$ and 10, respectively. When $\alpha = 1$, the curve is the same as the curve in the Langmuir isotherm equation. The α value modifies the curve. When α value increases, curvature increases. Usually, the α value reported in literature is less than 1 (proved later in this text). These low α values mean their isotherm equation curves are milder than those of the Langmuir isotherm equation.

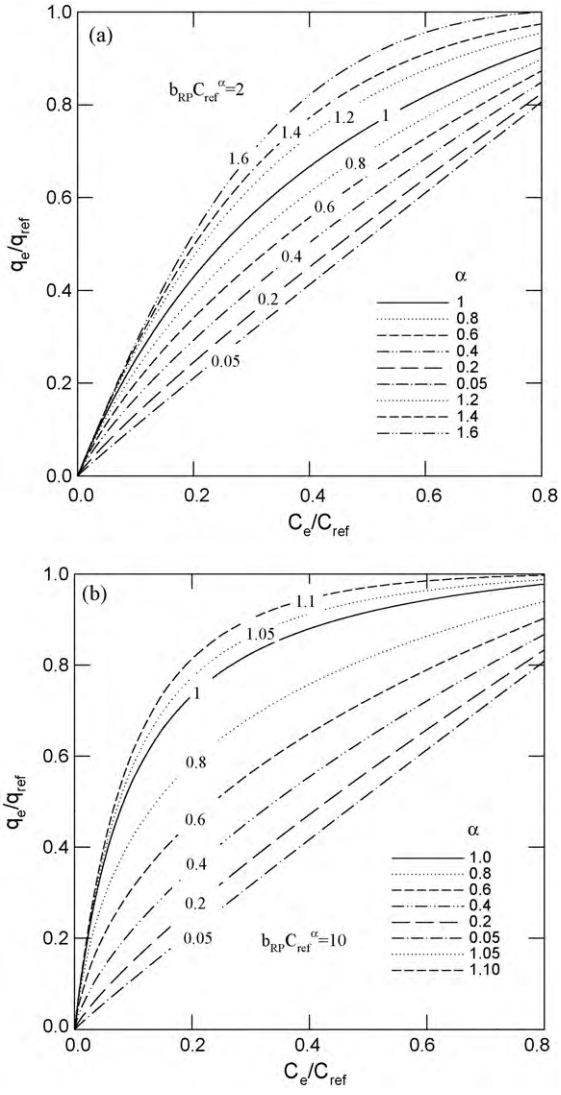

Fig. 1. Adsorption characteristic curves of dimensionless Redlich–Peterson isotherm equation (a) $b_{RP}C_{ref}^{\alpha}=2$ and (b) $b_{RP}C_{ref}^{\alpha}=10$.

3. Materials and method

3.1. Preparation of the activated carbon

Pistachio shells were dried at 110 °C for more than 24 h, then placed them into a high temperature, oxygen-deficient oven by introducing N_2 gas. The oven was kept at 450 °C for 2 h to carbonize the pistachio shells into char. The carbonization yield was 32.0 wt%.

Char was ground and sieved into a particle size ranging from 0.833 to 1.65 mm. The activation step was conducted by adding the char and NaOH in water, uniformly mixed, and dried at 130 °C. The solid was then placed in an oven that was heated and maintained at 780 °C for 1 h in the presence of N_2 gas. The product was neutralized by HCl (600 cm^3) having an equal equivalent to NaOH solution until most of CO_2 bubbles were disappeared. The product was kept at 80 °C for 1 h in water bath, and washed continuously with distilled water till the water was neutral. The samples were classified according to agent/char ratio as PSN2, PSN2.5, PSN3 and PSN3.5. The first two characters, PS, represent the Pistachio shells. The third character, N, represents NaOH activation. The number represents the weight NaOH/char ratio.

The BET surface area of the adsorbent (S_p) was measured from N_2 adsorption isotherms at 77 K with a sorptiometer (Porous Materi-

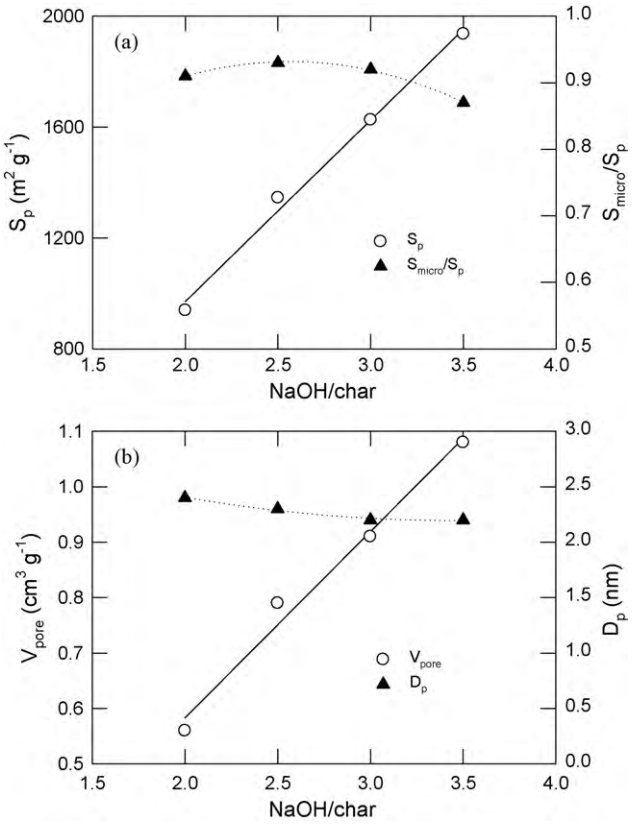

Fig. 2. (a) BET surface area and micropore surface area ratio, (b) total pore volume and mean pore diameter of activated carbon derived from pistachio shell by NaOH activation.

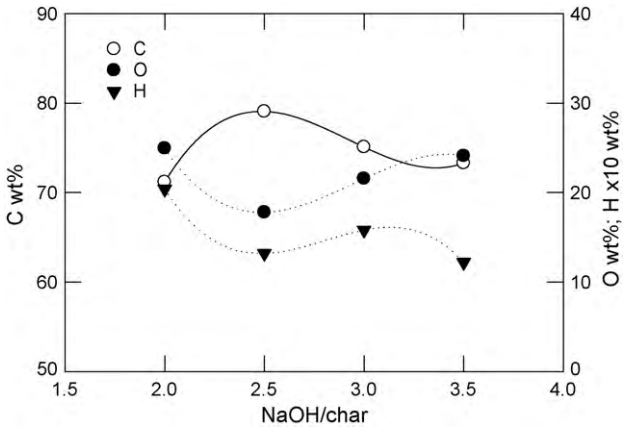

Fig. 3. Chemical compositions of activated carbon derived from pistachio shell by NaOH activation.

als Co., BET-202A), and the total pore volume (V_{pore}) was calculated using the manufacturer s software. The pore size distribution was also determined using the BJH theory [50]. On the other hand, the micropore volume (V_{micro}) and exterior surface area (S_{ext}) were calculated by the *t*-plot method [51,52] and, accordingly, the micropore surface area (S_{micro}) was obtained by subtracting S_{ext} from S_p [53].

3.2. Procedures for adsorption experiments

Analytical reagent grade acid blue 74 (AB74, Mw (molecular weight) = 466.4), basic brown 1 (BB1, Mw = 419.4), and methylene blue (MB, Mw = 284.3) were offered from Merck Co. and were used as received.

In the adsorption equilibrium experiments, an amount of the activated carbons (0.1 g) and 0.1 dm^3 of an aqueous phase were placed in a 0.25-dm^3 flask and stirred for 4 days in a water bath (Haake Model K-F3) at 30 °C. Preliminary tests showed that adsorption was complete after 3 days. The aqueous solution was prepared by dissolving solute to required concentration without pH adjustment. The molecular structures of MB, BB1, and AB74 have been described earlier [54]. The procedures for measuring adsorption isotherms and rates were identical to those described previously [55]. The concentrations of MB, AB74, and BB1 were determined with a Hitachi UV/visible spectrophotometer (U-2001). Each experiment was repeated at least three times under identical conditions. The amount of adsorption at equilibrium, q_e (g kg^{-1}), was calculated by

$$q_e = \left(\frac{C_0 - C_e}{W}\right) V \tag{9}$$

where C_0 and C_e are the initial and equilibrium liquid concentrations (g m^{-3}); V is the volume of the solution (m^3); and W is the weight of dried carbons used (kg).

In order to compare the validity of two isotherm equations, a normalized standard deviation Δq_e (%) is calculated,

$$\Delta q_e\,(\%) = 100\sqrt{\frac{\sum[(q_{e,\exp} - q_{e,cat})/q_{e,\exp}]^2}{N-1}} \tag{10}$$

where N is the number of data.

Table 1
Analysis of adsorptions of dyes on the activated carbons based on Langmuir and Freundlich equations.

Dyes	Carbon	Langmuir eq.			Freundlich eq.				
		q_{mon} (g/kg)	K_L (m^3/g)	r^2	Δq_e (%)	K_F^*	$1/n$	r^2	Δq_e (%)
BB1	PSN2	794	0.015	0.9982	2.98	182	0.217	0.970	2.85
	PSN2.5	1217	0.073	0.9995	6.20	429	0.181	0.926	15.7
	PSN3	1422	0.132	0.9998	5.90	531	0.186	0.864	11.4
	PSN3.5	1806	0.080	0.9981	8.83	413	0.298	0.954	8.02
MB	PSN2	394	0.029	0.9975	6.25	216	0.085	0.880	2.16
	PSN2.5	674	0.116	0.9996	25.1	410	0.081	0.989	1.25
	PSN3	745	0.218	0.9999	7.72	437	0.094	0.902	4.87
	PSN3.5	759	0.271	0.9999	14.3	474	0.099	0.880	6.56
AB74	PSN2	204	0.078	0.9996	2.63	116	0.093	0.995	0.45
	PSN2.5	465	0.052	0.9972	9.59	119	0.250	0.990	2.43
	PSN3	515	0.053	0.9979	2.77	115	0.283	0.953	5.80
	PSN3.5	574	0.068	0.9991	3.19	131	0.294	0.948	6.68

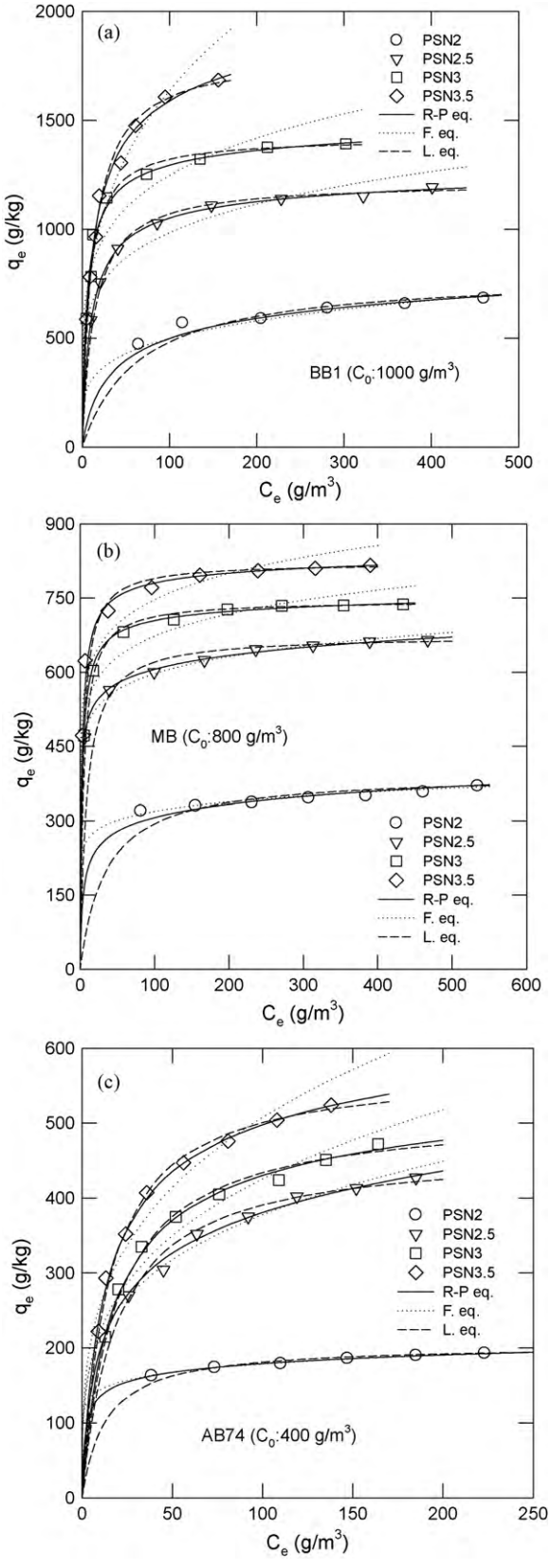

Fig. 4. Adsorption isotherm equilibrium of (a) BB1, (b) MB, and (c) AB74 at 30 °C on the activated carbons (PSN2 (○), PSN2.5 (▽), PSN3 (□), and PSN3.5 (◇), respectively).

4. Results and discussion

4.1. Properties of the activated carbons

This work prepared activated carbons from pistachio shells with NaOH at various NaOH/char ratios. Fig. 2(a) shows the relationships between BET specific surface area (S_p) and micropore surface area ratio (S_{micro}/S_p) and NaOH/char ratios. The data reveals that S_p increases with increased NaOH/char ratio, from 939 to 1936 $m^2\ g^{-1}$. S_{micro}/S_p values are between 0.87 and 0.93, a small variation. Fig. 2(b) shows the relationship between total pore volume (V_{pore}) and mean pore size (D_p) and NaOH/char ratios which reveals that V_{pore} increases with NaOH/char ratio, from 0.56 to 1.08 $cm^3\ g^{-1}$. D_p values are between 2.2 and 2.4 nm.

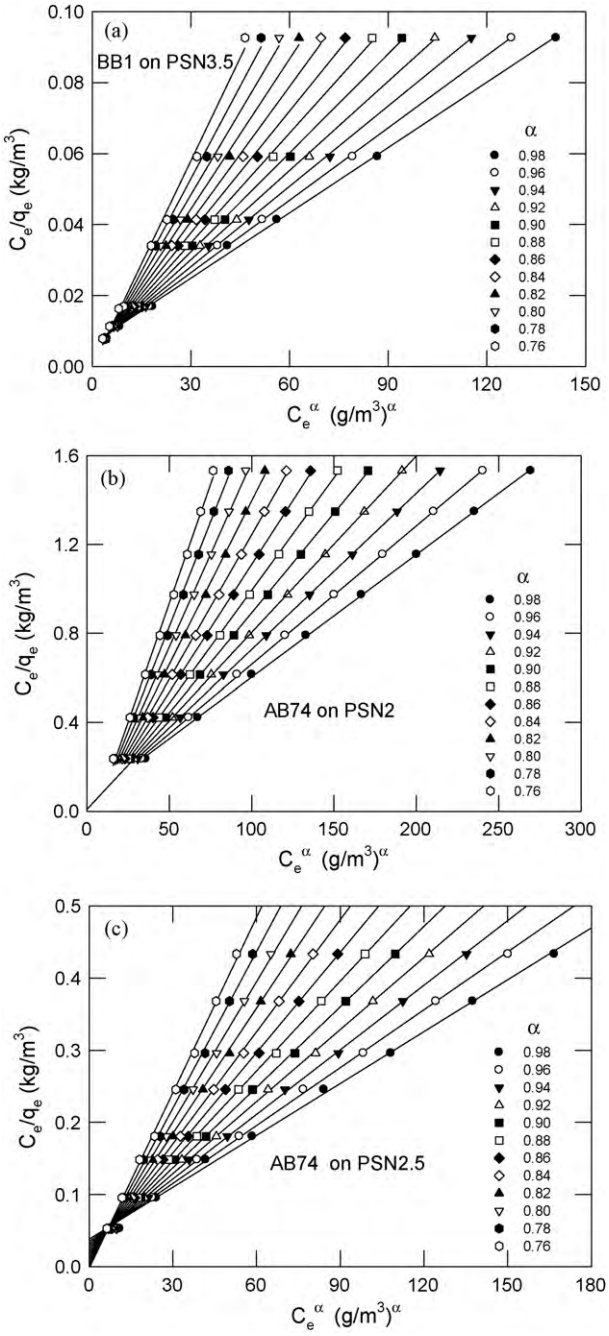

Fig. 5. Adsorption of (a) BB1 on PSN3.5, (b) AB74 on PSN2, (c) AB74 on PSN2.5 fitted with new linear form of Redlich–Peterson isotherm equation.

Table 2
Analysis of adsorptions of dyes on the activated carbons based on Redlich–Peterson equations.

Dyes	Carbon	q'_{mon}	b_{RP} $(m^3/g)^{\alpha}$	α	r^2	Δq_e (%)
BB1	PSN2	351	0.079	0.88	0.9993	1.79
	PSN2.5	953	0.134	0.96	0.9998	0.30
	PSN3	1130	0.244	0.96	0.9999	4.81
	PSN3.5	1194	0.169	0.92	0.9990	4.23
MB	PSN2	200	0.505	0.90	0.9975	1.88
	PSN2.5	463	1.912	0.94	0.9999	0.65
	PSN3	658	0.442	0.98	1.0000	0.86
	PSN3.5	728	0.469	0.98	1.0000	4.80
AB74	PSN2	126	1.031	0.92	1.0000	0.42
	PSN2.5	173	0.541	0.82	0.9991	2.16
	PSN3	373	0.092	0.94	0.9988	2.01
	PSN3.5	380	0.141	0.92	0.9997	2.58

BB1: C_{max} = 500 g m^{-3}, MB: C_{max} = 700 g m^{-3}, AB74: C_{max} = 400 g m^{-3}.

Analyzed elements [56,57] express the chemical properties of activated carbon prepared with chemical activation. Fig. 3 lists weight percents of C, O, and H of the activated carbons studied. Element C is 71–79 wt%; O, 18–25 wt%; H, 1.2–2.0 wt%.

4.2. Adsorption equilibrium of dyes and three isotherm equations

This isotherm equilibrium adsorption study used three dyes (BB1, MB, and AB74) as adsorbates and PSN2, PSN2.5, PSN3, and PSN3.5, the activated carbons of activated at different NaOH/char ratios, as adsorbents. Fig. 4(a)–(c) shows the experiment data results. Data were fitted with Langmuir and Freundlich isotherm equations, and the results summarized in Table 1. Table 1 shows that r^2 values of the Langmuir isotherm equation are larger than 0.9972. However, Δq_e (%) values are widely distributed, from 2.63% to 25.1%. This study excellently fitted the Freundlich isotherm equation to the experimental data of the adsorption systems of MB and AB74 on PSN2 and PSN2.5 with very small deviation values.

To fit the R–P isotherm equation to experimental data set, the linear regression lines were constructed by plotting C_e/q_e vs. C_e^{α} of Eq. (3) with various α values. In this paper, first, the value of C_e^{α} was calculated for each α value, and then the linear regression line associated with this specific α value was plotted.

Fig. 5(a)–(c) shows three families of regression lines for the adsorption systems of BB1 on PSN3.5, AB74 on PSN2, and AB74 on PSN2.5, respectively. The α values of the most suitable linear regression lines are, respectively, 0.92, 0.92, and 0.82. The interceptions of straight lines are all close to 0 for all α values. Table 2 lists the obtained values of q'_{mon}, b_{RP}, α, r^2, and Δq_e (%). All the r^2 values are larger than 0.9988 and all the Δq_e (%) values are less

Table 3
Redlich–Peterson and Langmuir isotherm equations of the adsorption of dyes obtained from the literature.

No.	Dyes	Adsorbent	Redlich–Peterson eq.			Langmuir eq. q_{mon} (g/kg)	Ref.
			α	b_{RP} $(m^3\ g^{-1})^{\alpha}$	q'_{mon}		
1	Malachite G	AC	1.00	0.0198	510	509	[32]
2	RR	Silica	1.00	0.012	16.1	16.2	[60]
3	MB	Lemon peel	1.00	0.0558	33.2	33.2	[61]
4	MB	AC	1.00	0.0442	401	401	[33]
5	RY	FS400	0.99	0.28	700	714	[16]
6	RB19	CC/OPA	0.965	0.04	434	667	[11]
7	BlueG	Beet pulp	0.96	0.020	194	250	[41]
8	MB	Red mud	0.949	0.261	1.93	2.64	[9]
9	BG	RHA	0.939	2.10	19.5	26.2	[34]
10	RR	FS400	0.93	0.02	180	213	[16]
11	Blue-G	Fungus	0.925	0.014	502	601	[62]
12	BB3	Peat	0.914	0.327	324	556	[8]
13	RR	Silica	0.91	0.184	15.9	24.1	[60]
14	RB	FS400	0.91	0.10	168	278	[16]
15	Blue-G	Fungus	0.901	0.16	324	555	[62]
16	Malachite G	Biomass	0.894	0.142	78.9	118	[31]
17	BY21	Peat	0.887	0.121	355	667	[8]
18	AR	AC	0.868	1.20	58.6	101	[24]
19	AB	AC	0.866	17.2	61.9	101	[24]
20	BR22	Peat	0.865	0.996	128	312	[8]
21	MV	BFA	0.861	2.35	17.5	26.2	[19]
22	BR22	Kudzu	0.843	0.293	69.8	192	[62]
23	AY	AC	0.825	32.89	74.3	129	[24]
24	MB	Fibres	0.74	0.77	2.48	5.56	[64]
25	MV	WC	0.677	0.71	38.4	240	[25]
26	AR	WC	0.657	1.93	35.4	246	[25]
27	CR	WC	0.648	0.35	31.9	234	[25]
28	OG	WC	0.602	0.66	25.9	236	[25]
29	Malachite G	ACL	0.618	3.29	7.36	42.2	[65]
30	Malachite G	ACC	0.597	68.6	1.21	8.27	[63]
31	Malachite G	BFA	0.583	89.2	25.5	170	[65]
32	RB	Alga	0.53	0.36	16.2	357	[57]
33	RB	Alga	0.48	0.51	17.6	476	[59]
34	OG	MV	0.458	182	3.10	26.2	[19]
35	RB19	CC/OPA	0.188	0.414	53.9	909	[11]

than those listed in Table 1 of the fitted Langmuir and Freundlich isotherm equations. Tables 1 and 2 show that Δq_e (%) values of MB adsorption on PSN2.5 are, respectively, 25.1%, 1.25%, and 0.65% for Langmuir, Freundlich, and R–P isotherm equations. The Freundlich, and R–P isotherm equations are much more suitable than the Langmuir equation in fitting to the data set of MB adsorption on PSN2.5. The value of b_{RP} in the R–P isotherm equation is high, up to 1.912, for MB adsorption on PSN2.5, much higher than 0.079–0.244 for BB1 adsorption as shown in Table 2. Fig. 4(a)–(c) shows the calculated curves of three isotherm equations and that the curvatures of the curves of the R–P equation are higher than those of the Freundlich equation but lower than those of the Langmuir equation. All α values were less than 1 (see explanation of Fig. 1), from 0.82 to 0.98 as listed in Table 2.

Table 3 lists thirty-five adsorption systems of dyes fitted with the R–P isotherm equation. Among them, four systems have an α value of 1 and the rest have an α value less than 1. Large molecule adsorption, as in dyes, is not easy in accordance with the theory of monolayer adsorption upon which the Langmuir isotherm equation is based. This is because impediments exist between pores and adsorbate so the α value is usually less than 1.

In a report of dye adsorptions on chitosan [14], the Langmuir isotherm equation was better fitted to the adsorptions of AG25, AR18, AR73, AO12. The Freundlich isotherm equation was better fitted to the adsorption of AO10 and the R–P isotherm equation to those of all dyes with the lowest error values. In a report of basic dye adsorption on kudzu [58], the error values of Freundlich and R–P isotherm equations in fitting to the adsorption of BY21 were both the same and the least. The error value of the R–P isotherm equation in fitting to the adsorption of BR22 was the least. In a report of reactive dye adsorption on green alga [59], the errors of Freundlich and R–P isotherm equations in fitting to RBB adsorption were both the same and the least. Consequently, the above reports show that error values of the R–P isotherm equation are either close to those of the Freundlich or the Langmuir equations, or less than those of both equations. These findings agree with the results of this study.

5. Conclusions

This study explains the special conditions of the R–P isotherm equation and proves they are in accordance with Langmuir and Freundlich isotherm equations. Thus the R–P isotherm equation is suitable for wide applications. This investigation deduces the dimensionless form of the R–P isotherm equation and plots curves of the dimensionless form. The current study explains that the α value modifies R–P isotherm equation curves. Microporous activated carbons with BET specific surface area (S_p) from 939 to 1936 $m^2\,g^{-1}$ were prepared from pistachio shells with NaOH activation for the adsorptions of three dyes (AB74, BB1, and MB). The linear regressions calculated are used to identify the most suitable α value. This is a simple and accurate calculation method. In fitting the data sets of the adsorptions of three dyes, the R–P isotherm equation is the best with α values being between 0.82 and 0.98. This investigation employed thirty-five adsorption systems of dyes reported in the literature to explain the existence of a solid impediment between pores and adsorbate, so the α value is usually less than 1 in the adsorption of large molecules.

Acknowledgments

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