

The Kinetics of Sorption of Basic Dyes from Aqueous Solution by Sphagnum Moss Peat

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The kinetics of sorption of three basic dyes, namely, Chrysoidine (BO2), Astrazon Blue (BB3) and Astrazone Blue (BB69) onto sphagnum moss peat have been investigated. The study focuses on the application of three sorption kinetic models for predicting the uptake of basic dyes. The sorption behaviour is found to be second order, based on the assumption of a pseudo-second order mechanism. The rate constant of sorption, the equilibrium capacity and initial sorption rate with the effect of various peat doses and initial dye concentrations have also been predicted.

On a étudié les cinétiques de sorption de trois teintures de base, à savoir Chrysoidine (BO₂), Astrazon Blue (BB3) et Astrazone Blue (BB69) sur une mousse de sphaigne. L'étude porte sur l'application de trois modèles cinétiques de sorption pour la prédiction de la consommation des teintures de base. On a trouvé que le comportement de sorption était du second ordre, en s'appuyant sur l'hypothèse d'un mécanisme de pseudo-second ordre. On a également prédit la constante de vitesse de sorption, la capacité d'équilibre et la vitesse de sorption initiale avec l'effet des diverses doses de tourbe et des concentrations de teintures initiales.

Keywords: peat, dye, kinetics and sorption.

The removal of dye from textile effluents is one of the significant environmental problems. Since water is a valuable material it should not be polluted by the discharge of industrial effluents. Sorption techniques produce high quality treated effluents and sorption processes have been investigated as a method to remove dyes from wastewater. McKay et al. (1981a,b,c; 1982; 1985; 1987) have used low cost sorbents such as peat, wood, silica, chitin, Fuller's earth and bagasse pith for the removal of acid and basic dyes from aqueous solutions.

In recent years, sorption has been accepted as one of the most appropriate processes, for the purification of water and wastewater. The prediction of batch sorption kinetics is necessary for the design of industrial sorption columns. However, sorption kinetics show a large dependence on the physical and/or chemical characteristics of the sorbent material which also influences the sorption mechanism. Other relevant factors include sorbate-sorbent chemical properties and system conditions. The sorption mechanism has usually been described by the pseudo-first order Lagergren equation (Khare et al., 1987; Namasivayam and Yamuna, 1992; Low et al., 1995; Mittal and Gupta, 1996; Banerjee et al., 1997) and multiple first order kinetics have been also represented in some sorption systems such as protein/silica (Sarkar and Chatteraj, 1993), mercury(II)/kaolinite (Singh et al., 1996) and persistent/kaolinite (Atun and Sismanoglu, 1996).

The sorption of dyes on various sorbents has been reported as pseudo-first order by many authors (Namasivayam et al., 1996; Atun and Sismanoglu, 1996; Lee et al., 1996). There is only limited second order data available for sorption kinetics on peat; pseudo-second order mechanisms (Gosset et al., 1986; Ho et al., 1994) have been proposed to explain metal ion sorption processes. Other researchers (Allen et al., 1995) have analysed dye and metal sorption on peat based

on an external mass transfer mechanism. The sorption of basic dyes on peat has been studied (McKay and Allen, 1980) and analysed using a diffusional mass transport model. However, no attempts have been undertaken to assess dye sorption onto peat using pseudo-second order sorption kinetic models.

The purposes of this paper are to propose and test a sorption model to predict the kinetic behaviour for the batch sorption of basic dyes. A pseudo-second order mechanism has been developed and applied to the sorption of three basic dyes and the effect of peat dose and initial dye concentration has been studied.

Materials and methods

SORBENT

The experiments were conducted with sphagnum moss peat obtained from Northern Ireland. The peat as supplied, was dried at a temperature of $85 \pm 5^\circ\text{C}$ for 3 h and stored in sealed containers. The sample was broken-up using a hammer mill and finally sieved into different fractions. The peat surface area was found to be $27 \text{ m}^2\text{g}^{-1}$ by BET analysis using nitrogen. The absolute peat density using hexane was $1250 \text{ kg}\cdot\text{m}^{-3}$ and its equilibrium moisture content was 14.5% by weight. The ash content was 1.8% by weight and an elemental analysis was determined: $C = 48.8\%$; $H = 5.6\%$; $N = 1.3\%$; $S = 0.7\%$.

The apparatus used to determine the contact time data has been described previously (McKay and Allen, 1980).

Adsorption isotherms were obtained by shaking 0.5 g samples of peat with 50 ml dye solution using concentrations from $10 - 2000 \text{ mg}/\text{dm}^3$ for a contact time of 5 d. After agitation the flasks are set aside to stand and then filtered using small quantities of glass wool. The first part of the filtered liquor was discarded to eliminate the effects of any adsorption on the glass wool. The concentration of colouring matter was determined using an Optica double beam

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TABLE I
Experimental Details

Variable	Basic Orange 2	Basic Blue 3	Basic Blue 69
Peat Mass (g)	0.294, 1.18, 1.77, 2.35. (Co = 105 mg/dm ³)	0.294, 0.397, 0.536, 0.735. (Co = 125 mg/dm ³)	0.556, 2.22, 3.33, 4.44. (Co = 200 mg/dm ³)
Dye Concentration (mg/dm ³)	26.3, 52.5, 78.8, 105. (mass = 0.5 g)	25, 50, 75, 100. (mass = 0.3 g)	20, 50, 100, 200. (mass = 1.0 g)
Agitation Speed (rpm)	400	400	400
Temperature (°C)	20 ± 2	20 ± 2	20 ± 2
Solution Volume (dm ³)	1.7	1.7	1.7
Particle Size (µm)	710–1000	710–1000	500–710

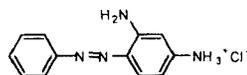
spectrophotometer and a 1 cm light path cell was employed for all determinations. The wavelength was selected so as to obtain maximum adsorbance for each dye. All tests were made at room temperature (20 ± 2°C) to eliminate any temperature effects.

Blank experiments were performed (i) without peat to check that no dye adsorbed onto the container jars, and (ii) with peat and water only to ensure that no leaching occurred which would interfere with the measurement of dye concentrations on the spectrophotometer.

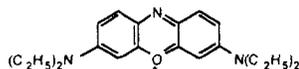
SORBATES

The sorbates and their structures used in the experiments are listed below. The dyestuffs were used as the commercial salts.

Chrysoidine GS (C.I. Basic Orange 2) was supplied by Ciba-Geigy.



Astrazon Blue BG (C.I. Basic Blue 3) was supplied by Ciba-Geigy.



Astrazone Blue FRR (C.I. Basic Blue 69) was supplied by Ciba-Geigy.

No structure is available for this dye. It belongs to the methine class, of which the chromophore is a conjugate chain of carbon atoms terminated by an equivalent unsaturated group.

SYSTEM VARIABLES

Batch agitation experiments were carried out to study the effect of peat mass and initial metal ion concentration. The experimental conditions are summarised in Table 1.

Results and discussion

EFFECT OF PEAT DOSE

The sorption kinetics of basic dyes, namely, Chrysoidine (BO2), Astrazon Blue (BB3) and Astrazone Blue (BB69) on peat are shown in Figures 1 to 3 respectively. The figures

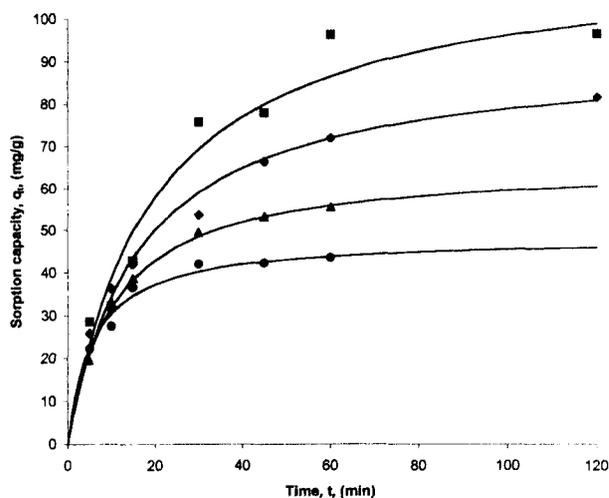


Figure 1 — Effect of peat dose on the sorption of BO2 onto peat.

show the amount of dye sorbed in mg/g peat against contact time using various masses of peat. These plots show that for all peat doses, the amount of dye sorbed increases rapidly with time in the beginning and very slowly towards the end of the reaction. Furthermore, a large fraction on the total amount of dye was removed within a short time. In the first step the sorbate molecules are sorbing onto the surface where there are no other such molecules and consequently the sorbate-sorbate interactions are negligible (Varshney et al., 1996). This is supported by the fact that in this initial region the equation for the first order reaction is applicable. Equation (1) represents the first order reaction proposed by Lagergren (1898):

$$\frac{dq_t}{dt} = k_1(q_1 - q_t) \dots \dots \dots (1)$$

integrating this for the boundary conditions $t = 0$ to $t = t$ and $q_t = 0$ to $q_t = q_t$. Equation (1) may be arranged for linearized data plotting as shown by Equation (2):

$$\log(q_1 - q_t) = \log(q_1) - \frac{k_1}{2.303} t \dots \dots \dots (2)$$

The pseudo-first order plot of $\log(q_1 - q_t)$ versus time for various peat doses is shown in Figure 4 for the initial 30 min for the sorption of BO2 and BB69. It can be seen in Figure 4

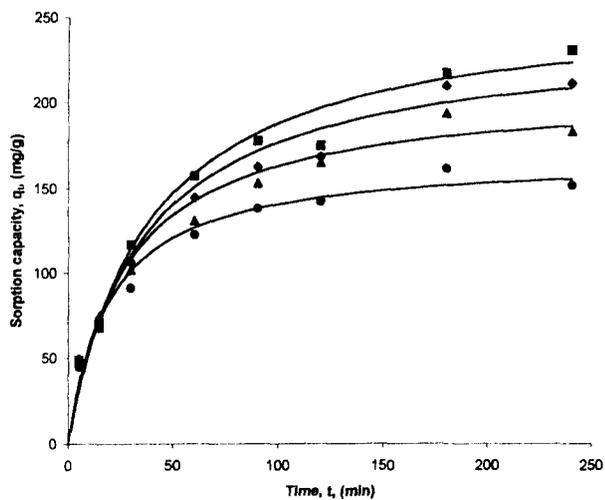


Figure 2 — Effect of peat dose on the sorption of BB3 onto peat.

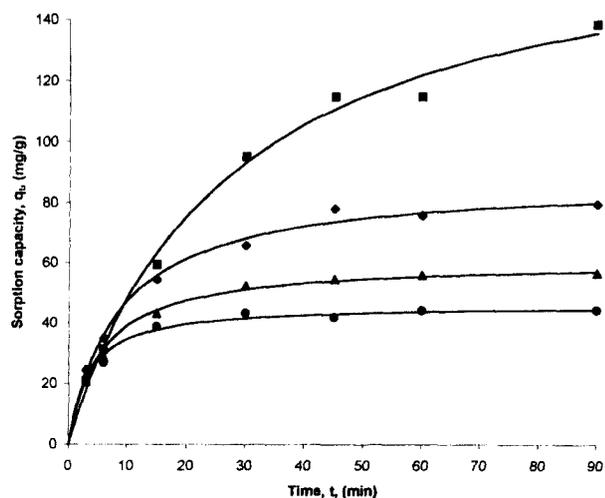


Figure 3 — Effect of peat dose on the sorption of BB69 onto peat.

the experimental points and theoretical lines deviate rapidly and extensively after 30 min. Table 1 shows that the first order correlation coefficients, r_1^2 , for these studies are greater than 0.959 for the first sorption step (30 min). Consequently these sorption data may follow a first order reaction mechanism in the early stages of the sorption process, but it is difficult to assess whether the mechanism suddenly changes from first order or the transition is gradual.

It is often incorrect to apply simple kinetic models such as first- or second-order rate equations to a sorption with solid surfaces which are rarely homogeneous and because effects of transport phenomena and chemical reactions are often experimentally inseparable (Sparks, 1989). Multiple first order kinetics have been represented in some sorption systems such as protein/silica (Sarkar and Chatteraj, 1993), mercury(II)/kaolinite (Singh et al., 1996) and persistent/kaolinite (Atun and Sismanoglu, 1996). In the multiple first order kinetics sorption process, the first stage is attributed to the initial binding or anchorage of protein molecules with the active spots of solid surface by removal and reorganization of surface-bound water leaving. The second stage is attributed to the denaturation and reorganization of the bound biopolymer at the interface, leading to the formation of spread films (Searcher and Chatterji, 1993). Atuna and Sismanoglu

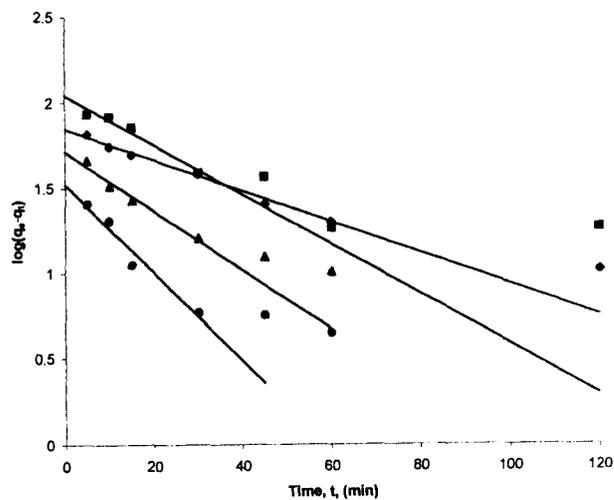


Figure 4 — First-order kinetic plot for the sorption of BO2 onto peat at various peat doses.

(1996) reported that in the case of two kinetics steps, the first step of sorption was more rapid than the second one and the sorption rate is controlled by either a film diffusion or a intraparticle diffusion. It might be because that kaolinite is a clay and its macroscopic structure is probably closer to a microporous 'honeycomb' than a macroporous 'capillary' system (Lockhart, 1980).

The kinetics of sorption processes are concerned with force fields between sites and dyes and form an important area of surface chemistry. The experimental data in this paper have been analysed using a pseudo-second order mechanism according to Equation (3), which on integration becomes Equation (4).

$$\frac{dq}{dt} = k(q_e - q_t)^2 \dots\dots\dots (3)$$

$$\frac{1}{(q_e - q_t)} = \frac{1}{q_e} + kt \dots\dots\dots (4)$$

where q_e is the amount of dye sorbed at equilibrium, (mg/g) and k is the equilibrium rate constant for pseudo-second order sorption, (g/mg·min). A pseudo-second order mechanism could occur due to the ability of dyes to form micelles in aqueous solution. If the dyes exist as dimeric micelles, then a possible pseudo-second order mechanism is shown:



Equation (4) can be arranged to obtain a linear form as shown:

$$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{1}{q_e}t \dots\dots\dots (5)$$

and as $t \rightarrow 0$, the initial sorption rate, h (mg/gmin) is:

$$h = kq_e^2 \dots\dots\dots (6)$$

It was found that the equilibrium sorption of dye is a function of peat dose. The rate constant, k , the equilibrium sorption,

TABLE 2
The Effect of Peat Dose on Basic Dye Sorption Data

	m_s	r^2	q_e	k	h	r_i^2	q_i	k_i	r_i^2	k_i
BO2	0.294	0.981	114	4.48×10^{-4}	5.86	0.963	110	3.35×10^{-2}	0.933	15.1
	1.18	0.996	91.6	6.59×10^{-4}	5.53	0.977	70.0	2.10×10^{-2}	0.987	8.37
	1.77	0.999	65.5	14.5×10^{-3}	6.21	0.975	51.7	4.01×10^{-2}	0.958	8.89
	2.35	0.997	47.8	3.69×10^{-3}	8.43	0.959	33.3	5.98×10^{-2}	0.934	6.25
BB3	0.294	0.984	258	1.04×10^{-4}	6.92	0.985	224	1.36×10^{-2}	0.979	18.8
	0.397	0.987	238	1.22×10^{-4}	6.91	0.995	199	1.28×10^{-2}	0.994	17.6
	0.536	0.991	207	1.80×10^{-4}	7.71	0.987	163	1.31×10^{-2}	0.996	15.0
	0.735	0.994	168	3.05×10^{-4}	8.62	0.995	130	1.76×10^{-2}	0.997	13.9
BB69	0.556	0.989	175	2.19×10^{-4}	6.67	1.000	166	2.45×10^{-2}	0.996	20.1
	2.22	0.998	87.2	1.37×10^{-3}	10.4	0.995	43.7	5.74×10^{-2}	0.974	11.1
	3.33	0.999	60.4	3.04×10^{-3}	11.1	0.971	66.1	3.92×10^{-2}	0.984	8.07
	4.44	0.999	46.1	6.54×10^{-3}	13.9	0.982	29.4	8.11×10^{-2}	0.947	5.95

m_s : g/dm³, q_e , q_i : mg/g, k : g/mg-min, h : mg/g-min, k_i : 1/min, k_i : mg/g-min^{0.5}.

TABLE 3
The Effect of Initial Concentration on Basic Dye Sorption Data

	C_0	r^2	q_e	k	h	r_i^2	q_i	k_i	r_i^2	k_i
BO2	26.3	0.992	73.8	4.10×10^{-4}	2.23	0.999	67.2	1.69×10^{-2}	0.991	6.52
	52.5	0.992	100	4.17×10^{-4}	4.19	1.000	91.5	2.54×10^{-2}	0.999	1.17
	78.8	0.994	128	2.16×10^{-4}	3.54	0.992	120	1.86×10^{-2}	0.977	1.22
	105	0.991	126	1.85×10^{-4}	2.93	0.987	118	1.38×10^{-2}	0.998	1.01
BB3	25.0	0.964	139	8.80×10^{-5}	1.70	0.904	140	9.61×10^{-3}	0.897	10.6
	50.0	0.984	232	5.06×10^{-5}	2.73	0.931	238	1.02×10^{-2}	0.927	19.0
	75.0	0.975	312	3.38×10^{-5}	3.29	0.955	307	8.17×10^{-3}	0.947	21.4
	100	0.978	375	2.51×10^{-5}	3.52	0.950	368	7.45×10^{-3}	0.940	24.0
BB69	20.0	0.998	32.6	2.18×10^{-3}	2.31	1.000	25.0	2.80×10^{-2}	1.000	3.39
	50.0	0.989	81.6	4.87×10^{-4}	3.24	0.978	65.4	1.54×10^{-2}	0.993	6.17
	100	0.995	145	2.38×10^{-4}	4.99	0.999	124	1.51×10^{-2}	0.999	11.2
	200	0.974	254	7.68×10^{-5}	4.94	0.989	235	1.34×10^{-2}	0.965	18.9

C_0 : mg/dm³, q , q_2 : mg/g, k : g/mg-min, h : mg/g-min, k_i : 1/min, k_i : mg/g-min^{0.5}.

q_e , and the initial sorption rate, h of sorption at various peat doses were calculated from the intercept and slope of the straight line plots of t/q_t versus t in Equation (5). Table 2 shows the rate constants k_1 and k for the pseudo-first order and pseudo-second order mechanisms proposed and obtained from the plots of Equation (3) and (5) respectively. The theoretical plots of qt versus time using the pseudo-second order model are shown by the solid lines in Figures 1, 2 and 3. A third model is also used as a basis for comparison and this assumes intraparticle diffusion. An intraparticle diffusion parameter, k_i , has been defined by Weber and Morris (1963) and is shown in Equation (7):

$$k_i = \frac{q_i}{t^{0.5}} \dots \dots \dots (7)$$

Table 2 shows that the equilibrium sorption of dye increased with a decrease in the peat dose and the initial sorption rate and rate constant increased with an increase in the peat dose. The intraparticle diffusion correlation coefficient, r_i^2 , is based on the results for the first hour only, as Figure 5 shows the experimental data and model line deviate very considerable after this initial period. The linear regression correlation coefficients for Equation (3), (5) and (7) are shown as r_1^2 , r^2 and r_i^2 respectively and over the sorption time range considered (first order, 30 min; second order, whole sorption period; intraparticle diffusion, 60 min) the experimental data are best represented overall by the pseudo-second order kinetic model.

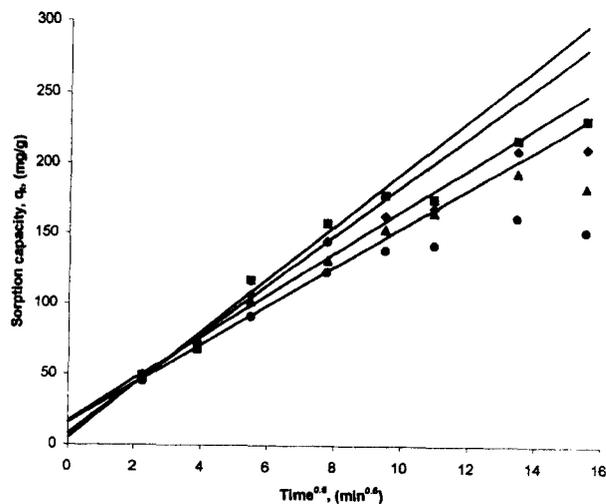


Figure 5 — Intraparticle diffusion kinetic plot for the sorption of BB3 onto peat at various peat doses.

EFFECT OF INITIAL CONCENTRATION

The effect of initial dye concentration on the rate of sorption for three systems was studied. The intraparticle diffusion coefficients, k_i were determined and Figure 5 shows the amount of BB3 per unit weight of peat against square root of time. This model generates best fit straight lines only in the initial 30 min for BO2 and BB69 and 60 min for BB3. This suggests that for dye sorption onto peat, the mechanism

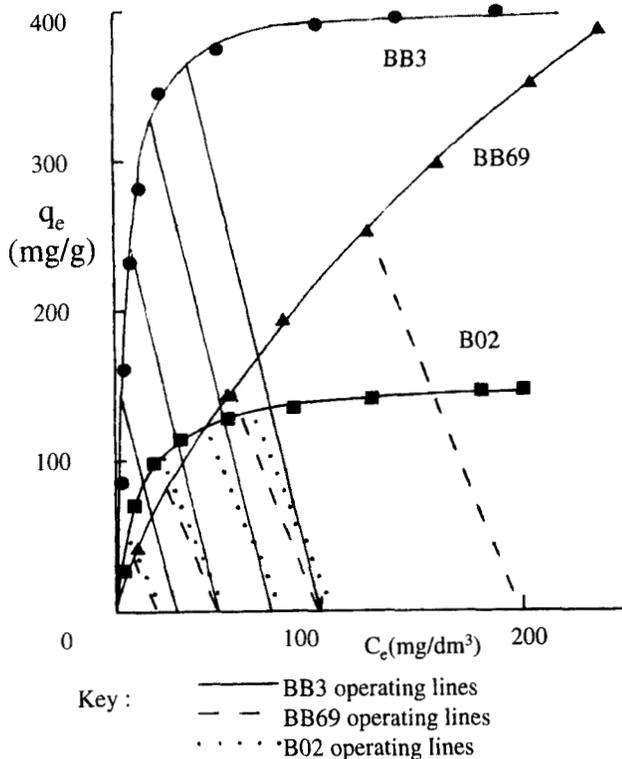


Figure 6 — Equilibrium isotherms showing contact time operating lines.

could be represented by intraparticle diffusion for the first step of sorption. Table 3 shows the intraparticle diffusion correlation coefficients, r_i^2 , and the intraparticle diffusion constants, k_i . Figure 5 shows how the correlation between experimental results and predicted data deviate after this initial time period. The mechanism may change due to the variation of internal pore sizes and a different approach is required to model the whole sorption period. The first order coefficients were also determined within the same time period in the beginning of the reaction and the first order correlation coefficients, r_1^2 , sorption capacities, q_1 and rate constants, k_p , are shown in Table 3. In the first 30 min of the sorption of basic dyes on peat, the mechanism follows the pseudo-first order sorption rate expression of Lagergren with a high correlation coefficient, r_1^2 . However, the fits between experimental results and predicted data become poor after this initial time period.

The rate constant, k , the equilibrium sorption, q_e , and the initial sorption rate, h , at various initial concentrations were calculated from the intercept and slope of the straight line plots of t/q_t versus t in Equation (5) using the pseudo-second order model. Table 3 shows that the equilibrium sorption, q_e , of dye increased with an increase in the initial dye concentration and the pseudo-second order rate constant, k , also increased as the initial dye concentration was decreased for the three basic dyes studied. The correlation coefficients r_2^2 , r_1^2 and r_i^2 are shown in Table 3. It was found that the equilibrium sorption of dye is a function of initial concentration. For the sorption of basic dyes and peat systems, chemical reaction seems significant in the rate controlling step and also the pseudo-second order chemical reaction kinetics provide the best correlation of the experimental data over the first four hours of the sorption process.

The isotherms for the three basic dyes are shown in Figure 6. The operating lines, slope $-V/m$ for each of the

TABLE 4
Comparison of Measured Isotherm q_e^* Values with Predicted q_1 and q_e Values

Dye	C_0	q_e	q_1	q_e^*
BO2	26.3	73.8	67.2	75
	52.5	100	91.5	105
	78.8	128	120	125
	105	126	118	130
BB3	25.0	139	140	135
	50.0	232	238	235
	75.0	312	307	330
	100	375	368	375
BB69	20.0	32.6	25.0	35
	50.0	81.6	65.4	85
	100	145	124	145
	200	254	235	255

twelve initial dye concentration experiments have been constructed on the three equilibrium curves. These isotherm solid phase equilibrium concentrations, q_e^* , for each experiment can be obtained directly from the operating lines starting from a point, C_0 , where $q_0 = 0$ for fresh peat. The q_e^* values are compared with the sorption kinetic models values in Table 4, for the pseudo-second order scheme, q . The agreement between q_e^* and q_e is very close providing further indirect support for a pseudo-second order model.

No studies were carried out relating to the recovery of dyes from peat. However, peat is used extensively as a cheap fuel and therefore by dewatering the spent peat plus dye, it can be burned and heat generated. The calorific value of the dye/peat was determined in this study and found to be 1250 kJ/kg on a dry weight basis. It may also be possible to recover the dyes by solvent extraction, however, the leaching out of other components from the peat is likely.

Conclusions

The study focuses on the application of three sorption kinetic models for predicting the uptake of basic dyes. A pseudo-second order mechanism has been considered which enables the rate constant of sorption, the equilibrium capacity and initial sorption rate to be determined as a function of various peat doses and initial dye concentrations. For all of the systems studied, a pseudo-first order and an intraparticle diffusion model fit the experimental data well for a short initial period. However, over a long reaction period the pseudo-second order model provides the best correlation between experimental results and predicted data over the whole sorption period.

Nomenclature

- C_e = liquid phase equilibrium dye concentration, (mg/dm³)
- C_0 = initial liquid phase dye concentration, (mg/dm³)
- h = initial sorption rate for pseudo-second order model, (mg/g·min)
- k = pseudo-second order rate constant, (g/mg·min)
- k_1 = pseudo-first order rate constant, (min⁻¹)
- k_i = intraparticle diffusion rate parameter, (mg/g·min^{0.5})
- q_e^* = experimental solid phase equilibrium dye concentration, (mg/g)
- q_1 = theoretical pseudo-first order model predicted equilibrium solid phase dye concentration, (mg/g)
- q_e = theoretical pseudo-second order model predicted equilibrium solid phase dye concentration, (mg/g)
- q_t = solid phase dye concentration at time, t , (mg/g)

- r^2 = linear regression correlation coefficient for pseudo-second order model
 r_i^2 = linear regression correlation coefficient for intraparticle diffusion model
 r_f^2 = linear regression correlation coefficient for pseudo-first order model
 t = time, (min)

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Erratum

We regret that the following papers printed in our June issue (Volume 76, Number 3) were not identified as having been presented orally during the 2nd International Symposium on Mixing in Industrial Processes held in Cancun, Mexico in November 1997:

Yoshihito Kato, Setsuro Hiraoka, Yutaka Tada and Toshio Nomura, "Solid-Liquid Mass Transfer in a Shaking Vessel for a Bioreactor with "Current Pole"", pp. 441-445.

M. Carmen Sánchez, Manuel Berjano, Antonio Guerrero, Edmundo Brito and Crispulo Gallegos, "Evolution of the Microstructure and Rheology of O/W Emulsions During the Emulsification Process", pp. 479-485.

Eduardo Vivaldo-Lima, Philip E. Wood, Archie E. Hamielec and Alexander Penlidis, "Calculation of the Particle Size Distribution in Suspension Polymerization using a Compartment-Mixing Model", pp. 495-505.

Lionel Choplin, Sandrine Torandell and Jacques Servoin, "In Situ Rheological Monitoring in Semi-batch Emulsification Process for Cosmetic Lotion Production", pp. 506-510.

Damien Rauline, Philippe A. Tanguy, Jean-Marc Le Blévec and Jacques Bousquet, "Numerical Investigation of the Performance of Several Static Mixers", pp. 527-535.

Yang Zhao and Robert S. Brodkey, "Averaged and Time-Resolved, Full-Field (Three-Dimensional), Measurements of Unsteady Opposed Jets", pp. 536-545.

Joseph P. Mmbaga and Chad P.J. Bennington, "The Use of Mixing-Sensitive Chemical Reactions to Study Mixing in Dispersed Fibre Systems: Adsorption of Reactants and Product Dyes on the Dispersed Phase", pp. 670-679.