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A kinetic study of metal complex dye sorption onto pine sawdust

Mahmut Özacar[a,*], İ. Ayhan Şengil[b]

[a] *Department of Chemistry, Science and Arts Faculty, Sakarya University, 54100 Sakarya, Turkey*
[b] *Department of Environmental Engineering, Enginering Faculty, Sakarya University, 54040 Sakarya, Turkey*

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Abstract

The removal of metal complex dyes, Metal Complex Blue (MCB) and Metal Complex Yellow (MCY), from aqueous solutions by pine sawdust has been studied in single component sorption systems. The aim of this study was to understand the mechanisms that govern metal complex dyes removal and find a appropriate model for the kinetics of removal in a batch reactor. In order to investigate the mechanism of sorption and potential rate controlling steps, pseuso first- and second-order equations, intraparticle diffusion equation and the Elovich equation have been used to test experimental data. Kinetic analysis of the four models has been carried out for system variables—particle size and mass of sawdust, pH and initial dye concentration. The rate constants for the four models have been determined and the correlation coefficients have been calculated in order to assess which model provides the best fit predicted data with experimental results. Both pseudo second-order equation and the Elovich equation provide the best fit to experimental data.

Keywords: Pine sawdust; Metal complex dyes; Sorption kinetics; Pseudo second-order; Elovich equation; Intraparticle diffusion

1. Introduction

Among various industries, textile industry ranks first in usage of dyes for coloration of products. The effluents of the textile industry are highly coloured and the disposal of these into natural water resources causes damage to the environment as they may significantly affect creatures in water because of reduced light penetration and may also be toxic to some aquatic life because of the presence of metals, chloride and others components in them. Thus, environmental research has paid special attention to dye compounds because of the extensive environmental contamination arising from dyeing operations. Textile effluents are usually treated by physical or chemical processes. Among several chemical and physical methods, a sorption process is often one of the effective methods to removes dyes from wastewater. Although activated carbon is the most widely used adsorbent for the removal of colour and treatment of textile effluents, carbon adsorption remains an expensive process, and this fact has prompted growing research interest into the production of low-cost alternatives to activated carbons in recent years. Consequently numerous low-cost alternatives have been proposed including peat [1,2], various pith [3,4], chitosan [5,6], clay [7,8], alunite [9,10], perlite [11], sawdust [12–14], and other [15,16].

The metal complex dye group one of the most important groups of dyes used in the textile dyeing industries. However, only a limited number of studies on the removal of metal complex dyes have been found in the literature [17,18].

The kinetic describes the solute uptake rate which in turn controls the resistance time of sorbate uptake at the solid-solution interface. It is important to be able to predict the rate at which pollutant is removed from aqueous solution in order to design appropriate sorption treatment systems. The study of sorption kinetics in wastewater treatment is also important as it provides deep understanding of the reaction pathways and the mechanism of sorption reactions. A knowledge of the rate law describing the sorption system is required in order to develop sorption kinetics [19].

The present paper uses pine sawdust, a waste/by product from the timber industry that is either used as cooking fuel or packing material. Four sorption kinetic models were tested in this study for the sorption of Lanasan brilliant blue CFB-A (Metal Complex Blue, MCB) and Lanasan Yellow CFB (Metal Complex Yellow, MCY) onto pine sawdust. To investigate the mechanisms of MCB and MCY sorption, characteristic constants of sorption were determined using

* Corresponding author. Fax: +90-264-3460371.
E-mail address: mozacar@hotmail.com (M. Özacar).

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a pseudo first- and second-order equation, intraparticle diffusion equation and the Elovich equation, respectively, and compared.

2. Kinetic models applied to the sorption of MCB and MCY onto pine sawdust

In order to examine the controlling mechanism of sorption process such as chemical reaction, diffusion control and mass transfer, several kinetic models are used to test experimental data. From a system design viewpoint, a lumped analysis of sorption rates is thus sufficient for practical operation [10,20,21].

2.1. Pseudo first-order equation

The pseudo first-order equation is generally expressed as follows [10,15,16,19,20]:

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

After integration and applying the boundary conditions, for $q_t = 0$ at $t = 0$ and $q_t = q_t$ at $t = t$, the integrated form of Eq. (1) becomes:

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

where q_1 and q_t are the amounts of dye sorbed at equilibrium and at time t (mg/g), respectively, and k_1 is the rate constant of pseudo first-order sorption (1/min).

2.2. Pseudo second-order equation

If the rate of sorption is a second-order mechanism, the pseudo second-order chemisorption kinetic rate equation is expressed as [10,15,16,19,20]:

$$\frac{dq_t}{dt} = k_2(q_2 - q_t)^2 \quad (3)$$

Integrating this equation for the boundary conditions, gives:

$$\frac{1}{q_2 - q_t} = \frac{1}{q_2} + k_2 t \quad (4)$$

which is the integrated law for a second-order reaction, where q_2 is the amount of dyes sorbed at equilibrium (mg/g), k_2 is the equilibrium rate constant of pseudo second-order sorption (g/mg min). Eq. (4) can be rearranged to obtain a linear form:

$$\frac{t}{q_t} = \frac{1}{k_2 q_2^2} + \frac{1}{q_2}t \quad (5)$$

and

$$h = k_2 q_2^2 \quad (6)$$

where h is the initial sorption rate (mg/g min).

2.3. Intraparticle diffusion equation

The fractional approach to equilibrium changes according to a function of $(Dt/r^2)^{1/2}$, where r is the particle radius and D the diffusivity of solute within the particle. The initial rate of the intraparticle diffusion is the following [4,20–22]:

$$q_t = f(t^{1/2}) \quad (7)$$

The rate parameter (k_{int}) for intraparticle diffusion can be defined as:

$$q_t = k_{int} t^{1/2} \quad (8)$$

where k_{int} is the intraparticle diffusion rate constant (mg/g min$^{1/2}$).

2.4. The Elovich equation

The Elovic equation is given as follows [23,24]:

$$\frac{dq_t}{dt} = \alpha e^{-\beta q_t} \quad (9)$$

The integration of the rate equation with the same boundary conditions as the pseudo first- and second-order equations becomes the Elovich equation.

$$q_t = \frac{1}{\beta}\ln(\alpha\beta) + \frac{1}{\beta}\ln t \quad (10)$$

where α is the initial sorption rate (mg/g min), and the parameter β is related to the extent of surface coverage and activation energy for chemisorption (g/mg).

3. Materials and methods

The pine sawdust was collected from a local saw-mill. Sawdust was washed to remove adhering dirt, rinsed thoroughly with distilled water and finally dried at 353 K for 24 h. After drying, the sawdust was sieved through 90–150, 150–250, 250–355, 355–500 and 500–710 μm size fractions using ASTM standard sieves. It was used directly for sorption experiments without any further treatment.

Two metal complex dyes used as sorbates were obtained from Clariant Turkey A.Ş. The dyes used were Lanasan brillant blue CFB-A (MCB, wavelength at which maximum absorbance occurs (λ_{max} = 584 nm)), Lanasan Yellow CFB (MCY, λ_{max} = 428 nm). These dyes were commercial grade and was used without further purification. All other chemicals used in the experiments were obtained from Merck Chemical Co.

Dye solutions were prepared by dissolving different dyes in distilled de-ionized water at desired concentrations. Sorption experiments were carried out by agitating 1 g of the pine sawdust with 1 l of dye solutions of desired concentration and pH, and at 298 ± 2 K temperature in a

magnetic stirrer operating 500 rpm. The pH of the dye solutions was adjusted with HCl or NaOH solution by using a pH meter. Samples (5 ml) were withdrawn at suitable time intervals, centrifuged for 15 min at 5000 rpm and then analyzed with an spectrophotometer (Shimadzu UV-150-02) for all the studies. All dye concentrations were measured at the wavelength corresponding to maximum absorbance, λ_{max}. The amount of dye sorbed onto pine sawdust, q_t (mg/g), was calculated by a mass-balance relationship (Eq. (11)):

$$q_t = (C_0 - C_t)\frac{V}{W} \tag{11}$$

where C_0 and C_t are the initial and time t liquid-phase concentrations of the dye (mg/l), respectively, V the volume of the solution (l) and W the weight of the dry pine sawdust used (g).

4. Results and discussion

4.1. Effect of particle size

The effect of pine sawdust particle size was studied on the two metal complex dyes. Fig. 1(a) shows a series of contact time curves of MCB with particle size from 90–150 to 500–710 μm. The results also shown in Fig. 2(a) as a plot of $\log(q_1 - q_t)$ versus t for sorption of MCB for the pseudo first-order model onto pine sawdust. Kinetic parameters for four kinetic model and correlation coefficients were calculated and listed in Table 1. The values of r^2 of pseudo second-order kinetic model for two dyes are extremely high (>0.998) and followed by those of the Elovich equation, pseudo first-order equation and intraparticle diffusion equation, respectively. Moreover the equilibrium sorption capacities for pseudo second-order are slightly more reasonable than those of the pseudo first-order when comparing

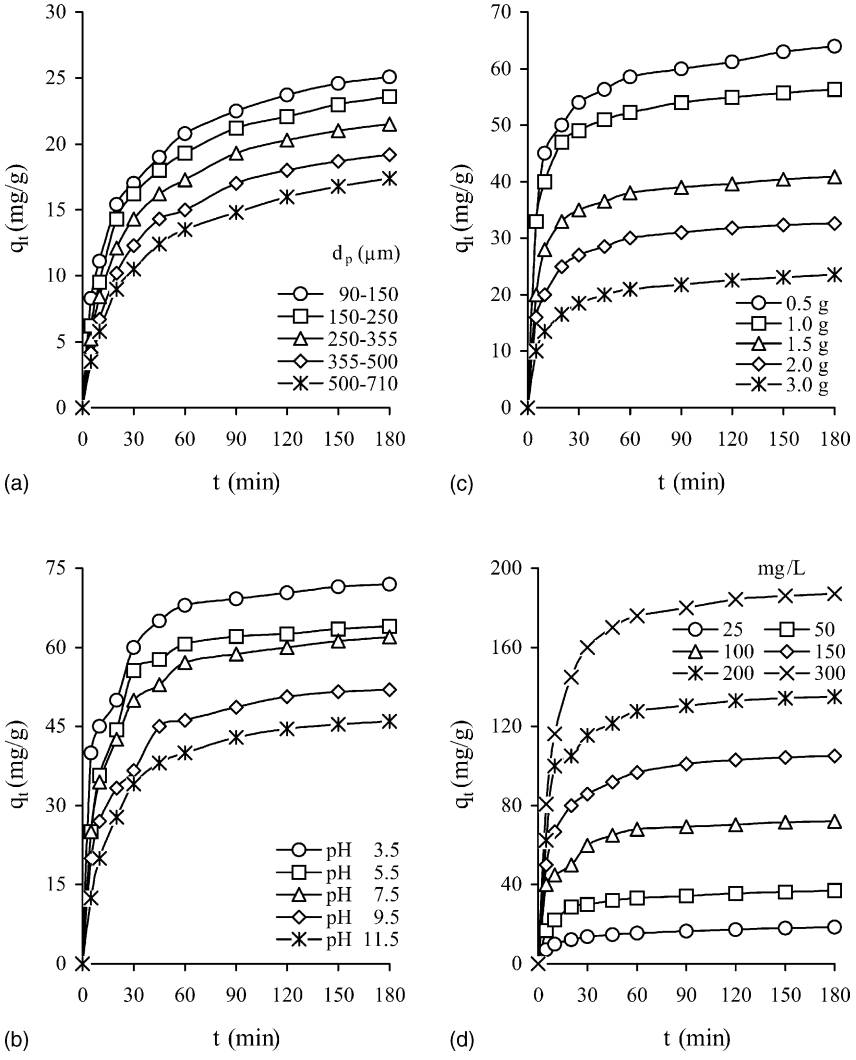

Fig. 1. Effects of sawdust particle size for MCB (a); pH for MCY (b); sawdust dose for MCB (c); and initial dye concentration for MCY (d) on sorption of metal complex dyes onto pine sawdust. Conditions: 100 mg/l dye concentration (a)–(c); 1 g/l dose (a), (b) and (d); 298 K temperature (a)–(d); 90–150 μm particle size (b)–(d); pH 7.5 (a); pH 3.5 (c) and (d).

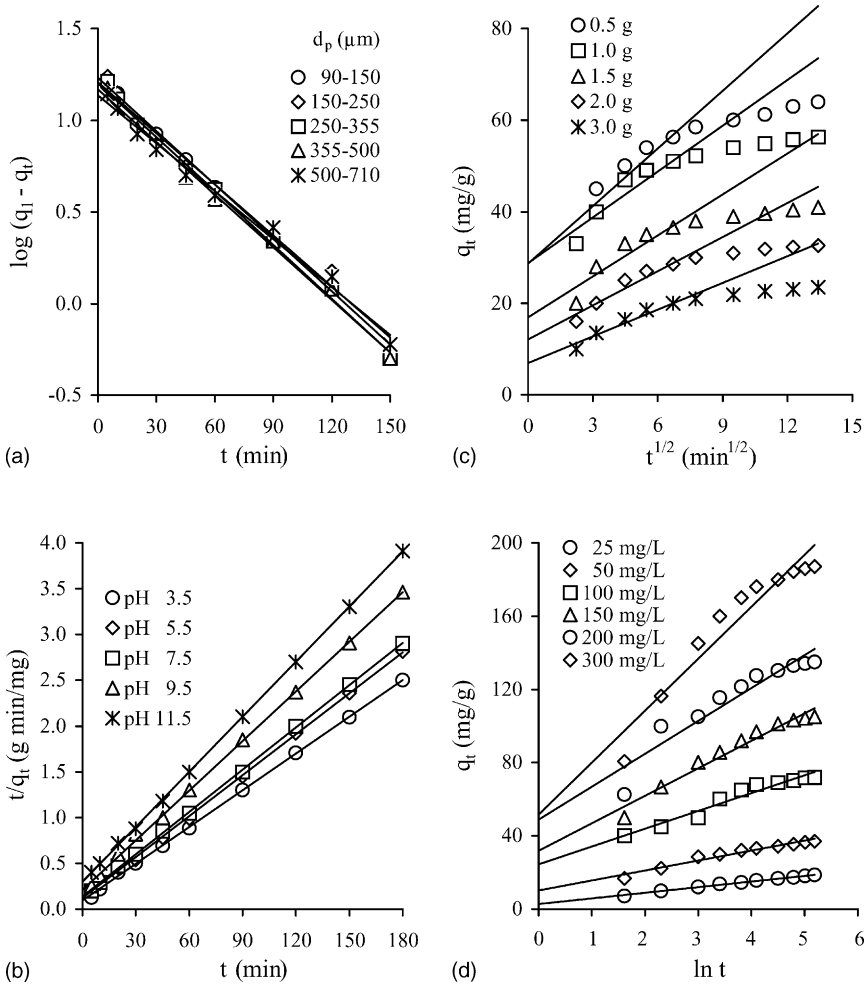

Fig. 2. Plots sorption kinetic equations for sorption of metal complex dyes onto pine sawdust, the pseudo first-order sorption kinetics of MCB at different particle size (a); the pseudo second-order sorption kinetics of MCY at different pH (b); the intrapaticle diffusion kinetics of MCB at different sawdust dose (c); the Elovich equation for sorption of MCY at different initial dye concentration (d).

predicted results with experimental data because all of the equilibrium sorption capacities, q_1, of pseudo first-order are lower than experimental results. The initial sorption rate, h, decreased with an increase in the pine sawdust particle size. While the initial sorption rate varied from 1.68 to 0.78 mg/g min as the pine sawdust particle size varied from 90–150 to 500–710 μm for MCB and from 7.06 to 2.64 mg/g min for the case of MCY.

4.2. Effect of initial pH

The experimental results of the sorption of MCY on pine sawdust at different initial pH are shown in Fig. 1(b). The sorption capacity, q_e, increases from 46 to 72 mg/g with a decrease in the pH from 11.5 to 3.5 for MCY and an increase from 15.6 to 56.3 mg/g with a decrease in the pH from 11.5 to 3.5 for MCB. Fig. 2(b) shows a plot of t/q_t versus t for the sorption of MCY for the pseudo second-order kinetic. The kinetic parameters obtained from linear plots of all the kinetic models and correlation coefficients are listed in Table 1 and are compared with r_2^2, the pseudo second-order rate parameters, k_2, and sorption capacity, q_2. The sorption of both of the metal complex dyes onto pine sawdust is best described by the pseudo second-order model. Other kinetic models fit the experimental results pseudo first-order equation, the Elovich equation and intraparticle diffusion equation, respectively, for the sorption of MCY and the Elovich equation, first-order equation and intraparticle diffusion equation, respectively, for the sorption of MCB. Again the equilibrium sorption capacities for second-order correlation are much more reasonable when compared with experimental results than those of the first-order system. Because most of the first-order q_1 values deviate by 30–70% from the experimental values, it suggests that the sorption of metal complex dyes onto pine sawdust follows the pseudo second-order model.

4.3. Effect of pine sawdust dose

The results of the effect of pine sawdust dose on experiments carried out using 100 mg/l initial dye concentration with particle size 90–150 μm for MCB are shown in

Table 1
Kinetic parameters for the effects of sawdust particle size and pH on sorption of metal complex dyes onto pine sawdust

Dye	Parameters	$q_{e,exp}$ (mg/g)	First-order kinetic equation			Second-order kinetic equation				The Elovich equation			Intraparticle diffusion equation	
			q_1 (mg/g)	k_1 (1/min) $\times 10^2$	r_1^2	q_2 (mg/g)	k_2 (g/(mg min)) $\times 10^4$	h (mg/(g min))	r_2^2	α	β	r_E^2	k_{int} (mg/(g min$^{1/2}$))	r_{int}^2
MCB	90–150 μm	25.1	17	4.21	0.989	27	2.31	1.682	0.998	5.41	0.2071	0.996	2.241	0.972
	150–250 μm	23.6	16.1	4.04	0.987	25.6	2.24	1.467	0.999	3.91	0.2044	0.991	2.370	0.949
	250–355 μm	21.5	15.9	4.26	0.993	23.6	2.18	1.214	0.999	3.06	0.2155	0.995	2.195	0.964
	355–500 μm	19.2	14.7	4.17	0.992	21.4	2.12	0.969	0.999	2.30	0.2315	0.995	2.014	0.967
	500–710 μm	17.4	13.6	4.21	0.989	19.5	2.05	0.781	0.999	1.88	0.2529	0.998	1.820	0.977
MCY	90–150 μm	62	31	4.65	0.973	64.9	1.68	7.062	0.999	30.53	0.0963	0.963	5.688	0.959
	150–250 μm	55.4	30.6	4.39	0.933	58.5	1.50	5.123	0.999	15.27	0.0953	0.954	5.912	0.946
	250–355 μm	50.1	33.3	4.60	0.962	53.8	1.29	3.745	0.999	10.62	0.1004	0.975	5.220	0.941
	355–500 μm	47.7	25.1	4.30	0.704	51.8	1.15	3.094	0.999	8.14	0.1007	0.988	4.963	0.959
	500–710 μm	44.3	32.3	4.56	0.987	48.8	1.11	2.638	0.999	6.36	0.1028	0.985	4.889	0.976
MCB	pH 3.5	56.3	17.8	4.26	0.970	57.5	3.52	11.64	0.998	426.8	0.1624	0.947	3.336	0.886
	pH 5.5	36	17.8	4.30	0.943	37.7	2.64	3.755	0.999	12.49	0.1537	0.941	3.741	0.941
	pH 7.5	25.1	17	4.21	0.989	27	2.31	1.682	0.998	5.31	0.2065	0.996	2.241	0.972
	pH 9.5	20	14.8	4.04	0.985	22	2.24	1.084	0.999	2.70	0.2301	0.993	2.085	0.975
	pH 11.5	15.6	13.1	4.04	0.981	17.6	2.16	0.669	0.999	1.59	0.2781	0.998	1.634	0.986
MCY	pH 3.5	72	30.9	5.12	0.970	74.6	1.97	10.98	0.999	120.8	0.1030	0.951	5.350	0.978
	pH 5.5	64	29	5.25	0.947	67.1	1.88	8.453	0.999	33.09	0.0919	0.919	6.488	0.934
	pH 7.5	62	31	4.65	0.973	64.9	1.68	7.062	0.999	30.53	0.0963	0.963	5.688	0.959
	pH 9.5	52	33.6	5.30	0.985	55.3	1.55	4.744	0.999	17.56	0.1071	0.977	4.805	0.977
	pH 11.5	46	31.1	4.95	0.991	49.8	1.36	3.385	0.999	8.70	0.1047	0.973	5.047	0.958

Table 2
Kinetic parameters for the effects of sawdust dose and initial dye concentration on sorption of metal complex dyes onto pine sawdust

Dye	Parameters	$q_{e,exp}$ (mg/g)	First-order kinetic equation			Second-order kinetic equation				The Elovich equation			Intraparticle diffusion equation	
			q_1 (mg/g)	k_1 (1/min) $\times 10^2$	r_1^2	q_2 (mg/g)	k_2 (g/(mg min)) $\times 10^4$	h (mg/(g min))	r_2^2	α	β	r_E^2	k_{int} (mg/(g min$^{1/2}$))	r_{int}^2
MCB	0.5 g/l	64	22.8	3.78	0.953	65.4	2.48	10.63	0.999	188.8	0.1276	0.948	4.190	0.876
	1.0 g/l	56.3	17.8	4.26	0.970	57.5	3.52	11.64	0.999	426.8	0.1624	0.947	3.336	0.886
	1.5 g/l	40.9	14.5	4.21	0.955	42	4.03	7.112	0.999	89.8	0.1883	0.925	2.982	0.865
	2.0 g/l	32.6	14.4	4.82	0.984	33.8	4.14	4.728	0.999	40.80	0.2170	0.964	2.485	0.935
	3.0 g/l	23.5	11.5	4.21	0.981	24.5	4.41	2.646	0.999	14.47	0.2693	0.977	1.947	0.950
MCY	0.5 g/l	79	31.6	4.82	0.932	82	1.73	11.63	0.999	67.68	0.0827	0.929	7.143	0.963
	1.0 g/l	72	30.9	5.12	0.970	74.6	1.97	10.98	0.999	120.8	0.1030	0.951	5.350	0.978
	1.5 g/l	53.3	11.2	3.82	0.868	54.1	5.51	16.13	0.999	2468	0.2081	0.906	2.891	0.906
	2.0 g/l	45	9.8	2.95	0.965	45.2	5.69	11.63	0.999	32918	0.3224	0.991	1.467	0.941
	3.0 g/l	32.7	8.6	4.26	0.956	33.2	7.61	8.398	0.999	732.3	0.3152	0.908	1.767	0.823
MCB	25 mg/l	15.1	8	3.86	0.945	15.8	5.52	1.378	0.999	5.008	0.3719	0.964	1.449	0.925
	50 mg/l	29.3	13	3.78	0.956	30.2	3.92	3.574	0.999	33.42	0.2444	0.972	2.043	0.889
	100 mg/l	56.3	17.8	4.26	0.970	57.5	3.52	11.64	0.999	426.8	0.1624	0.947	3.336	0.886
	150 mg/l	82	23.8	4.52	0.941	83.3	2.72	18.87	0.999	651.1	0.1112	0.920	5.251	0.908
	200 mg/l	103	36.3	4.82	0.954	105.3	1.76	19.57	0.999	265.5	0.0760	0.933	7.707	0.939
	300 mg/l	135	45.4	5.25	0.924	138.9	1.35	26.11	0.999	179.3	0.0513	0.879	12.25	0.896
MCY	25 mg/l	18.6	10	3.47	0.982	19.5	4.16	1.584	0.998	7.37	0.3230	0.994	1.467	0.944
	50 mg/l	37	15.8	4.04	0.963	38.2	3.33	4.864	0.999	36.18	0.1853	0.956	2.909	0.901
	100 mg/l	72	30.9	5.12	0.970	74.6	1.97	10.98	0.999	120.8	0.1030	0.951	5.350	0.978
	150 mg/l	105	48.7	5.20	0.993	108.7	1.29	15.20	0.999	123.7	0.0664	0.964	8.012	0.930
	200 mg/l	135	53.2	5.34	0.980	138.9	1.25	24.10	0.999	274.8	0.0558	0.894	10.11	0.824
	300 mg/l	187	84.5	5.65	0.978	192.3	0.788	29.16	1	174.6	0.0353	0.922	16.61	0.905

Fig. 1(c). The results are also shown in Fig. 2(c) as a plot of q_t versus the square root of t for the intraparticle diffusion rate model for the sorption of MCB onto pine sawdust. Kinetic parameters from linear plots of four kinetic models are given in Table 2. The data for MCB and MCY again show a good compliance with the pseudo second-order equation and the regression coefficients, r_2^2, for the linear plots were all high (>0.998). Again, the equilibrium sorption capacity, q_2, for second-order are slightly more reasonable than those of the first-order when comparing predicted results with experimental data because of the equilibrium sorption capacities, q_1, are lower than experimental results. Again, the pseudo second-order model provides the best correlation for all the sorption processes while the intraparticle diffusion model fits the experimental data well for an initial period of the sorption processes only. The overall rate of metal complex dyes sorption processes appear to be controlled by the chemical process in this case in accordance with the pseudo second-order reaction mechanism.

4.4. Effect of initial dye concentration

The experimental results of the sorption of MCY on pine sawdust at various concentrations are shown in Fig. 1(d). The sorption capacities at equilibrium, q_e, increases from 15.1 to 135 mg/g and from 18.6 to 187 mg/g with an increase in the initial dye concentration from 25 to 300 mg/l with a pine sawdust dose 1 g/l for MCB and MCY, respectively. Fig. 2(d) shows a plot of q_t versus ln t for the sorption of MCY for the Elovich equation. The characteristic parameters of all the kinetic models and correlation coefficients are tabulated in Table 2. Again correlation coefficients of second-order equation for all concentrations are higher than those of other kinetic models and its calculated equilibrium sorption capacities, q_2, fit well the experimental data. These suggest that the pseudo second-order sorption mechanism is predominant and that the overall rate of the metal complex dyes sorption process appear to be controlled by the chemical process. The similar phenomena have also been observed in biosorption of dyes RB2, RY2, and Remazol Black B on biomass [15,16]. For the pseudo second-order model, the rate constant decreases with an increasing of initial dye concentration, while the initial sorption rate, h, increases with an increasing of initial dye concentration (Table 2). An increase in initial dye concentration results in significant increasing the q_2.

A comparison of calculated and measured results for 100 mg/l initial dye concentration is shown in Fig. 3. The pseudo second-order equation provides the best correlation for all of the sorption process, whereas the Elovich equation also fits the experimental data well. The pseudo first-order and intraparticle equations do not give a good fit to the experimental data for the sorption of metal complex dyes.

This suggest that the sorption systems studied belong to the second-order kinetic model, based on the assumption that the rate limiting step may be chemical sorption

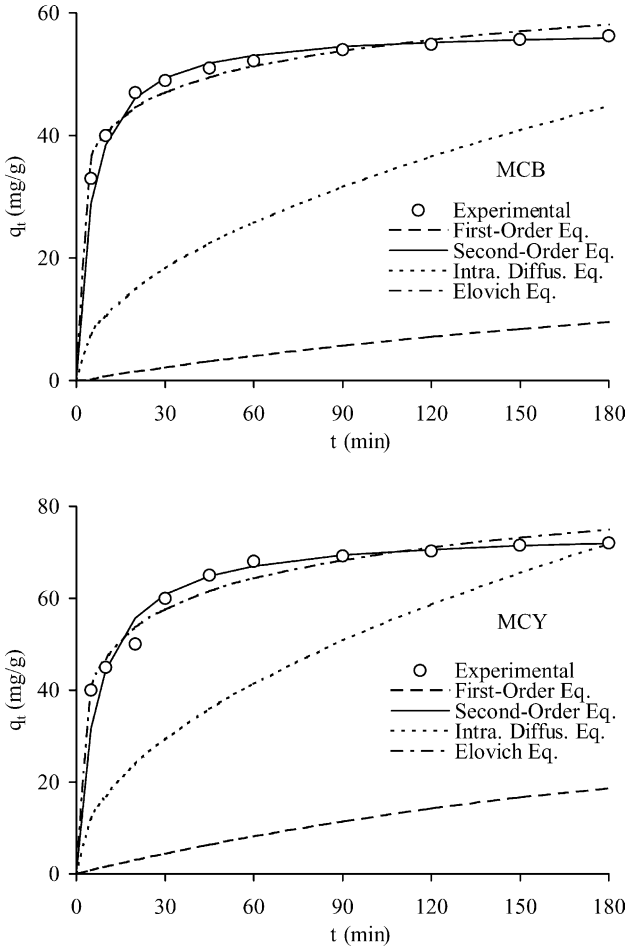

Fig. 3. Comparison between the measured and modeled time profiles for sorption of metal complex dyes (100 mg/l initial dye concentration) onto pine sawdust.

or chemisorption involving valency forces through sharing or exchange of electrons between sorbent and sorbate. The agreement of the Elovich equation with experimental data may be explained as below. The previous successful applications of the Elovich equation to heterogeneous catalyst surfaces helps to explain its success in predicting the sorption of metal complex dyes on pine sawdust. The general explanation for this form of kinetic law involves a variation of the energetics of chemisorption with the active sites are heterogenous sawdust and therefore, exhibit different activation energies for chemisorption [25]. Because the cell walls of sawdust mainly consist of cellulose and lignin, and many hydroxyl groups, such as tannins or other phenolic compounds [13].

5. Conclusion

The kinetics of sorption of MCB and MCY onto pine sawdust were studied by using pseudo first-order and pseudo second-order equations, intraparticle diffusion equation and

the Elovich equation. The sorption kinetics of MCB and MCY was studied as a function of particle size and mass of sawdust, pH and initial dye concentration. For all the systems examined, the pseudo second-order kinetic model provided the best correlation of the experimental data. The pseudo second-order equation is based on the sorption capacity on the solid-phase and is in agreement with a chemisorption mechanism being the rate controlling step. The sorption of metal complex dyes onto pine sawdust can also be successfully interpreted by the Elovich equation. This supports the heterogeneous sorption mechanism likely to be responsible for dye uptake.

All findings presented in this study suggest that MCB–MCY/pine sawdust systems can not be described by a first-order reaction and intraparticle diffusion model. Two kinetic models have been used extensively to describe the sorption of dyes onto sorbent. The main disadvantage of these models are that the linear first-order equation does not give theoretical q_e values which agree with experimental q_e values, and the plots of intraparticle diffusion model are only linear over the initial 60 min of the sorption process.

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