

Kinetic models for methylene blue adsorption using activated human hair waste as a biosorbent

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Adsorption kinetics of basic dyestuff (methylene blue) onto chemically treated human hair waste as a biosorbent material was studied. The rate of adsorption has been calculated under most important process parameters, initial dye concentration, agitation speed, adsorbent dose and contact time. The experimental data showed that the adsorption of dye was a gradual process. The rate of adsorption was found to be very rapid in the initial contact period (initial rapid range) and thereafter the process tends to proceed at a slower rate and finally reaches equilibrium (slower range). Higher percentage of dye was removed with decrease in the initial concentration of dye and increase in amount of adsorbent used. Lagergren pseudo first-order, pseudo second-order and Elovich model were used to fit the experimental data and a comparison of the three models has been carried out. Results show that the two latter models agree well with the experimental data, whereas the first one proposed fits the experimental data well for the initial period of process only. The highest correlation coefficients were obtained for the pseudo second-order kinetic model.

يهدف هذا البحث الي دراسة حركية امتزاز صبغة الميثيلين الأزرق (BB69) باستخدام مخلف شعر الانسان المعالج كيميائياً كمادة حيوية مازة. تم حساب معدل الامتزاز عند المتغيرات المهمة للعملية وهي التركيز الابتدائي للصبغة؛ سرعة التقلب؛ كمية المادة المازة وزمن العملية. وقد أوضحت النتائج أن عملية الامتزاز للصبغة تحدث تدريجياً ووجد أن معدل الامتزاز يكون سريع جداً في المرحلة الاولى من العملية (initial rapid range) ثم بعد ذلك تتم العملية بمعدل منخفض الي أن تصل لحالة الاتزان (slower range). وقد وجد أن النسبة المئوية لعملية الازالة تزداد مع نقص التركيز الابتدائي للصبغة وزيادة كمية المادة المازة المستخدمة. وقد استخدمت ثلاث نماذج رياضية في معالجة النتائج ومن ثم تم تقييم هذه النماذج من حيث أهم أكثر تطابقاً مع النتائج العملية والذي يستخدم فيما بعد لدراسة حركية امتزاز هذه الصبغة بواسطة الشعر المعالج كيميائياً وهم (Lagergren first-order, second-order and Elovich mode) وقد وجد أن كل من النموذج الثاني والثالث أكثر تطابقاً مع النتائج العملية بينما النموذج الأول يكون متوافق فقط خلال الفترة الابتدائية لعملية الامتزاز. اما أعلى معامل تصحيح (تطابق) فلقد وجد للنموذج الثاني (second-order mode).

Keywords: Keratinous waste, Human hair, Dyestuff, Kinetics, Adsorption

1. Introduction

Color is a visible pollutant and presence of even very minute amount of coloring substance makes it undesirable due to its appearance. Further, dye effluents may contain chemicals that exhibit toxic effects toward microbial populations and can be toxic and/or carcinogenic to mammalian animal. The removal of color from dye bearing effluents is one of the major problems due to the difficulty in treating such wastewaters by conventional treatment methods. Currently adsorption processes are proved to be an effective process for the removal of pollutants from wastewaters [1-3]. The treatment of dye wastewaters by adsorption technique is

receiving growing attention since the standards for quality of effluent disposal are becoming increasingly more rigid. The major advantages of an adsorption technique for water pollution control are less investment in terms of initial cost and land, simple design and easy operation, no effect by toxic substances, and superior removal of organic waste constituents as compared to conventional biological treatment processes.

Activated carbon is the most commonly used adsorbent and has proved to be an effective process for the removal of various pollutants from wastewaters. However the activated carbon processes are found to be an expensive adsorption process because of the high cost of activated carbon. The use of high

cost activated carbons based on the non-renewable and relatively expensive starting materials such as coal is unjustified in most pollution control processes. This has prompted several researches for searching low-cost alternative adsorbents such as wood [4], bagasse pith [5], maize cob [6], peanut hulls [7], clay [8], orange peel [9], bleaching earth [10], and human hair [11]. The use of such human hair waste as an adsorbent, biosorbent, would help to reduce the cost of wastewater treatment and would make a contribution to cleaning the environment.

In previous work [11, 12] preliminary testes were undertaken to assess the contact time necessary for dye-hair system to come to equilibrium and, for experimental purposes, each system was given a contact time in excess of this period. Experiments showed that such equilibrium was established within 100 minutes; however, all experiments were allowed to run for 180 minutes. A series of studies for adsorption of two dyestuffs (BB69 and AB25) onto natural and chemically treated human hair were determined through the adsorption isotherms. It was found that the adsorption capacity of natural human hair is limited, while activated hair exhibited high dye removals, also showing that the adsorption capacity was comparatively high for the basic dye and was lower for the acidic dye. The optimum conditions for activation human hair was determined according to maximum adsorption capacity, it was the treatment with 0.1N-NaOH/0.1N-Na₂S for 20 minutes at room temperature. The applicability of common isotherm models (i.e., Langmuir, Freundlich and Redlich-Peterson) was studied.

The main object of the present contribution is to study the adsorption kinetics of Basic Blue 69 onto activated human hair at different system variables. The variables investigated include initial dye concentration, agitation speed, and mass of human hair (dose). The usefulness of common adsorption kinetics models was also evaluated.

2. Theoretical approach

Several models can be used to express the mechanism of solute adsorption onto an adsorbent. In order to investigate the mechanism of dye-hair adsorption system, characteristic constants of adsorption were determined using a pseudo-first order, pseudo-second order, and the Elovich kinetic models. The conformity between experimental data and the model-predicted values was expressed by the correlation coefficients (R^2 , values close or equal to 1). A relatively high R^2 value indicates that the model successfully describes the kinetics of dye adsorption.

2.1. Pseudo-first order kinetic model

The pseudo-first order rate equation based on solid capacity given by Lagergren and Svenska [13] is defined as,

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

Integration of this equation for the boundary conditions $q_t=0$ at $t=0$ and $q_t=q_t$ at $t=t$, eq. (1) may be rearranged for linearized data plotting as shown by eq. (2):

$$\ln(q_e - q_t) = \ln(q_e) - k_1 t. \quad (2)$$

Therefore a plot of $\ln(q_e - q_t)$ versus time will give a straight line and from the gradient and intercept, values of k_1 and q_e can be obtained.

2.2. Pseudo-second order kinetic model

The pseudo-second kinetic model is expressed as [14],

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

Integration of this equation for the boundary conditions $t=0$ to $t=t$ and $q_t=0$ to $q_t=q_t$, gives:

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

which is the integrated rate law for a pseudo-second reaction.

Eq. (4) can be rearranged to obtain eq. (5), which has a linear form,

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

The plot of (t/q_t) versus t should give a linear relationship, where, the constants, q_e and k_2 , can be extracted from its gradient and intercept.

2.3. The Elovich kinetic model

The Elovich model equation is generally expressed as,

$$\frac{dq_t}{dt} = \alpha \exp(-\beta q_t), \quad (6)$$

where α is the initial adsorption rate and β is the desorption constant during any one experiment. To simplify the Elovich equation, Chien and Clayton [15] assumed $(\alpha\beta t) \gg t$ and by applying the boundary conditions $q_t=0$ at $t=0$ and $q_t=q_t$ at $t=t$, eq. (6) becomes:

$$q_t = \frac{\ln(\alpha\beta)}{\beta} + \frac{1}{\beta} \ln(t). \quad (7)$$

If the adsorption data fits the Elovich model, a plot of q_t versus $\ln(t)$ should be a linear relationship with a slope of $(1/\beta)$ and an intercept of $(1/\beta)\ln(\alpha\beta)$.

3. Materials and methods

The dye, adsorbate, used in the experiments was methylene blue (BB69). The dyestuff was supplied by Bayer and was used as the commercial salts. No structure is available for this dye. It belongs to the methane class of which chromophore is a conjugated chain of atoms terminated by an ammonium group, and in addition a nitrogen, sulphur or oxygen atom, or an equivalent unsaturated group. The dye was makeup in stock solution of concentration 2000 mg/dm³ and was subsequently diluted to the required

concentration using distilled water. The concentrations of the dye in aqueous solutions were determined using a spectrophotometer (Spectro-Plus MK1A). All measurements were made at the wave length corresponding to maximum absorbance, λ_{max} , which was 585 nm for BB 69.

The adsorbent used in this investigation was human hair treated chemically at optimum conditions. Human hair waste collected from various local barber shops, were mixed together, washed, clean of the adhering dirt with a detergent, rinsed several times with distilled water and finally dried in an electric oven at 100 °C before being used. For each pretreatment process, about 4 grams of the dried hair was weighed out and soaked in 250 ml of a pretreatment reagent, 0.1N-NaOH/0.1N-Na₂S, at room temperature. After a given soaking time, 20 minutes, the solution was filtered and the hair washed clean with tap water, deionized water and then dried again in an electric oven. The dried treated hair is stored in a clean plastic bin to avoid the adsorption of moisture from environment, after this it can be used directly. Details of the dye and human hair were presented in an earlier paper in which equilibrium adsorption isotherms for the dye-hair system were reported [12].

All the experiments of kinetics were carried out in a batch adsorber vessel with a standard tank configuration. The batch adsorber was a baffled 2 dm³ Perspex vessel and holding a volume of 1.7 dm³ dye solution in each run. Constant and uniform agitation was achieved using a variable speed motor and a six bladed (flat) impeller. Eight perspex baffles were evenly spaced around the circumference of the adsorber. Further design details of the batch adsorber used in the kinetic studies have been reported previously [16]. The variables studied were initial dye concentration, agitation speed and adsorbent dose. In the adsorbing stage, after all adsorption conditions were set, the dye concentration was immediately measured at specified time intervals until equilibrium was reached. The amount of dye adsorbed (q_t , mg/g) was determined as,

$$q_t = V(C_0 - C_t)/m. \quad (8)$$

4. Results and discussion

4.1. Effect of initial concentration

The influence of initial dye concentration on the rate and extent of adsorption of chemical treated human hair was studied at an adsorbent dosage of 4 g in 1.7 dm³ dye solution and agitation speed of 400 rpm, the experimental results obtained are depicted in fig. 1 as a plot of (q_t) against time. It is evident from fig. 1 that the amount of dye adsorbed gets increased with increasing initial dye concentration and contact time. It was observed that the rate of dye uptake was found to be very rapid for the initial contact period of 50 min and thereafter the dye uptake process tends to proceed at a slower rate and finally reaches saturation. Decreasing of adsorption rate with time is due to the continuous decrease in the dye concentration deriving force. Similar results have been observed for dye-jute fiber carbon system [17]. The predicted rate of adsorption, r , for both regions is shown in fig. 2. Rate of adsorption for different initial dye concentration fits eqs. (9 and 10) for the initial rapid range and the later slower range, respectively:

$$r_{\text{rapid range}} = 0.6164 \ln(C_0) - 2.768, R^2 = 0.99. \quad (9)$$

$$r_{\text{slower range}} = 0.0003 C_0 - 0.032, R^2 = 0.98. \quad (10)$$

Fig. 3 shows a plot of the linearized form of pseudo-first order model in eq. (2) at various initial dye concentrations. However, the experimental data deviate considerably from the theoretical data especially for high concentrations. Since, pseudo-first order model as a plot is not shown for the influence of other parameters. Values of k_1 , q_e and correlation coefficient (R^2) at different initial dye concentrations are presented in table 1, from which it will be seen that the kinetics of dye adsorption on activated human hair adsorbent is not successfully described by this model (lower values of correlation coefficient).

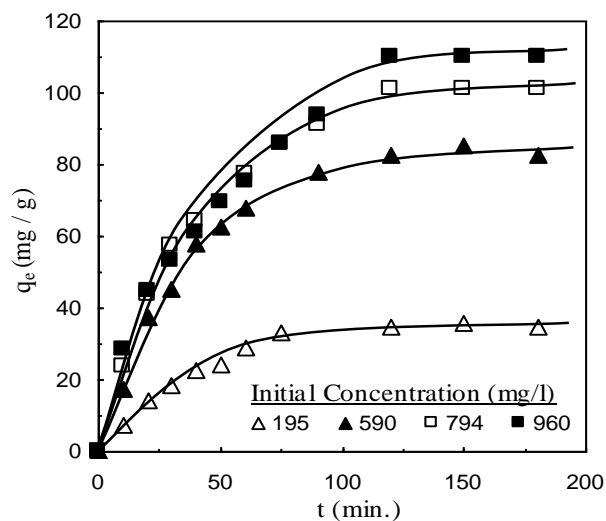


Fig. 1. Plots of adsorbed dye versus time at different initial dye concentrations.

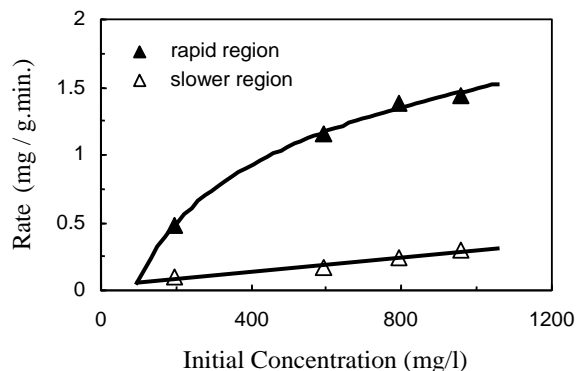


Fig. 2. Effect of initial dye concentration on the rate of dye uptake.

Table 1
Kinetic parameters for different models
(different initial concentrations)

Kinetic parameters	Concentration (mg.dm ⁻³)			
	195	590	794	960
<i>Pseudo-first order constants</i>				
$k_1 \times 10^2$	2.44	2.37	3.13	3.16
q_e	32.4	75.9	116.1	151.3
R^2	0.80	0.88	0.91	0.83
<i>Pseudo-second order constants</i>				
$k_2 \times 10^4$	5.42	2.62	2.09	1.46
q_e	45.1	104.2	126.6	142.9
R^2	0.99	0.98	0.99	0.99
<i>Elovich equation constants</i>				
α	2.218	5.806	6.103	6.935
β	0.095	0.042	0.035	0.032
R^2	0.96	0.97	0.98	0.97

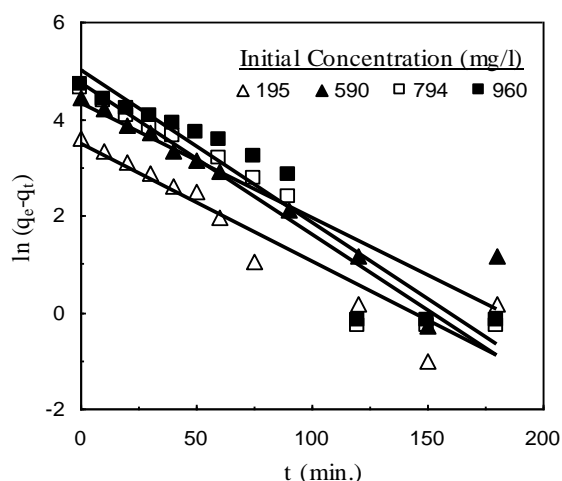


Fig. 3. Pseudo first-order adsorption kinetics at various initial dye concentrations.

The kinetic data were also analyzed using pseudo-second order model. Plots of the mode in linearized form are shown in fig. 4. The constants of eq. (5) that were obtained from slope and intercept of straight line plots of t/q_t against t and are listed in table 1, from which it is seen that the kinetics of the dye adsorption on the chemically treated human hair adsorbent follows this model with high values of correlation coefficients (0.98-0.99). Ho and McKay presented similar results for the adsorption of basic and acid dyes onto peat, wood and pith [14]. The adsorption capacity q_e increased from 45.1 to 142.9 mg/g when the initial dye concentration increased from 195 to 960 mg/dm³. Values of the rate constant were found to decrease from 5.42×10^{-4} to 1.46×10^{-4} g/mg.min for an increase in dye initial concentration from 195 to 960 mg/dm³.

Elovich model is the third one which was also checked in this investigation. The coefficients of this model, a and β , were calculated from the slope and intercept of the straight line plots of q_t against $\ln(t)$ which is represented in fig. 5. Table 1 lists the kinetic constants beside correlation coefficient of linear fitting. It is seen from the data that the values of a and β varied as a function of the initial dye concentration. Thus, on increasing initial dye concentration from 195 to 960 mg/dm³, the value of initial adsorption rate, a , increases from 2.22 to 6.94 mg/g.min, where β decreases from 0.095 to 0.032 g/mg.

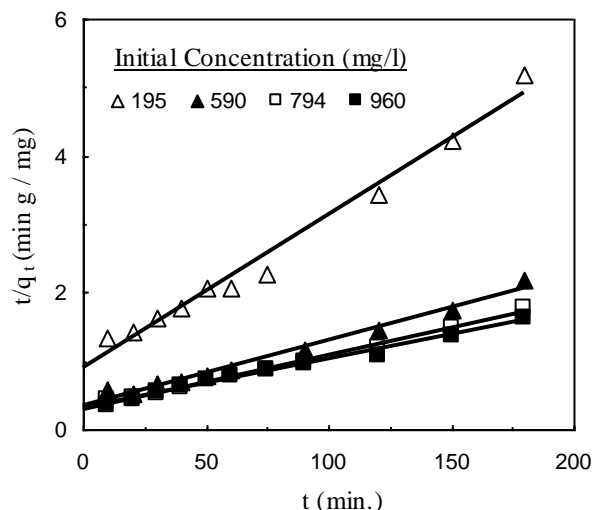


Fig. 4. Pseudo second-order adsorption kinetics at various initial dye concentrations.

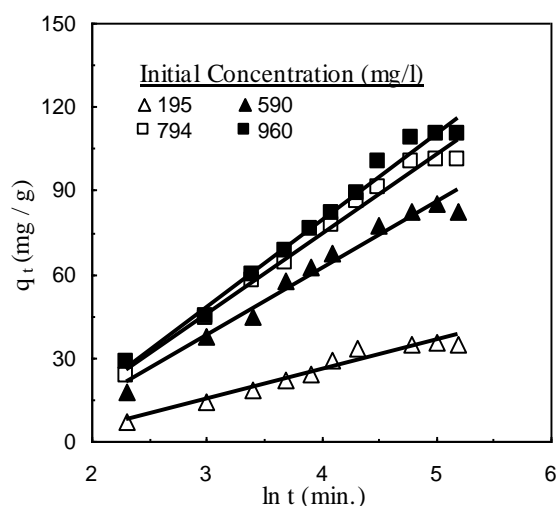


Fig. 5. Plot of Elovich adsorption kinetics model at various initial dye concentrations.

4.2. Effect of agitation speed

Fig. 6 shows the experimental results obtained from a series of contact time studies for the adsorption of BB69 onto chemically treated human hair at an adsorbent dosage of 4 g in 1.7 dm³ dye solution of 684 mg/dm³ concentration in which degree of agitation was varied from 200 to 1000 rpm. The results indicate that the rate of adsorption is controlled by the degree of agitation. The increase in the extent of removal can be explained on the following basis: (1) the dye

may get more activated due to a gain in kinetic energy and easily crosses the potential barrier, (2) the increase in agitation speed decreases the boundary layer resistance to mass transfer in the bulk solution and increases the driving force of solute ions. It was observed that the rate of adsorption decreased with time until it gradually approached a plateau at saturation, and it was classified into rapid and slower ranges. The effects of agitation speed on adsorption rate in both ranges are shown in fig. 7. It can be observed that, rate of adsorption in initial rapid and slower range was found to be increasing with increase in agitation speed. The influence of agitation speed on the rate of dye uptake in the initial rapid range and slower range fits the eqs. (11 and 12), respectively:

$$r_{\text{rapid range}} = 0.2544 \ln(\text{rpm}) - 0.199, R^2 = 0.96. \quad (11)$$

$$r_{\text{slower range}} = 0.00005 \text{ rpm} + 0.148, R^2 = 0.97. \quad (12)$$

The constants of pseudo-first order kinetic model were calculated and represented in table 2. Lower values for the correlation coefficient were observed.

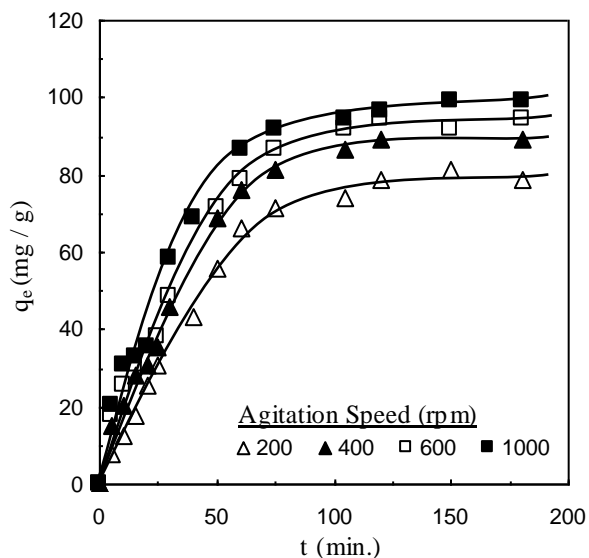


Fig. 6. Plots of adsorbed dye versus time at different agitation speeds.

Table 2
Kinetic parameters for different models (different agitation speeds)

Kinetic parameters	Agitation speed (rpm)			
	200	400	600	1000
<i>Pseudo-first order constants</i>				
$k_1 \times 10^2$	2.52	2.99	2.81	2.92
q_e	85.6	91.2	93.3	96.5
R^2	0.88	0.93	0.91	0.93
<i>Pseudo-second order constants</i>				
$k_2 \times 10^4$	1.21	1.94	2.35	2.62
q_e	119.1	117.7	117.6	120.5
R^2	0.95	0.97	0.99	0.98
<i>Elovich equation constants</i>				
α	4.440	6.060	7.254	8.413
β	0.041	0.039	0.039	0.038
R^2	0.95	0.94	0.95	0.95

Table 3
Kinetic parameters for different models (different human hair masses).

Kinetic Parameters	Human Hair Mass (g)		
	2	4	5
<i>Pseudo-first order constants</i>			
$k_1 \times 10^2$	2.19	2.41	2.52
q_e	144.3	119.4	106.5
R^2	0.77	0.89	0.91
<i>Pseudo-second order constants</i>			
$k_2 \times 10^4$	0.94	1.19	1.27
q_e	180.3	156.3	138.9
R^2	0.98	0.99	0.98
<i>Elovich equation constants</i>			
α	9.841	6.875	5.479
β	0.022	0.030	0.033
R^2	0.92	0.97	0.96

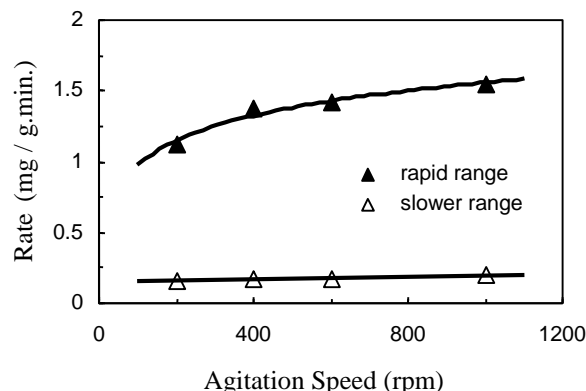


Fig. 7. Effect of agitation speed on the rate of dye uptake.

Constants of pseudo-second order kinetic model, k_2 , q_e and R^2 , were calculated from the intercept and slope of the straight line plots in fig. 8 and are listed in table 2. The data showed a good compliance with the pseudo-second order model and the correlation coefficients for the linear plots were very good (0.97-0.99). The value of rate constant, k_2 , were found to increase from 1.21×10^{-4} to 2.62×10^{-4} g/mg.min as the agitation speed increased from 200 to 1000 rpm. The data were also analyzed using Elovich model. The kinetic constants depending on Elovich model were obtained from the straight line plots shown in fig. 9, and presented in table 2. Thus, the initial adsorption rate, a , and the desorption constant, β , also varied as a function of agitation speed.

4.3. Effect of human hair mass

The influence of human hair mass on the adsorption rate has also been studied keeping the other experimental conditions constant (400 rpm and 684 mg/dm³). The results are shown in fig. 10 as a plot of q_t against t where adsorbent dosage was varied 2, 4 and 5 g in 1.7 dm³ dye solution. The results indicate that the rate of adsorption is affected by adsorbent mass as well as contact time. It was observed that the amount of dye adsorbed onto unit mass of adsorbent decreased with increasing human hair mass (dose), whereas the percent dye removal increases with increasing adsorbent dose. The decrease in q_t value may be due to the splitting effect of flux (concentration gradient) between adsorbate and adsorbent with increasing human hair concentration causing a decrease in amount of dye adsorbed onto unit weight of hair. The increase in percent dye removal is because, of the high human hair mass. There is a very fast superficial adsorption onto the hair surface that produces a lower dye concentration in the solution than when human hair mass is lower. On the other hand, the increase in the extent of removal indicated that with an increased mass of adsorbent, more surface area is made available and therefore the total number of sites increases.

For each specified adsorbent dosage, rate of dye uptake was found to be very rapid for the initial contact period, initial rapid range, and thereafter it decreases with time until equilibrium reached, (slower range). The effect of adsorbent mass on the rate of dye uptake in the rapid and slower ranges are shown in fig. 11. The adsorption rate in both ranges gets decreased with increasing human hair concentration. Similarly the effect of human hair mass on the rate of uptake in the initial rapid and slower range

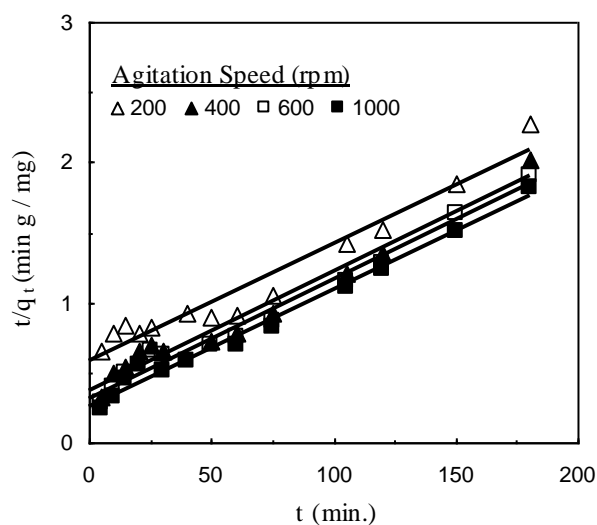


Fig. 8. Pseudo second-order adsorption kinetics at various agitation speeds.

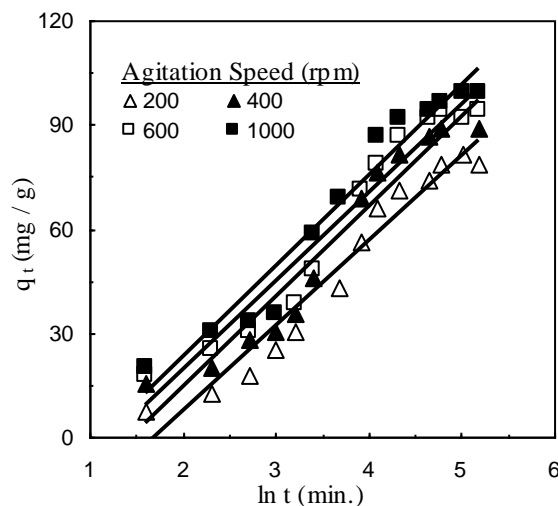


Fig. 9. Plot of Elovich adsorption kinetics model at various agitation speeds.

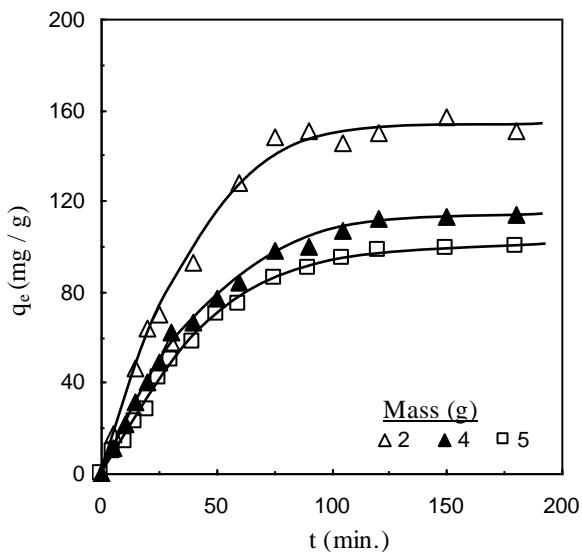


Fig. 10. Plots of adsorbed dye versus time at different human hair masses.

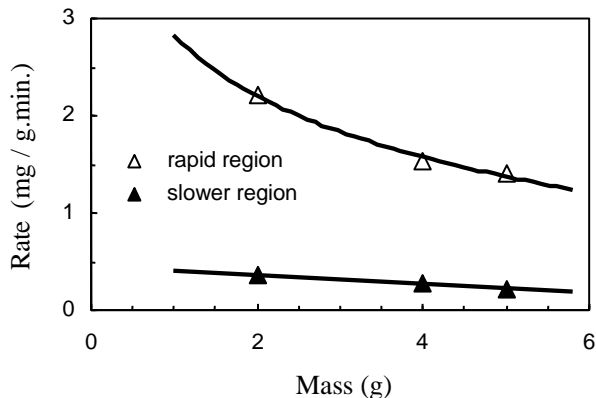


Fig.11. Effect of human hair mass (dose) on the rate of dye uptake.

were calculated and it fits the eqs. (13 and 14), respectively:

$$r_{\text{rapid range}} = -0.9036 \ln(m) + 2.828, R^2 = 0.99. \quad (13)$$

$$r_{\text{slower range}} = -0.0459 m + 0.4541, R^2 = 0.97 \quad (14)$$

The experimental data were fitted by the three models and the constants of each model are listed in table 3. The results showed that the constants varied as a function of mass of adsorbent. The straight line plots for pseudo-second order model are represented in fig. 12, where fig. 13 represents Elovich model at various adsorbent masses. Calculated

correlations are closer to unity for second-order kinetics model; therefore the adsorption kinetics is approximated more favorably by second-order kinetic model than other models. The results showed that the adsorption capacity for BB69 increases from 138.9 to 180.3 mg/g with the decrease in mass of human hair from 5 to 2 g.

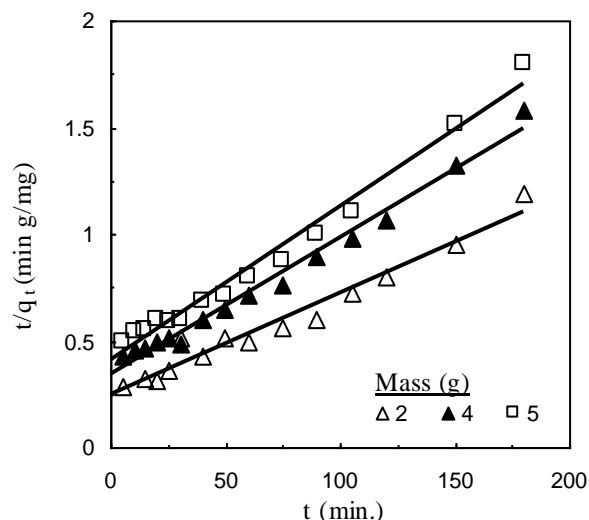


Fig. 12. Pseudo second-order adsorption kinetics at various human hair masses (dose).

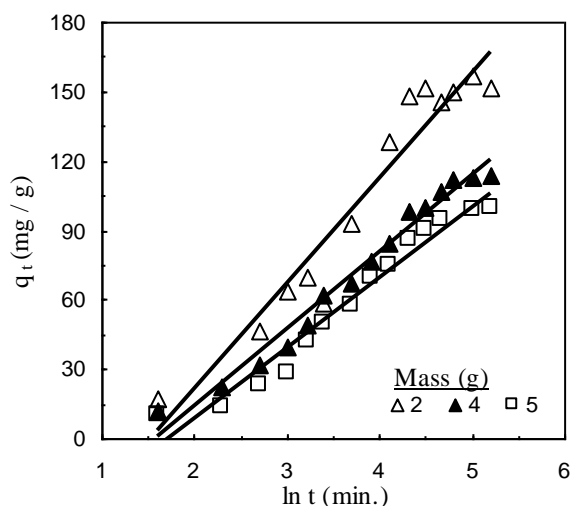


Fig.13. Plot of Elovich adsorption kinetics model at various human hair masses (dose).

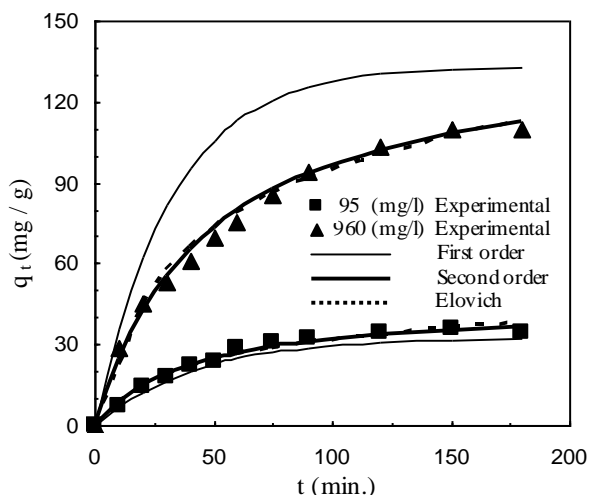


Fig. 14. Comparison of dye adsorbed predicted from kinetic models with experimental data at two different initial dye concentrations.

The experimental data, in order to establish which model yields the best fit. Obviously, it can be seen that, both pseudo-second order and Elovich kinetic models can be used successfully in simulating the kinetics of BB69 adsorption onto activated human hair, while pseudo-first order kinetic model is not agreement with experimental data. The same conclusion has been obtained before as reflected from correlation coefficient (R^2) in tables 1, 2 and 3.

5. Conclusions

The present study shows that activated human hair can be used as a potential adsorbent for the removal of basic dye (methylene blue) from aqueous solution. Batch kinetic studies performed on the hair-dye system at different operating conditions indicated the adsorption capacity. The amount of dye uptake (mg/g) was found to increase with increasing dye concentration, agitation speed as well as contact time and found to decrease with increase in human hair dosage. The rate of dye uptake was found to be very rapid for the initial contact period (initial rapid range) and thereafter the process tends to proceed at a slower rate (slower range) and finally reaches equilibrium. The kinetics data was fitted with three different models, pseudo first-order, pseudo second-

order and Elovich model. The kinetics data tended to fit very well by second-order kinetics model, confirming the chemisorptions of methylene blue on activated human hair. By second-order kinetic model, calculated q_t values agreed well with the q_t experimental values, supporting the chemisorptions and the applicability of second-order mode, since it recommended its use for design purposes.

Nomenclature

- C_0 initial dye concentration (mg/dm³),
- C_t dye concentration at time t (mg/dm³),
- k_1 rate constant of pseudo first-order adsorption (dm³/min),
- k_2 rate constant of pseudo second-order adsorption (g/mg min),
- m adsorbent dosage (g),
- q_e amount of dye adsorbed at equilibrium (mg/g),
- q_t amount of dye adsorbed at time t (mg/g),
- t contact time (min),
- V volume of solution to be treated (dm³),
- a initial adsorption rate, Elovich model (mg/g min), and
- β desorption constant, Elovich model (g/mg).

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