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Mass transfer kinetic model and removal capacity of acid blue 29 adsorptions onto activated carbon

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Mass transfer kinetic model and removal capacity of acid blue 29 adsorptions onto activated carbon

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Abstract. So far, the handling of dye pollution remains a challenge. One of the promising methods to reduce the dyes content from such waste is adsorption. The purpose of this study is to examine the suitability of kinetic models and to determine the removal capacity of acid blue 29 adsorptions onto activated carbon. Two mathematical models were developed to evaluate the adsorption kinetics. The first model only took into account the convective mass transfer of the dyestuff to the surface of the adsorbent, while the second model also took into account the diffusion of the dyes in the adsorbent pore. The result showed that the adsorption process followed the Langmuir equilibrium and the kinetic fits to the second model in which the adsorption rate is controlled by convection mass transfer in the liquid phase as well as the intra-particle molecular diffusion that occurs in the pore. The dye removal capacity inversely proportional to the dye concentration and reaches 86.6% when the initial dye concentration is 75 ppm.

1. Introduction

Up to recent decades, the contamination of industrial waste streams by synthetic dyes still becomes a problem which needs special attention [1]. Dyes are widely used in industries such as cosmetics, leather, printing, paper, food, and textiles. During the coloring process, approximately 10-15% of the dyes will enter the waste stream [2]. Dyestuff molecules are stable, so that when these chemicals enter the human or animal body or plant tissue. These substances will be stable in these tissues for a very long time[3].

The adverse effect of these chemicals on an aquatic ecosystem is to reduce the oxygen content and penetration of light in water. Besides, in the ecosystem, the pollutant inhibits photosynthesis and growth of biota [4]. Moreover, it was reported that exposure to a small amount of dye (<1 ppm) can cause several adverse effects such as skin irritation, dermatitis, allergy, cancer and even mutagenic effects on aquatic organisms and human [5].
Many methods and technologies were developed to remove these pollutants such as coagulation, precipitation, filtration, ion exchange, membrane technology, and adsorption. Among these methods, adsorption is seen as a very prospective choice because it is technically simple, high efficiency and economically competitive [6,7].

Studies of acid blue 29 adsorption using different adsorbents have been carried out by several researchers, among others: observation of the removal efficiency using activated carbon-clay composite [8], evaluation of the effect of dyes' structure on the efficiency of the adsorption [9] and

investigation of the adsorptive potential of chemically activated olive pomace boiler ash [10]. In this works, acid blue 29 adsorptions onto activated carbon was carried out in a batch system. Two kinetic models were developed. In the first model, the transport of adsorbate to be assumed only occurs from the bulk solution to the particle surface while the second model takes into account the intra-particle diffusion of adsorbate molecules to the active sites through the pore of the particles. The study was focused on getting a suitable kinetic model applied to this adsorption system.

2. Theory and experiment

2.1 Adsorption equilibrium correlation

Adsorption equilibrium is a condition where the solute concentration in the liquid phase at the interface is in equilibrium with the concentration of the solute at the sorbent at the interface. Three mathematical relationships are generally used to represent adsorption equilibrium, i.e: Langmuir (Eq. 1), Freundlich (Eq. 2) and Linear (Eq. 3).

$$\frac{1}{C_\mu} = \frac{1}{C_{\mu,max}} + \frac{1}{C_{\mu,max}k}\frac{1}{C_{eq}} \tag{1}$$

$$C_\mu = K\,C_{ceq}^{1/n} \tag{2}$$

$$C_{eq} = HC_\mu \tag{3}$$

where, C_μ: the amount of adsorbate adsorbed per mass of the adsorbent at equilibrium, $C_{\mu,max}$: the theoretical monolayer saturation capacity, C_{eq}: solute concentration in solution at equilibrium, k, K, n: constant, H: a distribution constant.

2.2 Modeling

In this study, two adsorption models were developed and observed. The first model is called model I, while the second model is called model II. The model I assumes that adsorption only occurs on the surface of the adsorbent and there is no intra-particle concentration gradient. Then model I adsorption mechanism consists of (1) mass transfer of the adsorbate from the solution bulk to the surface of the adsorbent through the film layer on the outer surface of the adsorbent; (2) adsorption of the adsorbate at the surface of the adsorbent.

The first model implies that, in the liquid phase, the rate of decrease in adsorbate concentration is equal to the rate of mass transfer of the solute from the solution to the adsorbent. Meanwhile, the solute mass transfer rate from the solution to the adsorbent will be equal to the rate of accumulation of the adsorbate on the adsorbent. Therefore, the following two equations apply to the first model.

$$\frac{dC_L}{dt} = -\frac{k_c A}{V}\left(C_L - C_L^*\right) \tag{4}$$

$$\frac{dC}{dt} = \frac{k_c A}{m}\left(C_L - C_L^*\right) \tag{5}$$

where, C_L: the solute concentration in liquid phase, k_c : the convective mass transfer coefficient, A: the outer surface area of the adsorbent, C_L^*: the concentration of the adsorbate at the liquid phase interface, V : the volume of solution, t : time, C : the concentration of adsorbate on the adsorbent and m : mass of adsorbent. The initial condition are: at t=0, $C_L = C_{Lo}$, C = 0. The boundary condition is $C_N = H\,C_L^*$, where C_N: concentration of solute at the solid surface.

The second model assumes that the adsorbent is porous media, so that the mass transfer of the adsorbate occurs with three mechanisms, namely: (1) mass transfer of the solute from the solution bulk to the surface of the adsorbent through the film layer of the liquid phase, (2) intraparticle diffusion of solute from the adsorbent surface to the center of the adsorbent through the pore, (3) adsorption of the solute at the active sites in the pore. Assuming the adsorbent particles are spherical and have uniform diffusivity and porosity, the following differential equations apply to the adsorption of the second model.

At the middle of the solid:

$$\left(-De\,4\pi\pi^2\,\frac{\partial C}{\partial r}\bigg|_r\right) - \left(-De\,4\pi\pi(+\Delta r)^2\,\frac{\partial C}{\partial r}\bigg|_{r+\Delta r}\right) = 4\pi\pi^2\Delta r\,\frac{\partial}{\partial t}\left(\varepsilon C + C_\mu \rho_{ads}\right) \tag{6}$$

At the center of the solid:

$$De\,4\pi\pi(\frac{\Delta r}{2})^2\,(\frac{\partial C}{\partial r}) = \frac{4}{3}\pi(\frac{\Delta r}{2})^3(\varepsilon + \frac{\rho_{ads}}{H})(\frac{\partial C}{\partial t}) \tag{7}$$

On the surface of the adsorbent:

$$k_c\,4\pi\pi^{\,2}\,(C_L - C_L*) = De\,4\pi\pi^{\,2}\,(\frac{\partial C_N}{\partial r})_{surface} \tag{8}$$

Where, C: concentration of solute at solid, ρ_{ads}: density of adsorbent, ε: ratio of the solution volume to the total volume, ρ_{ads}: adsorbent density, R: adsorbent radius, r: position in adsorbent in the radial system, D_e: effective diffusivity.

2.3. Experiment

The acid blue 29 dye was purchased from a local chemical supplier. Meanwhile, activated carbon was obtained from Merck Millipore. The activated carbon has a particle size <100 μm and bulk density: 150 - 440 kg/m³. Batch adsorption experiments were carried out using a 250 ml beaker with constant speed stirring using a magnetic bar. The volume of acid blue 29 solutions used in each batch is 100 ml with the initial concentration was varied by 75, 125, 185, 225 and 275 ppm. While the mass of activated carbon used was varied as much as 0.1, 0.1125, 0.125, 0.1375 and 0.15 g. Thus the ratio of the mass of activated carbon to the volume of the solution varies by 0.001, 0.001125, 0.00125, 0.001375 and 0.0015 g/ml. The concentration of acid blue 29 in solution at certain times and at equilibrium was measured using a UV-Vis spectrophotometer - Agilent 8453.

3. Results and discussions

3.1 Equilibrium analysis

In this experiment, to ensure that adsorption equilibrium has been reached, the batch adsorption experiment was carried out overnight. The acid blue 29 concentration in activated carbon and in solution at equilibrium was obtained from the experiment, which was marked as dots, and plot of Langmuir, Freundlich and Linear equilibrium correlations is shown in figure 1. While the values of the parameters of the equilibrium correlation are presented in table 1. The results showed that the Langmuir and Freundlich equilibrium correlations are better suited to the experimental data than the Linear correlation. Linear correlation generally only applies to low concentrations. Based on the value of the coefficient of determination, the correlation of Langmuir is the most suitable.

Since the Langmuir equilibrium correlation is developed based on the assumption that the adsorbent surface can only adsorb one layer of the adsorbate molecules, then this result confirmed that the acid blue 29 molecules absorbed on the surface of activated carbon form a single layer

(monolayer) with an adsorption capacity of 156.25 mg/g. Adsorption of several types of acid dyes with activated carbon followed the Langmuir correlation [11]. The adsorption capacity obtained in this study is higher than that obtained by Marrakchi et al. which is 104.83 mg/g using activated carbon – clay composite [8] and lower than that obtained by Auta et al. which is 453.12 mg/g using waste tea activated carbon [12].

The acid blue 29 contentration in the solution at equilibrium - C_{eq} (ppm)
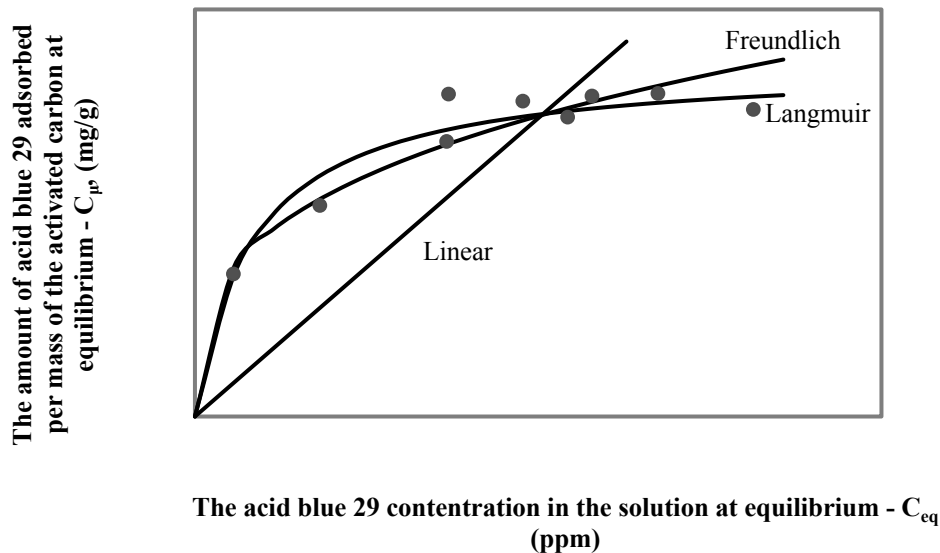
Figure 1. The acid blue 29 adsorptions equilibrium curve on activated carbon

**Table 1.** Equilibrium parameters of acid blue 29 adsorption on the activated carbon.

Equilibrium adsorption model	Parameters		The coefficient of determination (R^2)
Linear	H	0.063 g/L	0.523
Freundlich	K	31.728	0.910
	1/n	0.320	
Langmuir	k	0.067 L/mg	0.956
	$C_{\mu,max}$	156.25 mg/g	

3.2 Suitability of the model with experimental data

To solve the first and the second model, the finite difference method was applied. Then, the numerical computation was run to simultaneously solve a set of the differential equations using the Matlab program. In the computation, the adsorption equilibrium parameters obtained from the experiment were used. The numerical computational results, in which the acid blue 29 concentration change concerning to time during the adsorption process, are presented in figure 2 as a continuous line. Meanwhile, the related real values which were obtained from an experiment taken at several certain times were displayed as discrete data in the form of dots.

The results clearly show that the second model is more suitable than the first model. This finding has confirmed that acid blue 29 adsorptions onto activated carbon occurs with a mechanism that involves intra-particle diffusion through the pores of the activated carbon. Both external mass transfer and intra-particle diffusion play a role in dye adsorption. At the initial stages, the external mass

transfer is more significant, while at the final stages the mass transport is mainly controlled by intra-particle diffusion [13].
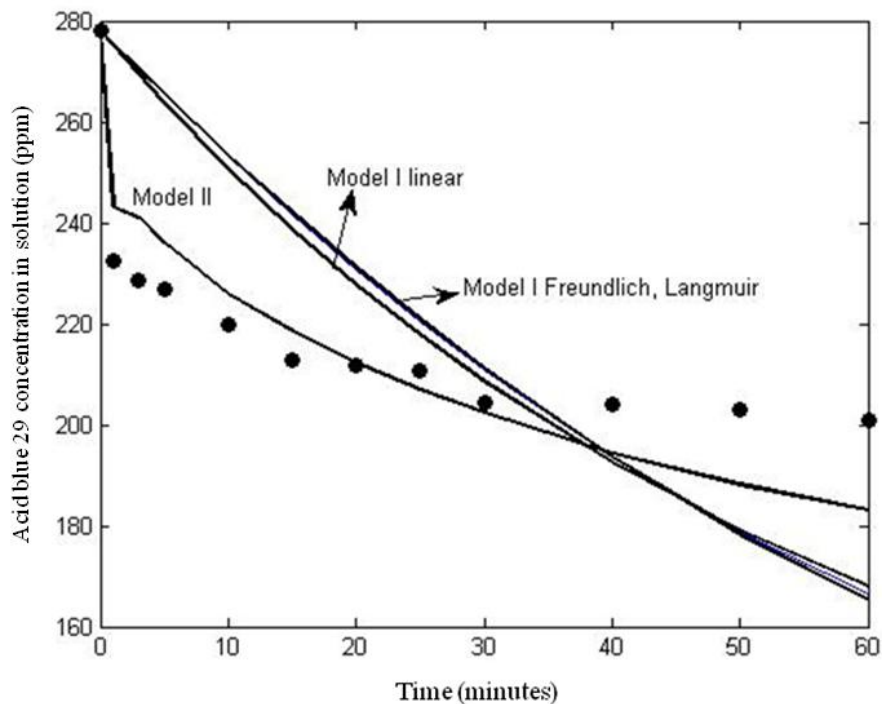
Figure 2. Decreasing the acid blue 29 concentration to the time for the initial concentration of 275 ppm and the ratio of the adsorbent to the solution of 0.001 g/ml.

3.3 The convective coefficient and the effective diffusivity

In observing the suitability of the first kinetic model, the mass transfer coefficient is obtained by solving the set of resulting differential equations. It was found that, even though it hasn't changed significantly, the coefficient decreases with the increasing initial concentration of the dye. For the mass of adsorbent to the volume of solution ratio of 0,001 g/ml, the convective coefficient is $6,40.10^{-2}$ cm/minute when the initial dye concentration is 75 ppm and it is $0,48.10^{-2}$ cm/minute when the initial dye concentration is 275 ppm. A similar trend also occurs in acid dye adsorption kinetics modeling developed by Aksu et al. [14]. Varying the ratio of the mass of adsorbent to the volume solution showed that, even though it doesn't change significantly, the convective coefficient increases with increasing the ratio. For initial dye concentration of 275 ppm, the convective coefficient is $0,48.10^{-2}$ cm/minute when the ratio is 0,001 g/ml and it increases to $0,62.10^{-2}$ cm/minute when the ratio is 0,0015 g/ml. Thus this study shows that the value of the convective coefficient is not much influenced by the concentration of the dye in the solution nor the ratio of the mass of adsorbent to the volume of the solution.

Since the second model takes into account the intra-particle diffusion in the adsorbent pores, the outputs of the second model solution are the convective coefficient of the dye in the liquid phase and the effective diffusivity of the dye in the adsorbent particle. From the computation results, itwas found that the convective coefficient is not affected by the initial concentration of the dye nor the ratio of the mass of adsorbent to the volume of the solution. The average value of the convection coefficient is 0.7248 cm/minute. Meanwhile, the effective diffusivity decreased with increasing acid blue 29 initial concentration, from 1.40×10^{-4} cm/minute at initial dye concentration 75 ppm to 1.64×10^{-6} cm/minute at dye initial concentration 275 ppm at mass adsorbent to solution volume ratio 0,001 g/ml. Besides, it was found that the effective diffusivity was not influenced by the changes of the mass adsorbent to the volume of solution ratio.

Decreasing the effective diffusivity at high dyestuff concentrations was also observed in the adsorption of tectilon red acid dyes on granule activated carbon which might be a result of collisions between dye molecules in the pore which caused additional diffusion barriers. Also, at high concentrations, the possibility of agglomeration of dye molecules that have large molecular sizes can also cause micropore-clogging and inhibit diffusion [15].

3.4 Removal percentage.
In this experiment, the removal percentage of the acid blue 29 is defined as the ratio of the difference between the initial dye concentration in the solution and the concentration at a certain time to the initial concentration. In this study, it was found that the higher the initial concentration of dyes, the smaller the percent removal of the acid blue 29. For radio of mass of adsorbent to solution volume of 0.001 g/ml, the percentage of removal reached 86.6% when the initial dye concentration was 75 ppm and decreased to 48.8% when the initial dye concentration was 275 ppm. It was reported that the removal percentage of dyes by adsorption was in the range of 68% – 99% [16].

4. Conclusions
Acid blue 29 dye adsorption onto activated carbon follows the Langmuir correlation and fits with the second model in which the adsorption rate is controlled by convection mass transfer in the liquid phase as well as the intra-particle molecular diffusion that occurs in the pore. It was found that the convective coefficient is independent to the initial concentration of the dye and the ratio of the mass of adsorbent to the volume of solution. On the other hand, the effective diffusivity depends on the initial concentration. The dye removal capacity increase when the initial dye concentration in the solution decrease and reaches 86.6% when the initial dye concentration is 75 ppm.

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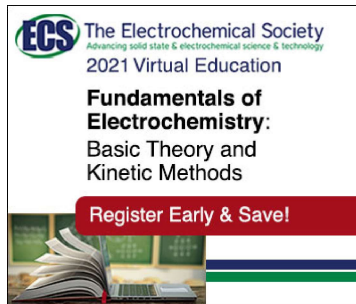
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Fiona McKenzie

Contact Fiona with questions about IOPP's current journals portfolio

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David McDade

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ASSOCIATE DIRECTOR, JOURNALSASSOCIATE DIRECTOR, APAC AND PARTNERSHIPS

Elaine Tham

Contact Elaine if you are a Society interested in working with IOPP in the co-publishing of your journal(s) and/or books

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Contact Liz with questions about the production process for journal, books and conference proceedings.

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ASSOCIATE DIRECTOR, CONTENT AND ENGAGEMENT MARKETING

Claire Webber

Contact Claire if you have questions about IOPP's marketing activities and conference attendance.

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