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## RESEARCH ARTICLE

### DYNAMIC MODELING OF THE TRANSPORT MECHANISM OF MALACHITE GREEN TO ADSORB ON USED BLACK TEA LEAVES

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#### ABSTRACT

Transport mechanism of Malachite Green (MG) to adsorb on used black tea leaves (UBTL) was studied by applying different transport model equations to the kinetic data of adsorption. The effects of particle size of adsorbent and the initial dye concentration on the rate of adsorption were studied in batch process at constant solution pH and temperature to distinguish the reaction-controlled and transport controlled. A significant effect of particle size of the UBTL on the adsorption rate indicated that the adsorption of MG by UBTL follows transport controlled mechanism. Again, the decreasing of the extent of adsorption with an increase in particle size supports the involvement of diffusion phenomena in this transport mechanism. Verification of parabolic diffusion model and film diffusion model suggested that the film resistance is insignificant to the transfer of MG to UBTL but it seems to be diffusion controlled with an increase in initial MG concentration in the solution. The intraparticle diffusion is the rate-determining step in the uptake of MG by the UBTL and small particle size of UBTL is more favorable for intraparticle diffusion controlled mechanism.

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#### INTRODUCTION

Environmental considerations are today imposing great restriction and closer control on the disposal of industrial effluents. These tasks together with an increase in total volume of effluent generated by various industrial processes have led to the development of efficient and adaptable treatment systems to reduce pollution. Adsorption is one of the important processes, and has greatly contributed to the removal of pollutants from industrial effluents. Increasing of textile dyes concentration in the various waste of electroplating, paints, dyes, chrome tanning, paper industries, etc., has been a great concern because of their toxic nature and other adverse effect on receiving waters. Malachite Green (MG) is one of the basic dyes commonly used in dyeing of cotton, silk, paper, bamboo, weed, straw and leather as well as in manufacturing of paints and printing inks, which pollutes the aquatic environment due to their improper disposal of such industries wastewaters in developing countries. MG is suspected of being a carcinogen as well as teratogen and respiratory poison (Mendez *et al.*, 2007). Several studies have been reported about the removal of MG by low-cost bio-sorption process (Dhaneshwar *et al.*, 2007; Mendez *et al.*, 2007; Gupta *et al.*, 2004; Khattri and Singh 1999). Most of these are less effective and their adsorption kinetics as well as mechanism is not well understood. In our recent study, the excellent capability of used black tea leaves (UBTL) for adsorption of MG from aqueous solutions has been pointed out, which is especially interesting for removal of MG from industrial wastewater (Hossain, 2012). Study on the adsorption kinetics in the water treatment is important in providing valuable insights into the reaction pathways and into the mechanism of adsorption reaction, which is

necessary for effective utilization of an adsorption system. Our previous studies showed that the pseudo-second order kinetic equation is successfully applicable to the adsorption of MG on the UBTL, in comparison with simple first order or pseudo-first order one (Hossain, 2012), but not sufficient for the complete description of adsorption mechanism especially in transport process (Ho and McKay, 2000). The aim of the present paper is to study the transport mechanism of MG from aqueous solution onto the UBTL, applying different mass transport models to the concentration decay curves for different initial concentrations, and to elucidate the limiting step in the overall uptake phenomenon.

#### MATERIALS AND METHODS

##### Adsorbent

The fresh tea leaves sample was collected from Dhaka city in Bangladesh. Used black tea leaves (UBTL) were obtained after extracting tea liquor from the fresh ones. The collected fresh tea leaves were boiled with distilled water for 8 h to discard tea liquor from leaves and residue tea leaves were dried at room temperature and then, 105°C in an electric oven. The dried tea leaves were sieved to receive different sizes of particle in the range of 300-355, 355-425 and 425-500  $\mu\text{m}$  and stored in air tight plastic containers. The prepared UBTL was characterized by determining apparent bulk density (0.34  $\text{g}/\text{cm}^3$ ) using specific gravity bottle (Mohan *et al.*, 2001). Elemental analysis using an energy-dispersed X-ray micro-analyzer (JED-2300 Analysis Station, JEOL, Japan) showed that the UBTL contain 65.3 % of C, 34.2 % of O, and 0.1 % of Ca.

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### Adsorbate

Malachite Green (MG) is a basic triphenyl methane dye. Molecular weight of MG (CAS no. 123333-61-9) is 382.93 g. A stock solution of 1000 mg/L was prepared by dissolving MG (Analytical grade) in distilled water from which different concentrated solutions were prepared. Computerized UV-visible spectrophotometer (UV-1650A, Shimadzu, Japan) was used to measure the absorbance of MG solution at predetermined  $\lambda_{\max} = 617$  nm and at optimum pH = 4.0. The solution pH was adjusted at 4.0 by adding either 0.5 (mol/L)  $\text{HNO}_3$  or 0.5 (mol/L) NaOH solution. All chemicals used were of analytical grade.

### Adsorption kinetic experiments

Batch adsorption kinetic experiments were carried out to obtain the role of mass transport in the MG uptake by the UBTL, by suspending 2.5 mg of the UBTL in 25 mL of 100 mg/L MG solution at an optimum pH 2.0 for UBTL (Hossain and Alam 2012) in a series of reagent bottles (60 mL). The suspensions were mixed on a thermostatic water bath shaker (HAAKE SWB20, Fissions Ltd., Germany) at 110 rpm and  $30.0 \pm 0.2^\circ\text{C}$ . The flasks were withdrawn at specified times and the solutions were analyzed at pH 4.0 using a UV-visible spectrophotometer at  $\lambda_{\max}$  617 nm. The amounts of MG adsorbed at time  $t$ ,  $q_t$  (mg/g), was calculated, based on the analysis of the remaining solutions and using the Equation (1)

$$q_t = (C_o - C_t) \times \frac{V}{W} \quad (1)$$

where,  $C_o$  and  $C_t$  are the concentrations of MG solution at initial and contact time  $t$  (mg /L), respectively,  $V$  is the volume (L) of aqueous solution containing MG dye, and  $W$  is the mass of UBTL adsorbent used (g). Similarly, batch adsorption kinetic experiments were performed with different particle sizes of the UBTL for fixed concentration of MG solution, and also for various initial concentrations of MG solution with a fixed particle size of UBTL.

### Theory and Kinetic Models

Adsorption process may be considered as the transport of solute from the bulk phase through the laminar liquid film to the surface, transport and onto the interior of the pores or onto active sites on the adsorbent surface where chemical transformation (chemisorption) takes place (Tse and Shang, 2002). Consequently, the rate of adsorption will depend on the mass transfer of solute into and within the adsorbent particles. Several kinetic models have been developed to analyze the adsorption of metal ions from water. Kinetically adsorption mechanism can be described by reaction-controlled or transport-controlled model (Tse and Shang, 2002; Jansson *et al.*, 1996; Onyango *et al.*, 2003). In numerous studies available in literature reports, first order, pseudo-first order or pseudo-second order kinetics have been applied to describe the reaction-controlled mechanism. Ho's pseudo-second order rate Equation (2) is a well-known one (Ho and McKay, 2000).

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

where,  $q_e$  is the amount adsorbed at equilibrium (mg/g),  $k$  the rate constant (g/(mg.min)) of pseudo-second order adsorption and  $kq_e^2$  ( $= h$ ) is the initial rate constant (mg/(g.min)).

Ho and McKay (1999) reported that if the interaction between the adsorbate and the binding sites is kinetically rate controlling, i.e., the reaction-controlled rate constant, and hence the adsorption rate, will be independent of the adsorbent particle size. On the other hand several studies (Onyango *et al.*, 2003; McKay *et al.*, 1983) have showed that the rate and extent of adsorption are significantly affected by the particle size for the transport-controlled processes. Various studies have been reported (Yiacoumi and Tien, 1995) about the transport-controlled model, in which adsorption is based on the following steps:

**Step 1:** transfer of the solute from the bulk solution through the boundary film to the adsorbent surface, which describes film mass transfer resistance, i.e., film diffusion;

**Step 2:** transfer of the solute from the adsorbent surface to the intra-particle active sites, which are related to the intraparticle diffusion model;

**Step 3:** uptake of the solute on the active sites on the adsorbent surface, via complexation, sorption and intra-particle precipitation, which are related to the pore diffusion or surface diffusion model.

A number of models, including single step of diffusion (external or intraparticle) or combined phenomena (Jansson *et al.*, 1996), have been extensively applied in batch reactors to successfully describe the molecule transport in the adsorbent particles (Wu *et al.*, 2001). Now we use the following kinetic models such as parabolic diffusion model, film diffusion model, and intraparticle diffusion model, etc., to analyze the transport mechanism of MG onto the UBTL.

### Parabolic diffusion model

The parabolic diffusion model may be stated empirically as follows (Sivasubramaniam and Talidudeen, 1972).

$$X = Dt^{1/2} + \text{constant} \quad (3)$$

where,  $X$  is the ratio of MG adsorbed ( $q/q_e$ ) at time  $t$  and equilibrium,  $D$  the overall diffusion rate coefficient and  $t$  the contact time. A plot of fraction adsorbed ( $X$ ) at time  $t$  against  $t^{1/2}$ , are often used to test a diffusion-controlled reaction rate (Sparks and Jardine, 1984).

### Film diffusion model

The film diffusion model proposed by van Lier (van Lier, 1989) is

$$\ln \frac{C_t}{C_o} = - \frac{K_f WS_w}{V_f} t \quad (4)$$

where,  $K_f$  is the mass transfer coefficient for film diffusion,  $S_w$  the specific external surface of adsorbent on weight basis,  $W$  adsorbent dosage, and  $V_f$  the volume of the fluid. A plot of  $\ln(C_t/C_o)$  vs. time should be linear if the adsorption mechanism is controlled by film diffusion (van Lier, 1989).

### Intraparticle diffusion model

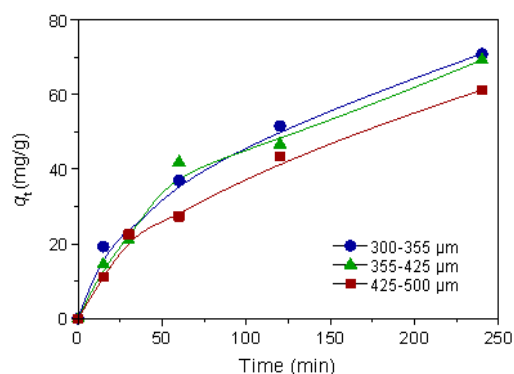
Intraparticle diffusion model proposed by Weber and Morris (Namasivayam and Ranganathan, 1995, Alkan *et al.*, 2007) can be expressed as

$$q_t = k_{id}t^{1/2} \quad (5)$$

where,  $k_{id}$  is the intraparticle diffusion constant, which can be obtained from the slope of the linear portion of the curve of  $q_t$  vs.  $t^{1/2}$  plot.

## RESULTS AND DISCUSSION

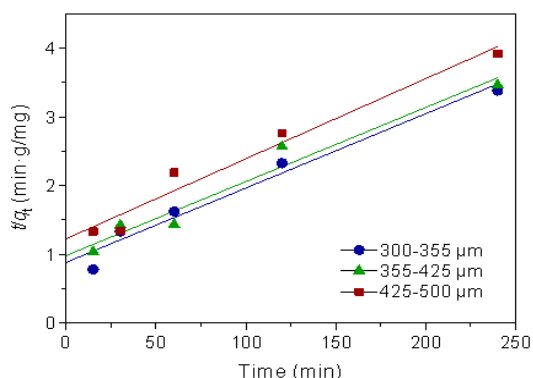
The results obtained through batch experiments for different initial concentrations of MG and particle sizes of the UBTL, were used to elucidate the controlling mechanism of adsorption processes such as mass transfer and chemical reaction, by linear regression with different kinetic models previously described. The conformity between the experimental data and the fit values of the model was expressed by the regression coefficient ( $R^2$ ).



**Figure 1** Effect of particle size on uptake of MG by UBTL at a temperature of  $30.0 \pm 0.2^\circ\text{C}$ ,  $C_0 = 104.35 \text{ mg/L}$ ,  $W_s = 0.0025 \text{ g/L}$  and  $\text{pH} = 2.0$

### Effect of particle size

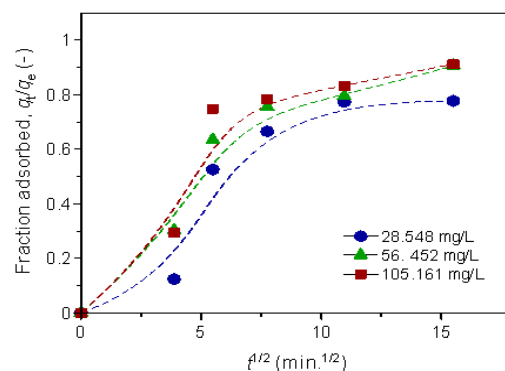
In order to identify the adsorption process as reaction-controlled or transport-controlled one, the effect of adsorbent size on the uptake of MG was studied. Figure 1 shows the uptake vs. time plots for three different sizes of the UBTL. This figure shows that the uptake depends on particle size, suggesting that the MG uptake by the UBTL follows the transport-controlled mechanism (Onyango *et al.*, 2003; McKay *et al.*, 1983). The rate parameters of the process were calculated from the well linear fit of pseudo second order kinetic equation (2) as the plots of  $t/q_t$  against  $t$  (Figure 2), and the kinetic parameters are presented in Table-1. Here the equilibrium amount adsorbed decreased as the particle size increased.



**Figure 2** Effect of particle size on the pseudo second order kinetic equation for the uptake of MG by UBTL at a temperature of  $30.0 \pm 0.2^\circ\text{C}$

The rate and the extent of adsorption, for a constant mass of adsorbent, are proportional to the specific surface area, which is higher for small particle. This is due to the fact that small particles provide more windows through which adsorbate species diffuse to

reach the active sites (Faust and Aly, 1987). The pore diffusion distances to the sites are also shortened. In fact, the mathematical models for external surface diffusion and intraparticle diffusion dictate that the adsorption rate parameters should vary with the reciprocal of the first power of the adsorbent particle diameter and with the reciprocal of some power of the adsorbent particle diameter, respectively (Onyango *et al.*, 2003). In our experimental results, decreasing of the extent of adsorption (equilibrium amount adsorbed) with an increase in particle size supports the involvement of diffusion phenomena in this transport mechanism.

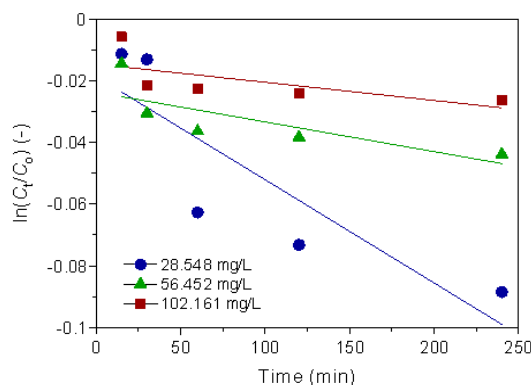


**Figure 3** Effect of initial concentration on the parabolic diffusion model for adsorption of MG on UBTL at a temperature of  $30.0 \pm 0.2^\circ\text{C}$  and  $\text{pH} = 2.0$

### Identification of Diffusion Phenomena

#### Test for parabolic diffusion model

Many researchers used empirical parabolic-diffusion model to describe the metal ion adsorption (Tse and Shang, 2002). The approach is particularly concerned with diffusion mechanisms: intraparticle (surface and/or pore) and film diffusion (Jansson *et al.*, 1996). Figure 3 shows our experimental data as a plot of fractional adsorbed ( $X = q_t/q_e$ ) against square root of time, based on the parabolic diffusion model, Equation (3). The figure shows that the plot became curvature for low concentration of MG but it changed to straight line at high concentration of MG after a short period of time. Thus the adsorption of MG on the UBTL seems to be diffusion-controlled when the concentration of MG is high, but when the concentration becomes lower; other mechanisms would begin to control the adsorption process. Reichenberg proposed that in case of ionic adsorption, at low concentration, rate is controlled by film diffusion and at high concentration by intraparticle diffusion (Reichenberg, 1953). Similarly, at higher initial concentration of MG ( $> 60 \text{ mg/L}$ ), diffusion might be control the adsorption of MG on the UBTL.



**Figure 4** Effect of initial concentration on the film diffusion model for the adsorption of MG on UBTL at a temperature of  $30.0 \pm 0.2^\circ\text{C}$  and  $\text{pH} = 2.0$ .

**Table 1** Effect of particle size on the pseudo-second order rate parameters for the adsorption of MG on UBTL

Adsorbent size (μm)	$q_e$ (mg/g)	$k \times 10^{-3}$ (g/(-mg·min))	$R^2$ (-)
300 – 355	92.593	1.32	0.969
355– 425	92.593	1.19	0.960
425 – 500	85.470	1.12	0.966

**Table 2** A comparison of the experimental data fitness to the pseudo second order kinetic and intra-particle diffusion kinetic equations for different initial concentrations of MG

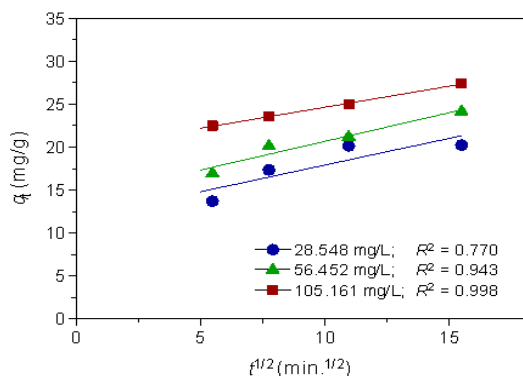
Initial concentration of MG, $C_0$ (mg/L)	Pseudo second order kinetic		Intraparticle diffusion	
	rate constant, $k_2 \times 10^{-3}$ (g/(mg·min))	$R^2$ (-)	constant, $k_{id}$ (mg/(g·min <sup>1/2</sup> ))	$R^2$ (-)
28.548	0.712	0.881	0.942	0.976
56.452	1.485	0.994	0.823	0.986
105.161	1.531	0.989	0.717	0.984

**Table 3** Effect of particle size of the fitness to intraparticle diffusion model

Particle size (μm)	Intraparticle diffusion rate constant, $k_{id}$ (mg/(g·min <sup>1/2</sup> ))	Values of $R^2$ (-)
300-355	4.607	0.993
355-425	4.454	0.970
425-500	4.191	0.990

**Test for film diffusion**

Film diffusion model, Equation (4) was applied to evaluate whether the initial stage of MG adsorption is controlled by the film diffusion. Figure 4 shows that all the data in a plot of  $\ln(C_0/C_t)$  vs. time, are non-linear, i.e., the film diffusion model shows a poor correlation of the experimental data with the low  $R^2$  values (0.75, 0.62 and 0.44 for 28.548, 56.452 and 102.161 mg/L of MG, respectively), suggesting that the film diffusion is less significant for the adsorption of MG on UBTL. By considering the agitation speed higher than 100 rpm used during the kinetic experiments, it is therefore not surprising that the film resistance does not control the uptake of MG.

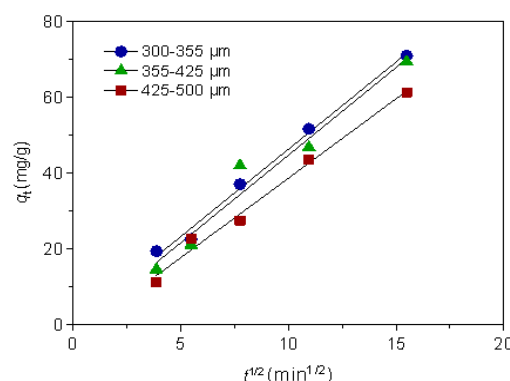


**Figure 5** Effect of initial concentration on the intraparticle diffusion model for the adsorption of MG on UBTL at a temperature of 30.0 ± 0.2°C and pH = 2.0

At the high agitation speed, the shear on the particle surface is large and the thickness of the boundary layer surrounding the particles is thin and the boundary layer resistance or film diffusion would not be a rate-controlling step. Cheung *et al.* observed similar result for the removal of Cd<sup>2+</sup> from effluent, using bone char (Cheung *et al.*, 2001). The first-order kinetic model has been often used to describe the film diffusion-controlled reactions (Tse and Shang, 2002), but our recent study (Hossain, 2012) showed considerable deviation of this adsorption data from linearity indicating that the adsorption MG by the UBTL is not controlled by film-diffusion.

**Test for intraparticle diffusion**

In order to clarify the possibility that intraparticle diffusion of MG on the UBTL is rate-controlling, amount adsorbed ( $q_t$ ) at time  $t$  for different initial concentrations against square root of time ( $t^{1/2}$ ) was investigated, based on Equation (5), as shown in Figure 5.



**Figure 6** Effect of particle size of UBTL on the intraparticle diffusion model for the adsorption of MG on UBTL at a temperature of 30.0 ± 0.2°C and pH = 2.0

Three plots for different initial concentrations have the same general features of linearity but the values of  $R^2$  are increased with the increase of initial concentrations of MG, reveals that the intraparticle diffusion becomes predominant at high concentration of MG for specified amount of the UBTL. A good correlation of the kinetic data with the intraparticle diffusion model suggests that the adsorption rate is mainly controlled by intraparticle diffusion. The value of rate constant for intraparticle diffusion ( $k_{id}$ ) was obtained from the slope of the linear plots of the  $q_t$  vs.  $t^{1/2}$  for each initial concentration and compare with the pseudo second order rate constant as shown in Table 2. The rate constant for the intraparticle diffusion decreases with increasing MG concentration whereas reverse values were observed for pseudo second order rate constant. At the lower concentration, monomeric MG molecules should be more easily transferred and adsorbed onto the surface of the UBTL, compared to polymeric MG molecules at higher concentration. Therefore, the value of intraparticle diffusion constant decreased with increasing the concentration of MG. There has been no report to-date about the adsorption of MG by UBTL, to be compared with the present values of intraparticle rate constant.

**Effect of particle size on intraparticle diffusion**

The effect of particle size of UBTL on the intraparticle diffusion of MG on the UBTL was investigated by performing kinetic experiments with three different particle size of UBTL using same concentration of MG solution. The amount adsorbed ( $q_t$ ) at time  $t$  for different particle size of UBTL against square root of time ( $t^{1/2}$ ) are shown in Figure 6. The intraparticle diffusion constant, calculated from the slope of the linear plot for different particles



size given in Table 3, decreased with increase of particle size of UBTL. Such observation might be due to the existence of large size of cavity on the large size of UBTL particles resultant the retardation of the movement of adsorbed species from the cavity (Hossain, 2003).

## CONCLUSIONS

Verification of different transport models to the kinetic decay of MG to adsorb on UBTL for different initial concentrations of MG and also for different particle sizes of UBTL reveals that the transport mechanism of MG to adsorb on UBTL is dominated by intraparticle-diffusion-controlled. The rate constant of intraparticle diffusion model decreases with increasing MG concentration. The effect of particle size indicated that the small particle size is favorable for intraparticle diffusion controlled.

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