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Isotherm and Kinetic Modelling of Azo Dyes Adsorption

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Abstract

Textile and dye industries' wastewaters are one of the major problem in the water pollution. This wastewater causes serious environmental pollution, because of non-biodegradable and toxic dye molecules. Azo dyes are widely used in the textile industry. In the anaerobic condition azo dyestuff decompose to toxic byproducts. The aim of this work is to understand the adsorption mechanisms of various azo dyestuff adsorbed by domestic wastewater treatment plant inactivated sludge. To determine the adsorption mechanisms, various isotherms and kinetics were used and constants of each isotherms and kinetics were calculated for each dyestuff. In this study, Reactive Black 5 (RB5), Reactive Blue 21 (RB21), Acid Brown 283 (AB283) and Basic Violete 3 (BV3) azo dyestuff adsorption data were used for isotherms and kinetics calculations. The results of this study showed that, the best isotherm which describe the adsorption process was Freundlich. This isotherm model assumes that heterogeneous sorption occurs on adsorbent surface, stated in other words adsorption power varies at every sorption point. For RB5 and AB283 dyes, the best kinetic model which describe the adsorption process was pseudo-second-order kinetic model. This kinetic model assumes that adsorption rate dependent to adsorbent material quantities and contact time.

Keywords

Azo Dyes, Dyestuff Adsorption, Equilibrium Isotherms and Kinetic Models

1. INTRODUCTION

Dye manufacturing is a large industry. There are more than 100,000 commercially available dyes in the world trade market [1].It is estimated that approximatively 1 million tones dyestuff produced annually, and 20-25% of these produced dyestuff is discharged to the receiving environment as waste [2]. Furthermore, Dyes are widely used in many industries, such as textile, packaging industry, automotive industry, food industry, etc. These industries colored wastewaters are caused serious aesthetic and environmental problems. The colored substances prevent the passes of the sun ray into the water. So that, photosynthetic reactions are reduced [3]. Dyes can cause mutagenic and carcinogenic effects on living organisms. At that, dyes can affect brain, central nervous, reproduction system, and organs such as kidney, lung, liver, etc. [4]. The majority of the synthetic dyes are resistant to biological degradation, because of its' complex structures, such as azo dyes [5]. Azo dyes have at least one double bounded nitrogen (N=N), and these dyes are named according to the number of double bounded nitrogen pairs. Azo dyes which have one double bound nitrogen molecules are called monoazo, and azo dyes which have 2 or 3 double bound nitrogen molecules are called diazo or triazo dyes [6].In the anaerobic conditions, azo dyes are degraded to colorless and toxic aromatic amines [7]. Many physical, chemical and biological techniques have been developed for dye removal. Adsorption is one of the most important techniques for dye removal. Many adsorbents have been as scientifically or commercially tested for dye removal. Peat, activated sludge, coir pith, waste organic peel, tree fern, red mud and minerals can bered given as examples for these adsorbents [8,9,10,11,12,13,14,15]. Biological activated sludge systems are one of the most common treatment method for colored wastewaters, particularly textile industries wastewater treatment [16].

One of the dye remove mechanisms that occur in activated sludge system is adsorption. Adsorbable substances can transferred into the cell, and take a part in metabolic / co-metabolic activities. In this reason, studies on adsorption of dyestuff with activated sludge, and understand of the adsorption mechanisms of dyestuff is important for colored wastewater treatment [9]. Isotherms and kinetic models are important to understand the adsorption mechanisms, identify optimum operation conditions, and design effective treatment systems.

The aim of this work is to understand the adsorption mechanisms of various azo dyestuff adsorbed by domestic wastewater treatment plant inactivated sludge.

2. MATERIAL AND METHOD

In this paper, some isotherms and kinetics used to understand Reactive Black 5 (RB5), Reactive Blue 21 (RB21), Acid Brown 283 (AB283) and Basic Violete 3 (BV3) azo dyestuff adsorption mechanisms by domestic wastewater plant inactivated sludge.

Table 1. RB5, RB21, AB283, and BV3 dyes properties

ColorIndex	RB5	RB21	AB283	BV3
Type	Anionic	Anionic	Anionic	Cationic
Chemical Property	Reactive	Reactive	Acidic	Basic
Chromophore Group	Azo	Azo	Azo	Azo
CAS	17095-24-8	12236-86-1	12219-66-8	42555
Molecular Weight	991.82	377.43	882.25	407.99
Molecule Formula	$C_{26}H_{21}N_5Na_4O_{19}S_6$	$C_{18}H_{15}N_7OS$	$C_{32}H_{19}CrN_8O_{11}S.H.Na$	$C_{25}H_{30}ClN_3$
λ_{maks}	579	626	328	590

The adsorption data used in the modeling was taken from a MS. Thesis of the year 2014 [17]. The material and method part can be stated in this MS. Thesis. To sum up, adsorption experiments were performed at room temperature, and the study was conducted at different concentrations of initial dye concentrations.

3. RESULT AND DISCUSSION

3.1. Adsorption Isotherms

Adsorption isotherms demonstrate adsorption phenomena during the adsorption process reaches an equilibrium state [18]. Design parameters can be determined fitting isotherm data to different isotherm models [19]. Adsorption isotherm is show the interaction between solutes and adsorbents. It is important for optimizing the use of adsorbents [18].

Langmuir, Freundlich, Temkin and Dubinin-Radushkevich isotherms were used in this study.

3.1.1. Langmuir Isotherm

Langmuir isotherm model assumes that adsorbent surface has adsorptive points and each point can adsorb one molecule. So that, mono layer occurred on the adsorbent and the layer disperse homogenous [18,20]. Langmuir isotherm equation linearized form is given as:

$$\frac{q_e}{C_e} = K_L q_{max} + K_L q_e \quad (1)$$

where; Ce is the equilibrium concentration of adsorbate in solution (mg/L), qe is the equilibrium solid phase concentration (mg/g), KLis the Langmuir constants. Ce/qe data is plotted against Ce to calculate KLand qmax[21,22].

3.1.2. Freundlich Isotherm

Freundlich isotherm model assumes that heterogeneous sorption occurs on adsorbent surface, stated in other words adsorption power varies at every sorption point [23]. Freundlich isotherm equation line arized form is given as:

$$\log(q_e) = \log K_f + \left(1/n\right) \log C_e \quad (2)$$

where; Ce is the equilibrium concentration of adsorbate in solution (mg/L), qe is the equilibrium solid phase concentration (mg/g), Kfand n are Freundlich constant. Kf (L/mg) represents the quantity of dye adsorbet onto adsorbent for a unit equilibrium concentration. n (unitless) shows that the degree of favourability of adsorption. 1/n value change between 0 and 1. Surface heterogeneity increase as 1/n value gets closer to zero. logqe data is plotted against log Ce to calculate Kf and 1/n [18,22,24].

3.1.3. Temkin Isotherm

Temkin isotherm model is evaluated interactions between adsorbed substances. This isotherm assumed that heat of adsorption decreases linearly with coverage due to adsorbate/adsorbent interactions [18,25].Temkin isotherm equation line arized form is give as:

$$q_e = \frac{RT}{b_T} \ln K_T + \frac{RT}{b_T} \ln C_e \quad (3)$$

where; Ce is the equilibrium concentration of adsorbate in solution (mg/L), qe is the equilibrium solid phase concentration (mg/g), 1/bT corresponds to the adsorption potential of the adsorbent (J/mol), KT is the Temkin isotherm constant (L/g), T is temperature (Kelvin) and R gas constant (8.314 J/mol.K). qe data is plotted against lnCe to calculate KT and bT [26].

3.1.4. Dubinin-Radushkevich Isotherm

Dubinin-Radushkevich isotherm model is based on the micro pore volume filling theory. This isotherm assumes that multilayer adsorption mechanisms [27].Dubinin-Radushkevich isotherm equation line arized form is given as:

$$\ln q_e = \ln q_{max} + B_D \varepsilon^2 \quad (4)$$

where; qe is the equilibrium solid phase concentration (mg/g), qmax is the maximum solid phase concentration (mg/g), BD is free energy adsorption constant. To calculate the equation (4) the initial value of Ɛ must be calculated with the below equation:

$$\varepsilon = RT \ln\left(1 + \frac{1}{C_e}\right) \quad (5)$$

where; Ce is the equilibrium concentration of adsorbate in solution (mg/L),T is temperature (Kelvin) and R gas constant (8.314 J/mol.K). lnqe data is plotted against Ɛ2 to calculate BD and qmax. Equation (6) is applied after the BD value find. In equation (6), E refers to free energy [27]:

$$E = \frac{1}{\sqrt{2B_D}} \quad (6)$$

If the free energy value range between 1-8 kj/mol, Van der Waals forces are effective and physical adsorption is happened. If free energy value greater than 8 kj/mol, chemical adsorption is happened [28].

3.1.5. Evaluation of Isotherms:

Calculated isotherm parameters summarized on the Table 2 for RB5, RB21, AB283 and BV3 dyes. The results on the Table 2 show that, the best isotherm which describe the adsorption process was Freundlich for all dyes, because of the higher regression coefficient (R2) values. This isotherm model assumes that heterogeneous sorption occurs on adsorbent surface, stated in other words adsorption power varies at every sorption point. Moreover, calculated “1/n” values were range between 0.378-0.957 (1/n<1). Its mean that chemical adsorption happened. Chemical adsorption is generally irreversible. For AB283 dye, calculated regression coefficient (R2=0.991) was very high for AB283 dye. It means that the heat of adsorption decreased linearly with coverage due to adsorbate/adsorbent interactions, so we assume that the adsorption process of AB283 by inactivated sludge is an endothermic reaction.

Table 2. Isotherms parameters of RB5, RB21, AB283 and BV3 dyes

Ishotherm	Parameters	RB5	RB21	AB283	BV3
Langmuir	q_{max}	2.956	-0.00295	0.0376	888
	K_L	0.0285	-0.0006	0.0237	0.0063
	R^2	0.715	0.0243	0.875	0.089

Freundlich	1/n	0.378	0.957	0.625	0.930
	K_F	3.01	0.247	3.128	6.045
	R^2	0.964	0.950	0.960	0.989
Tempkin	b_t	3655	213	172	83
	K_T	5.390	0.060	0.248	0.607
	R^2	0.915	0.836	0.991	0.865
Dubinin-Raduskevich	q_{max}	2.03	16.665	33.36	47.375
	B_D	$8*10^{-5}$	$9*10^{-5}$	$2*10^{-5}$	$1*10^{-5}$
	R^2	0.663	0.671	0.904	0.771

3.2. Adsorption Kinetics

Intraparticle diffusion model, Lagergren kinetic model, Pseudo second order kinetic model and Elovich kinetic model were used in this study. BV3 dye reached equilibrium point in 1 minute. So, BV3 dye data could not use in the kinetic models.

3.2.1. Intraparticle Diffusion Model

Intraparticle diffusion model assumes that adsorbate substances in solution enter the pores which are state at the adsorbate, and hold on the surface of the pores [29,30]. Intraparticle diffusion equation is given as:

$$q_t = k_p t^{0.5} \quad (7)$$

where; qt is theamount of material collected on the adsorbent during the t time (mg/g), t is the time (minute), kp is the intraparticle diffusion rate constant. qt data is plotted against t0.5 to calculate kp [31,22].

Intraparticle diffusion model parameters calculated and summarized on the Table 3 for RB5, RB21 and AB283 dyes. High regression coefficient values proved that the intraparticle diffusion theory is valid for all these dyes. It can be assumed, the adsorbent substances which come to the adsorbent pores were entered the pores and kept the pore surface.

Table 3. Intraparticle diffusion model parameters of RB5, RB21 and AB283 dyes

Initial Dye Concentration	Parameters	RB5	RB21	AB283
25 mg/L	R^2	0.994	0.974	0.961
	K_L (mg/g.min$^{0.5}$)	0.323	1.053	2.396
50 mg/L	R^2	0.973	0.968	0.979
	K_L (mg/g.min$^{0.5}$)	0.363	1.413	4.933
75 mg/L	R^2	0.978	0.994	0.958
	K_L (mg/g.min$^{0.5}$)	0.4513	1.834	6.002
100 mg/L	R^2	0.9821	0.995	0.964
	K_L (mg/g.min$^{0.5}$)	0.5327	3.013	7.893
150 mg/L	R^2	0.988	0.991	0.950
	K_L (mg/g.min$^{0.5}$)	0.6828	4.197	9.67

3.2.2. *Lagergren (First Order) Kinetic Model*

Lagergren equation is used to evaluate the relationship between adsorption rate and adsorption capacity [32]. This model generally in compliance with low adsorbent concentration process [33]. Lagergren kinetic model equation is given as:

$$\log \frac{q_e - q_t}{q_e} = -\frac{k_1 t}{2.303} \quad (8)$$

where; qe is the equilibrium solid phase concentration (mg/g), qt is the amount of material collected on the adsorbent during the t time (mg/g), k1 is the Lagergren rate constant adsorption (min-1), t is the time (min) [34, 18].

Table 4. Lagergren kinetic model parameters of RB5, RB21 and AB283 dyes

Initial Dye Concentration	Parameters	RB5	RB21	AB283
25 mg/L	R^2	0.996	0.934	0.962

	K_L (min^{-1})	0.171	0.149	0.150
50 mg/L	R^2	0.997	0.968	0.857
	K_L (min^{-1})	1.170	0.103	0.220
75 mg/L	R^2	0.981	0.981	0.922
	K_L (min^{-1})	0.210	0.0744	0.189
100 mg/L	R^2	0.976	0.996	0.897
	K_L (min^{-1})	0.216	0.076	0.171
150 mg/L	R^2	0.919	0.997	0.893
	K_L (min^{-1})	0.231	0.073	0.157

Lagergren kinetic model parameters calculated and summarized on the Table 4 for RB5, RB21 and AB283 dyes. Calculated regression coefficient values for RB21 were increased in direct proportion to initial dye concentration. However, RB5 and AB283 dyes regression coefficient values were decreased depending on the increase initial dye concentration. So, it can be said that Lagergren kinetic model is available for RB21 dye.

3.2.3. *.Pseudo Second Order Kinetic Model*

This kinetic model assumes that adsorption rate dependent to adsorbent material quantities and contact time [35]. This kinetic model equation linearized form is given as:

$$\frac{1}{q_t} = \frac{1}{q_e} + \left(\frac{1}{k_2 q_e^2}\right)\frac{1}{t} \quad (9)$$

where; qe is the equilibrium solid phase concentration (mg/g), qt is the amount of material collected on the adsorbent during the t time (mg/g), k2 is the pseudo second order rate constant (g/mg.min), t is the time (minute) [36,37].

Pseudo second order kinetic model parameters calculated and summarized on the Table 5 for RB5, RB21 and AB283 dyes. Calculated regression coefficient values for RB5 and AB283 were increased in direct proportion to initial dye concentration. However, RB21 dye regression coefficient values were decreased depending on the increase initial dye concentration. So, it can be said that adsorption rate of RB5 and AB283 dependent to adsorbent material quantities and contact time.

Table 5. Pseudo Second Order kinetic model parameters of RB5, RB21 and AB283 dyes

Initial Dye Concentration	Parameters	RB5	RB21	AB283
25 mg/L	R^2	0.990	0.958	0.922
	K_L (g/mg.min)	0.078	0.085	0.034
50 mg/L	R^2	0.998	0.990	0.909
	K_L (g/mg.min)	0.087	0.020	0.061
75 mg/L	R^2	0.996	0.976	0.964
	K_L (g/mg.min)	0.169	0.009	0.026
100 mg/L	R^2	0.994	0.978	0.964
	K_L (g/mg.min)	0.163	0.005	0.018
150 mg/L	R^2	0.898	0.991	0.971
	K_L (g/mg.min)	0.237	0.002	0.011

3.2.4. Elovich Kinetic Model

Elovich kinetic model has been determined the kinetics of the adsorption and desorption of inorganic substances on solid surface [38]. This kinetic model assumes that the solid (adsorbent) surface is heterogeneous, and adsorption kinetic not affected the low adsorption/desorption interaction [39]. This kinetic model equation linearized form is given as:

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

where; qt is the amount of material collected on the adsorbent during the t time (mg/g), t is the time (minute), α is the adsorption rate constant (mg/g.min). β is a constant related with to expand the activation energy for chemical sorption and the surface coverage (g/mg). qt data is plotted against lnt to calculate α and β [40,41].

Elovich kinetic model parameters calculated and summarized on the Table 5 for RB5, RB21 and AB283 dyes. High regression coefficient values proved that the Elovich kinetic model is valid for all these dyes. It can be assumed that desorption phenomena do not occur, and adsorption rate decrease with the coverage of adsorbent surface.

Table 6. Elovich kinetic model parameters of RB5, RB21 and AB283 dyes

Initial Dye Concentration	Parameters	RB5	RB21	AB283
25 mg/L	R^2	0.979	0.998	0.971
	α (mg/g.min)	0.486	17.121	22.623
	β (g/mg)	2.547	1.442	0.598
50 mg/L	R^2	0.994	0.992	0.992
	α (mg/g.min)	0.663	0.973	89182
	β (g/mg)	2.300	0.579	0.785
75 mg/L	R^2	0.996	0.967	0.988
	α (mg/g.min)	2.180	2.264	948
	β (g/mg)	2.809	0.319	0.397
100 mg/L	R^2	0.997	0.985	0.998
	α (mg/g.min)	3.237	3.460	604
	β (g/mg)	2.557	0.183	0.272
150 mg/L	R^2	0.967	0.989	0.985
	α (mg/g.min)	17.125	4.459	186
	β (g/mg)	2.796	0.119	0.176

4. CONCLUSION

The study was carried out to understand adsorption mechanisms of RB5, RB21, AB283 and BV3 dyes. Four different isotherms and four different kinetics were used and constants of each isotherms and kinetics were calculated for each dyestuff. The best isotherm which describe the adsorption process was Freundlich isotherm. The best kinetic model which has the highest regression coefficient values was Pseudo second order kinetic model for RB5 and AB283 dyes and Lagergren kinetic model for RB21 dye.

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