

JES

JOURNAL OF
ENVIRONMENTAL
SCIENCES

ISSN 1001-0742
CN 11-2629/X

March 1, 2013 Volume 25 Number 3
www.jesc.ac.cn

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Applicable models for multi-component adsorption of dyes: A review

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Received 17 October 2012; revised 26 December 2012; accepted 08 January 2013

Abstract

Adsorption is one of the several techniques that has been successfully used for dyes removal. Since most industrial colored effluents contain several components including dyes, having a strong knowledge about the scope of competitive adsorption process is a powerful key to design an appropriate system. This is mainly because of the complexity brought about by the increasing number of parameters needed for process description which complicates not only the process modeling but also the experimental data collection. A multi-component adsorption model should be based on fundamental soundness, speed, and simplicity of calculation. For such systems, competition will change the adsorbent-adsorbate attractions. Thus, there is major concern to develop an accurate and reliable method to predict dye adsorption behavior in multi-component systems. This article covers topics such as the theory of dyes adsorption in multi-component systems along with applicable models according to the consistent theories presented by researchers.

Key words: adsorption; dye; multi-component; isotherm models

DOI: 10.1016/S1001-0742(12)60194-6

Introduction

Developing countries are faced with serious water pollution because of unavailability of funds for investing in waste water treatment systems. Pollutants generally enter surface water through discharging effluents from a variety of industrial processes such as the petrochemical, food, dye and chemical industries. The presence of toxic pollutants in water sources has stimulated much attention in recent decades because of their potential to involve the environmental problems. Moreover, they lead to undesirable effects in the color, odor and taste of source waters (Attia et al., 2003). Colored dye wastewater is created as a direct result of the production of the dye and also as a consequence of its use in the textile and related industries. There are more than 100,000 tons commercially available dyes with over 700,000 tons of produced annually. It is estimated that 2% of the dyes are discharged in effluent from manufacturing operations, while 10% was discharged from textile and associated industries (Allen et al., 2004).

Among industries, textile factories consume large volumes of water and chemicals for processing of textiles. Wastewater stream from the textile dyeing operation contains unutilized dyes (about 8%–20% of the total pollution

load due to incomplete exhaustion of the dye) and auxiliary chemicals along with a large amount of water (Mukherjee et al., 1999). It is estimated that 10%–20% of dyes in the textile dyeing process will be lost in residual liquors through incomplete exhaustion and washing operations. The rate of loss is approximated to be 1%–10% for pigments, paper and leather dyes. Effluent treatment processes for dyes are currently able to eliminate only half of the dyes lost in wastewater streams. Therefore, hundreds tons daily find their way into the environment, primarily dissolved or suspended in water (Allen et al., 2004).

Dyes are synthetic aromatic compounds which are embodied with various functional groups. Some dyes are reported to cause allergy, dermatitis, skin irritation, cancer, and mutations in humans (Bhatnagar and Jain, 2005). Ignoring the aesthetic problem, the furthestmost environmental concern with dyes is their absorption and reflection of sunlight entering the water, which interferes with the growth of bacteria, limiting it to levels insufficient to biologically degrade impurities in the water. It is evident, therefore, that removal of such colored agents from aqueous effluents is of significant environmental, technical, and commercial importance to many countries of the world on both an environmental basis and for water reuse (Mckay and AL Duri, 1987). Textile wastewaters present

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considerable resistance to biodegradation due to presence of the dyes which have a complex chemical structure and are resistant to light, heat and oxidation agents (Ravikumar et al., 2006). Hence the removal of dyes in economic and effectual manner from textile industry remains as the most imperative problem. Physico-chemical processes are generally applied to treat colored wastewater. These include flocculation, electro-flotation, precipitation, electro-kinetic coagulation, ion exchange, membrane filtration, electro-chemical destruction, irradiation and ozonation. However, all these processes are costly and cannot be utilized by small industries to treat the wide range of wastewater (Indra et al., 2005). Hence, adsorption appears to be a superior means for the treatment of textile industry effluents (Vinod and Anirudhan, 2003). Increasing stringent legislation on the purity of water resources has created a growing interest in the cleansing of water, wastewater and polluted effluents by adsorption processes (Leitao and Serrão, 2005).

Researchers have studied different adsorption processes for wastewater treatment, especially for reducing COD and removing dyes. The major advantages of an adsorption system for water pollution control are less investment in terms of initial cost, simple design and easy operation, less energy intensiveness, no effect by toxic substances and greater removal of organic waste ingredients as compared to the conventional treatment methods (Rathi and Puranik, 1999).

1 Adsorption

Adsorption is an important unit operation in a number of natural and industrial systems such as, fundamental biological studies, separation and purification processes, recovery of chemical compounds and waste treatment processes. It can replace with other separation processes and contribute efficiently for removal of contaminants from aqueous solution (Jena et al., 2004).

Adsorption is a physico-chemical wastewater treatment process in which dissolved molecules are attached to the surface of an adsorbent by physical-chemical forces. **Figure 1** describes the progression of a molecule of an adsorbate from the bulk towards the site of adsorption.

Since adsorption is sometimes reversible, the regeneration of the adsorbent with consequential economy of

operation is also possible. Rather than attempting to remove the dye and degrade it at the same time, adsorption offers the alternative of splitting up these two steps once the dye has been adsorbed onto the substrate. Adsorption, as a treatment process, has fascinated considerable attention in this respect, since a well-designed adsorption system can produce an effluent with virtually no dyestuffs present (Choy et al., 2000).

Recently, adsorption techniques have been considered due to their efficiency in the removal of pollutants that could not be achieved by conventional methods. Adsorption produces a high quality product, and is a process, which is economically feasible. Decolonization may be a result of two mechanisms: adsorption and ion exchange, and is influenced by many physico-chemical factors, such as, dye/adsorbent interaction, adsorbent surface area, particle size, temperature, pH and contact time (Robinson et al., 2001).

It is widely accepted that the process of dye adsorption can be represented by four consecutive steps (Al-Godah, 2000; Baup et al., 2000; Sanghi and Bhattacharya, 2002): (1) diffusion/convection of dye molecules through the bulk of solution; (2) diffusion of dye molecules through a diffusional boundary layer (film diffusion); (3) diffusion of dye molecules from the surface into the interior of the adsorbent materials; (4) adsorption of dye molecules on the surface of the materials through molecular interactions.

The concentration of the dye and agitation may affect step 2. Step 3 is usually considered as the rate determining stage which certainly should affect the adsorption of dyes on the substrate. Step 4 is dependent on the nature of the dye molecules, such as anionic and cationic structures.

It is important to note that step 3 could involve two different phenomena: porous diffusion (the adsorbate first diffuses in the liquid filling the pores and then is adsorbed) and surface diffusion (the adsorbate is first adsorbed then diffuses from one site to another).

Several adsorbents have been studied to determine their ability to adsorb dyes from aqueous effluents (**Table 1**), however, the most widely used dye adsorbent and most readily available from commercial sources is activated carbon (Yeh and Thomas, 1995).

2 Appropriate isotherms for dyes adsorption

Equilibrium data, commonly known as adsorption isotherms, are the basic requirements for the design of adsorption systems. Obtaining equilibrium data for a specific adsorbate-adsorbent system can be performed experimentally, with a time-consuming procedure that is incompatible with the growing needs for adsorption systems design. The physical chemistry involved with the adsorption process may be complex and no single theory of adsorption could adequately present all experimental data satisfactorily. Although the assumptions on which

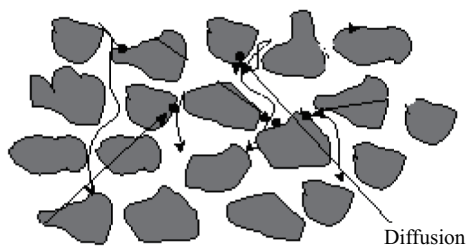


Fig. 1 Adsorption of particles into the porous media (Berefield and Weand, 1982).

Table 1 Some adsorbents used for dyes removal

Adsorbate	Adsorbent
Acid dyes	Cationized sawdust (Baouab et al., 2001)
	Compost sorbent (Tsui et al., 2003)
	Maize waste (Dávila-Jiménez, 2005)
	Orange peel (Sivaraj et al., 2001)
	Peat (Ramakrishna and Viraraghavan, 1996)
	Polysaccharides (Delval et al., 2002)
	Red mud (Namasivayam et al., 2001)
	Shale oil ash (Al-Godah, 2000)
	Bottom ash and de-oiled soya (Gupta et al., 2006)
	Bagass Flyash (Indra et al., 2005)
Cationic dyes	Blast Furnace Sludge (Bhatnagar and Jain, 2005)
	Clinoptilolite and Amberlite (Yener et al., 2006)
	Compost sorbent (Tsui et al., 2003)
	Dried Activated Sludge (Gulnaz et al., 2004)
	Fly Ash (Basava Rao and Mohan Rao, 2006)
	Giant duckweed (Waranusantigul et al., 2003)
	Linseed Cake (Liversidge et al., 1997)
	MaizeWaste (Dávila-Jiménez, 2005)
	Natural Zeolite (Wang et al., 2006; Wang and Ariyanto, 2007)
	Peat (Allen et al., 2004)
Direct dyes	Shale oil ash (Al-Godah, 2000)
	Silk worm pupa (Noroozi et al., 2007)
	Chitosan (Bhavani and Dutta, 1999)
	Coal based sorbent (Mohan et al., 2002)
Disperse dyes	Compost sorbent (Tsui et al., 2003)
	Fly Ash (Basava Rao and Mohan Rao, 2006)
Reactive dyes	Polysaccharides (Delval et al., 2002)
	Chitin (Mckay et al., 1984)
	Eggshell membrane (Koumanova et al., 2002)
	Eucalyptus bark (Morais et al., 1999)
	Maize waste (Dávila-Jiménez, 2005)
	Polysaccharides (Delval et al., 2002)
	Activated carbon (Al-Degs et al., 2007)
	Sugar beet pulp (Aksu and Isoglu, 2007)

they were based were seen in later years to be not entirely valid, some of the oldest theories of adsorption are still the most useful due to their simplicity. However, most industries discharge effluents containing several components; hence adsorption systems design must be based on multi-component effluents, making multi-component equilibrium data a necessity. Compared with single-solute isotherms, multi-component isotherms require a tedious procedure to be obtained experimentally; therefore many attempts were carried out to obtain the latter from single-solute equilibrium data. However, multi-component systems themselves are complicated due to the interaction and competition effects involved. These attempts were either too simplified to describe the complexity of a multi-component isotherm or too mathematically complicated to be used in practice. Many models have been published in the literature to describe the equilibrium relationship between solute and adsorbent in both single and multi-component systems. For single-solute systems, Langmuir and Freundlich isotherms have been the most common in addition to the BET (Brunauer et al., 1938) multi-layer model (Mckay and Al Duri, 1989; Khan et al., 1997).

Competitive interactions between adsorbate have been recognized as a factor that affects adsorption. The magnitude of these competitive interactions needs to be determined in order to better predict and model contaminant migration (Gutierrez and Fuentes, 1993). Multi-component equilibrium studies have passed through many stages since the beginning of this century when none of the multi-component equilibrium studies dealt with competitive adsorption and a few were only concerned with the selectivity of adsorption processes (Mckay and Al Duri, 1989). The complexity of multi-component adsorption is even severe when the adsorbent exhibits a microporous structure. This is mainly because of an inherent adsorption heterogeneity known for this type of adsorbent (Nguyen and Do, 2001).

3 Multi-component adsorption

Several methodologies have been developed. The earliest attempt (Butler and Ockrent, 1930) extended the Langmuir isotherm to account for competitive adsorption and this was later modified to include a separate term for adsorption without competition (Jain and Snoeyink, 1973). McKay and Al-Duri (1989) applied the competitive Langmuir isotherm to basic dyes on carbon. Prediction of the equilibrium adsorption of mixtures from single-solute isotherms is important both in theory of adsorption and for practical applications. Development and improvement of the mixture adsorption theories and models have attracted much attention in the past. The ideal adsorbed solution theory (IAST) of Myers and Prausnitz (1965) is one of typical such theories or models. It can be used to predict mixture adsorption equilibrium from corresponding single-solute isotherms. However, it is not suitable for mixture systems that show non-ideal behaviors. When the energetic heterogeneity of the adsorbent surface and/or interactions between adsorbed molecules is taken into account, improvements can be achieved to some extent. These theories include the real adsorbed solution theory (Krishna, 2001), the vacancy solution theory (Črňáková et al., 2008), the heterogeneous extended Langmuir model and the heterogeneous ideal adsorbed solution theory (Bai et al., 2003). For its simplicity, the extended Langmuir model remains the most widely used isotherm. Among the mentioned multi-component isotherm equations only some of them have been applied to model the dyes systems. **Table 2** summarizes the theories applied for the adsorption of multi-component systems of dyes.

At low concentrations, competitive interactions are not expected to play a significant role in the adsorption process. However, in the case of a highly concentrated accidental release, a full understanding of the competitive interactions among adsorbates would be necessary to predict contaminant migration. The study of competitive interactions also serves the purpose of defining the concen-

Table 2 Multi-component adsorption studies of dyes systems

Multi-component system	Adsorbent	Type of models applied	Reference
C.I. Basic Blue 41 C.I. Basic Red 18 Remazol Reactive Yellow Remazol Reactive Black Remazol Reactive Red	Activated carbon, Silkworm pupa Activated carbon	Langmuir, IAST Langmuir, Redlich- Peterson, Freundlich	Noroozi et al., 2008 Al-Degs et al., 2007
Malachite Green, Pb ions Gemazol Turquoise Blue-G reactive dye, copper(II) ions	Natural zeolite Dried sugar beet pulp	Freundlich, Langmuir Langmuir	Wang and Ariyanto, 2007 Aksu and Isoglu, 2007
Metanil Yellow, C.I. Reactive Blue 15 C.I. Basic Blue 3, C.I. Basic Yellow 21, C.I. Basic Red 22	Cross-linked chitosan Lignite	Langmuir IAST	Chiou and Chuang, 2006 Choy et al., 2005
Atrazine, Congo Red, Methylene Blue	Natural sediment	Freundlich	Tao and Tang, 2004
C.I. Acid Blue 80, C.I. Acid Red 114, C.I. Acid Yellow 117	Activated carbon	IAST, Langmuir	Porter et al., 1999; Choy et al., 2000, 2004a, 2004b
C.I. Basic Blue 3, C.I. Basic Yellow 21, C.I. Basic Red 22 Methylene Blue, Crystal Violet, Rhodamine B	Peat Humic acid	Freundlich, Langmuir, Redlich-Peterson Freundlich	Allen et al., 1988; Allen et al., 2004a Vinod and Anirudhan, 2003
Methylene Blue, Rhodamine B C.I. Reactive Red 4, C.I. Reactive Blue 19, C.I. Reactive Orange 16	Activated carbon Rhizopus arrhizus biomass	Freundlich, Langmuir Isotherm definition	Attia et al., 2003 O'Mahony et al., 2002
Atrazine and Congo Red C.I. Direct Blue 15, C.I. Direct Yellow 12	Activated carbon Cellulose membrane	Freundlich Diffusion	Pelekani and Snoeyink, 2001 O'Maekawa et al., 2000
C.I. Acid Blue 80, C.I. Acid Yellow 117	Activated carbon	IAST	Porter et al., 1999
C.I. Basic Blue 69, C.I. Basic Red 22, C.I. Basic Yellow 21 C.I. Basic Red 46, C.I. Basic Yellow 28 C. I. Acid Red 18, C. I. Acid Yellow 23	Activated carbon Bentonite Bentonite and modified bentonite	IAST, Redlich-Peterson, Langmuir, Freundlich Freundlich, Langmuir Langmuir	Mckay and AL Duri, 1987, 1988, 1989, 1991 Turabik, 2008 Qiao et al., 2009
C. I. Acid Blue 80, C. I. Acid Blue 324, C. I. Acid Green 25, C. I. Acid Green 27, C. I. Acid Orange 7, C. I. Acid Orange 8, C. I. Acid Orange 10, C. I. Acid Red 1	Husk of mango seed	Freundlich, Langmuir	Dávila-Jiménez et al., 2009
C. I. Acid Blue 25, C. I. Acid Yellow 117 C. I. Acid Blue 25,	Bamboo derived active carbon Tyre demineralised activated	Langmuir, Freundlich Redlich-Peterson, Sips Correlative extended Freundlich isotherm	Chan et al., 2009 Chan et al., 2012
C. I. Acid Yellow 117 Malachite Green, Methylene Blue	Rarasaponin-bentonite	extended Langmuir model	Kurniawan et al., 2012
Remazol Brilliant Blue R, C. I. Reactive Orange 16	Polysaccharides composite	Sheindorf-Rehbun-Sheintuch equation, extended Langmuir model	Janaki et al., 2012

trations at which the effect due to competitive interactions starts to be significant in adsorption models (Gutierrez and Fuentes, 1993).

Allen et al. (1988) in their results indicated that the presence of two or more dyes in solution can and will have an effect on the amount of the dyes adsorbed. The amount of any one dye adsorbed was reduced in the presence of a second or third dye. The extent of this effect varied with individual dyes. Factors which could be influencing mixture adsorption will be interactions between dyes in solutions; the effect of each dye in contributing to the surface charge on the adsorbent; competition between the different dyes for the available surface. It is evident from the equilibrium studies that equilibrium adsorption capacities decreased in multi-component systems (as compared to single dye systems). The different dye ions will experience different physical and electrical forces according to their structure, molecular size and functional groups. Hence, competition to take available sites cannot be a factor alone (Allen et al., 1988). Experimental data as shown in Fig. 2 indicate that competitive adsorption for active sites on the carbon surface results in a reduction in the overall uptake capacity of the cationic dyes investigated. In the other study (Turabik, 2008) it has been shown that the equilibrium uptakes of cationic dyes in the binary mixture decreased because of the presence of other dye due to the antagonistic interaction between dyes.

For the adsorption of dyes it is known that molecular characteristics of dyes and physico-chemical properties of adsorbents can affect the adsorption mechanism. It is found that different molecular weight and also the molecular size of dyes play a major role in the physio-sorption system; the smaller molecule will occupy the pores in the structure of adsorbent in advance. However, for the systems where the predominant adsorption mechanism is chemi-sorption the role of molecular size will not play a major role. For example, on comparing the structures of two different dyes, one can find that the availability of the ionic charges may be diverse due to their special positions in the structures

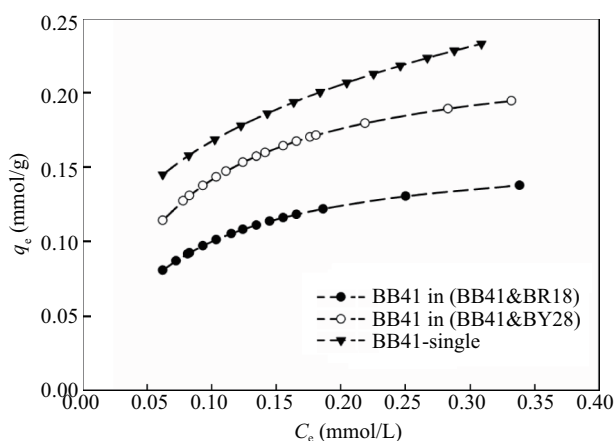


Fig. 2 Equilibrium adsorption isotherms for the adsorption of Basic Blue 41 from single and binary systems onto granular activated carbon.

affecting the adsorption efficiency. Furthermore the dye molecule could provide conditions for a chemical reaction between the adsorbent and adsorbate. As a result of fact, the chemical properties and functional groups of dye and adsorbent can alter the process of the adsorption and play the main role in adsorption of dyes (Noroozi et al., 2008).

4 Multi-component adsorption models

To develop a mathematical model that describes the multi-component adsorption dynamics, following information are generally required: (1) a complete description of equilibrium behavior, i.e. the maximum level of adsorption attained in a adsorbent-adsorbate system as a function of the adsorbate liquid phase concentration; (2) a mathematical representation of associated rate of adsorption, which is controlled by the resistances within the adsorbent particles (Jena et al., 2004). For multi-component systems experimental equilibrium data are difficult to obtain. Frequently the practical way of estimating multi-component adsorption equilibria is to predict mixture isotherms from isotherm data of the single-solutes. It was observed, however, that methods based solely on single solute equilibrium data often fail to calculate experimental mixture equilibria accurately. Several equations with additional empirical parameters which have to be determined from mixture experiments have been proposed (Seidel and Gelbin, 1988).

A model for competitive sorption based on the Langmuir equation was first developed by Butler and Ockrent (1930) to describe the sorption equilibrium in multi-component systems. This isotherm (extended Langmuir (EL)) is applicable when each component obeys Langmuir behavior in a single-solute system. It is widely used to calculate the Langmuir constant (Q_m), the amount of solute adsorbed per unit weight of adsorbent, in the multi-component systems. The common form for depicting the distribution is to correlate the amount of solute adsorbed per unit weight of adsorbent with the residual solute concentration remaining in an equilibrium state (Ho and McKay, 1998). This model assumed a homogeneous surface with respect to the energy of adsorption, no interaction between adsorbed species and that all adsorption sites are equally available to all adsorbed species. As this is rarely the case, this model had limited validity and was improved and modified (JSME) by Jain and Snoeyink (1973) who assumed that a portion of adsorption takes place without competition because not all adsorption sites are available to all species. This was attributed to the chemical nature of sites and adsorbates or the relatively large molecular size of the latter. The number of sites with noncompetitive adsorption is given by the difference between the maximum loadings of the species. Representing the Langmuir model as Eq. (1), the EL equation is shown in Eq. (2) and JSME equation given by Eqs. (3) and (4).

$$q_e = \frac{K_L^0 C_e}{1 + a_L^0 C_e} \quad (1)$$

where, C_e (mmol/L) is the concentration of adsorbate at equilibrium, q_e (mmol/g) is amount of solute adsorbed at equilibrium, K_L^0 (L/g) and a_L^0 (L/mmol) are constants.

$$q_{e,i} = \frac{K_{L,i}^0 C_{e,i}}{1 + \sum a_{L,i}^0 C_{e,i}} \quad (2)$$

where, K_L and a_L are the Langmuir isotherm constants obtained from single solute system.

$$q_{e,1} = \frac{(Q_{m,1}^0 - Q_{m,2}^0) a_{L,1}^0 C_{e,1}}{1 + a_{L,1}^0 C_{e,1}} + \frac{Q_{m,2}^0 a_{L,1}^0 C_{e,1}}{1 + a_{L,1}^0 C_{e,1} + a_{L,2}^0 C_{e,2}} \quad (3)$$

$$q_{e,2} = \frac{Q_{m,2}^0 a_{L,2}^0 C_{e,2}}{1 + a_{L,1}^0 C_{e,1} + a_{L,2}^0 C_{e,2}} \quad (4)$$

where, Q_m^0 (mmol/g) is the monolayer saturation capacity for the single solute Langmuir isotherm.

JSME model was valid for bisolute systems with wide molecular size difference or chemical properties with respect to the adsorbent. However, for more than two components this model is inapplicable. **Figures 3 and 4** confirm inconsistency of the EL and JSME equations on the experimental data for certain dyes removal by activated carbon.

The P -factor method is another correlative technique that has been developed (McKay and Al Duri, 1987) and applied to dye/carbon systems. It was found that if the Langmuir equation correlated all single-solute and multi-component data over the entire concentration range consequently a simple method could be used to correlate single-solute and multi-component isotherms using the

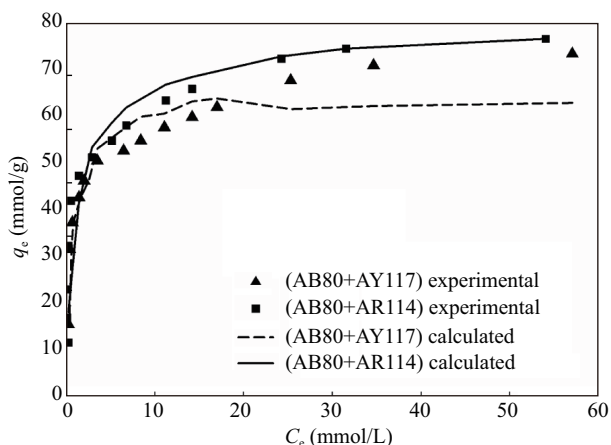


Fig. 3 Extended Langmuir analysis for Acid Blue 80 in the binary system (McKay et al., 2000).

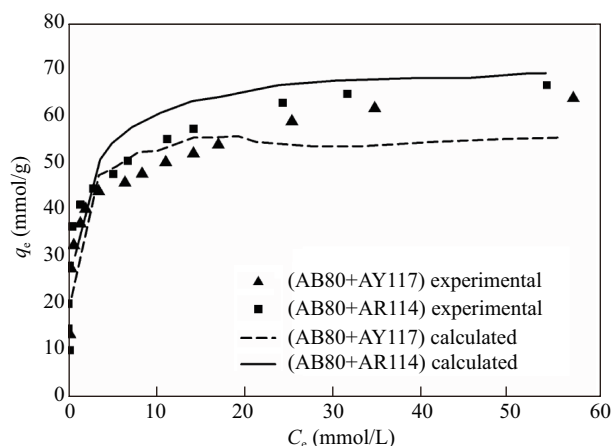


Fig. 4 Jain and Snoeyink modified extended (JAME) Langmuir analysis for Acid Blue 80 in the binary system (Choy et al., 2000).

Langmuir constants (K_L) and a_L (**Fig. 5**). The constant (K_L) represents the ratio of kinetic parameters and a_L is linked to the thermodynamic properties through the enthalpy of adsorption, therefore they decided to use the ratio K_L/a_L for their model. Furthermore, since the ratio K_L/a_L is a capacity factor, the single-solute solid-phase concentration q_e is correlated to the q_e values in the mixtures. The capacity factor, (P_i), has been made dimensionless by expressing it in the form of a ratio:

$$P_i = \frac{(K_{L,i}/a_{L,i})_{\text{single-solute}}}{(K_{L,i}/a_{L,i})_{\text{multi-solute}}} \quad (5)$$

where, $(K_{L,i}/a_{L,i})$ is the adsorbent monolayer capacity for component (i) in a multicomponent system while $(K_{L,i}/a_{L,i})$ is that in the single-solute system. This model assumes a Langmuir isotherm; hence, for each component (i) the multicomponent isotherm equation can be defined as:

$$q_{e,i,\text{multi}} = \frac{1}{P_i} \times \frac{K_{L,i}^0 C_{e,i,\text{multi}}}{1 + a_{L,i}^0 C_{e,i,\text{multi}}} \quad (6)$$

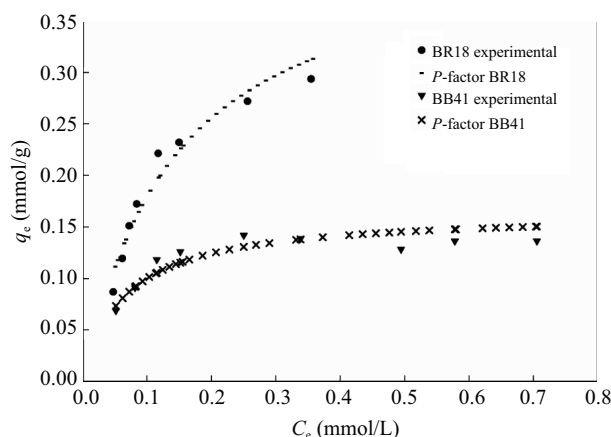


Fig. 5 P -factor analysis plots for a binary system of Basic Blue 41 and Basic Red 18 adsorption onto granular activated carbon.

The limitation of the P -factor model is its failure to predict the multi-component isotherm based on the single-solute data, i.e. it requires some experimental data in the multi-component system.

In general, Langmuir models are too simplified for the complexity of multi-component systems. This led Myers and Prausnitz (1965) to adopt the spreading pressure principle and apply it for gaseous mixtures, followed by Radke and Prausnitz (1972) who applied it for dilute liquid mixtures. They proposed that when multi-solutes adsorb simultaneously at the same temperature and spreading pressure as each species would, the adsorbed phase forms an ideal solution. This is called the ideal adsorbed solution theory (IAST). Radke and Prausnitz (1972) have used IAST theory on the bisolute adsorption data for dilute aqueous solutions on activated carbon. They reported an excellent agreement for the volatile organic solutes, acetone and propionitrile; for strongly adsorbing solutes, such as *p*-cresol and *p*-chlorophenol, the predicted results showed deviation from experimentally measured values at high concentration ratios because of non-idealities in the adsorbing phase (Khan et al., 1997).

The IAST is regarded as a model with the most thermodynamically accepted foundation. The IAST provides a thermodynamic consistent and practical method for predicting binary and ternary adsorption isotherms using single-solute isotherm data alone. Prediction of dyes adsorption by IAST using Freundlich, Langmuir and Myers equation has been reported by different researchers (McKay and Al Duri, 1988; Choy et al., 2004b, 2005; Noroozi et al., 2008). The basis of the theory has been demonstrated elsewhere (Lu and Sorial, 2004).

The IAST is based on the assumption that the adsorbed mixture forms an ideal solution at a constant spreading pressure, as shown in Eq. (7).

$$C_{e,i} = \frac{C_i^0(\pi_m, T)q_{e,i}}{\sum_{i=1}^{i=N} q_{e,i}} \quad (7)$$

where, $C_i^0(\pi_m, T)$ is equilibrium liquid-phase concentration of pure solute i at the same temperature T and spreading pressure π_m of the mixture with N components. Considering the two-dimensional adsorbed phase as an ideal solution leads to the Eq. (8) that relates the mixture equilibrium solid-phase concentration $q_{e,i}$ to the single-solute equilibrium concentrations q_i^0 and C_i^0 of solute i :

$$\sum_{i=1}^{i=N} \frac{q_{e,i}}{q_i^0} = \sum_{i=1}^{i=N} \frac{C_{e,i}}{C_i^0} = 1 \quad (8)$$

To utilize Eqs. (7) and (8) for predicting mixture adsorption equilibria, a further relation is needed between the spreading pressure and directly measurable quantities. This relation is given by the Gibbs adsorption equation, which

may be written in the following integral form:

$$\frac{A_s \pi_i}{RT} = \int_0^{q_i^0} \frac{q_i^0}{C_i^0} \frac{dC_i^0}{dq_i^0} dq_i^0 \quad (9)$$

where, A_s is external surface area per unit mass of adsorbent; and R is the gas constant. Integration of Eq. (9) may be done analytically through the use of various isotherms fitted to single-solute data. Although many isotherm equations are commonly used for modeling adsorption isotherms, however following Henry's law equation at low coverage is very important when predicting multi-component adsorption systems by the IAST (Eq. 7) because of the integration limits.

Figure 6 shows the comparison between the experimental data (McKay et al., 2004b) and the predictions based on the IAST model with different isotherm equations for the binary system of acid dyes. Visually, the IAST-Freundlich (IAST-F) model (based on Eq. (10) (Freundlich, 1906)) provides the worst fit for acidic dye systems. The IAST models based on the other three isotherms, Langmuir (Eq. (1)), Sips (Eq. (11)) (Choy et al., 2004b), and Redlich-Peterson (Eq. (12)) (Koumanova et al., 2002) isotherms produce a reasonable fit for the dye to the experimental data at certain equilibrium solute concentrations.

$$q_e = k_f C_e^n \quad (10)$$

where, q_e (mmol/g) is amount of adsorbate at equilibrium, C_e (mmol/L) is the adsorbate concentration in the solution at equilibrium and K_f and n are constants.

$$q_e = \frac{k_s C_e^{1/b_s}}{1 + a_s C_e^{1/b_s}} \quad (11)$$

where, q_e (mmol/g) is amount of adsorbate at equilibrium, C_e (mmol/L) is the adsorbate concentration in the solution at equilibrium and k_s is Sips constant related to energy

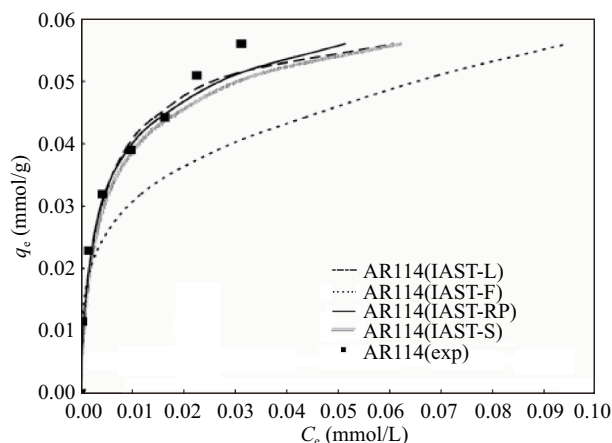


Fig. 6 IAST model analysis of Acid Red 114 in the Acid Blue 80 + Acid Red 114 binary system (Choy et al., 2004b).

of adsorption, and parameter b_S could be regarded as the parameter characterizing the system heterogeneity.

$$q_e = \frac{k_R C_e}{1 + a_R C_e^b} \quad (12)$$

where, q_e (mmol/g) is amount of solute adsorbed at equilibrium, k_R is the isotherm constant, C_e (mmol/L) is the equilibrium concentration of adsorbate at equilibrium and a and b are constant.

The effectiveness of the IAST based on Myers equation in predicting the binary adsorption of cationic dyes on activated carbon has been reported (Noroozi et al., 2008). **Figure 7** reveals that IAST provides desirable prediction for the binary adsorption system on granular activated carbon.

It should be noted that IAST can be applied only to the adsorption systems where the predominant adsorption mechanism is physi-sorption. This subject has been revealed previously (Lu and Sorial, 2004; Noroozi et al., 2008). Obtained data in **Fig. 8** for chemi-sorption of cationic dyes on silkworm pupa show that binary adsorption could not be predicted by IAST since the adsorption of the dye by silkworm pupa was suspected to occur by chemi-sorption.

The chemical nature of dyes is such that they are likely to undergo chemisorption depending on the adsorbent. Chemisorption differs from physisorption in the kind of bonding between the adsorbent and adsorbate. In the former these could be ionic or covalent bonds, in the latter they are Van der Waals. This makes the energy of adsorption much different, i.e. high bonding energy in the case of chemisorption. In chemisorption processes dye ions will possess a certain activity coefficient which could affect the chemical potential in the IAST thermodynamic analysis. However in a very dilute solutions the activity coefficients will be tending to unity (Mckay and Al Duri, 1988).

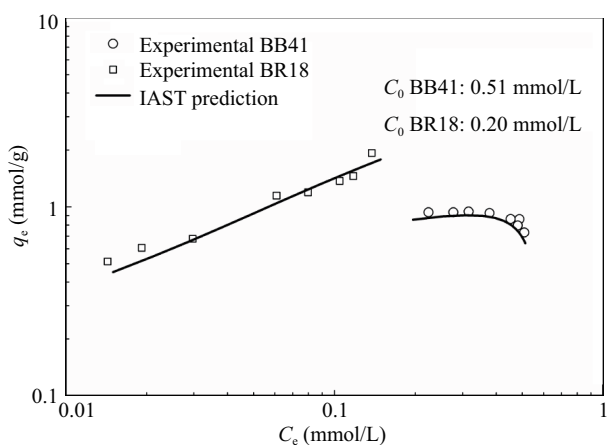


Fig. 7 Prediction of binary adsorption of Basic Blue 41 and Basic Red 18 on granular activated carbon using IAST based on Myers equation (Noroozi et al., 2008).

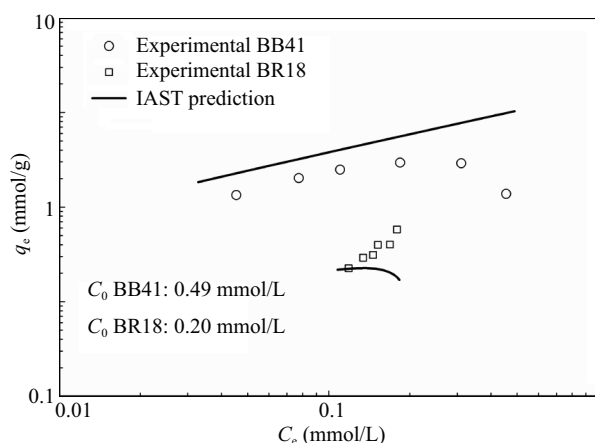


Fig. 8 Prediction of binary adsorption of Basic Blue 41 and Basic Red 18 on SWP using IAST based on Myers equation (Noroozi et al., 2008).

5 Conclusions

Adsorption produces a high quality product and is a process, which is economically feasible but should be controlled by influencing physio-chemical factors. Equilibrium dye adsorption isotherms play an important role in predictive modeling for analysis and design of the multi-component adsorption systems. The interactions and competitions between different adsorbates to reach the adsorbent, complicate the process in multi-component systems. The model with the most thermodynamically accepted foundation for physical adsorption of dyes in multi-component systems is IAST. Application of the IAST is not limited to any specific single-component isotherm model and most common adsorption isotherms can incorporate in IAST in diverse ways to simplify the solution for determining the key parameters. Any chemisorptions occurring because of chemical nature of dyes leads to complicated adsorption behavior and no models have been yet proposed to predict such multi-component systems. According to the obtained results and also the experience of the current article authors it is recommended to apply the proper founded model to the real adsorption systems containing dyes. Since, dyeing is conducted by using diverse auxiliaries, it is important to explore the effect of other chemicals on the equilibrium and kinetic adsorption of dyes in single and multicomponent systems. Furthermore, knowing the impact of different chemical dye class on the efficiency of adsorption in multicomponent solution will lead to a significant practical way to introduce the most appropriate adsorption models to describe the real conditions during adsorption process.

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Journal of Environmental Sciences (Established in 1989)

Vol. 25 No. 3 2013

Supervised by	Chinese Academy of Sciences	Published by	Science Press, Beijing, China
Sponsored by	Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences		Elsevier Limited, The Netherlands
Edited by	Editorial Office of Journal of Environmental Sciences P. O. Box 2871, Beijing 100085, China Tel: 86-10-62920553; http://www.jesc.ac.cn E-mail: jesc@263.net , jesc@rcees.ac.cn	Distributed by	Domestic Science Press, 16 Donghuangchenggen North Street, Beijing 100717, China Local Post Offices through China
Editor-in-chief	Hongxiao Tang	Foreign	Elsevier Limited http://www.elsevier.com/locate/jes
CN 11-2629/X	Domestic postcode: 2-580	Printed by	Beijing Beilin Printing House, 100083, China
		Domestic price per issue	RMB ¥ 110.00

ISSN 1001-0742



www.jesc.ac.cn