



Review:

Critical review in adsorption kinetic models^{*}

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Abstract: Adsorption is one of the most widely applied techniques for environmental remediation. Its kinetics are of great significance to evaluate the performance of a given adsorbent and gain insight into the underlying mechanisms. There are lots of references available concerning adsorption kinetics, and several mathematic models have been developed to describe adsorption reaction and diffusion processes. However, these models were frequently employed to fit the kinetic data in an unsuitable or improper manner. This is mainly because the boundary conditions of the associated models were, to a considerable extent, ignored for data modeling. Here we reviewed several widely-used adsorption kinetic models and paid more attention to their boundary conditions. We believe that the review is of certain significance and improvement for adsorption kinetic modeling.

Key words: Adsorption kinetics, Reaction models, Diffusion models, Film diffusion, Intraparticle diffusion

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INTRODUCTION

Adsorption is one of the most widely applied techniques for pollutant removal from contaminated medias. The common adsorbents include activated carbon, molecular sieves, polymeric adsorbents, and some other low-cost materials. When adsorption is concerned, thermodynamic and kinetic aspects should be involved to know more details about its performance and mechanisms. Except for adsorption capacity, kinetic performance of a given adsorbent is also of great significance for the pilot application. From the kinetic analysis, the solute uptake rate, which determines the residence time required for completion of adsorption reaction, may be established. Also, one can know the scale of an adsorption apparatus based on the kinetic information. Generally speaking, adsorption kinetics is the base to determine the performance of fixed-bed or any other flow-through systems.

In the past decades, several mathematical models have been proposed to describe adsorption data, which can generally be classified as adsorption reaction models and adsorption diffusion models. Both models are applied to describe the kinetic process of adsorption; however, they are quite different in nature. Adsorption diffusion models are always constructed on the basis of three consecutive steps (Lazaridis and Asouhidou, 2003): (1) diffusion across the liquid film surrounding the adsorbent particles, i.e., external diffusion or film diffusion; (2) diffusion in the liquid contained in the pores and/or along the pore walls, which is so-called internal diffusion or intra-particle diffusion; and (3) adsorption and desorption between the adsorbate and active sites, i.e., mass action. However, adsorption reaction models originating from chemical reaction kinetics are based on the whole process of adsorption without considering these steps mentioned above.

At present, adsorption reaction models have been widely developed or employed to describe the kinetic process of adsorption (Banat *et al.*, 2003; Sun and Yang, 2003; Aksu and Kabasakal, 2004; Hamadi *et al.*, 2004; Jain *et al.*, 2004; Min *et al.*, 2004; Shin *et*

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al., 2004; Namasivayam and Kavitha, 2005; Chen et al., 2008; Cheng et al., 2008; Hameed, 2008; Huang et al., 2008; Wan Ngah and Hanafiah, 2008; Rosa et al., 2008; Tan et al., 2008); however, there still exist some problems. For example, pseudo-second-order rate equation based on chemical adsorption was unsuitably employed to describe organic pollutants adsorption onto several non-polar polymeric adsorbents. This is essentially a process of physical adsorption (Meng, 2005). In addition, Lagergren (1898)'s models were still widely applied to data modeling, though no adsorption mechanisms could be reasonably available. Consequently, some famous journals like *Journal of Hazardous Materials and Separation and Purification Technology* began to decline adsorption manuscripts based on unsuitable or simple kinetic modeling as mentioned above (Tien, 2007; 2008).

Here we reviewed several widely-used batch kinetic models and paid more attention to their boundary conditions. We believe the subject review is of considerable significance to improve the current research on adsorption kinetic modeling.

ADSORPTION REACTION MODELS

Pseudo-first-order rate equation

Lagergren (1898) presented a first-order rate equation to describe the kinetic process of liquid-solid phase adsorption of oxalic acid and malonic acid onto charcoal, which is believed to be the earliest model pertaining to the adsorption rate based on the adsorption capacity. It can be presented as follows:

$$\frac{dq_t}{dt} = k_{p1}(q_e - q_t), \quad (1)$$

where q_e and q_t (mg/g) are the adsorption capacities at equilibrium and time t (min), respectively. k_{p1} (min^{-1}) is the pseudo-first-order rate constant for the kinetic model. Integrating Eq.(1) with the boundary conditions of $q_t=0$ at $t=0$ and $q_t=q_t$ at $t=t$, yields (Ho, 2004)

$$\ln\left(\frac{q_e}{q_e - q_t}\right) = k_{p1}t, \quad (2)$$

which can be rearranged to:

$$\log(q_e - q_t) = \log q_e - \frac{k_{p1}}{2.303}t. \quad (3)$$

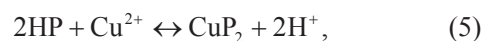
To distinguish kinetic equations based on adsorption capacity from solution concentration, Lagergren's first order rate equation has been called pseudo-first-order (Ho and McKay, 1998a). In recent years, it has been widely used to describe the adsorption of pollutants from wastewater in different fields, such as the adsorption of methylene blue from aqueous solution by broad bean peels and the removal of malachite green from aqueous solutions using oil palm trunk fibre (Hameed and El-Khaiary, 2008a; 2008b; Tan et al., 2008).

Pseudo-second-order rate equation

In 1995, Ho described a kinetic process of the adsorption of divalent metal ions onto peat (Ho and McKay, 1998b), in which the chemical bonding among divalent metal ions and polar functional groups on peat, such as aldehydes, ketones, acids, and phenolics are responsible for the cation-exchange capacity of the peat. Therefore, the peat-metal reaction may be presented as shown in Eqs.(4) and (5), which can be dominant in the adsorption of Cu^{2+} ions onto peat (Coleman et al., 1956):



and



where P^- and HP are active sites on the peat surface.

The main assumptions for the above two equations were that the adsorption may be second-order, and the rate limiting step may be chemical adsorption involving valent forces through sharing or the exchange of electrons between the peat and divalent metal ions. In addition, the adsorption follows the Langmuir equation (Ho and McKay, 2000).

The rate of adsorption described by Eqs.(4) and (5) is dependent upon the amount of divalent metal ions on the surface of peat at time t and that adsorbed at equilibrium. Therefore the rate expression may be given as

$$\frac{d(\text{P})_t}{dt} = k_{p2}[(\text{P})_0 - (\text{P})_t]^2, \quad (6)$$

or

$$\frac{d(\text{HP})_t}{dt} = k_{p2}[(\text{HP})_0 - (\text{HP})_t]^2, \quad (7)$$

where $(P)_0$ and $(\text{HP})_0$ denote the amount of equilibrium sites available on the peat, $(P)_t$ and $(\text{HP})_t$ denote the amount of active sites occupied on the peat at time t , and k_{p2} (g/(mg·min)) is the pseudo-second-order rate constant of adsorption (Ho and McKay, 1998b).

The driving force, $(q_e - q_t)$, is proportional to the available fraction of active sites (Ho, 2006). Then, it yields

$$\frac{dq_t}{dt} = k_{p2}(q_e - q_t)^2. \quad (8)$$

Eq.(8) can be rearranged as follows:

$$\frac{dq_t}{(q_e - q_t)^2} = k_{p2} dt. \quad (9)$$

Integrating Eq.(9) with the boundary conditions of $q_t=0$ at $t=0$ and $q_t=q_t$ at $t=t$, yields

$$\frac{1}{(q_e - q_t)} = \frac{1}{q_e} + k_{p2}t, \quad (10)$$

which can be rearranged as follows:

$$\frac{t}{q_t} = \frac{1}{V_0} + \frac{1}{q_e}t, \quad (11)$$

and

$$V_0 = k_{p2}q_e^2, \quad (12)$$

where V_0 (mg/(g·min)) means the initial adsorption rate, and the constants can be determined experimentally by plotting of t/q_t against t .

Similarly, Ho's second-order rate equation has been called pseudo-second-order rate equation to distinguish kinetic equations based on adsorption capacity from concentration of solution (Ho, 2006). This equation has been successfully applied to the adsorption of metal ions, dyes, herbicides, oils, and organic substances from aqueous solutions (Yan and Viraraghavan, 2003; Al-Asheh *et al.*, 2004; Petroni *et al.*, 2004; Pan *et al.*, 2007; Anirudhan and Rad-

hakrishnan, 2008; Cheng *et al.*, 2008; Hameed, 2008; Hameed *et al.*, 2008).

Elovich's equation

A kinetic equation of chemisorption was established by Zeldowitsch (1934) and was used to describe the rate of adsorption of carbon monoxide on manganese dioxide that decreases exponentially with an increase in the amount of gas adsorbed (Ho, 2006), which is the so-called Elovich equation as follows (Low, 1960):

$$\frac{dq}{dt} = ae^{-\alpha q}, \quad (13)$$

where q represents the amount of gas adsorbed at time t , a the desorption constant, and α the initial adsorption rate (Ho and McKay, 1998a). Eq.(13) can be rearranged to a linear form:

$$q = \left(\frac{2.3}{\alpha}\right) \log(t + t_0) - \left(\frac{2.3}{\alpha}\right) \log t_0, \quad (14)$$

with

$$t_0 = \frac{1}{\alpha a}. \quad (15)$$

The plot of q versus $\log(t+t_0)$ should yield a straight line with an apposite value of t_0 . Elovich equation is applied to determine the kinetics of chemisorption of gases onto heterogeneous solids (Rudzinski and Panczyk, 2000).

With the assumption of $\alpha at \gg 1$ (Chien and Clayton, 1980), Eq.(14) was integrated by using the boundary conditions of $q=0$ at $t=0$ and $q=q$ at $t=t$ to yield (Ho, 2006):

$$q = \alpha \ln(\alpha a) + \alpha \ln t. \quad (16)$$

Elovich's equation has been widely used to describe the adsorption of gas onto solid systems (Rudzinski and Panczyk, 2000; Heimberg *et al.*, 2001). Recently it has also been applied to describe the adsorption process of pollutants from aqueous solutions, such as cadmium removal from effluents using bone char (Cheung *et al.*, 2001), and Cr(VI) and Cu(II) adsorption by chitin, chitosan, and *Rhizopus arrhizus* (Sağ and Aktay, 2002).

Second-order rate equation

The typical second-order rate equation in solution systems is (Xu *et al.*, 2006)

$$\frac{dC_t}{dt} = -k_2 C_t^2. \quad (17)$$

Eq.(17) was integrated with the boundary conditions of $C_t=0$ at $t=0$ and $C_t=C_i$ at $t=t$ to yield

$$\frac{1}{C_t} = k_2 t + \frac{1}{C_0}, \quad (18)$$

where C_0 and C_t (mg/L) is the concentration of solute at equilibrium and at time t (min), respectively, and k_2 (L/(mg·min)) is the rate constant.

In earlier years, the second-order rate equations were reasonably applied to describe adsorption reactions occurring in soil and soil minerals. Recently, the equation has also been used to describe fluoride adsorption onto acid-treated spent bleaching earth (Mahramanlioglu *et al.*, 2002) and phosphamidon adsorption on an antimony(V) phosphate cation exchanger (Varshney *et al.*, 1996).

ADSORPTION DIFFUSION MODELS

It is generally known that a typical liquid/solid adsorption involves film diffusion, intraparticle diffusion, and mass action. For physical adsorption, mass action is a very rapid process and can be negligible for kinetic study. Thus, the kinetic process of adsorption is always controlled by liquid film diffusion or intraparticle diffusion, i.e., one of the processes should be the rate limiting step (Meng, 2005). Therefore, adsorption diffusion models are mainly constructed to describe the process of film diffusion and/or intraparticle diffusion.

Liquid film diffusion model

1. Linear driving force rate law

In liquid/solid adsorption systems the rate of solute accumulation in the solid phase is equal to that of solute transfer across the liquid film according to the mass balance law. The rate of solute accumulation in a solid particle clearly equals to $V_p \left(\frac{\partial \bar{q}}{\partial t} \right)$, where

\bar{q} represents the average solute concentration in the solid, and V_p the volume of the particle. Meanwhile the rate of solute transfer across the liquid film is proportional to the surface area of the particle A_s and the concentration driving force $(C-C_i)$. Therefore, it equals to $k_f A_s (C-C_i)$, where k_f represents the film mass transfer coefficient (Cooney, 1999). With the discussion above we can obtain

$$V_p \left(\frac{\partial \bar{q}}{\partial t} \right) = k_f A_s (C - C_i), \quad (19)$$

where C_i and C denote the concentration of solute at the particle/liquid interface and in the bulk of the liquid far from the surface, respectively. Eq.(19), thus, can be rearranged to

$$\frac{\partial \bar{q}}{\partial t} = k_f \frac{A_s}{V_p} (C - C_i). \quad (20)$$

The ratio A_s/V_p that is the particle surface area per unit particle volume can be defined as S_0 . Then, Eq.(20) can be written as

$$\frac{\partial \bar{q}}{\partial t} = k_f S_0 (C - C_i). \quad (21)$$

Eq.(21) is called as "linear driving force" rate law, which is usually applied to describe the mass transfer through the liquid film.

2. Film diffusion mass transfer rate equation

The film diffusion mass transfer rate equation presented by Boyd *et al.*(1947) is

$$\ln \left(1 - \frac{q_t}{q_e} \right) = -R^1 t, \quad (22)$$

$$R^1 = \frac{3D_e^1}{r_0 \Delta r_0 k'}, \quad (23)$$

where R^1 (min^{-1}) is liquid film diffusion constant, D_e^1 (cm^2/min) is effective liquid film diffusion coefficient, r_0 (cm) is radius of adsorbent beads, Δr_0 (cm) is the thickness of liquid film, and k' is equilibrium constant of adsorption.

A plot of $\ln(1-q_t/q_e) \sim t$ should be a straight line with a slope $-R^1$ if the film diffusion is the rate

limiting step. Then the corrected liquid film diffusion coefficient D_e^1 can be evaluated according to Eq.(21). The film diffusion mass transfer rate equation has been successfully applied to model several liquid/solid adsorption cases, e.g., phenol adsorption by a polymeric adsorbent NDA-100 under different temperature and initial concentration conditions (Meng, 2005).

Intraparticle diffusion model

1. Homogeneous solid diffusion model (HSDM)

A typical intraparticle diffusion model is the so-called homogeneous solid diffusion model (HSDM), which can describe mass transfer in an amorphous and homogeneous sphere (Cooney, 1999). The HSDM equation can be presented as

$$\frac{\partial q}{\partial t} = \frac{D_s}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial q}{\partial r} \right), \quad (24)$$

where D_s is intraparticle diffusion coefficient, r radial position, and q the adsorption quantity of solute in the solid varying with radial position at time t .

Crank (1956) gave an exact solution to Eq.(24) for the "infinite bath" case where the sphere is initially free of solute and the concentration of the solute at the surface remains constant. External film resistance can be neglected according to the constant surface concentration (Cooney, 1999). Then, Crank's solution is written as follows

$$\frac{q}{q_s} = 1 + \frac{2R}{\pi r} \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \sin \frac{n\pi r}{R} \exp \left(\frac{-D_s n^2 \pi^2 t}{R^2} \right), \quad (25)$$

where R is the total particle radius.

The average value of q in a spherical particle at any particular time, defined as \bar{q} , is presented as follows:

$$\bar{q} = \frac{3}{R^3} \int_0^R q(r) r^2 dr, \quad (26)$$

where $q(r)$ is the local value of the solid-phase concentration. By inserting the solution for $q(r)$ into Eq.(26), the following equation can be obtained:

$$\frac{\bar{q}}{q_\infty} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp \left(\frac{-D_s n^2 \pi^2 t}{R^2} \right), \quad (27)$$

where q_∞ represents the average concentration in the solid at infinite time.

For a short time, when $\bar{q}/q_\infty < 0.3$, Eq.(27) can be simplified to yield

$$\frac{\bar{q}}{q_\infty} = 6 \left(\frac{D_s}{R^2 \pi} \right)^{1/2} t^{1/2}. \quad (28)$$

A value of D_s from short-time data can be determined by plotting of \bar{q}/q_∞ against $t^{1/2}$. It can also be concluded from Eq.(28) that adsorption rate reduces along with increasing adsorbent particle size and vice versa.

For a long time, Eq.(27) may be written as follows:

$$\frac{\bar{q}}{q_\infty} = 1 - \frac{6}{\pi^2} \exp \left(\frac{-D_s \pi^2 t}{R^2} \right). \quad (29)$$

The linearization of Eq.(29) gives

$$\ln \left(1 - \frac{\bar{q}}{q_\infty} \right) = \frac{-D_s \pi^2}{R^2} t + \ln \frac{6}{\pi^2}. \quad (30)$$

Similarly, the value of D_s from long-time data can be determined by plotting of $\ln(1 - \bar{q}/q_\infty)$ vs t . However, the assumption of constant surface concentration for HSDM is likely to be violated at a long time. Therefore, the equation discussed above is generally somewhat valid in a short time (Cooney, 1999).

In recent years, HSDM has been applied to different kinds of adsorption systems (Cheng, 2004), such as the adsorption of salicylic acid and 5-sulfosalicylic acid from aqueous solutions by hypercrosslinked polymeric adsorbent NDA-99 and NDA-101. In the adsorption systems of pentachlorophenol (PCP) onto activated carbon, diffusion coefficient D_s evaluated from batch kinetic adsorption experiments has been applied to fixed-bed situation,

such as the prediction of the adsorption breakthrough curves and design of fixed beds for removal of PCP (Slaney and Bhamidimarri, 1998).

2. Weber-Morris model

Weber-Morris found that in many adsorption cases, solute uptake varies almost proportionally with $t^{1/2}$ rather than with the contact time t (Alkan et al., 2007):

$$q_t = k_{\text{int}} t^{1/2}, \quad (31)$$

where k_{int} is the intraparticle diffusion rate constant.

According to Eq.(31), a plot of $q_t \sim t^{1/2}$ should be a straight line with a slope k_{int} when the intraparticle diffusion is a rate-limiting step. For Weber-Morris model, it is essential for the $q_t \sim t^{1/2}$ plot to go through the origin if the intraparticle diffusion is the sole rate-limiting step. However, it is not always the case and adsorption kinetics may be controlled by film diffusion and intraparticle diffusion simultaneously. Thus, the slope is not equal to zero.

3. Dumwald-Wagner model

Dumwald-Wagner proposed another intraparticle diffusion model as (Wang et al., 2004)

$$F = \frac{q_t}{q_e} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp(-n^2 Kt), \quad (32)$$

where K (min^{-1}) is the rate constant of adsorption. Eq.(32) can be simplified as

$$\log(1 - F^2) = -\frac{K}{2.303} t. \quad (33)$$

A plot of $\log(1 - F^2) \sim t$ should be linear and the rate constant K can be obtained from the slope. Dumwald-Wagner model proved to be reasonable to model different kinds of adsorption systems, e.g., *p*-toluidine adsorption from aqueous solutions onto hypercrosslinked polymeric adsorbents.

Double-exponential model (DEM)

A double-exponential function proposed by Wilczak and Keinath (1993) was used to describe lead and copper adsorption onto activated carbon. In this case, the uptake process of both metals could be divided into two steps, namely a rapid phase involving

external and internal diffusions, followed by a slow phase controlled by the intraparticle diffusion. It was demonstrated that the two-step mechanism can be described fairly well with the double-exponential model (Chiron et al, 2003), which is presented as follows:

$$q_t = q_e - \frac{D_1}{m_a} \exp(-K_1 t) - \frac{D_2}{m_a} \exp(-K_2 t), \quad (34)$$

where D_1 (mmol/L) and K_1 (min^{-1}) are the adsorption rate parameters and diffusion parameters of the rapid step, respectively, and D_2 and K_2 for the slow step.

If $K_1 \gg K_2$, it means that the rapid process can be assumed to be negligible on the overall kinetics. Eq.(34) can then be simplified as

$$q_t = q_e - \frac{D_2}{m_a} \exp(-K_2 t), \quad (35)$$

and be rearranged to a linear form

$$\ln(q_e - q_t) = \ln \frac{D_2}{m_a} - K_2 t, \quad (36)$$

the parameters D_2 and K_2 can be determined by plotting of $\ln(q_e - q_t)$ against t , then D_1 and K_2 can be obtained from the following equation:

$$\ln \left(q_e - q_t - \frac{D_2}{m_a} \exp(-K_2 t) \right) = \ln \frac{D_1}{m_a} - K_1 t. \quad (37)$$

Then, Eq.(34) can be used to fit the kinetic process of adsorption.

Nevertheless, values of K_1 and K_2 are not sufficient to describe and interpret the influence of external and internal diffusions. As the rapid step involves both diffusion steps, these parameters can only allow a comparison of the respective adsorption rate of Pb(II) and Cu(II).

DEM can also describe a process where the adsorbent offers two different types of adsorption sites. On the first-type site rapid adsorption equilibration occurs within a few minutes, whereas on the second site type, adsorption is more slowly. DEM is

particularly suitable for modeling heavy metals adsorption, e.g., adsorption of Cu(II) and Pb(II) from aqueous solutions by activated carbon and grafted silica (Chiron *et al.*, 2003).

CONCLUSIONS AND PERSPECTIVES

Both adsorption reaction models and adsorption diffusion models are now widely employed for fitting kinetic data. Based on the above analysis, it can be seen that appropriate adsorption reaction models should be chosen according to the mechanism of adsorption process and the applicability of different models. Generally speaking, they cannot represent the real course of adsorption and thus, cannot offer useful information to gain insight in adsorption mechanism and to design fixed-bed systems. On the contrary, adsorption diffusion models based on three basic steps can represent the real adsorption course more reasonably, and intraparticle diffusion coefficient D_s , determined from these models are useful for design of fixed-bed systems.

Today, adsorption kinetic study still attracts considerable interest because of its particular significance in adsorbent evaluation and application, and more deliberate but complicated models will be proposed on the basis of adsorption mechanism and diffusion analysis. Meanwhile, new effective softwares are required to solve such complex mathematical models. These new models are expected to describe the performance of a given adsorbent more accurately, and to help to gain insight in adsorption mechanism.

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