



Evaluation of Kinetic Adsorption Models in Aquatic Environment, Models of Lagergren and Ho et al.

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Abstract: Heavy metal pollution has become one of the most crucial environmental obstacle today. The treatment of heavy metals is of special concern due to their refractoriness and resistance in the environment. In recent years, different procedures for heavy metal removal from wastewater have been widely studied. This paper revises the common procedures that have been applied to treat heavy metal wastewater and considers these techniques. Presently kinetic adsorption equations are widely used in adsorption processes. The main objective of the present research is to evaluate the linear and non-linear equations of the Lagergren and Ho et al. models in the kinetics of nickel adsorption by activated carbon. For this purpose, powdered activated carbon, the raw material of which is wood, was purchased from the "Merck" company (Germany). The optimum PH of the adsorption was obtained to be 6. As well, the kinetics of adsorption showed that for the initial nickel concentration of 2.5, 5, 10, 50 and 125 mg/L, equilibrium time was 40, 75, 120, 150, 50 minutes respectively. The maximum removal efficiency of nickel in the initial concentration of 2.5 mg/L was obtained to be 89.6%. The fitting of the four linear kinetic models of Ho et al. (1995), and its non-linear model, as well as the linear model of kinetic data of adsorption at different concentrations showed that the non-linear model of Ho et al. (1995) better describes the kinetic data of nickel adsorption by activated carbon than his linear models. Comparison of the non-linear models of Ho et al. and Lagergren (1893) showed that in low concentrations the non-linear model of Ho et al., and in high concentrations non-linear model of Lagergren better describes the kinetic data of adsorption. First and second-order rate equations were pragmatic to study adsorption kinetics. The most significant advantage of the offered model is its capability to predicate the balance time of adsorption, which is very essential to optimize the expense of the adsorption system design. The received consequences depict that the new model can analyze the experiential information very well. The equation is received clearly by changing the mass balance equation of a single step batch-type system into integrated first order rate equation. The proposed new formulation depends on maximum sorption (or removal) rather than equilibrium sorption, as given by Lagergren equation. The precise value of equilibrium sorption is not easy to describe in many cases, and it is exhibited that the modified formulation prepares better correlation with sorption data when the procedure is far from completion. A corrected formulation of the popular Lagergren pseudo first order rate equation is proposed. The equation is created clearly by changing the mass balance equation of a single step batch-type system into unified first order rate equation. The proposed new formulation depends on maximum sorption (or removal) rather than equilibrium sorption, as given by Lagergren equation. The precise value of equilibrium sorption is not simple to describe in many cases, and it is exhibited that the modified formulation provides better correlation with sorption data when the process is far from completion.

Keywords: Adsorption, Nickel, Linear and Non-linear Models, Activated Carbon, Kinetic Adsorption

Table 1. List of Abbreviations

$k\alpha$: the rate constant

V : the volume of the solution (L)

k_2 : the Sobkowsk and Czerwinski rate constant

q_t : the amount of ion sorbed at time (mg/g)

q_∞ : the amount of sorption after an infinite time

k_1 : the adsorption constant (1/min)

q_e : the amount of nickel ion sorbed at equilibrium (mg/g)

q_t : the amount of ion sorbed at time (mg/g)

C_0 : the initial concentration of metal ion (mg/L)

k_2 : the adsorption constant (g/mg.min)

c_t : the concentration of metal ion solution at time t (mg/L)

n_0 : the exchange capacity

m : the adsorbent weight (gr)

θ : the fraction of surface which is sorbed by gas

α_p : The Blanchard et al. rate constant

INTRODUCTION

Studies on the adsorption of heavy metals from industrial wastewaters have seen a remarkable growth in recent years due to the increasing use of such pollutants. Based on the objectives of adsorption studies various models for the analysis of adsorption processes are selected and used. One of the important goals of such studies is to determine the time required for optimal adsorption in the solid-liquid system (Jonathan et al., 2009:616). The mechanism of adsorption process and its time is called adsorption kinetics (Royer et al., 2009:1213; Lima et al., 2008:536). Adsorption kinetics is affected by various factors, such as concentration of the sorbed material, amount, size and special surface area of the adsorbent, pH of solution and ambient temperature (Shamohammadi, Heydari, 1386:126). For the best-fitting of the adsorption kinetic data models of Ho et al. (1995) and Lagergren (1893) are more efficient (Jonathan et al., 2009:616), where two types of equations, linear and non-linear, are used. Lagergren's model does not necessarily show the actual nature of adsorption, so the capability by this model to analyze the adsorption data is doubtful (Schiewe et al., 2008:1896). Ho et al. (2006) examined and evaluated the linear and nonlinear behavior of the Ho et al. (1995) model. Their results showed that the nonlinear model is better fitted with the adsorption data and provides a better ability to determine the parameters of the model. In the Table 2 the corrections of the Ho et al. model of adsorption kinetics are showed (Ho, 2006: 119).

At present, more than 40% of the nickel found in the environment is produced by industrial plants and battery factories (Ajmal et al., 2001:117), that have a great role in contaminating the surface waters (Dursun et al., 2006:187). Nickel concentration in industrial wastewater ranges from 3.4 to 900 mg/L (Patterson, 1985:217).

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It is possible with the adsorbents, such as active carbon (Erdogan et al., Akbar Esma'ili and Samira Qasemi, 2009:1818), zeolite (Prasanth et al., 2008:735), clay (Pratik et al., 2007:216), and sawdust (Rafatullah et al., 2009:969), to adsorb nickel in high capacity. The main objective of this research is to evaluate the linear and nonlinear equation models of Ho et al. and Lagergren, as well as to examine the kinetic parameters of nickel adsorption by activated carbon.

Table 2. Corrections of the Adsorption Kinetics Model of Ho et al.

References	Linear form of the model
Sobkowsk and Czerwinski (1974)	$\frac{\theta}{1-\theta} = \kappa_2 t$
Ritchie (1977)	$\frac{q_{\infty}}{q_{\infty} - 1} = \alpha t + 1$
Blanchard et al. (1984)	$\frac{1}{n_o - n} - \alpha_p - \kappa t$
Ho (1995)	$\frac{t}{q_t} = \frac{1}{\kappa q_e^2} + \frac{1}{q_e} t$

MATERIALS AND METHODS

In this research a standard nickel solution (Nickel Titrisol) manufactured by the German Merck company was purchased and a solution was prepared with the concentration of 1000 mg/L (stock solution). In all of the experiments the volume of the solution used was 50 ml, based on the 250 ml Erlenmeyer flask. The utensils were washed with acid and then with deionized water. All solutions were tested with Whatman filter paper grade 40. The pH was measured by pH meter (model PTR 79) and to adjust pH nitric acid and sodium hydroxide solutions were used. Measurement of ion adsorption was carried out by the Spectroquant meter of the German Merck company. During all of the experiments the room temperature was controlled to be in the range of 22 to 24°C.

Preparation of Adsorbent

Powdered activated carbon, the raw material of which is wood, was purchased from the German Merck company. Some characteristics of the used activated carbon and its consisting elements are presented in Table 3.

Table 3. Characteristics of Powdered Activated Carbon and its Consisting Elements

Characteristics	Amount
Amount of the dissolved ethanol	0.2%
Chloride	0.01%
Sulfate	0.01%
Lead	0.005%
Iron	150 mg/L
Amount of the dissolved hydrochloric acid	1%

Effect of the Optimal Adsorption PH

In eight 100 ml capacity Erlenmeyer flasks 1 g/L of weighted adsorbent was put inside of each of the Erlenmeyer flasks. Then, a metal solution with a concentration of 10 mg/L (Jamali Paqal'e, 1388:76) was added to each of the Erlenmeyer flasks. Different pH values (ranging from 2 to 8) were examined. The solutions were placed on a shaker rotating at the speed of 150 rpm (Aydin et al., 2008:37) in a controlled temperature for 12 hours (Babel et al., 2003:14).

Effect of Contact Time and Initial Nickel Concentration:

Twelve Erlenmeyer flasks were picked and in each of them nickel solution with a concentration of 2.5 mg/L was poured. Also, the amount of 0.05 grams of activated carbon adsorbent was added to each of the Erlenmeyer flasks and they with an optimal pH were put on the rotating shaker at the speed of 150 rpm for 5 to 240 minutes. This procedure was repeated with the concentrations of 5 mg/L, 10 mg/L, 50 mg/L and 125 mg/L.

Kinetic Adsorption Models

Lagergren’s model(1898): this model is expressed as a equation based on the adsorbent capacity $\frac{dq}{dt} = K_1(q_e - q)$. By integrating the previous equation and boundary conditions (q=0, t=0 and q=q, t=t), it was changed to equation $\ln(q_e - q) = \ln q_e - K_1 t$, that may be rearranged to the non-linear form. Also its linear form is written as $\text{Log}(q_e - q_t) = \text{Log}q_e - \frac{K_1}{2.303} t$ equation In order to fit the linear models with the experimental data, q_t and $\text{Log}(q_e - q_t)$ is written in front of t . In the linear form line slope $\frac{K_1}{2.303}$ and its y-intercept will be $\text{Log}q_e$. Therefore, it is possible to obtain k_1 and q_e from the slope and y-intercept of linear equation (Ho et al, 2002:797).

Ho et al. model (1995): this model is widely used in analyzing and examining kinetic adsorption data (Ho et al, 2001:241; Ho et al., 2000:189). It is possible to write this model in different linear forms (Makris et al., 2005: 4250; Duons et al., 2005:438; Wang et al., 2005:336; Janos et al., 2005:19). It has four linear forms (Yuh et al., 2006:119) (Table 4).

Differential form of the Ho et al. model (1996) is written as follows: $\frac{dq_t}{dt} = k_2(q_e - q_t)^2$. As well, for the boundary conditions (t=0 to t and q=0 to q) non-linear equation form is written like this: $q = \frac{q_e^2}{1 + q_e k_2 t}$

Table4. Linear Forms of Ho et al. Model

Type of Equation	Linear Equation Form of Ho et al.
1	$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{1}{q_e} t$
2	$\frac{1}{q_t} = \left(\frac{1}{kq_e^2}\right) \frac{1}{t} + \frac{1}{q_e}$
3	$q_t = q_e - \left(\frac{1}{kq_e}\right) \frac{q_t}{t}$
4	$\frac{q_t}{t} = kq_e^2 - kq_e q_t$

The linear equations of Ho and et al. model provide different amounts in determining the adsorption parameters (Ho, 2004:2115; Longhinoti et al., 1998:435; Kinniburgh, 1986:895). First type linear equation (see Table 4) is more known than the other types (Ho et al., 2006:119; Yuh at al., 2006:119). By fitting the models with the experimental data, in both linear and nonlinear forms, the coefficients of the Ho et al. model are obtained. Then q_t and t/q_t are written against t respectively, and in this way constants k_2 and q_e are calculated. As well, it is possible to calculate the q_t by using the following equation $q_t = \frac{c_e - c_t}{m} \times v$.

RESULTS AND DISCUSSION

Determining optimal PH

Environmental acidity, in which the adsorption process takes place, has a great influence on the absorption rate (Larous at al., 2005:483; Ahmad et al., 2008:127). Part of this effect is due to the adsorption of hydrogen

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ions (Jamali Paqal'e, 1388:76), which appear as competitors in the surface adsorption of cations (Addel Ghani et al., 2007:67; Srivastava et al., 2009:370). Figure 1 presents effect of pH on nickel adsorption efficiency. The maximum adsorption efficiency in pH=6 equals to 79.24%. The intensity of nickel adsorption from pH=2 to pH=6 is high and in pH=6 reaches its maximum rate. From pH=6 to pH=8 adsorption intensity remains almost constant. The relatively rapid increase of adsorption efficiency in pH<6 can be attributed to two factors: 1. In low pH rates the amounts of H⁺ ions found in solution, that compete with the nickel positive ions for the surface adsorption, their amount is enough; 2. For each metal ion apt to hydrolysis there is a critical pH range point, at which the metal adsorption efficiency from a very small value reaches its maximum value. This value is called surface adsorption threshold (Kumar et al., 2006:104). Constant value in pH>6 corresponds to the results of the reports of Vimal Chandra et al. (2006:257) and Semerijian (2009:155).

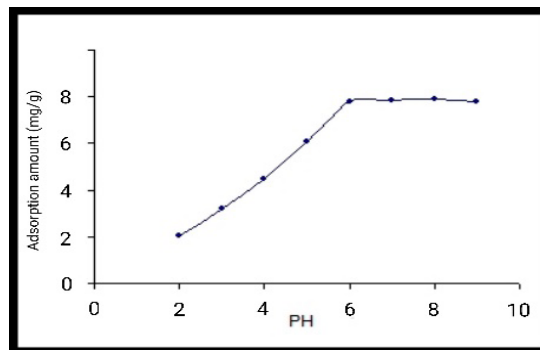


Fig1. Effect of PH on Nickel Adsorption Efficiency by Powdered Activated Carbon Adsorbent (Initial concentration of nickel - 10 mg/L, contact time - 5 hours, amount of adsorbent - 1 g/L)

Kinetic Adsorption Experiments

Effect of Contact Time and Initial Concentration

In Figure 2 the changes in the amount of nickel adsorption by the activated carbon adsorbent together with the time are presented. As can be seen, initially, the adsorption intensity rate is high in all concentrations, but over time, adsorption intensity decreases until finally it reaches a constant value. The period of time necessary for the adsorption amount to reach its maximum is called equilibrium time. At this point the maximum amount of adsorption occurs.

Maximum amount of adsorption in 2.5 mg/L, 5 mg/L, 10 mg/L, 50 mg/L and 125 mg/L concentrations is respectively 2.24 mg/g, 4.3 mg/g, 7.96 mg/g, 14.26 mg/g and 21.22 mg/g.

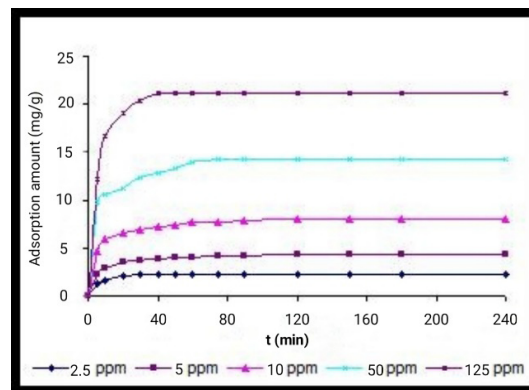


Fig2. Effect of Contact Time of Activated Carbon Adsorbent on Removing Nickel (Amount of adsorbent - 10 g/L and pH=6)

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Therefore, it may be concluded that for a certain amount of adsorbent by increasing nickel ion concentration from 2.5 to 125 mg/L, the adsorption amount decreased from 89.6% to 21.22%. These results are consistent with the results of studies performed by Jamali et al. (2008: 65).

Also, the results of the experiments of the effect of initial solution concentration on the equilibrium time of the nickel adsorption are shown in Figure 3.

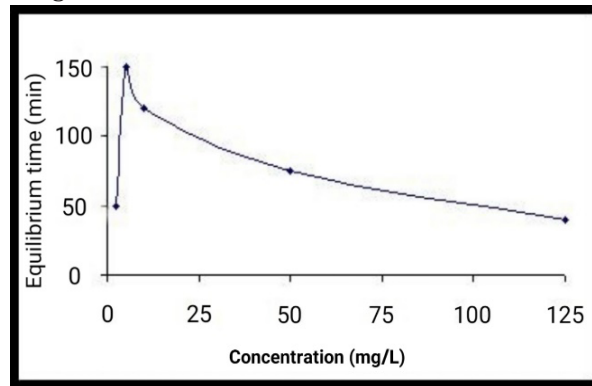


Fig3. Effect of Initial Nickel Concentration on Equilibrium Time by Powdered Activated Carbon

As can be seen, equilibrium time of nickel adsorption by activated carbon adsorbent in 2.5 mg/L, 5 mg/L, 10 mg/L, 50 mg/L, 125 mg/L respectively equals to 50, 150, 120, 75, 40 minutes.

According to the above Figure, it can be said that by increasing the concentration of nickel solution from 5 to 125 mg/L, and also by decreasing the concentration of solution from 5 to 2.5 mg/L, the equilibrium time of the adsorption process decreases. By reducing the initial concentration of the solution, it is expected that the time to achieve equilibrium will be increased due to a decreased chance of metal contact with adsorbent. However, given that in low concentrations the amount of metal ions found in the solution is low, this limited amount of ion is absorbed by the pores of the adsorbent. Therefore, the time to reach the equilibrium decreases by decreasing initial concentration of nickel. In this case (when concentrations are low), the factor determining equilibrium time is the concentration of solution. In high concentrations, due to the high accumulation of metal ions around the adsorbent and increased chance of metal contact with the adsorbent, it takes a little more time for the adsorbent capacity to reach its saturation and the process to reach equilibrium.

Therefore, in this case also, the factor determining the equilibrium time is the adsorbent's capacity. These results are consistent with the results of the studies performed by Jamali (1387:71), Jamali Paqal'e (1388:88) and (Azizian, 2004:47).

Kinetic Adsorption Equations

The transformation of the non-linear model of Ho et al. to linear equations may lead to an increase in the error variance and standard least squares (Kinniburgh, 1986:895; Ho, 2004:2115).

Figures 4 and 5 present respectively the fitting of non-linear models of Lagergren and Ho et al., and Figures 6, 7, 8, 9, 10 present respectively the fitting of the linear models of Lagergren and Ho et al. As well kinetic parameters of the linear and non-linear equations adsorption of the Lagergren and Ho et al. models respectively are shown in Tables 5 and 6. The results show that the numerical values of the kinetic parameters of adsorption q_e , k_1 (in each of the four linear models of Ho et al., Type 1, 2, 3 and 4) differ significantly, which can be explained by their non-linear regression coefficients. These results correspond to the studies performed by Yuh et al. (2006:119). Therefore, having a reliable non-linear model (Ho et al., 1995:1327), the usage of different linear equations of this model does not seem necessary.

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As well, the results of the fitting of the non-linear models of Lagergren and Ho et al. (Fig. 6) show that, although both models describe the data by the accuracy of 95%, the model of Ho et al. is more efficient for all concentrations used in the test. The reason of this might be due to the hypothesis of Ho et al. regarding the effect of the adsorbent capacity on the initial rate of adsorption (Jonathan et al., 2009:616). These results do not correspond to the results of Shamohmmadi and Heydari (1386:126), but is in correspondence with the results of the studies performed by Jamali at al. (1386:70), Jamali Paqal'e (1388:90), Jonathan et al. (2009:616), Mohama et al. (2009:12) and Ho (2004:2115).

Comparison of the non-linear models of Ho et al. with the Lagergren's model in different concentrations, presented in Table 6, shows that in low concentrations the Lagergren's model and in high concentration the Ho et al. model has better regression coefficient, and therefore describes the data better. These results correspond to the results of the study performed by Azizian (2004:47).

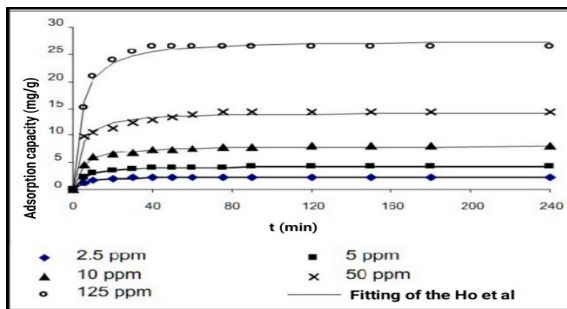


Fig4. Fitting of the Lagergren's non-linear model with kinetic adsorption data

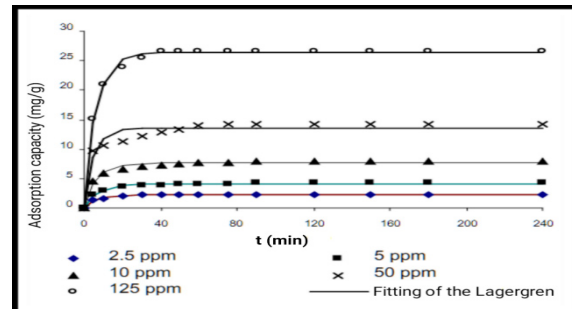


Fig5. Fitting of the Ho et al. non-linear model with kinetic adsorption data

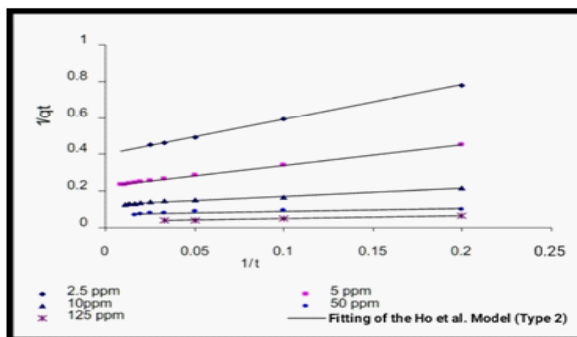


Fig6. Fitting of the Lagergren's linear model with kinetic adsorption data

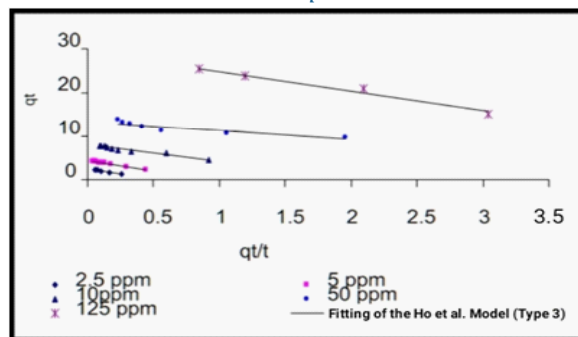


Fig7. Fitting of the Ho et al. Type 1 linear model with kinetic adsorption data

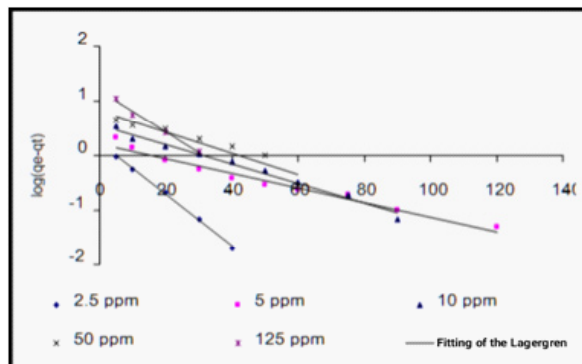


Fig8. Fitting of the Ho et al. Type 2 linear model with kinetic adsorption data

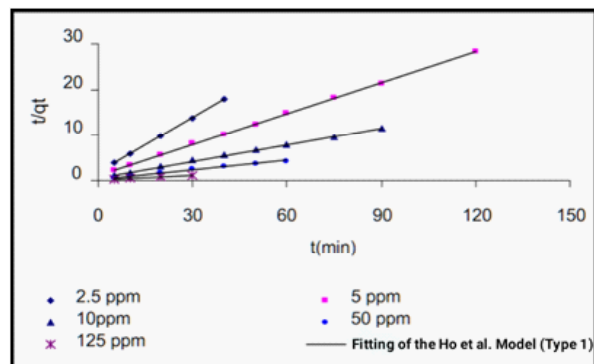


Fig9. Fitting of the Ho et al. Type 3 linear model with kinetic adsorption data

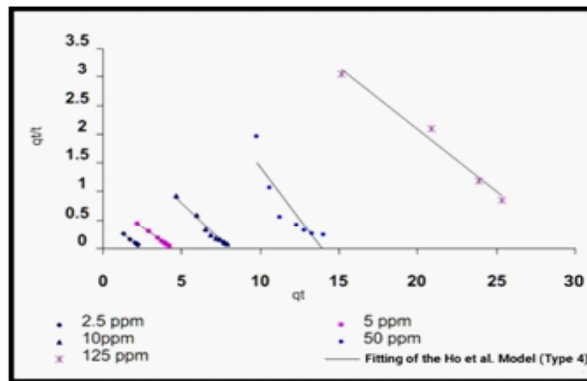


Fig10. Fitting of the Ho et al. Type 3 linear model with t kinetic adsorption data

The coefficient of the Ho et al. model (k_2) shows surface adsorption rate of the adsorbent before reaching equilibrium. The higher the coefficient, the greater is the surface adsorption rate of the metal ion by the adsorbent (Shahmohammadi Heydari, 1386:126). As is shown in Table 6, by increasing the solution concentration from 2.5 to 125 mg/L, the coefficient k_2 of the Ho et al. non-linear model is reduced. These results correspond to the studies of Aksu (2008:112), Mane et al. (2007:390), Han et al. (2009:496) and Azizian et al. (2006:112). Rate of the coefficient k_2 by increasing concentration in the Lagergren’s model at first increases and later decreases (there is no linear relation). As it is seen, this is not a linear relation and therefore in this case the Lagergren’s model has no good capacity in describing the data (Azizian, 2004:47). Thus, this result confirms the results of the comparison of the regression coefficients of the the Lagergren models and Hu et al. models.

Table5. Constants of the linear kinetic models for absorbing nickel by powdered activated carbon adsorbent

Ho et al. (Type 4)			Ho et al. (Type 3)			Ho et al. (Type 2)			Ho et al. (Type 1)			Lagergren			$q_{e\text{exp}}$	Concentration mg/l
R^2	q_e	k_2	R^2	q_e	k_2	R^2	q_e	k_2	R^2	q_e	k_2	R^2	q_e	k_1		
0.998	2.503	0.0831	0.1093	1.726	0.997	0.085	2.493	0.999	0.0835	2.501	0.999	0.0833	2.502	0.998	2.5	2.24
0.998	4.4	0.0456	0.0306	1.643	0.969	0.0428	4.430	0.999	0.0428	4.391	0.999	0.0457	4.399	0.998	5	4.3
0.969	8.035	0.0326	0.0414	3.667	0.989	0.025	8.244	0.999	0.0342	7.98	0.986	0.0338	7.999	0.969	10	7.96
0.79	13.861	0.0265	0.0446	6.652	0.969	0.016	14.513	0.994	0.0364	13.368	0.848	0.0345	13.469	0.790	50	14.26
0.981	29.632	0.0072	0.0882	15.649	0.883	0.008	29.06	0.999	0.007	29.85	0.992	0.0074	29.477	0.981	125	26.53

Table6. Constants of the non-linear kinetic models for absorbing nickel by powdered activated carbon adsorbent

Ho et al.			Lagergren			$q_{e\text{exp}}$	Concentration mg/l
R^2	q_e	k_2	R^2	q_e	k_1		
0.993	2.244	0.114	0.985	2.226	0.1518	2.24	2.5
0.996	4.411	0.004	0.977	2.344	0.1262	4.3	5
0.997	8.097	0.003	0.966	7.619	0.158	7.96	10
0.979	14.349	0.002	0.934	13.583	0.1958	14.26	50
0.993	27.705	0.001	0.995	26.338	0.1601	26.53	125

CONCLUSIONS

1. Non-linear model of Ho et al. (1995) better describes the kinetic data of nickel adsorption by activated carbon than their linear models.
2. In the non-linear model of Ho et al. (1995) in increasing concentration of nickel solution, adsorption rate is reduced and the equilibrium concentration on solid phase (q_e) is increased.
3. Comparison of the Ho et al. and Lagergren's non-linear models shows that in low concentrations the non-linear model of Ho et al. and in high concentrations the Lagergren's non-linear model better describes the kinetic data of nickel adsorption by activated carbon.
4. Adsorption procedures can be modeled on the basis of phenomenological theories using conservation, equilibrium and transport kinetics equations. However many times an experiential procedure is followed using pseudo first order and pseudo second order models to explain adsorption kinetics. A relation between the kinetic constants of both models is insulated, being possible to reveal they exhibit very different dependency with temperature whatever the porous structure of the adsorbent particle. Adsorption kinetic curves of the Lagergren's first-order (LFO) equation were classified into four areas pursuant to their ascending characteristics. Of the 85 adsorption procedures defined by Lagergren's first-order equation, 46% of the kinetic curves depended on zone II and 29% to zone III, these being good and fast. The standard models are found to be special states of the general adsorption-diffusion model for determined quantities of the dimensionless numbers. The kinetics of adsorption was described by applying Lagergren's pseudo-first order and pseudo-second order equations and the consequences have exhibited that the adsorption procedures follows pseudo-second order kinetics and the adsorption procedures belongs on both time and concentration.
5. The precise value of equilibrium sorption is not simple to describe in many cases, and it is exhibited that the modified formulation prepares better correlation with sorption information when the procedure is far from performance. In result, care must be taken when applying the shortcut procedure of pseudo first order expressions, taking into account their theoretical inconvenient attitudes.
6. A corrected formulation of the popular Lagergren pseudo first order rate equation is proposed. The equation is created clearly by changing the mass balance equation of a single step batch-type system into unified first order rate equation. The proposed new formulation depends on maximum sorption (or removal) rather than equilibrium sorption, as given by Lagergren equation. The precise value of equilibrium sorption is not simple to describe in many cases, and it is exhibited that the modified formulation provides better correlation with sorption data when the process is far from completion. The mechanistic route of the adsorption procedure was assessed with intraparticle diffusion model. The trace of heat of adsorption of the adsorbate onto the adsorbent material was defined using the thermodynamic parameters and the reusability of the adsorbent materials was defined with desorption studies.
7. Adsorption has been demonstrated to be an great way to treat industrial waste sewages, offering important superiorities like the relatively inexpensive, accessibility, profitability, easy of action and performance.

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Citation: Zahra Askari, Shayan Shamohammadi, Ali Hasantabar-Amiri, Kaveh Ostad-Ali-Askari, Saeid Eslamian, Vijay P. Singh, "Evaluation of Kinetic Adsorption Models in Aquatic Environment, Models of Lagergren and Ho et al.," *American Research Journal of Civil and Structural Engineering*, vol 2, no. 1, pp. 1-13.

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