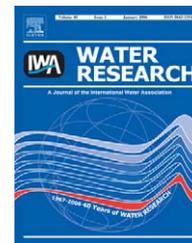


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Second-order kinetic model for the sorption of cadmium onto tree fern: A comparison of linear and non-linear methods

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ABSTRACT

A comparison was made of the linear least-squares method and a trial-and-error non-linear method of the widely used pseudo-second-order kinetic model for the sorption of cadmium onto ground-up tree fern. Four pseudo-second-order kinetic linear equations are discussed. Kinetic parameters obtained from the four kinetic linear equations using the linear method differed but they were the same when using the non-linear method. A type 1 pseudo-second-order linear kinetic model has the highest coefficient of determination. Results show that the non-linear method may be a better way to obtain the desired parameters.

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1. Introduction

Sorption of pollutants from wastewater has long been studied. The rate at which sorption takes place is of the utmost importance when designing batch sorption systems, consequently it is important to establish the time dependence of such systems under various process conditions. Previously, Sobkowski and Czerwiński (1974) used a second-order rate equation based on the sorption capacity of a solid for a higher concentration of the solid for the rate of reaction of carbon dioxide sorption on a platinum electrode. Ritchie used a second-order empirical equation to test the sorption of gases onto a solid (Ritchie, 1977). The Ritchie equation has also been applied in solution/solid sorption systems, for example, the sorption of cadmium ions onto bone char (Cheung et al., 2001) and the sorption of Cd(II) onto acid-treated jackfruit peel (Inbaraj and Sulochana, 2004). Blanchard et al. reported a similar rate equation for the overall exchange reaction of NH_4^+

ions fixed in zeolite by divalent metallic ions in solution (Blanchard et al., 1984). The second-order expression of Blanchard et al. was used to describe the kinetics of exchange processes between the sodium ions from zeolite A and cadmium, copper, and nickel ions from solutions (Biškup and Subotič, 2004). In recent years, Ho described sorption, which included chemisorption and provided a different idea to the second-order equation called a pseudo-second-order rate expression (Ho, 1995). Moreover, a theoretical analysis of the pseudo-second-order model was reported by Azizian (2004). The advantage of Azizian's derivation is that when the initial concentration of a solute is low, the sorption process obeys the pseudo-second-order model. The pseudo-second-order equation has the following advantages: it does not have the problem of assigning an effective sorption capacity; the sorption capacity, rate constant of pseudo-second-order, and the initial sorption rate can all be determined from the equation without knowing any parameter

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Nomenclature	
\bar{q}_t	is the average of q_t
Γ and Γ_∞	are the surface concentrations at time t and after completion of the reaction
θ	is the fraction of surface sites which are occupied by a sorbed gas, $\theta = \Gamma/\Gamma_\infty$
α	is the rate constant
α_B	is the Blanchard et al. rate constant
h	is the initial sorption rate (mg/gmin)
K	is the rate constant
k	is the pseudo-second-order rate constant (g/mgmin)
k_2	is the Sobkowsk and Czerwiński rate constant
n	is amount of M^{2+} fixed or the amount of NH_4^+ released at each instant
n_0	is exchange capacity
q	is the amount of sorption at time t
q_∞	is the amount of sorption after an infinite time
q_e	is the amount of cadmium ion sorbed at equilibrium (mg/g)
q_m	is the amount of cadmium ion on the surface of the tree fern at any time, t , obtained from the pseudo-second-order kinetic model (mg/g)
q_t	is amount of cadmium ion on the surface of the tree fern at any time, t , obtained from experiment (mg/g)

beforehand. Table 1 shows a comparison of the second-order rate equations of Sobkowsk and Czerwiński (1974), Ritchie (1977), Blanchard et al. (1984), and Ho (1995).

Tree fern is a complex material containing lignin and cellulose as its major constituents (Newman, 1997). Chemical sorption can occur due to the polar functional groups of lignin, which include alcohols, aldehydes, ketones, acids, phenolic hydroxides, and ethers which serve as chemical bonding agents (Adler and Lundquist, 1963). It is, therefore, concluded that tree fern can be a suitable sorbate for the sorption of metal cations because of its polar and acid characters. The removal of divalent metal ions, such as Zn(II), Cu(II), and Pb(II) using tree fern was first reported by Ho et al. (2002, 2004), as well as Cd(II) (Ho and Wang, 2004), and dyes (Ho et al., 2005).

Kinetic studies can be carried out by batch reactions at various initial sorbate concentrations, sorbent doses, particle sizes, agitation speeds, pH values and temperatures, and they also vary by the sorbent type, sorbate type. Linear regression was frequently used to determine the best-fitting kinetic rate equation. The linear least-squares method was also widely applied to the linearly transformed kinetic rate equations to confirm the experimental data and kinetic rate equations using coefficients of determinations. In this study, a comparison was made of the linear least-squares method and non-linear method of the widely used pseudo-second-order kinetic model to the experimental sorption of cadmium onto tree fern. A trial-and-error procedure was used for the non-linear method using the solver add-in with Microsoft's spreadsheet, Microsoft Excel.

Table 1 – A comparison of the second-order models

References	Linear form	Plot
Sobkowsk and Czerwiński (1974)	$\frac{\theta}{1-\theta} = k_2 t$	$\frac{\theta}{1-\theta}$ vs. t
Ritchie (1977)	$\frac{q_\infty - q}{q_\infty - q} = \alpha t + 1$	$\frac{q_\infty - q}{q_\infty - q}$ vs. t
Blanchard et al. (1984)	$\frac{1}{n_0 - n} - \alpha_B = K t$	$\frac{1}{n_0 - n}$ vs. t
Ho (1995)	$\frac{t}{q_t} = \frac{1}{k q_e^2} + \frac{1}{q_e} t$	$\frac{t}{q_t}$ vs. t

2. Materials and methods

Raw tree fern trunk was dried in an oven at 105 °C for 24 h. The sorbent was screened to obtain four different ranges of geometrical sizes of 43–53, 74–88, and 104–124 μm. Table 2 shows characters of the tree fern (104–124 μm). Stock cad-

Table 2 – Characters of the tree fern

<i>Surface area</i>	
Single point surface area at P/Po = 0.1606	0.1936 m ² /g
BET surface area	0.1516 m ² /g
Langmuir surface area	0.1776 m ² /g
BJH adsorption cumulative surface area of pores between 17.000 and 3000.000 Å diameter	1.185 m ² /g
BJH desorption cumulative surface area of pores between 17.000 and 3000.000 Å diameter	1.449 m ² /g
<i>Pore volume</i>	
Single point adsorption total pore volume of pores less than 1004.7057 Å at P/Po = 0.9803	0.01236 cm ³ /g
Single point desorption total pore volume of pores less than 1014.1506 Å at P/Po = 0.9805	0.01646 cm ³ /g
BJH adsorption cumulative volume of pores between 17.000 and 3000.000 Å diameter	0.022291 cm ³ /g
BJH desorption cumulative volume of pores between 17.000 and 3000.000 Å diameter	0.022288 cm ³ /g
<i>Pore size</i>	
Adsorption average pore width (4V/A by BET)	3260.9007 Å
Desorption average pore width (4V/A by BET)	4343.4703 Å
BJH adsorption average pore diameter (4V/A)	752.420 Å
BJH desorption average pore diameter (4V/A)	615.249 Å

mium solutions were prepared with distilled water and Cd(NO₃)₂·4H₂O. All working solutions were prepared by diluting the stock solution with distilled water.

All contact investigations were carried out using a baffled, agitated 2-dm³ sorber vessel. A predetermined mass of tree fern was added to each 1.7-dm³ batch volume of cadmium ion solution using a fixed agitation speed at 25 °C and an initial pH 5.4 for 60 min. Two and a half milliliters of the mixture was withdrawn at suitable time intervals, and filtered through a 0.45-μm membrane filter. These were analysed by an inductively coupled plasma atomic emission spectroscopy analyser (ICP-AES) for the concentration of cadmium. Experiments were carried out to study the effects of the tree fern dose, *m_s*, particle size, *d_p*, and agitation speed, *S*, on the sorption rate. The experimental conditions adopted are shown in Table 3.

3. Results and discussion

An expression of the pseudo-second-order rate based on the solid capacity has been presented for the kinetics of sorption of divalent metal ions onto peat (Ho, 1995; Ho and McKay, 2000):

$$q_t = \frac{q_e^2 kt}{1 + q_e kt}, \quad (1)$$

where *k* is the pseudo-second-order rate constant (g/mg min), *q_e* is the amount of cadmium ion sorbed at equilibrium (mg/g), and *q_t* is amount of cadmium ion on the surface of the tree fern at any time, *t*, (mg/g).

Eq. (1) can be rearranged to obtain

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

which has a linear form of

$$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{1}{q_e}t. \quad (3)$$

If the initial sorption rate, as *h* = *q_t*/*t* when *t* approaches 0, *h* (mg/g min), is

$$h = kq_e^2. \quad (4)$$

Eq. (2) can be rearranged to obtain

$$q_t = \frac{t}{1/h + t/q_e}, \quad (5)$$

and

$$\frac{t}{q_t} = \frac{1}{h} + \frac{1}{q_e}t. \quad (6)$$

In order to distinguish the kinetics equation based on the concentration of a solution from the sorption capacity of solids, this second-order rate equation has been called a pseudo-second-order one since it was represented. The pseudo-second-order rate constants can be determined experimentally by plotting *t/q_t* against *t*. Although there are many factors which influence the sorption capacity, including the initial sorbate concentration (Ho et al., 2004; Ho and McKay, 2000, 2003), the reaction temperature (Ho et al., 2004; Ho and McKay, 2000), the solution pH value (Ho et al., 1994, 1995), the sorbent particle size (Ho and McKay, 2003) and dose (Ho and McKay, 2000, 2003), and the nature of the solute (Ho

Table 3 – Experimental conditions

	C ₀ (mg/dm ³)	<i>m_s</i> (g)	<i>d_p</i> (μm)	<i>S</i> (rpm)
Effect of <i>m_s</i> (g)	92.2	5.95, 6.80, 9.44	53–61	300
Effect of <i>d_p</i> (μm)	93.4	6.8	43–53, 74–88, 104–124	300
Effect of <i>S</i> (rpm)	90.7	6.8	53–61	280, 300, 350

Table 4 – Pseudo-second-order kinetic model linear forms

Type	Linear form	Plot	Parameters
Type 1	$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{1}{q_e}t$	<i>t/q_t</i> vs. <i>t</i>	<i>q_e</i> = 1/slope <i>k</i> = slope ² /intercept <i>H</i> = 1/intercept
Type 2	$\frac{1}{q_t} = \left(\frac{1}{kq_e^2}\right)\frac{1}{t} + \frac{1}{q_e}$	1/ <i>q_t</i> vs. 1/ <i>t</i>	<i>q_e</i> = 1/intercept <i>k</i> = intercept ² /slope <i>h</i> = 1/slope
Type 3	$q_t = q_e - \left(\frac{1}{kq_e}\right)\frac{q_t}{t}$	<i>q_t</i> vs. <i>q_t/t</i>	<i>q_e</i> = intercept <i>k</i> = -1/(intercept × slope) <i>h</i> = -intercept/slope
Type 4	$\frac{q_t}{t} = kq_e^2 - kq_e q_t$	<i>q_t/t</i> vs. <i>q_t</i>	<i>q_e</i> = -intercept/slope <i>k</i> = slope ² /intercept <i>h</i> = intercept

et al., 2002; Ho and McKay, 2000), a kinetic model is concerned only with the effect of observable parameters on the overall rate.

In this study, the coefficient of determination, r^2 , was used to test the best-fitting of the kinetic model to the experimental data:

$$r^2 = \frac{\sum(q_m - \bar{q}_t)^2}{\sum(q_m - \bar{q}_t)^2 + \sum(q_m - q_t)^2}, \tag{7}$$

where q_m is amount of cadmium ion on the surface of the tree fern at any time, t , (mg/g) obtained from the second-order kinetic model, q_t , is the amount of cadmium ion on the surface of the tree fern at any time, t , (mg/g) obtained from experiment, and \bar{q}_t is the average of q_t , (mg/g).

3.1. Linear regression method

Linear regressions are frequently used to determine the best-fitting kinetic models, and the method of least squares is used for finding parameters of the kinetic models. However, the pseudo-second-order kinetic model can be linearized as four different types (Table 4), and a simple linear regression will result in different parameter estimates (Kinniburgh, 1986; Longhinotti et al., 1998; Ho, 2004). The most popular linear form used is type 1.

Figures 1–4 show experimental data with linear equations of the four pseudo-second-order kinetic models obtained by using the linear method for the sorption of cadmium onto tree fern at various tree fern doses. Similar patterns were obtained for the effect of tree fern particle size and agitation

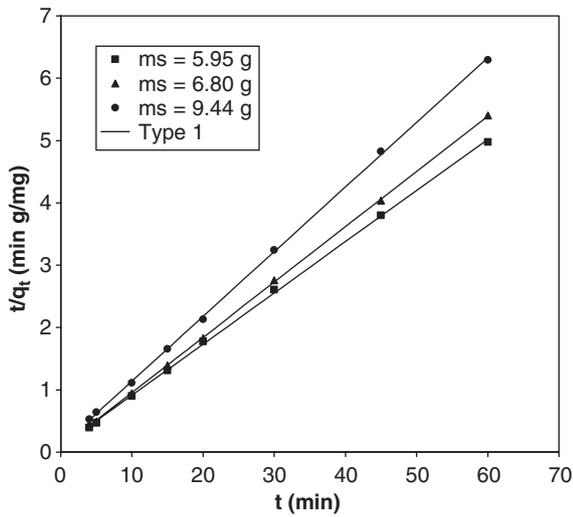


Fig. 1 – Type-1 pseudo-second-order linear equations obtained by using the linear method for the sorption of cadmium onto tree fern at various doses.

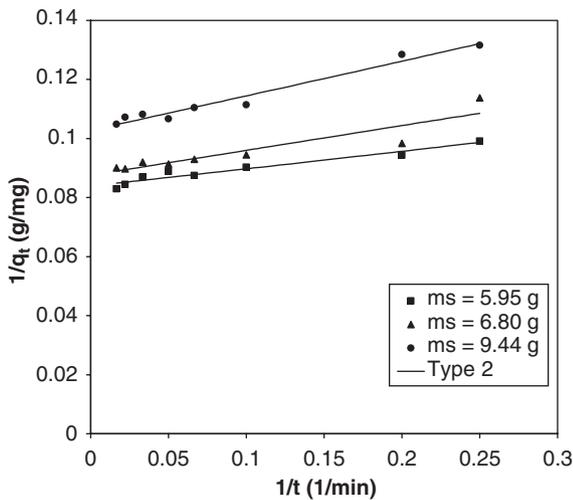


Fig. 2 – Type-2 pseudo-second-order linear equations obtained by using the linear method for the sorption of cadmium onto tree fern at various doses.

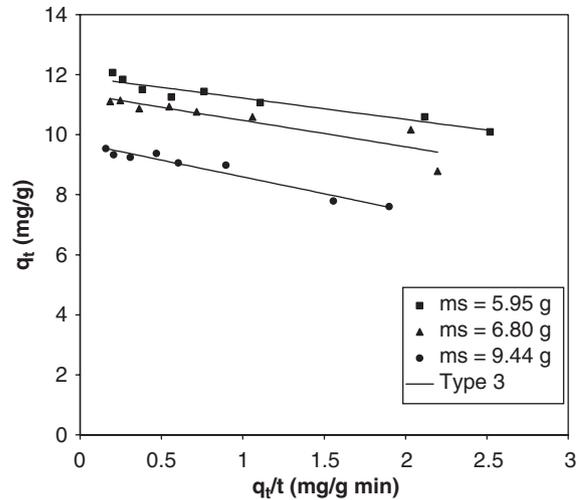


Fig. 3 – Type-3 pseudo-second-order linear equations obtained by using the linear method for the sorption of cadmium onto tree fern at various doses.

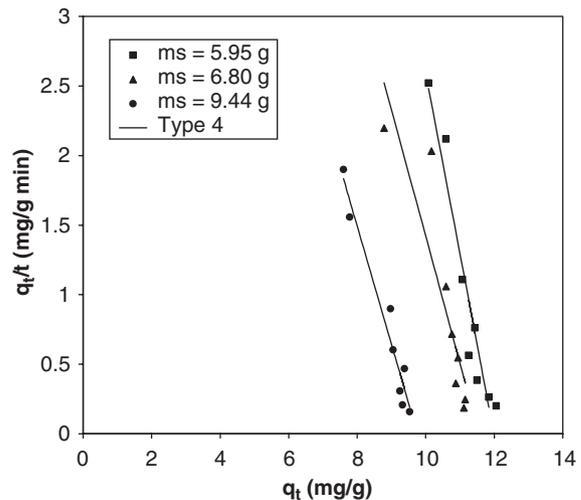


Fig. 4 – Type-4 pseudo-second-order linear equations obtained by using the linear method for the sorption of cadmium onto tree fern at various.

speed. Values of the pseudo-second-order kinetic model constant, k , the amount of cadmium ion sorbed at equilibrium, q_e , and the initial sorption rate, h , are listed in Tables 5–7 for the sorption of cadmium ion onto tree fern for different sorption conditions. These values of the extremely high coefficient of determinations, r^2 , obtained from type 1, indicate that there is strong positive evidence that the cadmium sorption onto tree fern follows the pseudo-second-order kinetic expression. It is clear that transformations of non-linear pseudo-second-order kinetic models to linear forms implicitly alter their error structure and may also violate the error variance and normality assumptions of standard least-squares method (Kinniburgh, 1986; Ho, 2004). In a linear analysis, different linear forms of the same model would significantly effect calculations of the parameters. Figure 5 shows the typical results for the sorption of cadmium

ions onto tree fern. The pseudo-second-order rate equations were obtained using the linear method at a temperature of 25 °C, an initial cadmium concentration of 93.4 mg/dm³, a tree fern dose of 6.8 g, a particle size of 104–124 μm, and an agitation speed of 300 rpm. The four kinetic curves in Fig. 5 almost overlap. However, constants q_e , k , and h as well as the coefficient of determination of type 1 to type 4 expressions differed. The best fit was obtained by using the type 1 expression because the highest coefficient of determinations was calculated from the fitted equation.

3.2. Non-linear method

In the case of the non-linear method, a trial-and-error procedure, which is applicable to computer operation, was developed to determine the pseudo-second-order rate

Table 5 – Pseudo-second-order kinetic parameters obtained by using the linear methods at different tree fern doses

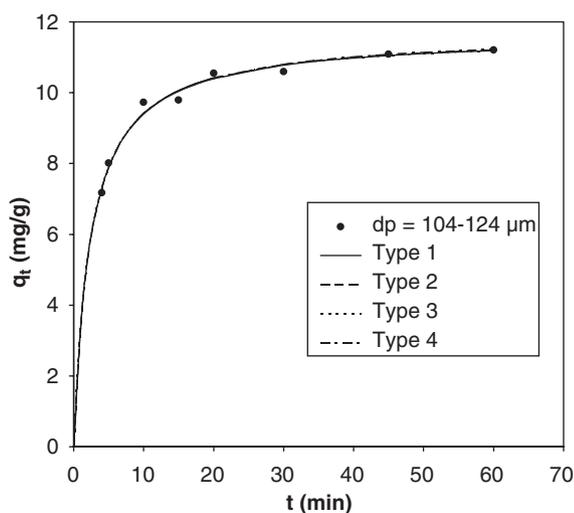
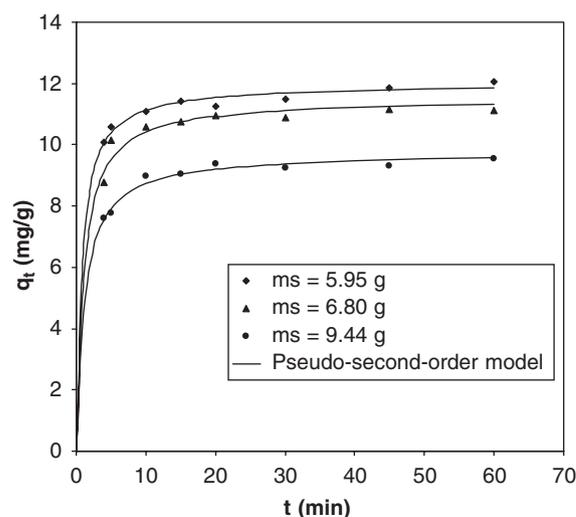
Type	Parameters	5.95 g	6.80 g	9.44 g
Type 1	q_e (mg/g)	12.2	11.3	9.64
	k (g/mg min)	0.0752	0.117	0.105
	h (mg/g min)	11.1	14.8	9.71
	r^2	1.000	1.000	1.000
Type 2	q_e (mg/g)	11.9	11.4	9.73
	k (g/mg min)	0.119	0.0914	0.0901
	h (mg/g min)	16.9	11.9	8.53
	r^2	0.939	0.844	0.968
Type 3	q_e (mg/g)	11.9	11.4	9.72
	k (g/mg min)	0.118	0.0998	0.0915
	h (mg/g min)	16.9	12.9	8.65
	r^2	0.923	0.805	0.953
Type 4	q_e (mg/g)	12.0	11.6	9.76
	k (g/mg min)	9.19	12.7	11.5
	h (mg/g min)	15.6	10.5	8.28
	r^2	0.923	0.805	0.953

Table 6 – Pseudo-second-order kinetic parameters obtained by using the linear methods at different tree fern particle sizes

Type	Parameters	104–124 μm	74–88 μm	43–53 μm
Type 1	q_e (mg/g)	11.6	11.8	12.4
	k (g/mg min)	0.0362	0.0622	0.0697
	h (mg/g min)	4.90	8.71	10.7
	r^2	1.000	1.000	1.000
Type 2	q_e (mg/g)	11.7	11.6	12.1
	k (g/mg min)	0.0358	0.094	0.111
	h (mg/g min)	4.87	12.5	16.1
	r^2	0.985	0.900	0.940
Type 3	q_e (mg/g)	11.6	11.6	12.1
	k (g/mg min)	0.0366	0.095	0.109
	h (mg/g min)	4.95	12.6	16.0
	r^2	0.974	0.877	0.926
Type 4	q_e (mg/g)	11.7	11.7	12.2
	k (g/mg min)	28.1	12.2	9.92
	h (mg/g min)	4.84	11.2	14.9
	r^2	0.974	0.877	0.926

Table 7 – Pseudo-second-order kinetic parameters obtained by using the linear methods at different agitation speeds

Type	Parameters	280 rpm	300 rpm	350 rpm
Type 1	q_e (mg/g)	11.3	11.7	11.6
	k (g/mg min)	0.0416	0.0436	0.0562
	h (mg/g min)	5.32	5.99	7.57
	r^2	1.000	0.999	1.000
Type 2	q_e (mg/g)	11.3	11.3	11.2
	k (g/mg min)	0.0431	0.073	0.0943
	h (mg/g min)	5.47	9.23	11.9
	r^2	0.981	0.900	0.862
Type 3	q_e (mg/g)	11.3	11.3	11.2
	k (g/mg min)	0.0436	0.0712	0.0926
	h (mg/g min)	5.53	9.07	11.7
	r^2	0.973	0.876	0.836
Type 4	q_e (mg/g)	11.3	11.4	11.4
	k (g/mg min)	23.6	16.2	13.1
	h (mg/g min)	5.40	8.05	9.93
	r^2	0.973	0.876	0.836

**Fig. 5 – Four second-order rate equations obtained by using the linear method for the sorption of cadmium onto tree fern.****Fig. 6 – Second-order rate equations obtained by using the non-linear method for the sorption of cadmium onto tree fern at various doses.**

parameters by optimization routine to maximize the coefficient of determination between experimental data and pseudo-second-order model using the solver add-in with Microsoft's spreadsheet, Microsoft Excel. The abilities of four linear pseudo-second-order equations to model the kinetic sorption data were examined. In all cases, the results from the four pseudo-second-order linear equations were the same. Table 8 shows the pseudo-second-order rate parameters obtained by using the non-linear method. By using the non-linear method, there is no problem with transformations of the non-linear pseudo-second-order rate equation to linear forms, and also they had contained the same error structures. Inversely, the linear regression produced very different outcomes. Unlike the linear method, different linear forms significantly values effect r^2 and impact the final

determination of parameters, where non-linear methods would be able to avoid such errors.

Figure 6 shows experimental data with the pseudo-second-order kinetic model obtained by using the non-linear method for the sorption of cadmium using tree fern at various tree fern doses. From the non-line method results in Table 8, the particle size, d_p , the dose of tree fern, m_s , and the agitation speed, S , were found to influence the sorption system. Values of q_e increased with increasing m_s but with decreasing d_p . This is to be expected because increasing the tree fern dose and decreasing the tree fern particle size provide greater a surface area (or a greater number of sorption sites). In addition, changing agitation speeds for the sorption system does not alter the surface area of the tree fern. It is clear that the equilibrium sorption of cadmium ion is a function of d_p

Table 8 – pseudo-second-order rate parameters obtained using the non-linear methods for different effects

Parameters	m_s (g)			d_p (μm)			S (rpm)		
	5.95	6.80	9.44	104–124	74–88	43–53	280	300	350
q_e (mg/g)	12.0	11.5	9.75	11.7	11.7	12.2	11.3	11.5	11.4
k (g/mg min)	0.107	0.0821	0.0879	0.0360	0.0806	0.0986	0.0422	0.0583	0.0724
h (mg/g min)	15.4	10.9	8.36	4.89	11.0	14.6	5.39	7.69	9.49
r^2	0.934	0.875	0.962	0.983	0.911	0.938	0.985	0.911	0.881

and m_s but not agitation speed. The values of h increased with increasing S but decreasing m_s and d_p . It was found that increasing the agitation speed enhanced the particle movement, while decreasing the tree fern particle size increased the rate of cadmium sorption. Moreover, decreasing the tree fern dose enhanced the initial sorption rate, a result counter to that of the sorption of lead by peat (Ho and McKay, 2000). The values of k increased with increasing S and decreasing d_p . However, there was no clear trend for changes in the reaction constants when tree fern doses were changed in this study.

4. Conclusions

- It is not appropriate to use the coefficient of determination of linear regression method for comparing the best-fitting of kinetic models.
- Non-linear method could be a better way to obtain the kinetic parameters.
- The results show sorption of cadmium onto tree fern during agitation by suspended shaking; a pseudo-second-order model can describe the sorption system.
- The sorption equilibrium capacity, the sorption rate constant, and the initial sorption rate are function of the tree fern dose, the particle size, and the agitation speed.

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