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Kinetic Analysis of the Adsorption of lead (II) onto Activated Carbon from *Tridax procumbens*

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ABSTRACT

Adults who are exposed to high levels of lead may develop hypertension and kidney damage. Lead exposure is dangerous for everyone, but it can have devastating effects on pregnant women and their babies. Methods such as membrane separation, ion exchange, precipitation, and biosorption are currently in use for the removal of lead pollution. Biosorption has the fewest negative aspects of these technologies due to its low operating costs, high efficiency at detoxifying low concentrations of toxicants, and small volume of disposal materials. The biosorption of the biosorption of lead (II) onto the activated carbon from *Tridax procumbens* is remodeled using nonlinear regression and the optimal mode was determined by a series of error function assessments. The best kinetic model for adsorption of lead (II) was Pseudo-1st order with a reasonable difference in terms of corrected Akaike Information Criterion to the next best model, which was pseudo-2nd order, and followed by the Elovich. However, the error function analyses especially the AICc was not conclusive in ranking the pseudo-1st order model as the best model due to the low (<5) absolute values of differences between the model. The pseudo-1st order kinetic constants obtained were q_e (mg/g) of 6.181 (95% confidence interval from 5.009 to 7.352) and k_1 (per min) of 0.007 (95% confidence interval from 0.004 to 0.009). Nonlinear modeling enables the determination of a 95 percent confidence interval for the uncertainty range, which can be used in model comparison and discriminant analysis.

INTRODUCTION

Heavy metals are increasingly being released into the environment as a byproduct of mining, electroplating, alloy preparation, pulp-paper, and fertilizer production, among other industrial processes. Heavy metal pollution has become a pressing issue because of the widespread accumulation of these elements in the food chain and the serious health concerns they cause to living organisms. Lead is a naturally occurring dangerous element that can be found all over the Earth's crust. Many countries' public health has suffered as a result of its pervasive use, which has polluted large swaths of land and exposed countless individuals.

Major sources of environmental pollution include the mining, smelting, manufacturing, and recycling industries, as well as the persistent use of lead-based paint and aviation fuel in some countries. More than 75% of all lead is used in the creation of lead-acid car batteries. Pigments, paints, solder, stained glass, lead crystal glassware, ammunition, ceramic glazes, jewelry, toys, cosmetics, and traditional remedies are just some of the many other products that use lead. Drinking water may contain lead if the pipes it travels through are made of lead or if the pipes themselves were joined using lead-based solder. Currently, recycled lead accounts for the vast majority of lead used in international trade. Lead is especially dangerous for young children because of the lasting, devastating effects it can have on their health, particularly on the development of their brain and

nervous system. Adults who are exposed to high levels of lead may develop hypertension and kidney damage. Lead exposure is dangerous for everyone, but it can have devastating effects on pregnant women and their babies. Anemia, hepatitis, encephalopathy, and nephritic syndrome are all conditions associated with lead levels in drinking water that are above the legal limit of 0.05 mg/L. As a result of its high biosorption capacity and selectivity, low cost, and low environmental risk, biosorption is a promising approach for the removal of heavy metals from wastewater.

To fully grasp the biosorption process of toxicants, it is crucial to correctly assign the kinetics and isotherms of biosorption. Estimating uncertainty of the parameters of the kinetics, which are often displayed as a 95 percent confidence interval range, can be made more challenging by the linearization of a clearly nonlinear curve, which can cause problems on the error structure of the data [1]. During the process of data transformation required for linearization, error in the independent variable may also be introduced. Weighting of data points can also affect the fitted parameter values for the linear and nonlinear versions of the model [2]. In this study the published data from the biosorption of lead (II) onto the activated carbon from *Tridax procumbens* [3] is remodeled using nonlinear regression of several kinetic models (**Table 1)** and in the end, a series of error function evaluations identified the best possible setting. Because a linearized modeling version was proposed for the kinetics in the aforementioned publication, this modeling analysis was required.

Table 1. Kinetic models and equation utilized in this study.

Model	Equation	Ref
Pseudo-1st order	$q_t = q_e(1 - e^{-K_1 t})$	[4]
Pseudo-2nd order	$q_t = \frac{K_2 {q_e}^2 t}{(1 + K_2 q_e t)}$	[5]
h value	$h = K_2 {q_e}^2$	[5]
Elovich	$q_t = \frac{1}{\beta ln\alpha\beta} + \frac{1}{\beta lnt}$	[6]

METHODS

Data acquisition and fitting

Data from **Figure 3** from a published work [3] were digitized using the software Webplotdigitizer 2.5 [7].The accuracy of data digitized with this program has been verified [8,9]. The data were first converted to q_t values and then nonlinearly regressed using the curve-fitting software CurveExpert Professional software (Version 1.6).

Statistical analysis

Commonly used statistical discriminatory methods such as corrected AICc (Akaike Information Criterion), Bayesian Information Criterion (BIC), Hannan and Quinn's Criterion (HQ), Root-Mean-Square Error (RMSE), bias factor (BF), accuracy factor (AF) and adjusted coefficient of determination (R^2). The RMSE was calculated according to **Eq. (1)**, [1], and smaller number of parameters is expected to give a smaller RMSE values. *n* is the number of experimental data, Ob_i and Pd_i are the experimental and predicted data while *p* is the number of parameters.

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}(Pd_i - Ob_i)^2}{n - p}} \qquad \text{(Eqn. 1)}$$

As R^2 or the coefficient of determination ignores the number of parameters in a model, the adjusted R^2 is utilized to overcome this issue. In the equation (**Eqns. 2** and **3**), the total variance of the y-variable is denoted by S_y^2 while RMS is the Residual Mean Square.

$$Adjusted\left(R^2\right) = 1 - \frac{RMS}{s_Y^2} \qquad \text{(Eqn. 2)}$$

$$Adjusted\left(R^2\right) = 1 - \frac{(1 - R^2)(n-1)}{(n - p - 1)} \qquad \text{(Eqn. 3)}$$

When it comes to evaluating data, the Akaike Information Criterion (AIC) uses principles from the field of information theory. It strikes a balance between a model's complexity and its goodness of fit [10]. To handle data having a high number of parameters or a smaller number of values corrected Akaike information criterion (AICc) is utilized [11]. The AICc is calculated as follows (**Eqn. 4**), where *p* signifies the quantity of parameters and *n* signify the quantity of data points. A model with a smaller value of AICc is deemed likely more correct [11].

$$AICc = 2p + n\ln\left(\frac{RSS}{n}\right) + 2(p+1) + \frac{2(p+1)(p+2)}{n - p - 2} \qquad \text{(Eqn. 4)}$$

Aside from AICc, Bayesian Information Criterion (BIC) (**Eqn. 5**) is another statistical method that is based on information theory. This error function penalizes the number of parameters more strongly than AIC [12].

$$BIC = n.\ln\frac{RSS}{n} + k.\ln(n) \qquad \text{(Eqn. 5)}$$

A further error function method based on the information theory is the Hannan–Quinn information criterion (HQC) (**Eqn. 6**). The HQC is strongly consistent unlike AIC due to the ln ln *n* term in the equation [11];

$$HQC = n \times ln\frac{RSS}{n} + 2 \times k \times ln(\ln n) \qquad \text{(Eqn. 6)}$$

Further error function analysis that originates from the work of Ross [13] are the Accuracy Factor (AF) and Bias Factor (BF). These error functions test the statistical evaluation of models for the goodness-of-fit but do not penalize for number of parameter (**Eqns. 7 and 8**).

$$\text{Bias factor} = 10^{\left(\sum_{i=1}^{n}\log\frac{(Pd_i/Ob_i)}{n}\right)} \qquad \text{(Eqn. 7)}$$

$$\text{Accuracy factor} = 10^{\left(\sum_{i=1}^{n}\log\frac{|(Pd_i/Ob_i)|}{n}\right)} \qquad \text{(Eqn. 8)}$$

RESULTS AND DISCUSSION

The absorption kinetics data of biosorption isotherm experiment from a published work [3] on the biosorption of lead (II) on the activated carbon from *Tridax procumbens* were analyzed using three models—pseudo-1st, pseudo-2nd and Elovich, and fitted using non-linear regression. The Elovich model was the poorest in fitting the curve based on visual observation (**Figs. 1-6**).

The optimal model was determined through the application of statistical analysis based on the root-mean-square error (RMSE), adjusted coefficient of determination (adjR2), accuracy factor (AF), bias factor (BF), Bayesian Information Criterion (BIC), corrected Akaike Information Criterion (AICc), and Hannan–Quinn information criterion (HQC).

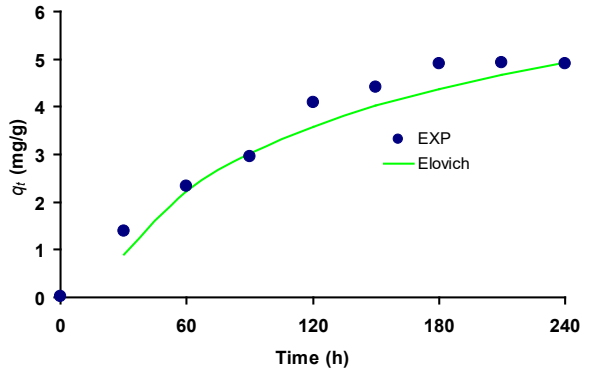

Fig. 1. Kinetics of on the biosorption of lead (II) onto activated carbon from *Tridax procumbens* modelled using the Elovich model.

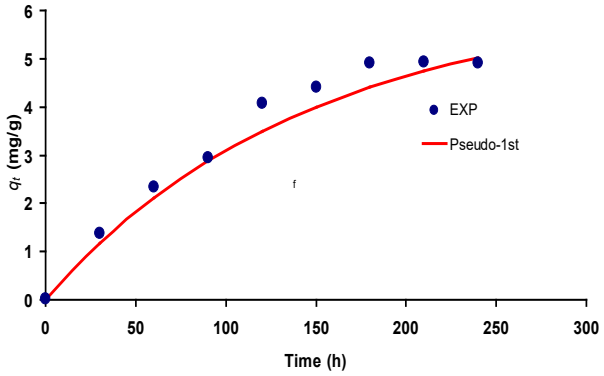

Fig. 2. Kinetics of on the biosorption of lead (II) onto activated carbon from *Tridax procumbens* modelled using the pseudo-1st order model.

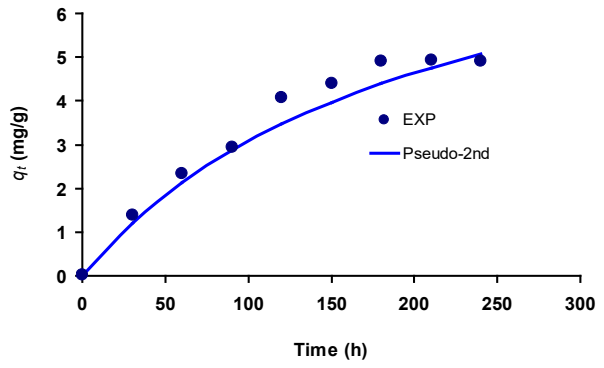

Fig. 3. Kinetics of on the biosorption of lead (II) onto activated carbon from *Tridax procumbens* modelled using the pseudo-2nd order model.

The best kinetic model for adsorption of lead (II) was Pseudo-1st ordernwith a reasonable difference in terms of corrected Akaike Information Criterion to the next best model, which was pseudo-2nd order, and followed by the Elovich (**Table 2**). The error function analyses especially the AICc was not conclusive in ranking the pseudo-1st order model as the best model due to the low (<5) absolute values of differences between the model. The kinetic constants for all of the models are shown in **Table 3**. The pseudo-1st order kinetic constants obtained were q_e (mg/g) of 6.181 (95% confidence interval from 5.009 to 7.352) and k_1 (per min) of 0.007 (95% confidence interval from 0.004 to 0.009). The h value, (mg/g.min) utilized to calculate the initial adsorption rate constant indicates the driving force to accelerate the diffusion of adsorbate from solution onto the adsorbent [14].

Table 2. Error function analysis for the kinetic models.

Model	p	RMSE	R^2	adR^2	AICc	BIC	HQC	AF	BF
Pseudo-1st order	2	0.360	0.961	0.948	-5.85	-16.25	-17.50	1.083	0.927
Pseudo-2nd order	2	0.376	0.958	0.944	-5.06	-15.47	-16.71	1.083	0.929
Elovich	2	0.381	0.957	0.943	-4.85	-15.25	-16.50	1.106	0.910

Note:
RMSE Root mean Square Error
p no of parameters
adR^2 Adjusted Coefficient of determination
BF Bias factor
AF Accuracy factor
AICc Adjusted Akaike Information Criterion
BIC Bayesian Information Criterion
HQC Hannan–Quinn information criterion

Table 3. Calculated constants for the kinetics models fitting the biosorption of lead (II) onto activated carbon from *Tridax procumbens*.

Model	Value	95% Confidence interval
Pseudo-1st		
k_1 (per min)	0.007	0.004 to 0.009
q_e (mg/g)	6.181	5.009 to 7.352
Pseudo-2nd		
k_2 (g/mg/min)	0.0010	0.0001 to 0.00094
q_e (mg/g)	9.411	6.636436 to 12.186386
h (mg/g.min)	0.046	0.023 to 0.077
Elovich		
Alpha (mg/g.min)	0.103	0.0835 to 0.122
Beta (g/mg)	0.514	0.421 to 0.607

Through the use of kinetic models, researchers have been able to analyze experimental data and gain insight into the sorption mechanism and potential rate controlling steps, such as chemical reaction and mass transport processes. These kinetic models incorporated not only the Elovich equation but also the pseudo-1st order and pseudo-2nd order equations. The initial adsorption rate (in mg/g min) and the surface coverage (in g/mg) are given by and respectively in the Elovich model. In the pseudo-1st order reaction, the adsorbate concentration is maintained at a constant saturation value. The adsorbate is adsorbed at a constant rate because its level is independent of the adsorbate concentration.

Under the control of film diffusion, the rate is inversely proportional to the particle size, the distribution coefficient, and the film thickness. The rate-limiting step here is diffusion, which is not concentration or reactant-dependent, so we call it physisorption (physical exchange) [15–19]. If the reaction is governed by a pseudo-2nd order reaction, then chemisorption occurs because a chemical reaction is in charge of the rate-controlling step. When the sorbate to sorbent ratio is small, the sorption kinetics is similar to a reversible second order reaction, while when it is larger, two competitive reversible second order reactions take place [20]. However, additional proofs, such as evaluation results of the activation energies via experiment repetition at different temperatures and checking the process rates dependences to the sizes of the adsorbent particle, should be provided to confirm the mechanism is a chemisorption [21].

The pseudo-1st order model has also been reported to be the best model for lead(II) sorption onto coco-peat biomass [22] and in several other metal sorption works [23–30]. On the other hand,

the pseudo-2nd order kinetics model has been reported to be the best model in several lead sorption studies such as lead(II) sorption by *Cephalosporium aphidicola* [31], on sesame leaf [32] and on *Spirodela polyrhiza* [33] to name a few. In general, the pseudo-2nd order was the best model for metal sorption such as the biosorption of Cr(VI) to magnetic iron oxide nanoparticle-multi-walled carbon nanotube [34], Cu(II) adsorption onto functionalized cellulose beads from Tunisian almond (*Prunus dulcis*) shell [35] and the sorption of Zn(II) by *Streptomyces ciscaucasicus* [36] and other heavy metals [20,24,37–43], indicating that the pseudo-2nd model is routinely reported to be better than the pseudo-1st order in fitting kinetics of xenobiotics sorption although this discrepancy has been attributed to the mathematically versatile properties of the pseudo-2nd order model [2,16,44].

CONCLUSION

In conclusion, the biosorption of the biosorption of lead (II) onto activated carbon from *Tridax procumbens* was successfully modelled using three models—pseudo-1st, pseudo-2nd and Elovich, and fitted using non-linear regression. Pseudo-1st order was the best kinetic model for lead (II) adsorption, with a significant difference in corrected Akaike Information Criterion between it and the next best model, pseudo-2nd order, and then the Elovich. Due to the low (5) absolute values of differences between the models, the error function analyses, especially the AICc, were unable to conclusively rank the pseudo-1st order model as the best model. Modelling using a nonlinear approach allows for the calculation of uncertainty range in terms of 95% confidence interval that would be useful for model comparison and discriminant in future studies.

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