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# Kinetic Analysis of the Adsorption of Chromium onto Calcium Alginate Nanoparticles

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#### ABSTRACT

The widespread use of chromium in industrial applications such as leather tanning, metallurgy, electroplating, and refractory materials has resulted in it being one of the most harmful trace elements to be introduced into surface and ground waters. The sorption isotherm of chromium sorption onto calcium alginate nanoparticles were analyzed using three models—pseudo-1<sup>st</sup>, pseudo-2<sup>nd</sup> and Elovich, and fitted using non-linear regression. The Elovich model was the poorest in fitting the curve based on visual observation followed by the pseudo-1<sup>st</sup> order. Statistical analysis based on root-mean-square error (RMSE), adjusted coefficient of determination (adj $R^2$ ), bias factor (BF), accuracy factor (AF), corrected AICc (Akaike Information Criterion), Bayesian Information Criterion (BIC) and Hannan–Quinn information criterion (HQC) that showed that the pseudo-1ST order model is the best model. Kinetic analysis using the pseudo-1st order model at 400 mg/L 4-BDE gave a value of equilibrium sorption capacity  $q_e$  of 31.89 mg g<sup>-1</sup> (95% confidence interval from 30.37 to 33.42) and a value of the pseudo-1st-order rate constant,  $k_l$  of 0.22 (95% confidence interval from 0.019 to 0.025). Further analysis is needed to provide proof for the chemisorption mechanism usually tied to this kinetic.

### INTRODUCTION

Because of its extensive usage in industrial applications like as leather tanning, metallurgy, electroplating, and refractory chromium is one of the most harmful trace elements introduced into surface waters and ground waters. The rising number of studies published regarding chromium toxicity over the past ten years demonstrates the increasing number of attempts to show and remediate chromium-bearing pollution in the environment. Using standard procedures, such as chemical precipitation, ion exchange, membrane filtration, coagulation/flocculation, and electrochemical treatment, trace elements may be removed from wastewaters [1–5].

These approaches, on the other hand, are inefficient and result in enormous amounts of waste production. Sorption technologies [6] are being considered as alternatives for the treatment of water polluted by metals. As the name implies, sorption is the process by which sorbate is transferred from its liquid phase to the surface of a sorbent. Different parameters, such as pH, temperature, the kind and quantity of sorbent used, starting metal concentration, ionic strength, as well as the presence of other pollutants, influence sorption efficacy. Physical (physiosorption) and/or chemical (chemisorption) interactions are formed between the sorbent and the sorbate in response to the attraction forces between the two. Whereas in the physiosorption process, the sorbate attaches to the sorbent surface by weak forces such as van der Waals forces [6–10].

The removal of trace elements from freshwater can be accomplished using a variety of sorbents, including nanomaterials with various forms of coating and chemical synthesis methods, as well as other approaches [11–16]. Nanomaterials, which are defined as materials and structures with a minimum size of 1-100 nm and a minimum thickness of 1-100 nm, exhibit a wide range of mechanical, optical, magnetic, and chemical characteristics when compared to particles and macroscopic surfaces with a comparable thickness. These properties are highly reliant on the form, size, surface features, and internal structure of the particle, and they are distinguishable from those of macroscopic surfaces and particles of equivalent composition in that they are not dependent on these factors.

Moreover, the physical-chemical features of the fluid in which nanoparticles are absorbed or desorption are dependent on the sorption processes of nanomaterials, which should be considered while designing nanomaterials [17-19]. Whenever nanomaterials are used as sorbents for the removal of toxic elements from wastewater, they must meet a number of requirements. These requirements include being nontoxic, having high sorption capacities, being selective to low concentrations of contaminants, being simple to remove the sorbed contaminant from their surface, and being recyclable. This question has been investigated whether harmful trace elements can be removed from aqueous solutions by using a variety of nanomaterials, including carbon nanotubes and carbon-based material composites such as graphene, nanometals or metal oxides, and polymeric sorbents. To date, all of these nanomaterials have met these requirements, with the exception of carbon nanotubes and carbon-based material composite materials such as graphene [20-23]. In a previous study, the sorption of chromium on calcium alginate nanoparticles was studied using linearized kinetic models which disrupt the error structure of the data and hindered efficient inference and comparison with current biosorption data that have begun to capitalize on the computing power that allow nonlinear regression to be carried out at ease.

Correct assignment of the kinetics and isotherms of biosorption is critical for understanding the mechanism of biosorption. This is especially true for understanding the mechanism of biosorption. The use of linearization to smooth out a clearly nonlinear curve causes the error structure of the data to be disrupted. This makes estimating the uncertainty of the parameters of the kinetics, which is generally provided in the form of a 95 percent confidence interval range, much more challenging [24]. Aside from that, the linearization procedure results in the introduction of error into the independent variable as well. Additionally, changes in the weights assigned to each data point may occur, which typically results in discrepancies in the fit parameter values between both the linear and nonlinear versions of the kinetics model, depending on the data set [25]. Thus, the aim of this study is to remodel the data using nonlinear regression.

#### **METHODS**

#### Data acquisition and fitting

Data from Figure 4 from a published work [26] were digitized using the software Webplotdigitizer 2.5 [27]. The data were then nonlinearly regressed using the curve-fitting software CurveExpert Professional software (Version 1.6). Digitization using this software has been acknowledged for its reliability [28,29]. The data were then nonlinearly regressed using the curve-fitting software CurveExpert Professional software (Version 1.6) using several models (**Table 1**).

Table 1. Kinetic models utilized in this study.

Model	Equation	Reference
Pseudo-1st order	$q_t = q_e (1 - e^{-K_{1t}})$	[30]
Pseudo-2nd order	$K_2 q_e^2 t$	[31]
Elovich	$q_t = \frac{1}{(1 + K_2 q_e t)}$ $q_t = \frac{1}{\beta \ln \alpha \beta} + \frac{1}{\beta \ln t}$	[32]

#### Statistical analysis

A battery of statistical discriminatory tests 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  $(R^2)$  were utilized in this work.

The RMSE was calculated according to Eq. (1), [24], 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.

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

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

The AICc is calculated as follows (**Eqn. 4**), where p signifies the quantity of parameters and n signify the quantity of data points. To handle data having a high number of parameters or a smaller number of values corrected Akaike information criterion (AICc) is utilized [33]. A model with a smaller value of AICc is deemed likely more correct [33]. The Akaike Information Criterion (AIC) is based on the information theory. It balances between the goodness of fit of a particular model and the complexity of a model [34].

$$AICc=2p+n\ln\left(\frac{RSS}{n}\right)+2(p+1)+\frac{2(p+1)(p+2)}{n-p-2}$$
 (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 [35].

$$BIC = n.\ln\frac{RSS}{n} + k.\ln(n)$$
 (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 [33];

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

Further error function analysis that originates from the work of Ross [36] 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**).

Bias factor = 
$$10^{\left(\sum_{i=1}^{n} \log \frac{(Pd_i / Ob_i)}{n}\right)}$$
 (Eqn. 7)  
Accuracy factor =  $10^{\left(\sum_{i=1}^{n} \log \frac{|(Pd_i / Ob_i)|}{n}\right)}$  (Eqn. 8)

Another error function analysis is the evidence ratio regarding the difference between the two lowest AICc values (Eqn. 9), where if it is the same, then each model will have an equal chance of being true.

If the difference in AICc scores is 6.0, model A has a 95% chance of being correct, making it 20 (95/5) times more likely than model B to be correct [24].

$$P_A = \frac{e^{0.5\Delta}}{1 + e^{0.5\Delta}} \tag{Eqn. 9}$$

#### **RESULTS AND DISCUSSION**

It is possible that a lack of chromium in the environment will have negative repercussions for the metabolism of both plants and animals. Chromium pollution from industrial sources, on the other hand, is becoming more significant in recent years. Chromium contamination in wastewater may be caused by a variety of sources, including the dye and pigment industries, wood preservation, electroplating, and leather tanning, among others. Chromium tanning processes are used in more than 80 percent of the country's tanneries, according to official figures. Approximately the vast majority of these facilities, according to current estimates, release untreated sewage into the environment. New chemicals are now being released into the environment as a consequence of current industrial activity patterns, which is interfering with the usual flow of materials in the ecosystem [37–41].

As defined by the International Atomic Energy Agency (IAEA), heavy metals are defined as metals having a density more than or equal to 5 g/mL and are classed as such. Heavy metal pollution of water is a significant concern in today's society, and it is a worldwide issue. According to the findings, the absorption of heavy metals by organisms is more reliant on the concentration of free metal ions in solution than on the concentration of total metal in solution, which was previously established. Since improved correlations between metal absorption and the concentration of free metal ions or labile metals have been established, the bioavailability and toxicity of heavy metals are now primarily governed by the number of heavy metals present in free metal ions, rather than the amount of heavy metals present in free metal ions. Because of their nonbiodegradability and high quantities of heavy metals, they are hazardous to human health and should be avoided.

World Health Organization (WHO) has recognised a handful of metals as the most important threats to human health in the contemporary period, according to the organisation. Cadmium, chromium, cobalt, copper, lead, nickel, mercury, zinc, arsenic, and tin are just a few of the heavy metals that may be found in the environment. In terms of neurotoxicity, the three most dangerous metals are mercury, cadmium, and lead, which are the most toxic of the three [42–46].

The absorption kinetics data were analyzed using three models—pseudo-1<sup>st</sup>, pseudo-2<sup>nd</sup> and Elovich, and fitted using non-linear regression. The Elovich model was the poorest in fitting the curve based on visual observation followed by Pseudo-2nd order (**Figs. 1-3**). Statistical analysis based on root-mean-square error (RMSE), adjusted coefficient of determination  $(adjR^2)$ , bias factor (BF), accuracy factor (AF), corrected AICc (Akaike Information Criterion), Bayesian Information Criterion (BIC) and Hannan–Quinn information criterion (HQC) that showed that the pseudo-first-order model is the best (**Table 2**). The calculated evidence ratio was 133 with an AICc probability value of 0.99 indicating that the best model was at least 133 times better than the nearest best model, which was pseudo-1<sup>st</sup>. Further analysis is needed to provide proof for the mechanism usually

tied to this kinetic. The sorption of chromium at various concentrations on calcium alginate nanoparticles was then fitted using the pseudo-1<sup>st</sup> model (**Fig. 4**).



Fig. 1. Kinetics of the sorption of chromium on calcium alginate nanoparticles as modelled using the Elovich model.



Fig. 2. Kinetics of the sorption of chromium on calcium alginate nanoparticles as modelled using the pseudo-1<sup>st</sup> order model.



**Fig. 3.** Kinetics of the sorption of chromium on calcium alginate nanoparticles as modelled using the pseudo-2<sup>nd</sup> order model.

Table 2. Error function analysis of regressed models.

Mode	el		p	RMSE	$adR^2$	AICc	BIC	HOC	AF	BF
Pseudo-1st order		2	0.541	0.995	1 49	-8.91	-10.16	1.018	0 999	
Pseudo-2nd order		2	0.932	0.986	11.28	0.87	-0.38	1.010	1 009	
Flovich		2	3 548	0 749	35 33	24.93	23.68	1 166	1.065	
Note:		-	01010	01712	00.00	21190	20.00		11000	
RMSE	Roo	t mean Sq	uare E	Error						
p	no c	of paramete	ers							
adR <sup>2</sup>	R <sup>2</sup> Adjusted Coefficient of determination									
AF	Acc	uracy facto	or							
AICc	Adj	usted Akai	ke In	formation	Criterio	1				
BIC	Bay	esian Info	rmatic	on Criterio	on					
HQC	Han	nan–Quin	n info	rmation c	riterion					
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Fig. 4. Fitting of the chromium sorption data at various concentrations onto calcium alginate nanoparticles using the pseudo-1<sup>st</sup> model (blue curve).

Using kinetic models to examine experimental data, researchers have been able to better understand the mechanism of sorption as well as probable rate-controlling phases, such as chemical reactions and mass transport mechanisms. Incorporating the pseudo-1st order equation, the pseudo-2nd order equation, and the Elovich equation, these kinetic models yielded the best results. It is necessary to achieve saturation of both the adsorbate concentration in the pseudo first-order process before it may proceed. The outcome is that the level of the adsorbate remains constant, and the adsorbate is adsorbed at a constant rate since the rate is reliant on a single concentration of the adsorbate is maintained constant. When the rate is controlled through film diffusion, there is an inverse connection between the rate and the particle size, the distribution coefficient, and the thickness of the film. In this case, the label physisorption is used since the rate-limiting phase is diffusion, which is independent of the concentrations of both reactants in the solution (physical exchange) [47-50].

When the reaction is governed by a pseudo second order reaction, the chemical reaction governs the rate-controlling step, and when this occurs, the process is referred to as chemisorption (chemical absorption). When this occurs, the sorption kinetics corresponds to a reversible second order reaction at low adsorbate/adsorbent ratios, and when this occurs at larger adsorbate/adsorbent ratios, two competing reversible second order reactions take place [51]. Although chemisorption has been demonstrated, additional evidence must be provided to support this conclusion, such as the evaluation of activation energies obtained by repeating the experiment at various temperatures, as well as the examination of process rates and their relationships with adsorbent particle size and size dependence [52]. The pseudo-1<sup>st</sup> order model has been called the Lagergren model [53] in honour of its developer [30]. A pseudo-1st order reaction suggests that physisorption is the primary mechanism involved rather than chemisorption in most cases. Adsorption in which the forces involved are intermolecular forces (van der Waals forces) is referred to as physisorption (also known as physical adsorption). Even though the model has been successfully used to describe the process of adsorption, numerous academics have voiced reservations about it in using either the pseudo-1<sup>st</sup> (PFO) or pseudo-2<sup>nd</sup> models (PSO) in explaining the mechanism of adsorption [25,54,55]. In the original published work [26], the authors did not report whether the pseudo-1<sup>st</sup> order or pseudo-2<sup>nd</sup> order is the best model.

PFO models have been used in investigations on the removal of copper (II) from water and effluent from the copper plating industry by adsorption onto peanut shell carbon, among other things [56], the use of spent black tea for the removal of nitrobenzene from aqueous media [57], carbon beads- Caalginate-activated for the removal of patulin from apple juice [58], adsorption of the reactive red 141 dye using sesame waste [59], adsorption kinetics of Acid Red on activated carbon from acrylic fibrous waste [60] an sorption of basic dyes onto water hyacinth roots [61].

## CONCLUSION

In conclusion, the chromium sorption data at various concentrations onto calcium alginate nanoparticles was successfully modelled using three models-pseudo-1st, pseudo-2<sup>nd</sup> and Elovich, and fitted using non-linear regression. Statistical analysis based on root-mean-square error (RMSE), adjusted coefficient of determination  $(adjR^2)$ , bias factor (BF), accuracy factor (AF), corrected AICc (Akaike Information Criterion), Bayesian Information Criterion (BIC) and Hannan-Quinn information criterion (HQC) showed that the pseudo-1st order model is the best model giving valuable parameters such as the equilibrium sorption capacity  $q_e$  and the pseudo-second-order rate constant,  $k_2$ , which can be further utilized in isothermal modelling analysis. Kinetic analysis using the pseudo-1st order model at 400 mg/L 4-BDE gave a value of equilibrium sorption capacity  $q_e$  of 31.89 mg g<sup>-1</sup> (95% confidence interval from 30.37 to 33.42) and a value of the pseudo-1st-order rate constant,  $k_1$  of 0.22 (95% confidence interval from 0.019 to 0.025). Further analysis is needed to provide proof for the mechanism usually tied to this kinetic. The nonlinear regression method allows for the parameter values to be represented in the 95% confidence interval range which can better allow comparison with published results.

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