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Kinetic Analysis of the Adsorption of Ethyl Violet onto Graphene Oxide Sheets Integrated with Gold Nanoparticles

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ABSTRACT

An example of biosorption is when the sorbent is made from a biodegradable material. Biosorption is now being seen as a simple, cost-effective, and environmentally acceptable alternative to traditional pollution treatment methods. Bioremediation is one of the branches of bioremediation that is used to minimise pollution in the context of incorrect dye waste disposal. The sorption isotherm of Ethyl Violet onto graphene oxide were analyzed using three models—pseudo-1st, pseudo-2nd 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) that showed that the pseudo-second-order model was the best which was the same finding from the original published work. The calculated evidence ratio was 11 with an AICc probability value of 0.91 indicating that the best model was at least 11 times better than the nearest best model, which was pseudo-1st. Further analysis is needed to provide proof for the mechanism usually tied to this kinetic. Nonlinear regression analysis using the pseudo-2nd order model for the highest concentration tested, which was 10 mM, gave values of equilibrium sorption capacity q_e of 30.928 mg/g (95% confidence interval from 29.328 to 32.527) and a value of the pseudo-2nd-order rate constant, k_2 of 0.020 (95% confidence interval from 0.011 to 0.028).

INTRODUCTION

Dry and wet fibre manufacturing methods are used by the dye, pigment, and textile industries. Among the two processes, sizing and de-sizing, bleaching, dying, and finishing, the wet process is the most critical. There is a significant amount of clean water used in these processes, which results in severely contaminated effluents. The dying process is essential to the transmission of textile goods. Buyers generally search for common product features including great optical fixation, abruptness, and cleanliness both during the initial purchasing process and after long-term use. Fibre dyes must have a high degree of consistency in colour and be resistant to fading as well as being reasonably priced [1–4]. The various stages of advanced dyeing technology are selected for use in supplies based on the origin of the fibre and the properties of the dyes and pigments, such as chemical constitution, group, commercial affordability, adhesive matters consistent with the desired substance to be coloured, and financial considerations. Dyeing and pigmenting are two of the most advanced dyeing methods. Dyes have altered over time but haven't progressed as much as some would have hoped. The process is broken down into three stages: preparation, dying, and finishing. Despite its limited use in the textile sector, the dye and pigment industry has been known to pollute ethyl violet [4–8]. Preparation is the process of removing any unwanted contaminants from the materials before to colouring them.

Alkaline washing and detergents, as well as enzymes, are used in this procedure. It is possible to eliminate the cloth's colour origin using hydrogen peroxide or a chlorine-based cleaning solution. An optical brightening agent is used to enhance the brightness of white materials (Mamun et al., 2017). Some of the most hazardous pollutants to aquatic life and human health are colourants, such as dyes and pigments, which are emitted with industrial effluents and can be poisonous or carcinogenic to both. Colors of industrial effluents can be mutagenic, toxic to aquatic

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life, and carcinogenic to humans even at low levels. Closing the dyeing process is known as textile finishing and is the final stage in the process of creating a finished textile product. Commission finishing and integrated textile enterprises are included in this subsector. Because of the large range of raw materials, production processes, and finished items accessible in the textile finishing industry, it is a diverse industry. An alternative perspective is to think of the process as a series of unit operations that may be combined to create a textile product that is tailored to the needs of its intended audience. Finishing is the process of using chemical chemicals to improve the appearance and feel of a textile. Permanent press procedures such as waterproofing and softening, as well as antistatic treatment, can extend the life of water and stain-resistant textiles [9–12].

Biosorption methods are essential in a variety of biotreatment treatments, both conventional and environmental. Biological materials, such as live or dead bacteria and their components, marine algae, plants, agricultural wastes, and naturally existing inhabitants, are used to extract or recover organic or inorganic stuff from solution. In a previous study, the sorption of Ethyl Violet onto graphene oxide was investigated using linearized kinetic models, which disrupted the error structure of the data and hampered efficient inference and comparison with current biosorption data, which has begun to capitalise on computing power, allowing nonlinear regression to be performed easily.

The correct assignment of biosorption kinetics and isotherms is crucial for understanding the mechanism of biosorption. This is particularly true when it comes to comprehending the process of biosorption. The use of linearization to smooth out an obviously nonlinear curve disrupts the data's error structure. This makes assessing the uncertainty of the kinetic parameters, which is often supplied in the form of a 95 percent confidence interval range, considerably more difficult [13]. Apart from that, the linearization technique introduces error into the independent variable as well. Furthermore, depending on the data set, variations in the weights allocated to each data point may occur, resulting in inconsistencies in the fit parameter values between the linear and nonlinear versions of the kinetics model [14]. 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 [15] were digitized using the software Webplotdigitizer 2.5 [16]. 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 [17,18]. 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	$q_t = q_e(1 - e^{-K_1 t})$	[19]
Pseudo-2nd order	$q_t = \frac{K_2 {q_e}^2 t}{(1 + K_2 q_e t)}$	[20]
Elovich	$q_t = \frac{1}{\beta ln\alpha\beta} + \frac{1}{\beta lnt}$	[21]

Statistical analysis

An extensive array of statistical discriminatory tests, including corrected AICc (Akaike information criterion) and Bayesian Information Criterion (BIC), Hannan and Quinn's Criterion (HQ), root-means square error (RMSE), bias factor, accuracy factors, and adjusted coefficient of determination were used in this study.

The RMSE was calculated according to **Eq. (1)**, [13], 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.

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

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

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 [22].A model with a smaller value of AICc is deemed likely more correct [22]. 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 [23].

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

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 [24].

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

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 [22];

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

Further error function analysis that originates from the work of Ross [25] are the Accuracy Factor (AF) and Bias Factor (BF). These error functions test the statistical evaluation of models for

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the goodness-of-fit but do not penalize for number of parameter (**Eqns. 7 and 8**).

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

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

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 [13].

$$P_A = \frac{e^{0.5\Delta}}{1+e^{0.5\Delta}} \qquad \text{(Eqn. 9)}$$

RESULTS AND DISCUSSION

People have used dyes in their daily lives since 3000 B.C. for a variety of purposes. Some countries, like Egypt and Romania, have thriving dying industries. Color compounds are sometimes referred to as a 'discernible contaminant,' despite their status as minor pollutants. Around 10,000 metric tonnes of commercial dyes and pigments are produced each year around the world, with the United States accounting for more than 70,000 metric tonnes of that total. Rather of dyes, mordants were utilised, which necessitated a lengthy dyeing process. With the invention of synthetic dyes in 1856 by W. H. Perkins, textiles of all hues and colours could be produced quickly and cheaply.

An entire multi-billion-dollar business has developed around "dye application." Synthetic dyes, on the other hand, are becoming less and less popular due to the negative impact they have on all forms of life. Dye has the potential to be toxic and to considerably diminish the effluent value of the effluent value [5]. Natural dye painting demanded a lot of water, which was a lot of water to use. An equal or even greater amount of dye is utilised than the amount of dyed fibre. About 80% of the colourant is left on the fabric after dyeing, while the remaining 20% is flushed away as effluent. Around 22% of all industrial wastewater in the United States is generated in the textile industry, according to that industry [4].

The absorption kinetics data 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 followed by the Pseudo-1st 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-2nd-order model was the best (**Table 2**) which was the same finding from the original published work.

The calculated evidence ratio was 11 with an AICc probability value of 0.91 indicating that the best model was at least 11 times better than the nearest best model, which was pseudo-1st. Further analysis is needed to provide proof for the mechanism usually tied to this kinetic. Nonlinear regression analysis using the pseudo-2nd order model for the highest concentration tested, which was 10 mM, gave values of equilibrium sorption capacity q_e of 30.928 mg/g (95% confidence interval from 29.328 to 32.527) and a value of the pseudo-2nd-order rate constant, k_2 of 0.020 (95% confidence interval from 0.011 to 0.028). The pseudo-2nd order model was then utilized to fit sorption curves of Ethyl Violet at various concentrations onto graphene oxide (**Fig. 4**).

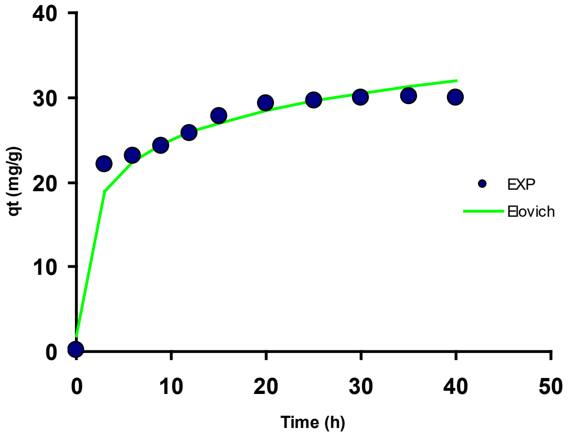

Fig. 1. Kinetics of the sorption of Ethyl Violet onto graphene oxide as modelled using the Elovich model.

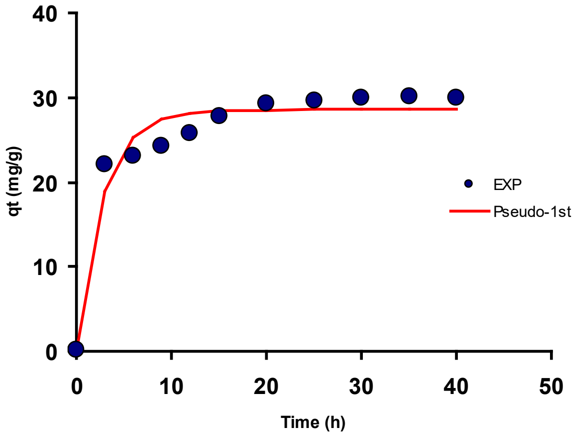

Fig. 2. Kinetics of the sorption of Ethyl Violet onto graphene oxide as modelled using the pseudo-1st order model.

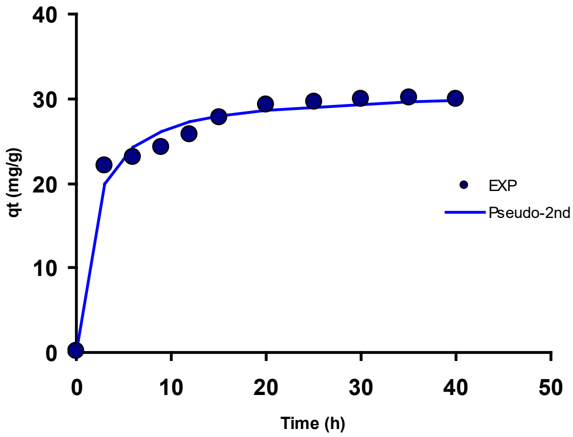

Fig. 3. Kinetics of the sorption of Ethyl Violet onto graphene oxide as modelled using the pseudo-2nd order model.

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Table 2. Error function analysis of regressed models.

Model	p	RMSE	*adR²*	AICc	BIC	HQC	AF	BF
Pseudo-1st order	2	2.076	0.935	27.29	18.65	17.36	1.065	0.998
Pseudo-2nd order	2	1.189	0.978	15.03	6.40	5.10	1.035	0.999
Elovich	2	1.475	0.967	19.78	11.14	9.85	1.029	0.981

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

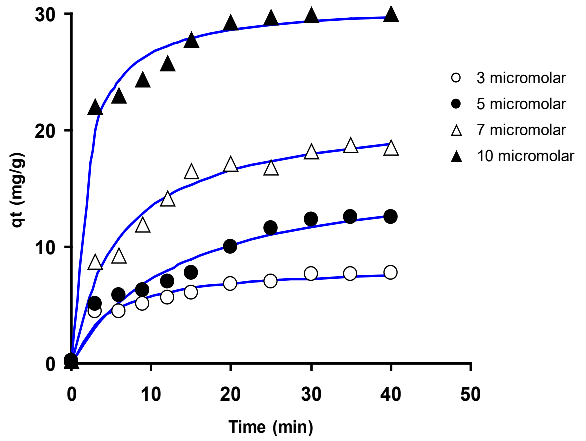

Fig. 4. Fitting of the kinetics of the sorption of Ethyl Violet at various concentrations onto graphene oxide as modelled using the pseudo-2nd order model.

Studying experimental data by employing kinetic models has helped researchers better understand the sorption mechanism as well as possible rate-controlling phases, like chemical reactions and mass transport mechanisms. The best results were obtained using kinetic models that included the pseudo-1st order equation, the pseudo-2nd order equation, and the Elovich equation. The adsorbate concentration must be saturated before the pseudo first-order process may begin. Because of this, a consistent concentration of adsorbate is maintained, resulting in a constant absorption rate. As film diffusion is in charge, the rate is inversely proportional to particle size, distribution coefficient, and film thickness. In the event that both reactants are present in a solution, the rate-limiting phase of diffusion will be called physisorption" (physical exchange) [26–29].

The rate-controlling step is controlled by the chemical reaction when it is driven by a pseudo second order reaction, and this is referred to as chemisorption (chemical absorption). A second-order reaction happens when this occurs at low adsorbate/adsorbent ratio and two competing second-order reactions happen at higher adsorbate/selective sorbent ratios [30]. There are numerous examples where the pseudo-2nd order has been found to fit kinetics data from many dyes sorption works [31–39]. However, additional evidence is needed to support this conclusion, such as the evaluation of activation energies obtained by re-running the experiment at various temperatures, and an examination of procedure rates in regards to adsorbent particle size, as well as their reliance on the adsorbent's particle size [40].

CONCLUSION

In conclusion, the Ethyl Violet onto graphene oxide was successfully modelled using three models—pseudo-1st, pseudo-2nd 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) that showed that the pseudo-2nd-order model was the best which was the same finding from the original published work. The calculated evidence ratio was 11 with an AICc probability value of 0.91 indicating that the best model was at least 11 times better than the nearest best model, which was pseudo-1st. Further analysis is needed to provide proof for the mechanism usually tied to this kinetic. Nonlinear regression analysis using the pseudo-2nd order model for the highest concentration tested, which was 10 mM, gave values of equilibrium sorption capacity q_e of 30.928 mg/g (95% confidence interval from 29.328 to 32.527) and a value of the pseudo-2nd-order rate constant, k_2 of 0.020 (95% confidence interval from 0.011 to 0.028). 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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