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Sustainable Removal of Dyes from Wastewater Using Eggshell-Derived Calcium Carbonate Nanoparticles: Adsorption Isotherms, Kinetics, and Thermodynamic Analysis Supporting Sustainable Development Goals (SDGs)

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ABSTRACT

This study explores eggshell waste as a low-cost, eco-friendly adsorbent for removing Aniline Blue dye from water. Batch and continuous experiments under various conditions showed effective performance. FTIR and SEM analyses confirmed porosity and functional groups. The adsorption followed the Sips isotherm and pseudo-first-order kinetics. The porous calcium carbonate structure enhanced dye removal. Thermodynamic results indicated an endothermic and spontaneous process. Lower flow rates improved performance in fixed-bed columns. The study supporting environmentally friendly solutions aligned with the Sustainable Development Goals (SDGs).

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1. INTRODUCTION

Wastewater management remains one of the most persistent environmental challenges, particularly due to the release of dye-laden effluents from industries such as textiles and paper. These effluents are often difficult to treat because of the presence of stable and toxic dyes in low concentrations, which can significantly degrade water quality and threaten aquatic ecosystems [1]. Among various remediation strategies, adsorption is considered an effective method due to its operational simplicity, low cost, and high efficiency in dye removal [2].

One promising adsorbent is eggshell, a food industry by-product that constitutes about 11% of the total egg mass and is often discarded as waste [3]. Eggshell is rich in calcium carbonate and contains beneficial properties such as porosity, antibacterial activity, and a high surface area, making it suitable for adsorbing chemical pollutants from aqueous solutions [4, 5]. With millions of tons generated annually (such as the 13,103 tons estimated in Taiwan alone), eggshell waste represents an untapped source for environmental technology applications [4].

In recent years, research has shifted toward converting agricultural waste into functional adsorbents (**Table 1**). They are through chemical or thermal modification. Studies have demonstrated the effectiveness of eggshell ash and calcium oxide derived from eggshells in removing dyes like methylene blue and toluidine blue under varied environmental conditions [6, 7]. These findings confirm the eggshell's capacity to function as a bio-based adsorbent when enhanced with scientific treatment and modeling techniques such as FTIR, SEM, and adsorption kinetics [8].

Table 1. Previous studies on adsorbents using biomass.

No	Title	Ref.
1	How to calculate adsorption isotherms of particles using two-parameter monolayer adsorption models and equations	[9]
2	Adsorption of sodium in an aqueous solution in activated date pits	[10]
3	Conversion of Indonesian coal fly ash into zeolites for ammonium adsorption	[11]
4	Review on adsorption and photocatalysis of dyes for wastewater treatment	[12]
5	Trends and developments in research on adsorption in education: Bibliometric analysis	[13]
6	Utilization of orange peel-derived biochar for ammonia adsorption	[14]
7	Red onion peel biomass carbon microparticles for ammonia adsorption	[15]
8	Biochar microparticles from pomegranate peel waste	[16]
9	Utilizing cassava peel-derived carbon biochar for ammonia adsorption	[17]
10	Carbon Biochar Microparticles from Mango Peel	[18]
11	Carbon particle size from galangal rhizomes	[19]
12	Silica microparticles from bamboo leave waste	[20]
13	How to conduct paired-t-test SPSS	[21]
14	Isotherm and kinetic adsorption of rice husk particles	[22]
15	Characteristics of ammonia adsorption on calcium carbonate microparticles	[23]
16	Assessment of the Congo Red and Crystal Violet toxic dyes adsorption	[24]
17	How to purify and experiment with dye adsorption using carbon	[25]
18	How to calculate and measure solution concentration using UV-vis spectrum analysis	[26]
19	Rice husk for adsorbing dyes in wastewater	[27]
20	Sustainable biochar carbon microparticles based on mangosteen peel	[28]
21	Adsorption isotherm characteristics of calcium carbon microparticles	[29]
22	Computational calculation of adsorption isotherm characteristics	[30]
23	Analysis of adsorption isotherm characteristics using avocado seed waste	[31]
24	Removal of curcumin dyes using jackfruit seeds	[32]

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Table 1 (continue). Previous studies on adsorbents using biomass.

No	Title	Ref.
25	Olive Industry solid waste-based biosorbent	[33]
26	Sustainable carbon-based biosorbent particles from papaya seed waste	[34]

Therefore, the purpose of this study is to determine the adsorption performance of eggshells against Aniline Blue (AB) dye under varying operational parameters, and to evaluate the process through kinetic, isothermal, and thermodynamic frameworks. **Figure 1** presents the molecular structure of AB, a pH-sensitive dye widely used for diagnostic staining and cytological applications. The novelty lies in highlighting eggshell biowaste as a low-cost, scalable, and eco-sustainable solution for water purification through the lens of environmental chemistry, adsorption science, and green engineering. While numerous biosorbents have been explored, the novelty of this study lies in its dual-mode investigation, a comprehensive batch and continuous flow analysis of AB dye removal using untreated, thermally processed eggshells. Unlike other works focusing on photocatalytic or nanocomposite eggshell derivatives, this research evaluates the raw material's intrinsic adsorption potential, making it more applicable and cost-effective for developing regions. Additionally, the study integrates scientific modeling (Sips isotherm, pseudo-first-order kinetics, and Yoon-Nelson prediction) and material characterization to uncover the thermodynamic and kinetic behavior of dye interaction.

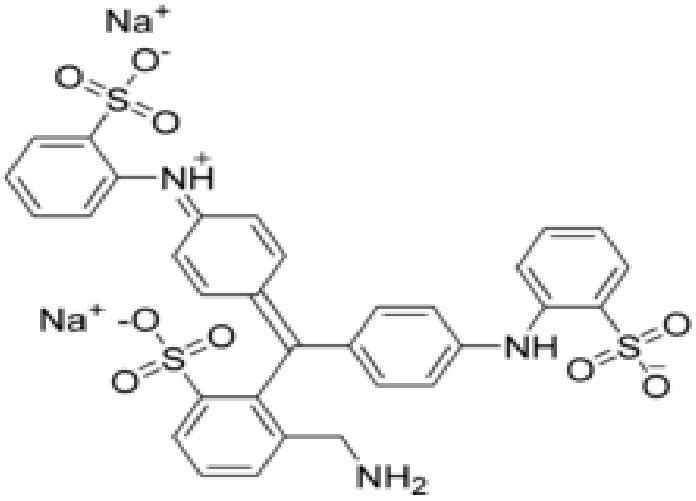

Figure 1. Chemical structure of AB dye.

2. METHODS

2.1. Batch Adsorption Experimental Work

Various amounts of the adsorbent (0.5-2.5 g/L) were mixed with different concentrations of the dye (10-50 ppm) in 100 ml containers. The experiments were conducted using a complete factorial design of experiments, with a fixed temperature of 25℃ and an initial pH of 5.8. The initial set of trials involved shaking containers at 150 rpm for a given amount of time, with each step ranging from 30 to 180 minutes. How temperature and pH affected the results was examined using a different set of full factorial designs. The temperature range was (20-60) ℃, step 10, while the pH range was (3-11), step 1. The ideal values of the factors examined in the first set were maintained. The absorbance was measured at a wavelength of 594 nm using a UV-visible spectrophotometer (Shimadzu 1800) to determine the equilibrium

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concentrations of AB dye. Detailed information on how to do adsorption and how to calculate the results is explained elsewhere [9, 25, 26].

Figure 2 shows that the absorbance against the concentration curve was linear. This curve will be used as the standard to indicate the dye concentration based on the absorbance obtained at 594 nm for all investigations employing a UV-Vis spectrophotometer. This particular wavelength was selected as it lies above the maximum absorbance of the AB dye, in order to calculate as accurately as possible, the concentration of the dye in solution. The concentration (c) vs absorbance curve was linear. Since the Beer-Lambert relationship is linear in this concentration range, this confirms that the applied spectrophotometric method is reliable. The obtained curve will be taken as an indication of the dye concentration as a function of the absorbance at 594 nm for all studies in which a UV-Vis spectrophotometer was used. The calibration curve is paramount in the ensuing experiments where dye removal percentage is quantified, thus enabling a clear understanding of the adsorption process. Using the following equations, it will be possible to determine the equilibrium concentrations and the efficiency of dye removal [14] (see equations (1) and (2)).

$$\mathrm{Qe} = (\mathrm{Co} - \mathrm{Ce}) \times \frac{\mathrm{V}}{\mathrm{M}} \quad (1)$$

$$\%\ \mathrm{removal} = \frac{Co-Ce}{Co} \times 100 \quad (2)$$

The equilibrium concentration of the dye is equal to *Ce*, while the initial concentration is *Co*, measured in mg/L. At equilibrium, the amount of dye per gram of adsorbent is denoted by *Qe*. The dye solution volume, in L, is denoted by *V*. *M* stands for the adsorbent mass in grams.

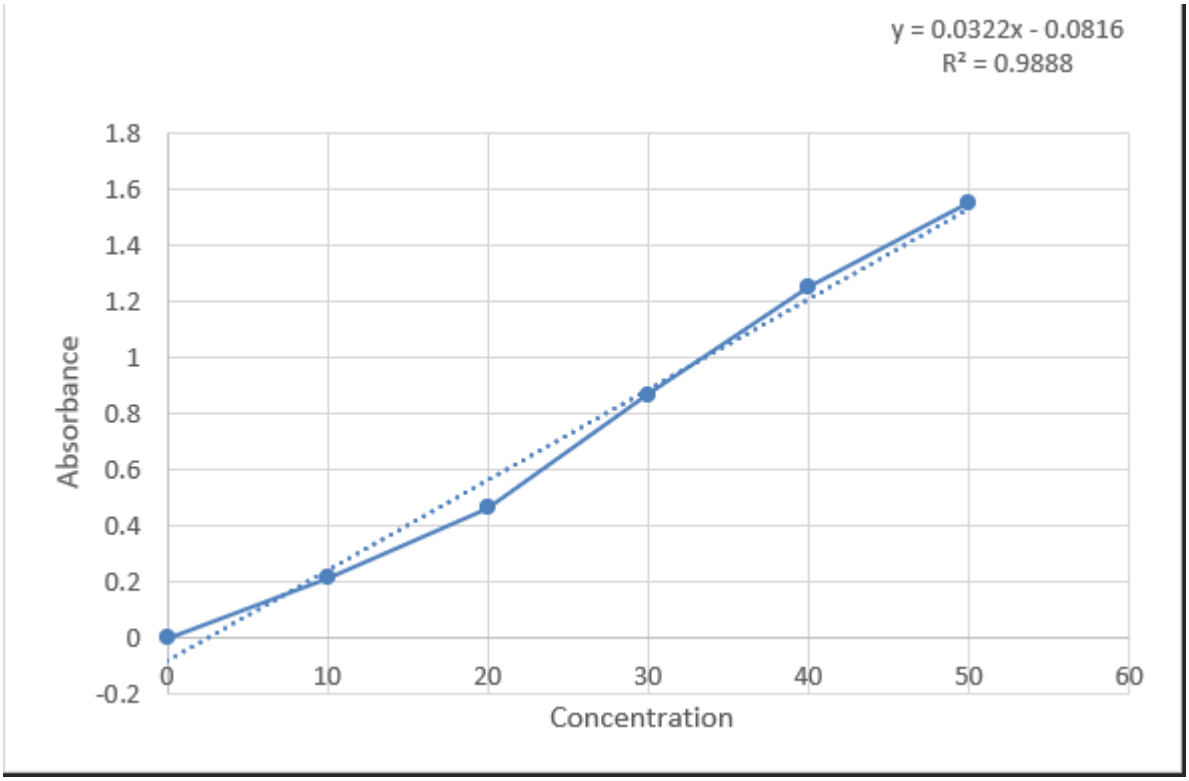

Figure 2. Calibration curve of AB concentration against spectral absorbance.

2.2. Dye Solution Preparation

To generate a stock solution of 1000 ppm, 0.25 g of AB dye was dissolved in 250 mL of distilled water. No filtration was performed before use. Adding a certain percentage of

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distilled water into the mixture created dye solutions with varying concentrations ranging from 10 to 50 ppm.

2.3. Adsorbent Preparation

The eggshells were cleaned multiple times to remove any dust or other contaminants, then dried in an oven set at 105℃ for 24 hours. The samples were ground and sieved after drying to achieve a particle size of less than 125 µm. They were then dried in an oven set at 105°C for a whole night to remove any remaining moisture. Finally, they were kept in desiccators that had been cleaned and dried for future use.

2.4. Continuous Mode Study

To evaluate the effect of flow rate on the system performance, a series of experiments were conducted in continuous mode operation under the optimum conditions of the AB dye. A tank with a capacity of 15 L, two packed bed columns (radius of 10 cm and height of 26 cm), a filter cartridge of polypropylene with a filtration capability of 5 microns, and two valves to control the flow rate, a pump to provide the dye solution through the columns, formed the experimental setup. A volume of 10 L of dye solution was prepared at a concentration of 20 ppm. To prepare 10 L of the 20% (v/v) stock solution, 200 mL was taken from the stock solution and diluted with distilled water. Asia Scientific Products imported the eggshells and rolled them in tissue before packing, so no packing material would be lost if the temperature changed during operation. The complete experimental design and procedure for preparing and conducting the tasks are summarized in **Figure 3**. It helps for the overall picture of the system components and the methodology it has implemented in the continuous mode, before diving into great detail.

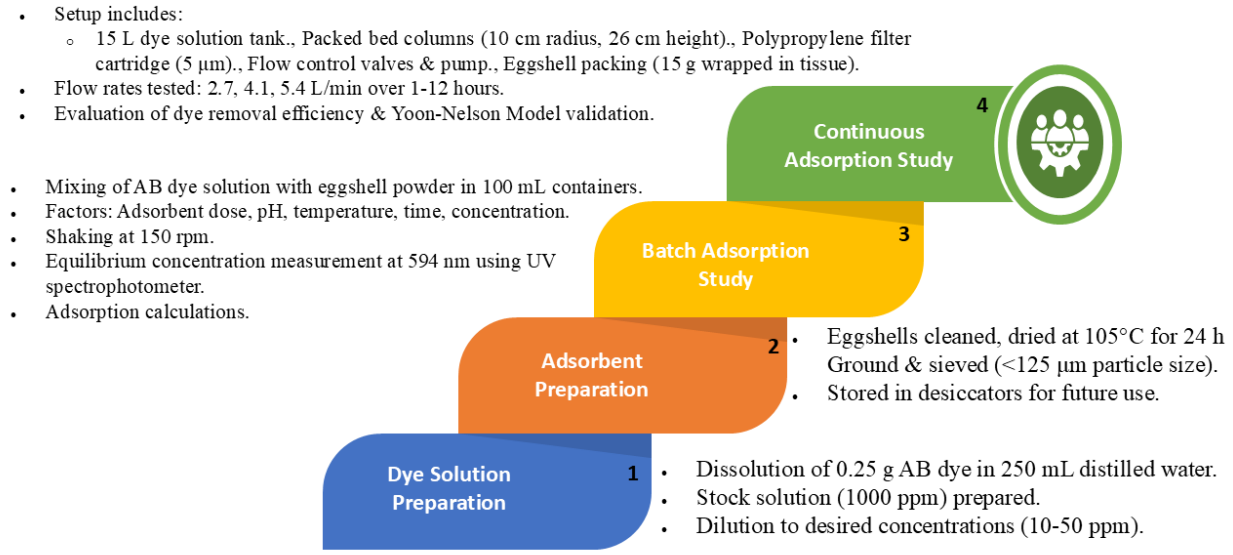

Figure 3. Experiment steps diagram.

3. RESULTS AND DISCUSSION

3.1. Batch Study (Characterization of Eggshells)

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Fourier transform infrared spectroscopy was used to describe the eggshells. The eggshells have been significantly characterized with Fourier transform infrared spectroscopy (FTIR) to better understand their functional groups and chemical composition. Detailed information regarding FTIR is explained elsewhere [35-37].

Figures 4 and **5** show the detailed FTIR spectra. The strong absorption peak of carbonate at 1433 cm^{-1} is evidence of the alkyl group, which is also an important factor in the eggshells. Likewise, the absorption due to OH was found at 3414 cm^{-1} in **Figure 4**, indicating that moisture or hydroxylated species are present on the eggshell surface, which may influence its adsorptive characteristics. Moreover, the characteristic absorption peak in calcite is visible at 876cm^{-1}, which further confirms the mineral composition of the eggshells. It is important to highlight the substantial shift in the peak positions and a change in the band intensity (**Figure 5**) after loading the eggshells with the AB dye. This variation might suggest the possible interactions between the eggshells and the dye molecules, inferring changes in the chemical environment around the functional groups. Interactions like these would be crucial in unraveling the adsorption mechanisms involved. The FTIR analysis confirms the structural features of the eggshells, illustrating their potential for functioning as effective adsorbents in numerous applications, especially in capturing dyes. The findings highlight the need for additional investigations into the adjunct adsorption capacities of these bulk natural adsorbents and kinetics.

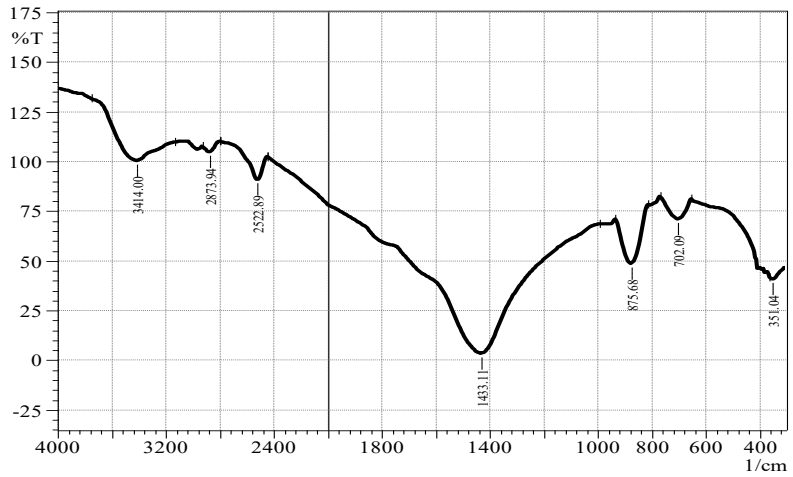

Figure 4. FTIR spectrum of eggshell adsorbent.

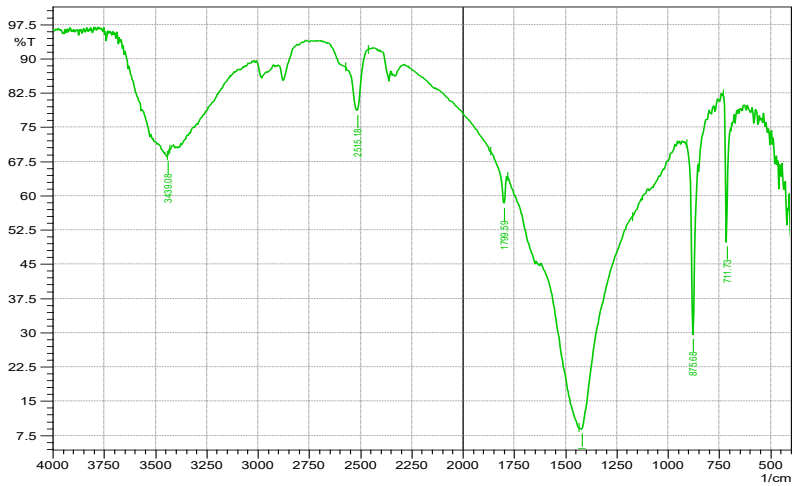

Figure 5. FTIR spectrum of AB dye-loaded eggshell adsorbent.

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The sorption involved an adsorption process, as evidenced by the surface alteration of the adsorbent, and the primary interaction was of the eggshells with the AB dye. A Scanning Electron Microscope (SEM) was used to picture the morphological aspect of the surface-luminous eggshell particles, which are displayed in **Figure 6**. A high-resolution image of the eggshell material surface morphology and structural features that are essential for this adsorption ability. The SEM shows that the rough and very porous morphology of the eggshell biosorbent is an appropriate material for dye sorption. The adsorption capacity of Himalayan salt is supported by two structural factors: the existence of active spots available for interaction with AB dye molecules, and the large surface area [38]. Additionally, eggshells contain calcium carbonate, which aids in their ionic and covalent interaction with dye constituents. This interaction is especially relevant in wastewater treatment applications, where the removal of organic pollutants is of utmost importance. The fact that eggshells can act as a biosorbent demonstrates not just their potential in the context of environmental remediation but also the need to investigate different treatment conditions (e.g., pH, temperature, and contact time) for optimal adsorption efficiency. Future research could investigate better conventional knowledge on modifying surfaces for improved adsorption efficacy and selectivity towards the desired contaminants. Moreover, exploring the regeneration potential of the biosorbent will give insight into the sustainability and cost-effectiveness of the biosorbent in large-scale applications.

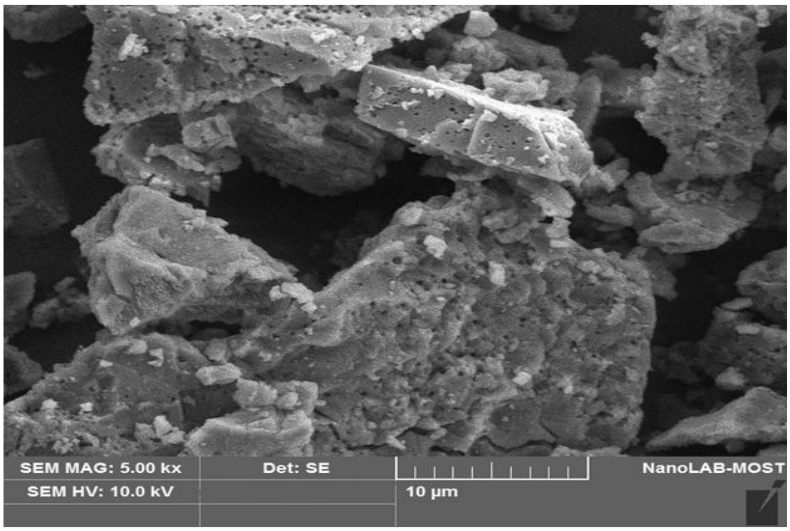

Figure 6. SEM of bio-sorbent (eggshell).

3.2. Effect of AB Concentration

The results of experiments showed an increase in the dye concentration with a growing adsorption capability until reaching equilibrium [39]. **Figure 7** shows that the percentage removal of the dye at various concentrations (10-50 ppm) with the same adsorbent amount (0.05 g) increases with the treatment of the dye concentration, as the gradient concentration is higher at lower dye concentration, which is the driving force of adsorption [40]. The increase in adsorption can be explained by the high concentration of dye molecules present in the solution, which promotes the interaction between the dye and the binding sites on the adsorbent. A higher concentration means that there are more dye molecules available to fill in the active sites of the adsorbent, leading to a higher rate of adsorption. Note that this trend may not last forever, however. The high concentration will significantly fill the adsorption sites. Thus, the increase in concentration may decrease the percentage removal efficiency. This saturation phenomenon can lead to a plateau in the adsorption capacity, as

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the increase in dye concentration does not contribute to a proportionate increase in removal efficiency. Moreover, the adsorption process can be influenced by the presence of competing ions in the solution or other substances that may compete with the dye for the active sites of the adsorbent, potentially affecting the overall performance of the dye removal process. **Table 2** is the percentage removal of AB dye.

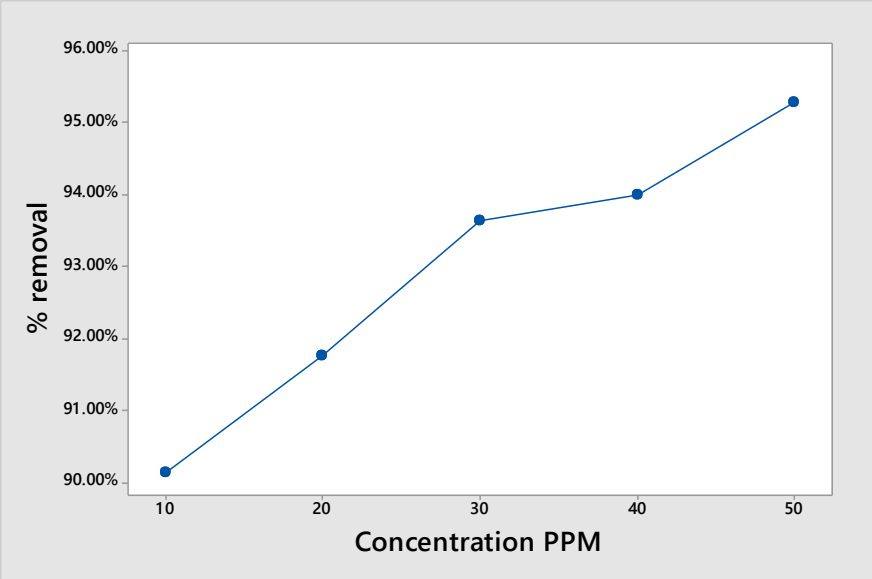

Figure 7. The relationship between dye concentration and elimination percentage at (10-50ppm) dye concentration, 0.05-mg adsorbent dosage, 20℃, pH of 5.4, and a contact time of 3 hrs.

Table 2. Percentage removal of AB dye.

Conc.	10	20	30	40	50
% Removal	90.14	91.76	93.63	94	95.29

In controlled conditions (0.05 g adsorbent, 20℃, pH of 5.4, and 3 hours contact time), the percentage of dye removal of AB dye at 10 ppm improves significantly to 90.14 and 95.29 % at 50 ppm. Since the removal efficiencies at all studied concentrations were consistently high (>90%), eggshell biosorbents are very good dye removers. The trend observed suggests that a denser concentration is generated with higher concentrations, to improve the passage of the dye molecules on the adsorbent surface. Thus, to enhance the overall adsorption efficiency, according to the experimental results, the concentration of AB dye should be increased. This beneficial effect is attributed to a more severe concentration gradient and more interaction with accessible surface sites. However, when adsorption approaches equilibrium at higher doses, there may be saturation of binding sites. These promising results further support the potential of eggshell biosorbent as a sustainable and cost-effective alternative for dye removal, specifically in wastewater treatment applications.

3.3. Adsorbent Dosage Effect

In the study of the adsorbent dosage effect on the adsorption capacity, upon investigation of the adsorbent dosage effect, it was observed that increasing amount of adsorbent (0.5-2.5 g/L) also increased the removal efficiency, at which point vacant sites were available during the sorption process, leading to an increase in the surface area of contact with the dye [18]. Moreover, as the pH of the solution increases from 5.4 to 9.8, it decreases the adsorption

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capacity. The dye removal percentage grew from 94.8 to 98.1%. **Figure 8** demonstrates the dye removal as a function of adsorbent dosage.

The substantial increase in dye molecule adsorption with an increased adsorbent dosing (0.5–2.5 g/L) is attributed to an increase in the number of active sites available for interaction with dye molecules. Higher specific surface area increases dye-adsorbent contact, resulting in higher dye removal percentage. Very high dosages lead to adsorption equilibria, where the addition of an adsorbent does not increase the removal efficiency. For a more economical approach, it is important to comprehend this process because it can reduce the quantity of adsorbent used while still being able to attain minimum dye removal [41].

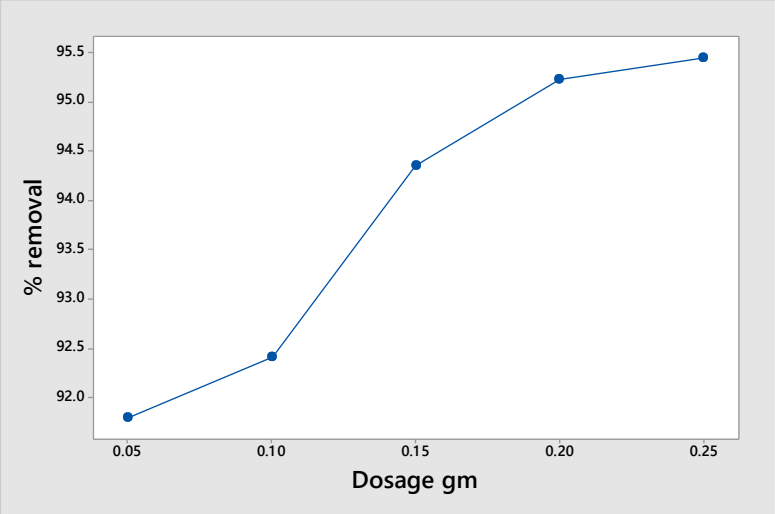

Figure 8. Investigating the relationship between adsorbent dosage and dye removal percentage at 20°C, 20 ppm dye concentration, pH of 5.4, and three hours.

3.4. Impact of Duration

Effect of adsorption time (60, 120, and 180 min) on the percent Removal of AB dye. The adsorption also increases from 73.8 to 94.8% with increasing time until it attains equilibrium, the state after which no increase occurs. This was expected, as the adsorbate dye occupied a greater number of free active sites, thereby promoting their accessibility [42]. The results are shown in **Figure 9**. **Figure 9** is the effect of contact time on the percentage of dye removal, 20°C, pH of 5.4, 0.05 g adsorbent dosage, and 20 ppm dye concentration. However, the solution pH (5.4–9.8) resulted in a drop in their adsorption capacity. The reduction can be attributed to the electrostatic repulsion between the dye molecules and the adsorbent surface charge due to increased pH, thereby minimizing their contact. However, the removal ratio changed from 94.8 to 98.1% during the process, indicating that other forces might also contribute. Such factors may be higher dye solubility or changes in the surface characteristics of the adsorbent [43]. In addition, the effect of temperature and mixing speed for the adsorption process must also be investigated, as both parameters have a key role in dye removal efficiency and kinetics. Higher temperatures typically increase dye molecular mobility, thereby promoting interaction with the adsorbent. Moreover, optimal mixing can enhance the interaction between the dye and the adsorbent, which will result in a greater removal efficiency.

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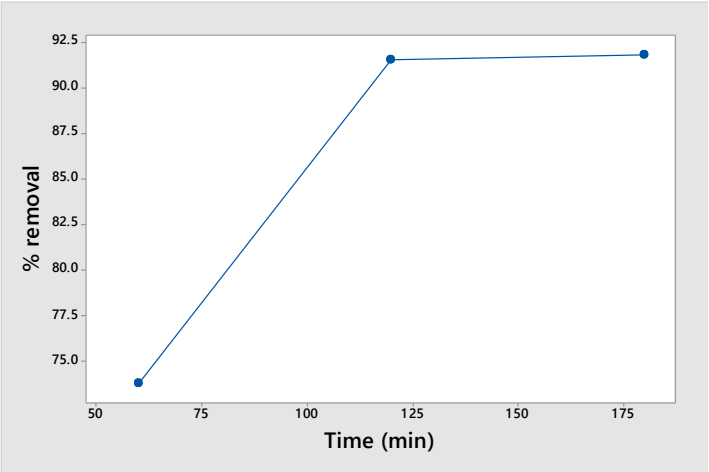

Figure 9. The impact of contact time on the percentage of dye removal at 20oC, pH of 5.4, 0.05g adsorbent dosage, and 20 ppm dye concentration.

3.5. Impact of pH

The effect of pH in the neutral, essential, and acidic media areas of (3, 5, 7, 9, 11) was studied by modifying the samples with NaOH and HCl solutions. It was observed that the increase in the percentage removal (74.8, 86.5, 92.2, 97.6, and 98.2%) with increasing pH; therefore, dye removal can be more efficient in an alkaline medium, and this increases the number of ionized sites [44]. **Figure 10** is about plot of pH versus the percentage removal of dye. The acidic condition provides higher removal efficiency because it contains more ionized adsorption sites on the surface of the adsorbent. At higher pH, the adsorbent surface is negatively charged, which enhances its electrostatic interaction with positively charged dye molecules. This increased allure thus optimizes the adsorption process for the extraction of dyes in alkaline media [45]. In addition, pH could also affect the stability of the adsorbent and the integrity of dye molecules, and thus, it needs to be taken into account as well. The above structural changes in the adsorbent may occur in the presence of highly acidic or basic conditions, affecting in turn the effective overall adsorption capacity. Further investigation may be made on the best pH for operation at the point of maximum stability of the adsorbent in real and/or field conditions by exploring more about the effectiveness of the adsorbent after several cycles of regeneration after dye removal. This insight will help improve dye removal processes and will be useful in improving wastewater treatment processes.

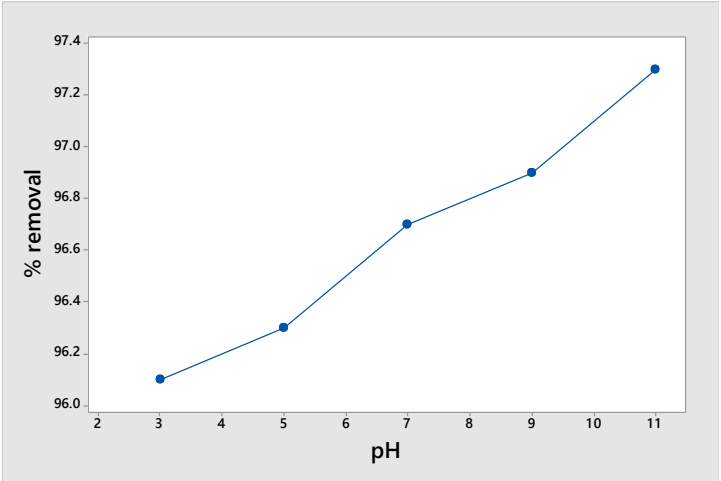

Figure 10. The impact of pH on the percentage of dye removal at 20oC, 20ppm dye concentration, 0.05g adsorbent, and 3 hours.

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3.6. The Effect of Temperature

The effects of temperature on the adsorption of AB dyes were studied in a temperature range of 20, 30, 40, 50, and 60 °C. The results show that the dye adsorption increases with increasing temperature due to the enlarged surface area, which means the adsorption process is endothermic [46]. The temperature effect on the adsorption process is presented in **Figure 11**. Thus, favor the above aspects or increase adsorption capacity at higher temperatures. First, at a high temperature, the kinetic energy of dye molecules is raised, thus increasing the frequency of collisions between dye molecules and the adsorbing surface, enhancing adsorption. The larger pore size and surface area at higher temperatures also improve the diffusion of the dye molecules into the active sites of the adsorbent. Since exothermic adsorption processes involve weaker dye-adsorbent interactions, the favourable effect of temperature indicates that chemisorption could be significant in the adsorption mechanism [47]. In addition, via temperature manipulation, the ability to improve adsorption can result in a decrease of operational costs and a decrease in treatment times, being these vital parameters are in large-scale treatment stations. This way, sectors can achieve better compliance with environmental regulations and reduce their ecological footprint by incorporating the knowledge gained through these data into such concepts. Thermal and chemical systems alone have achieved a considerable amount of efficacy in pollutant removal, which has also been utilized to develop hybrid systems; however, the insights from this study can help in developing hybrid systems as well.

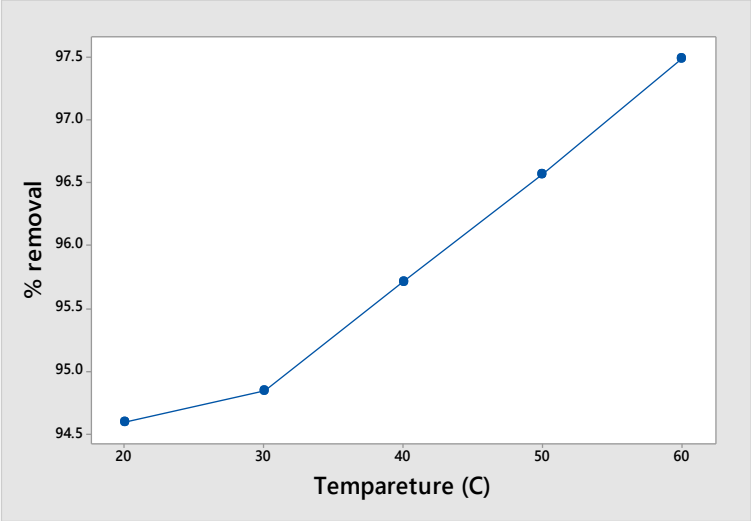

Figure 11. Variation in dye removal as a function of temperature at 3 hours. Contact time, 20ppm dye concentration, 0.05 g adsorbent dosage, and pH of 5.4.

3.7. Adsorption Isotherm

To verify the experimental results, the equilibrium relationship among adsorbent quantity and adsorbate concentration as a function of adsorption duration is described by the Langmuir, Freundlich, and Sips isotherms. Detailed information for the calculation is explained elsewhere [25, 26].

Assumptions made by the Langmuir isotherm include the following: the surface is energetically homogenous, the adsorbed molecules do not interact with one another, and the maximal adsorption rate occurs exclusively on localized spots on the surface, leading to the formation of a monolayer. The following equation can represent the linear form of the Langmuir isotherm [48, 49] (equation (3)).

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$$\frac{Ce}{q_e} = \frac{1}{K_l q_m} + \frac{Ce}{q_m} \quad (3)$$

In this context, q_e is the amount of dye adsorbed at equilibrium (in mg/g), q_m is the amount adsorbed at saturation (in mg/g), *Ce* is the equilibrium dye concentration (in mg/L), and K_l is the Langmuir constant. Freundlich isotherm can be used to describe the allocation of solute between solid and aqueous phases at saturation [50]. The linear form of the Freundlich equation is expressed as (equation (4)).

$$\ln q_e = \ln k_f + \frac{1}{n} \ln C_e \quad (4)$$

The Freundlich constants, K_f and *n*, are used to measure the adsorption capacity and intensity, respectively. The Sips isotherm results from the Langmuir and the Freundlich isotherms. The model is employed for targeted adsorption in the absence of adsorbate interactions [51]. At low equilibrium concentrations (C_e), the Sips isotherm effectively reduces to the Freundlich isotherm, but at high C_e, it predicts the Langmuir monolayer sorption characteristic. Here is the expression for the Sips linear equation model (equation (5)).

$$q_e = \frac{q_s k_s C_e^{1/m}}{1 + k_s C_e^{1/m}} \quad (5)$$

In this context, q_s (mg/g) is the maximal adsorbate uptake per unit mass of adsorbent according to Sips, K_s (L/mg)1/m is the Sips constant for the energy of adsorption, and *m* is the Sips parameter that describes the system's heterogeneity. As indicated in **Table 3**, the model constants (K_l and q_m) and correlation coefficient (R^2) were determined by linear regression by graphing (C_e/Q_e) vs (C_e) according to the Langmuir isotherm. These findings disprove the Langmuir isotherm for AB adsorption using eggshells as an adsorbent. Other regression analyses of the observed data using STATISTICA 10.0® software give the results that are shown based on Freundlich and Sip's assumption for a batch mode study. **Table 3** shows that the Sips isotherm is followed by the AB adsorption by the eggshell adsorbent.

Table 3. At 30°C, the adsorption isotherm parameters for AB on eggshells.

Langmuir	$Ce/qe = 1/kLq_m + Ce/q_m$
q_m	-75.188
KL	-0.1114
R2	0.2826
Freundlich	ln qe = ln KF + 1/n ln Ce
n	0.928
K_f	10.564
R2	0.9662
Sips	qe = (qsKsCe1/m) / (1+ KsCe1/m)
Ks	0.7175
m	0.3729
R2	0.9958

3.8. Adsorption Kinetics

The absorption rate of AB dye on eggshells was researched using Elovich's equation, pseudo-second-order, and pseudo-first-order models. The pseudo-first-order kinetic model can be represented by equation (7).

$$\ln (q_e - q_t) = \ln q_e - (k_1)\, t \quad (7)$$

Here, q_e and q_t are the adsorbent quantities (in mg/g) at equilibrium and time *t* (in minutes), respectively, and k_1 is the rate constant of adsorption (in min^{-1}). Pseudo-second-order equation is also stated as [48] (equation (8)).

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$$\frac{t}{q_t} = \frac{1}{(k_2\, q_e)} + \left(\frac{1}{q_e}\right) t \quad (8)$$

The pseudo-second-order adsorption rate constant, denoted as k_2 (g/mg.min), The Elovich kinetic equation, in its linearised version, is written as [51] (equation (9)).

$$q_t = \frac{1}{\beta}\ln(\alpha\beta) + \frac{1}{\beta}\ln(t) \quad (9)$$

Here, q_t denotes the quantity of gas adsorbed at time t, and β represents the constant of desorption. The preliminary rate constant for adsorption is denoted by α. Various kinetic models, including Pseudo-first, second-order, and Elovich, can be fitted to experimental data to examine the adsorption rate, describe the process, and anticipate information about the adsorbent/adsorbate interface [52]. The correlation coefficient (R^2) represented the degree of agreement between the model value and the experimental data. The alteration in the proportion of dye elimination with time is displayed as adsorption kinetics. **Figure 12** is for a linear representation of the Lagergren plot of log (q_e-q_t) with the adsorption time (t), which might be used to get the first-order rate constant. Plotting (t/q_t) and (q_t) against adsorption time (t), as illustrated in **Figures 13** and **14**, respectively, through the values of correlation coefficients (R^2), yields the pseudo-second-order and Elovich's constants, respectively. The kinetic models' characteristics, constants, and correlation coefficients are displayed in **Table 4**.

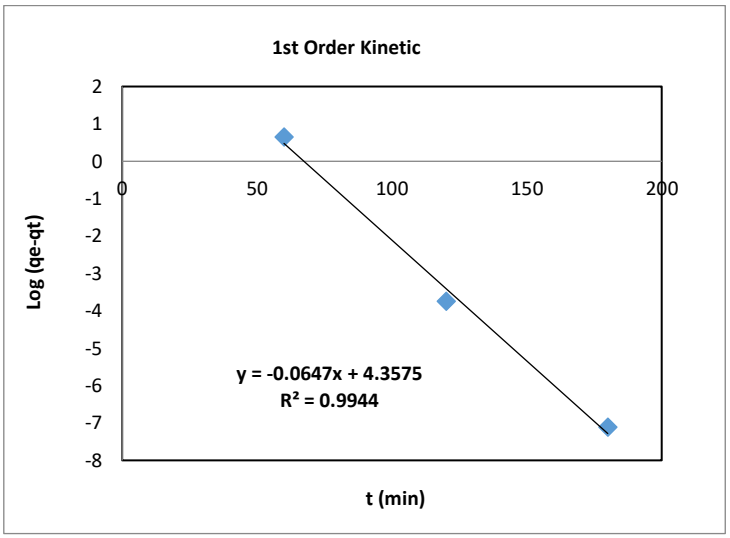

Figure 12. Eggshell adsorption of AB in a pseudo-first-order plot.

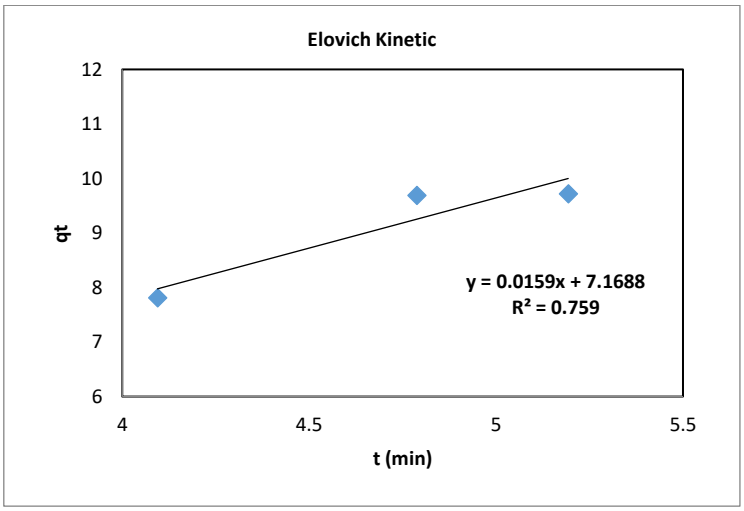

Figure 13. Eggshell adsorption of AB in a pseudo-first-order plot.

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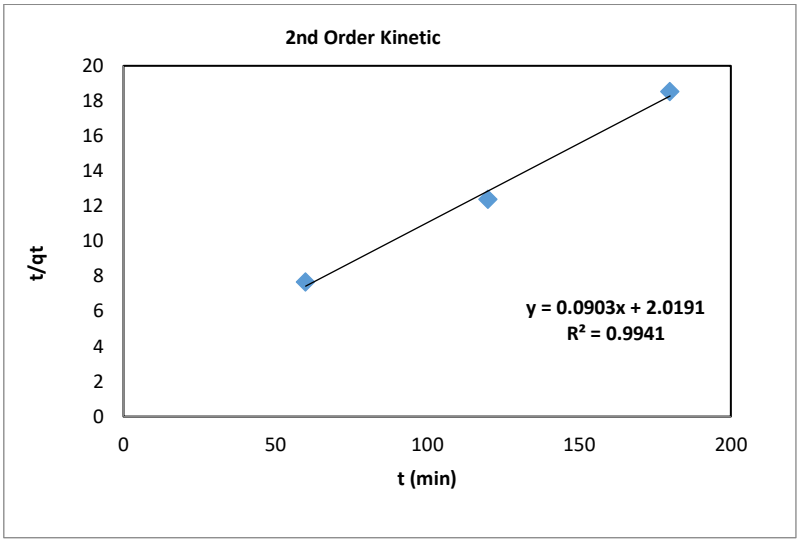

Figure 14. Elovich's graph shows the rate of AB adsorption on eggshells.

Table 4. Values of the kinetic parameters for AB adsorption concerning R^2.

Pseudo first order	**$\ln(q_e-q_t) = \ln(q_e) - K_1 t$**
q_e	78.0617
k_1	0.0647
R^2	0.9944
Pseudo second order:	**$t/q_t = (1/k.q_e)+(1/q_e)t$**
q_e	11.0742
k_2	0.00404
R^2	0.9941
Elovich	**$q_t = (1/\beta) \ln (\alpha \beta) + (1/\beta) \ln (t)$**
α	∞
β	62.893
R^2	0.759

The findings support using the pseudo-first-order kinetic model to explain the sorption of AB dye using eggshell adsorbent.

3.9. Adsorption Thermodynamics

The parameters for the thermodynamic adsorption of AB onto eggshells were extracted using experimental data collected at 293, 303, 313, 323, and 333 K. The following equations were used to compute the standard change in enthalpy (ΔH°), Gibbs free energy (ΔG°), and entropy (ΔS°) [53, 54] (equations (10), (11), (12), and (13)).

$$K_d = \frac{q_e}{C_e} \quad (10)$$

$$\Delta G^o = - RT \ln K_d \quad (11)$$

$$\ln K_d = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \quad (12)$$

$$\ln k = \ln A - \frac{E_a}{R}\frac{1}{T} \quad (13)$$

where k_d is the distribution coefficient, q_e is the concentration of AB adsorbed on eggshell at equilibrium (mg/L), C_e is the equilibrium concentration of AB (mg/l), R is the universal gas constant (8.314 J/mol K), T is the absolute temperature (K), k is the rate constant obtained

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from the PFO kinetic data, *A* is Arrhenius pre-exponential factor and E_a is the activation energy. The slope and intercept of the Van't Hoff plots of ln kd vs. 1/T were used to compute the values of ΔH°, ΔG°, and ΔSo, respectively.

Table 5 contains the thermodynamic parameters. The fact that the values of ΔG° are negative and fall within the range of (-20-0) KJ/mol suggests that the adsorption of AB on eggshell surfaces occurs naturally. The fact that the enthalpy of physisorption of eggshell-AB changes for the better suggests that the system is endothermic. There appears to be some disturbance or loss in the binding between the AB molecules and the surface of the eggshell, as indicated by the positive change of the entropy (70.05376) J/mol [55]. The Arrhenius equation [40] determined the adsorption activation energy. The physisorption mechanism is indicated by an E_a magnitude in the range of (5-40) KJ/mol, which means the kind of adsorption. A chemisorption mechanism is indicated by a value larger than 40–800 KJ/mol [56]. The mechanism is physisorption, as evidenced by the value of E_a (2.081 kJ/mol), which agrees with the thermodynamic data provided in **Table 5**.

Table 5. Apparent values of the thermodynamic parameters for the adsorption of AB dye.

Temperature(K)	K_d	ΔG° (KJ/mol)	ΔH°(KJ/mol)	ΔS° (J/mol)	k	E_a (KJ/mol)
293	7.314286	-4.84723			0.168572	
303	8.905263	-5.50846			0.179971	
313	11.76727	-6.41546	15.67272	70.05376	0.182416	2.081
323	12.53333	-6.7898			0.184425	
333	16.1863	-7.70813			0.18907	

3.10. Effect of Flow Rate

The effect of flow rate (2.7, 4.1, 5.4) L/min was studied on the adsorption of AB dye. **Figure 15** shows the flow rate's impact on the dye's percentage removal. Both the adsorption efficiency and the capacity were shown to decrease with increasing flow rate, yet to rise with increasing duration. This is because at a higher flow rate, the amount of dye adsorbed increases, and the residence time decreases, which leads to faster saturation of the adsorbent material [57]. Another plot is commonly used to describe the performance of continuous adsorption using a packed bed. The breakthrough curve is a characteristic of the process obtained by plotting the ratio of concentration (Ct/Co) (effluent to influent ratio of concentrations) versus time [58]. **Figure 16** shows that the breakthrough curve is steeper in the first three hours due to the mass transfer fundamentals [59], and then the adsorption is slower with time. Higher flow rates accelerate breakthrough by reducing contact time and adsorption efficiency. Due to longer residence time and slower flow rates, dye removal is better. The sharp initial breakthrough is owing to rapid mass transfer, while the slower latter phase is due to decreasing active sites and diffusion limits [60].

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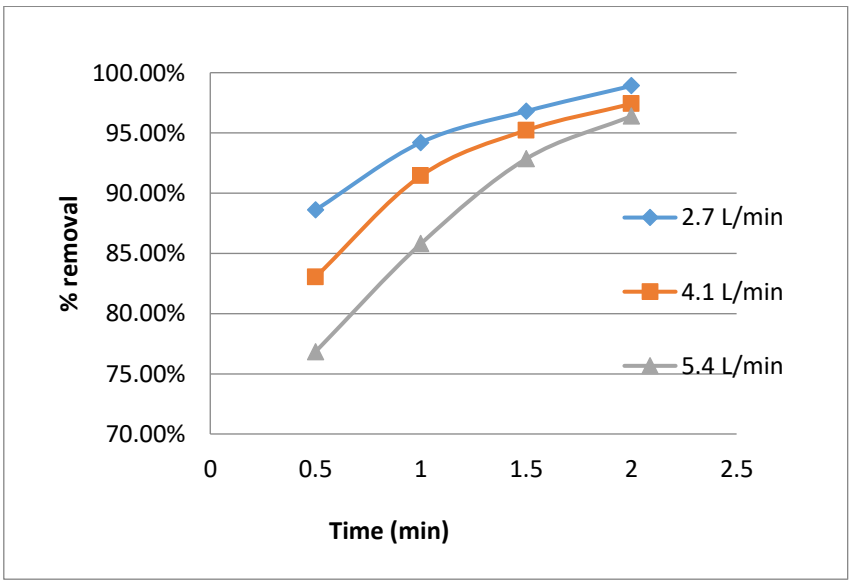

Figure 15. Effect of flow rate on the removal efficiency of AB dye.

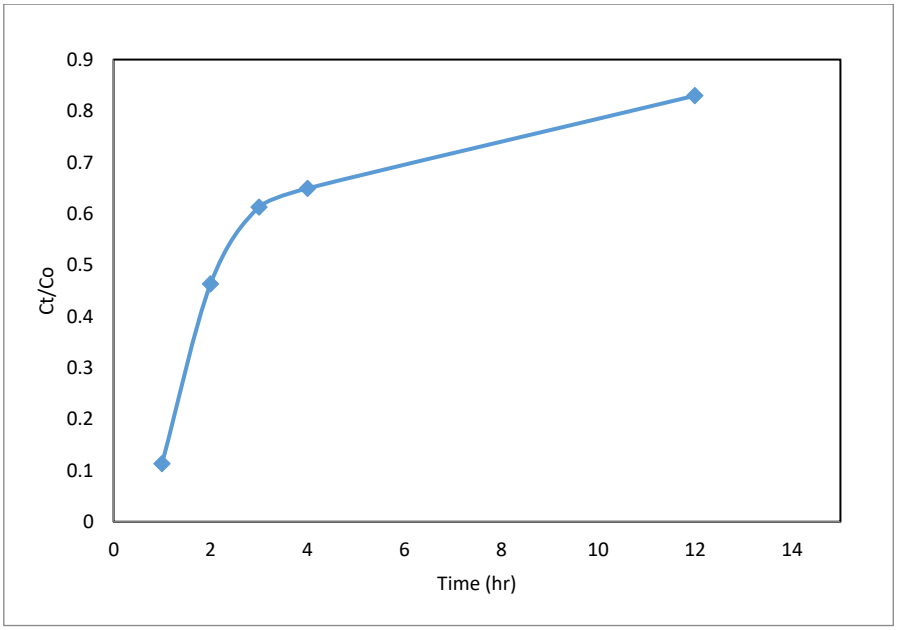

Figure 16. Breakthrough curve of the adsorption of AB Dye at 20ppm dye concentration.

3.11. Yoon-Nelson Model

The model postulates that the likelihood of adsorbate breakthroughs on the adsorbent is directly related to the pace at which the adsorption probability decreases for each molecule of the adsorbate [3]. When contrasted with the other models, this one is simpler. Under the premise of a one-component system, the Yoon-Nelson model can be represented as follows [41].

$$\ln\left(\frac{Ct}{Co-Ct}\right) = k_{YN}t - \tau k_{YN} \quad (14)$$

τ (min) is the time needed to break down 50% of the adsorbate, and k_{YN} is the adsorption rate constant (min^{-1}). By plotting the linear form of ln $[C_t/(C_o - C_t)]$ versus t according to equation 14, values of the equation constants, k_{YN} and τ, can be determined from the slope and intercept as shown in **Figure 17** and **Table 6** with a high correlation coefficient (R^2).

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The time required for a 50% breakthrough (τ) was increased with increasing feed concentration (C_o). The rate constant (k_{YN}) increased with decreasing dye concentration. The observed results agreed with the data, as reported in [61].

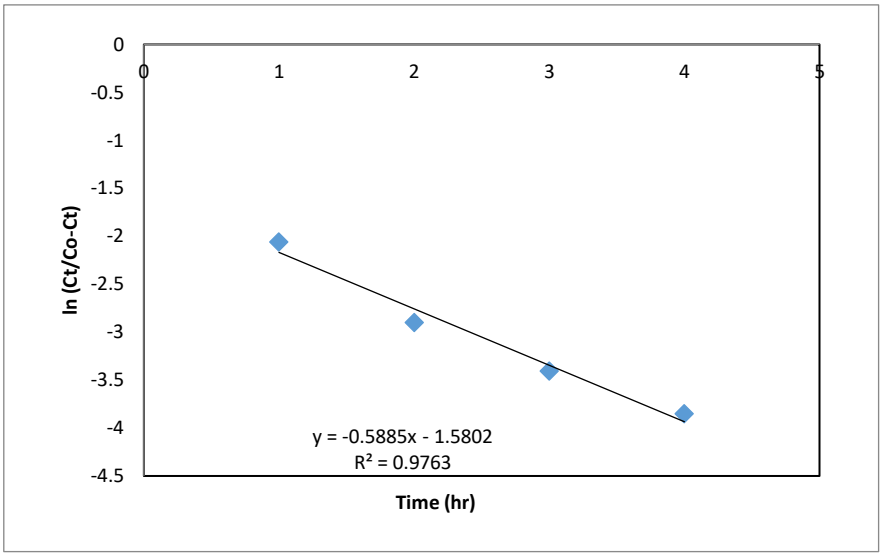

Figure 17. The isotherm constants for AB adsorption at 30oC in continuous mode.

Table 6. Yoon Nelson model parameters.

Yoon Nelsmodel in $\ln\left(\frac{Ct}{Co-Ct}\right) = k_{YN}t - \tau k_{YN}$	
K_{YN}	-0.5885
$\tau\ (hr)$	2.6851
R^2	0.9763

3.12. Relevance of Findings to the Sustainable Development Goals (SDGs)

The results of this study strongly align with the United Nations Sustainable Development Goals, particularly SDG 6 (Clean Water and Sanitation) and SDG 12 (Responsible Consumption and Production). The fabricated calcium oxide nanoparticles derived from eggshell waste have demonstrated excellent adsorption capacity for methylene blue dyes, as reflected in the favorable isotherm fits and kinetic models. These findings present a sustainable alternative for dye-contaminated wastewater treatment. By employing eggshells (a widely available and underutilized biowaste) as the precursor material, this research promotes resource efficiency and waste minimization. The valorization of biogenic waste into high-value adsorbents supports circular economy principles under SDG 12, reducing the environmental burden associated with landfill disposal and encouraging responsible material usage. This approach simultaneously addresses industrial wastewater treatment challenges by offering a low-cost and eco-friendly solution, contributing to the achievement of SDG 6. Furthermore, the use of simple calcination and eco-compatible synthesis techniques enhances the practical relevance of this method for decentralized or small-scale applications in low-resource settings. This makes the technique not only technologically feasible but also socially inclusive, as it can be adopted in rural or developing regions without advanced infrastructure. The integration of green chemistry principles and local waste reuse also positions this work within the broader framework of SDG 13 (Climate Action), by reducing the carbon footprint associated with conventional chemical adsorbent production. The experimental findings and methodological choices in this study provide a solid foundation for integrating nanotechnology, waste

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valorization, and sustainable water treatment in alignment with global sustainability goals. Finally, this study adds new information regarding SDGs as reported elsewhere (**Table 7**).

Table 7. Previous studies on SDGs.

No	Title	Ref.
1	Low-carbon food consumption for solving climate change mitigation: Literature review with bibliometric and simple calculation application for cultivating sustainability consciousness in facing sustainable development goals (SDGs)	[62]
2	Towards sustainable wind energy: A systematic review of airfoil and blade technologies over the past 25 years for supporting sustainable development goals (SDGs)	[63]
3	Assessment of student awareness and application of eco-friendly curriculum and technologies in Indonesian higher education for supporting sustainable development goals (SDGs): A case study on environmental challenges	[64]
4	A study on sustainable eggshell-derived hydroxyapatite/CMC membranes: Enhancing flexibility and thermal stability for sustainable development goals (SDGs)	[65]
5	Integrating multi-stakeholder governance, engineering approaches, and bibliometric literature review insights for sustainable regional road maintenance: Contribution to sustainable development goals (SDGs) 9, 11, and 16	[66]
6	Computational engineering of malonate and tetrazole derivatives targeting SARS-CoV-2 main protease: Pharmacokinetics, docking, and molecular dynamics insights to support the sustainable development goals (SDGs), with a bibliometric analysis	[67]
7	Innovative nanofluid encapsulation in solar stills: Boosting water yield and efficiency under extreme climate supporting sustainable development goals (SDGs)	[68]
8	Modernization of submersible pump designs for sustainable irrigation: A bibliometric and experimental contribution to sustainable development goals (SDGs)	[69]
9	Sustainable development goals (SDGs) in engineering education: Definitions, research trends, bibliometric insights, and strategic approaches	[70]
10	Sustainable packaging: Bioplastics as a low-carbon future step for the sustainable development goals (SDGs)	[71]
11	Production of wet organic waste ecoenzymes as an alternative solution for environmental conservation supporting sustainable development goals (SDGs): A techno-economic and bibliometric analysis	[72]
12	Hazard identification, risk assessment, and determining control (HIRADC) for workplace safety in manufacturing industry: A risk-control framework complete with bibliometric literature review analysis to support sustainable development goals (SDGs)	[73]
13	Techno-economic analysis of production ecobrick from plastic waste to support sustainable development goals (SDGs)	[74]
14	Techno-economic analysis of sawdust-based trash cans and their contribution to Indonesia's green tourism policy and the sustainable development goals (SDGs)	[75]
15	The influence of environmentally friendly packaging on consumer interest in implementing zero waste in the food industry to meet sustainable development goals (SDGs) needs	[76]

4. CONCLUSION

Eggshells are a readily available, inexpensive biowaste adsorbent in Iraq; this study explored the possibility of using them to adsorb AB dye in both batch and continuous modes. With a starting AB concentration of 20 ppm, a pH of 11, an adsorbent dose of 0.05, a temperature of 60 °C, and a contact time of 3 hours, the ideal circumstances were pinpointed by screening design tests. The elimination of AB dye by sorption was enhanced as the pH, adsorbent dosage, temperature, contact time, and dye concentration were increased. At equilibrium, the data agreed with the Sips isotherm and the adsorption rate by pseudo-first-order kinetics (r = 0.9944). According to the thermodynamic data, this is an endothermic process for physisorption and spontaneous adsorption. A decreased flow rate improved the adsorption system's performance in the continuous mode investigation, and the data were

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well-fit by the Yoon-Nelson model (R = 0.97). The results demonstrate that using eggshells as an adsorbent for bio-waste is a cost-effective and efficient way to remove AB dye from water-based solutions. It is worth mentioning the importance of the developed approach in different industrial applications, such as wastewater treatment plants, where it is possible to obtain excellent results in this field. One of this research's most important contributions and findings is its application-oriented, efficient, and cost-effective approach, highlighting eggshell-derived sorbents as an excellent, scalable, and low-cost solution for wastewater treatment. Compared to other sorbents, eggshells provide an environmentally friendly alternative, using many waste materials in line with the circular economy concept. Further research is needed to address some limitations regarding future work and challenges. We suggest that future studies focus on regeneration and reuse, study the regeneration efficiency of eggshell adsorbents for multiple absorption cycles to ensure long-term reusability, and study the implementation of larger-scale experiments to evaluate the real-world performance in wastewater treatment plants. It is important to conduct an economic and environmental evaluation to compare eggshell adsorption's economic feasibility and its environmental impact versus conventional treatment methods. Addressing these challenges will establish eggshell-based adsorption as a sustainable and viable solution for dye and toxic material removal for the broader use of eggshells in wastewater treatment.

5. AUTHORS' NOTE

The authors declare that there is no conflict of interest regarding the publication of this article. Authors confirmed that the paper was free of plagiarism.

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