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Effective Cyanide Adsorption in Wastewater Using Buckthorn Leaves: A study on Removal Efficiency and Kinetic Analysis.

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Abstract

Cyanide is an extremely toxic compound that prevents cellular respiration by binding to cytochrome c oxidase, that causes a speedy oxygen deficiency and potentially fatal consequences for affected organisms. Therefore, the elimination of cyanide from wastewater is of pronounced health and environmental position. The current study focuses on investigating the removal of cyanide by adsorption practice by means of buckthorn leaves as low-cost and an available adsorption medium. The cyanide removal process is conducted in a batch mode unit and under different operating conditions of temperature (25-50 °C), contact time (5-180 min), agitation speed (100-500 rpm), initial concentration (1-150 ppm), pH (1-10), and adsorbent dosage (0.1-5 g). The obtained results show that the removal efficacy is proportional to all factors except the initial concentration, and that the highest cyanide recovery rate of 95.5% is attained at pH of 8, 400 rpm for agitation speed, initial concentration of 100 ppm, adsorbent dose of 4 g, contact time of 150 min, and 50 °C of temperature. According to the correlation coefficient, the isothermal study confirm that the Langmuir model is the closest to representing the experimental data with a value of 0.9994, slightly ahead of the Freundlich and Temkin models. Also, the pseudo-second-order model records to be the best in representing the data kinetically with a correlation coefficient of 0.9999, which is ahead of the pseudo-first-order model (the Elovitch model), and the intra-particle diffusion model. Thermodynamically, the adsorption is endothermic and positively entropic with values of 159 kJ/mol and 571.8 J/mol K, respectively, and spontaneous at all temperatures studied.

Paper type: Research paper

Keywords: Adsorption; Buckthorn leaves; Batch unit; Cyanide; Removal efficiency.

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Introduction

Water pollution can be considered as one of the most serious challenges threatening human health and living organisms Mujtaba *et al.* (2017). Rapid population growth, industrial extension, and increased agricultural activities can lead to a weighty increase in the quantity of pollutants discharged into waterways around the globe (du Plessis, 2022). These pollutants range from toxic chemicals, heavy metals, organic pollutants, and inorganic toxins that contribute to the worsening of water quality, directly impacting aquatic ecosystems, and consequently human health (Singh *et al.*, 2024). Among the toxic chemical pollutants that affect water quality and stance a severe threat to human health and living organisms, cyanide ions be prominent as one of the most hazardous toxic substances released into the aquatic environment (Vaca-Escobar *et al.*, 2024). Cyanide composes of the element carbon and nitrogen (CN^-) and can be in the form of various compounds such as potassium cyanide or sodium cyanide. Lachowicz *et al.* (2024) stated that specific chemical reactions are commonly used to produce cyanide. For instance, the reaction of carbon-containing compounds with ammonia or nitrogen is one of the most utilised apprach. Specifically, the reaction of methane with ammonia can effeiciently produce the hydrogen cyanide (HCN) while

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using range of operational temperatures between 500-600 °C following to the Andrussow process (Olaya-Abril *et al.*, 2024). Also, the eaction of hydrocyanic acid (HCN) with ammonia or associated salts of sodium cyanide (NaCN) or potassium cyanide (KCN) can efficiently produce the cyanide. Furthermore, the synthesize of cyanide can be made electrically by utilising compounds of nitrogen and carbon. As an example of this reaction, Monga *et al.* (2022) presented the reaction of ammonia and sodium or potassium carbonate in the presence of an electric current.

Several industrial applications such as the mining industry use cyanide as a reagent to extract metals such as gold and silver from rocks. Also, cyanide is used to synthesize pesticides suh as phosphine and hydrogen cyanide, and some medicines like sodium nitroprusside, which can be considered as a key material in the treatment of high blood pressure (Muderawan *et al.*, 2023). Although cyanide is of prosperous advantages, cyanide still is a very toxic substance of a notable risk to the public health and environment (Zuhra and Szabo, 2022). Cyanide has a direct effect on hindering energetic enzymes like cytochrome oxidase, that discourages the use of oxygen in cells, which could produce serious damage to the respiratory system and causes a respiratory failure of a speedy death (MacLennan and Moiemen, 2015).

The removal of toxic pollutants such as cyanide from aqueous solutions is therefore vital for both the public health and environment whch requires effected and reliable technologies (MA Al-Obaidi *et al.*, 2018; Mudhar A Al-Obaidi *et al.*, 2018). Among these technologies, the adsorption can be considered one of the most widely used and energetic tactics to treat contaminated aqueous solutions (Botz *et al.*, 2016; Al-Obaidi *et al.*, 2020). The conception of this treatment is characterized by its capacity to adsorb pollutants and toxic ions while prevebnting them to spread into water. Several advantages can highlight the prosperity of the adsorption technique if compared to other associated techniques such as electrolysis and ion exchange. Chen *et al.* (2023) endorsed the adsorption as a cost-effective and less complicated technology compared to other available technologies. Natural and manufactured materials are used in the adsorption technology. In this regard, the fruit peels and rice and algae are examples of agricultural waste and charcoal and sawdust stand are organic materials that magnificently absorb a wide range of toxic pollutants (Badran *et al.*, 2023). More lately, there has been a progressive interest in developing new and powerful adsorbents to combat water pollution. Many researches are published in the open literature that introduce the utilisation of natural materials of agricultural waste, algae and charcoal, besides synthetic materials of nanomaterials as effective adsorbents to eliminate a wide set of pollutats from aqueous solutions (Jaramillo-Fierro *et al.*, 2023). Furthermore, the effects of several operational factors including the initial cyanide concentration, pH, temperature, and contact time on the adsorption efficiency were studied while signifying the optimum operational conditions to achieve the highest removal efficiency (Yang *et al.*, 2022). The recent research assured the success of mitigating the cyanide removal from water, but the challenge of having a balance between the materials cost and the overall performance is still an existed. On top of this, it is vital to consider the associated relationship between the environmental and public health standards and the competence of cyanide removal (Alalwan *et al.*, 2020).

This research attempts to delve into the efficiency of buckthorn leaves as a low-cost and freely available material for cyanide ions adsorption from aqueous solutions, besides focusing on analysing the influential factors of the process. In this regard, different adsorption models are used to analyse the relationship between the ion concentration on the surface, reaction time, and changes in some environmental parameters. Through this research, the greatest conditions for adsorption are ascertained, which contribute to the development of more active and less costly cyanide-contaminated water treatment technologies.

1. Experimental Work

1.1 Adsorbent medium (Buckthorn leaves)

The buckthorn leaves of scientific name (*Rhamnus cathartica* L.) utilised in the current investigation, which were collected from an area near Amman, Jordan. The collected leaves were characterised by their clarity, completeness, and the absence of spots, scars, or any indication of disease or deformity, in addition to their bright green color. Firstly, the leaves were washed with an excess of tap water to remove any dirt or impurities, and then washed with distilled water. The washed leaves were dried naturally in the open air, and then dried using an oven at 50 °C to avoid charring. The drying process continued until the weight was constant. Finally, the clean and dried leaves were stored in their natural state without any further treatment in amber jars equipped with two covers to prevent moisture until use.

1.2 Stock solution

To be aware of any interference with any other materials or pollutants present in the real wastewater, the adsorption of cyanide ions using buckthorn leaves was studied using laboratory-prepared contaminated aqueous solutions with known concentrations of the pollutant (cyanide ion). To achieve this goal, a 1000 mg/l stock solution of cyanide ions was prepared using sodium cyanide. The concentration were ranged between 0-6 mg/l, which examined using a spectrophotometer at 481 nm wavelength, and the correlation coefficient R^2 of calibration curve is 0.9999

1.3 Adsorption unit

The adsorption process was achieved in a batch adsorption unit, that is a water bath shaker. Samples of aqueous solutions contaminated with cyanide at known concentrations were prepared by diluting specific volumes of the stock solution and placed in clean, sterile 0.1 L flasks. To adjust the pH value of each sample, 1 M solutions of hydrochloric acid (HCl) and sodium hydroxide (NaOH) were utilised, before a specific amount of buckthorn leaves, representing the dose of the adsorbent, was placed in each of the flasks. Each flask inside the shaking water bath is tightly exposed with aluminum foil's layer, to prevent the samples from being exposed to light. After adjusting the agitation speed and temperature, the experiment begins and continues until the desired time. Then, the samples were extracted and the buckthorn leaves were carefully isolated, and then the treated solution was filtered using Whatman ® filter paper. The filtered solution was stored in 10 ml test tubes and examined using a spectrophotometer and the remaining cyanide concentration was determined by the calibration curve prepared in advance Figure 1. The operating parameters under investigation range from 1-10 for pH, 100-500 rpm for agitation speed, 1-150 ppm for initial cyanide concentration, 0.1-5 g of adsorbent, 5-180 min for contact time, 25-50 °C for temperature. Through Eqs. 1 and 2, the cyanide recovery efficiency and adsorption capacity can be calculated, respectively:

$$\%R = \left(1 - \frac{C_f}{C_\circ}\right) \times 100 \quad (1)$$

$$q = \left(C_\circ - C_f\right)\frac{V}{m} \quad (2)$$

2. Results and Discussion

2.1 Impact of pH on the cyanide adsorption

This section focuses on exploring the effect of pH on the cyanide adsorption. The experiments were achieved within the range of pH of 1-10, while considering fixed values of 400 rpm, 10 ppm, 1 g, 180 min, 50°C of the agitation speed, initial cyanide concentration, adsorbent dosage, contact time, and temperature, respectively. The acquired findings of Figure 1 shows that the efficiency of removing cyanide ions from contaminated aqueous solutions using buckthorn leaves as an adsorbent is clearly impacted by pH's change. Specifically, **Figure 1** shows a significant increase in the removal efficiency with increasing pH until reaching a stable state at pH=8. It is noted that at low pHs (1-4), the environment is highly acidic, causing an increase in the concentration of hydrogen ions (H+) that compete with cyanide ions (CN^-) for the active sites in the adsorbent. Also, a high acidity causes a large portion of the cyanide ions to convert to hydrocyanic acid (HCN) molecules, which are uncharged and therefore less efficient to bind to the active sites on the adsorbent. Moving to moderate values (pH = 5-7), the competition from hydrogen ions decreases, and the concentration of charged cyanide ions (CN^-) increases, which improves the electrostatic interactions between the adsorbent and cyanide ions. In turn, this leads to a significant increase in the removal efficiency. At basic conditions (pH = 8-10), cyanide ions are almost entirely in the form of charged ions (CN^-), which enhances their attraction to the active sites on buckthorn leaves. At pH ≥ 8, the adsorbent reaches a saturation state with the active sites, resulting in a stable removal efficiency of 95.5%. These results reflect the dynamic interaction between the nature of the adsorbent, the properties of cyanide ions, and the effect of pH value in promoting or hindering the adsorption process. These were elucidated in the finding of Alalwan *et al.* (2020).

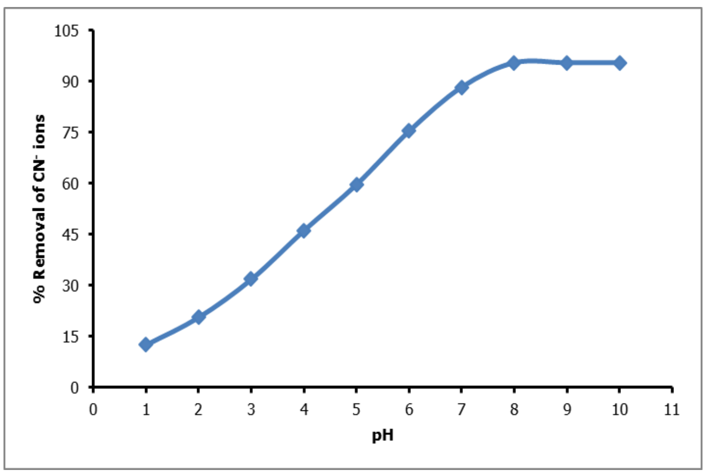

Fig. 1. Impact of pH on cyanide adsorption using buckthorn leaves at (m=1 g /l, As=400 rpm, t=150 min, T=50 ºC)

2.2 Impact of agitation speed on the cyanide adsorption

Within the range of 100-500 rpm, and with fixed operating parameters of 8 pH, 10 ppm initial concentration, 1 g buckthorn leaves, 180 min contact time, and temperature of 50 °C, the effect of agitation speed on the competence of cyanide ion removal from contaminated solutions was investigated using buckthorn leaves as a low-cost adsorption medium. The results show that the efficiency of cyanide ion removal using buckthorn leaves as an adsorbent is greatly affected by the agitation speed, as the removal efficiency gradually rises with an increase in the agitation speed until it grasps a steady state of 95. 5 % at a speed of 400 rpm **Figure 2**. At low speeds between 100-250 rpm, the removal efficiency is relatively low due to limited solution mobility, which reduces the chance of contact between cyanide ions and active sites on the surface of buckthorn leaves. Also, the effect of the static boundary layer surrounding the adsorbent at these speeds hinders the effective transport of ions to the active surface, limiting the adsorption efficiency. With increasing the agitation speed to the average values of 300-350 rpm, the improved stirring has resulted in enhanced homogeneity of the distribution of the adsorbent and cyanide ions in the solution. The higher speed also diminishes the effect of the boundary layer surrounding the adsorbent, which increases the diffusion rates and ion transport to the active sites. These improvements have reflected a significant increase in the removal efficiency, which reaches 89.1 % at 350 rpm. At high speeds ≥ 400 rpm, the removal efficiency reaches its peak value of 95.5 %. In this range, the cyanide ions are saturated throughout the active sites on the surface of the adsorbent, which endorses poor removal efficiency with increasing the agitation speed above 400 rpm. This might be the response to no additional active sites to house more ions. However, the efficiency can be fixed in this range with utilising a high speed that can contribute to conserving a homogeneous distribution in the solution. These results would indicate the importance of the agitating speed as a significant parameter in improving the adsorption action, as it has a role in lessening the resistance subsequent from the boundary layer and promotion the contact between the adsorbent and the cyanide ions (Kadhim *et al.*, 2020). However, further speed upsurge become uncreative after achievement the saturation point at a specific speed, signifying the dynamic nature of the adsorption process and its impact on the availability of active sites and mass transport factors of ions. These findings are in a agreement with the results of Ghulam *et al.* (2020).

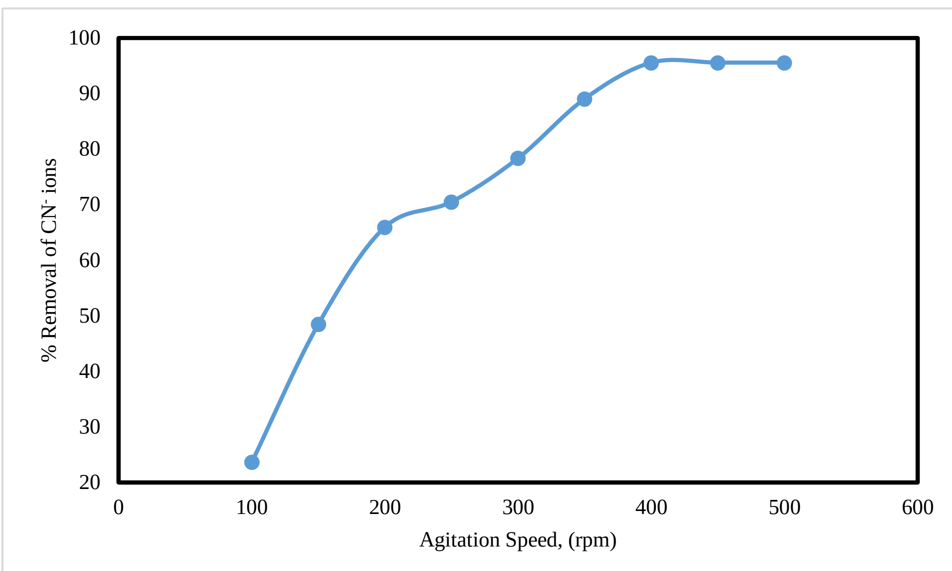

Fig. 2. Effect of agitation speed on cyanide adsorption utilising the buckthorn leaves at (m=1 g /l, pH=8, t=150 min, T=50°C)

2.3 Impact of initial concentration on the cyanide adsorption

The impact of initial cyanide concentration on the removal competence using buckthorn leaves is deliberated in this section over a wide range to perceive the action of this variable. Experiments were conducted with fixed other remaining variables of the optimum values of 8, 400 rpm, 1 g, 180 min, and 50 °C. **Figure 3** shows that the efficiency of cyanide ion removal using buckthorn leaves as an adsorbent gradually decreases with increasing initial concentration, while the adsorbed concentration increases until it reaches a steady state at high concentrations. At low initial concentrations (1-10 ppm), the removal efficiency is very high, reaching 100 % at 1 and 5 ppm and 95.5 % at 10 ppm. This excellent performance is attributed to the availability of a sufficient number of active sites on the surface of the adsorbent compared to the number of cyanide ions in the solution, which allows most of the ions to be adsorbed with high efficiency. At this stage, the system is in an unsaturated state, where the adsorbent is able to handle all of the ions in the solution, resulting in a relative match between the adsorbed concentration and the initial concentration. As the initial concentration increases to the intermediate range (20-50 ppm), the removal efficiency begins to gradually decline, decreasing from 70.1 % at 20 ppm to 45.9 % at 50 ppm. In this range, the number of cyanide ions in the solution increases at a faster rate than the availability of active sites, leading to partial saturation of the adsorbent. However, the adsorbed concentration (C_{in}-C_{out}) continues to increase, reaching 22.9 ppm at concentration of 50 ppm, reflecting the ability of the adsorbent to adsorb increasing amounts of cyanide despite the decreasing removal percentage. At high initial concentrations (60-150 ppm), the adsorbed concentration stabilizes at around 27.5 ppm, while the removal efficiency drops significantly, reaching 18.4 % at 150 ppm. This is a result to the complete saturation of the active sites on the surface of the buckthorn leaves, where the adsorbent becomes unable to absorb more ions regardless of the rise in the initial concentration. At this point, the process is limited by the maximum adsorption capacity of the adsorbent, as all active sites are filled with ions, which limits the adsorption efficiency and leads to a noteworthy decrease in the percentage of removal despite the continued presence of an excess of ions in the solution. These results highlight the dynamic relationship between the initial concentration of cyanide ions and the existence of active sites on the adsorbent. At low concentrations, the system shows high efficiency due to the proportionality between the number of available ions and the number of active sites, ensuring complete or near-complete removal of the ions. However, as the initial concentration increases, competition between ions for access to the active sites increases, causing a gradual reduction in removal competence. At high concentrations, the adsorbent becomes completely saturated, limiting its ability to adsorb excess ions. Therefore, the concentration used in the other parameter studies is 100 ppm. Similar findings were assured by Alalwan *et al.* (2021).

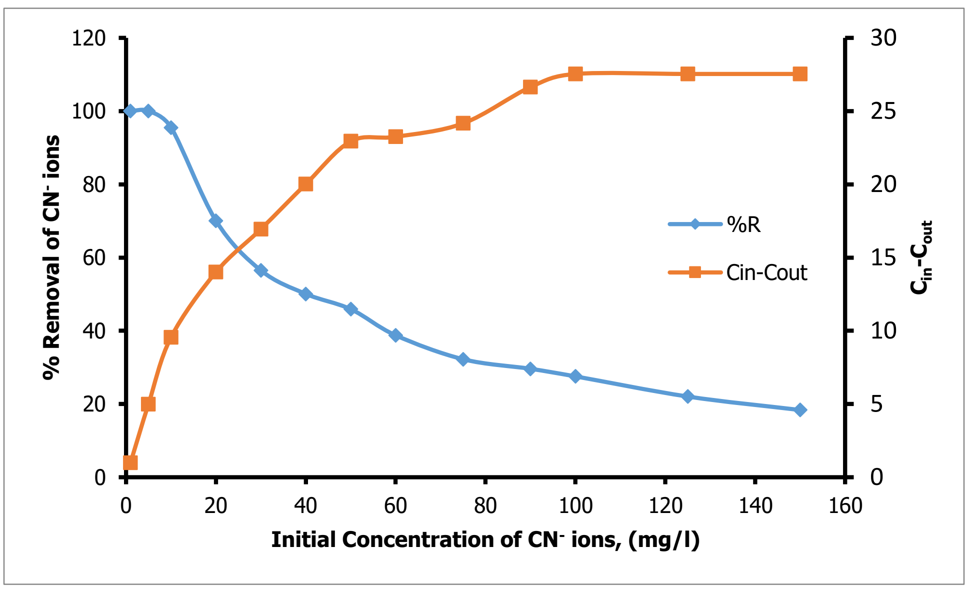

Fig. 3. Impact of initial concentration on cyanide adsorption usin buckthorn leaves at (m=1 g/l, As=400 rpm, pH=8, t=150 min, T=50 ºC)

2.4 Impact of adsorbent dose on cyanide adsorption

Different masses of buckthorn leave as adsorption media were tested in this section at fixed values of pH, agitation speed, initial concentration, contact time and temperature, which are 8, 400 rpm, 100 ppm, 180 min and 50 °C, respectively. The results show that the efficiency of removing cyanide ions increases significantly with increasing the dose of the adsorbent, as it rose from 27.6 % at a dose of 0.1 grams to 95.5 % at 4 grams and beyond **Figure 4**. This rise in the efficacy is attributed to the availability of a greater number of active sites on the surface of buckthorn leaves as the dose increases, allowing for the adsorption of a greater number of cyanide ions. Initially, at low doses of 0.1 g, the adsorption sites on the adsorbent are not saturated, causing in complete or near-complete adsorption of the ions into the solution, thus showing a significant increase in the removal efficiency. As the dose increases to 1 and 1.25 g, the removal efficiency gradually increases to 85.4 %, as the number of active sites that can bind to ions increases. However, the removal efficiency starts to slow down when we reach doses higher than 1.5 g, although the number of active sites continues to increase, the removal efficiency slows down due to the gradual decrease in the ratio of ions adsorbed per unit mass of adsorbent. This slowdown is due to the fact that most of the cyanide ions in the solution have already been adsorbed, thus increasing the dosage is too much and leads to a decrease in the specific efficiency per unit mass of the adsorbent. At the 4 g dosage, the efficiency stops increasing and stabilizes at 95.5 % although the specific adsorption capacity (q) continues to decrease, which decreased from 27.6 mg/g to 1.911mg/g at the 5 g dosage. The reduction in specific adsorption capacity with growing dose can be explained in the context of gradual saturation of the adsorbent. With each rise in dose, the active sites on the adsorbent become more abundant compared to the amount of cyanide ions in the solution, resulting in the adsorbed ions being distributed over a larger surface area, and thus the amount adsorbed per unit mass is reduced. This means that although increasing the dosage improves the removal efficiency in percentage terms, the number of ions adsorbed per unit weight of adsorbent decreases with higher dosages, causing a lessening in the specific adsorption capacity. The removal efficiency stabilizes at about 95.52 % at the dose of 4 g and beyond, indicating that the adsorbent has reached the saturation point in its ability to adsorb cyanide ions. At this stage, most of the active sites have been desorbed, and there is no further improvement in cyanide removal despite increasing the dose. This result specifies that the optimum dosage of adsorbent is around 4 g, where the maximum cyanide removal competence is attained without extreme adsorbent loss or lessening in specific adsorption capacity. Accordingly, it is clear that there is a balanced association between dosage, removal efficacy, and specific adsorption capacity. Choosing the suitable dosage of adsorbent is vibrant to accomplish the best removal effectiveness without causing additional adsorbent loss or unsuccessfully dropping the adsorption capacity. Interestingly, there is an agreement between the aforementioned results and those presented by Hasan *et al.* (2021).

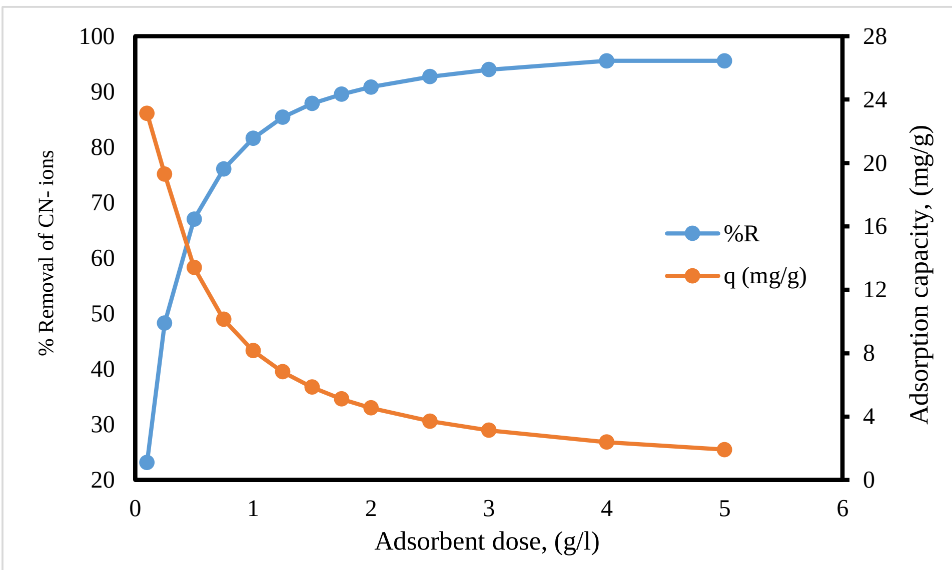

Fig. 4. Effect of adsorbent dose on the cyanide adsorption utilising the buckthorn leaves at (pH=8, As=400 rpm, t=150 min, T=50 ºC)

2.5 Impact of contact time on cyanide adsorption

Fixed values of 8, 400 rpm, 100 ppm, 4 g, 50 °C of pH, agitation speed, initial concentration, adsorbent dosage, and temperature respectively, were considered in this analysis to specifically investigate the influence of contact time ranges between 5-180 min on the cyanide removal efficacy. **Figure 5** depicts the influence of contact time on the removal of cyanide ions from aqueous solutions utilising buckthorn leaves as an adsorbent. This shows that the removal efficiency can be enlarged with rising the contact time until it reaches the supreme, where the percentage stabilizes at 95.5 % after 150 minutes. This result can be explained based on the change in the ratio (Ce/C_{in}), which indicates the concentration of cyanide ions in the solution after adsorption (C_e) compared to the initial concentration (C_{in}). Initially, at a short contact time (5 min), the cyanide removal rate was very low at 19.6 %, and the final concentration of ions (C_e) was still relatively high compared to the initial concentration, as the ratio (C_e/C_{in}) was 0.8. This indicates that the adsorbent has not yet exhausted most of its active sites at the beginning of contact, so the removal efficiency is low. As time went on and the contact period increases to 15 and 30 min, a significant increase in the removal efficiency is observed, reaching 60.7 % at 30 min. At this stage, the ion concentration in the solution decreases further, with the ratio (C_e/C_{in}) dropping to 0.39. This reflects the onset of more ions being absorbed by the adsorbent as time expands, which allows for greater mixing between the ions and the active sites on the surface of the buckthorn leaves. As the time continues to 60 and 90 min, the removal efficiency continues to improve, reaching 86.7 % at 90 min, with a continuous decrease in the ratio (C_e/C_{in}) to 0.13. This reflects the capacity of the adsorbent to adsorb more ions over time, with the remaining ions in the solution remaining relatively few, indicating effective adsorption of ions. With the contact time increases to 120 min and 150 min, the removal efficiency reaches 95.5 %, and remains stable up to 180 min, with the ratio (C_e/C_{in}) remaining at 0.045. This indicates that most of the ions are almost completely adsorbed from the solution, and the adsorbent becomes saturated, and therefore no significant improvement in removal efficiency is observed after 150 min. It can also be concluded that increasing the contact time can contribute to increasing the adsorption efficiency until the adsorbent reaches saturation at 150 min. After this point, there is no further significant improvement in cyanide ion removal, highlighting that the system has reached an equilibrium. This suggests that the optimum contact time in this system is about 150 minutes to achieve maximum cyanide removal. The same results were obtained by Mohanty *et al.* (2022).

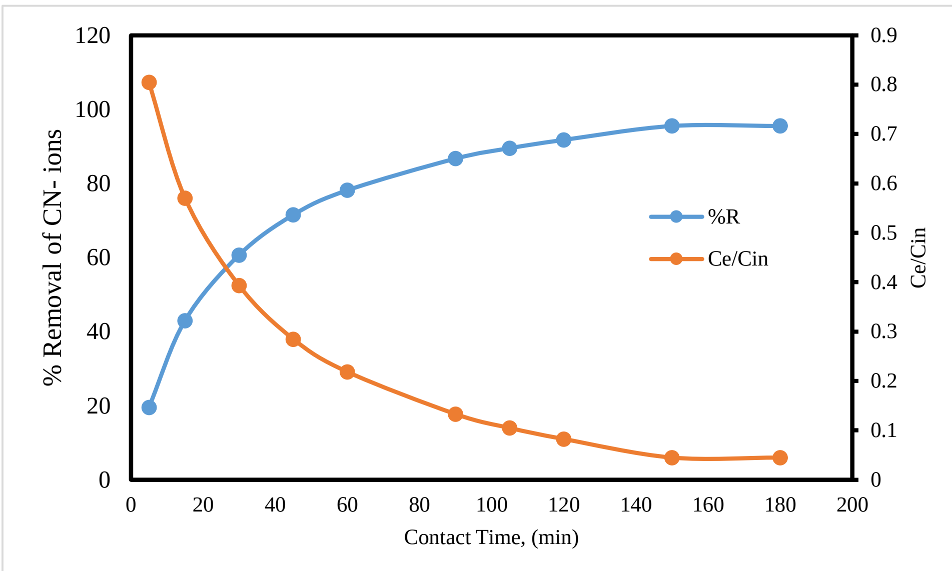

Fig. 5. Effect of contact time on the cyanide adsorption utilising the buckthorn leaves at (pH=8, As=400 rpm, m=4 g/l, T=50 ºC)

2.6 Impact of temperature on cyanide adsorption

Temperature is a dominant parameter not only in the performance of the adsorption process, but also in defining the thermodynamic features of the entire process. Thus, it is an energetic parameter that was studied within the range of 25-50 °C, while fixing the other design parameters at their optimum values of (pH=8, As=400 rpm, m=4 g/l, t=150 min). The effect of temperature on the removal of cyanide ions using buckthorn leaves as an adsorbent indicate a gradual increase in the removal efficiency with increasing temperature until it hits the maximum value of 45 °C **Figure 6**. These results assure that the process is positively affected by increasing temperature, signifying a significant effect in adsorption effectiveness at higher temperatures. At 25 °C, the removal ability is 26.8 %, the lowest in the experiments for this parameter. However, the removal competence rises to 38.9 % with growing the temperature into 27.5 °C, which represents a minor positive effect of temperature on the eliminating cyanide ions. Further increase of temperature to 30 °C and 32.5 °C results in further improvement of removal efficiency, reaching 52 % and 64.5 %, respectively. This increase indicates that the increase of temperature helps to stimulate chemical and physical reactions on the surface of the adsorbent, such as improving permeability and increasing the mobility of ions in the solution, which enhances the adsorption process. Using 35 °C and 37.5 °C, a greater increase in the removal efficiency is observed, reaching 75.3 % and 83.1 %, respectively. These high percentages indicate that an increase in the operational temperature would improve the capacity of the adsorbent to attract and adsorb ions more effectively, as heat increases the kinetic energy of molecules, which enhances the adhesion of ions to the surface of the adsorbent. With the temperature continuing to be raised to 40 °C and 42.5 °C, the removal efficiency reaches 88.9 % and 92.5 % respectively, reflecting the great influence of temperature on the process. This result indicates that the greater temperature can upgrade the adsorption capacity of the adsorbent, as many adsorption processes are driven by temperature, which further increase in the removal efficiency. The removal efficiency reaches its maximum of 95.5 % at 45 °C and 50 °C, with no significant change in efficiency when the temperature increases above this point. This is an indication that the system has reached its saturation state, where most of the active sites on the buckthorn leaves are saturated with adsorbed ions, and increasing the temperature no longer leads to further improvement in removal efficiency. These findings also indicate that an increase in the temperature has expressively enhanced the competence of removing cyanide ions from aqueous solutions using buckthorn leaves, and this is believed to be due to the impact of temperature in accelerating the physical and chemical processes that upgrade the adsorption ability of the adsorbent. However, increasing the temperature beyond 45 °C has not led to further enhancement, highlighting that this temperature is optimal for achieving maximum removal efficacy. The same perspective of these results was ascertained by Alsarayreh *et al.* (2024).

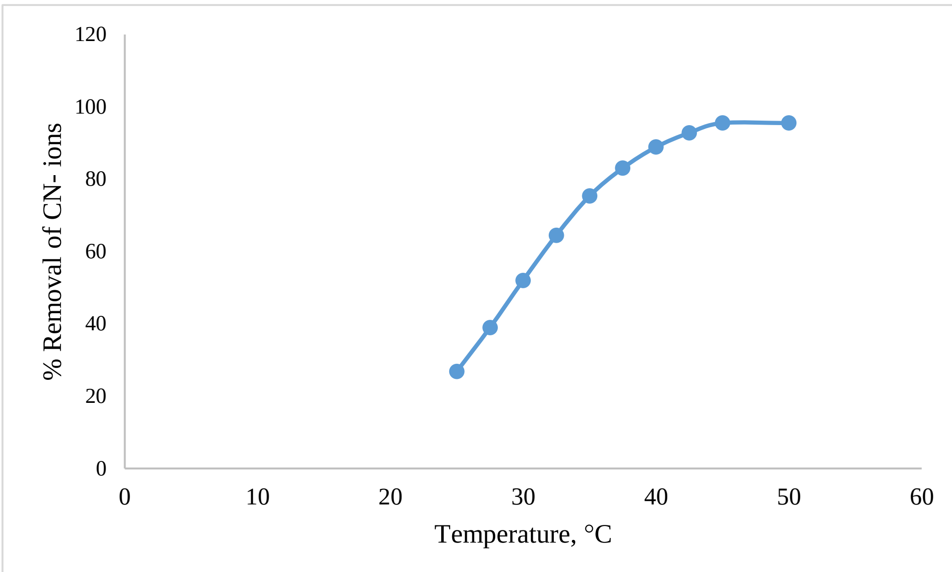

Fig. 6. Impact of temperature on cyanide adsorption using buckthorn leaves

2.7 Adsorption behavior of cyanide ions using buckthorn leaves
2.7.1 Isotherm study

The isotherm study is one of the fundamental tools for perceiving and analysing adsorption processes, as it focuses on clarifying how contaminants are distributed between the liquid and the adsorbent at a fixed temperature and pressure. This study is vital because it delivers visions into how the adsorbent interacts with ions or molecules in solution, which aids to improve the design of adsorption systems by defining the optimal adsorption capacity of the adsorbent materials. Isotherms display how the concentration of pollutants in a solution can impact the number of contaminants adsorbed, providing information about the competence and life of the adsorbent (Shadhan *et al.*, 2024). The accompanying studies are indispensable in industrial applications such as water and wastewater treatment, where the most appropriate adsorbent can be nominated based on its isothermal properties. Among the most widely utilised models for isothermal analysis is the Langmuir model, which presumes that adsorption occurs at fixed, specific sites on the adsorbent and that there is a finite ability of the adsorbent that cannot be exceeded. It assumes that adsorption occurs regardless of the attractive force between the adsorbent molecules. In contrast, the Freundlich model takes into account the heterogeneous surface of the adsorbent, assuming that the adsorption capacity varies with concentration and that adsorption occurs variably at different sites on the adsorbent surface. However, the Temkin model is used to explain the complex interactions between adsorbent molecules, which assumes that adsorption does not occur at fixed sites but is affected by dynamic interactions between molecules. These models can help to classify the adsorption behavior under different conditions, which would enhance the ability to improve industrial processes and increases their efficiency in removing pollutants from solutions. **Table 1** shows the details of the above models (Ali *et al.*, 2023).

Table 1. Details of isothermal models

Isotherm Model	Form of model's equation		Slop Term	Intercept Term	Augmented Parameter
	General	Linear			
Langmuir	$q_e = \frac{q_{max}.K_L C_e}{1 + K_L C_e}$	$\frac{1}{q_e} = \frac{1}{q_{max} K_L}\frac{1}{C_e} + \frac{1}{q_{max}}$	$\frac{1}{q_{max} K_L}$	$\frac{1}{q_{max}}$	$R_L = \frac{1}{1 + K_L C_e}$
Freundlich	$q_e = K_F C_e^{\frac{1}{n}}$	$ln\ q_e = ln\ K_F + \frac{1}{n} ln\ C_e$	$\frac{1}{n}$	$ln\ K_F$	–
Temkin	$q_e = \frac{RT}{b} ln\ K_T C_e$	$q_e = \frac{RT}{b} ln\ K_T + \frac{RT}{b} ln\ C_e$	$\frac{RT}{b}$	$\frac{RT}{b} ln\ K_T$	–

The findings of the isothermal study, shown in **Figures 7-9** and **Table 2**, obtained using the Langmuir, Freundlich, and Tamkeen models, show high values of the correlation coefficient that vary between the different models. These are 0.9994 for the Langmuir model, 0.9963 for the Freundlich model, and 0.9357 for the Temkin model. These results can be interpreted in detail based on the assumptions of each model and its mechanism of action in explaining the adsorption behavior. The Langmuir model assumes that adsorption occurs on a surface composed of homogeneous sites, i.e., all the sites on the surface of the adsorbent are equally capable of absorbing ions. It is also assumed that the adsorption occurs in only one layer (monolayer adsorption) and that there is no overlap between molecules adsorbed on the same site. It is also believed that the adsorption capacity is limited and the adsorbents can only saturate the sites when the maximum capacity of the adsorbent is reached. The high correlation coefficient of 0.9994 in this model indicates an excellent agreement between the experimental data and the Langmuir model. This means that the adsorption process in this study largely follows this homogeneous behavior associated with a limited capacity of the adsorbent. This suggests that the Buckthorn Leaves used as adsorbent may have homogeneous adsorption sites that effectively accept ions until the maximum capacity is reached. The Freundlich model reflects the adsorption behavior on a heterogeneous surface where the interaction between the adsorbent molecules and the adsorbent sites varies. This model does not assume that all sites on the adsorbent are homogeneous, but rather reflects the variation in the adsorbent sites that can have different adsorption capacities. The interaction between ions on the adsorbent surface also reflects heterogeneous interactions, and the adsorption efficiency is expected to increase with increasing concentration. The correlation coefficient of 0.9963 in this model is also high, showing that there is a good fit between the data and the model. This indicates that part of the adsorption process corresponds to a heterogeneous surface, where the ability of the sites to adsorb varies according to different concentrations. In this case, it can be said that some sites on the adsorbent are more effective than others in adsorbing ions, which reflects a complex, heterogeneous behavior. The Temkin model assumes that adsorption depends not only on surface interactions between the adsorbent and the ions, but also on thermal interactions, where thermal forces play a vital role in determining the extent of adsorption of ions on the surface. It also proposes that the adsorption can be impacted by temperature and interactions between ions in the solution. The correlation coefficient of 0.93 in this model is the lowermost among the other three models, highlighting that the experimental data are less associated with the assumptions of the Temkin model than the Langmuir and Freundlich models. This indeed proposes that the impact of temperature and intermolecular interactions cannot be the main factor in this investigation, or that non-thermal interactions are the most influential in the adsorption process. The high correlation coefficient results of the Langmuir and Freundlich models signify that the adsorption process follows a complex behavior that can be partially consistent with both. The Langmuir model delivers the superlative explanation of the data due to the stronger correlation, that means that there is a limited adsorption capacity and homogeneous sites on the adsorbent surface. However, the Freundlich model reveals the impact of variability in adsorption behavior on a heterogeneous surface, which means that some sites have a superior adsorption capacity than others, particularly at higher concentrations. On the other hand, the lower correlation coefficient of the Temkin model specifies that this model is not the most proper for elucidating the data in this investigation, as thermal interactions or intermolecular forces may have less effect on the adsorption process in a comparison to other factors.

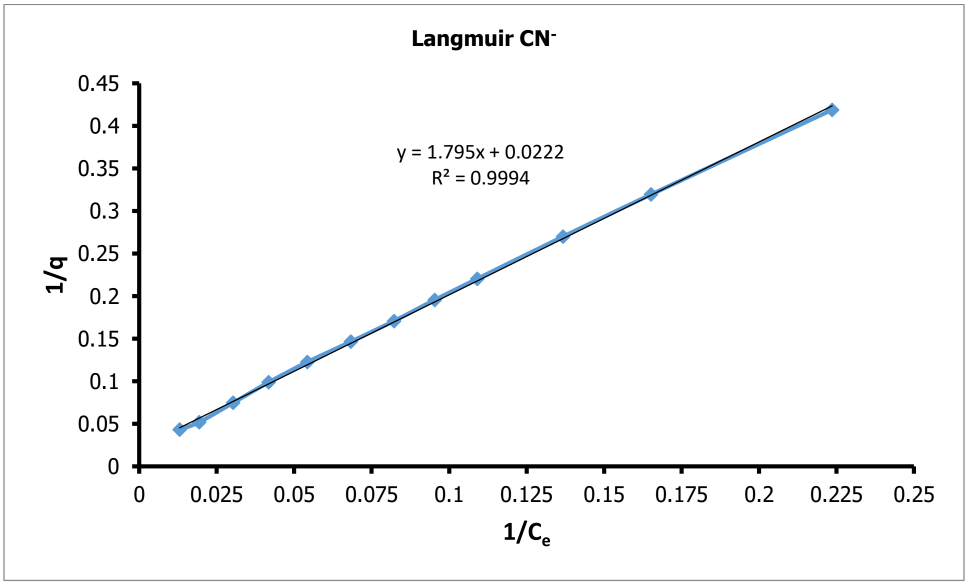

Fig. 7. Langmuir isotherm model

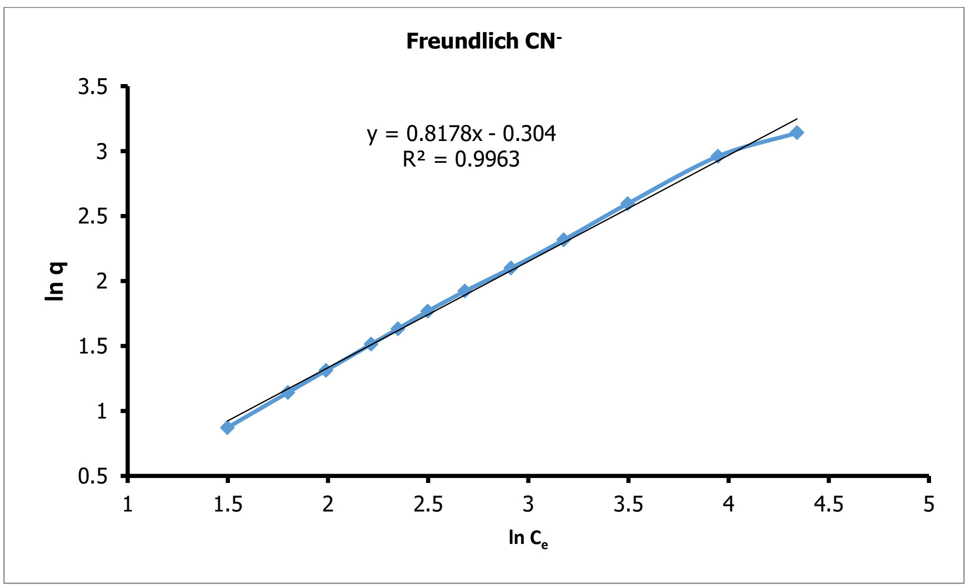

Fig. 8. Freundlich isotherm model

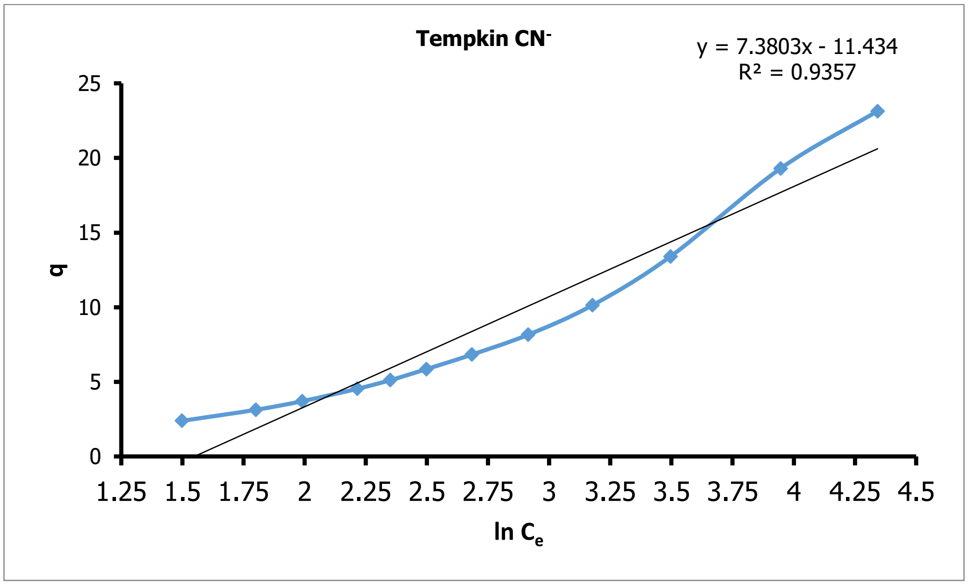

Fig. 9 Temkin isotherm model

Table 2. Constants of isothermal models used in the current study

Langmuir isotherm model				Freundlich isotherm model			Temkin isotherm model		
q_{max}	K_L	R_L	R^2	K_F	n	R^2	K_T	b	R^2
45.045	0.0124	0.447	0.9994	0.7379	1.2223	0.9963	0.9691	0.363	0.9357

2.7.2 Kinetic study

The kinetic study is one of the basic tools for understanding the mechanism and process of adsorption in a comprehensive manner, as it aims to determine the speed of removing pollutants from aqueous solutions and to study the effect of different factors such as time, concentration, and temperature on the adsorption rate. Kinetic studies are of countless reputation in enhancing the effectiveness of adsorbents, as they aid to allocate the optimal conditions that guarantee maximum elimination of contaminant ions or molecules with the least possible time and effort. Through multiple kinetic models, researchers can have exact details about how the contaminant interacts with the adsorbent, whether through diffusion within the molecules or across the surface. Among the most extensively utilised models in kinetic studies is the pseudo-first-order kinetic model, which presumes that the rate of adsorption is proportionate to the concentration of the pollutant in the solution at the start of the process. This model presumes that adsorption occurs principally on the surfaces of the adsorbent, and that the rate of reaction is not impacted by other factors beyond a specific point. The pseudo-second-order kinetic model is based on the statement that the interaction between the pollutant and the adsorbent depends primarily on the chemical interactions between the pollutant ions and the active site on the adsorbent (Shadhan *et al.*, 2024). This model specifies that the final adsorption rate is faster at first, then slows down as the concentration increases until equilibrium is reached. This model is considered more precise in enlightening the adsorption finding that happen as a result of the chemical interactions between the pollutant and the adsorbent. The intra-particle diffusion model emphases on the supposition that the adsorption process is not limited to the surface of the adsorbent, but also comprises the diffusion of pollutant ions within the adsorbent itself. This model displays that diffusion within the molecular pores of the adsorbent may be the limiting factor in the adsorption rate in some cases, particularly when the adsorbent has small pores, which hampers the diffusion process and diminishes the adsorption competence. Lastly, the Elovich model mirrors the assumption that adsorption does not occur steadily but changes over time, and is more flexible in labeling adsorption behavior in cases involving changes in the surface activity of the adsorbent. This model is grounded on the hypothesis that there are complicated interactions between the contaminant ions and the adsorbent, where the adsorption capacity may not persist constant throughout the process, but rather declines with time. The Elovich model is beneficial in adsorption cases that include multi-step reactions or reactions of a nonlinear nature, such as those happening in adsorption systems on porous materials. **Table 3** shows information of the kinetic models used in this study (Ali *et al.*, 2023).

Table 3. Information of kinetic models

Kinetic Model	Form of model's equation		Slop Term	Intercept Term
	Differential	Linear		
Pseudo first order	$\frac{dq_t}{dt} = k_1(q_e - q_t)$	$ln\,(q_e - q_t) = ln\,q_e - k_1 t$	$-k_1$	$ln\,q_e$
Pseudo second order	$\frac{dq_t}{dt} = k_2(q_e - q_t)^2$	$\frac{t}{q_t} = \frac{1}{k_2 {q_e}^2} + \frac{1}{q_e} t$	$\frac{1}{q_e}$	$\frac{1}{k_2 {q_e}^2}$
Elovich model	$\frac{dq_t}{dt} = \alpha e^{-\beta q_t}$	$q_t = \frac{1}{\beta} ln\,\alpha\beta + \frac{1}{\beta} ln\,t$	$\frac{1}{\beta}$	$\frac{1}{\beta} ln\,\alpha\beta$
Intra-particle diffusion	–	$q_t = k_P t^{0.5} + I$	k_P	I

The findings of the kinetic study are pictured in **Figures 10-13** and **Table 4**. These signify that the pseudo second order kinetic model is the most consistent with the obtained data, as it obtained the highest correlation coefficient of 0.9999, indicating that it is the most accurate model in explaining the adsorption behavior. This signifies that the adsorption process in this case is controlled by chemical interactions between the contaminant ions and the active sites on the surface of the adsorbent. In this model, the final adsorption rate is presumed to slow down over time as the reaction growths, with the system reaching equilibrium when the adsorption process is completed. This model is perfect for adsorption cases that happen as a result of chemical interactions between the pollutant and the adsorbent. In contrast, although the pseudo first-order kinetic model displays a high correlation coefficient of 0.9968, it was in lesser agreement with the data than the second-order model. This highlights that the first-order model is not adequate to fully elucidate the adsorption behavior, as this model is based on the supposition that the velocity is only linked to the concentration of the pollutant ions in the solution, and does not considered the complicated chemical interactions that may happen between the adsorbent and the pollutant. Thus, the first-order model may only be appropriate for short periods of adsorption before the process reaches equilibrium. As for the Elovich model, it displays a correlation coefficient value of 0.9974, which is a good figure mirroring a good agreement with the data, demonstrating that it can define the adsorption behavior in cases characterized by nonlinear or complex interactions. The Elovich model presumes that the surface activity of the adsorbent changes with time, ensuing in a change in the rate of adsorption, where adsorption starts quickly and then progressively declines as the process progresses. This model is therefore proper for defining processes involving multiple reactions or non-stationary modes of adsorption. Consequently, the intra-particle diffusion model stands out with the lowermost correlation coefficient among the models, at 0.9296. This specifies that diffusion within the molecular pores of the adsorbent is not the leading factor in the adsorption process in this study. Although intra-particle diffusion can be significant in some cases with microporous adsorbents, the outcomes of this investigation elucidate that surface interactions and chemical interactions between the contaminant and the adsorbent are more central. Referring to these analyses, it is obvious that the pseudo second order kinetic model is the best fit to the kinetic data, highlighting that the chemical interaction between the pollutant and the adsorbent is the main factor shaping the adsorption rate.

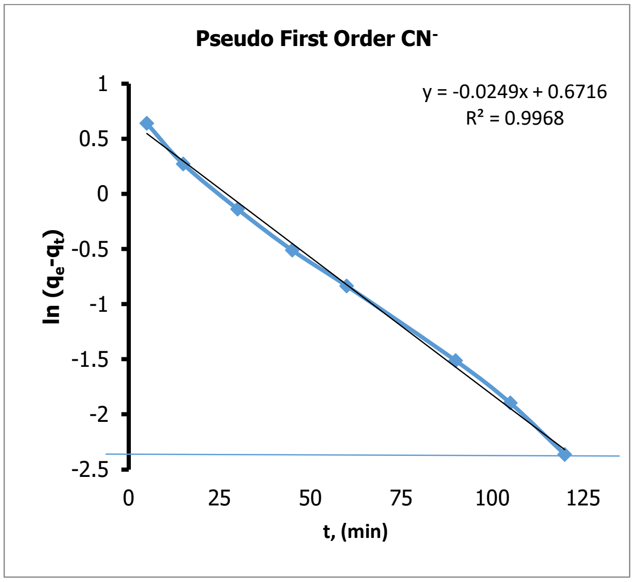

Fig. 10. Pseudo First Order Model

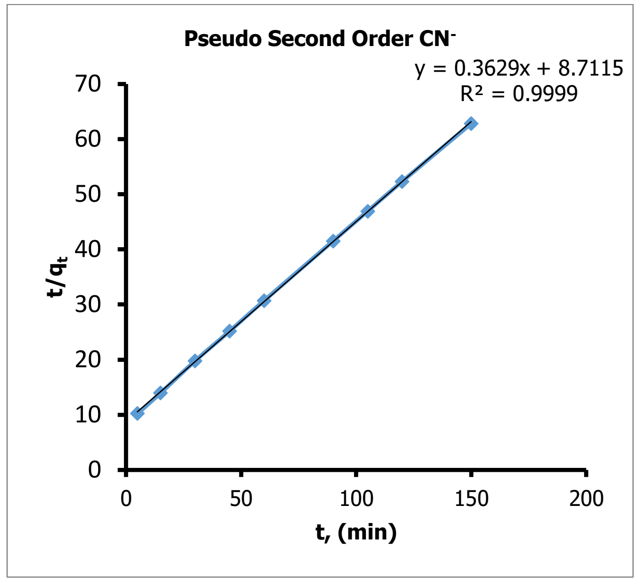

Fig. 11. Pseudo Second Order Model

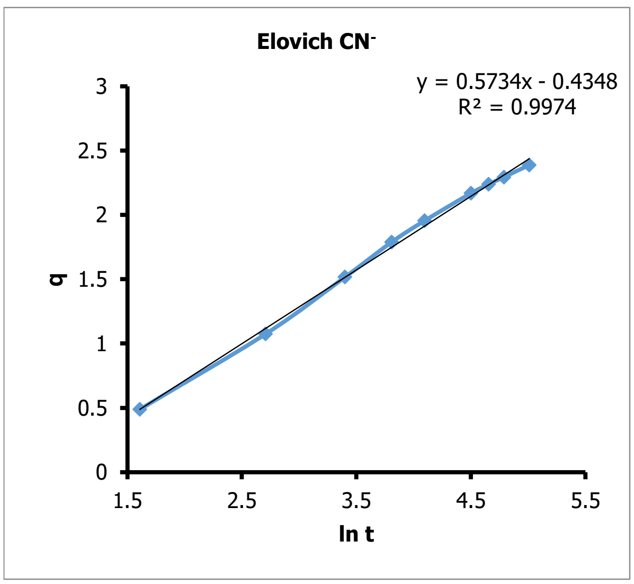

Fig. 12 .Elovich Model

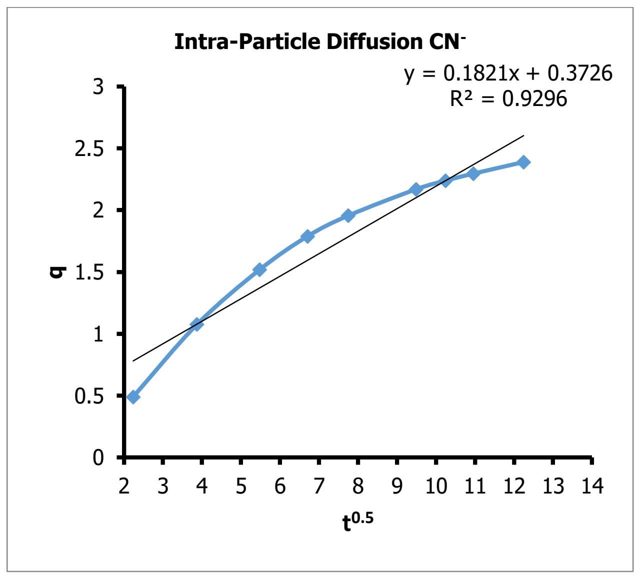

Fig. 13. Intra-Particle Diffusion Model

Table 4. Constants of kinetics models

Pseudo first order			**Pseudo second order**			**Elovich model**			**Intra-particle diffusion**		
k_1	q_e	R^2	k_2	q_e	R^2	α	β	R^2	k_P	I	R^2
0.0249	1.9573	0.9968	0.0151	2.7556	0.9999	0.2686	1.744	0.9974	0.1821	0.3726	0.9296

2.7.3 Thermodynamic study

Thermodynamic study is an essential part of understanding adsorption processes, as it helps to determine the nature of the interaction between ions and adsorbents, and offers information about the spontaneity of the process, the energy of the interaction, and the changes in randomness and order. This study depends on determining the distribution constants such as kd and different temperatures, in addition to calculating the values of enthalpy (ΔH), free energy (ΔG), and entropy (ΔS) to evaluate the thermodynamics of the reaction. Eqs. 3 and 4 are used to measure the free energy and the distribution constant at adsorption equilibrium, where negative values of ΔG indicate a spontaneous reaction, while positive values of ΔH indicate an endothermic process (Ali *et al.*, 2023; Shadhan *et al.*, 2024).

$$\Delta G = \Delta H - T\Delta S \quad (3)$$

$$ln\ k_d\ = -\frac{\Delta H}{R}\frac{1}{T} + \frac{\Delta S}{R} \quad (4)$$

The results of the thermodynamic study of cyanide ion removal using buckthorn leaves show that the adsorption process is significantly affected by increasing temperature (**Table 5**), as the distribution constant increases with increasing operational temperature from 25 °C to 50 °C, signifying that the adsorption becomes more efficient at higher temperatures. This reflects the endothermic nature of the process, as the process requires energy for the reaction between the ions and the adsorbent to occur, as evidenced by the constant value of the enthalpy ΔH, which is estimated at 159.214 kJ/mol. This means that the reaction takes place in an environment with added energy, which in turn enhances the reaction at high temperatures. As for the entropy ΔS, it remains constant at 588.64 J/mol K at different temperatures, indicating that the process leads to an increase in randomness or chaos in the system. This increase in randomness can be interpreted as an expression of the scattering of ions on the surface of the adsorbent after the reaction, reflecting a state of dynamic stability that enhances the capacity of the adsorbent to adsorb ions. On the other hand, the free energy values ΔG show interesting results, as all values are negative, which resources that the adsorption process is spontaneous and its spontaneity rises with growing temperature, as the free energy values range between -16.29 kJ/mol at 25 °C to -31.01 kJ/mol at 50 °C. This is an indication that the reaction becomes more stable and occurs faster with increasing temperature. Referring to these results, it can be stated that the adsorption of cyanide ions by buckthorn leaves is an endothermic process, and its effectiveness and efficiency increase with increasing temperature. **Table 6**. Shows a comparison between the obtained results with resuls reported in literature.

Table 5. Thermodynamic properties of cyanide adsorption using buckthorn leaves

Temperature, (K)	ΔH (kJ/mol)	ΔS (J/mol K)	ΔG (KJ/mol)
25			-11.2692
27.5			-12.6987
30			-14.1282
32.5			-15.5577
35			-16.9872
37.5	159.2136	571.8022	-18.4167
40			-19.8462
42.5			-21.2757
45			-22.7052
47.5			-25.5642
50			-11.2692

Table 6. a comparison between the obtained results with resuls reported in literature.

adsorbent used	conditions	uptake capacity	Reference
Activated carbon	pH=4 T=45 C Dose=20g/L t=48 h C_{in}=50 mg/L	7.91 mg/g	(Dash *et al.*, 2009)
pistachio hull wastes	pH=10 C_{in}=100 mg/L Dose=1.5g/L t=60 min	156.2 mg/g	(Moussavi *et al.*,2010)
nanocrystalline ZnO/NiO	pH=10 C_{in}=2.5 mg/L Dose=1.0g t=9 min	0.245 mg/g	(Jawad *et al.*, 2020)
Tectona grandis leaves powder	pH=7 T=45 C Dose=20g/L t=120 min C_{in}=100 mg/L	18.45 mg/g	(Dwivedi *et al.*, 2016)

Conclusions

The results of the current study elaborated that buckthorn leaves are an operative and low-cost adsorbent for the elimination of cyanide ions from contaminated aqueous solutions, which enhances their use as a sustainable environmental solution for water treatment. The experiments signified the actual effects of the operational variables on the cyanide removal efficiency. Specifically, it was found that increasing the pH significantly improves the adsorption, with maximum removal efficiency achieved at basic pH values. Also, it was concluded that the agitation speed affects the distribution of the adsorbent and improves the removal efficiency up to a saturation point at certain speeds. As for the initial cyanide concentration, the findings elucidated that the removal efficiency decreases with increasing the initial concentration, while the adsorption efficiency was directly related to temperature and contact time. More importantly, the optimum removal was achieved at the optimum operating conditions of 8, 400 rpm, 100 ppm, 4 g, 150 min, 50 °C for pH, agitation speed, initial concentration of cyanide, adsorbent dosage, contact time and temperature, respectively. Isothermal studies revealed a good agreement with the Langmuir model, indicating that adsorption occurs on a homogeneous surface with a maximum of active sites. The kinetic study showed a strong agreement with the pseudo-second-order model, highlighting that the process is controlled by chemical adsorption. The thermodynamic study confirmed that the adsorption process is spontaneous and endothermic, with the adsorption efficiency increasing with increasing temperature, with negative Gibbs free energy values and positive values for both enthalpy and entropy, reflecting the increased randomness of the system and the preference for reaction at higher temperatures. Overall, this study demonstrated that sea buckthorn leaves represent a promising and effective solution for treating cyanide-contaminated water, opening new horizons for their application in industrial and environmental systems, especially in areas requiring sustainable and low-cost solutions.

Conflict of Interest

The author declares that he has no conflict of interest

Nomenclature

R	the removal efficiency of cyanide ions	[%]
$C_{\circ}$	the initial concentrations of cyanide	[ppm]
C_f	the final concentrations of cyanide	[ppm]
q	the adsorption capacity of buckthorn leaves for cyanide ions, expressed in	$[\frac{\text{mg}}{\text{g}}]$
V	the volume of the solution	[L]
m	the mass of buckthorn leaves exploiting in each experiment	[g]
rpm	revolution per minute.	[rpm/min]
C_e	the equilibrium adsorbed concentration	$[mg/g]$
q_e	the adsorption capacity at equilibrium state	$[mg/g]$
q_{max}	the maximum adsorption capacity of Langmuir model	$[mg/g]$
R_L	the separation factor in Langmuir model	[dimensionless]
K_L	the constant of Langmuir adsorption isotherm model, expressed the binding sites	$[l/\text{m}g]$
K_F	the constant of Freundlich adsorption isotherm model	$[(mg/g).(l/\text{m}g)^{1/n}]$
n	the intensity of the adsorption in Freundlich model	[dimensionless]
K_T	the Temkin isotherm equilibrium binding constant	$[l/\text{m}g]$
b	constant in Temkin isotherm model	[dimensionless]
R	the universal gas constant	$[8.3144\, J/mol.K]$
T	the absolute temperature	[K]
q_t	is the adsorption capacity at any time	$[mg/g]$

k_1	the first order rate constant	[min^{-1}]
k_2	the second order rate constant	$[g/mg.min]$
α	the initial adsorption rate in Elovich model	$[mg/g.min]$
k_P	the rate constant in intra-particle diffusion model	$[mg/g.min^{0.5}]$
β	the desorption constant in Elovich model	$[g/mg]$
I	the thickness of boundary layer	$[mg/g]$
k_d	the adsorption equilibrium coefficient	[–]
ΔG	the Gibbs free energy	$[kJ/mol]$
ΔH	the enthalpy change	$[kJ/mol]$
ΔS	the entropy change	$[J/mol\ K]$

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