











ORIGINAL

Thermodynamic and Kinetic Assessment of Cobalt II Adsorption Using Green Synthesized NiO/ γ -Al₂O₃ Nanoparticles

Evaluación termodinámica y cinética de la adsorción de cobalto II mediante nanopartículas de NiO/ γ -Al₂O₃ sintetizadas ecológicamente

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Cite as: Hassan KH, Al-hussaniy HA, Ibraim Oraibi A, Bashar Al-Qazzan M, Zainee HY, Al-Jashamy K, et al. Thermodynamic and Kinetic Assessment of Cobalt II Adsorption Using Green Synthesized NiO/ γ -Al₂O₃ Nanoparticles. Salud, Ciencia y Tecnología - Serie de Conferencias. 2025; 4:1167. <https://doi.org/10.56294/sctconf20251167>

Submitted: 26-02-2024

Revised: 12-08-2024

Accepted: 22-12-2024

Published: 01-01-2025

Editor: Prof. Dr. William Castillo-González 

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ABSTRACT

Introduction: disposal of heavy metals into the water as a result of industrial development might cause a threat Health and the environment.

aim of this study was to assess the uptake of Co²⁺ from aqueous solutions via NiO/ γ -Al₂O₃ nano catalysts.

Method: the main variables that affect the percentage of metal removal were assessed. It took about 50 minutes to attain equilibrium for the elimination of Co²⁺ ions. It was discovered that raising the adsorbate concentration and increasing the surface weight somewhat lowered the removal of cobalt ions.

Results: the removal of cobalt ions was shown to depend on temperature, due to ecothermic natural of this proress increasing temperature associated with decrease the elimination.

Conclusions: the adsorption seems to be spontaneous, exothermic, and less random according to calculated values of the thermodynamic functions (ΔG , ΔH , and ΔS) of the adsorption. After the data were fitted into a number of kinetic models, including the Elovich model, pseudo-first order, pseudo-second order, and intraparticle diffusion equations, it was discovered that the pseudo-second-order model performed the best at describing the adsorption, with a high correlation factor (R²).

Keywords: Thermodynamics; Kinetics; Adsorption; Cobalt II; Nio/ γ -Al₂O₃ Nanoparticles.

RESUMEN

Introducción: la eliminación de metales pesados en el agua como resultado del desarrollo industrial puede causar una amenaza para la salud y el medio ambiente.

El objetivo de este estudio fue evaluar la absorción de Co²⁺ de soluciones acuosas a través de nanocatalizadores de NiO/ γ -Al₂O₃.

Método: se evaluaron las principales variables que afectan el porcentaje de eliminación de metales. Se tardó aproximadamente 50 minutos en alcanzar el equilibrio para la eliminación de iones Co²⁺. Se descubrió que aumentar la concentración de adsorbato y aumentar el peso de la superficie reducía un poco la eliminación de iones de cobalto.

Resultados: se demostró que la eliminación de iones de cobalto depende de la temperatura, debido a la

naturaleza ecotérmica de este proceso, el aumento de la temperatura se asocia con una disminución de la eliminación.

Conclusiones: la adsorción parece ser espontánea, exotérmica y menos aleatoria según los valores calculados de las funciones termodinámicas (ΔG , ΔH y ΔS) de la adsorción. Después de ajustar los datos a una serie de modelos cinéticos, incluido el modelo de Elovich, el pseudo-primer orden, el pseudo-segundo orden y las ecuaciones de difusión intrapartícula, se descubrió que el modelo pseudo-segundo orden tenía el mejor desempeño para describir la adsorción, con un alto factor de correlación (R^2).

Palabras clave: Termodinámica; Cinética; Adsorción; Cobalto II; Nanopartículas de NiO/ γ -Al₂O₃.

INTRODUCTION

The biological processes produce large amounts of wastewater, which represents a serious threat to the survival of living things in the environment since it is contaminated with cobalt and its compounds. The compounds of cobalt are an element in an environment in its mineral structural forms like arsenicals, arsenic hydrates, and even oxides.⁽¹⁾ There is a biological toxic effect of high concentrations of cobalt compounds on living organisms. Printing and ceramic industry working people who use cobalt dyes as paint materials, anti-insect, and agriculture might suffer from health problems.^(2,3) However, ways to remove cobalt ions from aqueous media and water are done by precipitation means, electrochemical techniques, and membrane processes and are most commonly used of adsorption and ion exchange.^(4,5,6,7) Basically, the surface of the nanoparticle's functionality might show a high surface area to increase both adsorption capacity and efficiency.^(8,9) Recently, researchers reported various nanomaterial particles for heavy metal removal by adsorption. Therefore, heavy metals might adsorb easily from aqueous systems using nano-size metal oxides like ferric oxides, manganese, cerium, and aluminum oxides.⁽¹⁰⁾ In recent years, extensive and rapid technological advances have been introduced in the field of nanotechnology. This evolution has raised a considerable demand for the production of efficient adsorbents for removing some toxic as well as heavy metal pollutants. It is quite imperative to focus on the principal advantages of nanoparticles adsorption technologies for removing cobalt ions from industrial wastewater. Nickel oxide nanoparticles garnished γ -aluminum oxide nanoparticles were synthesized and their adsorption efficiency was employed to expunge poisonous cobalt metal ions from an artificial wastewater. The nanoparticles were green synthesized via a chemical method using AlCl₃ to feed a colloidal mixture solution acting as γ -langoustine seeds and the gene principle of natural mimic the enzymatic technology was employed to grow nickel oxide nanoparticles.⁽¹¹⁾ The nanomaterial particles were intensively experimented with as very active and efficient absorbents to remove many heavy metal ions.⁽¹¹⁾ Cobalt removal studies employed magnetite-based nanocomposite.⁽¹²⁾ The present study aims to assess the Co²⁺ uptake from aqueous solutions via (NiO/ γ -Al₂O₃) nano catalysts.

METHOD

Adsorbates of Study

An adsorption process involves the surface accumulation of an adsorbate (in this case, Co²⁺) on a porous solid (the adsorbent, known here as nanoparticles), followed by its molecular bonding under various applied conditions (temperature, concentration). For Co²⁺ ions adsorption, functionality such as NiO/ γ -Al₂O₃ or any combination of its components was used as adsorbents in the literature so far. In this work, NiO/ γ -Al₂O₃ nanoparticles are considered, which are practical as well as economically important adsorbents. Hydroxy forms of Ni- and Fe-based hydrotalcite-like materials (eco-friendly synthesized ones), as well as other simple binary-based eco-synthesized metal oxides, are also good and efficient/desirable adsorbents for Co²⁺ removal from wastewaters. Several kinds of metal oxides (binary, nickel foam cartridges modified with iron oxide or magnetite) adsorbents for Co²⁺ recovery have been described in the literature with positively received outcomes. From the majority of this kind of literature, it is evident that there is no detailed work on the Co²⁺ ions adsorption performed from a thermodynamics or kinetics point of view (like we have done in this work) with synthesized NiO/ γ -Al₂O₃ nanoparticles.

Adsorbent of Study

The adsorbent used is Nano NiO/ γ -Al₂O₃ catalyst, which was prepared and characterized previously from nickel oxide nanoparticles that were synthesized by the Arundo Donax Leaves Extract route and alumina (γ -Al₂O₃) being prepared by co-precipitation method.⁽¹³⁾

Factors Affecting the Adsorption

In this investigation, the important factors that influenced the study were the amount of NiO/-Al₂O₃

adsorbent, the influence of the original concentration of Co^{+2} ions, and the role of temperature. To study the impact of temperature on adsorption, Co(II) ion removal from aqueous solutions is performed in a series of adsorption experiments at various temperatures (293, 303, 313, 323, and 333K).

Metal Removal Methods

The previously published formula was used to compute the removal percent of metal ($R\%$)^(14,15).

Kinetic Adsorption of Co^{+2} ions

At a regular interval of 5, 15, 25, 35, and 45 minutes, (0,1) g of produced $\text{NiO} / \gamma\text{-Al}_2\text{O}_3$ is shaken at a speed of 150 rpm at a temperature of 298 K to study the kinetics of cobalt ion adsorption. Once again, the experiment was conducted using the adsorbent at temperatures (308, 313, 318, and 323) K. The concentration of eliminated Co^{+2} ions was then determined using the same protocols as before. The (pseudo/first order) is one of four kinetic models and adsorbate/adsorbent interaction, were used to assess the order of the interaction. Pseudo/second order: intra particle diffusion: and finally, Elovich models one) described by equations (1-4).

$$[\ln(q_e - q_t)] = (\ln q_e) - (k_1 t) \quad (1)$$

$$[t/q_t] = 1/(k_2 q^2) + [1/(q_e)]t \quad (2)$$

$$(q_t) = k_d t^{1/2} + (\text{constant, } C) \quad (3)$$

$$qt = \beta \ln(\alpha \beta) + \beta \ln t \quad (4)$$

RESULTS

Adsorption of Co(II) ions on $\text{NiO} / \gamma\text{-Al}_2\text{O}_3$ NanoCatalyst

Effect adsorption on time of contact

The influence of contacting time on the Co^{+2} ion's adsorption on ($\text{NiO} / \gamma\text{-Al}_2\text{O}_3$) revealed that the adsorption equilibrium time is almost (50) min. The percentage removal of Co^{+2} ions from surfaces also decreases at the beginning of contact time before gradually increasing, with the quick initial increase in rate followed by a slowing in rate at the later period, which may be caused by the presence of extra adsorption on the adsorbent materials sites.^(14,15,16) According to the authors, the first high adsorption rate may have been caused by ion exchange, which was achieved by a gradual reaction between the sample ions' active groups as the available empty surface positions could not be filled easily because of repulsive force.⁽¹⁷⁾ To do so, metal ions must migrate much further and also deeper into the pore where it under go or encounter much greater resistance.⁽¹⁷⁾

Role of Amount of Adsorbent Quantity on Adsorption

The impact of adsorbent amount on the removal, Co^{+2} ions on ($\text{NiO} / \gamma\text{-Al}_2\text{O}_3$) was investigated via different amounts quantity (0,06- 0,08- 0,1- 0,12 and 0,15)g and at 298K with the same Co^{+2} ions concentration (100ppm), time of contact being (50) min. For the effect of surface amount on the absorption of metal ions demonstrates that the removal of metals increases along with the amount of $\text{NiO} / \gamma\text{-Al}_2\text{O}_3$ prepared using the [Arundo Leaves Extract method]. This increase indicates that the concentration of oxide nanoparticles increases to the percentage of metal elimination from the surface due to an increase in surface area.

Effect of Original Adsorbate Amount on Adsorption

Co^{+2} ions adsorption on $\text{NiO} / \gamma\text{-Al}_2\text{O}_3$ was first investigated at optimal conditions using different initial concentrations (50-100-150-200 and 250) ppm of the aqueous solution. The data showed that the initial concentration rise caused a decrease in removal. Since the nanoparticles have a big surface area and are tiny in size, they have a large number of active adsorption sites that are available for adsorption. These factors may be the cause of the drop in removal % at higher doses.⁽¹⁸⁾

Temperature effect on adsorption

The temperature might affect the extent of Co(II) ion adsorption on $\text{NiO} / \gamma\text{-Al}_2\text{O}_3$ was investigated at different temperatures that involved 293, 303, 313, 323, and 333K with an initial concentration of (100) mg/l, (0,1g) is the quantity of adsorbent, fixed time of contact (50min). Experimental data and Co (II) removing shape are presented (table 1 and figure 1).

It is obvious that the removal rate is lower with the temperature increasing. Based on these data, it can be observed that the adsorption is an exothermic process in studied cases; the lowering of the removal by adsorption raise by increasing temperature might be due to the weakness of the force of interaction between adsorption surface active sites and adsorbate ions.⁽¹⁹⁾

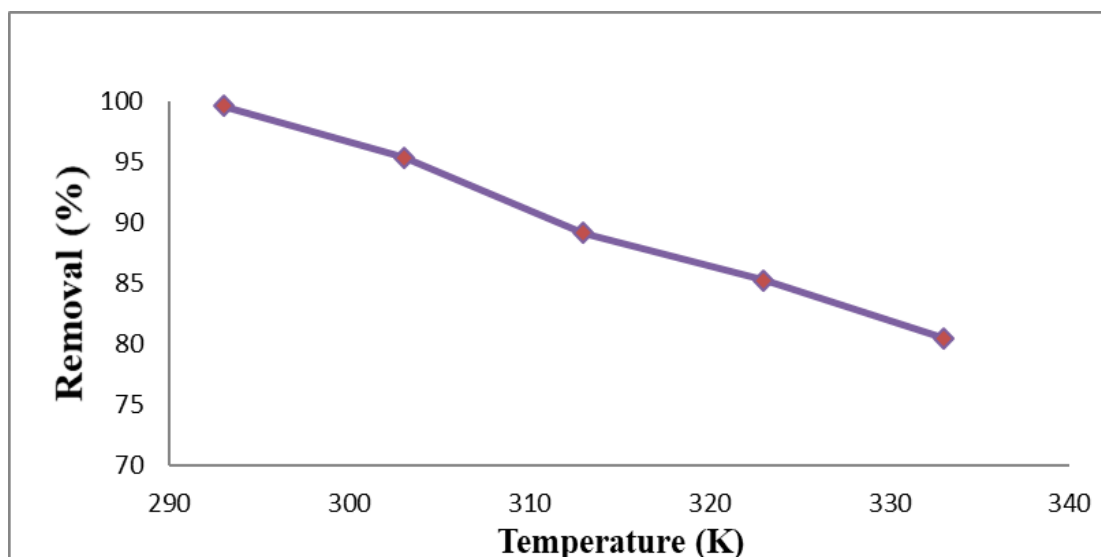
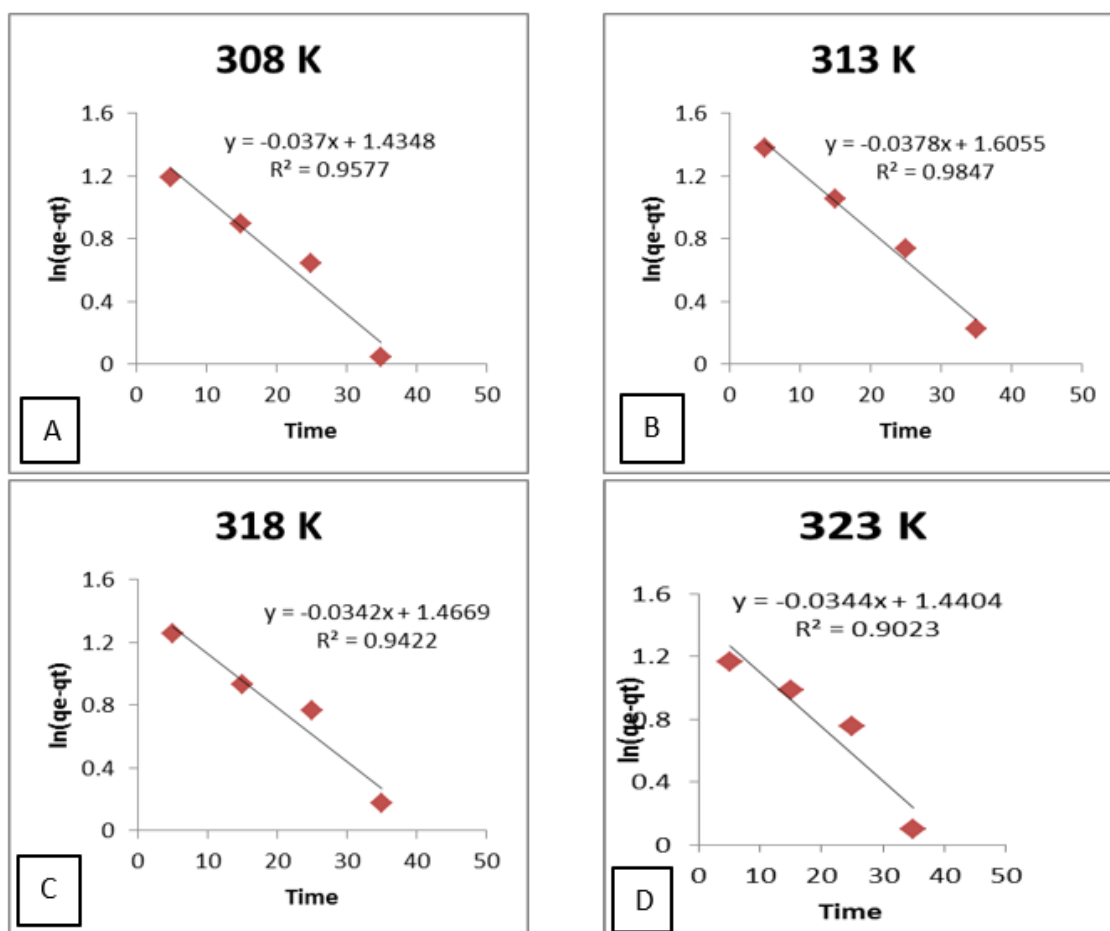


Figure 1. Relation of temperature and percent removal of Co (II) ion on NiO/ γ -Al₂O₃ nano catalyst

Kinetics Studies of Adsorption Process for Cu+2 and Co+2 ions

Utilizing the experimental circumstances of an initial concentration of 100 ppm, several temperatures (308-313-318- and 323), 150 rpm, and time intervals (5-15-25-35- and 45) minutes, an adsorption kinetic analysis was conducted. C_e represents the concentration of the ions followed the adsorption in (t) time (ppm), q_t is the value adsorbed at t time (ppm), q_e is the adsorbed quantity at equilibrium (mg/g, ppm), $q_e - q_t$, $\ln(q_e - q_t)$, t which is time (min), $t^{1/2}$ square root of time (min^{1/2}), These data were applied for four kinetic models, which are pseudo-first order, pseudo-second order, Intraparticle diffusion, and Elovich models described by equations 2, 3, 4, and 5 with the plots shown in figures 2, and 3 in different temperatures for the four mention models respectively.



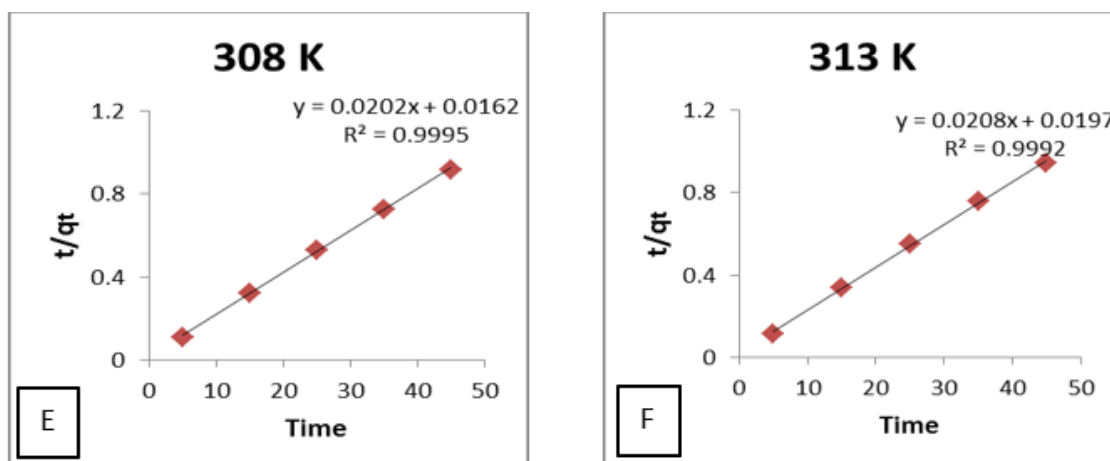
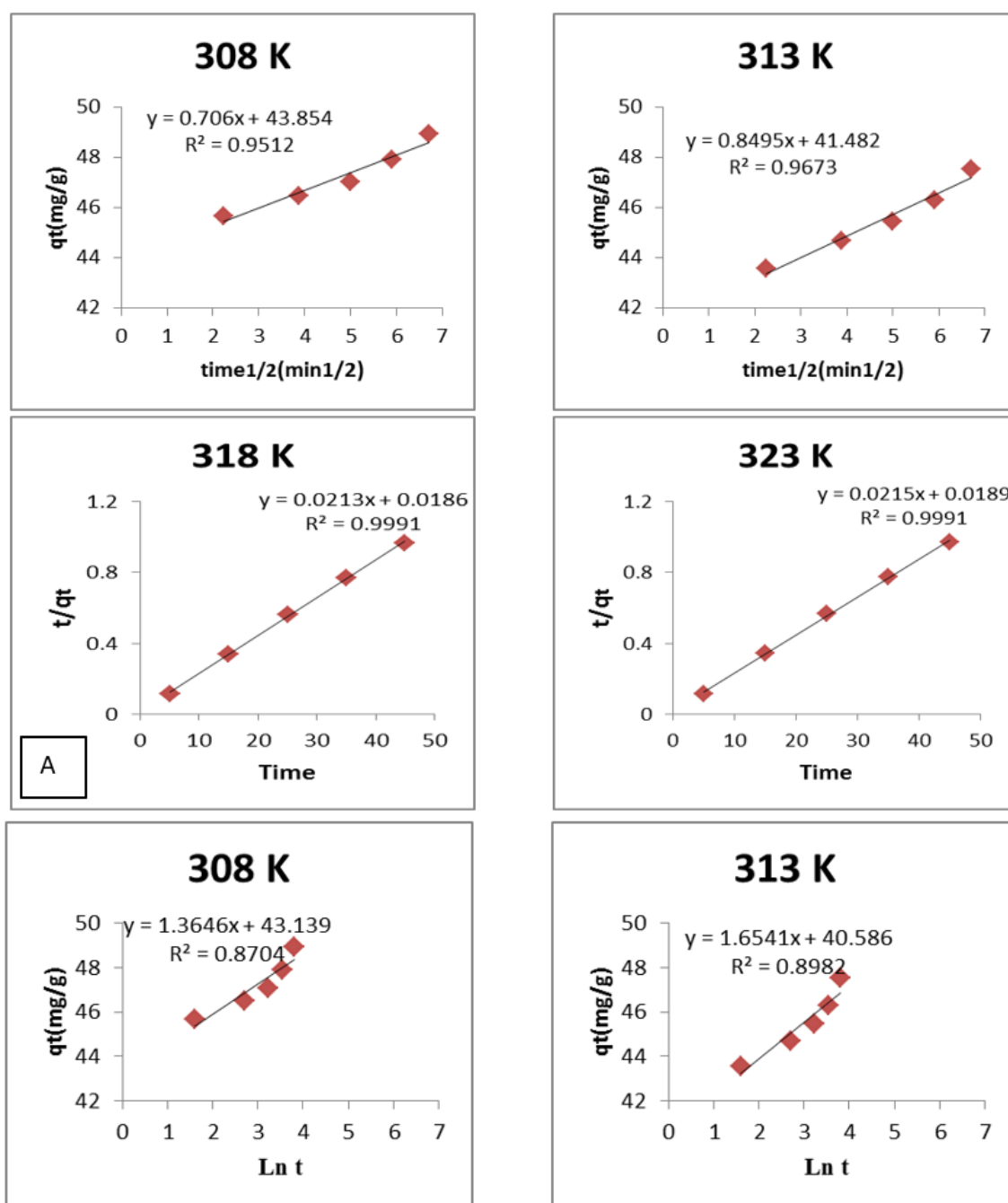


Figure 2. The Co+2 adsorption on NiO/γ-Al₂O₃ nanocatalyst: from (A- D) pseudo-1st-order kinetic model, (E , F) pseudo-2nd-order kinetic model



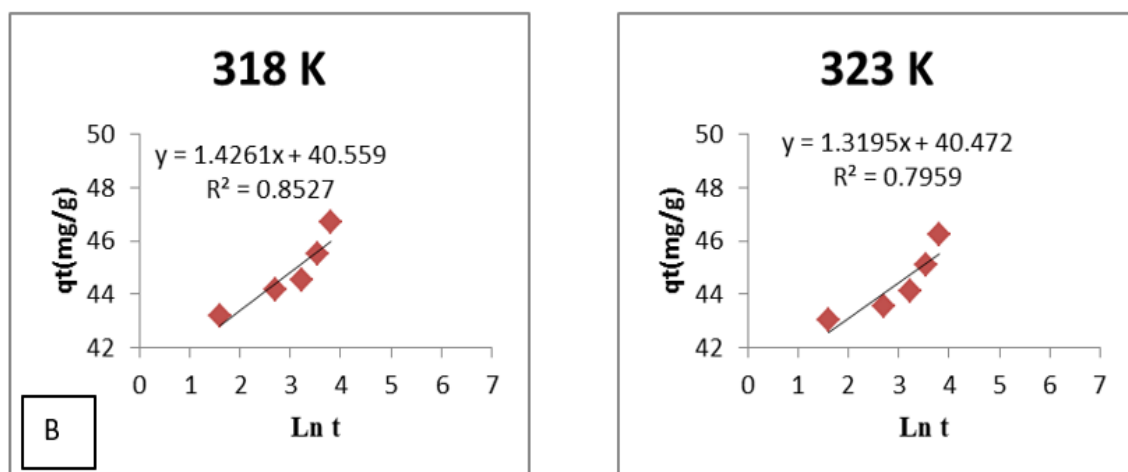


Figure 3. Diffusion model (A) and Elovich Mode (B) of intraparticle for Co+2 adsorption on NiO/γ-Al₂O₃ nanocatalyst

The values (k_1 ; k_2 ; k_p ; $k_{elovich}$) and correlation coefficients of adsorption for Co(²⁺) on NiO/γ-Al₂O₃ adsorbents are given in table 1. The results show that the pseudo-2nd-order correlation coefficient is more than one of pseudo-1st, intraparticle diffusion models and even Elovich model.

T(K)	q _e (Pseudo 1 st order		Pseudo-2 nd order		Intra particle-diffusion		Elovich-model	
		K ₁ in 1/min	(R) ²	K ₂ in g/mg.min	(R) ²	K _d in mg/g.min ⁻¹	(R) ²	k _{elovich} in (mg/g.s)	(R) ²
308,0	48,9450	-0,0370	0,9577	0,0252	0,9995	0,7059	0,9512	73132	0,8704
313,0	47,5515	-0,0378	0,9847	0,0220	0,9992	0,8494	0,9673	74733	0,8982
318,0	46,6895	-0,0342	0,9422	0,0243	0,9991	0,7385	0,9339	32029	0,8527
323,0	46,2300	-0,0344	0,9023	0,02433	0,9991	0,6955	0,9029	27612	0,7959

Thermodynamic study of adsorption for NiO/γ-Al₂O₃ nanocatalyst

The impact of temperature on the removal of Co²⁺ ions from NiO/γ-Al₂O₃ at different temperatures that included 293, 303, 313, 323, and 333 K was tested and used to evaluate the change in the thermodynamic basis function of the free-energy; [ΔG], enthalpy; [ΔH] and also the entropy; [ΔS] of adsorption. The equilibrium constant [K] is well-thermodynamically explained using the equation of Van 't Hoff (figure 4). The value of equilibrium constants[K] was computed at any studied temperature by ⁽²³⁾. The Gibbs-free-energy change [ΔG°] has been evaluated according to the previously reported equation ⁽²⁴⁾. Finally, values of (ΔH) and (ΔS) were calculated from both slope & intercept by plotting (lnk) vs (1/T).

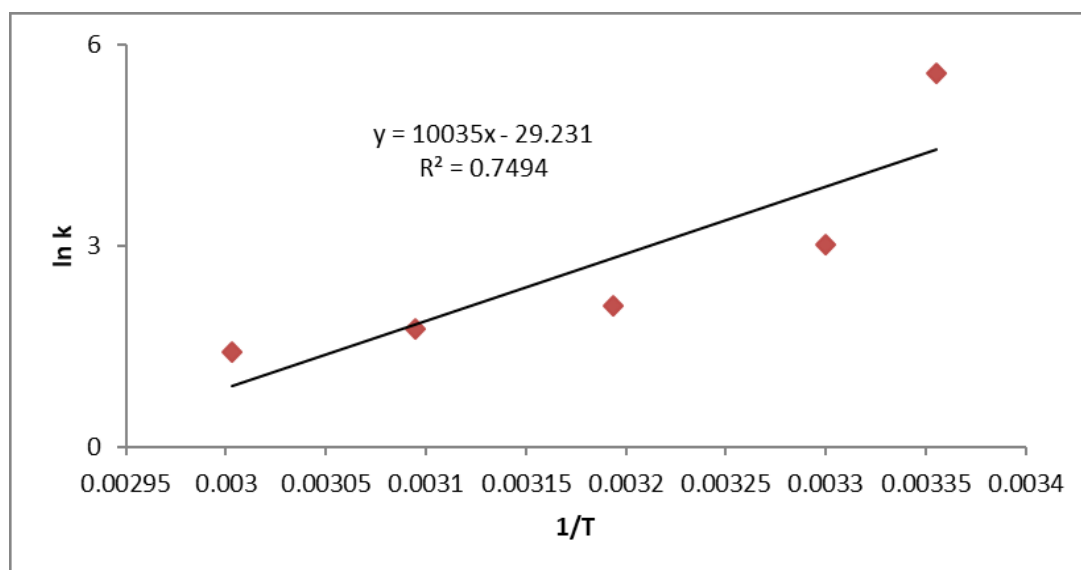


Figure 4. Plot of Van-'t Hoff adsorption plot of Co (II) by NiO/γAl₂O₃ catalyst

Table 2 illustrates the variations in $[(\Delta G); (\Delta H); \text{ and } (\Delta S)]$, where the negative ΔG values validate the appropriateness and spontaneous nature of the adsorption process.^(25, 26) While negative $[\Delta S]$ implies a decrease in randomness at the (solid-solution) interface due to ion adsorption by NiO/ γ -Al₂O₃ catalysts, negative ΔH suggests that the adsorption of ions onto NiO/ γ -Al₂O₃ catalysts was an exothermic process.

Table 2. Values of the general thermodynamic function of the adsorption for Co (II) on NiO/ γ -Al ₂ O ₃ nanocatalysts under various temperatures				
Ions	Temperature in, (K)	$\Delta G, (KJ/mol)$	$\Delta H, (KJ/mol)$	$\Delta S, (KJ/mol.K)$
Co ⁺²	293	-13565,851	-83428,87	-243,0282
	303	-7609,3203		
	313	-5483,2684		
	323	-4711,0358		
	333	-3923,3292		

DISCUSSION

Thermodynamic Insights into Co(II) Adsorption

The thermodynamic assessment of Co(II) ion adsorption onto NiO/ γ -Al₂O₃ nanoparticles reveals significant insights into the nature and feasibility of the process. The negative values of Gibbs free energy change (ΔG) across all studied temperatures confirm the spontaneous nature of the adsorption process. The increasingly negative ΔG with decreasing temperature suggests that lower temperatures favor adsorption, reinforcing the exothermic nature of the reaction as indicated by the negative enthalpy change ($\Delta H = -83428,87$ KJ/mol). This exothermic behavior implies that the adsorption process releases heat, making it less efficient at higher temperatures where thermal agitation may disrupt the interaction between adsorbent and adsorbate.^(21,22)

Additionally, the entropy change ($\Delta S = -243,0282$ KJ/mol.K) is negative, indicating a decrease in randomness at the solid-liquid interface during adsorption. This can be attributed to the structured arrangement of Co(II) ions on the nanoparticle surface, reducing the system's overall disorder. The negative ΔS further suggests that the adsorption involves a more ordered process, possibly due to the specific interaction between Co(II) ions and active sites on the NiO/ γ -Al₂O₃ nanoparticles.^(23,24,25)

Kinetic Modeling and Adsorption Mechanism

The kinetic analysis of Co(II) adsorption demonstrates that the process follows a pseudo-second-order kinetic model more accurately than pseudo-first-order, intraparticle diffusion, and Elovich models. The high correlation factor (R^2) for the pseudo-second-order model indicates that the rate-limiting step may involve chemisorption, where the adsorption rate is proportional to the square of the number of unoccupied sites. This suggests that the adsorption mechanism is likely dominated by chemical interactions between Co(II) ions and the active sites on the NiO/ γ -Al₂O₃ nanoparticles.^(26,27)

The kinetic parameters obtained from the pseudo-second-order model further support the chemisorption mechanism, as they provide a better fit to the experimental data compared to other models. The intraparticle diffusion model, while useful, shows that diffusion is not the sole rate-limiting step, indicating that the overall adsorption process involves multiple stages, including surface adsorption and pore diffusion.⁽²⁸⁾

Impact of Adsorption Variables

The study identifies several key factors affecting Co(II) adsorption efficiency, including contact time, adsorbent dosage, initial ion concentration, and temperature. The equilibrium time for maximum adsorption is approximately 50 minutes, after which no significant increase in adsorption is observed. This rapid initial adsorption rate followed by a plateau suggests that the process is initially driven by the availability of active sites, which become saturated over time.^(28,29,30)

The adsorbent dosage positively influences Co(II) removal efficiency, with higher amounts of NiO/ γ -Al₂O₃ leading to increased adsorption due to the greater availability of active sites. However, at very high dosages, the removal efficiency plateaus, possibly due to overlapping of adsorption sites and agglomeration of nanoparticles, which reduces the effective surface area as in chen study.⁽³¹⁾

The initial concentration of Co(II) ions shows an inverse relationship with removal efficiency, where higher concentrations result in lower percentage removal. This can be attributed to the saturation of available adsorption sites at higher concentrations, indicating that the adsorbent's capacity is finite and can be overwhelmed by excessive amounts of adsorbate.^(32,33,34,35)

Temperature variations significantly impact the adsorption process, with higher temperatures leading to decreased removal efficiency. This reinforces the exothermic nature of the adsorption, where increased kinetic

energy at higher temperatures may hinder the stable interaction between Co(II) ions and the adsorbent surface.

CONCLUSION

It has been concluded from the present investigation that adsorption needs 50 minutes till removal of all Co²⁺ and get equilibrium. It also seems that raising the adsorbate concentration and increasing the surface weight somewhat decreased the removal of cobalt ions. It was discovered that the elimination of cobalt ions was temperature-dependent, and that the process was exothermic as the temperature was raised. The estimated values of the thermodynamic functions (ΔH and ΔS) of the adsorption are -83428,87 KJ/mole and -243,0282 KJ/mole for ΔH and ΔS , respectively. These values suggest that the adsorption is spontaneous, exothermic, and less random. Fitting the data into many kinetic model equations such as pseudo-first order / pseudo-second order / intraparticle diffusion and finally Elovich model demonstrated that pseudo-second-order describes adsorption with high correlation factor (R^2) and is better than other kinetic models.

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FINANCING

No financing.

CONFLICT OF INTEREST

None.

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