



## Original Article



# Kinetic, Isotherm and Thermodynamic Studies on the Removal of Zn (II) From Aqueous Solution using Silica Adsorbent

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Email: [hshe2503@rediffmail.com](mailto:hshe2503@rediffmail.com)**Abstract**

**Background:** Water is essential for all life, but pollution, including heavy metals, poses severe health risks. The adsorption technology, known for being safe, cost-effective, and eco-friendly, uses silica to remove zinc ions from water. This paper detailed the kinetics, isotherms, and thermodynamics of this process.

**Methods:** A stock solution of zinc sulfate in distilled water was prepared. Silica oxide, with its high surface area, was used to remove zinc ions from solutions of 5, 10, 15, and 20 mg/L concentrations. A double beam atomic absorption spectrophotometer (AAS) was used to obtain the calibration curve for the experiments.

**Results:** The study investigated the removal efficiency of Zn (II) using SiO<sub>2</sub> as an adsorbent. Factors such as initial concentration, contact time, adsorbent dose, pH, agitation speed, and temperature were examined in a batch process. Equilibrium was reached in 60 minutes for all concentrations.

**Conclusion:** The highest Zn (II) adsorption (88%) was achieved with a 10 mg/L solution at pH 6, 180 rpm, and 303 K, using 1 g/L of silica. The adsorption kinetics followed a first-order rate mechanism with a rate constant of  $3.91 \times 10^{-2}$  1/min at 30 °C. Both Langmuir and Freundlich isotherms fit the data well, with rate constants of 0.089 and 0.248, respectively. The maximum adsorption capacity (q<sub>max</sub>) from the Langmuir isotherm was 0.9416. The negative free energy change ( $\Delta G^\circ = -3938.21$  kJ/mol at 298 K) indicated high capacity and affinity for Zn (II) removal, confirming the process's feasibility and spontaneity.

**Keywords:** Zn (II), SiO<sub>2</sub>, Batch adsorption, Isotherms, Adsorption capacity

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**Introduction**

The degradation of water quality due to heavy metal contamination has rapidly increased, driven by unethical practices such as the discharge of municipal, urban, and industrial waste, population growth, land conversion, industrial and mining activities, transportation, fertilizer use, construction residues, and combustion processes.<sup>1-4</sup> In addition, heavy metals originating from natural paedogenic processes, such as the weathering of parent rock, forest fires, volcanic eruptions, erosion, and other geological events, further contribute to increasing water pollution. Zinc, an essential micronutrient for human health, plays a role in over 100 immuno-regulatory and antioxidant reactions. However, exceeding the permissible zinc levels in potable water can cause severe health issues,<sup>5-7</sup> such as pancreatic and gastrointestinal problems, sexual dysfunction, appetite loss, immunity disruption,

disruption of brain and central nervous system cells, imbalance of Cu-Fe levels in the body, delayed wound healing, anemia, skin irritation, and protein metabolism disturbances. Zinc's antiviral properties have also led to its use in treating COVID-19. Despite government regulations and environmental acts that set maximum zinc limits, the toxicological profile of this heavy metal poses a significant public health risk. Effective zinc removal from aquatic systems is therefore essential for environmental protection. Numerous techniques for heavy metal removal from aqueous solutions have been reported, including coagulation, chemical oxidation/reduction, ion exchange, membrane filtration, electrodialysis, reverse osmosis, ultrafiltration, electrochemical treatment, and adsorption.<sup>8-10</sup> However, many of these methods are expensive, less efficient, sensitive to specific conditions, and involve complex disposal processes. In contrast,



adsorption technology is the simplest, safest, most eco-friendly, and cost-effective method, offering high removal efficiency and bulk availability of adsorbents. This technique has been widely used in effluent treatment plants and wastewater treatment. Traditional adsorbents such as zeolite, clay, red mud, fly ash, coal, metal oxides, activated carbon, agricultural waste, and biosorbents have been commonly used for heavy metal removal.<sup>11-14</sup> In recent years, advanced materials, including modified layered structures and nano-structured adsorbents like graphene, chitosan, and nitrilotriacetic acid (NTA)-based oxides, have shown superior efficiency and faster rates of heavy metal removal. Adsorption studies on zinc removal from contaminated water have extensively reported the use of materials with high surface area, swelling capacity, and mechanical strength.<sup>15-17</sup> The presence of high levels of heavy metals in water poses a significant threat to aquatic ecosystems and human health, as these metals are biologically toxic, non-degradable, bioaccumulative, and can enter the food chain, leading to long-lasting negative impacts. In this study, a batch adsorption technique was employed, known for its simplicity, effectiveness, experimental accuracy, and the potential for reusing the adsorbent. The objective was to remove Zn (II) from wastewater using a high-surface-area adsorbent (SiO<sub>2</sub>). The isotherms, kinetics, and thermodynamics of Zn (II) adsorption were thoroughly investigated in this work.

## Materials and Methods

ZnSO<sub>4</sub>·7H<sub>2</sub>O (Merck, India) was used to prepare a stock solution with double-distilled water. Working solutions with concentrations of 5, 10, 15, and 20 mg/L were then prepared from the stock solution using double-distilled water throughout the study. The adsorbent, SiO<sub>2</sub>, was purchased from Crysti Chemical, Gandhinagar, Gujarat, India. HCl and NaOH were obtained from CDH, India.

The pH of the solutions was adjusted using 0.01N NaOH and HCl with a Systronic Digital pH meter. All apparatus, including glass bottles and Erlenmeyer flasks, were cleaned and dried in an oven prior to use. Solutions of known Zn (II) concentrations were prepared, and experiments were conducted using an ELICO SL194 double beam atomic absorption spectrophotometer (AAS)<sup>18,19</sup> to obtain a calibration curve for atomic absorption. Each experiment was performed in triplicate to ensure accuracy.

### Batch adsorption experiment

First, 50 ml of the working solutions of Zn (II) with concentrations of 5, 10, 15, and 20 mg/L were taken in 250 ml Erlenmeyer flasks. To each flask, 1 g of SiO<sub>2</sub> was added. The flasks were sealed and shaken using a Shinetec shaker with digital temperature control for a duration ranging from 10 to 120 min. At specific intervals, the supernatant solution was examined using an AAS<sup>20-22</sup> after separating the adsorbent through filtration or decantation. After reaching equilibrium, experiments were conducted under varying conditions: pH 5-8 (measured with a Systronic

pH meter-335), temperature 30 to 40 °C, agitation speed 120-220 rpm, and adsorbent dose of 1-4 g/L. The kinetics and isotherms of Zn (II) adsorption were measured.

## Results and Discussion

### Contact time and effect of concentrations

Different concentrations of Zn (II) ion solutions were prepared at 5, 10, 15, and 20 mg/L at 30 °C, and 1 g of SiO<sub>2</sub> was added to each adsorbate solution. The initial pH was adjusted to 6, and the mixtures were shaken at a speed of 180 rpm at different time intervals of 10, 20, 30, 40, 60, 90, and 120 minutes to determine the effect of contact time. Rapid adsorption of Zn (II) ions occurred within 10-60 minutes, reaching equilibrium at 60 minutes, as shown in Figure 1. It was observed that the maximum removal of Zn (II) was 80, 85, 78.6, and 72.5% for the 5, 10, 15, and 20 mg/L adsorbate solutions, respectively. The adsorbate concentration of 10 mg/L at 60 minutes of equilibrium time showed the maximum exposure of adsorption sites. The removal percentage of Zn (II) can be calculated using equation 1. The quality of the adsorbent and the availability of its sorption sites affect the time needed to reach equilibrium.

$$\text{Adsorption/Removal efficiency (\%)} = \frac{C_o - C_e}{C_e} \times 100 \quad (1)$$

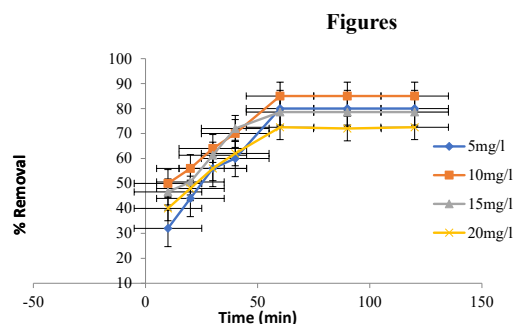
where, C<sub>o</sub> is initial concentration and C<sub>e</sub> is the equilibrium concentration of the solution.

### Effect of Adsorbent Dose

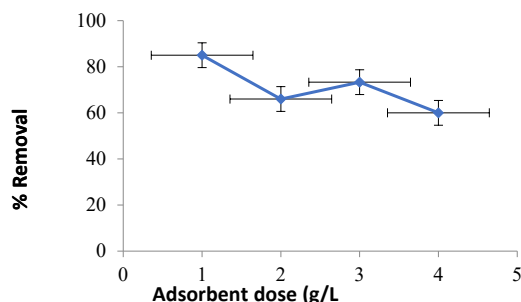
Figure 2 depicts the effect of adsorbent concentration. The concentration of silicon dioxide used as an adsorbent in the batch experiment ranged from 1 to 4 g/L. At a concentration of 1 g/L, 85% of Zn (II) ions were removed. However, at an adsorbent dose of 2 g/L, adsorption decreased to 66%. Increasing the adsorbent dose to 3 g/L slightly increased adsorption to 73.3%, but it decreased again at 4 g/L. This zigzag pattern indicates maximum adsorption at 1 g/L of silicon dioxide. Therefore, subsequent experiments were conducted at an adsorbent concentration of 1 g/L.

### Effect of pH

Figure 3 shows the optimization of pH within the range of 5 to 8. The maximum removal efficiency of 84% was observed at pH 6. As illustrated in Figure 3, there is a gradual increase in Zn (II) ion adsorption with increasing pH. However, beyond pH 6, further increases to pH 7 and 8 did not significantly affect the adsorption rate, which remained nearly constant. Changes in the pH of the solution chemically alter the binding sites of the adsorbent, affecting their affinity for specific metal ions and, in turn, impacting metal ion removal. At lower pH levels, protonation of hydroxyl groups leads to a positive surface charge on the adsorbent's active sites, causing electrostatic repulsion of Zn (II) ions and resulting in lower adsorption efficiency. The concept of zero-point



**Figure 1.** Removal Efficiency of Zn<sup>2+</sup> on Silicon Dioxide Powder as a Function of Different Adsorbent Concentrations, (pH=6.0, silica dose=1 gm/L, agitation speed=180 rpm and temp=303 K)



**Figure 2.** Effect of Silica Dosage on Removal Efficiency of Zn<sup>2+</sup> (C<sub>0</sub>=10 mg/L, pH=6, agitation speed=180 rpm and temp=303 K)

charge (pH<sub>zpc</sub>) was applied to determine the charge state of the adsorbent as a function of pH. At higher pH values, above the pH<sub>zpc</sub>, the adsorbent surface becomes negatively charged, making it more favorable for binding with metal cations.

**Effect of Agitation Speed**

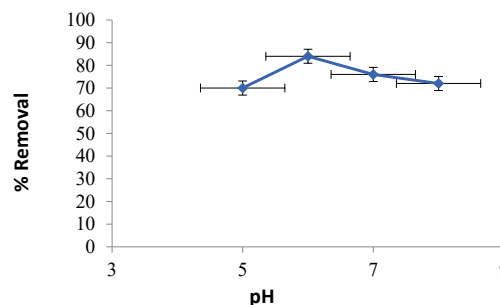
Agitation speed plays a crucial role in metal adsorption. Higher shaking rates increase the likelihood of interactions between aqueous Zn (II) ions and the binding sites of the adsorbent. In this study, the adsorption of Zn (II) ions was evaluated by varying the mixing rate of the reaction mixture from 120 rpm to 220 rpm, while keeping other variables constant. As shown in Figure 4, removal efficiency improved with increasing mixing rates, reaching a maximum of 86% at 180 rpm. This enhanced efficiency is likely due to increased interactions between Zn (II) ions and the adsorbent’s binding sites.

**Effect of Temperature**

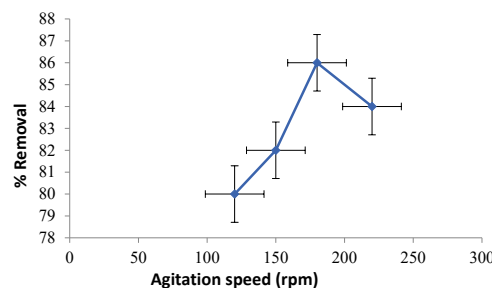
Figure 5 illustrates the influence of temperature on the removal efficiency of Zn (II) ions. The experiment was conducted at temperatures ranging from 298 to 313 K, while keeping other variables constant at their optimal values. The maximum removal efficiency of 88% was observed at 303 K, after which adsorption decreased. The rise in temperature may disrupt the surface of the adsorbent, adversely affecting the adsorption process.

**Adsorption Isotherm**

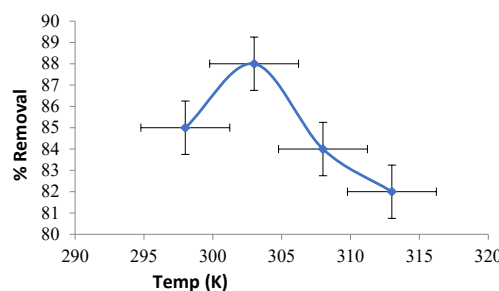
The removal of Zn (II) ions can be mathematically



**Figure 3.** Removal Efficiency of Zn<sup>2+</sup> With pH (C<sub>0</sub>=10 mg/L, agitation speed=180 rpm, temp=303 K, silica dose=1 g/L and time 60 min)



**Figure 4.** Effect of Agitation Speed on Percentage Removal of Zn<sup>2+</sup> (C<sub>0</sub>=10 mg/L, pH=6, silica dose=1 g/L, time=60 min and temp=303 K)

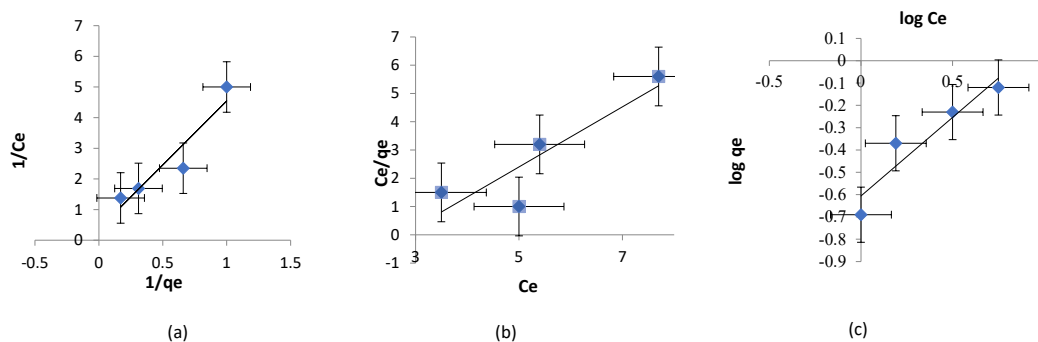


**Figure 5.** Effect of Temperature on Percentage Removal of Zn<sup>2+</sup> (C<sub>0</sub>=10 mg/L, pH=6, silica dose=1 g/L, time=60 min and agitation speed=180 rpm)

expressed using the classical Langmuir and Freundlich adsorption isotherm models, with the adsorption data typically fitting one of these models. The Langmuir adsorption equation is expressed as equation 2:

$$\frac{1}{q_e} = \frac{1}{Q_0} + \frac{1}{bQ_0C_e} \tag{2}$$

where, C<sub>e</sub> is the equilibrium concentration of the adsorbate (mg/L) and q<sub>e</sub> is the amount of the adsorbate adsorbed per unit mass of the adsorbate at equilibrium (mg/g), while Q<sub>0</sub> and b are the Langmuir constants related to the adsorption capacity and energy of adsorption, respectively. Figure 6a shows the linear plot of 1/q<sub>e</sub> versus 1/C<sub>e</sub>, illustrating the applicability of monolayer adsorption of Zn (II) ions on the adsorbent, following the Langmuir adsorption model. The Langmuir constants, Q<sub>0</sub> and b, were determined from these plots and are presented in Table 1. An RL value between 0 and 1 indicates favorable adsorption.<sup>23</sup> In this case, the RL value is 0.529, confirming favorable Langmuir adsorption of Zn (II) onto silicon dioxide. Figure 6b displays the plot of C<sub>e</sub>/Q<sub>e</sub> versus C<sub>e</sub>,



**Figure 6.** (a) Application of the linearized Langmuir adsorption isotherm to the data for the adsorption of Zn<sup>2+</sup> on SiO<sub>2</sub>. (Co=10 mg/L, pH=6, silica dose=1 g/L, time=60 min and agitation speed=180 rpm). (b) Experimental data fitted to the Langmuir model (Co=10 mg/L, pH=6, silica dose=1 g/L; time=60 min, agitation speed=180 rpm, temp=303 K). (c) Freundlich plot for the adsorption of Zn (II) on SiO<sub>2</sub> adsorbents (Co=10 mg/L; pH=6, silica dose=1 g/L; time=60 min, agitation speed=180 rpm and temp=303 K)

**Table 1.** Langmuir and Freundlich Constants for Zn (II) Adsorption on SiO<sub>2</sub> adsorbent

Isotherm	Parameters	Removal % Yield
Langmuir	Q <sub>o</sub> =2.688 b=0.089 R <sub>l</sub> =0.529 q <sub>max</sub> =0.9416 R <sup>2</sup> =0.889	88
Freundlich	n=1.422 1/n=0.703 K <sub>f</sub> =0.248 R <sup>2</sup> =0.891	

used to determine q<sub>max</sub>, the maximum adsorption capacity (mg/g), using equation 3:

$$\frac{C_e}{q_e} = \frac{C_e}{q_{max}} + \frac{1}{b q_{max}} \tag{3}$$

The Freundlich adsorption isotherm was also applied to the equilibrium concentration of Zn (II), and Figure 6c shows the linear plot of log qe versus log Ce, based on equation 4:

$$\text{Log}q_e = \log K_f + \frac{1}{n} \log C_e \tag{4}$$

where, q<sub>e</sub> is the amount of Zn (II) adsorbed at equilibrium (mg/g), C<sub>e</sub> is the equilibrium adsorbate concentration, and K<sub>f</sub> and n are the Freundlich constants. The K<sub>f</sub> value represents the overall adsorption capacity of the adsorbent (mg/g), while n indicates the adsorption intensity. Table 1 presents the values of K<sub>f</sub> and n. According to Abdus-Salam and Adekola, n values predict whether the adsorption of a given ion is favorable. In this study, since n > 1 for the adsorption of Zn (II), the adsorption intensity is considered favorable at higher concentrations.<sup>24</sup>

**Kinetics of Adsorption**

The pseudo first-order and pseudo second-order kinetics model was utilized to determine the kinetics of adsorption of Zn (II) ions on SiO<sub>2</sub> adsorbent. The rate constant of the reaction was calculated by using the first-order and second-order kinetics equation.

**Pseudo-first Order Kinetics Model**

It can be represented by equation 5:

$$\log(q_e - q_t) = \log q_e - \left( \frac{k_1}{2.303} \right) t \tag{5}$$

where, q<sub>e</sub> is the amount of metal ions adsorbed per unit weight of adsorbent at equilibrium (i.e., adsorption capacity, mg/g), q<sub>t</sub> is the amount of adsorbate adsorbed (mg/g) at any time t, and k<sub>1</sub> is the rate constant. The k<sub>1</sub> values were calculated from the slope of the linear plot of log (q<sub>e</sub> - q<sub>t</sub>) versus t, as shown in Figure S1, and found to be 0.0391. The R<sup>2</sup> value was 0.984, which is closer to 1 than the value obtained for the second-order model. Therefore, Zn (II) adsorption followed pseudo-first-order kinetics more closely than pseudo-second-order kinetics.

**Pseudo-second Order Model**

The Pseudo-second order model can be given by means of equation 6:

$$\frac{t}{q_t} = \left( \frac{1}{k_2 q_e^2} \right) + \left( \frac{1}{q_e} \right) t \tag{6}$$

where, k<sub>2</sub> is the rate constant, which can be calculated by plotting t/q<sub>t</sub> versus time as shown in Figure S2. The plot of t/q<sub>t</sub> versus t gives a linear curve with 1/q<sub>e</sub> and 1/k<sub>2</sub>q<sub>e</sub><sup>2</sup> as slope and intercept, respectively. As depicted in Table 2, the rate constant k<sub>2</sub> was found to be 0.1377 and q<sub>e</sub> was 0.505mg/g. R<sup>2</sup> was 0.968.

**Intra-particle Diffusion Model**

The overall kinetics of Zn (II) adsorption was close to the pseudo-first-order kinetics model. However, the rate-limiting step (the slowest step of the reaction) could not be determined by these models. The intra-particle diffusion suggests that the adsorbate uptake by the adsorbent is in proportion to the square root of the time. Weber and Morris proposed equation 7:

$$q_t = K_d t^{0.5} + C \tag{7}$$

where, K<sub>d</sub> is the intra-particle diffusion constant (mg /g min<sup>1/2</sup>) and qt is the adsorption capacity at any time t,

**Table 2.** The Kinetic Parameters for Adsorption of Zn (II) on SiO<sub>2</sub> Adsorbent at 303 K and pH 6

Experimental	Pseudo-first Order Constants			Pseudo-second Order Constants		
	q <sub>e</sub> (mg/g)	k <sub>1</sub> (min)	R <sup>2</sup>	q <sub>e</sub> (mg/g)	k <sub>2</sub> (g/mg/min)	R <sup>2</sup>
0.425	0.1	0.0391	0.984	0.505	0.1377	0.968

and C is the thickness of the boundary layer. The plot of q<sub>t</sub> versus t<sup>0.5</sup> is linear then the mechanism follows intra-particle diffusion. The rate-limiting factor is due to intra-particle diffusion if the plot passes through the origin. In this case, the plot is not linear and does not pass through the origin, indicating that the mechanism involved differs from simple intra-particle diffusion. Figure S3 shows the plot of q<sub>t</sub> versus t<sup>0.5</sup>, which is nonlinear over the entire time range. The adsorption process appears to occur in two stages: the first linear portion corresponds to macro-pore diffusion, while the second linear portion represents micro-pore diffusion. Therefore, it can be concluded that the adsorption of Zn (II) ions onto the silica adsorbent involves more than one process, with intra-particle diffusion not being the rate-limiting factor.

### Thermodynamics of Adsorption

Thermodynamic analysis of adsorption was applied at four different temperatures i.e., 298 K, 303 K, 308 K, and 313 K. The thermodynamics parameter can be obtained by using the equations: 8-10:

$$K_d = C_e / q_e \quad (8)$$

$$\Delta G^0 = -Rt \ln K_{ad} \quad (9)$$

$$\log K_d = \frac{\Delta S^0}{2.303R} - \frac{\Delta H^0}{2.303RT} \quad (10)$$

where, K<sub>d</sub> is the equilibrium constant, q<sub>e</sub> is the amount of metal ions adsorbed per unit weight of adsorbent at equilibrium (i.e., adsorption capacity, mg/g), and C<sub>e</sub> is the equilibrium adsorbate concentration in solution (mg/g). R is the gas constant (8.314 J/mol/K), ΔG<sup>0</sup> is the change in Gibbs free energy (kJ/mol), ΔH<sup>0</sup> is the change in enthalpy (kJ/mol), ΔS<sup>0</sup> is the change in entropy (J/K/mol), and T is the temperature (K). ΔG<sup>0</sup> at different temperatures can be determined using equation 9. The changes in entropy and enthalpy was then calculated from the intercept and slope of the plot of log K<sub>d</sub> versus 1/T as shown in Figure S4. The ΔG<sup>0</sup> values for the adsorption of Zn (II) ions at all temperatures were found to be negative, indicating that the adsorption process is feasible and spontaneous. The negative value of ΔH<sup>0</sup> confirms the exothermic nature of the adsorption. When ΔH<sup>0</sup> is less than 20 kJ/mol, the process involves physisorption, while values greater than 20 kJ/mol suggest chemisorption, indicating ion exchange interactions.<sup>25</sup> The negative entropy change reveals a decrease in randomness during the adsorption of Zn (II) on SiO<sub>2</sub>. All thermodynamic parameters are summarized in Table 3.

**Table 3.** Thermodynamic Parameters for Adsorption of Zn (II) on SiO<sub>2</sub>

Temperature (K)	ΔG <sup>0</sup> (kJ/mol)	ΔH <sup>0</sup> (kJ/mol)	ΔS <sup>0</sup> (J/mol K)	K <sub>d</sub> (C <sub>e</sub> /q <sub>e</sub> )
298	-3988.21			5
303	-2521.18			2.72
308	-3419.16	-30,307	-758.418	3.80
313	-2310.95			2.43

### Conclusion

This study presents the versatility of silica as an adsorbent and its potential application for metal adsorption in wastewater treatment. In this paper, silica proved to be an excellent adsorbent for the removal of Zn (II) ions from aqueous solutions. The adsorption and removal efficiency of the adsorbent were found to depend on several factors, including temperature, contact time, pH, silica dose, Zn (II) concentration, and the agitation speed of the aqueous solution. To examine the effect of these factors, a series of batch experiments was conducted at different concentrations (5–20 mg/L). The maximum removal efficiency of Zn (II) ions, 88%, was achieved at pH 6, with a contact time of 60 minutes, at a temperature of 30 °C, using an adsorbent dose of 1 g/L, and agitated at 180 rpm with a Zn (II) concentration of 10 mg/L. The isotherms, kinetics, and thermodynamics of Zn (II) adsorption were also analyzed. The results indicated that the adsorption followed both the Langmuir and Freundlich models and obeyed pseudo-first-order kinetics. Thermodynamic studies revealed that the adsorption process was exothermic and spontaneous. The negative entropy suggests a reduction in the degree of freedom, indicating a decrease in adsorbate concentration at the solid-solution interface.

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### Authors' Contribution

**Conceptualization:** Himanshu Shekhar.

**Data curation:** Vivekanand Choudhary.

**Formal analysis:** Twinkle Keshari.

**Funding acquisition:** Vivekanand Choudhary.

**Investigation:** Vivekanand Choudhary.

**Methodology:** Twinkle Keshari.

**Project administration:** Himanshu Shekhar.

**Resources:** Himanshu Shekhar.

**Software:** Twinkle Keshari.

**Supervision:** Himanshu Shekhar.

**Validation:** Twinkle Keshari.

**Visualization:** Himanshu Shekhar.

**Writing—original draft:** Twinkle Keshari.

**Writing—review editing:** Twinkle Keshari.

### Competing Interests

None.

### Ethical Approval

Departmental Research Committee

**Funding**

Veer Kunwar Singh University, Ara, Bihar

**Supplementary files**

Supplementary file 1 contains Figures S1-S4.

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