

Interpretation of the Tungstate Anion Adsorption Behavior on Bio-Rex 5 resin in $^{188}\text{W}/^{188}\text{Re}$ Generator System

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Abstract: Rhenium-188 (^{188}Re), a β^- -emitting radionuclide, holds considerable promise for targeted radionuclide therapy due to its favorable physical and chemical properties. This study explores the adsorption behavior of tungstate ions onto Bio-Rex 5 anionic resin under various conditions to enhance the efficiency of $^{188}\text{W}/^{188}\text{Re}$ generator. Batch adsorption experiments revealed that the maximum adsorption capacity was achieved within 15 min. of contact time and at an initial tungstate concentration of 250 ppm. Kinetic modeling indicated that the adsorption process aligns with a pseudo-second-order model, suggesting chemisorption as the dominant mechanism. The Langmuir model provided the best description of adsorption isotherms, suggesting monolayer adsorption on a homogeneous surface. Thermodynamic analysis demonstrated an endothermic process with a positive ΔH and ΔS along with a negative ΔG , confirming spontaneity. These findings provide valuable insights into the optimization of tungstate ion adsorption, paving the way for the development of an efficient and reliable $^{188}\text{W}/^{188}\text{Re}$ generator system in theranostic applications.

Keywords: Rhenium-188, $^{188}\text{W}/^{188}\text{Re}$ generator, Kinetics, Adsorption Isotherm, Thermodynamics.

1 Introduction

The $^{188}\text{W}/^{188}\text{Re}$ generator system plays a crucial role in the field of nuclear medicine, providing a reliable and continuous supply of rhenium-188 (^{188}Re) for various therapeutic and diagnostic applications. ^{188}Re is a high-energy beta-emitting radioisotope with a maximum energy of 2.12 MeV, allowing for effective penetration in solid tumors. Its decay also produces a 155 keV gamma emission, which is used for imaging and dosimetry studies, allowing for real-time tracking of the administered therapeutic agents, making ^{188}Re a dual-purpose theranostic agent [1, 2]. Furthermore, ^{188}Re has a half-life of 17.0 h. This relatively short half-life is advantageous as it allows

for timely use in medical procedures without prolonged radiation exposure to patients. The ability to generate ^{188}Re on-site through the $^{188}\text{W}/^{188}\text{Re}$ generator system significantly reduces logistical challenges and the dependency on centralized production facilities, making this technology vital for clinical settings that require rapid access to ^{188}Re for patient treatment [3, 4]. ^{188}Re is chemically similar to technetium, a widely used radionuclide in nuclear medicine, enabling the development of numerous radiopharmaceuticals for targeted therapy. Its chemical properties allow for the easy labeling of biomolecules, such as antibodies or peptides, enhancing the specificity of ^{188}Re -based radiopharmaceuticals for targeting cancer cells or other pathological tissues [5].

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The production of ^{188}Re is typically achieved through the irradiation of ^{186}W in a research reactor. Upon irradiation, ^{186}W undergoes double neutron capture, transforming into ^{188}W , which then decays into ^{188}Re through β^- decay [6, 7].

The preparation of efficient $^{188}\text{W}/^{188}\text{Re}$ generators requires the use of ion-exchange resins to separate and purify ^{188}Re from ^{188}W . Ion exchange resins, such as Bio-Rex 5, are essential for this process as they offer excellent selectivity for tungstate ions (WO_4^{2-}). Bio-Rex 5 is a weak anion-exchange resin, known for its high efficiency and capacity to selectively bind tungstate ions while allowing the passage of other ions. This characteristic makes it an ideal candidate for use in the development of $^{188}\text{W}/^{188}\text{Re}$ generators. The advantages of using Bio-Rex 5 include its cost-effectiveness, ease of regeneration, and efficient separation of ^{188}Re from tungsten-188, thereby optimizing the yield of ^{188}Re for theranostic applications [8].

The adsorption mechanism of tungstate ions onto Bio-Rex 5 is a complex process that can be understood through kinetic modeling, isotherms, and thermodynamic studies. Kinetic models describe the rate at which tungstate ions are adsorbed onto the resin, providing insights into the nature of the adsorption process. Isotherm studies are used to describe the equilibrium distribution of tungstate ions between the Bio-Rex 5 and solution, allowing for the determination of adsorption capacities and affinities. Additionally, thermodynamic parameters, including enthalpy (ΔH), entropy (ΔS), and Gibbs free energy (ΔG), are crucial for understanding the spontaneity and temperature dependence of the adsorption process [09-13].

In this study, we aim to investigate the factors influencing the adsorption of tungstate ions onto Bio-Rex 5. Key parameters such as contact time, initial ion concentration, adsorbent dosage, and temperature will be systematically examined to understand their impact on the adsorption efficiency. Furthermore, the kinetics of adsorption, isotherm models, and thermodynamic analyses will be studied to enhance our understanding of the adsorption mechanism, offering valuable insights for the development and optimization of efficient $^{188}\text{W}/^{188}\text{Re}$ generator systems for therapeutic and diagnostic applications.

2 Experimental Section

2.1 Chemicals

Bio-Rex 5 anionic resin was purchased from Bio-Rad Laboratories, USA. Tungsten trioxide, WO_3 , and sodium tungstate dihydrate ($\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$) were obtained from Sigma-Aldrich, Germany. All other chemicals were of analytical grade and used without further purification.

2.2 Preparation of ^{187}W radiotracer

A 1 g sample of natural tungsten oxide (WO_3) was

irradiated for 48 hours in the Egyptian Second Research Reactor (ETRR-2) with a thermal neutron flux of $1.8 \times 10^{14} \text{ n/cm}^2/\text{s}$. Following irradiation, the sample was left to cool for 5 days and then dissolved in 10 mL of 5.0 M NaOH solution with the addition of a few drops of H_2O_2 , and the solution was gently heated until it was completely dry. The residue was subsequently dissolved in 10 mL of deionized water to obtain a solution containing ^{187}W , which was used as a radiotracer for adsorption experiments. The ^{187}W radioactivity was measured at 685.8 keV by a calibrated high-purity germanium coaxial detector (HPGe) with a multichannel analyzer (MCA) (GX2518 model, Canberra, USA) [8, 14].

2.3 Effect of contact time on tungstate adsorption

The effect of contact time on tungstate adsorption onto Bio-Rex 5 resin was investigated using a batch adsorption experiment. A fixed amount of resin (0.01 g) was added to a series of 10 mL vials containing 10 mL of a solution of pH 4 spiked with 100 μL of the irradiated tungstate target solution (initial concentration of 250 mg/L). The vials were then agitated at a constant temperature of 25 $^\circ\text{C}$ using a thermostated shaker (140 rpm). At predetermined time intervals (5, 10, 20, 30, 60, and 120 minutes), samples were withdrawn from the vials and analyzed for residual tungstate radioactivity. To guarantee reproducibility, the experiments were carried out three times.

The amount of tungstate adsorbed onto the resin (q) at time t was calculated using the following equation:

$$q = (C_0 - C_t) V/m \quad (1)$$

Where q (mg/g) is the amount of tungstate adsorbed per unit mass of adsorbent, C_0 (mg/L) is the initial concentration of tungstate solution, V (L) is the volume of solution, m (g) is the mass of adsorbent, and C_t (mg/L) is the concentration of tungstate solution at time t and is obtained from the following formula:

$$C_t = C_0 (A_f / A_0) \quad (2)$$

Where, A_0 (counts/min) is the initial radioactivity of ^{187}W in the solution, and A_f (counts/min) is the radioactivity of ^{187}W in the solution after adsorption

2.4. Kinetic modeling

The adsorption kinetics of tungstate anions onto Bio-Rex 5 anionic resin were analyzed using three kinetic models: pseudo-first-order, pseudo-second-order, and intraparticle diffusion. The experimental data were fitted to these models to determine the rate-controlling steps and the nature of the adsorption mechanism.

2.4.1. Pseudo-First-Order Model

This model assumes that the rate of adsorption is directly proportional to the number of available adsorption sites. It is expressed as:

$$\ln (q_e - q_t) = \ln q_e - K_1 t \quad (3)$$

Where q_t (mg/g) is the adsorption capacity at time t (min), q_e (mg/g) is the equilibrium adsorption capacity, and k_1 (min^{-1}) is the rate constant. A plot of $\ln(q_e - q_t)$ versus t was used to determine k_1 and q_e .

2.4.2. Pseudo-Second-Order Model

This model assumes that the rate-limiting step is chemisorption involving the sharing or exchange of electrons between the adsorbent and the adsorbate. It is given by this equation:

$$t / q_t = 1 / k_2 q_e^2 + t / q_e \quad (4)$$

Where k_2 (g/mg.min) is the rate constant. The slope and intercept of the t/q_t vs. t plot were used to get the values of q_e and k_2 .

2.4.3. Intraparticle Diffusion Model

This model describes the diffusion of adsorbate molecules within the pores of the adsorbent. To evaluate the contribution of diffusion within the resin pores, the intraparticle diffusion model was applied:

$$q_t = k_{id} t^{1/2} + C \quad (5)$$

Where K_{id} ($\text{mg/g.min}^{0.5}$) is the intraparticle diffusion rate constant, and C (mg/g) represents the thickness of the boundary layer. A plot of q_t versus $t^{0.5}$ was used to evaluate k_{id} and C .

All the kinetic parameters were determined using non-linear or linear regression analysis, and the model with the highest correlation coefficient (R^2) and best agreement between experimental and predicted adsorption capacities was considered the most suitable for describing the adsorption process.

2.5. Effect of initial tungstate concentration on ^{187}W adsorption

The effect of initial tungstate ion concentration on the adsorption of ^{187}W onto Bio-Rex 5 anionic resin was

studied under controlled experimental conditions. Solutions of tungstate ions with initial concentrations ranging from 25 ppm to 500 ppm were prepared by diluting a stock solution. The pH of each solution was adjusted to 4 using dilute HCl or NaOH. ^{187}W was added to each solution to achieve a constant initial radioactivity. Adsorption experiments were conducted at room temperature ($25 \pm 2^\circ\text{C}$) with a contact time of 15 min. to ensure equilibrium. 0.01 g of Bio-Rex resin was added for each batch experiment, with constant agitation to promote uniform interaction between the tungstate ions and the resin. After the adsorption process, the solution was separated from the resin, and the remaining radioactivity of ^{187}W in the supernatant was measured. The adsorption capacity (q_e) of the resin at each initial tungstate concentration was calculated using equation (1).

2.6. Adsorption Isotherm Models

To evaluate the adsorption behavior of tungstate anions onto Bio-Rex 5 anionic resin, four isotherm models - Langmuir, Freundlich, Dubinin-Radushkevich (D-R), and Temkin - were applied to analyze the equilibrium data [15-19]. The linearized forms of the isotherm equations were used to fit the experimental data.

2.6.1. Langmuir Isotherm

This model assumes monolayer adsorption onto a homogeneous surface with a finite number of identical adsorption sites. The linearized equation is:

$$1/q_e = 1/q_{\max} + 1/q_{\max} K_L C_e \quad (6)$$

Where C_e (mg/L) is the equilibrium concentration of ^{187}W , q_e (mg/g) is the equilibrium adsorption capacity, q_{\max} (mg/g) is the maximum adsorption capacity, and K_L (L/mg) is the Langmuir constant.

2.6.2. Freundlich Isotherm

This model describes heterogeneous adsorption with multilayer formation. The linearized form is:

$$\ln q_e = \ln K_F + 1/n \ln C_e \quad (7)$$

Where, K_F (L/mg) is the Freundlich constant, and n is a measure of adsorption intensity.

2.6.3. Dubinin-Radushkevich (D-R) Isotherm

This model explains adsorption with a Gaussian energy distribution and is expressed as:

$$\ln q_e = \ln q_s - (K_{ad} \varepsilon^2) \quad (8)$$

Where q_s (mgg⁻¹) is the theoretical maximum capacity, K_{ad} (mol²/KJ²) is the D-R constant related to the mean free energy of adsorption, and ε is the Polanyi potential, calculated as:

$$\varepsilon = RT \ln (1 + 1/C_e) \quad (9)$$

Where R (KJ/molK) is the ideal gas constant, T (K) is the absolute temperature.

2.6.4. Temkin Isotherm

According to this model, the heat of adsorption decreases linearly with surface coverage. The linearized equation is:

$$q_e = (R_T/b_t) \ln a_t + (R_T/b_t) \ln C_e \quad (10)$$

Where R_T/b_t (B_T) (KJ/mol) is the Temkin constant related to the heat of adsorption, a_t (L/mg) is the Temkin equilibrium binding constant.

The parameters of each isotherm model were determined by linear regression analysis of the experimental data.

2.7. Effect of adsorbent dosage on tungstate adsorption

The influence of the Bio-Rex 5 adsorbent dosage on the adsorption of tungstate ions was evaluated by varying the volume-to-mass (V/m) ratio in the range of 0.066–2.0 Lg⁻¹. Adsorption experiments were conducted under controlled conditions, ensuring consistent initial tungstate concentrations, temperature, and pH. The adsorption capacity was determined for each V/m ratio.

2.8. Effect of temperature on tungstate adsorption

The effect of temperature on the adsorption of tungstate ions by Bio-Rex 5 was studied at five different temperatures: 30°C, 40°C, 60°C, 80°C, and 100°C. Equilibrium data were used to calculate thermodynamic parameters, including the changes in enthalpy (ΔH), entropy (ΔS), and Gibbs free energy (ΔG) at each temperature, using the Van't Hoff equation and standard thermodynamic relationships.

$$\Delta G = -RT \ln K_c \quad (11)$$

$$\Delta G = \Delta H - T \Delta S \quad (12)$$

$$\ln K_c = \Delta S/R - \Delta H/RT \quad (13)$$

Where, R (KJ/mol) is the universal gas constant, T (Kelvin) is the absolute temperature, and K_c is the equilibrium constant for adsorption.

By plotting $\ln K_c$ against $1/T$, the slope of the line gives $-\Delta H/R$ and the intercept provides $\Delta S/R$.

3 Results and Discussion

3.1. Effect of contact time on tungstate adsorption

The impact of contact time on tungstate anion adsorption onto Bio-Rex 5 anionic resin was investigated, and the results are shown in Figure 1. The figure clearly illustrates a rapid increase in adsorption, reaching 93.32% within just 15 minutes, indicating a strong affinity between tungstate anions and the resin's functional groups. This sharp increase suggests a high number of available active sites at the beginning of the adsorption process. Beyond this point, the adsorption capacity remained relatively stable, fluctuating slightly around 93% up to 180 min. The minor variations observed after equilibrium may be attributed to dynamic surface interactions, such as slight desorption-re-adsorption processes. The negligible difference in adsorption beyond 15 minutes suggests that equilibrium is achieved quickly, making this resin highly efficient for tungstate adsorption. These findings indicate that an optimal contact time of 15 minutes is sufficient to achieve maximum adsorption efficiency, reducing the need for extended processing durations in practical applications.

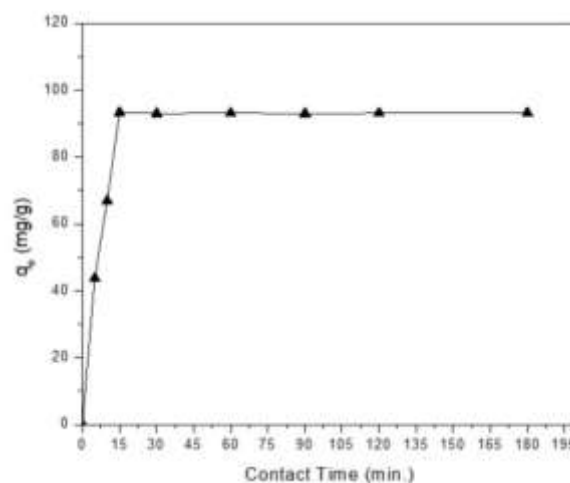


Fig. 1. Effect of contact time on the adsorption of ¹⁸⁷W on Bio-Rex 5 anionic resin.

The kinetics of tungstate anion adsorption onto Bio-Rex 5 resin was evaluated using three models: pseudo-first-order, pseudo-second-order, and the intraparticle diffusion model, and the results are shown in Table 1. The pseudo-first-order model showed a poor fit to the experimental data, with a

low correlation coefficient ($R^2 = 0.475$), and the calculated adsorption capacity ($q_e = 7.75$ mg/g) deviated significantly from the experimental value ($q_e = 93.32$ mg/g). This suggests that the adsorption process is not solely governed by physisorption. In contrast, the pseudo-second-order model provided an excellent fit to the data with a high correlation coefficient ($R^2 = 0.999$). The calculated $q_e = 95.06$ mg/g from this model closely matched the experimental value. This indicates that the adsorption process is likely governed by chemisorption, involving valence forces through the sharing or exchange of electrons between tungstate anions and active sites on the resin. The rate constant ($k_2 = 0.004$ mg/g.min) indicates a rapid adsorption rate. The intraparticle diffusion model revealed that diffusion within the resin pores contributes to the overall adsorption process but is not the sole rate-controlling step, as the plots did not pass through the origin. The k_{id} value was determined to be 2.99 mg/g.min^{0.5}, and the relatively high C value (62.55 mg/g) suggests significant surface adsorption before intraparticle diffusion becomes significant. These findings collectively indicate that the adsorption mechanism involves a combination of surface reactions and diffusion, with chemisorption playing a dominant role [20].

Table 1. Kinetic parameters for the adsorption of ^{187}W onto Bio-Rex 5 resin.

Kinetic Model	Kinetic parameters	
Pseudo-First Order	$q_e(\text{mg/g})$	7.75
	$K_1 (\text{min}^{-1})$	0.029
	R^2	0.475
Pseudo-Second Order	$q_e(\text{mg/g})$	95.06
	$K_2 (\text{g/mg.min})$	0.004
	R^2	0.999
Intraparticle Diffusion	$K_{id} (\text{mg/g.min}^{0.5})$	2.99
	$C (\text{mg/g})$	62.55
	R^2	0.325

3.2. Adsorption capacity as a function of initial tungstate concentration

The effect of initial tungstate concentration on the adsorption of ^{187}W onto Bio-Rex 5 anionic resin was investigated within the range of 25 ppm to 500 ppm, and the results are presented in Figure 2. The figure indicates that the adsorption capacity increased gradually with increasing initial tungstate concentration up to 250 ppm, reaching a maximum value of 93.32 mg/g. At lower concentrations, such as 25 mg/L, the resin exhibited minimal adsorption (12.25 mg/g), likely due to the limited availability of tungstate anions for interaction with active sites. A significant enhancement in adsorption was observed with increasing tungstate concentration, reaching

84.88 mg/g at 200 mg/L. Beyond 250 mg/L, the adsorption efficiency stabilized around 93.33%, with negligible fluctuations up to 500 mg/L, suggesting equilibrium was reached. This trend suggests that at higher concentrations, the resin's active sites become saturated, leading to an equilibrium where additional tungstate ions in solution cannot be effectively adsorbed. These results indicate that Bio-Rex 5 resin exhibits optimal adsorption performance at approximately 250 mg/L, beyond which additional increases in concentration do not significantly enhance adsorption capacity.

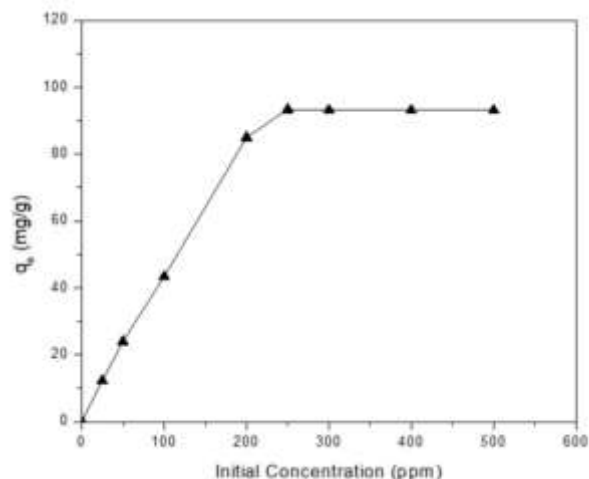


Fig.2. Effect of initial tungstate concentration on ^{187}W adsorption onto Bio-Rex 5 anionic resin.

To understand the adsorption mechanism of tungstate anion on Bio-Rex 5 anionic resin, the equilibrium data were analyzed using Langmuir, Freundlich, Dubinin-Radushkevich (D-R), and Temkin isotherm models. The results of all these models are summarized in Table 2. The Langmuir isotherm exhibited the best fit among the models, with a high correlation coefficient ($R^2=0.998$). The maximum adsorption capacity (q_{max}) was determined to be 95.42 mg/g, indicating the strong affinity of Bio-Rex 5 resin for tungsten species and the likelihood of monolayer adsorption on a homogeneous surface. The Langmuir constant ($K_L=0.169$ L/mg) further supports favorable adsorption interactions. The Freundlich model exhibited a moderate correlation ($R^2=0.903$), with a Freundlich exponent ($n=3.03$) greater than 1, signifying favorable multilayer adsorption on heterogeneous surfaces. However, the Dubinin-Radushkevich (D-R) model showed a poor correlation ($R^2=0.642$), suggesting that the adsorption process is not primarily governed by pore-filling mechanisms. The high mean free energy ($E=1.43 \times 10^3$ kJ/mol) suggests that the process is strongly driven by chemisorption rather than physical adsorption. The Temkin model showed a moderate fit ($R^2=0.887$), with an adsorption energy parameter ($B_T=0.170$ kJ/mol) indicating weak interactions between tungsten ions and the resin surface. Overall, the adsorption data suggest that the Langmuir model provides the most appropriate description

of tungstate adsorption on Bio-Rex 5 anionic resin, implying that monolayer adsorption dominates the process [21-22].

Table 2. Adsorption Isotherm parameters for the adsorption of ^{187}W onto Bio-Rex 5 resin.

Adsorption model	Adsorption parameters	
Langmuir	q_{\max} (mg/g)	95.42
	K_L (L/mg)	0.169
	R^2	0.998
Freundlich	n	3.03
	K_F (L/mg)	18.72
	R^2	0.903
Dubinin–Radushkevich (D–R)	E (KJ/mol)	1.43E+03
	K_{ad} (mol ² /KJ ²)	2.43E-07
	R^2	0.642
Temkin	a_t (L/mg)	1.367
	B_T (KJ/mol)	0.170
	R^2	0.887

3.3. Adsorption capacity of Bio-Rex 5 as a function of volume-to-mass ratio

The adsorption capacity of Bio-Rex 5 for tungstate ions showed a significant dependence on the V/m ratio. Figure 3 indicates that as the V/m ratio increased, the adsorption capacity consistently increased. This trend is attributed to the higher availability of tungstate ions in the solution relative to the adsorbent mass at higher V/m ratios, resulting in more efficient utilization of the available adsorption sites. At lower V/m ratios, the excess adsorbent relative to the solution volume led to underutilization of the adsorption capacity, as many sites remained unsaturated. These results demonstrate that optimizing the V/m ratio is critical to maximizing tungstate uptake by Bio-Rex 5.

3.4. Thermodynamic analysis of tungstate adsorption

The effect of temperature on the adsorption of ^{187}W onto Bio-Rex 5 anionic resin demonstrated a significant increase in adsorption capacity with rising temperature, as shown in Figure 4. The adsorption capacity increased from 93.33 mg/g at 30°C to 123.07 mg/g at 100°C, indicating an endothermic process where higher temperatures enhance molecular mobility, diffusion rates, and interaction between tungstate anions and the resin's active sites. The thermodynamic parameters, summarized in Table 3, further support this behavior. The enthalpy change ($\Delta H=59.39$

kJ/mol) confirms the endothermic nature of the adsorption, as positive ΔH values indicate energy absorption. Additionally, the positive entropy change ($\Delta S=0.197$ kJ/mol.K) suggests increased randomness at the solid-liquid interface, likely due to the displacement of water molecules from the resin surface by tungstate ions, favoring adsorption at elevated temperatures. The Gibbs free energy change (ΔG) values, as presented in Table 3, were negative at all studied temperatures, confirming the spontaneous nature of the adsorption process. Moreover, ΔG became more negative with increasing temperature, from -0.29 kJ/mol at 303 K to -8.17 kJ/mol at 343 K, indicating enhanced spontaneity at higher temperatures. This trend supports the hypothesis that higher temperatures strengthen the adsorbate-adsorbent interactions, likely through chemisorption mechanisms. These findings confirm that the adsorption of ^{187}W onto Bio-Rex 5 is a favorable, entropy-driven, and endothermic process [23-25].

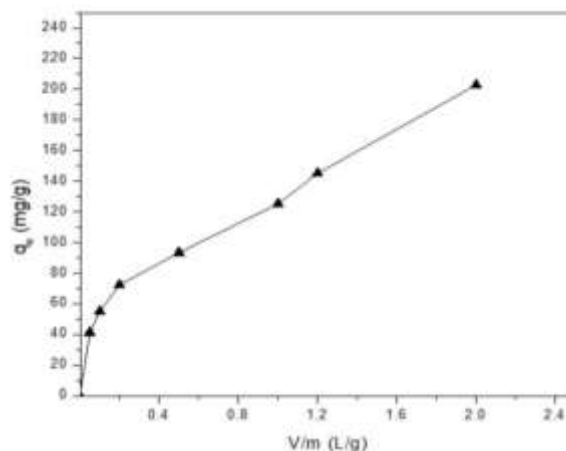


Fig. 3. Effect of V/m ratio on the adsorption of ^{187}W onto Bio-Rex 5 resin.

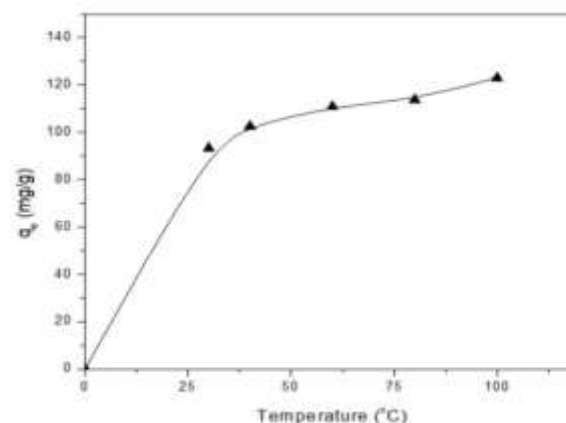


Fig. 4. Effect of temperature on the adsorption of ^{187}W onto Bio-Rex 5 resin.

Table 3. Thermodynamic parameters for the adsorption of ^{187}W onto Bio-Rex 5 resin.

ΔH (kJ/mol)	ΔS (kJ/mol·K)	ΔG (kJ/mol)				
		303	313	323	333	343
59.39	0.197	-	-	-	-	-
		0.29	2.26	4.23	6.20	8.17

4 Conclusions

This study demonstrated the efficacy of Bio-Rex 5 anionic resin in adsorbing tungstate ions under optimized conditions. The adsorption process was rapid, with equilibrium reached within 15 min., and followed a pseudo-second-order kinetic model, indicating chemisorption. The Langmuir isotherm model highlighted a monolayer adsorption mechanism and thermodynamic analysis confirmed the endothermic and spontaneous nature of the process. These findings suggest that Bio-Rex 5 is a promising adsorbent for use in the $^{188}\text{W}/^{188}\text{Re}$ generator system, offering potential improvements in efficiency and reliability for radionuclide therapy applications.

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