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Nitrate Adsorption onto Unmodified Calcium–Alginate Beads: Equilibrium Behaviour and Adsorption Constraints

Mieow Kee Chan, Daria Kudritskaya, Nii Chien Ng, Yan Zu Ng, Tengku Faris Imran bin Tengku Amirruzaini

Centre for Water Research, Faculty of Engineering, Built Environment and Information Technology, SEGi University, Jalan Teknologi, Kota Damansara, 47810 Petaling Jaya, Selangor Darul Ehsan, Malaysia
mkchan@segi.edu.my, 0000-0002-0805-9395

Corresponding email: mkchan@segi.edu.my

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Abstract: Excessive nitrate contamination in water bodies due to agricultural runoff and aquaculture effluents poses a serious threat to aquatic ecosystems and drinking water safety. Thus, a sustainable and cost-effective treatment technology is required. This study investigated the effectiveness and the adsorption mechanism of neat alginate beads for nitrate removal from aqueous solution. Alginate beads were synthesized via crosslinking a 2 wt% alginate solution in a 5 wt% $CaCl_2$ solution and subsequently tested in batch experiments using nitrate solutions ranging from 10 to 100 ppm. The objectives of this study were to identify the most effective adsorbent dosage, ranging from 1 to 20 g, and determine the best-fitting equilibrium isotherm model. The results showed that the nitrate removal efficiency peaked at ~35% at the most effective dosage of 5 g. No significant improvement was observed at higher dosages due to bead aggregation. Isotherm analysis indicated the Langmuir model provided the best fit of R^2 = 0.9701. This confirmed that adsorption was dominated by a monolayer mechanism on uniform sites. The calculated maximum adsorption capacity for the neat alginate was 0.414 mg/g. This was lower than the capacities achieved by modified alginate composites reported in literature. This indicated that the inherent functional groups of pure alginate beads were inadequate for practical, high-efficiency nitrate remediation. This highlighted the critical need for chemical modification or utilization of alginate in immobilized biological systems to enhance affinity and achieve competitive performance for nitrate removal.

Keywords: *adsorption, nitrate removal, Langmuir, dosage, alginate beads*

1. INTRODUCTION

Fish wastewater, generated primarily from aquaculture and fish processing, presents a significant environmental challenge due to its high concentration of nitrogen compounds, particularly nitrates. This discharge is a major contributor to eutrophication, an ecological process where excessive nutrient enrichment leads to harmful algal blooms and severe hypoxic conditions that deplete dissolved oxygen, thereby threatening aquatic ecosystems and biodiversity (Akinnawo, 2023). Effective and sustainable technologies are therefore crucial for treating this high-nitrogen effluent before its release into natural water bodies.

Current technologies for nitrate removal from wastewater face significant practical and economic constraints. Biological denitrification is the mainstream approach due to its simplicity and ability to convert nitrate to nitrogen gas (Zheng et al., 2024; Azis et al., 2025). However, traditional heterotrophic denitrification requires external organic carbon sources to sustain, and the type and dosage of carbon strongly influence denitrification efficiency and by-product formation. Incomplete denitrification often leads to the emission of nitrous oxide (N_2O). Furthermore, excessive carbon addition results in increased sludge production, significantly raising operational and disposal costs and limiting practical application. Moreover, biological systems are sensitive to temperature and pH, making them challenging to manage in fluctuating environmental conditions (Chan et al., 2021a).

Physicochemical methods such as ion exchange (IE) and reverse osmosis (RO) are also widely employed due to its excellent nitrate removal performance (Ahmer & Uddin, 2024; Zirrahi et al., 2024). Zeng et al. (2023) reported that the macroporous IE resin removed 91.8% of nitrate form the secondary effluent of wastewater treatment plants. Meanwhile, Cohen et al. (2025) found that the spiral-wound RO membrane rejected >95% of nitrate and > 96% of salt from the well water. Nevertheless, IE processes often generate a concentrated brine slurry that is difficult and costly to dispose, and the capital and operational cost are high (Li et al., 2025). RO membranes are costly and are characterized by high energy demands and an increased environmental footprint (Kantor et al., 2025).

Adsorption has emerged as a promising and versatile wastewater treatment technology, favoured for its simplicity, cost-effectiveness, and high-removal efficiency across various contaminants. Li et al. (2025) studied the use of amine-functionalized biowaste-derived adsorbent for nitrate removal. A maximum nitrate adsorption capacity of ~66 mg/g was recorded. Rahmati et al. (2026) found that the sugar-based aluminum coordination polymer adsorbent removed 95% of nitrate from a 200 mg/L of nitrate solution in 30 minutes. Among the diverse materials explored, alginate beads, derived from abundant seaweed, stand out as a green adsorbent (Chiew et al., 2022). This is due to their inherent low toxicity, biodegradability, relatively low cost, and a naturally high surface area. They function by capturing pollutants, like nitrate ions, through surface adhesion via physical forces or chemical bonds. Although many studies report modified alginate systems, this work is novel in establishing a quantitative baseline for nitrate adsorption using neat calcium–alginate beads. This enables objective evaluation of adsorption mechanisms and provides a reference framework against which the benefits of chemical modification can be assessed. This investigation first identifies the most effective dosage of alginate beads necessary for achieving efficient nitrate removal, and subsequently, it will calculate the equilibrium adsorption capacity of the beads across varying initial nitrate concentrations. Finally, the researchers seek to identify the most suitable adsorption isotherm model including Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich (D-R) that accurately describes the adsorption equilibrium and mechanism governing this specific nitrate-alginate system.

2. METHODOLOGY

Calcium chloride and sodium nitrate were purchased from Ever Gainful Enterprise, Malaysia, while sodium alginate was obtained from Friedemann Schmidt, Germany. Sodium nitrate was used as the nitrate precursor for the preparation of aqueous nitrate solutions. All chemicals were of analytical grade and were used as received without further purification.

Alginate beads were prepared by dropping a 2 wt% sodium alginate solution into a 5 wt% calcium chloride solution, following the method described by Chan et al. (2021b). The formed beads were subsequently stored at 4°C for 24 hours to ensure complete crosslinking and stabilization. Following the curing process, 1 to 20 g of alginate beads were dispersed in 100 –150 mL of nitrate solutions with concentrations ranging from 10 to 100 ppm, at pH 7 and room temperature. The mixtures were stirred at a constant speed of 250 rpm for an hour. The nitrate concentration of the solution was determined using a Shimadzu UVmini 1240 UV spectrophotometer at a wavelength of 220 nm (Chan et al., 2018). Prior to the experiment, a nitrate calibration graph with R^2 of 0.976 was prepared using standard nitrate solutions to enable accurate determination of nitrate concentrations.

The nitrate removal rate was calculated using Equation (1), where C_i is the initial nitrate concentration and C_f is the final nitration concentration of the solution.

$$\text{Nitrate removal} = \frac{(C_i - C_f)}{C_i} x\ 100\% \quad (1)$$

Four isotherm models, namely Langmuir, Freundlich, Temkin and Dubinin-Radushkevich (D-R) were selected for this study (Table 1). The best-fitting model was determined by comparing the correlation coefficient (R^2) obtained from the linear regression analysis (Table 2). The model with the R^2 value closest to 1 is considered the most appropriate fit.

Table 1. Linearized adsorption-isotherm models and parameter definitions used in this study.

Model	Linearized Equation	Description
Langmuir	$\frac{C_e}{q_e} = \frac{1}{q_m K_L} + \frac{1}{q_m} C_e$	q_m= maximum adsorption capacity (mg/g) K_L= Langmuir constant (L/mg)
Freundlich	$lnq_e = lnK_F + \frac{1}{n} lnC_e$	K_F= Freundlich capacity factor (mg/g)(L/mg)$^{1/n}$ n = Freundlich intensity parameter (dimensionless)
Temkin	$q_e = B \ln A_T + B lnC_e$ $B = \frac{RT}{b}$	A_T = Temkin binding constant (L/mg) b = Temkin constant related to heat of adsorption (J/mol) R = Gas constant (8.314J/(mol.K)) T= Absolute temperature (K)
D-R	$lnq_e = lnq_{m,DR} - K_{DR}\varepsilon^2$ $\varepsilon = RT \ln(1 + \frac{1}{C_e})$	$q_{m,DR}$ = maximum adsorption capacity (mg/g) K_{DR}= D-R constant J^{-2}) R = Gas constant (8.314J/(mol.K)) T= Absolute temperature (K)

Table 2. Basis for data acceptance and isotherm model selection.

Criterion	Description
Calibration validity	$R^2 \geq 0.95$
Model selection	Highest R^2 value (closest to 1)
Mechanistic relevance	Consistency with adsorption theory

The equilibrium adsorption capacity (q_e) was calculated using Equation (2), where C_e is the nitrate concentration at equilibrium point, V is the volume of the solution (150 mL) and m is the mass of alginate beads used (5 g).

$$q_e = \frac{(C_i - C_e)\, x\, V}{m} \qquad (2)$$

3. RESULTS AND DISCUSSION

Alginate is a natural, anionic polysaccharide composed of β-D-mannuronic (M) and α-L-guluronic (G) acid units (Tordi et al., 2024). In the presence of divalent cations, such as Ca^{2+}, alginate chains undergo ionotropic gelation to form mechanically stable hydrogel beads widely used for water treatment. Recent reviews emphasize alginate's biocompatibility, low toxicity, and renewability, simple bead fabrication process, making it a green material for adsorption and immobilization (Abka-khajouei et al., 2022; Tordi et al., 2024). Figure 1 illustrates the influence of alginate bead mass on nitrate removal efficiency. As the mass of alginate beads increased from 1 g to 5 g, the nitrate removal rate improved significantly, increasing from approximately 4% to 35%. This increment is due to the higher number of available active binding sites on the surface of the beads at higher dosages (Tai et al., 2022). An increased number of active sites enhances the likelihood of nitrate ions interacting with the adsorbent, thereby accelerating the overall adsorption process. However, further increases in bead mass to 10, 15, and 20 g resulted in only marginal improvements in nitrate removal. This plateau effect is likely due to the aggregation of beads under shaker-flask conditions, which can lead to the shielding or blockage of active sites. Beads aggregation reduces the effective surface area available for adsorption per unit mass, thereby diminishing the overall efficiency of the additional adsorbent.

These findings are consistent with the observations reported by Yasir et al. (2022), where the phenol adsorption capacity of ionic liquid agar–alginate beads increased from 25% to 60% as the bead concentration increased from 5 to 30 mg/mL. Beyond this point, further increase in dosage did not result in significant improvements in phenol removal. Similarly, Hosseini et al. (2023) identified 3 g/L of Fe-exchanged nanoporous clinoptilolite as the optimal dosage for nitrate removal, achieving a 90% removal rate. Increasing the adsorbent mass beyond this level led to less than a 1% improvement in performance. In the present study, a dosage of 5 g of alginate beads was determined to be the most effective dosage for treating 100 mL of a 30 ppm nitrate solution.

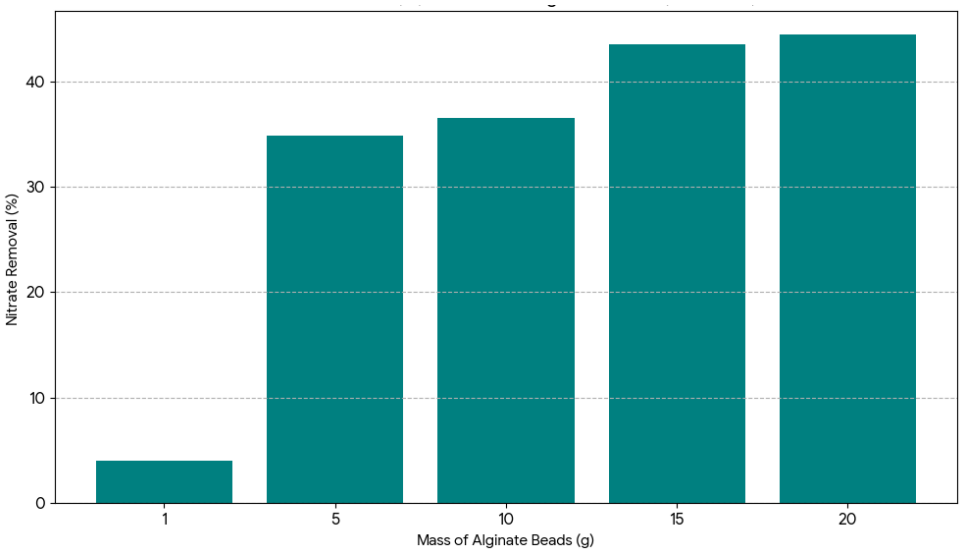

Figure 1. Effect of alginate dosage on nitrate removal efficiency

The experimental data for nitrate adsorption onto alginate beads were analysed using the linearized forms of the Langmuir, Freundlich, Temkin, and D-R isotherm models (Figure 2 and Table 3). The best fit for each model is determined by the R^2 value. The Langmuir model provides the best fit to the experimental data with the highest R^2 value of 0.9701. This high linearity suggests that the adsorption of nitrate onto the alginate beads is dominated by a monolayer adsorption process on a surface with a finite number of uniform adsorption sites (Langmuir, 1918). Based on the Langmuir model, the maximum monolayer adsorption capacity was determined to be 0.414 mg/g. This value represents the theoretical limit of nitrate that can be removed by 5 g of alginate beads under experimental conditions.

Table 3. Isotherm model parameters and the corresponding R^2 for nitrate adsorption onto alginate beads

Model	Constants	R^2
Langmuir	q_m= 0.414 mg/g K_L= 0.485 L/mg	0.9701
Freundlich	K_F= 0.188 (mg/g)(L/mg)$^{1/n}$ n = 4.879	0.5988
Temkin	A_T = 10.837 L/mg b = 3.670 x 10^4 J/mol	0.4701
D-R	$q_{m,DR}$ = 0.378 mg/g K_{DR}= 7.698 x 10^{-7} J^{-2}	0.6135

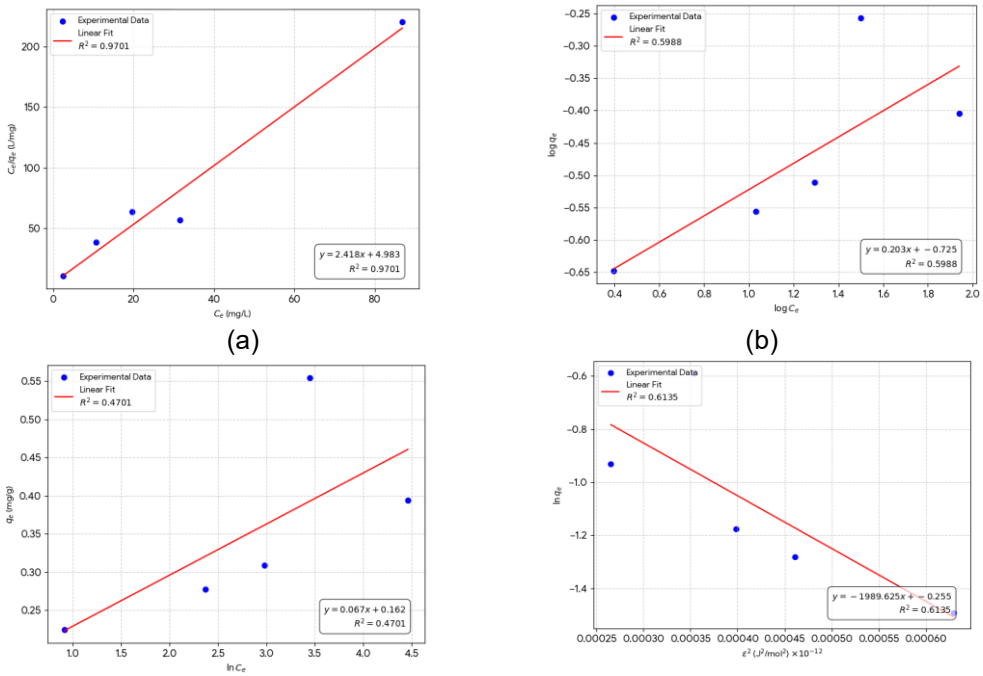

Figure 2. The linearized graph for (a) Langmuir isotherm (b) Freundlich isotherm (c) Temkin isotherm and (d) D-R isotherm.

A higher value of the Langmuir constant ($KKLL$) indicates a stronger affinity of the adsorbate for the 172 adsorbent, which is generally considered better for the removal process (Langmuir, 1918). In this study, 173 $KKLL$ is 0.485 L/mg indicating a moderate attraction between the nitrate ions and the alginate surface. 174 Both the Freundlich (R2 = 0.5988) and D-R (R2 = 0.6135) models showed a moderate fit, indicating that 175 heterogeneous adsorption, contributed minor impact to the adsorption process. Meanwhile, Temkin 176 model demonstrated a poor fit of nitrate adsorption data, indicating that the assumption of a linear 177 change in the heat of adsorption does not accurately describe this adsorption process (Temkin & 178 Pyzhev, 1940).

Based on the general trend in adsorption studies involving alginate-based materials for nitrate removal, it is reasonable to conclude that the Langmuir isotherm model provides a good fit for the equilibrium data. This suggests a dominant monolayer adsorption mechanism, which aligns with the typical chemical modification strategies used for alginate-based materials to target nitrate anions. Abo-El-Enein et al. (2017) modified alginate beads with nano-hydroxyapatite and the data demonstrated that the nitrate adsorption typically follows a monolayer adsorption mechanism. This is because alginate itself contains carboxyl (-COOH) and hydroxyl (-OH) groups (Shahrbabak et al., 2025; Yan et al., 2024). For effective nitrate removal, it is typically modified by incorporating metal oxides such as Fe or other functional groups, including quaternary

amines (Hosseini Nami & Mousavi, 2023). These modifications create new, dedicated, and often specific NO_3^- binding sites, making the surface more homogeneous for the targeted adsorbate, which is the key assumption of the Langmuir model.

The q_m of the alginate beads for nitrate removal in this study, was found to be 0.414 mg/g. Compared to recent studies utilizing modified alginate and other high-performance adsorbents published within the last decade, this capacity is considerably lower, as presented in Table 4. This indicates that the native carboxylate/hydroxyl functionality and texture of unmodified alginate provide only limited affinity and accessible sites for NO_3^- under the tested conditions. For instance, Fe-clinoptilolite/alginate composites achieved 24.39 mg/g to 36.63 mg/g of nitrate (Hosseini Nami & Mousavi, 2023). This suggested that the superior performance reported in these studies arises not from the alginate backbone itself. Introducing functionalities such as additional cationic sites, metal-oxide surfaces, or porous inorganic support explains the superior nitrate adsorptive performance. Han et al. (2024) developed a novel polyvinyl alcohol (PVA)/sodium alginate (SA) hydrogel bead entrapping the salt-tolerant composite bioagent AHM-M3, exhibiting complete nitrate removal within 48 hours in batch experiments. Similarly, the PVA/SA entrapped biochar fabricated by Wang et al. (2022), removed 94.64% of nitrate. The outcome of this study establishes the baseline limit for unmodified Ca–alginate and quantitatively justify the need of chemical modification or bio-immobilisation is required to move from modest removal and low q_m to competitive performance. This quantitative baseline therefore provides a reference framework for rational design and benchmarking of future alginate-based nitrate adsorbents under comparable isotherm analyses.

Table 4. Comparison of maximum adsorption capacities q_m for nitrate removal using different adsorbents

Adsorbent	q_m	References
Chitosan based adsorbents	0.247 – 250.63 mg/g	Italiya & Subramanian, 2022
Zeolite based adsorbents	0.395 – 125 mg/g	Italiya & Subramanian, 2022
Fe-clinoptilolite/alginate composites	24.39 - 36.63 mg/g	Hosseini Nami & Mousavi, 2023
Biochar-Supported Al-Substituted Goethite	96.147 mg/g	Wang et al., 2022
Alginate–Moroccan clay bio-composite	52.63 mg/g	Aziam et al., 2023
Alginate	0.414 mg/g	This study

4. CONCLUSIONS AND RECOMMENDATIONS

The finding of this study confirmed that the mass of alginate beads used as adsorbent is a critical factor, with an effective dosage of 5 g, exhibits a maximum nitrate removal efficiency of approximately 35% before the onset of saturation and bead aggregation effects. Analysis of the equilibrium data revealed that the Langmuir isotherm model provided the best fit with a R^2 of 0.9701. This suggested that nitrate adsorption onto the alginate surface is primarily governed by a monolayer adsorption mechanism on uniform sites. Future research must focus on modifying the alginate

beads to enhance their affinity for the anionic nitrate species. Incorporating cationic species such Fe-oxides, or high-surface-area materials like biochar, would create dedicated and stronger binding sites necessary to drastically increase capacity, approaching the competitive performance levels demonstrated by engineered alginate composites. Besides, material characterisation using techniques such as scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), and Brunauer–Emmett–Teller (BET) analysis is also recommended to achieve a more comprehensive understanding of the surface morphology, functional groups, and surface area of the alginate beads. This provides deeper insight into the structure–performance relationship that governs the nitrate adsorption. Lastly, a wider range of nitrate concentrations, additional operational parameters, including pH, contact time, and temperature, will be investigated in future studies to further strengthen the robustness of the adsorption modelling.

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