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RECEIVED 07 December 2023

ACCEPTED 08 January 2024

PUBLISHED 23 January 2024

## CITATION

Wang M, Wang N, Zhou J, Yuan J, Jiang L, Lu L, Yan J, Gong S, Zeng Y, Zhang R, Zhi L and Li G (2024) Removal of chromate in aqueous solutions by termite nests and reduction chromate accumulation in *Brassica chinensis* L.. *Front. Sustain. Food Syst.* 8:1351964. doi: 10.3389/fsufs.2024.1351964

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# Removal of chromate in aqueous solutions by termite nests and reduction chromate accumulation in *Brassica chinensis* L.

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Chromate [Cr(VI)] is a highly toxic heavy metal element, representing one of the most prevalent sources of wastewater contamination. It poses a significant threat to human health and food safety. Therefore, effective treatment before discharging wastewater is of paramount importance. In this study, termite nests (*Coptotermes formosanus* and *Odontotermes formosanus*), as natural biomass materials, were used to adsorb Cr(VI) ions in wastewater as a strategy to reduce environmental pollution and minimize poisoning by Cr. Structural and morphological characterizations were performed using scanning electron microscope (SEM), Fourier Transform Infrared Spectroscopy (FT-IR) and X-ray photoelectron spectroscopy (XPS) analyses. Batch adsorption experiments were conducted to evaluate the influence of termite nest dosage, coexisting ions, and pH. To gain further insight, detailed studies on adsorption kinetics, adsorption isotherms, and adsorption thermodynamics were undertaken. The results indicate that under acidic pH conditions, both termite nests exhibit the highest adsorption capacity for Cr(VI), with an optimal adsorbent dosage of 10 g/L. The maximum adsorption capacities of *C. formosanus* nest and *O. formosanus* nest for Cr(VI) were found to be 48.52 mg/g and 35.99 mg/g, respectively. Thermodynamic studies confirmed the spontaneous and endothermic nature of the adsorption process. In the rapeseed cultivation experiment, the growth status of *Brassica chinensis* L. post-adsorption treatment was markedly improved compared to the untreated group. Additionally, the concentration of Cr(VI) in the plants was significantly reduced. This demonstrates both the inhibitory effect of Cr(VI) on the growth of oilseed rape and the effectiveness of water remediation techniques. In addition, both types of termite nests can be effectively reused by 0.1 mol/L HCl. To the best of our knowledge, this is the first report of adsorption removal of Cr(VI) by *C. formosanus* nest and *O. formosanus* nest. Compared to traditional natural biomass adsorbents, termite nests exhibit a relatively higher adsorption capacity for Cr(VI). The results of this study demonstrate that subterranean termite nests can efficiently remove Cr(VI) from wastewater, offering the potential for a cost-effective and reusable bioremediation agent with the advantages of ease of operation.

## KEYWORDS

chromate, termite nest, adsorption, removal, *Brassica chinensis* L.

# 1 Introduction

Water, crucial for human life, health, and agriculture, is a fundamental resource. Due to the rapid expansion of industrial production, water resources are facing severe threats, with particular attention being drawn to the issue of heavy metal pollution. Chromium is an element belonging to Group VIB of the periodic table, and it exists primarily in two stable forms: trivalent chromium [Cr(III)] and hexavalent chromium [Cr(VI)] (Sharma et al., 2022). It is commonly generated during industrial processes such as electroplating, leather manufacturing, wood preservativation, and steel production (Singh et al., 2022). The toxicity and mutagenicity of Cr(VI) are, 100 and 1,000 times greater than that of Cr(III), respectively (Benimeli et al., 2011). When irrigating farmland with chromium-contaminated water, chromium accumulates within the plant tissues. Furthermore, the breakdown of organic matter in the soil is inhibited, resulting in reduced agricultural yields (Hayat et al., 2012). The ingestion of hexavalent chromium by the human body can potentially lead to acute and chronic health issues, including disruptions in the digestive system, lung damage, carcinogenicity, and genetic toxicity (den Braver-Sewradj et al., 2021). Therefore, the removal of hexavalent chromium contamination is an urgent task in safeguarding food security and public health.

Different methods for the removal of heavy metals from aqueous solutions have been proposed (Qin et al., 2020). These methods include solvent extraction, ion exchange filtration, membrane separation, reverse osmosis, chemical precipitation, and coagulation. However, each of these methods has its drawbacks, including incomplete removal of metals, high energy consumption, costly reagents, disposal of large volumes of organic solvents, and inefficiency at metal concentrations below 10 mg/L (Amer et al., 2010). Consequently, the adsorption process is emerging as a viable alternative, devoid of the limitations associated with the previously mentioned techniques. Present research concentrates on adsorbents originating from natural or modified materials, alongside synthetic sources. These adsorbents are comprised of bacteria, agricultural products, and other nonliving biomass such as chitosan and clays (Loukidou et al., 2004; Baran et al., 2007). Compared to synthetic adsorbents, natural materials offer high efficiency and low operating costs, making them an economical alternative that can help minimize pollutant inputs to the environment. In natural ecosystems, termite nests constitute a widely prevalent yet underexploited biomass resource. However, research into the utilization of termite nests as biomass adsorbents remains in its nascent phase. Currently, there are no research reports on the use of *C. formosanus* nest and *O. formosanus* nest as biomass adsorbents for treating aqueous media containing Cr(VI). Exploring this area may provide new perspectives and methods for bioremediation of heavy metal pollution.

Termites are eusocial insects (Isoptera: Termitidae), and constitute 10% of all animal biomass (Van Huis, 2017), with approximately 3,105 species found worldwide (Lertlumnaphakul et al., 2022). Around 185 species have been identified as pests, posing a significant threat to crops and buildings, especially in tropical and subtropical regions (Verma et al., 2009). Termites possess the ability to break down cellulose, which is one of

the primary components of wood (Ahmad et al., 2021a). It is estimated that each year termites cause a noteworthy economic loss of more than 40 billion USD worldwide (Subekti et al., 2015). Termites damage the agricultural and horticultural crops directly (through feeding on the bark and underlying tissue) and indirectly (the damaged plants become susceptible to pathogenic microbes) resulting in plant death (Paul et al., 2018). Infestations start from the root and then spreads to the whole plant. Furthermore, certain species of termites, such as *O. formosanus*, constitute a significant threat to the structural integrity of dams (Tian et al., 2009). Developing scientific methods for the prevention, control, and utilization of these hazardous termites represents a significant research challenge.

The management of termites using physical and mechanical means is a common practice worldwide to keep the population at a certain level (Ahmad et al., 2021b). According to the current Chinese local “Technical code of practice for prevention and control of termite in forest” (DB32/T 4588-2023) and “Technical specification for termite prevention and control in water conservancy projects” (DB42/T 768-2011), the nest-digging method involves digging out the termites from the nest, removing the termite kings and queens, and simultaneously killing the living organisms in the termite nest. The methods for termite control primarily involve nest excavation and the use of synthetic chemical agents, which often result in the generation of substantial amounts of discarded termite nests and additional environmental impacts. If these abandoned termite nests can be effectively utilized, it would not only achieve the reuse of resources but also help to reduce the use of synthetic chemical reagents, playing a positive role in environmental protection and resource recycling. Termite nests typically consist of cellulose components, saliva, excreta, and soil. The composition and unique structure make them potentially suitable as biomass adsorbents (Araújo et al., 2013), modified termite nests for the adsorption of cadmium ions, lead ions, and organic compounds (Cunha et al., 2018), and termite mounds for adsorbing lead and zinc ions, among others (Abdus-Salam and Bello, 2015). However, the exploration of termite nest biomass as an adsorbent material remains under-researched.

Therefore, this study proposes the use of termite nests from two common subterranean termite species (*C. formosanus* and *O. formosanus*) for remediating water contaminated with Cr(VI). The research involved the chemical characterization of this novel adsorbent and investigated its mechanism for Cr(VI) removal. This method transforms waste biomass into valuable components, giving new purpose to these bio-wastes while addressing the challenges of their safe disposal. This study aims to provide a novel, sustainable solution for overcoming heavy metal pollution issues while promoting the integrated development of environmental protection, resource recycling, and public health.

## 2 Materials and methods

### 2.1 Materials and chemicals

In this study, nest samples of the *C. formosanus* nests were collected from Nanning, Guangxi, while samples of the

*O. formosanus* nests were obtained from Huangshi, Hubei. The termite nests were washed with deionized water, air-dried at room temperature ( $25 \pm 2^\circ\text{C}$ ), and sieved through a 60-mesh screen (Araújo et al., 2013). The resulting material was used in the adsorption experiments.

Potassium dichromate ( $\text{K}_2\text{Cr}_2\text{O}_7$ ), sulfuric acid ( $\text{H}_2\text{SO}_4$ ), hydrochloric acid (HCl), phosphoric acid ( $\text{H}_3\text{PO}_4$ ), sodium hydroxide (NaOH), and diphenylcarbazide ( $\text{C}_{13}\text{H}_{10}\text{N}_2\text{O}_2$ ) were all of the analytical grade and supplied by Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). In this research, the pH values of all solutions were measured using a pH meter (PHSJ-4F, Leici Shanghai) and adjusted with sodium hydroxide (NaOH) and hydrochloric acid (HCl) as needed. All solutions were prepared with deionized (DI) water.

## 2.2 Adsorption experiments

To investigate the adsorptive properties of termite nest material from *C. formosanus* and *O. formosanus*, all batch experiments were conducted in Erlenmeyer flasks maintained at a controlled temperature of  $25 \pm 2^\circ\text{C}$  (Jing et al., 2021). In each flask, a designated amount of termite nest material was combined with a fixed concentration of Cr(VI) solution and agitated on a shaker at a speed of 150 rpm. The primary variables adjusted included the concentration of Cr(VI) affecting adsorption (100–1,000 mg/L), pH value (2–12), termite nest dosage (1–12 g/L), contact time (5–2,160 min), and coexisting ions ( $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{3-}$ ). After the predetermined adsorption period, the aqueous solution was filtered through a membrane with a pore size of 0.45  $\mu\text{m}$ . Subsequently, a 5 mL sample of the filtered aqueous solution was taken for analysis. The initial and final concentrations of Cr(VI) were determined by the colorimetric diphenylcarbazide (DPC) method (Xia et al., 2018a,b). Each step of the experiment was repeated thrice for accuracy. The amount of Cr(VI) adsorbed by the termite nest material was calculated according to Equation (1):

$$q_t = \frac{(C_0 - C_t)V}{m} \quad (1)$$

where  $q_t$  is the adsorbed capacity after time  $t$ ,  $C_0$  and  $C_t$  are the initial and residual concentrations after time  $t$ , respectively,  $V$  is the volume of the solution, and  $m$  is the dry weight of the adsorbent.

## 2.3 Adsorption kinetics and adsorption isotherms experiments

The investigation of adsorption kinetics is critical as it characterizes the solute uptake rate, predicts the rate-determining steps, and elucidates the adsorption mechanism. For the adsorption kinetics analysis, *C. formosanus* and *O. formosanus* nests (each at 4 g/L) were incubated with Cr(VI) solutions at varying concentrations (50, 75, and 100 mg/L) under optimal pH conditions at 298 K for 24 h. Samples were collected at predetermined intervals. The quantification of residual Cr(VI) ions was conducted by Method 2.2.

Adsorbent capacity is determined through equilibrium studies. The Freundlich, Langmuir, and Temkin isotherms represent the simplest known models to describe the adsorption isotherms. In the analysis of adsorption isotherms, *C. formosanus* and *O. formosanus* nests (each at 4 g/L) were incubated with Cr(VI) solutions at various concentrations under optimal pH conditions and temperatures of 298, 308, and 318 K for 24 h. The concentration of residual Cr(VI) was quantified using the aforementioned method.

## 2.4 Characterization of the termite nest

Surface morphology was identified by a scanning electron microscope (SEM: HITACHI, Japan). The specific surface area ( $S_{\text{BET}}$ ), total pore volume ( $V_{\text{tot}}$ ), and average pore size ( $D_p$ ) were studied using a gas adsorption analyzer (Micromeritics, USA). All pH measurements were performed with a pH meter (INESA, China). The pH at the point of zero charges ( $\text{pH}_{\text{pzc}}$ ) was determined using the method described by Li et al. (2021). The functional groups of the material were studied using Fourier transform infrared spectroscopy (FT-IR: Thermo, USA). The surface elements of the adsorbent were analyzed qualitatively and quantitatively using X-ray photoelectron spectroscopy (XPS: Kratos, UK).

## 2.5 Desorption experiments

In the desorption study, 50 mg/L Cr(VI) solutions were adsorbed under optimized conditions using 4 g/L of both *C. formosanus* nests and *O. formosanus* nests. Subsequently, samples were collected through centrifugation at a speed of 12,000 rpm for 10 min. The samples were then resuspended and the adsorbents were regenerated using HCl (0.1 mol/L) (Xia et al., 2018a,b). The efficiency of desorption was calculated using the following equation:

$$\text{Desorption percentage (\%)} = \frac{\text{Concentration desorbed}}{\text{Concentration adsorbed}} \times 100\% \quad (2)$$

Five consecutive adsorption/desorption cycles were performed to test the reusability of the adsorbents from *C. formosanus* and *O. formosanus* nests. The removal efficiency for each cycle was calculated as follows:

$$\text{Removal percentage (\%)} = \frac{\text{Concentration adsorbed}}{\text{Initial concentration}} \times 100\% \quad (3)$$

# 3 Results and discussion

## 3.1 Characterization of the termite nest

The porous structures of the *C. formosanus* nest and the *O. formosanus* nest are presented in Table 1. Both types of termite nests possess a considerable specific surface area, total pore volume, and average pore diameter. This can likely be attributed to the similarity in the primary components of the two nests, which both

TABLE 1 Porous characteristics of the termite nest.

Adsorbents	$S_{\text{BET}}$ ( $\text{m}^2/\text{g}$ )	$V_{\text{tot}}$ ( $\text{cm}^3/\text{g}$ )	$D_p$ (nm)
<i>C. formosanus</i> nest	2.2951	0.0166	28.8727
<i>O. formosanus</i> nest	1.7418	0.0112	25.6438

predominantly contain lignocellulosic materials. Notably, the *C. formosanus* nest exhibits greater porosity and accessible surface area, which may enhance its adsorption capacity for Cr(VI) (Song et al., 2016).

The surface morphology of the termite nest before and after the adsorption of Cr(VI) was examined using a scanning electron microscope (SEM). SEM images of the nests of *C. formosanus* and *O. formosanus* before the adsorption of Cr(VI) depicted a rough surface; the irregular texture and varying porosity within the termite nests provided high surface area and suitable sites for the adsorption of Cr(VI) (see Figures 1A, C). Following the adsorption of Cr(VI), a significant area of the termite nests was covered by adsorbed molecules. Post-adsorption SEM images of both types of termite nests displayed surfaces smoother than before, indicative of the trapping and adsorption of heavy metal ions on their surfaces (see Figures 1B, D) (Liu et al., 2020).

The mode of the bonding in metal compounds can be determined by the infrared spectra. Comparative analysis of the infrared spectra of nests from *C. formosanus* and *O. formosanus* reveals that the primary absorption bands are fundamentally identical, with notable absorption peaks occurring near wavelengths of 3,400, 2,900, 1,400, and 1,050  $\text{cm}^{-1}$  (see Figure 2A). However, certain variations in peak shapes and absorption intensities were observed. These suggested that while the samples share similar structures and functional groups, their relative concentrations vary. Examining the infrared spectra of both termite nests before and after adsorption shows slight shifts in the intensity and position of these absorption peaks, indicating the potential involvement of these functional groups in the adsorption process (Jing et al., 2021). The broad absorption peak between 3,500 and 3,200  $\text{cm}^{-1}$ , attributable to the stretching vibrations of phenolic hydroxyl groups, alters slightly in intensity and position post-adsorption, likely due to metal ions complexing with hydroxyl groups on the termite nest surfaces through surface adsorption (Meng et al., 2014). Absorption peaks near 1,750, 1,450, and 2,900  $\text{cm}^{-1}$ , originating from hydroxyl O-H or C-H stretching vibrations, indicate the presence of numerous hydroxyl and other oxygen-containing functional groups in the termite nests, conducive to ion exchange reactions (Mansour et al., 2020). A noticeable decrease in the intensity of these peaks post-adsorption suggests ion exchange between the termite nest and metal ions. The ion exchange between metal and mineral ions facilitates electrostatic outer-sphere complexation and coprecipitation/inner-sphere complexation (Qiu et al., 2008). The strong absorption peaks between 1,100 and 1,000  $\text{cm}^{-1}$ , associated with C-O bond stretching vibrations, change in intensity after heavy metal adsorption, indicating the involvement of ether-type oxygen-containing groups in the adsorption process (Shukla and Vankar, 2012). In conclusion, the positions of the absorption peaks of

hydroxyl, carboxyl, and C-O oxygen-containing functional groups in both termite nests changed significantly after the adsorption of heavy metals, indicating that the adsorption process in the termite nests was primarily due to the complexation mechanism between Cr(VI) ions and these functional groups.

The isoelectric points of *C. formosanus* nest and *O. formosanus* nest are shown in Figure 2B. From the graph, the isoelectric points of *C. formosanus* nest and *O. formosanus* nest are 5.93 and 4.90, respectively. This indicates that the surfaces of both termite nests have a relatively higher abundance of acidic functional groups. When the solution pH is below the isoelectric point, the adsorbent surfaces carry a positive charge, which is favorable for the electrostatic attraction-based adsorption of anions as the predominant mechanism. On the other hand, when the pH is above the isoelectric point, the adsorbent surfaces become negatively charged, facilitating the electrostatic attraction-based adsorption of cations as the predominant mechanism (Araújo et al., 2013).

The study employed X-ray Photoelectron Spectroscopy (XPS) to observe changes in the binding energy and valence states of elements within two types of termite nests before and after adsorption, as depicted in Figure 3. Compared to the pre-adsorption state, the XPS spectra of the termite nests post-adsorption of Cr(VI) revealed a new binding energy peak region, Cr2p, confirming the adsorption of Cr(VI) onto the surfaces of the termite nests (see Figure 3A). In the post-adsorption Cr2p spectra of both termite nests, a binding energy of 579 eV corresponding to Cr(VI) was observed, along with a weaker binding energy at 575 eV, indicative of a minor presence of Cr(III) (Shi et al., 2018). This observation suggests a partial reduction of Cr(VI) to Cr(III) during adsorption, likely facilitated by the interaction with functional groups on the termite nest surface (see Figure 3B). This reduction process is possibly attributable to the presence of hydroxyl and carbonyl groups in the adsorbent. Under acidic conditions, these groups can act as electron donors, thereby reducing Cr(VI) to its trivalent form (Suksabye et al., 2009).

In summary, the experimental findings suggest that the adsorption of Cr(VI) by the two types of termite nests primarily involves electrostatic interactions, complexation, and the partial reduction of Cr(VI) to Cr(III), which is then followed by complexation with surface functional groups.

### 3.2 Adsorption of Cr(VI) under different conditions

Figure 4A demonstrates that as the solution pH increases, the adsorption capacity for Cr(VI) by *C. formosanus* and *O. formosanus* termite nests initially decreases rapidly and then stabilizes. This trend can be explained by several factors. When the solution pH is below the isoelectric point, the high concentration of  $\text{H}^+$  ions protonates the functional groups on the surface of the termite nests, resulting in electrostatic attraction to Cr(VI) in the solution. The existence form of Cr(VI) varies with pH: as  $\text{H}_2\text{CrO}_4$  at a pH below 1, as  $\text{HCrO}_4^-$  between pH 1 and 6, and as  $\text{CrO}_4^{2-}$  at a pH above 6.  $\text{HCrO}_4^-$  requires fewer active adsorption sites compared to

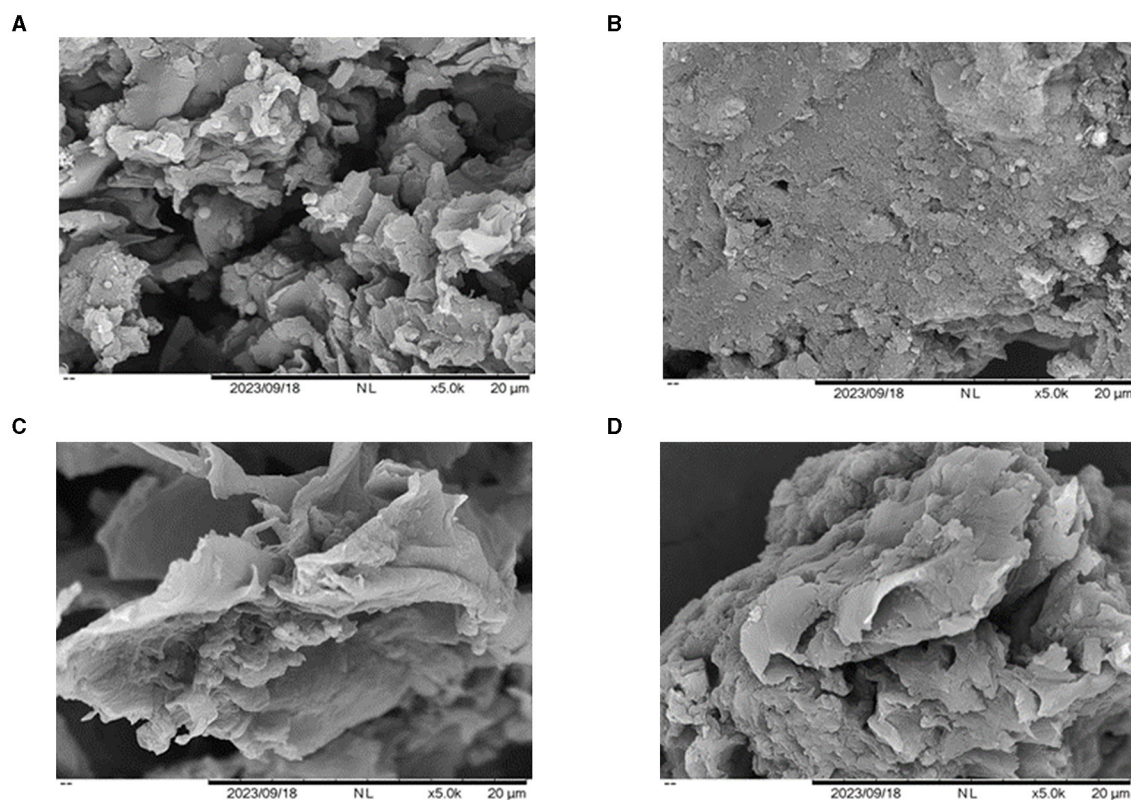


FIGURE 1 SEM micrographs of the termite nest adsorbent (×5,000). (A) *C. formosanus* nest before adsorption, (B) *C. formosanus* nest after Cr(VI) adsorption, (C) *O. formosanus* nest before adsorption, (D) *O. formosanus* nest after Cr(VI) adsorption.

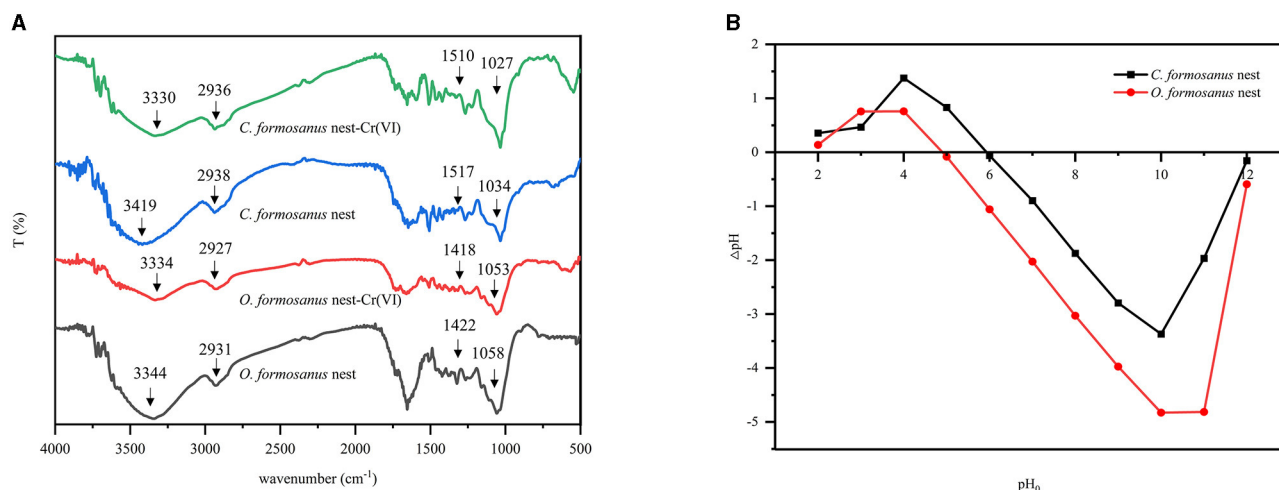
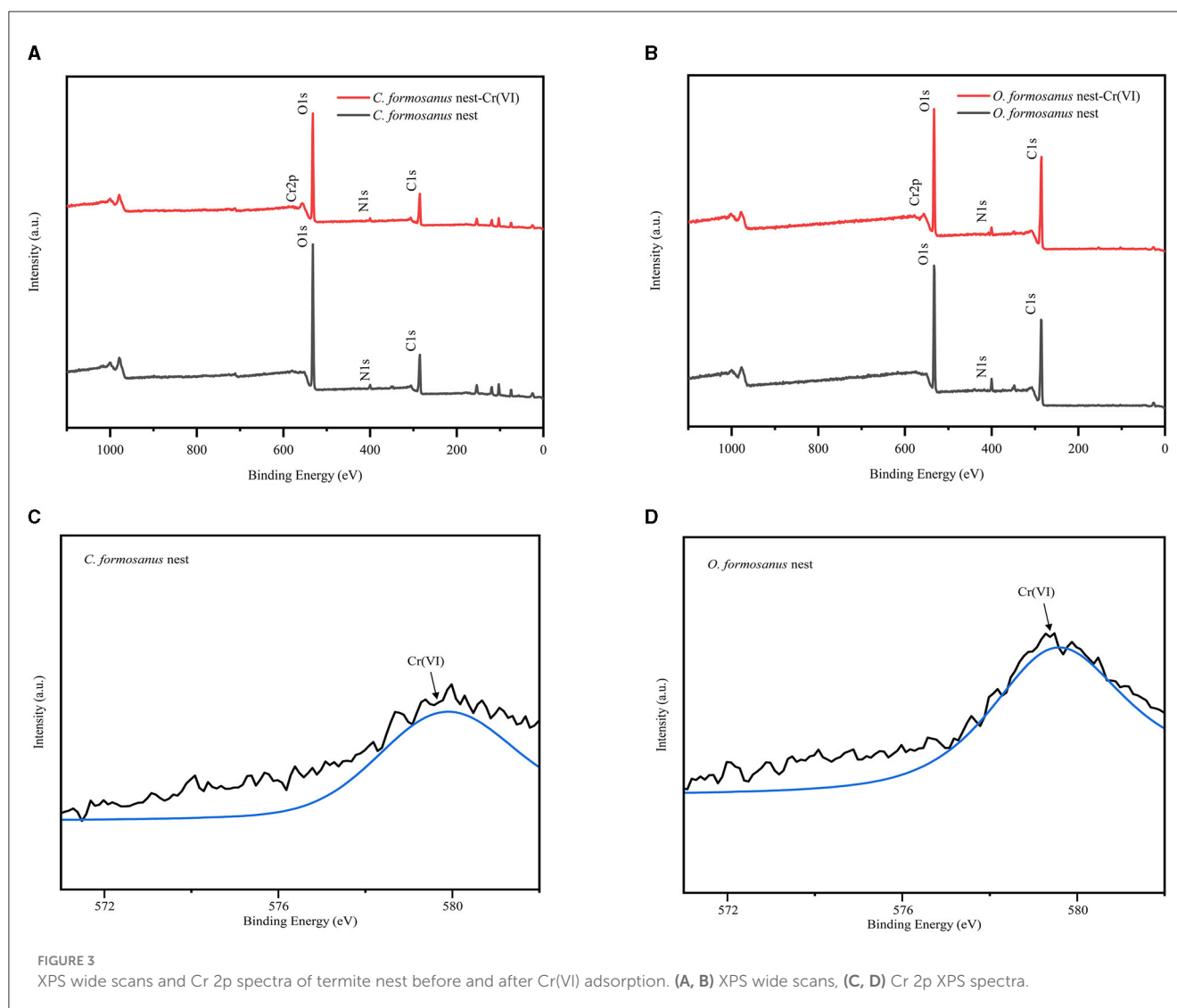


FIGURE 2 FTIR spectra and zero-charge points of termite nest. (A) FTIR spectra of fresh termite nest and Cr(VI) ions loaded termite nest, (B) determination of the point of zero charge of the termite nest.

CrO<sub>4</sub><sup>2-</sup>, hence favoring adsorption at lower pH values (Araújo et al., 2013). As the pH increases, the adsorption of Cr(VI) decreases. However, even under strongly alkaline conditions, both types of termite nests still maintain a certain degree of Cr(VI) adsorption,

indicating the involvement of specific functional groups that are complex with Cr(VI).

Industrial wastewater commonly contains various ions such as Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and PO<sub>4</sub><sup>3-</sup>, making it crucial to study their



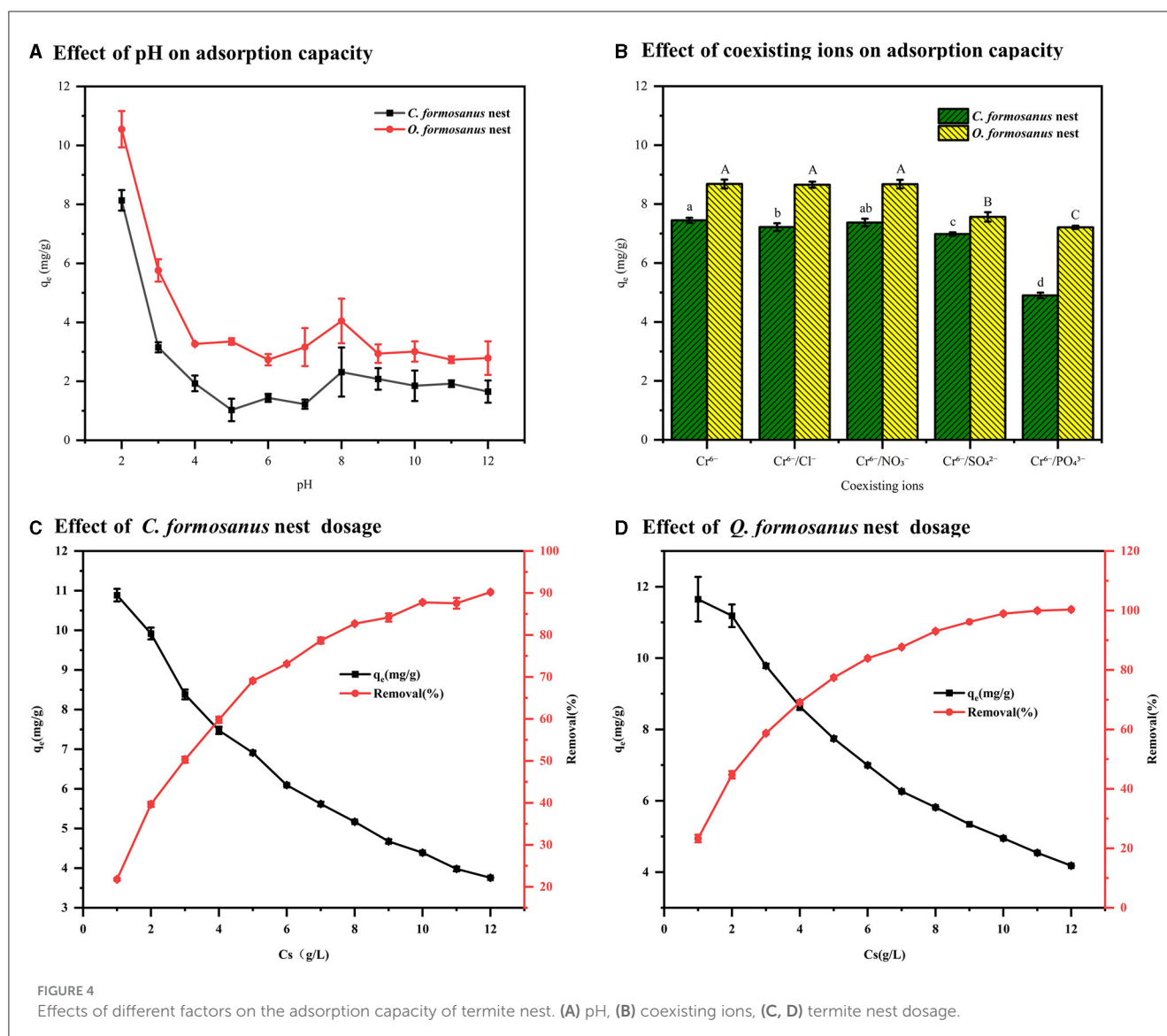
impact on the adsorption process. As detailed in Figure 4B, our experiments were conducted with a constant ion concentration of 20 mg/L and an adsorbent dose of 4 g/L, under conditions of pH 2 and a temperature of 25°C. We found that  $\text{Cl}^-$  and  $\text{NO}_3^-$  ions had minimal impact on the adsorption of Cr(VI) by both types of termite nests. In contrast,  $\text{SO}_4^{2-}$  and  $\text{PO}_4^{3-}$  significantly influenced the adsorption of Cr(VI): the removal efficiency of the *C. formosanus* nest decreased by 6.3% and 34.2%, respectively, while the *O. formosanus* nest showed a more pronounced decrease of 12.9% and 16.9%. This variation may be due to the molecular size of  $\text{SO}_4^{2-}$  and  $\text{PO}_4^{3-}$  anions being similar to the ionic radius of  $\text{HCrO}_4^-$  (Cai et al., 2019). Overall, the impact of coexisting ions on Cr(VI) adsorption by termite nests is relatively minor.

Figures 4C, D demonstrate that as termite nest dosage increases, the adsorption capacity decreases, but the removal efficiency of Cr(VI) ions from water improves. At a loading of 10 g/L, the removal efficiencies for *C. formosanus* and *O. formosanus* nests reach 87.77% and 98.95%, respectively, with adsorption capacities of 4.39 mg/g and 4.95 mg/g. This trend is attributed to the increased quantity of absorbance, offering more

functional groups for adsorption and a larger specific surface area (Castro-Castro et al., 2020). However, removal efficiency plateaus with further loading increase, likely because excessive adsorbent leads to agglomeration, reducing the contact surface area and active adsorption sites (Chen et al., 2020). Additionally, reductive substances in the termite nests react with  $\text{CrO}_4^{2-}$ , further enhancing Cr(VI) removal. Therefore, a 10 g/L loading is optimal for maximum removal efficiency, whereas a 4 g/L loading is more cost-effective.

### 3.3 Adsorption kinetics

To explore the adsorption kinetics of Cr(VI) by *C. formosanus* and *O. formosanus* nests, we examined the effect of contact time on adsorption capacity at three different Cr(VI) concentrations, using an optimal pH of 2. The findings, presented in Figure 5A, show that both nest types adsorbed Cr(VI) quickly at first but took longer to reach equilibrium. For *C. formosanus*



nesses, equilibrium was achieved at 12, 20, and 24 h for initial Cr(VI) concentrations of 50, 75, and 100 mg/L, respectively. *O. formosanus* nests reached equilibrium faster, within 6, 15, and 20 h at these concentrations. This suggests that the termite nests' Cr(VI) adsorption is influenced by the initial concentration (Xia et al., 2018a,b).

Furthermore, pseudo-first-order, pseudo-second-order and intraparticle diffusion models were performed to elucidate the adsorption kinetics. The three models were calculated as follows:

Pseudo-first-order kinetic model:

$$\log(q_e - q_t) = \log q_e - \frac{k_1 t}{2.303} \quad (4)$$

Pseudo-second-order kinetic model:

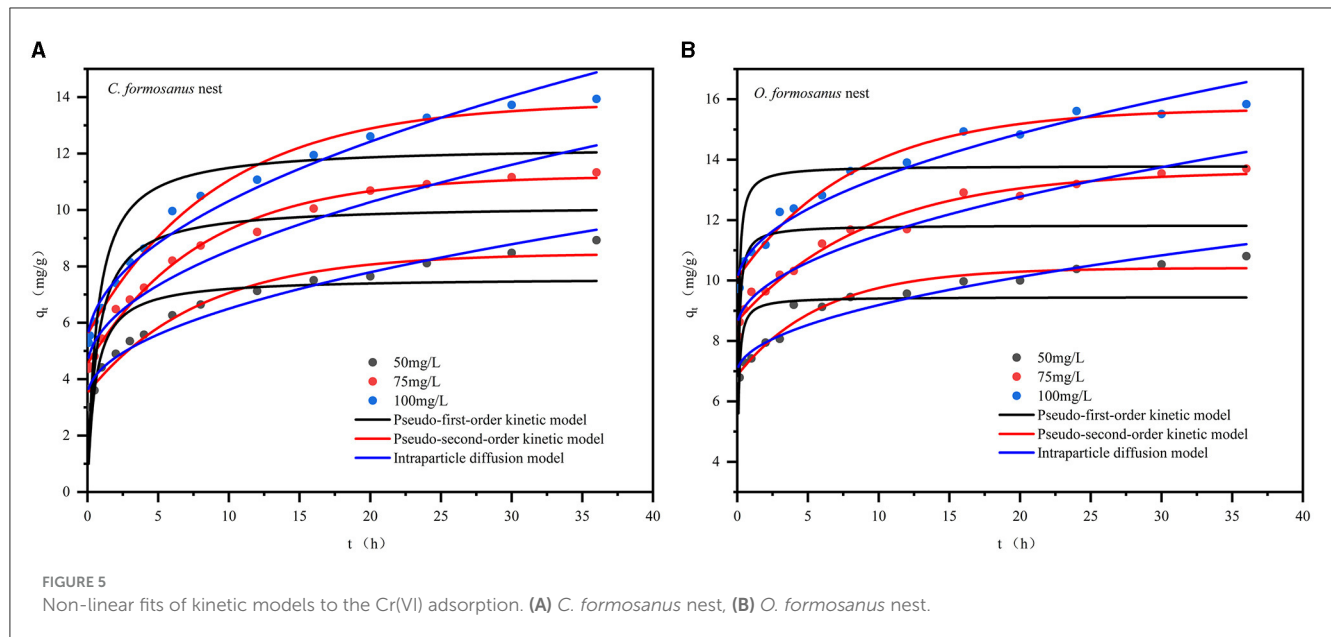
$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (5)$$

Intraparticle diffusion model:

$$q_t = k_3 t^{0.5} + c \quad (6)$$

where  $q_e$  and  $q_t$  are the amount of adsorbed Cr(VI) per unit mass of adsorbent ( $\text{mg g}^{-1}$ ) at equilibrium and time  $t$ , respectively.  $k_1$ ,  $k_2$ , and  $k_3$  are the rate constants of the pseudo-first-order adsorption ( $\text{h}^{-1}$ ), pseudo-second-order adsorption ( $\text{g mg}^{-1} \text{h}^{-1}$ ) and intraparticle diffusion ( $\text{mg g}^{-1} \text{h}^{-0.5}$ ), respectively.  $c$  indicates the thickness of the boundary layer.

Table 2 presents the kinetic parameters, and Figure 5 displays the fitting curves for Cr(VI) concentrations of 50, 75, and 100 mg/L. The  $R^2$  values for the pseudo-second-order equation ranged from 0.9748 to 0.9991 for *C. formosanus* nests and 0.9661 to 0.9828 for *O. formosanus* nests, outperforming the pseudo-first-order and intraparticle diffusion models (see Table 2). Moreover, the equilibrium adsorption values derived from the pseudo-second-order model closely align with the experimental data. This indicates



that Cr(VI) adsorption by both *C. formosanus* and *O. formosanus* nests adheres to a pseudo-second-order kinetic model. This model, which includes film diffusion, surface adsorption, and intraparticle diffusion stages, more accurately captures the complexity of the adsorption process (Sarfranz et al., 2022). Notably, similar kinetic behaviors are observed in the adsorption of heavy metals by tree termite nests, highlighting a shared characteristic among different types of nests (Araújo et al., 2013).

### 3.4 Adsorption isotherms

Comprehending the equilibrium adsorption isotherm is crucial for a thorough understanding of the termite nest's adsorption characteristics (Chen et al., 2020). To achieve this, we applied the experimental data of the termite nest to three key isotherm models: Langmuir, Freundlich, and Temkin. This approach allows for a comprehensive analysis of the adsorption properties.

Langmuir equation:

$$\frac{c_e}{q_e} = \frac{c_e}{q_m} + \frac{1}{K_L q_m} \quad (7)$$

Freundlich equation:

$$q_e = K_F C_e^{\frac{1}{n}} \quad (8)$$

Temkin equation:

$$q_e = A + B \ln C_e \quad (9)$$

where  $q_e$  (mg/g) is the equilibrium adsorption capacity of Cr(VI) on the termite nest,  $C_e$  (mg/L) is the equilibrium Cr(VI) concentration in solution,  $q_m$  (mg/g) is the maximum

capacity of the adsorbent,  $K_L$  (L/mg) is the Langmuir adsorption equilibrium constant related to the energy of adsorption,  $K_F$  (L/mg) and  $1/n$  are the Freundlich constant representing the adsorption capacity and adsorption intensity and  $A$  and  $B$  are Temkin constants.

Experimental adsorption data were collected at three temperatures: 298 K, 308 K, and 318 K, as shown in Figure 6. The isotherm fitting parameters are detailed in Table 3, with fitting curves for *C. formosanus* and *O. formosanus* nests displayed in Figures 6A, B. Correlation coefficients ( $R^2$  values, see Table 3) indicate that the adsorption behavior aligns more closely with the Freundlich model than with the Langmuir and Temkin models. The Langmuir model predicts maximum adsorption capacities ( $q_m$ ) of 48.5189 mg/g for *C. formosanus* and 35.9910 mg/g for *O. formosanus*. Additionally, the Freundlich constants ( $n$ ),  $> 1$  for both termite nests, suggest favorable and heterogeneous adsorption. This finding implies that the adsorption of Cr(VI) on termite nests under our experimental conditions likely involves both monolayer biosorption and heterogeneous surface conditions (Charazińska et al., 2022). Consequently, the overall Cr(VI) adsorption process on termite nests appears complex, encompassing mechanisms such as ion exchange, electrostatic attraction, and surface complexation, as evidenced by the characterization results.

### 3.5 Desorption and reusability study

The desorption and reusability of adsorbents are crucial for practical applications. This study investigated the reusability of termite nests as adsorbents. We employed 0.1 mol/L HCl for desorbing Cr(VI) from the termite nests. Their reusability was evaluated through consecutive adsorption/desorption cycles. After five cycles, the Cr(VI) removal efficiency of *C. formosanus* and *O. formosanus* nests decreased by  $\sim 19.00\%$  and  $28.75\%$ , respectively,



TABLE 2 Adsorption kinetics parameters fitted by the three models.

	Adsorbents	<i>C. formosanus</i> nest			<i>O. formosanus</i> nest		
	$C_0$ (mg/L)	50	75	100	50	75	100
First order	$R^2$	0.6854	0.6733	0.6382	0.5247	0.4981	0.5041
	$k_1$	0.1258	0.1251	0.1092	0.1679	0.1078	0.1200
	$q_e$ (mg/g)	8.4620	11.2106	13.810	10.4160	13.6274	15.6937
Second order	$R^2$	0.9748	0.9911	0.9911	0.9661	0.9773	0.9828
	$K_2$	0.2399	0.1474	0.1212	1.8449	1.3838	1.1833
	$q_e$ (mg/g)	7.5960	10.1756	12.2638	9.4566	11.8336	13.7978
Intraparticle diffusion	$R^2$	0.9656	0.9672	0.9781	0.9301	0.9674	0.9609
	$K_3$	1.3180	0.9866	1.6071	0.9301	0.9674	0.9609
	$c$	4.3820	3.3787	5.5334	6.9414	8.4252	9.8712

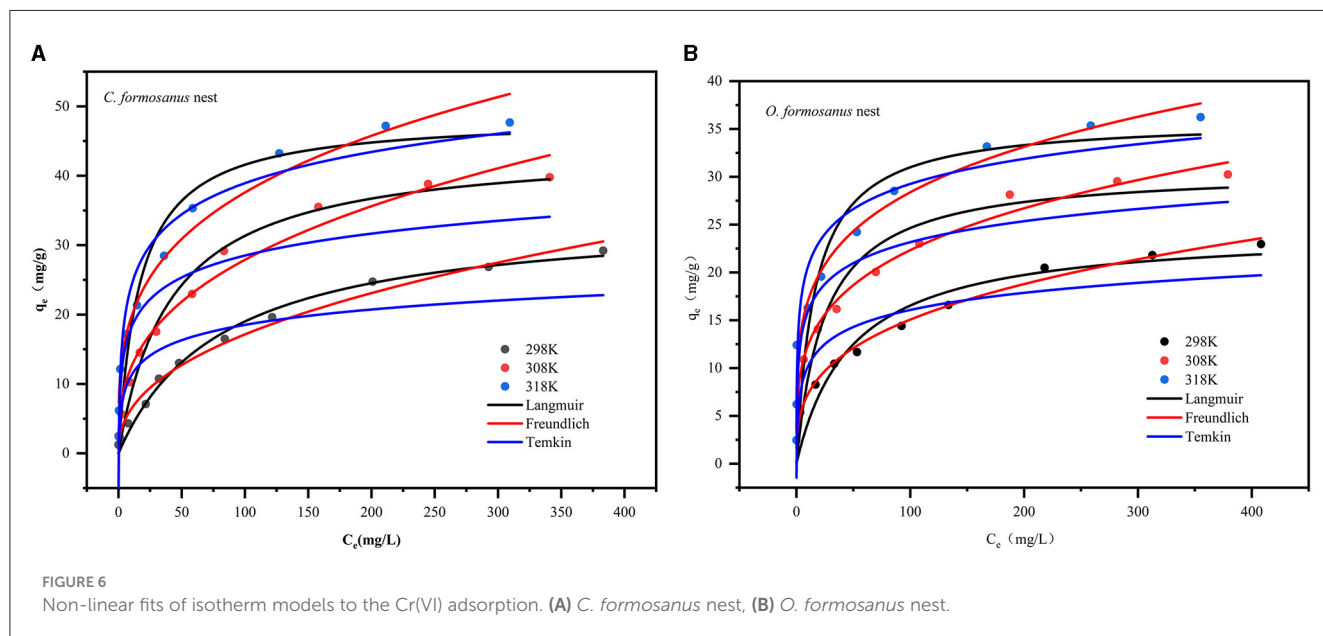
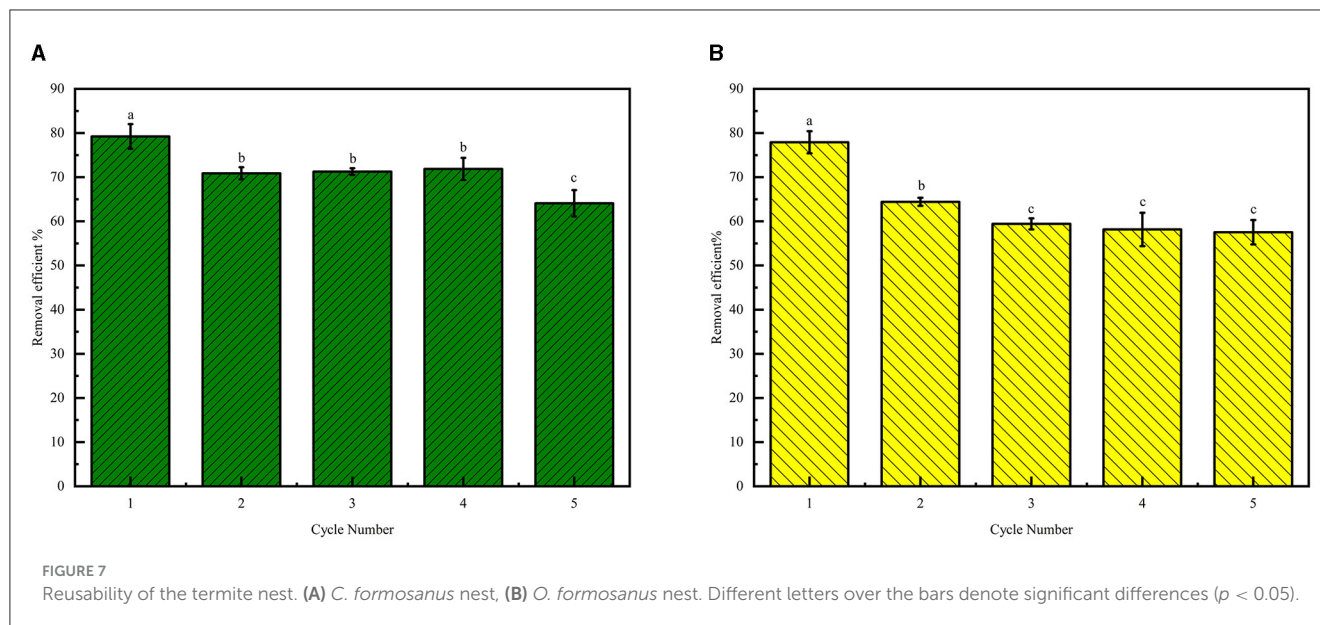


TABLE 3 Parameters of isotherm models.

	Adsorbents	<i>C. formosanus</i> nest			<i>O. formosanus</i> nest		
	Temperature (K)	298	308	318	298	308	318
Langmuir	$R^2$	0.9899	0.9770	0.9408	0.9278	0.8707	0.8459
	$K_L$ (L/mg)	0.0123	0.0242	0.0600	0.0208	0.0401	0.0620
	$q_m$ (mg/g)	34.5340	44.2923	48.5189	24.4745	30.7797	35.9910
Freundlich	$R^2$	0.9884	0.9793	0.9754	0.9931	0.9855	0.9698
	$K_F$ (mg/g)	2.3718	5.5136	10.1870	3.4825	6.7939	10.1449
	$n$	2.3278	2.8401	3.5265	3.1438	3.8698	4.4763
Temkin	$R^2$	0.7506	0.7936	0.9637	0.8395	0.9003	0.9410
	$A$	3.8326	7.5506	9.0671	4.2044	8.6058	11.8350
	$B$	-3.1893	-4.5510	-6.4859	-2.5790	-3.1616	-3.7827



as illustrated in Figure 7. The results show that both nests can be reused.

### 3.6 Pot cultivation of rapeseed

In the cultivation experiment, rapeseed (*Brassia campestris* L.) seeds were cultivated with untreated 50 mg/L Cr(VI) simulated wastewater and water treated with two types of termite nests (10 g/L) to assess their Cr(VI) adsorption or remediation efficiency. As shown in Figure 8, the rapeseed grown in the treated water exhibited notably longer stems and roots compared to the control (see Figure 8A). This suggests that Cr has an inhibitory effect on the growth of *Brassia campestris* L. The bioaccumulation of Cr by different parts of canola seedlings was further determined. The results showed that Cr accumulation was significantly reduced after termite nest adsorption treatment. For rapeseed grown in water associated with the control group, the Cr contents within roots and aerial parts were 15.6567 mg/kg and 6.4233 mg/kg, respectively; in the *C. formosanus* nest treatment group, these contents were 0.1267 mg/kg and 0.0267 mg/kg, respectively. In the *O. formosanus* nest treatment, the measured Cr contents in the roots and aerial parts of canola seedlings were 0.0733 mg/kg and 0.0067 mg/kg, respectively (see in Figure 8B). The substantial decrease in Cr levels, falling below the 0.5 mg/kg limit of China's national food safety standards, demonstrates the treatment's effectiveness (Yang et al., 2020).

### 3.7 Comparison with other biosorbents

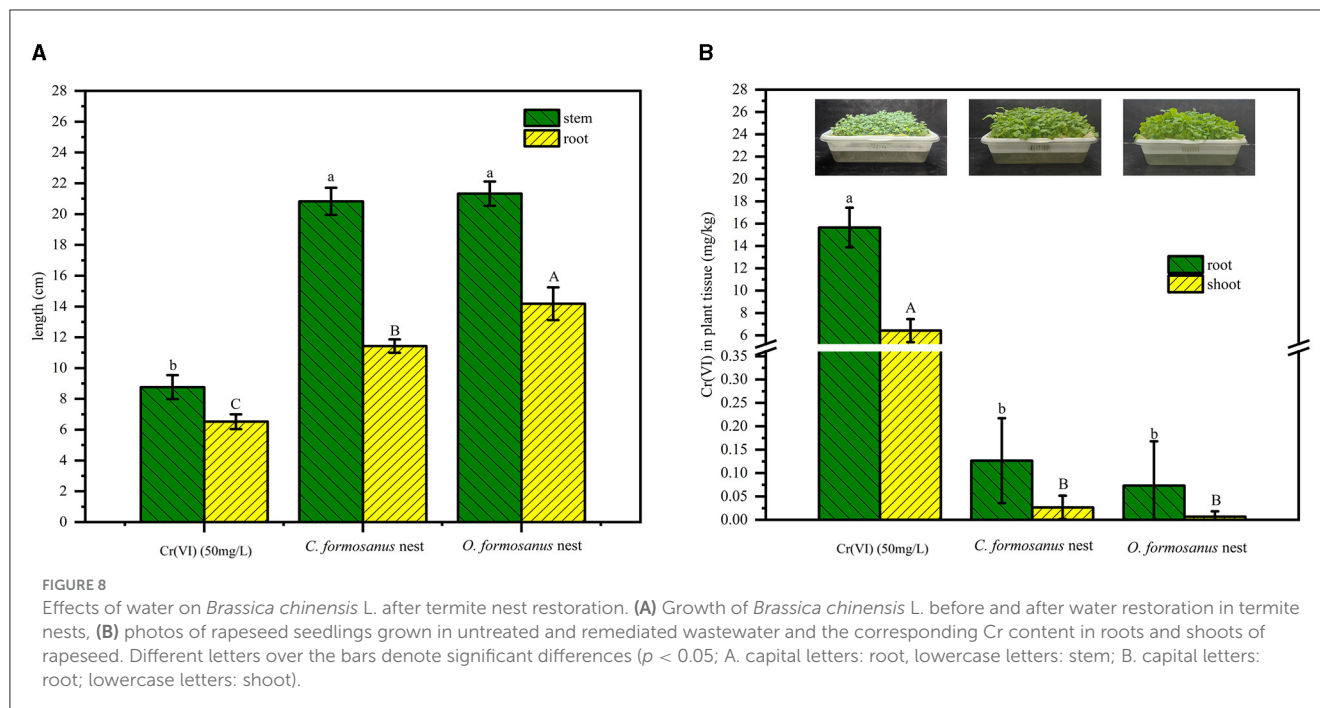
The study's findings reveal that nests of *C. formosanus* and *O. formosanus* have similar principles in Cr(VI) adsorption, yet differ in their capacities. The *C. formosanus* nest shows greater adsorption capacity and better regeneration, but is slower in adsorption rate and less efficient in removal compared to the *O. formosanus* nest. These variations may stem from the *C. formosanus* nest's larger

surface area and more adsorption sites. However, *C. formosanus* nest's tendency to aggregate during adsorption may impede Cr(VI) access to these sites, leading to a prolonged time for reaching equilibrium. This underscores the need to choose the right termite nest type for practical use, considering specific adsorption needs and conditions (Chen et al., 2020).

The biosorption capability of termite nests for Cr(VI) was compared with other low-cost biosorbents, as detailed in Table 4 from various literature sources. While direct comparisons can be challenging due to differences in experimental conditions like temperature, pH, and adsorbent dosage, our findings indicate that *C. formosanus* and *O. formosanus* nests demonstrate a reasonable Cr(VI) adsorption capacity compared to other biosorbents. Notably, termite nests are often by-products of pest control efforts and are renewable and reusable. In the field of termite control, the nest excavation method is considered an effective approach for controlling large nesting termites, especially for controlling large soil-dwelling termites in dams. This process usually requires the use of synthetic chemical reagents with potential environmental risks. In this context, an innovative proposal was made in this study: instead of using synthetic chemical reagents to exterminate the living organisms in termite nests discarded during termite control, the nests were directly collected and used for the preparation of natural biomass adsorbents. This approach not only utilizes resources that would otherwise be wasted, but also helps to reduce the use of chemical reagents, thereby reducing potential risks to the environment.

## 4 Conclusions and perspectives

The study addresses the critical issue of Chromate [Cr(VI)], a highly soluble and environmentally harmful substance that poses significant risks to food safety and human health. It necessitates finding effective adsorbents for wastewater treatment. The



**TABLE 4** Comparison of adsorption capacity of Cr(VI) anionic by some biosorbents.

Biosorbent	$q_e$ (mg/g)	Reference
Tea ( <i>Camellia sinensis</i> ) waste	7.29	Malkoc et al., 2006
Almond	10.61	Pehlivan and Altun, 2008
Saw dust	15.82	Dakiky et al., 2002
Plantain waste	16.10	Idowu et al., 2011
Opuntia cladodes of cactus pear fruits	16.40	Fernández-López et al., 2014
Ectoderm of cactus pear fruits	18.50	Fernández-López et al., 2014
Tree termite nest	18.60	Araújo et al., 2013
Ficus carica fiber	19.68	Gupta et al., 2013
Pine needles	21.50	Dakiky et al., 2002
Tannin-immobilized activated clay	24.09	Li et al., 2012
Olive cake	33.44	Malkoc et al., 2006
Wool	41.15	Owlad et al., 2009
Eucalyptus bark	45.00	Sarin and Pant, 2006
<i>C. formosanus</i> nest	48.52	This work
<i>O. formosanus</i> nest	35.99	This work

research successfully developed an innovative, effective, and eco-friendly adsorbent using *C. formosanus* and *O. formosanus* nests, demonstrating excellent Cr(VI) adsorption. Key characteristics of these termite nests include their natural origin and construction by insects, rendering them environmentally benign and free of harmful chemicals. They show superior adsorption efficiency,

especially at pH 2 and a Cr(VI) concentration of 50 mg/L, with an optimal adsorbent dose of 4 g/L. Kinetic analysis aligns their adsorption with the pseudo-second-order model, and their maximum adsorption capacities are 48.52 mg/g for *C. formosanus* and 35.99 mg/g for *O. formosanus*. Analytical techniques like SEM, point of zero charge, FTIR, and XPS reveal that chemical adsorption is the primary mechanism, involving electrostatic interactions, complexation, and partial reduction of Cr(VI) to Cr(III), followed by complexation with adsorbent surface groups. Hydroponic experiments with *Brassica chinensis* L. indicate both termite nests significantly mitigate the impact of Cr(VI) on plant growth and Cr content. The regeneration experiments reveal that both *C. formosanus* and *O. formosanus* nests maintain robust adsorption capabilities even after five cycles. This finding highlights termite nests significant reusability as adsorbents.

In summary, this research demonstrates that termite nests offer a practical, affordable, and reusable solution for removing Cr(VI) from wastewater. Their use as biological adsorption material leverages biomass waste and diminishes the need for pesticides in termite control. Future studies should delve into the optimization of termite nest treatment processes, exploring the potential of chemical or physical modification techniques to enhance adsorption efficiency. Investigating the interaction mechanisms between termite nests and various pollutants could also provide insights into broadening the scope of their application in environmental remediation. Additionally, the study's implications extend beyond mitigating chromium pollution, offering benefits in environmental protection, resource utilization, cost savings, and pesticide reduction. A deeper analysis of the economic and environmental impacts of large-scale implementation could further substantiate the practicality of this approach. Moreover, examining the regeneration and reuse cycles of termite nest-based adsorbents will be crucial

in assessing their long-term viability and operational costs. These findings significantly bolster efforts toward sustainable development and environmental conservation, paving the way for innovative uses of termite nests in addressing complex environmental challenges.

## Data availability statement

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding author.

## Author contributions

GL: Funding acquisition, Supervision, Writing – review & editing. MW: Writing – original draft, Data curation, Investigation, Methodology. NW: Writing – review & editing. JZ: Writing – review & editing. JYu: Writing – review & editing. LJ: Writing – review & editing. LL: Writing – review & editing. JYa: Writing – review & editing. SG: Writing – review & editing. YZ: Writing – review & editing. RZ: Writing – review & editing. LZ: Writing – review & editing.

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## Funding

The author(s) declare that financial support was received for the research, authorship, and/or publication of this article. This work was supported by the National Natural Science Foundation of China (31601891), the Natural Science Foundation of Hubei Province of China (2023AFB1005), and the Hubei Key Laboratory of Edible Wild Plants Conservation and Utilization (EWPL201615).

## Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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