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Efficient Removal Of U(VI) Ions from Aqueous Solutions by Tannic Acid/Graphene Oxide Composites

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Abstract: Tannic acid/graphene oxide (TA/GO) composites were prepared in the present research, and their properties and sorption performance were evaluated by corresponding characterization methods and bath sorption experiments, respectively. The applications of TA/GO to remove U(VI) from aqueous solution were investigated with the maximum adsorption capacity of 87.8 mg·g⁻¹ at low pH (pH = 3.6 ± 0.03). The sorption of U(VI) ions on TA/GO followed the Langmuir model because of the complexation of oxygen-containing functional groups on the surface of TA/GO composites and uranium ions. TA/GO manifested excellent selective adsorption toward uranium ions with other metal ions (Cs⁺, Sr²⁺, Co²⁺). Furthermore, TA/GO as an effective adsorbent was reused to remove a large amount of U(VI) ions from aqueous solution. Therefore, TA/GO is an ideal material to remove highly toxic U(VI) ions from wastewater.

Keywords: tannic acid; graphene oxide; uranium; adsorption

1. Introduction

With the rapid development of modern industries, the generation of pollutants (organic pollutants, heavy metals, and radionuclides) is posing major threats to environmental ecosystems and human health [1–3]. To solve the problem of the global energy crisis, nuclear energy is widely adopted as a new energy source [4]; however, it also causes a certain side effect, nuclear wastes. Radioactive nuclides in nuclear wastes, such as ⁶⁰Co, ¹⁵⁴Eu, ²³²Th, ²³⁵U, ²³⁵Np, ²³⁹Pu, ²⁴¹Am, and ²⁴⁷Cm, directly pollute surface water and groundwater resources [5,6]. These radionuclides accumulate through food chains and cause radiation and biochemical damages to human organs due to their long-term radioactivity [7]. Hence, the safe disposal of nuclear wastes is a public concern [8]. Numerous methods, such as chemical precipitation, ion exchange, membrane separation, and adsorption, have been used to separate and preconcentrate radionuclides from contaminated wastewater [9–14]. In comparison to other methods, the adsorption technology is often used to eliminate radionuclides due to its high efficiency, low cost, and ease of operation [15]. A variety of artificial adsorbents, such as metal hydroxides [16,17], clay minerals [18,19], metal–organic framework material [20,21], nanoparticles [22,23], and carbon nanotubes [18], has been developed to process radionuclides. The influences of different environmental factors (pH, ionic strength, and humus) on the adsorption of radionuclides by artificial adsorbents have been extensively studied [15–18]. However, the adsorption capacity of these adsorbents can only be used under higher pH conditions, thus hindering their practical applications [8]. To preconcentrate and solidify radionuclides from wastewater, efficient and environment-friendly adsorbents are urgently needed [24].

Graphene oxide (GO) has the advantages of large specific surface area, high acid and alkali resistance, and excellent irradiation resistance [4,15,16]. However, the low content of characteristic functional groups on the surface of graphene oxide leads to an adsorption capacity of only 97.5 mg/g at pH = 5.0 [25]. Therefore, the limited adsorption capacity and layer aggregation of GO in aqueous solutions limit its practical application. To improve the above deficiency, some suitable characteristic functional groups are generally grafted on the graphene oxide surface.

Tannic acid has a large number of reactive oxygen-containing functional groups (hydroxyl, carboxyl); thus, it can efficiently capture organic and inorganic pollutants through π - π or electrostatic interactions and hydrogen bonding [25–27]. In comparison to polystyrene or silica, tannic acid has an easy-to-control synthesis scheme and possesses natural high-reactive surface functional groups (catechol) [8].

In the present study, by combining the high specific surface area of graphene oxide and the high-level of active sites of tannic acid, tannic acid/graphene oxide composites as high selectivity and good recyclability adsorbents were prepared to increase the adsorption capacity of graphene oxide on radionuclides from wastewater at low pH, providing a certain basis for the safe treatment and disposal of radioactive wastewater.

2. Materials and Methods

2.1. Synthesis and Purification of Graphene Oxide (GO) and Tannic Acid/Graphene Oxide (TA/GO)

Graphene oxide was prepared according to the modified Hummer's method [28]. First, 120 mg of graphene oxide were added to 80 mL of deionized water and ultrasonically dispersed for 30 min. Subsequently, 32 mg of tannic acid were added to the resultant dispersion and stirred for 30 min. Finally, 5 mL of epichlorohydrin were added for crosslinking, and the solution was continuously stirred for three hours. The synthesized material was washed several times with deionized water and then lyophilized.

2.2. Characterization of Graphene Oxide (GO) and Tannic Acid/Graphene Oxide (TA/GO) Composites

The corresponding characterization of GO and TA/GO were performed by corresponding characterization methods. The images of scanning electron microscope (SEM) was used to observe the surface morphology of the sample. In addition, the characteristic functional groups of all samples were detected by Fourier Infrared spectrometer (FTIR) and X-ray photoelectron spectroscopy (XPS). The spectra of FTIR were obtained with the range from 4000 to 500 cm^{-1} . XPS measurement was performed using the ESCALab220i-XL surface microanalysis system (VG Scientific) equipped with Al K α ($h\nu = 1486.6 \text{ eV}$), and the pressure in the chamber was 3×10^{-9} mbar. The C1s peak at 284.4 eV was used as a reference. The Raman spectroscopy analysis of the sample was performed by using a LabRam HR Raman spectrometer under Ar⁺ laser at 514.5 nm.

2.3. Batch Sorption Experiment

The adsorption of U(VI) on TA/GO composites was performed by batch adsorption experiments at room temperature [15]. In order to achieve sufficient equilibrium of adsorption, the suspension was shaken for 2 days; then, the solid phase was separated from the liquid phase through a 0.22- μm filter membrane. The uranium concentration of filtrate was measured by spectrometry with arsine-azo-III as the chromogenic agent at 652 nm.

3. Results

3.1. Characterization of TA/GO Composites

SEM images were obtained to observe the surface morphology and dimensions of the samples. The folding nature of graphene nanosheets was observed throughout the morphology, and a smooth

layered structure with an average size of about several micrometers was detected (Figure 1a,c). In comparison to graphene oxide, the surface morphology and size of TA/GO were not significantly different (Figure 1b,d), indicating that tannic acid modification did not destroy the basic structure of graphene oxide.

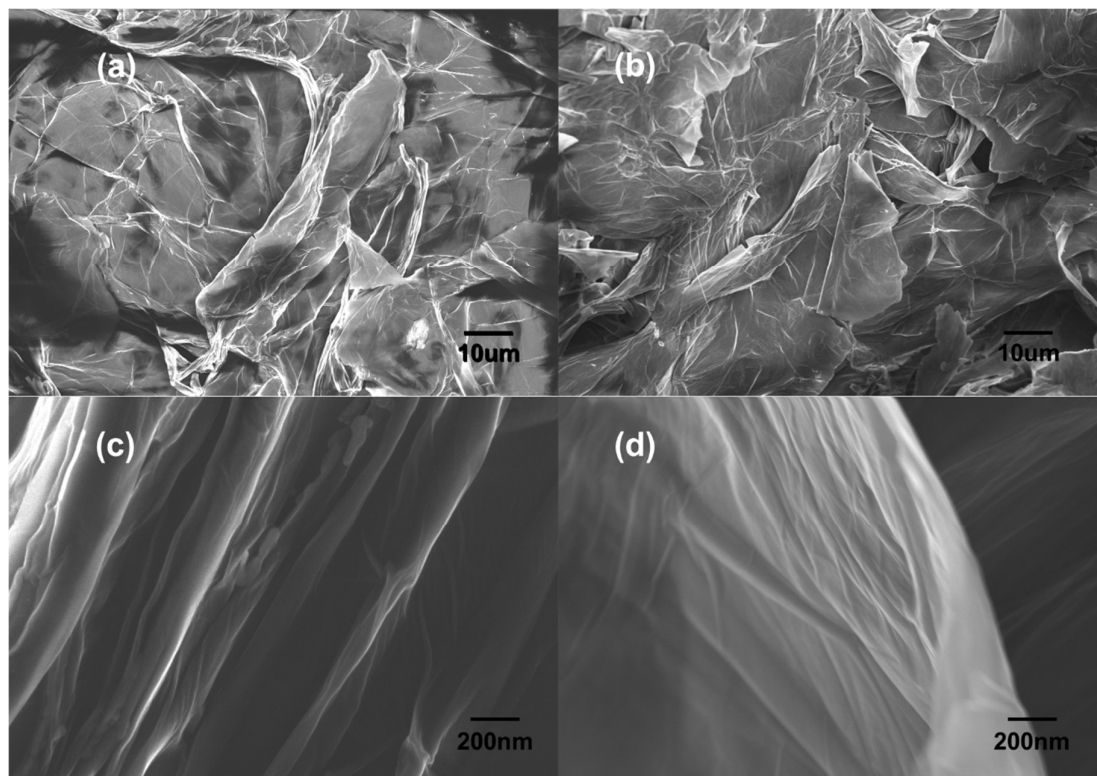


Figure 1. SEM images of GO (a,c) and TA/GO (b,d).

The structural defects and electronic characteristics of GO and TA/GO were detected by Raman spectroscopy (Figure 2). The peaks at 1345 cm^{-1} (D band) and 1588 cm^{-1} (G band) can be assigned to disordered structures (defects and disordered sp^3 carbon atoms and/or other impurities) and the graphite structure (sp^2 -hybridized carbon atoms in graphene sheets), respectively [29]. The peak intensity ratio of D and G bands (I_D/I_G) reflects the relative defect degree of graphitic materials [27]. In comparison to graphene oxide ($I_D/I_G \sim 0.96$), the I_D/I_G ratio of TA/GO (~ 0.89) was reduced by about 0.07, indicating that tannic acid/graphene oxide (TA/GO) composites were successfully prepared. Therefore, SEM images and Raman spectroscopy results reveal that the structural integrity of graphene oxide was not destroyed during the tannic acid modification process.

The characteristics of GO, TA, TA/GO, and TA/GO after uranium adsorption were analyzed by FTIR (Figure 3a). In comparison to graphene oxide, some new peaks were detected on TA/GO at 1717 cm^{-1} , 1610 cm^{-1} , 1442 cm^{-1} , 1318 cm^{-1} , 759 cm^{-1} , and 582 cm^{-1} [26,30]. These new peaks were also detected on TA, indicating that tannic acid was successfully grafted on the surface of graphene oxide. After uranium adsorption, a new characteristic absorption peak was detected on TA/GO at $\sim 920\text{ cm}^{-1}$ due to the presence of UO_2^{2+} ions, indicating that this material successfully adsorbed uranium [15].

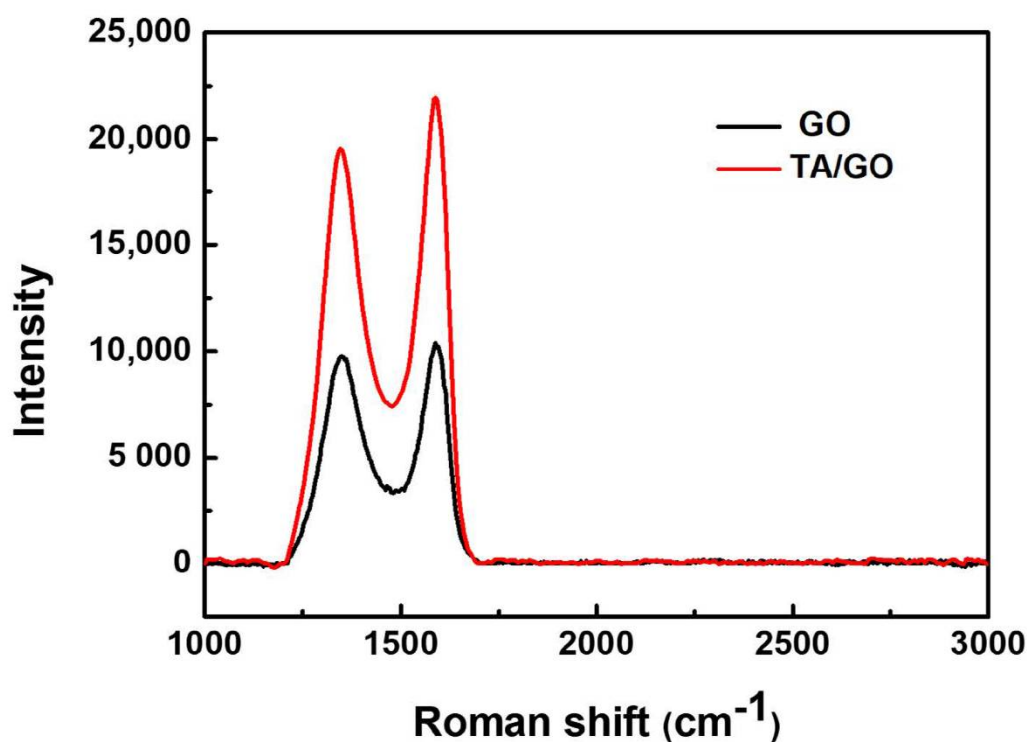


Figure 2. Roman spectra of GO and TA/GO.

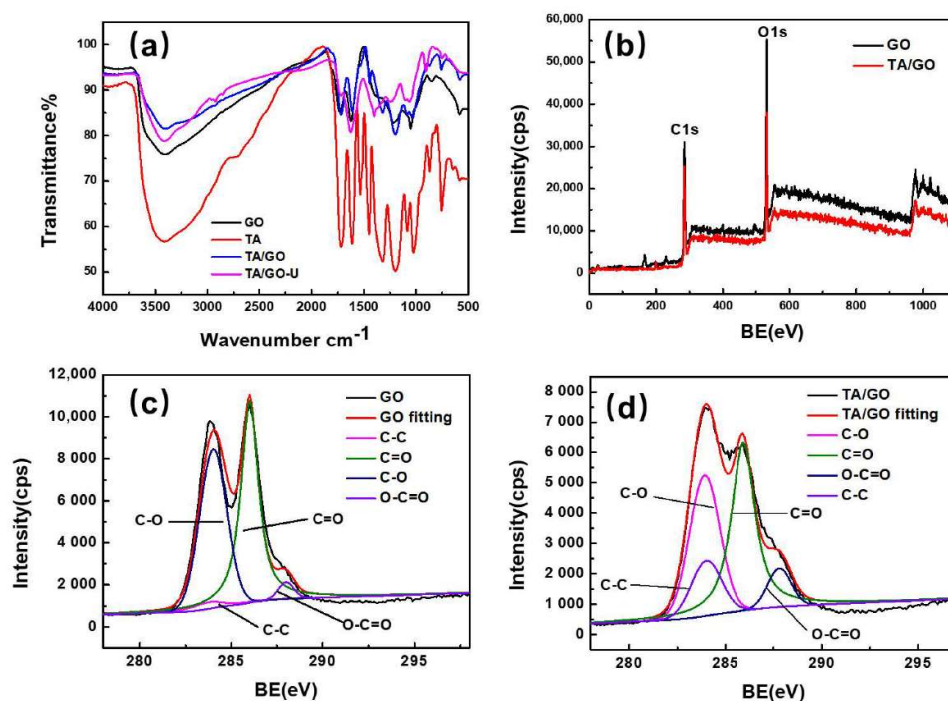


Figure 3. (a) The FTIR of GO, TA, TA/GO, and TA/GO-U(VI); (b) XPS full survey spectra; (c) C1s XPS spectra of GO; (d) C1s XPS spectra of TA/GO composites.

Further, different functional groups on sample surfaces were detected by XPS. The wide-scan spectrum scan results for GO and TA/GO are displayed in Figure 3b. It is noticeable that carbon (C1s at 284.0 eV) and oxygen (O1s at 532.0 eV) peaks appeared in the wide-scan spectra of both GO and TA/GO; it happened because these two materials contained the same functional groups. Figure 3c,d exhibits the high-resolution XPS C1s spectra of GO and TA/GO, respectively. The C1s XPS spectrum of

GO was composed of four parts with the binding energies of 283.7 eV (C–C), 284 eV (C–O), 286 eV (C=O), and 288 eV (O–C=O) [24]. The C 1s XPS spectrum of TA/GO consisted of four parts with the binding energies of 283.91 eV (C–C), 284 eV (C–O), 285.9 eV (C–O), and 287.8 eV (C=O). It is noticeable from Table 1 that the content of O–C=O increased significantly, indicating that TA was successfully grafted on the GO surface.

Table 1. Curve fitting results of XPS C1s spectra of GO and TA/GO composites.

Sample	Peak	Position (eV)	FWHM	%
GO	C–C	283.7	2.06	2.899753
	C–O	284	1.749	43.48051
	C=O	286	1.167	51.02671
	O–C=O	288	1	2.593018
TA/GO	C–C	283.91	1.862	13.47373
	C–O	284	1.854	34.69049
	C=O	285.922	1.449	44.46788
	O–C=O	287.8	1.429	7.367895

3.2. Sorption Kinetics

Adsorption kinetics reflects the rate and efficiency of adsorption of uranium ions by the adsorbent [15,31,32]. The uranium adsorption kinetics of TA/GO from an aqueous solution as a function of contact time at pH = 3.6 ± 0.05 is displayed in Figure 4. It can be seen from Figure 4 that the percentage of adsorption increases rapidly with time during the first 120 min and then remains constant. The fast adsorption indicates that the main mechanism of adsorption is strong chemisorption or surface complexation [15]. To analyze the adsorption kinetics in detail, the obtained experimental data were fitted by pseudo-first- and pseudo-second-order rate equations [33,34].

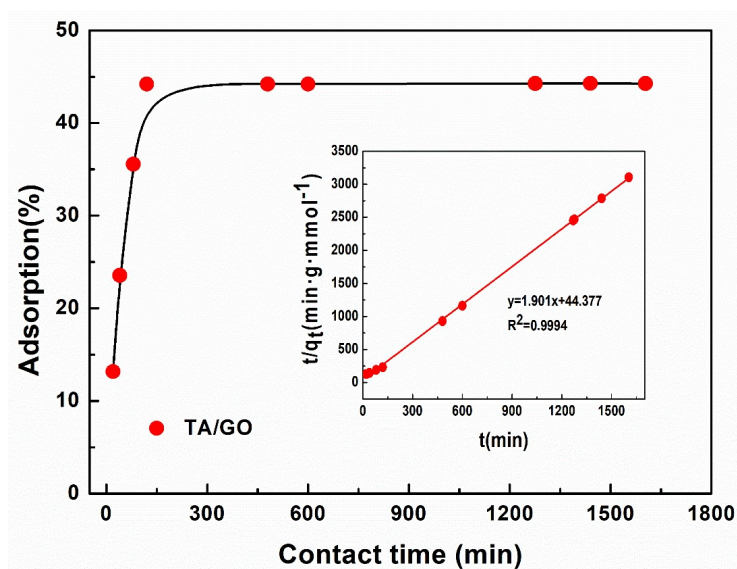


Figure 4. Time-dependent sorption at pH = 3.60 ± 0.05, m/V = 0.6 g/L, and I = 0.001 M NaCl.

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (1)$$

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

where q_t ($\text{mg} \cdot \text{g}^{-1}$) is the adsorption capacity of uranium ions on TA/GO at time t (h), q_e ($\text{mg} \cdot \text{g}^{-1}$) is the adsorption capacity at equilibrium, and k_1 (h^{-1}) and k_2 ($\text{g} \cdot \text{mg}^{-1} \cdot \text{h}^{-1}$) are the pseudo-first-order rate

constant and the pseudo-second-order rate constant, respectively. Table 2 presents the relevant kinetic parameters obtained by fitting. It is clear that the correlation coefficient (R^2) of the pseudo-secondary-level model was higher than that of the pseudo-first-level model, indicating that the pseudo-secondary model could better describe adsorption kinetics.

Table 2. Parameters for adsorption kinetics.

Parameters	Pseudo-First-Order	Pseudo-Second-Order
q_e (mmol g ⁻¹)	0.0430	0.526
k_1 (h ⁻¹)	0.0022	
k_2 (g mmol ⁻¹ h ⁻¹)		0.0814
R^2	0.7144	0.9994

3.3. Effect of Solid Content

Solid content is an important factor for uranium adsorption. The use of a small amount of adsorbent can achieve a considerable adsorption percentage, resulting in a low adsorption cost [15]. The effects of different solid contents of GO and TA/GO are displayed in Figure 5. In comparison to GO, the adsorption percentage of TA/GO was significantly higher at the same solid content ($m/V < 2.0$ g/L), indicating the improved uranium adsorption capacity of TA/GO composites because of its higher number of oxygen-containing functional groups. When the solid content of TA/GO increased from 0.2 g·L⁻¹ to 2.5 g·L⁻¹, the adsorption percentage of uranium increased from ~20% to ~96%. It can be ascribed to the increase of the content of available functional groups in TA/GO; therefore, the available adsorption sites for uranium also increased.

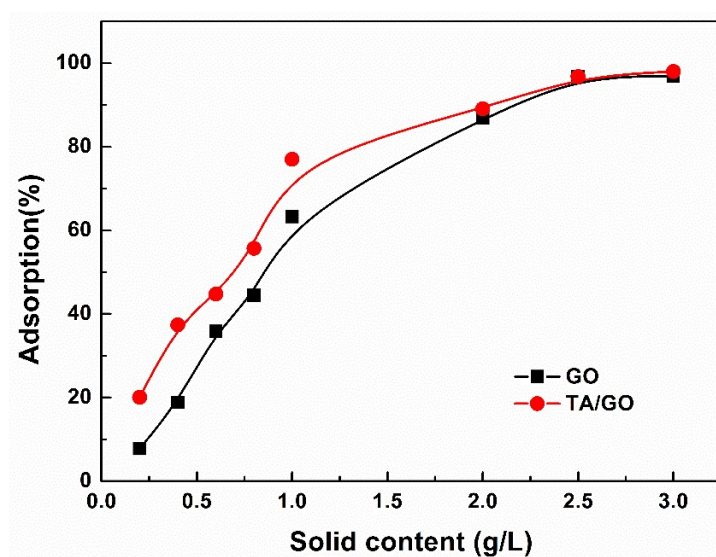


Figure 5. Effects of different solid contents at $pH = 3.6 \pm 0.05$ and $I = 0.001$ M NaCl.

3.4. Effects of pH and Ionic Strength

The distribution of uranium ions and the protonation degree of functional groups on the adsorbent strongly depend on the pH value of the solution [35–37]. Figure 6 presents the adsorption behavior of uranium ions on TA/GO as a function of pH in 0.001 M and 0.1 M NaCl solutions. When the pH value was less than 6.0, free UO_2^{2+} ions were the main form of uranium. Because of the interaction of adsorption sites between H^+/Na^+ and UO_2^{2+} ions, the adsorption of uranium ions by the adsorbent was inhibited at $pH < 6.0$ [15,27]. With the increase of pH, the concentration of H^+ ions in the solution gradually decreased and more surface functional groups became deprotonated [15]; thus, the adsorbent

surface carried a large amount of negative charges. Therefore, when the pH value increased from 2.0 to 6.0, the adsorption percentage of U(VI) ions gradually increased.

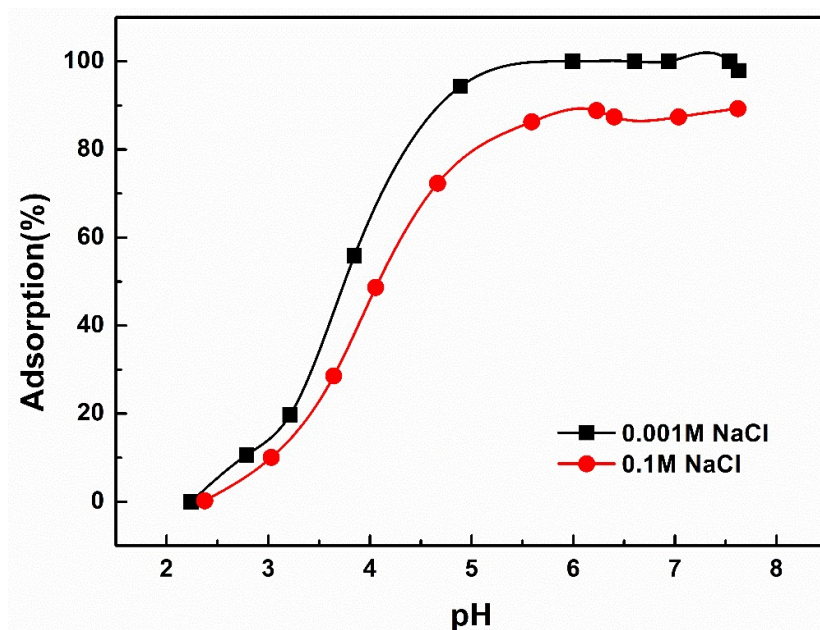


Figure 6. Effects of ionic strength on U(VI) ions adsorption at $m/V = 0.6$ g/L and $pH = 3.6 \pm 0.05$.

Figure 6 expresses that the adsorption of U(VI) ions by TA/GO strongly depended on the ionic strength. The concentration of Na^+ ions affected the thickness and interface energy of the electron double layer and also bound species [15]. It is reported that, as compared to outer spherical surface complexation and cation exchange, inner spherical surface complexation is insensitive to ionic strength [38]. It was found that the adsorption percentage of U(VI) ions decreases with increasing ionic strength, which may be due to the following reasons [15]: (1) the increase of NaCl concentration decreases the activity coefficient of uranium ions, thus limiting the transfer of uranium ions from the solution to the adsorbent surface and (2) the formation of electron double-sided complexes between uranium ions and TA/GO was beneficial for the adsorption of uranium ions under low NaCl concentrations; thus, ion exchange occurred between uranium ions and TA/GO complex functional groups. Therefore, the adsorption of U(VI) ions on TA/GO composites occurred due to ion exchange and complexation with the outer spherical surface of oxygen-containing functional groups on the TA/GO composite surface.

3.5. Adsorption Isotherms

The adsorption isotherms of U(VI) ions on TA/GO are displayed in Figure 7, and the adsorption capacity was calculated from these isotherms. The Langmuir and Freundlich models were used to fit the experimental data to better understand the adsorption mechanism of U(VI) ion on TA/GO [39,40]. The Langmuir isotherm model was used to describe the monolayer adsorption, and its fitting equation can be expressed as [39]:

$$C_s = \frac{bC_{s\max}C_e}{1 + bC_e} \quad (3)$$

The above equation can be further transformed into the following linear form:

$$\frac{C_e}{C_s} = \frac{1}{bC_{s\max}} + \frac{C_e}{C_{s\max}} \quad (4)$$

where C_s ($\text{mol} \cdot \text{g}^{-1}$) and $C_{s\max}$ ($\text{mol} \cdot \text{g}^{-1}$) are the amount of uranium ions adsorbed by the adsorbent and the maximum amount of uranium ions adsorbed per unit weight of the adsorbent, respectively,

and b (L mg^{-1}) is the Langmuir adsorption coefficient representing the enthalpy of adsorption. The Freundlich isotherm model is an empirical equation based on the adsorption behavior on a heterogeneous surface, and it can be expressed as [40]:

$$C_s = K_F C_e^n \quad (5)$$

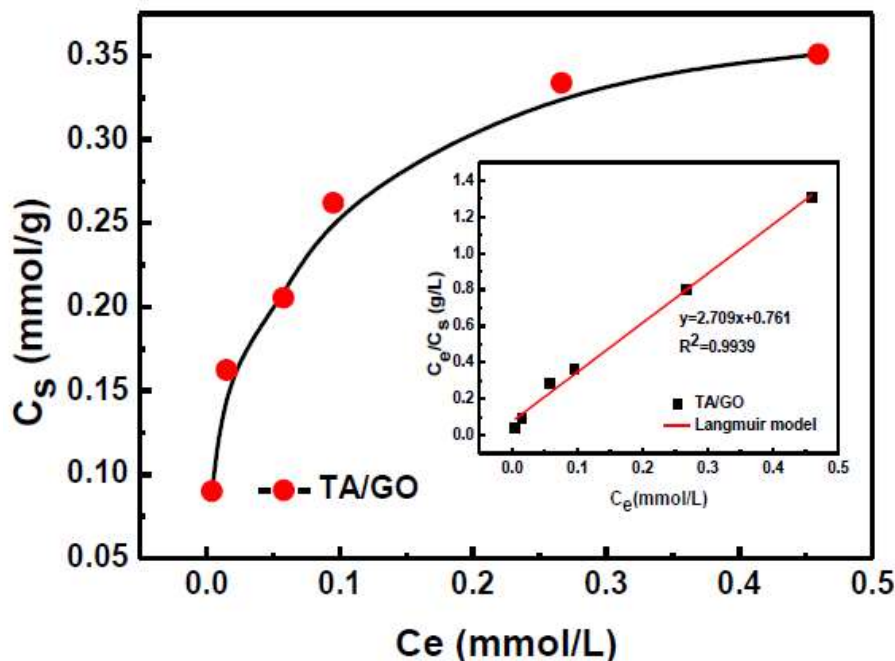


Figure 7. Sorption isotherms of U(VI) ions on TA/GO at $\text{pH} = 3.6 \pm 0.03$, $T = 298 \text{ K}$, $m/V = 0.6 \text{ g/L}$, $I = 0.001 \text{ M NaCl}$.

The above equation can be further transformed into the following linear form:

$$\log C_s = \log K_F + \frac{1}{n} \log C_e \quad (6)$$

where K_F ($\text{mol}^{1-n} \cdot \text{L}^n/\text{g}$) and n are the adsorption strength at a specific temperature and the Freundlich constant of the adsorption capacity when the equilibrium concentration of metal ions is equal to 1, respectively. The linear fitting results for the Langmuir models of U(VI) ions on TA/GO composites are presented in Figure 7, and the relevant parameters are listed in Table 3. It is noticeable from Table 3 that the correlation coefficient (R^2) of the adsorption isotherm fitted by the Langmuir model was larger than that fitted by the Freundlich model, which indicates that the Langmuir model could better fit the isotherm; hence, the adsorption of uranium ions by TA/GO occurred in a single-layer coverage mode [4]. According to the fitting curve of the Langmuir model, the maximum adsorption capacity ($C_{s, \max}$) of TA/GO was obtained as 87.8 mg/g at $\text{pH} = 3.6 \pm 0.03$ and $T = 298 \text{ K}$. The adsorption capacities of uranium ions on other adsorbents are shown in Table 4. It can be seen from the table that the adsorption capacity of TA/GO is 266.56 mg/g when the experimental conditions are $\text{pH} = 5.0$ and $T = 298 \text{ K}$. Compared with other adsorbents, the adsorption capacity of TA/GO is the largest. Oxygen-containing functional groups on the TA/GO surface reduced the aggregation of TA/GO particles through van der Waals forces, resulting in a good dispersion performance [27]. In addition, in comparison to GO (BET specific surface area = $131.9678 \text{ m}^2/\text{g}$), TA/GO has a larger specific surface area ($150.9678 \text{ m}^2/\text{g}$). The above results indicate that TA/GO is a suitable material to remove uranium ions from aqueous solutions.

Table 3. Parameters of the Langmuir and Freundlich models for U(VI) ion sorption on TA/GO composites.

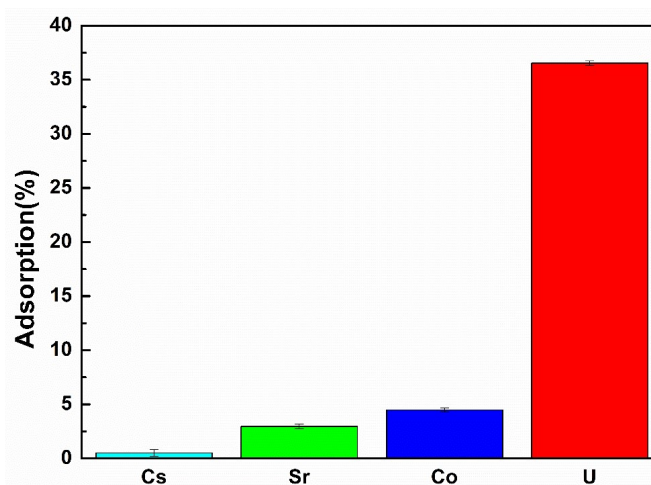
Sorbent	Langmuir Model			Freundlich Model		
	b (L mg ⁻¹)	C _{s, max} (mg g ⁻¹)	R ²	K _F (mg ¹⁻ⁿ L ⁿ g ⁻¹)	1/n	R ²
TA/GO	0.1497	87.8	0.9939	3.3620	0.2833	0.9655

Table 4. Comparison of U(VI) sorption capacity of TA/GO composites with other sorbents.

Sorbents	Experimental Conditions	C _{s, max} (mg g ⁻¹)	Ref.
Activated Carbon	pH = 3.0 T = 293 K	28.3	[36]
PPY	pH = 5.0 T = 298 K	28.6	[15]
GO nanosheet	pH = 5.0 T = 293 K	97.5	[41]
PPY/GO	pH = 5.0 T = 298 K	147.06	[15]
PANI	pH = 3.0 T = 298 K	31.42	[16]
TA/GO	pH = 3.6 T = 298 K	87.8	This study
TA/GO	pH = 5.0 T = 298 K	266.56	This study

3.6. Evaluation of Selectivity

The selectivity of TA/GO composites for uranium ion adsorption was determined in the presence of UO₂²⁺, Co²⁺, Sr²⁺, and Cs⁺ ions. It is noticeable from Figure 8 that the uranium ion adsorption percentage of TA/GO was about 38%, which was much higher than those for Cs (1%), Sr (3.4%), and Co (4%); therefore, TA/GO was highly selective toward uranium ions, and it can be attributed to the difference in the ion radius and charge of these ions [27].

**Figure 8.** Selective sorption capacities of coexistent ions on TA/GO composites at C_{initial} = 1.0 × 10⁻⁴ M.

3.7. Regeneration Experiment

The stability of the resultant TA/GO composites was evaluated by multiple adsorption/desorption cycles. It is evident from Figure 9 that the U(VI) adsorption capacity of TA/GO did not change greatly. After the cycle test, TA/GO composites were separated and dried, and the weight was calculated as 115 mg (a loss of about 4%). It indicates that TA/GO can be reused as an effective adsorbent to remove a large amount of U(VI) ions from aqueous solutions.

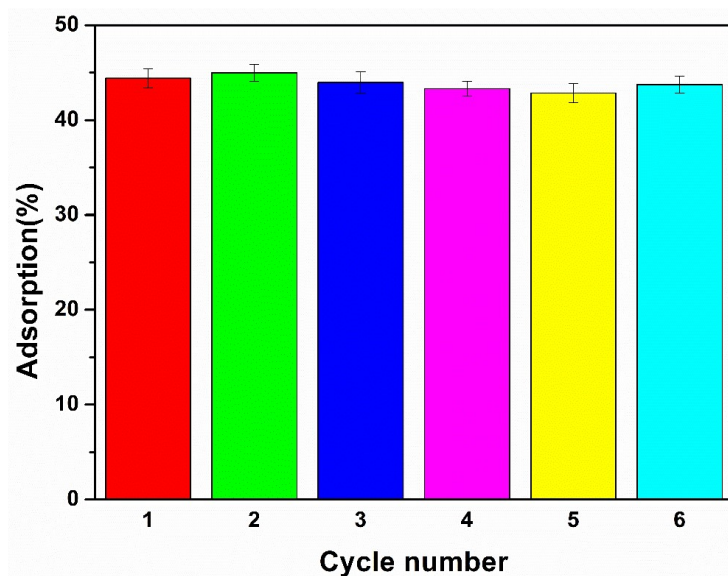


Figure 9. Recycling of TA/GO for the removal of U(VI) ions ($C_{\text{initial}} = 7.0 \times 10^{-4}$ M, $\text{pH} = 3.6 \pm 0.05$, $m/V = 0.6$ g/L, $T = 298$ K).

4. Conclusions

TA/GO composites were successfully synthesized, and their adsorption behavior toward U(VI) ions was studied by batch sorption experiments. The resultant TA/GO composites had excellent adsorption capacity in concentrating or removing U(VI) ions from aqueous solutions at low pH. The adsorption of U(VI) ions on TA/GO was controlled by contact time, pH, ionic strength, and the solid–liquid ratio. In addition, the adsorption of U(VI) ions on TA/GO followed pseudo-second-order kinetics and the Langmuir model. Moreover, TA/GO composites have outstanding selectivity for uranium ions in aqueous solutions containing other ions (Cs^+ , Sr^{2+} , Co^{2+}) and good recyclability for uranium adsorption. Therefore, TA/GO has high selectivity, excellent adsorption, and good recyclability adsorbents is an ideal material to remove highly toxic U(VI) ions from wastewater.

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