Graphene Oxide-based Nanomaterials for Uranium Adsorptive Uptake

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Abstract

The growth of nuclear power generation and the necessity to acquire uranium reserves for energy security and pollution regulation for environmental protection put much emphasis on the removal and recovery of uranium from aqueous solutions. Adsorption has been proved to be a promising method for this purpose method because of its high adsorption efficiency, easy operation, low cost, reusability and availability of massive adsorbents. Among a wide variety of adsorbents, graphene oxide (GO) has demonstrated excellent adsorption potential for uranium uptake and recovery due to its unique 2D structure, high specific surface area and abundant oxygen-containing functional groups. Regarding the functional groups, it can make GO with high dispersion and hydrophilicity and participate in the complexation of uranium, leading to high adsorption efficiency for uranium. In this review, the research status and progress of GO-based nanomaterials for uranium adsorption are summarized. Their adsorption capacities, influencing factors, kinetics, isotherms and thermodynamics are compared and discussed. The microscopic mechanisms of uranium adsorption onto these GO-based nanomaterials are elaborated at molecular level by spectral analysis, surface complexation models, and theoretical calculations. Meanwhile, the challenges and research trends in the study of uranium adsorption by GO-based nanomaterials are pointed out. We believe that our focused review provides not only a summarizing reference on the current status of uranium removal and recovery by GO-based nanomaterials, but also future directions for related follow-up research and practical applications.

Keywords: Uranium; Nanomaterials; Graphene oxide; Adsorption; Removal and recovery.

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1. Introduction

The world's energy demand is increasing continually with the rapid development of economy and the fast-growing population. The extensive use of fossil energy has recently caused serious environmental issues, and traditional fossil fuels are also facing the shortage problems.^[1-3] The global energy shortage and environmental problems have promoted the development of efficient and clean energy.^[4-6] Nuclear power plays a significant role in addressing the energy shortage^[7-10] and mitigating environmental problems such as the climate change.^[11,12] Uranium, as a critical radioactive element, has been widely used to produce nuclear power, which is increasing the necessity for uranium as an energy resource.^[13-16] However, uranium and its radioactive isotopes contribute to a high-level of nuclear waste, which needs to be

properly disposed. Therefore, we carried out speciation (*e.g.* oxidation state and coordination geometry) study of uranium ion doped in $\text{RE}_2\text{Hf}_2\text{O}_7$ serves as a prerequisite for their capability for nuclear waste immobilization and eventually safe nuclear energy and sustainable environment.^[15,16] Moreover, the terrestrial deposits of uranium are predicted to become a shortage owing to the booming energy demand. Seawater also contains uranium at low concentrations (~ 3.3 ppb).^[17] Although the uranium concentrations are extremely low, seawater represents an inexhaustible source of uranium which is estimated at approximately 4.5 billion tons—nearly 1,000 times of the entire available terrestrial deposits of uranium. Development of highly efficient and selective adsorbents capable of recovering uranium from seawater would be valuable to ensuring the availability of uranium resources.

On the other hand, the vigorous exploitation of nuclear power has also posed potential harms to ecological environment and human health. Due to uranium mining and processing activities, nuclear accidents and improper nuclear waste treatment, a large amounts of uranium species have

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Table 1. Adsorption capacities and main parameters of uranium adsorption by GO-based nanomaterials

Adsorbents	pH	Ambient temperature (K)	Adsorption capacity (mg g ⁻¹)	Isotherm model	Thermodynamics	Ref.
GO	3.45	298	94.25	Langmuir Isotherm	endothermic /spontaneous	[76]
GO	6.8	293	99	Langmuir Isotherm	endothermic	[77]
GO	4.4	298	330.82	Langmuir Isotherm	endothermic /spontaneous	[78]
GO	6.0	298.15	309.09	Langmuir Isotherm	endothermic /spontaneous	[79]
GO	5.0	293	97.5	n.a.	endothermic /spontaneous	[80]
GO	4.0	303	208.33	Langmuir Isotherm	endothermic /spontaneous	[81]
GO	4.0	RT	299	Langmuir Isotherm	endothermic /spontaneous	[82]
rGO	4.0	RT	47	n.a.	n.a.	[82]
CGO	4.5	298	240.1	Langmuir Isotherm	n.a.	[70]
EGO	4.5	298	75.2	Langmuir Isotherm	n.a.	[70]
BGO	4.5	298	68.8	Langmuir Isotherm	n.a.	[70]
OGO	5.5	303	291.8	n.a.	endothermic /spontaneous	[71]
GOC	5.0	303	101.0	Freundlich Isotherm	n.a.	[46]
GO-NH ₂	5.5	298.15	215.2	Langmuir Isotherm	endothermic /spontaneous	[83]
GONRs	4.5	298	394.1	Langmuir Isotherm	endothermic /spontaneous	[84]
GONRs/CTS	5	298	320	Freundlich Isotherm	endothermic /spontaneous	[47]
GO-CNTs	5.0	298.15	86.1	Langmuir Isotherm	endothermic /spontaneous	[85]
GO-ACF	5.5	298	298	Langmuir Isotherm	endothermic /spontaneous	[86]
MIL-68/GO	8.0	298	594.66	Langmuir Isotherm	endothermic	[87]
rGO/ZIF-67	4.01	298	1888.55	Langmuir Isotherm	endothermic /spontaneous	[10]
GZA	7.0	298	602.41	Freundlich/ Langmuir	endothermic /spontaneous	[88]
GCZ8A	8.0	298.15	361.01	Freundlich Isotherm	n.a.	[43]
CMC/MGOs	5.5	301	188.97	Langmuir Isotherm	endothermic	[89]
AOMGO	5.0	298	284.9	Langmuir Isotherm	endothermic /spontaneous	[90]
PAO/rGO	4.0	293	872	Langmuir Isotherm	endothermic /spontaneous	[91]

continued

Adsorbents	pH	Ambient temperature (K)	Adsorption capacity (mg g ⁻¹)	Isotherm model	Thermodynamics	Ref.
PA-GO	5.5	298	124.3	Langmuir Isotherm	n.a.	[92]
HO-CB[6]/GO	5.0	298	301.6	Langmuir Isotherm	endothermic /spontaneous	[93]
CD/GO	5.0	288	97.3	Langmuir Isotherm	n.a.	[93]
PANI/GO	5.0	293	1960	n.a.	n.a.	[94]
PAM/GO	5.0	295	166.12	Langmuir Isotherm	endothermic /spontaneous	[95]
GO-PDA	5.5	298	718	Langmuir Isotherm	endothermic /spontaneous	[96]
GO/PDA/PAO-0.2	6	298	502.5	Langmuir Isotherm	endothermic /spontaneous	[73]
GO-pDA-PEI	5.0	298	530.6	Langmuir Isotherm	exothermic /spontaneous	[97]
PEI-rGO/MoS ₂	8.0	298	572.05	Langmuir Isotherm	endothermic /spontaneous	[98]
CAs-3 PEI/GO	7	303	145.14	Langmuir Isotherm	n.a.	[49]
MBTA-GO	3.5	298	264	Langmuir Isotherm	n.a.	[99]
GO/PEDOT: PSS	4.5	298	384.51	Langmuir Isotherm	endothermic	[100]
GO-DM-AO	8.02	298	935	Langmuir Isotherm	endothermic /spontaneous	[75]
GO-CS-AO	6.0	308	248.75	Langmuir Isotherm	endothermic	[101]
GO-CMC-AO	6.0	308	327.86	Langmuir Isotherm	endothermic	[101]
MCGO	5.0	313	204.1	Langmuir Isotherm	endothermic /spontaneous	[102]
FH/GOA	5.0	293	288.64	Langmuir Isotherm	endothermic /spontaneous	[45]
Lysinibacillus-GO	4.5	303	149.3	Langmuir Isotherm	n.a.	[103]
GO-TW	5	298	111.61	Langmuir Isotherm	endothermic /spontaneous	[104]
rGO/Fe ₃ O ₄ /TW	5	298	103.84	Langmuir Isotherm	endothermic /spontaneous	[104]
Fe ₃ O ₄ /GO	5.0	298	28.32	Langmuir Isotherm	endothermic /spontaneous	[105]
Fe ₃ O ₄ /GO	5.5	293	69.49	n.a.	endothermic	[106]
CB[6]/GO/Fe ₃ O ₄	5.0	298	66.8	n.a.	n.a.	[107]
MnO ₂ -Fe ₃ O ₄ -rGO	6.0	328	108.7	Langmuir Isotherm	endothermic /spontaneous	[108]

continued

Adsorbents	pH	Ambient temperature (K)	Adsorption capacity (mg g ⁻¹)	Isotherm model	Thermodynamics	Ref.
CoFe ₂ O ₄ -rGO	6.0	298	227.2	Langmuir Isotherm	endothermic /spontaneous	[109]
Fe-Ni/GO	5.5	298	384.62	Langmuir Isotherm	endothermic /spontaneous	[110]
Ag-L-Arg-rGH	6.0	298	434.78	Langmuir Isotherm	endothermic /spontaneous	[111]
GO/bentonite	7	303	234.19	Freundlich Isotherm	endothermic /spontaneous	[50]
GO@sepiolite	5.0	298	161.29	Langmuir Isotherm	endothermic /spontaneous	[112]

*RT: Room Temperature. n.a.: not application.

released into the water system. Uranium in aqueous solutions mainly exists as soluble uranyl ions which are highly toxic and easily migrate.^[18-20] The exposure to excessive uranium can result in DNA damage and dangerous health issues such as liver disease and lung cancer.^[21-23] Thus, it is greatly important to remove and recycle uranium from aqueous solutions for environmental conservation, human health, and sustainable development of nuclear energy.^[24, 25]

In the last few decades, various treatment techniques including biological treatments,^[26] membrane treatment,^[27,28] adsorption,^[29-32] advance oxidation processes,^[33] chemical precipitation processes,^[34] ion exchange,^[35-37] and electrochemical method^[38-40] have been developed to uptake uranium from aqueous solutions. However, most of these methods have various drawbacks. For example, although chemical precipitation is comparably simple and cost-effective, they have difficulty reducing metal ion concentrations below legally limited levels and often produce large amounts of sludge. Among these methods, adsorption is a promising and effective method because of its high adsorption efficiency, easy operation, low cost, reusability and availability of massive adsorbents.^[41,42] Recent advances of nanomaterials have provided ample opportunities to develop and employ more efficient radioactive wastewater treatment technologies. Graphene oxide (GO)-based nanomaterials have attracted significant interests because of their unique physicochemical properties such as high specific surface area, high chemical stability and abundant oxygen-containing functional groups.^[43,44] These properties make GO-based nanomaterials to be promising candidates for adsorption uranium. For example, Li *et al.* assembled fungal hypha/GO aerogel as high-performance adsorbents for U(VI) removal.^[45] This fungal hypha/GO aerogel exhibited an excellent capability on U(VI) removal due to its large specific surface area (894 m²/g) and abundant functional groups. Yang *et al.* found that GO-cellulose (GOC) composites were promising adsorbents for uranium wastewater treatment.^[46] Hu *et al.* also found that the GO nanoribbons/chitosan (GONRs/CTS) composite membrane was suitable for separating and

recovering uranium from wastewater.^[47]

To date, the adsorption of uranium by GO-based nanomaterials has been extensively studied.^[48] This relevant research work has also been carried out in our laboratory.^[49-51] However, there have been few reviews that have specially focused on GO-based nanomaterials for uranium adsorption.^[52-67] Thereby, a needful review on the recent progress of GO-based nanomaterials for uranium adsorption is expected to promote the development of GO-based nanomaterials in the removal and recovery applications of uranium from aqueous solution. In this review, the adsorption capacities of GO-based nanomaterials are introduced. The influencing factors, kinetics, isotherms and thermodynamics of GO-based nanomaterials for uranium adsorption are analyzed. Moreover, the interaction mechanisms between uranium ions and the GO-based nanomaterials are expounded. Meanwhile, the challenges and research trends in studying the uranium adsorption by GO-based nanomaterials are pointed out. We believe that this focused review for the emerging research on uranium adsorption by GO-based nanomaterials is important for the future uranium removal and recovery technology development. This will also provide an important reference for relevant follow-up research and practical applications.

2. Adsorption capacities of GO-based nanomaterials

GO has a wide variety of functional groups on their surface and edges, such as hydroxyl group, epoxy group, carboxyl, carbonyl, *etc.*^[14,47] Due to these oxygen-containing functional groups, GO possesses features such as high dispersion, hydrophilicity, and compatibility, making it a suitable support material to combine with other chemical functional groups or composites as demonstrated in our previous studies among many others.^[68,69] Meanwhile, the functional groups of GO can provide active adsorption sites for environmental pollutants, such as uranium and other radioactive species in wastewater. For the past few years, a large number of studies on GO-based nanomaterials as adsorbents of uranium have been carried out. The GO-based nanomaterials for uranium adsorption along

their capacities are summarized in Table 1.

The adsorption capacities of the GO-based nanomaterials for uranium strongly depend on the functional groups on GO and experimental conditions. According to the results in Table 1, pristine GOs have different adsorption capacities due to its distinct synthesis procedures and factors affecting adsorption. Regarding synthesis procedures, different oxidation processes of the Hummers method (the major method to synthesize GO) can affect the type and distribution of functional group on GO, leading to different uranium adsorption capacities. The uranium adsorption capacity of GO is commonly higher than reduced GO (rGO), which is because there are much more functional groups on GO than rGO. CGO, EGO, and BGO were synthesized from chemical oxidation, electrolysis, and ball-milling of graphene sheets by the modified Hummers method. The adsorption capacity of the CGO (240.1 mg/g) was three times higher than those of the EGO (75.2 mg/g) and the BGO (68.8 mg/g). The CGO with high performance was due to that the amount and type of oxygen functional groups on it was much more abundant than those of the EGO and BGO, which was proved by SEM, FT-IR, Raman, XRD and XPS measurements.^[70] OGO was generated from GO oxidized by ozone. The oxidization of ozone made the OGO with high adsorption performance due to higher oxygen-containing functional groups.^[71] These results indicated that the functional groups on GO played an important role in the removal and recovery of uranium.^[72] In addition, the adsorption capacities of modified GOs can generally be enhanced by various modification methods, which could be due to improve the dispersion and stability as well as the introduction of more surface-active sites and functional groups. For example, through the assembly of zeolitic imidazolate framework-67 (ZIF-67) on rGO hydrogel, the rGO/ZIF-67 aerogel exhibited an excellent uranium adsorption capacity of 1888.55 mg/g, which was much higher than that of the initial rGO (134.23 mg/g).^[10] The adsorption capacity of GO/PDA/PAO-0.2 was 502.5 mg/g, which was higher than that of the initial GO (342.5 mg/g). The XPS results revealed that U(VI) coordinated with amidoxime and imine/amine groups, indicating that the U(VI) was adsorbed on PAO and PDA groups.^[73] Graphene oxide /hydroxyapatite (GO/HAP) was synthesized by directly assembling GO and HAP through a facile hydrothermal method. The GO/HAP composite had an outstanding adsorption capacity for U(VI) (373 mg/g) which was higher than that of GO (85 mg/g). XRD, FT-IR and XPS results indicated that the phosphate group from HAP was responsible for U(VI) retention and incorporation. The GO/HAP composite's enhanced U(VI) adsorption capacity was most likely due to the synergistic effect after functionalizing with nanostructured HAP.^[74] Similarly, through post-decoration with amidoxime functionalized diaminomaleonitrile (DM-AO) onto GO, the U(VI) adsorption capacity of the GO-DM-AO reached 935 mg/g, which was increased by 209% compared with that of the pristine GO. The increased adsorption capability of the GO-DM-AO was due to

the introduction of organic groups on the GO to increase the number of binding spots and the activation of the inactive points on the GO by cycloaddition reaction.^[75]

In conclusion, the adsorption capacities of the GO-based nanomaterials for uranium strongly depend on the functional groups on GO and experimental conditions. Pristine GOs have different adsorption capacities affected by their distinct synthesis procedures. The adsorption capacities of modified GOs are generally enhanced by their specific modification methods.

3. Affecting factors, kinetics, isotherm and thermodynamic

Generally, the performance of GO-based nanomaterials for uranium adsorption are affected by many factors, such as the solution pH value, temperature, contact time, ionic strength, co-existing ions, *etc.* The batch experiments of the effect of affecting factors on the uranium adsorption has been extensively carried out for the application to the treatment of uranium pollution. To some extent, the interaction mechanism can also be expounded based on the results of the effect of affecting factors.^[57] For example, the kinetic parameters from time-dependent adsorption experiments, the isotherms and thermodynamic parameters from the fittings of the temperature-dependent adsorption experiments can provide further information about the interactions between the adsorbents and adsorbates. The effect of adsorption affecting factors on GO-based nanomaterials for uranium is discussed below:

3.1. Effect of solution pH value

Solution pH value is one of the most important factors affecting uranium adsorption by GO-based materials. The pH value of uranium-containing solutions can affect uranium species in the solution and the surface charge of GO-based nanomaterials, thus affecting the interaction between such GO-based nanomaterials and uranium species, which leads to different adsorption efficiencies at various pH values. For example, Zhao *et al.*^[90] found that the adsorption efficiency of U(VI) by amineoxime magnetic GO composites (AOMGO) gradually increased from about 50% to 99% with increasing the pH value from 3 to 7, and began to decline when the pH value was higher than 8. Tan *et al.*^[108] studied the influence of pH value on the adsorption of uranium by MnO₂-Fe₃O₄-rGO. The adsorption amount of uranium by the MnO₂-Fe₃O₄-rGO first increased and then decreased within the range of pH value of 2–12. Dai *et al.*^[73] found the adsorption capacity of GO/PDA/PAO increased rapidly at pH from 3 to 6 and then decreased with the increase of pH value from 6 to 8 due to the change of the species distribution of U(VI) and the surface of GO/PDA/PAO with increasing pH value (Fig. 1a, 1c, 1d). Similarly, Zhu *et al.* studied the effect of solution pH value for the adsorption of uranium by Ag-L-Arg-rGH composites in the range of 2–9.^[111] The results showed that the adsorption capacity was increased at pH range from 2 to 6 and the highest adsorption capacity was obtained at pH = 6, and then the

uranium (VI) removal capability reduced with the pH value higher than 6. Other relevant literature also found that the adsorption of GO-based nanomaterials for uranium was strongly dependent on solution pH value.^[82,83]

Generally, the adsorption efficiency of uranium by GO-based nanomaterials is poor at low solution pH value. As the pH value increases, the adsorption efficiency of uranium by GO-based nanomaterials increases, while it begins to decline when the pH value further increases. This trend is mainly because the solution pH value can affect the U(VI) species and the surface properties of GO-based nanomaterials. At low solution pH value, high concentration of H^+ competed with uranyl cations for binding sites on the surface of the adsorbent of GO-based nanomaterials,^[114] thus reducing the adsorption efficiency of uranium(VI). With the increase of solution pH value, the surface of the adsorbent of GO-based nanomaterials becomes negatively charged due to the deprotonation process, at the same time, positive uranyl ionic complexes such as UO_2OH^+ , $(UO_2)_2(OH)_2^{2+}$, and $(UO_2)_3(OH)_5^+$ are formed,^[115,116] and then the electrostatic forces enhance between the positive uranyl complexes and the negatively

charged surface of the GO-based nanomaterials as adsorbents to improve the adsorption efficiency of uranium. Meanwhile, further increase of the solution pH value can induce the hydrolysis of uranium ions, which leads to the formation of anionic complexes such as $UO_2(CO_3)_3^{4-}$, $UO_2(CO_3)_2^{2-}$, $(UO_2)_3(OH)_7^-$, $UO_2(OH)_3^-$, or $UO_2(OH)_2$ precipitation, and eventually to the reduction of the uranium adsorption efficiency.

3.2. Effect of contact time and kinetic studies

The contact time required to reach the adsorption equilibrium is an important factor to reflect the adsorption properties of adsorbents for the uranium. The change of adsorption capacity with the contact time represents the adsorption dynamics. The high specific surface area and abundant functional groups on the surface of GO-based nanomaterials play important roles to improve the adsorption rate of U(VI), and so the U(VI) adsorption of GO-based nanomaterials can reach adsorption equilibrium quickly. For example, the adsorption capacity of GO-DM-AO enhanced with the contact time, and reaches adsorption equilibrium at 30 min.^[75] HO-CB[6]/GO for U(VI)

Fig. 1 (a) Effect of solution pH value on the removal of U(VI) by GO/PDA/PAO ($C_o = 10$ mg/L). (b) Effect of ionic strength on the removal of U(VI) by GO/PDA/PAO-0.2 ($C_o = 1$ mg/L). (c) Distribution of U(VI) species at different pH levels. (d) Zeta potentials of GO/PDA/PAO-0.2 ($m = 5$ mg, $V = 150$ mL, $t = 24$ h, $T = 298$ K). (Reproduced with the permission from [113], Copyright 2019 American Chemical Society)

adsorption was very fast, and it can complete 90% of uranium adsorption within 5 min and reached the adsorption equilibrium within 20 min.^[93] The adsorption of U(VI) onto Fe-Ni/GO was rapid because of many adsorption sites at the initial stage, and the adsorption equilibrium of U(VI) was reached after 80 min.^[110] Li *et al.* found that the adsorption capacity of GO nanosheets had a sharp upsurge with the contact time until reaching an equilibrium within 60 min.^[72]

Generally, adsorption kinetics of GO-based nanomaterials for U(VI) consist of two stages. The initial stage is the external surface adsorption stage, in which U(VI) is absorbed rapidly to contribute significantly to the adsorption equilibrium as GO-based nanomaterials as the adsorbent has a large number of available adsorption sites. During the second stage, the adsorption process is slow and the adsorption rate is generally controlled by intra-particle diffusion until the adsorption reached equilibrium.^[117,118] In order to study the adsorption kinetics of GO-based nanomaterials for U(VI) species, pseudo-first-order kinetic and pseudo-second-order kinetic and intra-particle diffusion models are often used. For example, Zong *et al.* used the three models to analyze the sorption kinetic data of U(VI) ions onto CMC/MGOs. And the kinetics of sorption of U(VI) onto the CMC/ MGOs composites best fit the pseudo-second-order kinetics model

(Fig. 2).^[71] A large number of literature papers reported that the adsorption of GO-based nanomaterials for U(VI) fits well to the pseudo-second-order kinetic model, which indicates that the adsorption process of GO-based nanomaterials for U(VI) is mainly controlled by chemical adsorption.

3.3. Effect of temperature, isotherm and thermodynamic analysis

Temperature is also one of the important environmental factors affecting the adsorption of uranium by GO-based nanomaterials. It has different effects on a series of chemical-physical processes such as adsorption-desorption, precipitation-dissolution and oxidation-reduction of uranium. Therefore, the change of temperature can lead to the variation of adsorption capacity of GO-based nanomaterials for uranium. In order to understand the thermodynamic behaviors of uranium on GO-based nanomaterials, adsorption isotherm models have been extensively used to provide information about the amount of adsorbed uranium ions by certain GO-based nanomaterials as the adsorbents and the interaction between GO-based nanomaterials as the adsorbent and the uranium species as the adsorbate. Langmuir, Freundlich, Dubinin Radushkevich, Brunauer–Emmett–Teller, and Redlich–Peterson adsorption isotherm models have been used

Fig. 2 (A) Influence of contact time on different initial concentration U(VI) ions adsorption onto CMC/MGOs composites. (B) Pseudo first-order model, (C) pseudo second-order model, and (D) intra-particle diffusion model fitting curve. $T = 301$ K, $\text{pH} = 5.5 \pm 0.1$, $m/V = 0.25$ g/L, $I = 0.01$ mol/L NaNO_3 . (Reproduced with the permission form [71], Copyright 2016, Science China Press and Springer-Verlag Berlin Heidelberg)

to describe the adsorption behaviors of GO-based nanomaterials for uranium.^[119] Langmuir and Freundlich models are the most common ones having been used. Langmuir isotherm assumes that all binding sites have equal affinity for an adsorbate, so a monolayer of adsorbed molecules forms with the saturation of the adsorbent. Freundlich isotherm describes adsorption onto heterogeneous surfaces that provide adsorption sites of varying affinities while the adsorbent will not be saturated.^[120] Based on the list in Table 1, it is obvious that the Langmuir isotherm model fits better for uranium adsorption by GO-based nanomaterials, indicating the adsorption procedure could be dominated by a monolayer adsorption.

Meanwhile, the values of the thermodynamic parameters, such as entropy (ΔS_0), enthalpy (ΔH_0), and Gibb's free energy (ΔG_0), are used to express the solution temperature effects.^[121] In general, the temperature has a positive effect on the adsorption capacity of uranium on GO-based nanomaterials as shown in Table 1, in which we can see most of the adsorption process of GO-based nanomaterials for uranium was endothermic and spontaneous. Specifically, Zhang *et al.* found that the U(VI) adsorption on Fe-Ni/GO followed the Langmuir isotherm, and higher removal capacity of Fe-Ni/GO could be acquired at higher temperature.^[122] The adsorption of U(VI) by Fe-Ni/GO was a spontaneous endothermic process proved by the positive ΔH_0 value and the negative ΔG_0 value (Fig. 3). Zhao *et al.* reported that the adsorption capacity of rGO/ZIF-67 aerogel enhanced as the

temperature increased from 293 to 313 K, and the adsorption process was endothermic and spontaneous.^[10] Zhang *et al.* studied the adsorption of uranium by sulfonated GO (GOS) within the temperature range of 288.15–323.15 K.^[123] Their results showed that better adsorption ability of uranium achieved at higher temperature. Similarly, Yang *et al.* found that the increase of temperature from 303 K to 333 K was beneficial to enhance the adsorption efficiency of GO-TW and rGO/Fe₃O₄/TW.^[104]

3.4 Effect of ionic strength and co-existing ions

The ionic strength of the solution containing uranium affects the existing species of uranyl ions. It also affects the dual electrostatic layer thickness of GO-based nanomaterials and changes the number of binding sites of GO-based nanomaterials as the adsorbent, thus affecting their adsorption of uranium.^[124] The adsorption of different GO-based nanomaterials for uranium is affected by ion strength in different ways. The adsorption efficiency of some GO-based nanomaterials for uranium is sensitive to ionic strength change.^[93] While that of some other GO-based nanomaterials for uranium is insensitive. Bai *et al.* used NaClO₄ as an inert electrolyte to study the effect of ionic strength on uranium sorption by PAO/GH.^[14] Their results showed that the ionic strength had very weak influence on the uranium adsorption by the PAO/GH, even when Na⁺ concentration was about 265 times of uranium. Zhang *et al.* found that the effect of ionic strength on uranium adsorption by GO and sulfonated GO was

Fig. 3 Adsorption of U(VI) by Fe-Ni/GO at pH = 5.5 and $m/V = 0.1$ g/L: (A) Adsorption isotherms at 298, 308 and 318 K, (B) representation of the linear Langmuir and (C) Freundlich isotherm models, (D) linear plots of $\ln K_d$ vs C_e for U(VI) adsorption, and (E) Van't Hoff plots for U(VI) adsorption. (Reproduced with the permission from [122], Copyright 2019 Elsevier Ltd)

Fig. 4 Effect of co-existing ions on the removal of uranium by Ag-L-Arg-rGH in terms of (a) removal rate and (b) adsorption capacity q_e and distribution coefficient K_d . (Reproduced with the permission from [122], Copyright 2019 Elsevier Inc.)

not significant.^[123] Dai *et al.* found negligible influence of ionic strength on the U(VI) adsorption onto GO/PDA/PAO-0.2 as shown in Fig. 1b.^[113] Zhao, *et al.* found that the adsorption efficiency of U(VI) by AOMGO was almost unaffected by ionic strength, which was mainly due to the formation of inner layer surface complexes between U(VI) and amidoxime and other oxygen-containing functional groups on the surface of the AOMGO rather than the formation of outer surface complexes or ion exchange.^[90]

On the contrary, some other studies reached different conclusions. Wang *et al.* reported that the uranium adsorption capacity of GO nanoribbons (GONRs) was sensitive to the changes of ionic strength in solution.^[125] Song *et al.* found that the increase NaClO₄ concentration could cause the uranium adsorption capacity of cyclodextrin/GO (CD/GO) to decrease, which was due to high ionic strength inhibited the transfer of U(VI) to the surface of the CD/GO.^[126] In addition, high ionic strength could reduce the electrostatic repulsion between the CD/GO, which could lead to the agglomeration of the CD/GO and then reduce the adsorption capacity of U(VI).

Considering the practical applications of GO-based nanomaterials for U(VI) removal, it is important to examine the competition of co-existing ions in uranium-containing aqueous solution. For example, in order to evaluate the selectivity performance of Ag nanoparticles and L-Arginine functionalized rGO composites (Ag-L-Arg-rGH) for the adsorption of U(VI), adsorption experiments were performed in the aqueous solutions with competitive ions of Ba²⁺, K⁺, Mg²⁺, Co³⁺, Ni²⁺, Sr²⁺, and Cu²⁺. The Ag-L-Arg-rGH showed higher removal rate (Fig. 4a), a larger adsorption capacity q_e , and greater distribution coefficient K_d (Fig. 4b) for U(VI) than other ions which indicated that the Ag-L-Arg-rGH could selectively adsorb U(VI).^[111] Li *et al.* found that common anions (Cl⁻, NO₃⁻, SO₄²⁻, CO₃²⁻, *et al.*) and cations (Cu²⁺, Ca²⁺, Cs⁺, Mn²⁺, Sr²⁺, *et al.*) had little effect on U(VI) removal, showing that FH/GOA-800 had good selectivity for U(VI).^[45] Dai *et al.* applied polyamidoxime/polydopamine-decorated GO (GO/PDA/PAO) for U(VI) adsorption from actual mine radioactive wastewater.^[73] The removal rate and K_d of the metal ions in the actual wastewater are listed in Fig. 5. The

Fig. 5 (a) Removal rate and (b) K_d of various metal ions in actual mine radioactive wastewater by polyamidoxime/polydopamine-decorated GO (GO/PDA/PAO). (Reproduced with the permission from [73], Copyright 2019 American Chemical Society.)

Table 2. Adsorption mechanisms of GO-based nanomaterials for uranium

Adsorbents	Adsorption mechanisms	Analysis techniques	Refs
APT+GO	Surface complexation, ion exchange and electrostatic attraction	SEM and XPS analyses	[132]
PEI/GO	Surface complexation	FT-IR and XPS analyses	[49]
FH/GOA	Surface complexation and electrostatic attraction	FT-IR and XPS analyses	[45]
MIL-68/GO	Surface complexation	XPS analyses	[87]
Ag-L-Arg-rGH	Surface complexation	FT-IR and XPS analyses	[111]
rGO/ZIF-67 Aerogel	surface complexation and electrostatic attraction	Zeta potential, FT-IR and XRD analyses	[10]
Fe-Ni/GO	Reduction, surface complexation and sorption	FT-IR, EDS and XPS analyses	[110]
GO/PDA/PAO-0.2	Surface complexation	FT-IR and XPS analyses	[73]
MBTA-GO	Surface complexation	SEM, FT-IR, XPS, EXAFS analyses and DFT calculations	[99]
GZA	Electrostatic interaction, intraparticle diffusion and coordination	FT-IR and XPS analyses	[88]
GCZ8A	Electrostatic interaction, intraparticle diffusion and coordination	FT-IR and XPS analyses	[43]
GO-pDA-PEI	Coordination and intramolecular diffusion	XPS and FT-IR analyses	[97]
GO/PEDOT:PSS	Surface complexation and immobilization	TEM-EDX, FT-IR and XPS analyses	[100]
GO- DM-AO	Chelation, Coordination and ion diffusion	FT-IR and XPS analyses	[75]
Lysinibacillus-GO	Surface complexation	XPS analyses	[103]
AO/mGO	Surface complexation	EXAFS analyses and surface complexation modeling	[133]
M-GO	Reduction and surface complexation	XPS, XANES analyses, and DFT calculations	[134]
MLGO	Surface complexation	Batch Experiments and Surface complexation modeling	[135]
GO-CS	Inner-sphere surface complexation	Batch Experiments and FT-IR, XPS, EXAFS analyses	[136]

removal rate and K_d by the GO/PDA/PAO for U(VI) were higher than those for other metal ions, which indicated that the GO/PDA/PAO had selective adsorption capability for U(VI) from the actual wastewater. These results indicated that GO-based nanomaterials as absorbents showed better selectivity after GO modified by other functional groups or formed composites. However, it is still a key challenge to remove and recover uranium from aqueous solution, especially in seawater. For example, vanadium has been proved to be the main competing ion for uranium recovery from seawater because of the similar size and charge of the vanadium ion with uranyl ion. Most of adsorbents showed an inferior selectivity between

uranium and vanadium.^[14,127,128] Developing GO-based nanomaterials as absorbents with excellent high selectivity for uranium in harsh conditions is still needful.

In summary, the uranium adsorption efficiency and capacity of GO-based nanomaterials are greatly affected by various factors, such as the pH value, temperature, contact time, ionic strength, and co-existing ions of solutions. From time- and temperature-dependent adsorption experiments, kinetics, isotherm, and thermodynamic models can be established to understand the interactions between uranium and GO-based nanomaterials. Based on the above results, the kinetic models reveal that the uranium adsorption by GO-

based nanomaterials is most fitted with pseudo-second order kinetic equations. The isotherm and thermodynamic models demonstrate that uranium adsorption by GO-based nanomaterials generally is fitted well with the Langmuir isotherm model and the adsorption process is endothermic and spontaneous.

4. Microscopic analysis of adsorption mechanisms

Different GO-based nanomaterials have different adsorption mechanisms with uranium. Their adsorption mechanisms usually include surface complexation or coordination,

chelation, electrostatic attraction, sorption, ion exchange, ion diffusion, precipitation, reduction reaction, *etc.*^[129] The possible mechanisms between uranium and GO-based nanomaterials in recent studies are shown in Table 2. Generally, adsorption mechanisms are expounded from the results of batch techniques, spectroscopic analysis, surface complexation modeling, and theoretical calculations. From batch experimental results, adsorption mechanisms can be explained, such as inner-sphere surface complexation is mostly ionic strength-independent and pH-dependent while outer-sphere surface complexation or ion exchange is mostly

Fig. 6 Spectroscopy studies of post-GO and post-GO/PEDOT:PSS: (a) survey scans of XPS spectra and high-resolution XPS spectra of (b) U 4f and (c, d) O 1s, and (e, f) FT-IR spectra. (Reproduced with the permission form [100], Copyright 2019 Elsevier Ltd.)

pH-independent and ionic strength-dependent.^[57,130,131] We can also understand adsorption mechanisms from batch experimental adsorption data of the adsorption affecting factors that were discussed in Section 3 with the adsorption isothermal models and adsorption thermodynamics of GO-based nanomaterials for uranium summarized in Table 1. In the following sections, we will discuss the interaction mechanisms between uranium ions and GO-based nanomaterials according to spectroscopy analysis, surface complexation modeling and computational theoretical calculations.

4.1 Spectroscopy analysis

The adsorption mechanisms of GO-based nanomaterials for uranium can be elaborated by analyzing the species and the microstructure of uranium at molecular level and the functional groups of the GO-based nanomaterials as the adsorbent which form surface complexes with uranium. Various advanced diffraction and spectroscopic techniques have been employed, such as X-ray diffraction (XRD), X-ray photoelectric spectroscopy (XPS), Fourier transformed infrared spectroscopy (FT-IR), X-ray absorption fine structure (XAFS) spectroscopy, which includes X-ray absorption near edge structure (XANES) and extended X-ray absorption fine structure (EXAFS) spectroscopy, *etc.* These diffraction and

spectroscopic studies are of great help to deeply understand the uranium adsorption mechanism at solid-liquid interfaces. For example, Song *et al.* found that the C-O/S-O bonds participated in surface complexation reaction with uranium may originate from the carboxyl/sulfonate groups instead of the alcohol hydroxyl group by FT-IR and XPS analysis (Fig. 6).^[100] The adsorption mechanism of U(VI) onto the surface of GO-DM-AO composites including chelation, coordination and ion diffusion was analyzed by FT-IR and XPS techniques (Fig. 7).^[75] In Fig. 7a, the XPS spectra of adsorption U(VI) showed that uranium was attached to the GO-DM and GO-DM-AO. In Fig. 7b and 7c, the varied binding energies of N 1s and O 1s after adsorption of U(VI) indicated that the adsorption behavior can be attributed to N-containing and O-containing functional groups. In Fig. 7d, the FTIR spectra of GO-DM-AO showed obvious blue shift at 3433 cm^{-1} and 995 cm^{-1} , indicating the coordination or chelation between U(VI) and -OH, -NH₂, AO groups of the GO-DM-AO.^[75] Huang *et al.* used FT-IR, XPS and EXAFS spectroscopic methods to find the inner-sphere surface coordination structure of U(VI) with -COOH, -OH, and -NH₂ groups of the GO-CS samples.^[136] In addition, FT-IR spectral results showed the broadening extent increased with the increase of solution pH, suggesting the increasing involvement of CS-NH₂ groups in the U(VI) coordination at high pH. Hu *et al.* investigated the

Fig. 7 Spectroscopy studies of GO-U, GO-DM-U and GO-DM-AO-U: (a) XPS survey scans, (b) N 1s and (c) O 1s XPS spectra after adsorption of U(VI), and d) FT-IR spectra. (Reproduced with the permission form [75], Copyright 2019 Elsevier B.V.)

Fig. 8 Uranium LIII-edge XANES (A) and EXAFS (B) analysis of U-loaded AO/mGO under different conditions, $C_0 = 10$ mg/L, $m/v = 0.5$ g/L, pH 4.0, $I = 0.01$ mol/L, $T = 293$ K. (Reproduced with the permission from [140], Copyright 2014 American Chemical Society)

enrichment of amidoxime/magnetic graphene oxide (AO/mGO) for U(VI) by XANES and EXAFS analysis.^[127] The k^2 -weighted U L_{III} -edge XANES and Fourier transforms (FT) EXAFS spectra was shown in Fig. 8A and B, respectively. The adsorption energy of near-edge of U loaded on the AO/mGO were 17175.1 eV located between that of U(VI) (17173.8 eV) and U(IV) (17176.2 eV) (Fig. 8A), indicating that the adsorbed U(VI) was partly reduced to U(IV) by AO/mGO. In Fig. 8B, two FT features at 1.91 Å and 1.40 Å can be well fitted by five equatorial oxygen shell ($U-O_{eq}$ at 2.35 Å) and two axial oxygen shell ($U-O_{ax}$ at 1.78 Å), respectively. For AO/mGO, FT feature at 3.01 Å can be fitted by 0.2 U-Fe shell at 3.42 Å, revealing the formation of inner-sphere surface complexation. The other FT feature at 3.51 Å was consistent with the U-U shell of $UO_2(s)$, indicating that the adsorbed U(VI) was partly reduced to U(IV) by structural Fe(II) of AO/mGO. The results of XANES and XAFS spectra demonstrated that adsorbed U(VI) was reduced to U(IV) by AO/mGO. Furthermore, the effective adsorption of U(VI) on AO/mGO was due to the formation of inner-sphere surface complexation based on EXAFS analysis. These mentioned results indicate that spectroscopic techniques are generally helpful to understand the uranium coordinated with the functional groups on GO-based adsorbing nanomaterials.

4.2 Surface complexation modeling

Surface complexation models (SCMs) are effective approaches to interpret the interaction mechanisms between the solid-liquid interface of adsorbates and adsorbents.^[137] Usually, SCMs including non-electrostatic model (NEM), constant capacitance model (CCM), three-layer model (TLM) and diffuse layer model (DLM) are extensively used for the simulation of interaction mechanisms. The different surface complexes are proposed to happen depending on the various

chemical factors such as ionic strength, pH, and temperature. Results from surface complexation modeling can be compared with or verify the spectroscopic results of EXAFS, XPS, *etc.*, which can be used to interpret their interaction mechanisms in more depth. Sun *et al.* used surface complexation modeling to verify their EXAFS analysis that uranium mainly formed inner surface complexes on GOs and $HOOC-GO$ while outer surface complexes on rGO.^[138] Hu *et al.* used surface complexation modeling and found that pH-dependence and isothermal adsorption of U(VI) on AO/mGO can be well fitted by diffuse layer model with two inner-sphere complexes such as $SOUO_2^+$ and $SOUO_2(CO_3)_2^{3-}$ species (Fig. 9).^[133] Duster *et al.* tested whether three different U(VI) to MLGO ratios affected the ability of NEM and DLM and predicted U(VI) adsorption behaviors over the pH range of 2 to 9.5 and ionic strength from 1 to 100 mM.^[135] NEM and DLM formulations assumed interactions between discrete surfaces sites of the MLGO and the abundant aqueous U(VI) complexes over a given pH range. They determined that the extents of U(VI) binding required adsorption of U(VI) species, such as UO_2^{2+} and uranyl hydroxides and/or carbonate, on the MLGO and calculated the stability constants for the essential U(VI)-MLGO surface complexes.

4.3 Computational theoretical calculations

Computational chemistry calculations can be used to obtain the electronic structure, species distribution, coordination properties and thermodynamic properties of complexes formed between uranium radionuclides and GO-based nanomaterials at the molecular level, which play important roles in elucidating their solid-liquid interaction mechanisms.^[139] The comparison and mutual verification of the solid-liquid interaction data between uranium and the adsorbing GO-based nanomaterials obtained by theoretical

Fig. 9 Surface complexation modeling of U(VI) adsorption on AO/mGO at (A) different pH, $C_0 = 10$ mg/L, $m/v = 0.5$ g/L, $I = 0.01$ mol/L, and $T = 293$ K (B) U(VI) concentrations with $C_0 = 10$ mg/L, $m/v = 0.5$ g/L, pH = 4.0, $I = 0.01$ mol/L, and $T = 293$ K. (Reproduced with the permission from [133], Copyright 2018 Elsevier B.V.)

calculations and experimental spectroscopic results can make the experimental studies on the uranium adsorption by GO-based nanomaterials more convincing and reliable. In addition, some experiments are difficult to carry out because of the radioactivity and toxicity of radionuclides, while theoretical calculations can provide an effective way to supplement the relevant adsorption research.

Density functional theory (DFT) is one of the important tools for theoretical calculations, which can calculate relevant basic data of bonding structures and adsorption energies of complexes between radionuclides and adsorbing materials in an effective way and has been well applied in the field of

computational chemistry.^[140,141] For example, Yu *et al.* studied the U(VI) adsorption mechanism of 3D GA/TiO₂ by DFT calculations and obtained a Ti–O3 bond length of 1.972 Å between 3D GA and TiO₂ surface (Fig. 10).^[142] Also, the adsorption energy of the 3D GA/TiO₂ for U(VI) was calculated to be 1.60 eV, which indicated a strong chemisorption. The strong chemisorption was further confirmed by the formation of surface complexes with U–O bond lengths of 2.348 Å (U–O1) and 2.638 Å (U–O2). Moreover, the adsorption mechanism was illustrated by calculating differential charge density. Charge transfer was found to occur between oxygen atom of the 3D GA/TiO₂ and

Fig. 10 Theoretical analysis on the adsorption process of U(VI) by 3D GA@TiO₂. (Reproduced with the permission from [142], Copyright 2019 Elsevier Ltd.)

Fig. 11 The optimized structures of GO(–O–)/uranyl and GO(–COOH)/uranyl at different pH levels. Bond lengths are in Å. (Reproduced with the permission form [143], Copyright 2018 Elsevier B.V.)

the uranium atom, which proved the electrostatic attraction between U(VI) and the 3D GA/TiO₂. Using DFT, Ding *et al.* calculated the binding energies of [GO–O...UO₂]⁺ and [GO–COO...UO₂]⁺ as 23.5 kcal/mol and 51.1 kcal/mol, respectively, which were high. It revealed that the carboxyl and hydroxyl groups were responsible for the high removal efficiency of U(VI) by the magnetite-graphene oxide composites (M–GO).^[134]

Ai *et al.* explained the interaction mechanism of uranium and GO with varying pH levels of the solution by means of high-level DFT with an implicit water model.^[143] A further look at the bonding structures and adsorption energies of 12 GO/uranyl complexes showed that the major adsorption sites of GO were the functional groups such as carboxyl group, epoxy group, and hydroxyl group. Moreover, it was found that high pH value was more beneficial to the adsorption process of uranyl ions onto GO because of stronger electrostatic attraction between uranyl ions and negatively charged O atom. The optimized structures of GO(–O–)/uranyl, GO(–COOH)/uranyl, GO(–OH)/uranyl and GO(–OH–OH)/uranyl

at different pH levels were shown in Fig. 11 and Fig. 12. Taking carboxyl functionalized GO as an example to discuss in detail, their optimized structures (Fig. 11) of [GO–COO...UO₂]⁺, [GO–COOH...UO₂]²⁺ and [GO–COOH₂...UO₂]³⁺ were represented for high, neutral and low pH values, respectively. The change of U–O_{ax} bond in these complexes followed the trend: it became longer as 1.746/1.747 Å in [GO–COO...UO₂]⁺, 1.748/1.735 Å in [GO–COOH...UO₂]²⁺, and 1.768/1.727 Å in [GO–COOH₂...UO₂]³⁺ after the uranyl ions adsorption. The U–O_G bond length of the [GO–COO...UO₂]⁺ was 2.291 Å, and it lengthened to 2.402 Å and 2.674 Å with the increasing of acidity. The shorter U–O_G bond length of the [GO–COO...UO₂]⁺ than [GO–COOH...UO₂]²⁺ and [GO–COOH₂...UO₂]³⁺ indicated a stronger interaction between the uranyl ion and GO at high pH level. Moreover, the GO with bi-hydroxyl groups demonstrated the highest adsorption capacity for uranyl ions by comparing with other forms of GO. For the [GO–OH–OH...UO₂]²⁺, the uranyl ions were fastened strongly by two U–O_G bonds of 2.444 Å and 2.463 Å as shown

Fig. 12 The optimized structures of GO(–OH)/uranyl and GO(–OH–OH)/uranyl at different pH levels. Bond lengths are in Å. (Reproduced with the permission form [143], Copyright 2018 Elsevier B.V.)

in Fig. 12. This was different from the mono-substituted case, where the uranyl ions kept away ~ 3.267 Å from the GO surface. In the meantime, the $[\text{GO}-\text{OH}-\text{OH}_2\cdots\text{UO}_2]^{3+}$ had the same uranyl ion situation that was fastened by a U–O_G bond of 2.914 Å instead of leaving away from the GO surface. Therefore, more oxygen-containing groups might improve the adsorption performance for uranyl ions.^[143]

Based on the results discussed above, GO-based nanomaterials for high uranium adsorption are mainly attributed to their surface functional groups. However, the influence degree of different functional groups of GO-based nanomaterials on uranium adsorption stays unclear due to various functional groups on GO-based nanomaterials. The binding energy attained from DFT calculations can provide some valuable information about the contribution of different groups of GO-based nanomaterials for uranium adsorption, i.e., higher binding energy means stronger interaction between uranium and functional groups of GO-based nanomaterials.^[57] For example, Sun *et al.* utilized DFT calculations to show evidence that the –COOH group formed much stronger surface complexes with U(VI) than other oxygen-containing functional groups.^[138] Their DFT calculations showed that the binding energy value of $[\text{GOs}-\text{COO}\cdots\text{UO}_2]^+$ was 50.5 kcal/mol, which was much higher than that of $[\text{rGOs}\cdots\text{UO}_2]^+$ (8.1 kcal/mol). The contribution of different functional groups for binding uranium on GO-based nanomaterials can be revealed by DFT calculations at molecule level, which is difficult to be achieved from batch adsorption experiments, spectroscopy analysis and surface complexation modeling. Therefore, theoretical calculations are a good supplemental method for microscopic analysis of uranium adsorption mechanisms onto GO-based nanomaterials.

In summary, the microscopic adsorption mechanisms between uranium and GO-based nanomaterials have been clearly revealed by current studies using microscopic spectral technology, surface complexation model and theoretical calculations. It has been found that the adsorption mechanisms include surface complexation or coordination, chelation, electrostatic attraction, sorption, ion exchange, ion diffusion, precipitation, reduction reaction, *etc.* Other new chemical functional groups or composites introduced onto GO by surface modifications can greatly affect the interaction mechanisms. GO-based nanomaterials exhibited different interactions with uranium due to the modifications by different functional groups or composites. For example, if functional groups, such as amino and carboxyl groups are introduced onto GO, surface complexation or coordination should be the main adsorption mechanisms. If reductive substances, such as zero-valent iron or Fe–Ni galvanic cell, are introduced onto GO, reduction should play an important role in elimination of U(VI). Nonetheless, among these above-mentioned mechanisms, surface complexation or coordination is the most common interaction mechanisms between uranium and GO-based nanomaterials because of various functional groups on GO or the new functional groups introduced can participate in

complexing or coordinating with uranium.

5. Conclusions and perspectives

In summary, establishing and understanding an adequate design model for the removal of uranium species from aqueous media, isotherm, thermodynamic, and kinetics studies have been performed as essential basic prerequisites for GO-based nanomaterials as adsorbents. In common, uranium adsorption by GO-based nanomaterials mostly fits well with the Langmuir isotherm model and the adsorption process is endothermic and spontaneous. Also, kinetic modeling reveals the potential rate-controlling steps such as mass transport or chemical reaction processes. The most commonly fitted model for uranium by GO-based nanomaterials is the pseudo-second order kinetic equations. In addition, these models obtained from macroscopic batch experiments, combined with microscopic spectral technology, surface complexation model and theoretical calculations, have been applied to study the adsorption behaviors of uranium by GO-based nanomaterials, which have made the interaction mechanisms between uranium and GO-based nanomaterials more deeply and clearly revealed.

Despite great advances in GO-based nanomaterials for uranium adsorption have been attained in recent years, there are still some challenges to overcome and many issues to be solved:

(1) Hummers method is the most commonly used method for the synthesis of GO. This method faces with the problems of explosion risk, environmental pollution and long reaction cycle. Development of safe, green and rapid synthesis methods of GO is needed to reduce the preparation cost and promote the industrial applications of GO, *e.g.* uranium recovery and removal discussed in this article.

(2) Improving the selectivity of GO-based nanomaterials is needed for removal and recovery of uranium because actual wastewater and seawater have high ionic strength and many co-existing ions. Especially, it is a quite great challenge to uptake uranium from seawater. Therefore, the introduction of GO-based nanomaterials with appropriate functionalities, such as amidoxime and phosphoryl groups, enables the exploration of GO-based nanomaterials for uranium with desirable selectivity, which will require further intensive research in the future.

(3) The structure of a material determines its property.^[144] Regulating the structure of GO-based nanomaterials through optimizing synthesis techniques and experimental conditions to improve their uranium adsorption performance require further studies. Moreover, the correlative relationship between molecular structure and adsorption performance should be clarified for further design of favorable GO-based nanomaterials for uranium recovery and removal.

(4) Current studies have found that most of GO-based nanomaterials with high adsorption efficiency for uranium are mainly due to their surface functional groups. However, because of the diversity of the functional groups of GO, such

as hydroxyl, carboxyl, carbonyl, epoxy group, etc., as well as some new functional groups introduced by surface functionalized modifications, further theoretical calculations combined with experimental studies are needed to determine which functional groups on uranium adsorption plays a role or the influence degree of the different groups for uranium adsorption.

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Supporting Information

Not Applicable.

Conflict of Interest

There is no conflict of interest.

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