In order to deal with the increasingly serious pollution of graphene oxide (GO) to the environment. In this paper, the use of red sandstone to treat GO-contaminated aqueous solution is proposed for the first time, and the adsorption capacity and adsorption mechanism of red sandstone to GO are discussed. The controlled variable method was used to explore the optimal pH, concentration, and quality of red sandstone for GO aqueous solution. The adsorption isotherm, thermodynamics, and adsorption kinetics were fitted. Adsorption characterization tests were performed using XRD, AFM, XPS, FT-IR, SAP, laser particle size analyzer, and SEM. The results show that when $T = 303 \text{ K}$, the optimum adsorption condition of red sandstone for GO is $\text{pH} = 4$, the mass of the adsorbent is 40 mg, and when the concentration of GO is 80 mg/L, the adsorption capacity is 90 mg/g. The adsorption isotherm model fits the Langmuir model. The adsorption thermodynamic experiments and fitting results show that the reaction is endothermic. XRD and FT-IR tests showed that CaCO$_3$ in red sandstone was involved in the adsorption of GO. SEM, TEM, and AFM microscopic results showed that GO was adsorbed on the surface of red sandstone particles. The XPS test showed that Ca$^{2+}$ in red sandstone and C=O bond in GO undergo ionic or coordination reaction. The adsorption kinetics fit a pseudo-second-order kinetic model. This study will provide some references for the removal of GO in the environment and the interaction mechanism with natural minerals.
adsorption capacity of GO by different cations and different kinds of clay minerals. The results showed that the adsorption of GO by different ions was related to water and radius of ions, and the hydroxyl and metal-oxygen bonds of clay minerals can interact with GO. Different types of clay minerals have different cohesive capacities for GO due to their different electrical properties in aqueous solutions, and their adsorption capacities are as follows: nanoalumina > kaolin > bentonite > montmorillonite [15]. Wang et al. studied the adsorption capacity of GO by different layered double hydroxides and compared the effects of different ratios of Mg/AL- and Ca/(Mg+AL)-layered double hydroxides on GO adsorption. It was found that the adsorption capacity of GO varied under different Mg/AL and Ca/(Mg+AL) ratios. When the ratio of Mg/AL was 4, the adsorption effect was the best; when the ratio of Ca/(Mg+AL) was 1, the best effect was achieved. The temperature and ions in the solution had little effect on GO adsorption [16]. Zhao et al. found that the adsorption between GO and negatively charged rocks was mainly electrostatic attraction and difficult to desorption by studying the interaction between GO and montmorillonite and kaolinite [17]. Feriancikova and Xu studied the adsorption capacity of GO by saturated sand and found that a small amount of retained particles occurred at low ionic strength (e.g., 1 mL of NaCl). When the ionic strength was raised to 100 mL of NaCl, the retention of degranulation increased significantly but was limited by its retention capacity. The Langmuir model can describe the migration behavior of saturated sand to degranulate [18]. In the study of Zhou and others, the effect of iron tailing sand on GO adsorption was investigated. The experimental data showed that pH, iron tailing quality, GO concentration, and temperature all had an effect on the removal of GO. The adsorption model conforms to the Langmuir monolayer adsorption model, and the adsorption process is a spontaneous process. Physical adsorption [19]. The removal of contaminated GO from water using calcareous sand from the South China Sea is studied by Lv et al. The experimental results show that the adsorption of GO on calcareous sand can reach the adsorption equilibrium within 5 h [20]. The above scholars have conducted in-depth research on the adsorption of GO by geotechnical minerals. Whether there are other geotechnical minerals that can react with GO needs to be further explored.

Red sandstone is a widespread rock that has been used as a building material since ancient times due to its bright colors [21]. Red sandstone is composed of feldspar, quartz, and clasts with a porous structure [22]. Many scholars have studied the properties of red sandstone [23–26] mostly on its mechanical and physical properties, but there are few studies on its adsorption properties. The natural porous structure of red sandstone has good adsorption properties. Chen et al. studied the kinetic characteristics of arsenic (V) adsorption on red soil after sandstone weathering. The results showed that chemical adsorption was the main adsorption process, and the adsorption effect was the best when pH = 7.0, T = 293 K, and As(V) concentration was less than 4.0 mg/L, and the hydraulic retention time was not less than 180 min [27]. Song et al. studied the adsorption kinetics and thermodynamic of Ni(II) by Pisha sandstone. The results showed that the adsorption was carried out when T = 293 K and pH = 7, the equilibrium was reached at 30 min, and the adsorption capacity of Ni(II) was 23.688 mg/g. The adsorption process of Ni(II) by Pisha sandstone can be described by the Langmuir pseudo-second-order kinetic model, which is mainly ion exchange and chemical adsorption, while chemical precipitation plays a little role in this process [28].

In summary, although some scholars have studied the adsorption of heavy metal ions on red sandstone, few people have paid attention to the adsorption of GO on red sandstone. As a relatively cheap and easily available adsorbent, red sandstone has many characteristics of a good adsorbent, such as high adsorption capacity, large specific surface area, and large volume [29]. It has broad application prospects in adsorption. Therefore, red sandstone was designed as adsorbent to remove GO in aqueous solution, and the adsorption effect of different pH, temperature, GO concentration, adsorbent mass, and different time was studied. At the same time, laser particle size analyzer (Mastersizer3000) X-ray diffractometer (XRD), Fourier Transform Infrared Spectroscopy (FTIR), scanning electron microscopy (SEM), Atomic Force Microscopy (AFM), high-resolution transmission electron microscopy (TEM), and X-ray photoelectron spectroscopy (XPS) were used to analyze the microstructure and characterization of the adsorbed samples and reveal its adsorption mechanism.

2. Test Materials and Methods

2.1. Materials. The red sandstone used in the experiment was collected from Yueyang City, Hunan Province, China. It is brownish red with fine-grained structure and uniform texture. The main components are quartz, clasts, and feldspar. The cementation composition is mainly iron, which is a hard brittle sedimentary rock. Figure 1(a) shows the crushed red sandstone. The proportion of grain size of red sandstone is shown in Figures 1(b) and 1(c). It can be seen from the particle size distribution diagram that the proportion of powdery red sandstone with particle size less than 90 μm is 84.8%, and the particle size of powdery red sandstone is mainly concentrated between 1 μm and 100 μm, and the average particle size is 56.8 μm.

The GO aqueous solution is produced by Suzhou Tanfeng Technology Co., Ltd., with a concentration of 2 mg/mL and a specific surface area of 2630m²/g. Its chemical element composition is shown in Table 1.

2.2. Test Section

2.2.1. Test Scheme. Red sandstone powder was taken as adsorbent, and the adsorption rate of GO by red sandstone was investigated by different pH, temperature, GO concentration, and adsorbent mass. The test steps are as follows:

Crush the obtained samples and put them in an oven to continue drying for 24 hours, taking into account the effect of particle size on the adsorption capacity. This paper chose to use a standard sieve with a 0.074 mm aperture to screen...
fine particles (Figure 1). It is used to reduce the effect of par-
ticle size on adsorption [30].

When using, first sonicate the GO aqueous solution with an ultrasonic device for 30 minutes, so that it can be uni-
formly dispersed in the aqueous solution. Then, differ-
ent volumes of GO solution and deionized water were taken with a pipette and placed in a volumetric
flask, so that the total volume of GO and deionized water was 50 mL. Sodium hydroxide solution and nitric acid solution with negligible volumes were added to adjust to different pH, and then, red sandstone powder with different mass was added.

Oscillation mixing. The above-prepared solution was poured into a volumetric flask and then put into a shaker
at different temperatures. After shaking for 3 h at 240 rpm, the volumetric flask was taken out and put into a constant
temperature curing box to stand for 24 h at different temperatures.

Absorbance test. 1 mL of the middle-layer clear liquid after adsorption was put into a test tube, and then 24 mL of deionized water was added to make a 25 mL solution. After standing for 10 min, the absorbance was measured by an ultraviolet spectrometer (incident wavelength \( \lambda = 210 \) nm). The relationship between absorbance and concentra-
tion was obtained by linear regression analysis. The relation-
ship between absorbance \( (y) \) and concentration \( (x) \) is the
 calibration curve. GO was selected as (0 mg/L, 1 mg/L,
2 mg/L, 3 mg/L, 4 mg/L, and 5 mg/L) solutions of 6 different
concentrations, and then, the absorbance was measured with an ultraviolet/visible spectrophotometer (UV75N). The measured data can be linearly fitted and then brought into the calculation to obtain the concentration of the measured
solution [31], and the average value was taken through three
time measurements. The adsorption capacity, adsorption rate, and distribution coefficient were obtained from formulas (1), (2), and (3), respectively [32].

\[
R = \frac{C_0 - C_e}{C_0} \times 100\% , \quad (1)
\]

\[
Q_e = \frac{C_0 - C_e}{m} \times V , \quad (2)
\]

\[
K_d = \frac{C_0 - C_e}{C_0} \times \frac{V}{m} , \quad (3)
\]

where \( C_0 \) represents GO concentration (mg/L), \( C_e \) represents solution concentration at adsorption equilibrium (mg/L), \( Q_e \) represents adsorption capacity (mg/g), \( m \) represents adsorbent mass (mg), \( V \) represents solution volume (mL), and \( K_d \) is distribution coefficient.

2.2.2. Microscopic Characterization. Constant temperature shaker (THZ-100B), UV/Vis spectrophotometer (UV75N), pH meter (FE28), electronic analytical balance (FA324C), X-ray diffractometer (XRD, Empyrean), Fourier transform infrared spectrometer (FTIR, NEXUS), scanning electron microscope (SEM, JSM-6360LV), atomic force microscope (AFM, SPA400), high-resolution transmission electron microscope (TEM, JEM-2100F), X-ray photoelectron spectroscopy (XPS, Thermo ESCALAB 250XI), a specific surface area analyzer (Tristar 3020II), and a laser particle sizer (Mastersizer 3000) were used to analyze the microscopic and characterization of the adsorbed precipitate samples.

3. Results and Analysis

3.1. Morphology Analysis

3.1.1. XRD. XRD can analyze the crystal structure and material composition of red sandstone and GO as well as the adsorbed products [33]. Cu target Kα was used as the radiation power source to characterize GO, red sandstone, and GO/red sandstone derivatives by XRD, as shown in Figure 2. PDF cards refer to the ICSD standard. The characteristic peaks before and after adsorption are compared. There is an obvious (001) characteristic peak at 10.08° in the GO spectrum. In the XRD pattern of red sandstone, 20.84° and 26.26° can be marked as SiO₂ (046), 27.88° can be marked as (Na,Ca)Al(Si,Al)₃O₈(041), and 29.44° can be marked as CaCO₃ (005) [34]. In the diffraction pattern of GO/red sandstone, the peak intensity of CaCO₃ (005) decreases from 18090.5 cm⁻¹ to 8169.5 cm⁻¹, and that of (Na,Ca)Al(Si,Al)₃O₈(041) also decreases from 18801.1 cm⁻¹ to 11042.4 cm⁻¹, compared with that of red sandstone. The diffraction peak of GO disappears, indicating that CaCO₃ and (Na,Ca)Al(Si,Al)₃O₈ (041) in red sandstone may be involved in the adsorption of GO, rather than the simple stacking of GO on the surface of red sandstone.

3.1.2. Brunauer-Emmett-Teller (BET) Surface Area Analysis. The specific surface area and pore size are the main factors affecting the adsorption [35], so we measured the specific surface area and pore size of the red sandstone. At the same time, in order to reduce the influence of the adsorbent moisture on the experimental results of the comparative surface area. All samples should be dried at 300°C for 3 hours and vacuumed. In the experiment, high-purity (99.99%) N₂ was used as the adsorbate, and the amount of N₂ adsorption at different relative pressures was measured in a low-temperature environment. The final measurement results are shown in Table 2: we found that the specific surface area of red sandstone is 10.0729 m²/g, which is larger. The average pore volume is 0.019024 cm³/g, and the average pore size is 5.69122 nm. This shows that red sandstone is a natural large specific surface area and an excellent adsorbent.

3.1.3. SEM and TEM. The samples before and after adsorption were observed by scanning electron microscopy (SEM) and high-resolution transmission electron microscopy (TEM), and the obtained microscopic morphologies are shown in Figure 3 [36, 37]. Figures 2(a) and 2(b) are the SEM and TEM pictures of red sandstone, through which the sharp edges and irregular shapes of red sandstone can be clearly seen. Figures 2(c) and 2(d) are the SEM and TEM images of GO, through which we can find the multilayered two-dimensional flake-like structure of GO, with some folds on the edges and smooth in the center [38]. Figures 2(e) and 2(f) are the SEM and TEM morphology of GO/red sandstone after adsorption, and the film-like GO attached to the surface of red sandstone particles can be clearly seen.

3.1.4. FT-IR. In order to further analyze which chemical bonds in GO are involved in the adsorption, FT-IR was used...
to analyze the changes of functional groups and the migration and disappearance of chemical bonds of GO, red sandstone, and GO/red sandstone derivatives after adsorption [39], as shown in Figure 4. GO exhibits stretching vibration of the -OH group at 3403 cm$^{-1}$, stretching vibration of C=O at 1734 cm$^{-1}$ [40], stretching vibration of C=C on the benzene ring skeleton structure at 1628 cm$^{-1}$ [41], stretching vibration of C-OH at 1219 cm$^{-1}$, and vibration peak of C-O at 1049 cm$^{-1}$ [42]. Red sandstone presents characteristic peaks of CO$_3$-out-of-plane deformation vibration in CaCO$_3$ at 875 cm$^{-1}$ and 712 cm$^{-1}$ [43], and symmetrical stretching vibration peak of Si-O bond in SiO$_2$ at 468 cm$^{-1}$ [44]. The peak of GO/red sandstone weakens at 1628 cm$^{-1}$, disappears at 1734 cm$^{-1}$, 1219 cm$^{-1}$, 1049 cm$^{-1}$, 875 cm$^{-1}$, 712 cm$^{-1}$, and 590 cm$^{-1}$, and strengthens at 468 cm$^{-1}$. The disappearance of the peak at 1734 cm$^{-1}$ may be caused by the combination of Ca$^{2+}$ in red sandstone and O-C=O bond in GO, resulting in the coagulation of GO [45], which is consistent with the change rule of the peak value of CaCO$_3$ in XRD pattern, indicating that GO has undergone ionic reaction or coordination reaction on the surface of red sandstone [46].

3.1.5. XPS and AFM. XPS is an effective characterization method that can qualitatively and quantitatively detect elements. Through the XPS spectra, the position of the characteristic spectral line is used to qualitatively analyze the elemental composition of material; the elemental content is quantitatively analyzed by the intensity of the photoelectron spectral line; the chemical state of each element can be further determined by peak fitting [47]. The characterization results are shown in Figure 5(a). Through the spectra, various strong peaks can be clearly seen, such as O1s, C1s of GO, O1s, C1s, Ca2p, Si2p, and Al2p of red sandstone and GO/red sandstone, which conforms to the results of XRD.
analysis. The peak of C1s in the spectra of red sandstone is lower (7898.35), and that in the spectra of GO/red sandstone is higher (17768.9), but it is still much lower than that in the spectra of GO (30106.2). Therefore, by performing peak fitting of C1s of GO and GO/red sandstone, the C1s spectra can be deconvolved and integrated into three components at 284.8 eV, 286.2 eV, and 289.0 eV [48]. The fitting results are shown in Figures 5(b) and 5(c). It can be found through fitting that the three convolutions of C1s of GO/red sandstone had obvious changes compared with that of GO. The most obvious changes are the bonds of C-O and O-C=O, which change from 286.2 eV and 289.0 eV to 286.69 eV and 287.02 eV, respectively. The proportion of the peak area of C-O decreases from 43% to 16%, and that of O-C=O increases from 15% to 39%. From the position changes of the three convolutional components of C1s and the proportion change of the peak area before and after adsorption, it can be seen that GO has solidified on the surface of red sandstone [49], which is consistent with the results of FT-IR analysis.

AFM can further explore the height and morphology of GO and GO solidified on the surface of red sandstone [50]. The surface thicknesses of GO and GO/red sandstone were characterized, as shown in Figure 6. It can be seen from Figure 6 that GO exhibits a single-layer or double-layer structure [51], and the maximum thickness is only 2.4 nm as shown in Figures 6(a) and 6(c). For the GO/red sandstone samples, we can clearly see red sandstone particles with a height of 4.5 nm and GO attached to the surface of red sandstone particles, as shown in Figures 6(b) and 6(d), which is consistent with the results measured by SEM and TEM, indicating that GO is adsorbed on the surface of red sandstone, which further indicates that red sandstone can effectively remove GO.

3.2. The Effect of Solution pH on Adsorption. pH is an important factor affecting the interaction of substances [52]. Figure 7 shows the effect of different pH on the adsorption performance of GO by red sandstone when the adsorbent mass is 40 mg, the solution concentration is 80 mg/L, \( T = 303 \) K after 24 h standing. It can be seen from the figure that with the increase of pH, the adsorption capacity of GO by red sandstone shows a trend of increasing first and then decreasing. The adsorption effect is the best when \( pH = 4 \), the adsorption rate is up to 90%, and the adsorption capacity is also 90 mg/g. The adsorption capacity at \( pH = 3 \) is smaller than that at \( pH = 4 \). Although GO itself has a certain self-coagulation capacity at acidic pH [53], its adsorption capacity is still lower than that at \( pH = 4 \), indicating that the adsorption capacity of GO by red sandstone is inhibited to a certain extent under higher acidic conditions. When \( pH = 4 \), CaCO\(_3\) in red sandstone reacts with H\(^+\) in acidic solution to form Ca\(^{2+}\). Because GO has more oxygen-containing functional groups, it has electronegativity. Particularly, Ca\(^{2+}\) can be directly adsorbed by oxygen-containing functional groups through the double electric layer in addition to compressing the double electric layer, further neutralizing the electronegativity of GO [54]. Studies have found that metal cations are easier to combine with oxygen-containing functional groups, such as epoxy groups and carboxyl groups [55]. Therefore, when \( pH > 4 \), the adsorption capacity of GO by red sandstone gradually decreases with the increase of pH. When \( pH = 10 \), the adsorption capacity and adsorption rate are only 2.47 mg/g and 2.47%, respectively. This is because the gradual decrease of H\(^+\) in the solution leads to the decrease of Ca\(^{2+}\), and the gradual increase of OH\(^-\) in the solution also competes with GO for the adsorption sites on red sandstone, thereby reducing the adsorption effect.

3.3. The Effect of Adsorbent Mass on Adsorption. Figure 8 shows the effect of the red sandstone mass on the adsorption of GO. When GO concentration is 80 mg/L, \( pH = 4 \), and \( T = 303 \) K, the red sandstone mass is set to 10 mg, 20 mg, 30 mg, 40 mg, 50 mg, and 60 mg, respectively. After standing for 24 h, with the increase of red sandstone mass, the removal rate of GO by red sandstone shows a trend of increasing first and then decreasing. When the red
sandstone mass is 40 mg, the removal rate reaches the maximum which is 90%, and the adsorption capacity is 90 mg/g. When the red sandstone mass exceeds 40 mg, its adsorption capacity and adsorption rate decrease compared with that of 40 mg, indicating that the optimal red sandstone mass is 40 mg when GO concentration is 80 mg/L. The reason for the decrease of adsorption capacity and adsorption rate with the increase of adsorbent mass may be that with the continuous increase of red sandstone mass, red sandstone can provide more and more adsorption sites, but as GO concentration is fixed, these adsorption sites can be only combined with limited GO, resulting in an insignificant change in the late adsorption rate compared with that of red sandstone of 40 mg.

3.4. The Effect of GO Concentration on Adsorption. On the basis of the above experiments, the red sandstone mass was set to 40 mg, pH = 4, and $T = 303$ K, and the effect of GO concentration on the adsorption of GO by red sandstone was studied after standing for 24 h. GO concentrations were set to 40 mg/L, 60 mg/L, 80 mg/L, 100 mg/L, and 120 mg/L, respectively. The test results are shown in Figure 9. It can be seen from the figure that with the continuous increase of GO concentration, the absorption capacity of GO by red sandstone generally shows a trend of first increasing and then decreasing. The best GO concentration was 80 mg/L. When the ratio of red sandstone mass to GO concentration was 0.5, the adsorption effect was the best, the adsorption capacity was 90.43 mg/g, and the distribution coefficient was 11.30. With the increase of GO concentration, the adsorption rate gradually decreased, but the adsorption capacity increased gradually. When GO concentration was 120 mg/L, the adsorption rate was only 83%, but the adsorption capacity was 125.64 mg/g, and the distribution coefficient was 6.44. It shows that the adsorption sites of red sandstone are not effectively utilized at low concentration, but with the increase of GO concentration, the adsorption sites are effectively utilized, making the adsorption capacity
continue to increase, but the adsorption efficiency begins to decline. The reason may be that high GO concentration makes the adsorption sites on red sandstone more easily combined with GO molecules, making it adsorb more GO. However, a certain red sandstone mass can provide a certain adsorption site, which makes the adsorption rate and distribution coefficient show a downward trend, indicating that high GO concentration has a certain inhibition effect on the adsorption capacity to a certain extent.

3.5. Adsorption Kinetics and Adsorption Thermodynamics. The above test results show that when $T = 303$ K, the optimal pH for GO adsorption by red sandstone is 4, and the optimal adsorbent mass is 40 mg. Thermodynamic experiments were carried out under the optimal pH and adsorbent mass. Three temperatures (293 K, 303 K, and 313 K) and five GO concentrations (40 mg/L, 60 mg/L, 80 mg/L, 100 mg/L, and 120 mg/L) were set for thermodynamic experiments. Three control groups were set for each temperature and concentration variable. The optimal GO concentration was 80 mg/L. Therefore, the optimal pH, optimal adsorbent mass, and GO concentration were selected as the experimental conditions of adsorption kinetics.

3.5.1. Adsorption Isotherm. In nature, the temperature of an aqueous solution is different at different times. In order to explore the adsorption capacity of GO by red sandstone at different temperatures, when pH = 4, the red sandstone mass was...
40 mg, and under three different temperatures (293 K, 303 K, and 313 K), GO solutions with different concentrations (40 mg/L, 60 mg/L, 80 mg/L, 100 mg/L, and 120 mg/L) were taken for adsorption tests. The test results are shown in Figure 10. When GO concentration is 120 mg/L, the adsorption capacity is up to 137.0 mg/g at 313 K, while that at 293 K and 303 K is only 125 mg/g and 128 mg/g, respectively. It indicates that the adsorption capacity of GO by red sandstone with the same GO concentration increases with the increase of temperature. This is because with the increase of temperature, GO molecules in the aqueous solution are more active, which greatly increases the probability of GO molecules
contacting the adsorption active sites on red sandstone, which leads to the increase of adsorption capacity.

In order to further explore the adsorption morphology of GO on the surface of red sandstone, Langmuir and Freundlich adsorption isotherm equations were used to fit the adsorption process, as shown in formulas (4) and (5) [56].

The fitted image is shown in Figure 11, and the fitting results are shown in Table 3. The correlation coefficients $R^2$ of the fitting results by the Langmuir equation are higher than those by Freundlich. It can be seen that the Langmuir equation can more accurately describe the adsorption process of GO by red sandstone, which is mainly based on...
uniform monolayer adsorption \[57\]. GO attached to the surface of red sandstone can be clearly seen from the SEM and TEM images in Figures 3(e) and 2(f). The maximum adsorption capacity \(Q_{\text{max}}\) in the fitting results of the Langmuir equation increases with the increase of temperature, indicating that the increase of temperature promotes the adsorption reaction.

**Langmuir Isothermal Adsorption Model**: 

\[
Q_e = \frac{bQ_{\text{max}}C_e}{1 + bC_e}, \tag{4}
\]

**Freundlich Isothermal Adsorption Model**: 

\[
\ln Q_e = \ln K_F + \frac{\ln C_e}{n}, \tag{5}
\]

where \(Q_e\) (mg/g) represents the adsorption capacity at equilibrium; \(C_e\) (mg/L) represents the concentration of residual adsorbent in the solution after adsorption; \(Q_{\text{max}}\) (mg/g) represents the saturated adsorption capacity obtained from the fitting results; \(b\) is the adsorption equilibrium constant. \(K_F\) and \(n\) are the Freundlich adsorption equilibrium constants.

3.5.2. Adsorption Thermodynamics. In order to analyze the effect of temperature change on the adsorption of GO by red sandstone, the experimental results were calculated by fitting the thermodynamic parameters, and the energy changes during the adsorption process were analyzed to determine whether the adsorption was a spontaneous reaction or a nonspontaneous reaction \[58\].

Thermodynamic parameters such as standard free energy (\(\Delta G^0\)), enthalpy change (\(\Delta H^0\)), and entropy change (\(\Delta S^0\)) were calculated by formulas (6), (7), and (8), and the effect of temperature changes on the adsorption process was analyzed. Table 4 and Figure 12 shows the thermodynamic fitting curves and calculation parameters. At 293 K, 303 K, and 313 K, the standard free energy (\(\Delta G^0\)) of GO adsorption experiments by red sandstone with different GO concentrations are all negative, indicating that the adsorption process is spontaneous. With the same GO concentration, the absolute value of standard free energy (\(\Delta G^0\)) increases with the increase of temperature, indicating that the increase of temperature is beneficial to the adsorption test. However, both \(\Delta H^0\) and \(\Delta S^0\) are positive, indicating that the reaction is an endothermic reaction \[59\]. There are two reasons: (1) the first reason may be that with the increase of temperature, the diffusion rate of GO increases and the contact rate with minerals increases \[60\]; (2) the increase of temperature is conducive to the formation of chemical bonds generated, which is beneficial for chemisorption.

**Langmuir Isothermal Adsorption Model**: 

\[
\ln K_d = \frac{\Delta S^0}{R} - \frac{\Delta S^0}{RT}, \tag{6}
\]

**Freundlich Isothermal Adsorption Model**: 

\[
\Delta G^0 = \Delta H^0 - T\Delta S^0, \tag{7}
\]

**Distribution Coefficient**: 

\[
K_d = \frac{C_0 - C_e}{C_e} \times \frac{V}{m}, \tag{8}
\]

where \(K_d\) is the distribution coefficient; \(R\) is the standard molar gas constant (8.3145 J/mol·K\(^{-1}\)); \(T\) (K) is the absolute temperature of Kelvin.

3.5.3. Adsorption Kinetics. The adsorption rate is an important index to evaluate the adsorption performance, which
is generally studied by adsorption kinetics. Adsorption kinetics can reveal the diffusion mechanism, adsorption steps, and controlling factors of adsorption [61]. At the same time, it can also reveal the time when the adsorbent reaches the adsorption equilibrium. In order to explore the above factors, the pseudo-first-order kinetic model and pseudo-second-order kinetic model are usually adopted [62], as follows:

pseudo-first-order kinetic equation: \[ \ln (Q_e - Q_t) = \ln Q_e - k_1 t, \] (9)

pseudo-second-order kinetic equation: \[ \frac{t}{Q_t} = \frac{t}{Q_e} + \frac{1}{k_2 Q_e^2}, \] (10)

where \( Q_e \) represents the adsorption capacity at equilibrium, mg/g; \( Q_t \) represents the adsorption capacity at time \( t \), mg/g; \( k_1 \) and \( k_2 \) are constants, g/(mg·min); \( t \) represents the adsorption time, min.

In order to reduce the change of the solution during the sampling process, thereby increasing the concentration of GO and causing experimental errors, we set up six groups of parallel samples to sequentially extract the solution in

| Table 3: Isotherm pseudo parameters. |  |
|-----------------------------------|--|
| pH | Temperature (K) | \( Q_m \) (mg/g) | Langmuir | \( R^2 \) | Freundlich | \( n \) | \( R^2 \) |
| 293 K | 293 | 0.0179 | 0.979 | 21.99 | 1.096 | 0.956 |
| 4 | 303 K | 594 | 0.034 | 0.976 | 16.05 | 1.34 | 0.959 |
| 313 K | 559 | 0.030 | 0.997 | 12.67 | 1.27 | 0.983 |

| Table 4: Thermodynamic fitting parameter table. |  |
|-----------------------------------------------|--|
| \( C_0 \) (mg/L) | 293 K | \( \Delta G^0 \) (kJ·mol\(^{-1}\)) | 303 K | 313 K | \( \Delta H^0 \) (kJ·mol\(^{-1}\)) | \( \Delta S^0 \) (J·mol\(^{-1}\)·K\(^{-1}\)) |
| 40 | -4.35 | -5.43 | -6.51 | 27.18 | 0.979 | 107.66 |
| 60 | -4.52 | -5.55 | -6.58 | 25.65 | 1.044 | 103.01 |
| 80 | -4.65 | -5.85 | -7.04 | 30.34 | 1.270 | 119.47 |
| 100 | -4.96 | -5.92 | -6.88 | 23.12 | 1.270 | 95.86 |
| 120 | -5.10 | -5.96 | -6.81 | 19.96 | 1.270 | 85.55 |

**Figure 12:** Thermodynamic fitting diagram of GO adsorption by red sandstone.
each time period. After solution extraction, centrifuge the extracted solution [63]. Figure 13 shows the change of adsorption capacity over time, when GO concentration is 80 mg/L, $T = 303$ K, and pH = 4, and the red sandstone mass is 40 mg. It can be seen from Figure 13 that the adsorption rate gradually decreases over time, the adsorption equilibrium is reached at 2800 min, and the adsorption capacity is 90.03 mg/g. This is mainly because the number of effective adsorption sites on the surface of red sandstone particles is relatively fixed. As the reaction time increases, there are not enough active sites on the surface of red sandstone particles for adsorption, resulting in a gradual decrease in the adsorption rate.

At the same time, the test data were fitted by kinetic formulas (9) and (10). The fitting results are shown in Figure 13 and Table 5. It can be seen from the figure that the equilibrium adsorption capacity fitted by the pseudo-first-order kinetic model is 76.2 mg/g, while that fitted by the pseudo-second-order kinetic model is 89.88 mg/g, which is closer to the actual measured data of 90.03 mg/g. In addition, the correlation coefficients $R^2$ of the pseudo-first-order and pseudo-second-order kinetic equations are 0.969 and 0.976, respectively. Therefore, the pseudo-second-order kinetic model equation is more suitable for describing the adsorption process of GO by red sandstone, indicating that the adsorption process belongs to chemical adsorption [64].

![Figure 13: Graph of adsorption capacity over time and fitting curve of the pseudo-first-order model and pseudo-second-order model.](image)

| pH | Temperature (K) | $Q_e$ (mg/g) | $k_1$ (g/(mg·min)) | $R^2$ | $Q_e$ (mg/g) | $k_2$ (g/(mg·min)) | $R^2$ |
|----|----------------|-------------|-------------------|-------|-------------|-------------------|-------|
| 4  | 303 K          | 76.2        | 0.047             | 0.969 | 89.88       | 0.004             | 0.976 |

### 4. Conclusion

Red sandstone, GO, and GO/red sandstone were characterized by microscopic tests such as SEM, TEM, XPS, FT-IR, AFM, BET, and XRD, and the adsorption isotherm, adsorption thermodynamics, and adsorption kinetic models were combined to systematically reveal the adsorption mechanism of GO by red sandstone. The following main conclusions are drawn:

1. The experimental results show that the red sandstone has a good adsorption capacity for GO. $T = 303$ K, GO concentration 80 mg/L, pH = 4, red sandstone mass 40 mg, and equilibration time 24 h. The adsorption effect is the best. The adsorption rate was 89.08%, the adsorption capacity was 89.08 g/mg, and the partition coefficient was 11.22. The adsorption of GO on red sandstone is mainly due to the coordination reaction of Ca$^{2+}$ dissolved in the solution with C-O and O-C=O bonds, which makes GO condense on the surface of red sandstone.

2. The adsorption of GO on red sandstone conforms to the Langmuir model and belongs to monolayer adsorption. The adsorption of GO to red sandstone is endothermic, and the amount of adsorption increases with increasing temperature. The adsorption of GO on red sandstone is more in line with
the pseudo-second-order kinetic equation, which belongs to chemical adsorption. Kinetic experiments show that the adsorption equilibrium time of red sandstone is 2800 min

(3) This study did not discuss the pore size, specific surface area, and polarity of red sandstone for the adsorption capacity of GO. We will continue to explore in the next exploration

The results of this study contribute to a better understanding of the adsorption behavior of GO with natural materials in aqueous environment, which is crucial for handling GO in aqueous solution and reducing the hazard of GO in nature.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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