Removal of Pb(II) ions from aqueous solutions on few-layered graphene oxide nanosheets†

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Few-layered graphene oxide (FGO) was synthesized from graphite by using the modified Hummers method, and was characterized by scanning electron microscopy, atomic force microscopy, powder X-ray diffraction, X-ray photoelectron spectroscopy and Raman spectroscopy. The prepared FGO was used to adsorb Pb(II) ions from aqueous solutions. The abundant oxygen-containing groups on the surfaces of FGO played an important role in Pb(II) ion adsorption on FGO. The adsorption of Pb(II) ions on FGO was dependent on pH values and independent of ionic strength. The adsorption of Pb(II) ions on FGO was mainly dominated by strong surface complexation. From the adsorption isotherms, the maximum adsorption capacities (C_{max}) of Pb(II) ions on FGO calculated from the Langmuir model were about 842, 1150, and 1850 mg g^{-1} at 293, 313, and 333 K, respectively, higher than any currently reported. The FGO had the highest adsorption capacities of today’s nanomaterials. The thermodynamic parameters calculated from the temperature dependent adsorption isotherms indicated that the adsorption of Pb(II) ions on FGO was a spontaneous and endothermic process.

1. Introduction

Graphene, one or several atomic layers of graphite, possesses a special two-dimensional (2D) structure and excellent mechanical, thermal, and electrical properties. Graphene oxide can be considered as graphene oxidate. It is exfoliated by strong oxidants, such as concentrated sulfuric acid and potassium permanganate, and consists of a hexagonal network of covalently linked carbon atoms with oxygen-containing functional groups attached to various sites. The 2D structure and the lateral sheets extending to up to several micrometers gives graphene oxide a very high aspect ratio and specific surface area. Considering the basal planes decorated mostly with epoxide and hydroxyl groups, in addition to carboxyl and carboxyl groups located presumably at the edges, graphene oxide would serve as an effective adsorbent for the removal of heavy metal ions. From this viewpoint, graphene oxide is an ideal adsorbent, and the adsorption mainly occurs on its planar surface. In interface science, there is much research on the theoretical calculation of small molecule, biomolecule and metal ion adsorption on graphene. Yang et al. found that the adsorption capacity of Cu(II) on graphene oxide was 10 times higher than that of Cu(II) on active carbon. In our previous study, we found that sulfonated graphene nanosheets had an adsorption capacity of ~2.3–2.4 mmol g^{-1} for naphthalene and 1-naphthol, which was one of the highest capabilities of today’s nanomaterials for organic pollutants.

Lead ions, arising from both natural and anthropogenic sources, are detrimental to humans and living things. Long-term drinking of lead polluted water will cause serious disorders, such as anaemia, cancer, kidney disease and mental retardation. Due to their high toxicity, the elimination of Pb(II) ions from aqueous solution is crucial for the environmental pollution cleanup. Many techniques have been applied to remove Pb(II) ions such as flocculation, membrane filtration, chemical precipitation, adsorption.

Herein, we report the synthesis of few-layered graphene oxide (FGO) nanosheets from graphite and use the synthesized FGO to remove Pb(II) ions from aqueous solutions. We demonstrate that the prepared FGO has a high adsorption capability toward Pb(II) ions in aqueous solutions, which would scale up for the practical application of graphene oxide in environmental pollution cleanup if graphene oxide could be synthesized on a large scale and at a low price in the near future. The novelty of this study is that the synthesized FGO with a high adsorption capacity in the removal of Pb(II) ions from aqueous solutions is introduced and at the same time a meaningful application for graphene oxide as an effective adsorbent has been developed. The results suggested that the prepared FGO had a higher adsorption capacity than any of today’s nanomaterials in the removal of Pb(II) ions from aqueous solutions.
2. Experimental Details

2.1 Materials

Few-layered graphene oxide was prepared using the modified Hummers method from the flake graphite (average particle diameter of 20 mm, 99.95% purity, Qingdao Tianhe Graphite Co. Ltd., China). Briefly, two grams of graphite and 1.5 g of NaNO₃ (A.R.) were placed in a flask. Then, 150 mL of H₂SO₄ (A.R.) was added with stirring in an ice–water bath, and 9.0 g of KMnO₄ (A.R.) was slowly added over about 1 h. The stirring was continued for 2 h in the ice–water bath and then it was continually stirred for 5 d at room temperature. Then, 280 mL of 5 wt% H₂SO₄ was added over about 1 h with stirring with the temperature kept at 98 °C. The resultant mixture was further stirred for 2 h at 98 °C. After the temperature was reduced to 60 °C, 6 mL of H₂O₂ (30 wt%) was added in the suspension, and the mixture was stirred for 2 h at room temperature. After centrifugation at 8000 rpm, the solid phase was redispersed using vigorous stirring and bath ultrasonication for 30 min at a power of 140 W. The centrifugation and ultrasonication were recycled several times, and then the sample was rinsed with deionized water until the solution was neutral. The desired products were dried in a vacuum tank at room temperature, and thus the FGO material was derived, characterized and used in the following experiments.

2.2 Characterization of FGO

FGO was characterized by scanning electron microscopy (SEM), atomic force microscopy (AFM), powder X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), Raman spectroscopy, acid–base titration and N₂-BET surface area analysis. The AFM images were obtained in air using a Digital Instrumental Nanoscope III in tapping mode. The XRD patterns were measured on a D/max2500 with a Cu-Kα source (λ = 1.541 Å). The XPS measurements were conducted with a ESCALab 220i-XL system. Raman spectra were recorded with a Renishaw in Via Raman spectrometer (Renishaw plc, UK). All samples were prepared by dripping the FGO suspension onto a silicon wafer, followed by drying in the draught cupboard. The laser excitation was provided by a regular model laser operated at wavelength of 514 nm. The acid–base titrations were conducted using a computer controlled titration system (DL50 Automatic Titrator, Mettler Toledo) in 0.01 M NaClO₄ background electrolyte under argon conditions. The data sets of pH versus the net consumption of H⁺ or OH⁻ were used to calculate intrinsic acidity constants in diffuse-layer model with the aid of FITEQCL 3.2. The surface area was obtained with a Micromeritics ASAP 2020 M + C accelerated surface area analyzer. The sample was firstly degassed overnight at 120 °C, and then the obtained N₂ adsorption–desorption isotherms were evaluated to give the N₂-BET specific surface area (SSA).

2.3 Batch Adsorption Experiments

Analytical-grade lead nitrate was employed to prepare a Pb(II) stock solution. The batch experiments of Pb(II) adsorption on FGO were carried out in polyethylene test tubes, where the stock suspensions of FGO, Pb(II) and NaClO₄ solutions were added in the polyethylene test tubes to achieve the desired concentrations of different components. The desired pH values of the suspensions in each tube were adjusted by adding negligible volumes of 0.01 mol L⁻¹ HClO₄ or NaOH solutions. After the suspensions were shaken for 24 h to achieve adsorption equilibration, the solid phase was separated from the solution using 0.22 μm membrane filters.

The concentration of Pb(II) in the filtrate was determined by spectrophotometry at the wavelength of 616 nm using a Pb(II)–Chlorophosphonazo(III) complex. All of the experimental data were the average of duplicate determinations, and the relative errors were about 5%. The amount of Pb(II) adsorbed on FGO was calculated from the difference between the initial concentration (C₀) and the equilibrium one (Cₑ) (Adsorption% = (C₀ – Cₑ)/C₀ × 100%, where Cₑ = (C₀ – Cₘ)/m_adsorbed × V, where Cₑ is the concentration of Pb(II) adsorbed on FGO, V is the volume of the suspension, and m_adsorbed is the mass of FGO in the system).

3. Results and discussion

3.1 Characterization of FGO

Fig. 1 shows the SEM and AFM images of the synthesized FGO. It can be seen from the AFM image that the thickness of FGO is about 2.98 nm, suggesting that 2- or 3-layered graphene oxide nanosheets are formed in the modified Hummers method from the flake graphite. The FGO nanosheets have lateral dimensions of several micrometres with the small holes caused by overexposure to sonication. The SEM image shows that the FGO is partially transparent, also suggesting that 2- or 3-layered graphene oxides are formed, although the SEM image does not estimate the layer numbers of the synthesized graphene oxide exactly.

![Fig. 1](image-url) SEM image (A) and AFM image (B) of FGO.
normal graphite spacing (002) of the graphite plane, disappeared in the synthesized graphene oxide in the oxidation process. The broad and relatively weak diffraction peak at $2\theta = 10.03^\circ$ with $d = 0.87$ nm corresponding to the typical diffraction peak of FGO is attributed to the (002) plane. The $c$-axis spacing increases from 0.34 to 0.87 nm after the graphite is modified to graphene oxide nanosheets during the oxidation process, which may be due to the creation of the oxygen-containing functional groups on the surfaces of FGO,\textsuperscript{21–24} and the functional groups are evidenced by the following XPS analysis.

As is shown in Fig. 2C, the C1s XPS spectrum of FGO indicates a considerable degree of oxidation with different functional groups: the nonoxygenated ring C (284.5 eV), the C atom in C–O bond (286.2 eV), the carbonyl C (287.8 eV), and the carboxylate carbon (O–C=O) (289.0 eV).\textsuperscript{25,26} The relative contents of different functional groups from the curve fitting of the C1s XPS spectrum are showed in Fig. 2C. Compared with FGO, there are no oxygen-containing groups on the surface of graphite (Fig. 2B). The ratio of carbon to oxygen (C/O) is about 2.37 in the graphene oxide, which indicates the high oxidation of the synthesized FGO by the oxidant. The specific peak area noted in Fig. 2C shows that the main oxygen-containing groups are C–O and C=O, which are expected to form strong surface complexes with metal ions on the surfaces of FGO. The synthesized FGO also has very high dispersion properties in aqueous solutions in a graphitic 2D hexagonal lattice and D band (related to the vibrations of sp$^3$ carbon atoms of defects and disorder). The G band is associated with the vibration of sp$^2$ carbon atoms in a graphitic 2D hexagonal lattice, and the D band at around 1350 cm$^{-1}$ is related to the vibrations of sp$^2$ carbon atoms of defects and disorder. These strong bands also existed in some previous papers.\textsuperscript{27–31} The weak and broad 2D peak at 2700 cm$^{-1}$ is another indication of disorder due to an out of plane vibrational mode, and the cooperation between D and G peaks also gives rise to an S3 peak near 2950 cm$^{-1}$. Both of the 2D and S3 peaks are similar to the results from previous papers.\textsuperscript{27–31}

The N$_2$-BET surface areas of the graphite and FGO are 3 and 120 m$^2$ g$^{-1}$, respectively. Because the N$_2$-BET surface area was strongly related to the degassed temperature, the samples were degassed overnight at 120 °C for SSA measurements. Since FGO powders tend to aggregate together, the SSA is normally lower than the theoretical value, and gas adsorption gives a much lower SSA value than that of liquid adsorption (dispersed in solvent, the
stable colloidal suspensions of FGO.\textsuperscript{32} It is reasonable that the N$_2$-BET surface area of FGO is relatively lower. From the acid-base titration results, the pH$_{pc}$ (point of zero charge) value of 2- or 3-layered graphene is 4.1, and that of FGO is 3.8. The surface charge is positive at pH < pH$_{pc}$, and is negative at pH > pH$_{pc}$. The surface site densities are $2.36 \times 10^{-3}$ mol g$^{-1}$ for FGO and $1.11 \times 10^{-3}$ mol g$^{-1}$ for 2- or 3-layered graphene. One can see that the surface site density of FGO is much higher than that of 2- or 3-layered graphene. The surface site density of FGO is about five times higher than that of oxidized multiwalled carbon nanotubes (MWCNTs) ($4.2 \times 10^{-4}$ mol g$^{-1}$).\textsuperscript{30}

### 3.2 Adsorption of Pb(II) on FGO

#### 3.2.1 Effect of pH and ionic strength

The adsorption of Pb(II) ions on FGO was investigated at different pH values. As shown in Fig. 4, the adsorption of Pb(II) ions on FGO increases with the pH increasing from 1 to 8, and then decreases with increasing pH values at pH > 8. It is necessary to note that the pH in Fig. 4 corresponds to the pH of the system after reaching adsorption equilibrium. The initial and final pH values are listed in Table 1. One can see that the final pH values are lower than the initial pH values at the initial pH values lower than 9.50. The oxygen-containing functional groups on the surface of FGO are deprotonated during the adsorption process, and part of the H$^+$ ions are released to the solution. The adsorption of Pb(II) is partly attributed to ion exchange of Pb$^{2+}$ with –H on FGO surfaces, and the exchanged H$^+$ ions are released to solution thereby resulting in the decrease of pH values. From the hydrolysis constants of Pb(II) ions,\textsuperscript{31} the Pb(II) ions present in the species of Pb$^{2+}$, Pb(OH)$^+$, Pb(OH)$_2^-$, Pb(OH)$_3^-$, at different pH values. At low pH values, the main species of Pb(II) ions are Pb$^{2+}$ and Pb(OH)$^+$, while at high pH values, the predominant Pb(II) species is Pb(OH)$_2^-$, which is difficult to be adsorbed on FGO due to the negative surface charge of FGO. At high pH values, the functional groups are progressively deprotonated to form a negative surface charge, and thereby the negatively charged Pb(II) species is difficult to adsorb on the negative charged surfaces of FGO.

The desorption of Pb(II) from FGO was also investigated at different pH values. After the sorption equilibrium, half of the supernatant was pipetted out and an equal volume of background electrolyte solution with the same pH value was added. Then the mixture was shaken and centrifugation was done under the same conditions as in the sorption experiments. The results are listed in Table S1 (ESI$^+\text{).}$ Part of Pb(II) can be desorbed from FGO at low pH values, and the desorption percentage of Pb(II) from FGO decreases with increasing pH values, which is attributed to the physical sorption of Pb(II) at low pH and strong surface complexation at high pH values.

It can also be seen from Fig. 4 that the adsorption of Pb(II) ions on FGO is not affected by NaClO$_4$ concentrations obviously at pH ranging from 1 to 13 under the experimental conditions. The adsorption of Pb(II) ions on FGO is strongly dependent on pH values and independent of ionic strength. The adsorption of Pb(II) ions on FGO is mainly dominated by strong inner-sphere surface complexation rather than by outer-sphere surface complexation.\textsuperscript{49}

From the precipitation constant of Pb(OH)$_2$(s) $(1.2 \times 10^{-19})$, at a Pb(II) initial concentration of 120 mg L$^{-1}$, Pb(II) ions begin to form a precipitation at pH ~8.1. However, more than 98% Pb(II) ions are adsorbed on FGO at pH 7. Thereby it is impossible to form a precipitation because of the very low concentration of Pb(II) ions remaining in aqueous solution. Therefore, the adsorption of Pb(II) ions on FGO is not attributed to the precipitation of Pb(OH)$_2$(s) at high pH values. The high adsorption of Pb(II) ions on FGO is dominated by the strong surface complexation of Pb(II) ions with the oxygen-containing groups on the surfaces of FGO.

#### 3.2.2 Adsorption isotherms

The adsorption isotherms of Pb(II) ions on FGO at $T = 293$, 313, and 333 K are shown in Fig. 5A. The adsorption isotherm is the highest at $T = 333$ K and is the lowest at $T = 293$ K, indicating that the adsorption of Pb(II) ions on FGO is promoted at higher temperatures. Herein, the widely used Langmuir and Freundlich isotherm models are applied to simulate and understand the adsorption mechanism. The Langmuir and Freundlich isotherm models are expressed as:\textsuperscript{32,34}

\begin{equation}
C_s = \frac{C_{max} \cdot K_T \cdot C_e}{1 + K_T \cdot C_e}
\end{equation}

\begin{equation}
C_e = K_F \cdot C_{eq}^{1/n}
\end{equation}

where $C_s$ is the equilibrium concentration of Pb(II) ions in aqueous solution (mg L$^{-1}$), $C_e$ is the amount of Pb(II) ions adsorbed on FGO (mg g$^{-1}$), $C_{max}$ is the maximum amount of Pb(II) ions adsorbed per unit weight of FGO to form complete monolayer coverage on the surface at high equilibrium Pb(II) concentration, $K_T$ represents the enthalpy of adsorption and should vary with temperature, $K_F$ and $n$ are Freundlich constants related to the adsorption capacity and adsorption intensity, respectively. The Langmuir and Freundlich isotherm parameters calculated from the adsorption isotherms are listed in Table 2. The high values of the correlation coefficients indicate that the Langmuir equation

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**Table 1** The initial pH and final pH in the adsorption system of Pb(II) onto FGO. $T = 293 \pm 1$ K, $[\text{Pb}^{2+}]_{\text{initial}} = 120.0$ mg L$^{-1}$, $m \cdot V^{-1} = 0.1$ g L$^{-1}$, $[\text{NaClO}_4] = 0.01$ mol L$^{-1}$.

| Initial pH | 1.20 | 2.50 | 3.10 | 4.40 | 5.44 | 6.00 | 6.54 | 7.47 | 8.71 | 9.50 | 10.05 | 10.88 | 11.32 | 11.56 | 12.11 | 12.57 |
| Final pH  | 1.19 | 2.43 | 3.07 | 4.31 | 4.86 | 5.95 | 6.05 | 6.24 | 6.89 | 7.61 | 8.71 | 9.50 | 10.05 | 10.88 | 11.32 | 11.56 | 12.11 | 12.57 |
The adsorption of Pb(II) ions on FGO is strongly dependent on temperature. Comparing to other kinds of adsorbents, the adsorption capacity of Pb(II) ions on FGO is the highest of the considered methods at 293 K. The adsorption isotherm of Pb(II) ions on FGO is much higher than that of Pb(II) ions on the 2- or 3-layered graphene. The maximum adsorption capacity ($C_{\text{max}}$) of Pb(II) ions on the 2- or 3-layered graphene is about 400 mg g$^{-1}$, which is about half of that of Pb(II) ions on FGO at the same experimental conditions. The difference of FGO and the 2- or 3-layered graphene is that there are abundant oxygen-containing functional groups on FGO. From the acid-base titration, the surface site density of FGO is 2.36 $\times$ 10$^{-3}$ mol g$^{-1}$ and that of 2- or 3-layered graphene is 1.11 $\times$ 10$^{-3}$ mol g$^{-1}$. The surface site density of FGO is about two times of that of 2- or 3-layered graphene. The enhanced adsorption of Pb(II) ions on FGO is attributed to the oxygen-containing functional groups, which form strong surface complexes with Pb(II) ions on FGO.

The adsorption isotherms of Pb(II) ions on FGO at three temperatures, pH = 6.0 $\pm$ 0.1, C[NaClO$_4$] = 0.01 mol L$^{-1}$, m V$^{-1}$ = 0.1 g L$^{-1}$; (B) adsorption isotherms of Pb(II) ions on FGO and on 2- or 3-layered graphene at $T$ = 293 K, solid points: FGO; open points: 2- or 3-layered graphene; pH = 6.0 $\pm$ 0.1, C[NaClO$_4$] = 0.01 mol L$^{-1}$, m V$^{-1}$ = 0.1 g L$^{-1}$.

Table 2: Adsorption constants for Langmuir and Freundlich isotherm models

<table>
<thead>
<tr>
<th>Adsorbents</th>
<th>$C_{\text{max}}$ (mg g$^{-1}$)</th>
<th>$K_L$</th>
<th>$R^2$</th>
<th>$K_F$</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Langmuir</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>293 K</td>
<td>842</td>
<td>0.36</td>
<td>0.985</td>
<td>5.33</td>
<td>0.906</td>
</tr>
<tr>
<td>313 K</td>
<td>1150</td>
<td>0.37</td>
<td>0.986</td>
<td>5.86</td>
<td>0.978</td>
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<tr>
<td>333 K</td>
<td>1850</td>
<td>0.29</td>
<td>0.993</td>
<td>4.72</td>
<td>0.957</td>
</tr>
<tr>
<td>Freundlich</td>
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Table 3: Maximum adsorption capacity of Pb(II) ions on various adsorbents

<table>
<thead>
<tr>
<th>Adsorbents</th>
<th>$C_{\text{max}}$ (mg g$^{-1}$)</th>
<th>Conditions</th>
<th>References</th>
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<td>Activated carbon</td>
<td>21.80</td>
<td>pH 6.0, T = 303 K</td>
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<tr>
<td>Sawdust</td>
<td>34.25</td>
<td>pH 7.0, T = 303 K</td>
<td>[36]</td>
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<td>Iron oxide</td>
<td>36.00</td>
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<td>[37]</td>
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<td>GMZ bentonite</td>
<td>23.83</td>
<td>pH 5.2, T = 293 K</td>
<td>[38]</td>
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<td>Hazelnut shell</td>
<td>28.18</td>
<td>pH 6.0, T = 298 K</td>
<td>[39]</td>
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<tr>
<td>Oxidized MWCNTs</td>
<td>2.06</td>
<td>pH 6.0, T = 293 K</td>
<td>[40]</td>
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<tr>
<td>Functionalized graphene</td>
<td>406.6</td>
<td>pH 5.1, T = 293 K</td>
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<tr>
<td>FGO</td>
<td>842</td>
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<td>This work</td>
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<tr>
<td>2- or 3-layered graphene</td>
<td>400</td>
<td>pH 6.0, T = 293 K</td>
<td>This work</td>
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</table>

The inferred formation of the inner-sphere surface complexes of Pb(II) ions on FGO can be described in the schematic diagram (Fig. 7). Considering the electronic configuration of Pb$^{2+}$, 1s$^2$ 2s$^2$ 2p$^6$ 3s$^2$ 3p$^6$ 4s$^2$ 3d$^{10}$ 4p$^6$ 5s$^2$ 4d$^{10}$ 5p$^6$ 6s$^2$ 4f$^{14}$ 5d$^{10}$ 6p$^2$, the proper coordination number of Pb$^{2+}$ is 3. The number of the valence electrons on the outer valence orbit is 8, which is in good agreement with the octet rule.
Fig. 6  (A) FIR spectra of FGO before and after adsorption of Pb(II) ions. (B) The $k^2$-weighted $\chi(k)$ spectra of Pb(II) adsorbed on FG and FGO samples; and (C) the corresponding Fourier transformed radial distribution functions of Pb(II) adsorbed on FG and FGO samples.

with the octet stability theory. At higher pH values, the adsorbed species include Pb(OH)$^+$, which is also shown in Fig. 7. What’s more, the abundant oxygen-containing groups on the surfaces of FGO make the 2 or 3 adjacent oxygen atoms available to bind Pb(II) ions. The special planar structure of FGO and the high available surface area of FGO in the FGO suspension also provide enough sites for the adsorption of Pb(II) ions. From the XRD analysis, the $c$-axis spacing of FGO is 0.87 nm, it is enough for the Pb$^{2+}$ ions ($r \sim 0.48$ nm) to enter into the interlayer space of FGO. In the system, the graphene is the Lewis base and the Pb(II) ion is the Lewis acid. The delocalized $\pi$ electron systems of graphene layer can act as Lewis base and form electron donor–acceptor complexes with Pb(II) ions. A complex between the graphene and Pb(II) ion occurs through Lewis acid–base interaction, which also contributes to Pb(II) ion adsorption on graphene or FGO. From the interpretation mentioned above, the strong interaction between Pb(II) ions and FGO is reasonable and the high-efficiency removal of Pb(II) ions from large volumes of aqueous solutions is understandable.

3.2.3 Thermodynamic analysis. The thermodynamic parameters ($\Delta H^o$, $\Delta S^o$, and $\Delta G^o$) for Pb(II) adsorption on FGO can be obtained from the temperature dependent adsorption isotherms. The average standard enthalpy change ($\Delta H^o$) and standard entropy change ($\Delta S^o$) can be calculated from the slope and $y$-intercept of plot of $\ln K_d$ versus $1/T$ (Fig. 8) using the van’t Hoff equation:

$$\ln K_d = -\frac{\Delta H^o}{RT} + \frac{\Delta S^o}{R}$$

where $K_d$ is the adsorption equilibrium constant.

![Fig. 7 A schematic diagram of the formation of the surface complexes of Pb(II) ions on the surfaces of FGO.](image1)

![Fig. 8 Linear plots of $\ln K_d$ vs. $C_e$ for the adsorption of Pb(II) ions on FGO at $T = 293, 313, 333$ K, pH $= 6.0 \pm 0.1$, $C[NaClO_4] = 0.01$ mol L$^{-1}$, $m$ V$^{-1} = 0.1$ g L$^{-1}$.](image2)

The free energy change ($\Delta G^o$) is calculated by the following equation:

$$\Delta G^o = \Delta H^o - T\Delta S^o$$

Table 4 lists the thermodynamic parameters calculated from the adsorption isotherms at pH 6.0 at three different temperatures.
The positive values of $\Delta H^0$ indicate that the adsorption of Pb(II) ions on FGO is an endothermic process. The interpretation to the endothermicity of the enthalphy of Pb(II) adsorption is that Pb(II) ions are well solvated in water. In order for the Pb(II) ions to adsorb on FGO, they are to some extent denuded of their hydration sheath, and this dehydration process needs energy. The energy of dehydration exceeds the exothermicity of the ions attaching to FGO surface. The removal of water molecules from Pb(II) ions in aqueous solutions and may suggest some structure values of the entropy change reflect the affinity of FGO toward adsorption process is spontaneous with high affinity. The positive values of $\Delta H^0$ with the increase of temperature indicates more efficient adsorption at higher temperatures. At higher temperatures, ions are readily dehydrated, and therefore their adsorption becomes more favorable. The positive entropy change also indicates that the adsorption process is spontaneous with high affinity. The positive values of the entropy change reflect the affinity of FGO toward Pb(II) ions in aqueous solutions and may suggest some structure changes in the adsorbents.

### Table 4 The thermodynamic parameters of Pb(II) ion adsorption on FGO

<table>
<thead>
<tr>
<th>$C_0$ (mg L$^{-1}$)</th>
<th>$\Delta H^0$ (kJ mol$^{-1}$)</th>
<th>$\Delta S^0$ (J mol K$^{-1}$)</th>
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<td>293 K</td>
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<td>160</td>
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### Acknowledgements

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### References