

## Adsorptive removal of lead ion from aqueous solution by activated carbon/iron oxide magnetic composite

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The adsorption features of activated carbon and the magnetic properties of iron oxides have been combined in a composite to produce magnetic adsorbents. These magnetic particles can be used as adsorbent for the removal of heavy metals such as lead from aqueous solution and can subsequently be removed from the medium by a simple magnetic procedure. The prepared samples have been characterized by SEM, N<sub>2</sub> adsorption-desorption and powder XRD. The results show that the presence of iron oxides caused a decrease in the surface area and microporous volume of the activated carbon. The effects of initial solution pH, contact time, adsorbent dosage and initial concentration of lead ion on the adsorption behaviour of lead ion onto the synthetic magnetic adsorbent have been investigated. Kinetic studies show that the overall adsorption rate of lead ion is illustrated well by the pseudo-second-order kinetic model. The applicability of the Langmuir and Freundlich models for the lead ion adsorption data is tested. The results show that the Langmuir equation gives a better fit than Freundlich equation to the adsorption isotherms. Moreover, the prepared composite exhibits paramagnetic behaviours and could be easily recovered from the medium by a simple magnetic process. It can therefore be potentially applied for the treatment of water contaminated by heavy metal pollutants.

**Keywords:** Activated carbon, Iron oxide, Adsorption, Magnetic composite, lead ion

Heavy metals are generally recognized to be a threat toward humans and ecosystems because of their high potential toxicity. They could not be biologically decomposed into harmless materials and, to matters worse, were accumulated in the organisms<sup>1</sup>. Lead, as well as mercury, cadmium, chromium and arsenic, is in the group of serious hazardous heavy metals. Drinking those that contain Pb<sup>2+</sup> for a long term, even if in a very low concentration, could lead to a wide range of spectrum health problems, such as renal failure, coma, nausea, cancer, convulsions and subtle effects on metabolism and intelligence<sup>2,3</sup>. Several methods have evolved over the years on the removal of heavy metals present in water and wastewater. These are chemical

precipitations, conventional coagulation, reverse osmosis, ion exchange and adsorption. One of which, adsorption method, is simple and cost-effective, thus has been widely used<sup>4-7</sup>. Among various adsorbents, adsorption onto activated carbon has proven to be one of the most effective and reliable physicochemical treatment methodologies<sup>8-11</sup>. Due to its high surface area and porous structure it can efficiently adsorb gases and compounds dispersed or dissolved in liquids<sup>12</sup>. The adsorption of several organic contaminants in water, such as pesticides, phenols and chlorophenols, has recently been reported<sup>13-15</sup>. Moreover, activated carbon can easily be functionalized and used as an efficient adsorbent for heavy metal cationic contaminants<sup>16</sup>. However, the application of activated carbon powders in water treatment system is limited because it is difficult to separate after the treatment process and reuse the tiny particles. The application of magnetic particle technology is one of the choices for field application of the activated carbon adsorbent.

The application of magnetic particle technology to solve environmental problems has received considerable attention in recent years<sup>17-19</sup>. Magnetic particles can be used to adsorb contaminants from aqueous or gaseous effluents, and after adsorption can be separated from the medium by a simple magnetic process.

The objectives of present study were (a) to prepare a high surface area and high adsorption capacity magnetic composite based on activated carbon/iron oxide; (b) to investigate the adsorption of lead ion on the prepared magnetic composite; (c) to study the effect of different parameters such as contact time, initial pH, adsorbent dosage and initial Pb<sup>2+</sup> concentration on adsorption process; (d) to find optimum adsorption isotherm as well as the rate of adsorption kinetics. The regeneration of activated carbon is one of key steps for practical applications. The separation problem of the prepared adsorbent has been solved, after adsorption, the magnetic composite can be separated from the medium by a simple magnetic process, so the prepared adsorbent can be reused easily.

### Experimental Section

#### Chemicals

All the chemicals used in the study were of analytical grade. All the solutions in the study were

prepared using de-ionized water. All glasswares were cleaned by rinsing with hydroxylamine hydrochloride, soaking in 10% HCl, and rinsing with de-ionized water.

The activated carbon (AC) (AC12×40, China Calgon) was used in this study. This kind of AC has moisture content of 1.2%, ash content of 10.3%, iodine values of AC adsorption of 1029 mg/g, the hardness of 96.2%, and the density of 480 g/L. Grain sizes of AC were: less than 1.7 mm in diameter and more than 0.425 mm in diameter. The virgin activated carbon was firstly rinsed with de-ionized water to remove dirties, and then was washed by 0.01 mol·L<sup>-1</sup> HCl solution to remove all salts precipitated in its pores. Then, the AC was repeatedly washed with de-ionized water to remove all traces of the acid. Subsequently, the washed activated carbon was modified by 10% HNO<sub>3</sub> for 12 h at the room temperature and repeatedly washed with de-ionized water, finally, the modified activated carbon was oven-dried at 85°C for 24 h to volatilize the organic impurities.

#### Adsorbent synthesis

The composite adsorbent used in this study was synthesized using a slightly modified procedure from that reported in the literature<sup>18</sup>. Briefly, the composite adsorbent was prepared from a suspension of the modified activated carbon in a 400 mL solution of FeCl<sub>3</sub> (7.8 g, 28 mmol) and FeSO<sub>4</sub> (3.9 g, 14 mmol) at 70°C. NaOH solution (100 mL, 5 mol/L) was added dropwise to precipitate the iron oxides. Later, the obtained material was washed with de-ionized water until rinsing water became neutral, then the adsorbent was dried in an oven at 100°C for 8 h and finally stored in polystyrene bottles for further usage. To determine the Fe(III) loading quality of composite adsorbent, about 0.5 g of the composite was calcined at 600°C and then digested with 25 mL of concentrated hydrochloric acid. The digestion solutions were analyzed for Fe(III) by a Shimadzu Atomic Absorption spectrophotometer (AA-6601F) unit with flame atomization. The composition of the prepared samples was determined to be iron oxide: activated carbon = 8.2:91.8.

#### Characterization

The BET specific surface area and pore volumes of adsorbent before and after loading iron oxide were obtained by the cumulative adsorption of nitrogen at 77K using a Micromeritics 2000 instrument (ASAP

2000, Micromeritics, USA). The morphologies of activated carbon/iron oxide composite and activated carbon were examined by a scanning electron microscope (SEM, Holland Philips, JSM-5800). X-ray diffraction pattern was taken from a Cu target X-ray diffractometer (Rigaku D/max-r B).

#### Batch adsorption experiments

A batch technique was used to investigate Pb<sup>2+</sup> adsorption. Batch experiments included: the kinetic studies, adsorption isotherms and some operating parameters.

The adsorption capacities of activated carbon and activated carbon/iron oxide composite were determined by batch adsorption isotherms at room temperature (20±1°C) in aqueous solution. Lead ion was hardly adsorbed onto any of the adsorbents in solutions having values of pH less than 3, at values of pH higher than 7, the adsorption experiments failed as a result of precipitation of lead hydroxide. Thus, in adsorption experiments we fixed the pH values of the initial solution at 4, 5 and 6. In several glass vials, 100 mL of solution containing various Pb<sup>2+</sup> concentrations (200, 400, 600, 800, 1000 mg/L) were contacted with 5.0 g of composite adsorbent. The vials were placed in a water bath at 20°C and shaken at 150 r/min for approximately 24 h to ensure equilibrium was reached, and the pH was adjusted by adding 0.1 mol/L NaOH or HNO<sub>3</sub> until it remained constant (±0.10). Then, each solution was filtered with a polycarbonate membrane of 0.22 µm of pore diameter, and the filtrates were analyzed for Pb<sup>2+</sup> equilibrium concentration by an atomic fluorescence spectrometer (AFS) (PS Analytical Ltd., Kent, UK). The amount of lead ion adsorbed was calculated with the following equation:

$$q_e = V(c_0 - c_e) / W \quad \dots (1)$$

where  $q_e$  is the amount adsorbed (mg/g);  $c_0$  and  $c_e$  are the initial and equilibrium lead ion concentrations in the solution (mg/L), respectively;  $V$  is the solution volume (L); and  $W$  is the mass of adsorbent (g).

The adsorption kinetic study was performed for Pb<sup>2+</sup> in solution at pH 6.0 and room temperature (20±1°C). Several glass vials were used to hold 100 mL Pb<sup>2+</sup> solution of known initial concentration (200, 400, and 600 mg/L) and 5.0 g/L of composite adsorbent, and shaken at 150 r/min for a duration ranging from 0 to 480 min. At certain period of time, each vial was removed from the shaker, and the

solution was then filtered with a polycarbonate membrane of 0.22  $\mu\text{m}$  of pore diameter. The filtrates were analyzed for residual  $\text{Pb}^{2+}$  concentration.

To determine the effects of different parameters on  $\text{Pb}^{2+}$  adsorption, experiments were performed at various initial  $\text{pH}$ , ranging between 3.5 and 6.5. Initial  $\text{Pb}^{2+}$  concentration of 100 mg/L and composite adsorbent dosage 5.0 g/L were employed. The effects of adsorbent dosage and contact time were conducted.

## Results and Discussion

### Characterization of adsorbents

The microstructure changes of pure iron oxide, pure activated carbon and activated carbon/iron oxide composite adsorbent were listed in Table 1. As shown in Table 1, the deposited iron oxide contributes to a decrease in the  $\text{N}_2$ -BET surface area, total pore volume and average pore diameter. As iron oxide has a relatively small surface area and microporous volume (63.7  $\text{m}^2/\text{g}$  and 0.009  $\text{cm}^3/\text{g}$ , respectively) its presence in the composites should cause a decrease in the surface area and microporous volume compared to pure activated carbon.

Figure 1 shows the SEM micrographs of activated carbon and composite adsorbent. It can be found from Fig. 1 that there are a few macropores in activated carbon, and small aggregates are observed from the general view of the composite, which appear brighter, supported on the darker surface of the activated carbon.

To obtain information on the crystal structure of the prepared composite adsorbent, X-ray diffraction patterns were measured. The XRD patterns of pure iron oxide and composite adsorbent were shown in Fig. 2. XRD analyses of pure iron oxide suggest the presence of a cubic iron oxide phase, which may be related to the presence of maghemite ( $\gamma\text{-Fe}_2\text{O}_3$ ) and magnetite ( $\text{Fe}_3\text{O}_4$ ). So the prepared iron oxides are magnetic. For the composite the iron oxide maintained cubic spinel structure. This illuminated that the magnetic properties of iron oxide were basically invariable, which makes the composite adsorbent can be separated more easily by an applied

Table 1 — Microstructure of pure iron oxide, pure activated carbon and activated carbon/iron oxide composite

Sample	$S_{\text{BET}}$ ( $\text{m}^2/\text{g}$ )	Average pore diameter(nm)	Total pore volume ( $\text{cm}^3/\text{g}$ )
Iron oxide	62.8	0.926	0.009
Activated carbon	1022.6	1.859	0.861
Composite adsorbent	678.3	1.688	0.632

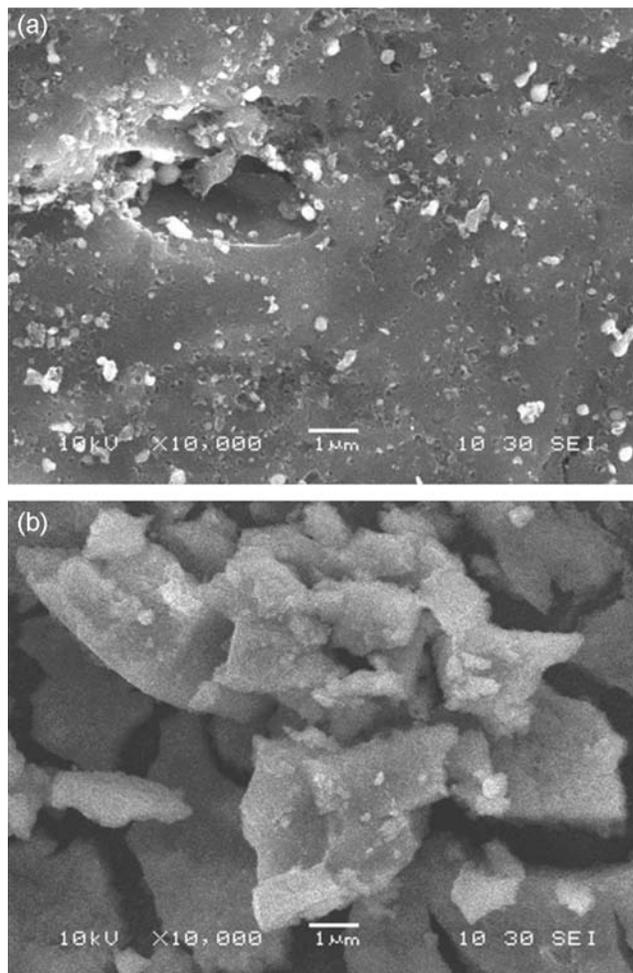


Fig. 1 — SEM micrographs of activated carbon (a), activated carbon/iron oxide composite (b).

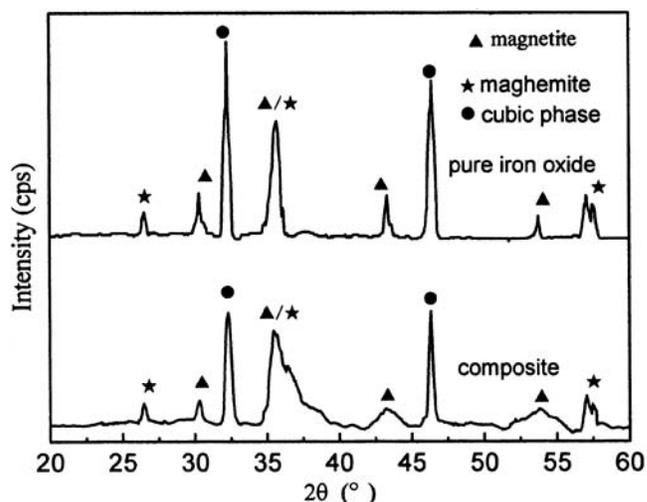


Fig. 2 — Powder XRD for pure iron oxide and activated carbon/iron oxide composite.

magnetic filter. It could also be seen from Fig. 2 that the peaks of cubic iron oxide phase in the composite appear broader, suggesting a smaller crystallite size.

#### **Effect of initial pH on adsorption process**

The comparison result on the uptake of  $\text{Pb}^{2+}$  by pure activated carbon and activated carbon/iron oxide composite showed that the removal rate of  $\text{Pb}^{2+}$  is 86.26% and 92.35%, respectively. It can be seen that the adsorption capacity of the composite is higher than that of pure activated carbon. Moreover, the separation problem of the prepared adsorbent has been solved, after adsorption, the magnetic composite can be separated from the medium by a simple magnetic process, and the prepared adsorbent can be reused easily. So the performances of the composite adsorbent were investigated in this study.

The experimental results were obtained from lead ion adsorption onto activated carbon/iron oxide under different initial pH conditions. Activated carbon/iron oxide dosage (5.0 g/L), lead ion concentration (100 mg/L) and agitation speed (150 r/min) were kept constant in all experiments. The lead ion adsorbed by activated carbon/iron oxide was sensitive to pH variation over the examined range of 3.5 to 6.5. The adsorption amount increased with increasing pH to reach a maximum at pH 6.0. At  $\text{pH} < 6.0$ ,  $\text{H}^+$  ions compete with lead ions for the surface of the adsorbent, which would restrict the approach of lead ions due to the repulsion<sup>20</sup>. Hence, the metal removal is the lower amount presumably due to the enhanced competition of proton with lead ions for ligand binding sites and complex formation. The condition of  $\text{pH} > 6.0$ , noted in adsorption decrease, may be attributed to precipitation of the lead ions as hydroxides<sup>20</sup>. For this reason, the optimal pH value was selected to be 6.0.

#### **Effect of contact time on adsorption process**

Contact time is one of the effective factors in batch adsorption process. The effect of contact time on lead ion adsorption efficiency was studied in this work. The experimental results showed that the adsorption was very fast and equilibrium between the aqueous solution and activated carbon/iron oxide was established within about 1 h. There was no significant change in lead ion removal rates after 1 h up to 3 h. Based on the result, 1 h was taken as the equilibrium time in adsorption experiments. The removal of lead ion from aqueous solutions by adsorption on activated carbon/iron oxide increases with contact time, till the equilibrium is attained.

#### **Effect of adsorbent dosage on adsorption process**

The effect of adsorbent dosage on percentage adsorption of  $\text{Pb}^{2+}$  was studied. The increase in adsorbent dosage from 1.0 to 5.0 g/L resulted in an increase from 25.8 to 89.7% in adsorption of  $\text{Pb}^{2+}$ . This may be due to the availability of more and more adsorption sites for  $\text{Pb}^{2+}$  adsorption during the adsorption reaction. A further increase in adsorbent dosage ( $> 5.0$  g/L) did not cause significant improvement in  $\text{Pb}^{2+}$  adsorption. This may be due to the adsorption of almost all  $\text{Pb}^{2+}$  to the adsorbent and the establishment of equilibrium between the  $\text{Pb}^{2+}$  molecules adsorbed to the adsorbent and those remaining unadsorbed in the solution<sup>21</sup>. Thus 5.0 g/L of activated carbon/iron oxide was chosen for next study.

#### **Effect of initial lead ion concentration on adsorption process**

Initial concentration is one of the effective factors on adsorption efficiency. The experiments were carried out with variable initial lead ion concentrations (50, 100, 150 and 200 mg/L) and constant temperature (20°C), pH (6.0), contact time (1 h) and adsorbent dosage (5.0 g/L). It can be observed from the experimental results that the  $\text{Pb}^{2+}$  removal rate decreased with the increase of initial  $\text{Pb}^{2+}$  concentration, the percentage adsorption of  $\text{Pb}^{2+}$  on activated carbon/iron oxide decreased from 99.02 to 66.78% as the initial  $\text{Pb}^{2+}$  concentration was increased from 50 to 200 mg/L. At lower  $\text{Pb}^{2+}$  concentrations, the ratio of the available adsorption sites of adsorbent to the initial number of  $\text{Pb}^{2+}$  molecules is large and subsequently the fractional adsorption becomes independent of initial concentration. However, at higher concentrations, the available sites of adsorption become fewer, and hence the percentage removal of  $\text{Pb}^{2+}$  which depends upon the initial concentration, decreases<sup>22</sup>.

#### **Adsorption Isotherms**

In the sorption technology, it is utmost essential to determine an adsorption, as it shows how the adsorption molecules are distributed in the liquid phase and with the adsorbent. For the design purpose, it is necessary to fit the isotherm data with the models. Langmuir and Freundlich isotherm equations are most widely used for the equilibrium study. Lead ion adsorption isotherm obtained for modified AC composite adsorbent was shown in Fig. 3. These isotherms represent the adsorption behavior of lead ion on the adsorbent as a function of increasing aqueous lead concentration for a contact time of 24 h

at  $pH=4.0, 5.0$  and  $6.0$ . These isotherms showed that the adsorption capacity increases with increasing equilibrium concentration of lead ion.

The results of lead ion adsorption on adsorbent (Fig. 3) were analyzed by using the Langmuir model to evaluate parameters associated to the adsorption behavior. The linear form of Langmuir equation at a given temperature is represented by:

$$q_e = q_m \cdot b \cdot c_e / (1 + b \cdot c_e) \quad \dots (2)$$

where  $c_e$  is the aqueous phase ion equilibrium concentration (mg/L),  $q_e$  is the amount of lead ion sorbed onto 1 g of the considered adsorbent (mg/g),  $b$  is the adsorption constant (L/mg) related to the energy of adsorption and represents the affinity between the adsorbent and adsorbate,  $q_m$  is the maximum adsorption capacity (mg/g).

Equation (2) can be rearranged to obtain:

$$c_e/q_e = 1/(b \cdot q_m) + c_e/q_m \quad \dots (3)$$

Experimental isotherm data acquired at different  $pH$  fit to the linear form of the Langmuir equation can be obtained by plotting of  $c_e/q_e$  against  $c_e$ . This linear plot was employed to obtain the values of  $q_m$  and  $b$  from the slope and intercept of the plot, the isotherm parameters related to the model obtained from the experimental data are listed in Table 2. It can be seen that both  $q_m$  and  $b$  increase with increasing  $pH$  from 4.0 to 6.0 (Table 2). High values of  $b$  are reflected in

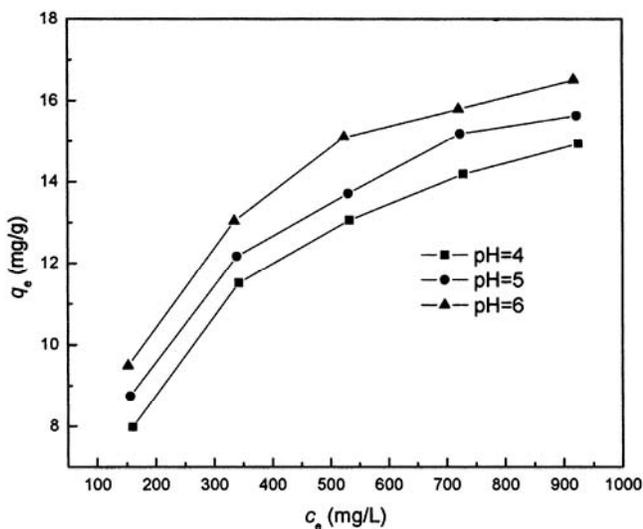


Fig. 3 — Adsorption isotherms of lead ions by activated carbon/iron oxide composite. (adsorbent dosage=5.0 g/L, adsorption time=24 h, agitation speed=150 r/min)

the steep initial slope of an adsorption isotherm, indicating desirable high affinity. Therefore, activated carbon/iron oxide composite adsorbent performed well in lead adsorption at  $pH 6.0$  compared to other  $pH$  values examined.

The Freundlich isotherm model is also used to analyze the results of lead ion adsorption on activated carbon/iron oxide adsorbent (Fig. 3). The Freundlich model can be expressed by the following equation:

$$q_e = k_f \cdot c_e^{1/n} \quad \dots (4)$$

where  $k_f$  and  $n$  are constants related to the adsorption capacity and affinity, respectively. The equation is conveniently used in the linear form by taking the logarithm of both sides as:

$$\lg q_e = \lg k_f + (1/n) \lg c_e \quad \dots (5)$$

The adsorption data fit to the linear form of the Freundlich equation can be obtained by plotting of  $\lg q_e$  against  $\lg c_e$ . Experimental isotherm data acquired at different  $pH$  were fitted with the Freundlich model and the isotherm parameters related to the model obtained from the experimental data were listed in Table 2.

The data showed that the  $k_f$  constant is increased with the increase of  $pH$  values, at  $pH 6.0$ ,  $k_f$  reached its corresponding maximum value, and  $1/n$  value at  $pH 6.0$  is smaller than that at other  $pH$  values. These imply that the binding capacity reaches the highest value and the affinity between the adsorbent and lead ions is also higher than other  $pH$  values. The correlation coefficients ( $R^2$ ) given in Table 2 also show that the Langmuir equation gives a better fit than Freundlich equation to the adsorption isotherms.

#### Kinetic study

In order to obtain the adsorption kinetic information of lead ion on the activated carbon/iron oxide adsorbent, the change of lead ion concentration with adsorption time was recorded for an initial concentration of 200, 400, 600 mg/L and a fixed  $pH$

Table 2 — The parameters of Langmuir and Freundlich equation

Initial $pH$	Langmuir equation			Freundlich equation		
	$q_m$ (mg·g <sup>-1</sup> )	$b$ (L·mg <sup>-1</sup> )	$R^2$	$1/n$	$K_f$	$R^2$
4.0	18.21	$4.884 \times 10^{-3}$	0.9997	0.35794	1.340	0.9565
5.0	18.82	$5.411 \times 10^{-3}$	0.9986	0.32573	1.749	0.9583
6.0	19.37	$6.315 \times 10^{-3}$	0.9994	0.3106	2.065	0.9584

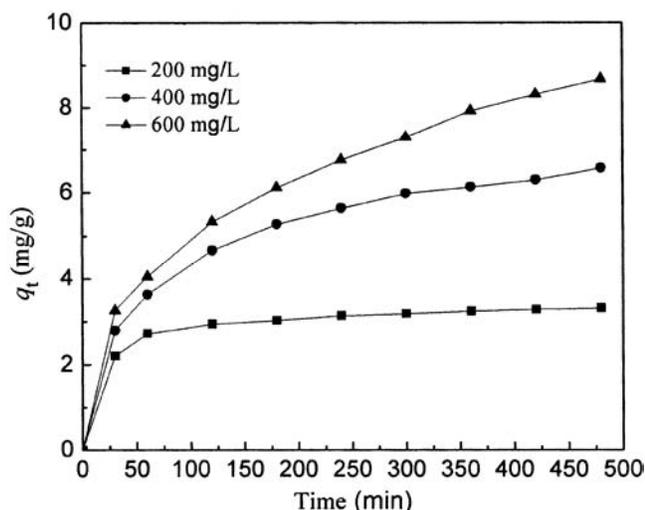


Fig. 4 — Adsorption kinetics of lead ions by activated carbon/iron oxide composite. (solution pH=6.0, agitation speed=150 r/min, adsorbent dosage=5.0 g/L)

solution of 6.0, adsorbent dosage 5.0 g/L (Fig. 4). It is evident from Fig. 4 that the adsorption of lead ion increases rapidly with time as well as with the increase of the initial ion concentrations. However, the time to reach the adsorption equilibrium takes longer with an increase in the concentration.

To investigate the mechanism of adsorption, the pseudo-second-order kinetic model was applied to experimental data. The pseudo-second-order kinetic equation can be derived as<sup>23</sup>:

$$dq/dt = k_2(q_e - q_t)^2 \quad \dots (6)$$

Separating the variables in equation (6) gives

$$-d(q_e - q_t)/(q_e - q_t)^2 = k_2 \cdot dt \quad \dots (7)$$

Integrating both sides for the boundary conditions  $t = 0$  to  $t = t$  and  $q_t = 0$  to  $q_t = q_t$  gives the integrated rate law for a pseudo-second-order reaction,

$$1/(q_e - q_t) = 1/q_e + k_2 \cdot t \quad \dots (8)$$

Equation (8) can be rearranged to obtain:

$$t/q_t = 1/(k_2 \cdot q_e^2) + t/q_e \quad \dots (9)$$

The kinetic constant,  $k_2$ , can be determined by plotting of  $t/q_t$  against  $t$ .

The kinetic experimental data of lead ion on the activated carbon/iron oxide composite adsorbent is simulated by pseudo-second-order rate equation (9).

Table 3 — Kinetic parameters for lead ion adsorption by iron oxide/activated carbon composite

$C_0$ (mg/L)	$q_e$ (mg/g)	$k_2$ (L/(mg.min))	$R^2$
200	3.3718	$22.20 \times 10^{-3}$	0.9986
400	6.8904	$3.52 \times 10^{-3}$	0.9878
600	9.2704	$1.73 \times 10^{-3}$	0.9664

The rate constants ( $k_2$ ) determined by the pseudo-second-order rate equation were listed in Table 3.

Remarkably, the kinetic data can be described well by the pseudo-second-order rate equation with a minimum linear correlation coefficient of 0.9911 (see Table 3), which was based on the assumption that the rate limiting step may be chemical sorption or chemisorptions involving valency forces through sharing or exchange of electron between adsorbent and adsorbate<sup>24</sup>. It can also be seen that the values of the pseudo-second-order rate constant decrease with increasing the initial lead ion concentrations.

## Conclusion

A magnetic composite adsorbent is successfully prepared with activated carbon and iron oxide as raw materials for the removal of heavy metals such as lead from aqueous solution. The performances of the composite adsorbent are compared to those of pure activated carbon, the composite adsorbent show higher adsorption capacities. Moreover, the prepared composite exhibits paramagnetic behavior, so the adsorbent can be separated more easily by an applied magnetic filter and reused without any mass loss. The adsorption properties of the composite adsorbent for lead ion depend on contact time, initial solution pH, adsorbent dosage and initial lead ion concentration. The adsorption kinetic data of lead ion can be illustrated very well by the pseudo-second-order rate equation. The equilibrium data obtained from this study is well presented by Langmuir model.

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