Behavior of chromium and arsenic on activated carbon

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ABSTRACT

The simultaneous adsorption of hexavalent chromium (Cr(VI)) and trivalent arsenic (As(III)) in single component and binary systems has been studied by activated carbon (AC). The capacity of Cr(VI) in the single experiment is greater than that of As(III) onto AC. The effects of various parameters like initial concentration, pH and temperature have been considered in the experiment. Cr(VI) removal is pH dependent and found to be maximum at pH 2.0. While, As(III) is found to be maximum at pH 7.0 in the single adsorption experiment. In the binary adsorption of As(III), the uptake of As(III) is generally higher than the single uptake. In the single adsorption the maximum adsorption rate of As(III) is 34% and in the binary metal mixtures the maximum adsorption rate of As(III) is 40% while the initial concentration is 5 mg/L. So in the binary system the Cr(VI) and As(III) are thought to be synergistic with respect to the single As(III) situation.

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1. Introduction

The pollution of heavy metal in aqueous has become severe with the development of economy. People have paid attention to it which will make us sick such as cancer or even make us to die. There are many methods about how to deal with the heavy metal pollution, such as chemical precipitation, electroanalysis, ion exchange and adsorption.

Chromium compounds are widely used in industries such as leather tanning, electroplating, manufacturing of dye, paint and paper. The industrial wastewater contained 150.8 tonnes Cr(VI) in China in the year of 2004. Chromium exists in the aqueous mainly in two states Cr(III) and Cr(VI). The hexavalent form has been considered to be more hazardous due to its carcinogenic properties [1]. The permissible limit for Cr(VI) in industrial wastewaters is 0.5 mg/L in China and 0.1 mg/L in other developed countries. Different methods such as coir pith adsorption, calcined bauxite, ferrochromium slag, reduction and precipitation, nanofiltration, electrolytic, iron exchange [2–8] have been reported for the removal of hexavelant chromium. Adsorption of Cr(VI) by AC is popularly used in recent years for its efficient and low cost. AC could be prepared from agricultural waste material, green alga, industrial solid waste, etc. It could be treated with chemical agent in advance. A batch system of experiments considering the effects of various parameters like contact time, initial concentration, pH and temperature indicate that the AC could be efficiently used for the treatment of Cr(VI). These investigations show that the Cr(VI) adsorption decreases by increasing the pH and increases by increasing the initial concentration and temperature [9–13]. AC has high surface areas and functional hole structures which make it to have great capacity of adsorbing substance. It is used abroad because of its characters that it is resistant to acid, alkali and high pressure.

Arsenic is a common pollution in industries by human activities and natural phenomena. The wastewater from the industries such as metallurgy, mining, chemical pharmacy, chemical plant, pesticide produce and leather tanning often include the arsenic pollution, while it has four stable oxidation states, i.e. +3, +5, −3 and 0, it occurs in aqueous solutions as As(III) and As(V) in the form of arsenites and arsenates, respectively. The industrial wastewater contained 306.1 tonnes arsenic in China in 2004. The arsenites is considered to be more hazardous than arsenates. Many diseases such as lung cancer, skin cancer, bladder cancer, tummy bug, etc. [14–17] may be caused by contacting with arsenic in aqueous or any other way. The permissible limit for arsenic in industrial wastewaters is 0.5 mg/L in China and 0.01 mg/L in other developed countries as Japan. There are some methods, such as coagulate, precipitation, ion exchange, biologic methods that had suggested for the removal of arsenic in countries. The adsorption of arsenic by coprecipitated aluminum, hydrous ferric oxide incorporated diatomite, iron-treated AC and zeolites have been reported [18–20]. Also the iron-impregnated or chemical modification of AC has been found to be effective in arsenic removal. In these reports the AC had been treated with irons or other chemical agents beforehand and as much iron had been to load within the carbon pores as much as possible for sorbing arsenic [21,22].

The objective of this study is to assess the uptake of Cr(VI) and As(III) from aqueous solutions onto coaly activated carbon. The adsorption mechanism at different temperatures, pH and

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concentration are discussed. However, the adsorption of As(III) or Cr(VI) separately has been reported, the admixture of them has been studied little. The behavior of Cr(VI) and As(III) onto AC is studied in this experiment. It could provide some basic data for the treatment of industrial wastewater which contains Cr(VI) and As(III).

2. Materials and methods

2.1. Materials

The coaly AC that bought from the company were washed with deionized water and boiled for 20 min, then dried at 120 °C in oven for 24 h. They were used for the experiment of chromium and arsenic adsorption. The main physical characteristics of AC which were given by the company was listed in Table 1. Cr(VI) solution was prepared by dissolving 0.2829 g $K_2Cr_2O_7$ in 1 L distilled water and the As(III) was prepared in super-clean laboratory with As_2O_3 and sub-packed in 20 ml ampoules. The concentration of this standard solution in ampoule was 100 mg/L (GSB07-1275-2000).

2.1.1. Batch experiments

Batch adsorption studies were carried out by adding 0.2 g of AC to 100 ml of a Cr(VI) solution at concentration (ranging from 5 mg/L to 75 mg/L) and pH (ranging from 1.0 to 9.0) and the temperature (ranging from 20 °C to 40 °C) in a 250 ml Erlenmeyer flask on a mechanical shaker. The initial pH was adjusted with solutions of 1 M HCl or 1 M NaOH. The agitation speed of the shaker was fixed at 130 rpm for all batch experiments. Samples at different time intervals were taken. After 6 h of contact, the concentration of the samples was analyzed in a spectrophotometer (UV-1201 model) using 1,5-diphenylcarbazide as the complexing agent at the wavelength of 540 nm (GB7467-87).

The adsorption of As(III) was carried out by adding 0.2 g of AC to 100 ml of a As(III) solution at concentration (5 mg/L and 10 mg/L) and pH (ranging from 1.0 to 9.0) and the temperature (ranging from 20 °C to 40 °C) in a 250 ml Erlenmeyer flask on a mechanical shaker. The initial pH was adjusted with solutions of 1 M HCl or 1 M NaOH. The agitation speed of the shaker was fixed at 130 rpm for all batch experiments. After 6 h of contact, the concentration of the arsenic samples was analyzed in the spectrophotometer (UV-1201 model) at the wavelength of 510 nm (GB7485-87). All the chemicals used in the experiment were analytical reagent grade.

2.1.2. Recycling studies

First the anaerobic sludge (SV30 = 27.0%) and aerobic sludge (SV30 = 36.5%) were carried from the wastewater treatment plants in China. Both of the sludge were put into two 1000 ml beakers and the volume of sludge was 600 ml. Everyday nutriments (such as C, N, P) were added into the sludge in order to maintain the activation of the sludge. Also the Cr(VI) and As(III) were added into the beakers everyday and the concentration was increased gradually. But the volume of Cr(VI) and As(III) was fixed at 1 ml respectively everyday. First 2 days Cr(VI) and As(III) were added into the beakers while the concentration of Cr(VI) and As(III) were 0.5 mg/L. The concentration was increased as follows 1 mg/L, 2 mg/L, 5 mg/L and 10 mg/L. After

Table 1
Physical characteristics of AC

Constituents	AC
Surface area (m^2/g)	>1000
Pores volume (cm^3/g)	>0.9
Intensity (%)	≥90
pH	≥7
Filling density (g/cm^3)	0.38–0.45
Iodine value (mg/g)	≥1000

10 days, both of the activated sludge suspension was taken out. The activated carbon which had been used for adsorption experiment was divided into three parts. The first part was kept, the second was added into the anaerobic sludge suspension and the third part into the aerobic sludge suspension for 48 h. After 48 h, all the carbons were dried at 120 °C for 2 h. Then the three kinds of activated carbon were used for adsorbing Cr(VI) and As(III) again. The concentration of Cr(VI) and As(III) admixture was 10 mg/L.

2.2. Adsorption isotherms

Adsorption data are generally analyzed by Langmuir isotherms. The general form of this model is presented in Eq. (1):

$$q_e = \frac{K_L b C_e}{(1 + b C_e)} \tag{1}$$

$$\frac{C_e}{q_e} = \frac{1}{(bK_L)} + \frac{C_e}{K_L} \tag{2}$$

where q_e is the amount of metal ions adsorbed per unit mass of adsorbent (mg/g), C_e is the equilibrium concentration of solute in the bulk solution (mg/L), K_L and b are the Langmuir constants related to the adsorption capacity and the energy of adsorption, respectively. Values of K_L and b can be determined from the linear form of the Langmuir model Eq. (2), the intercept is $1/(bK_L)$ and the slope is $1/K_L$ [23].

3. Results and discussion

3.1. Effect of pH

The effect of pH on the single adsorption of Cr(VI) and As(III) was studied in the range 1.0–9.0 at the temperature of 30 °C. According to the harm of Cr(VI) and As(III) and the literature, 10 mg/L was chosen for the initial concentration and 0.2 g activated carbon for the experiment accordingly. The contact time is 6 h. The result of the effect of pH on adsorption is shown in Fig. 1.

When the aqueous is acidic, the removal of Cr(VI) is better and the maximum of Cr(VI) adsorption occurs at pH 2.0 that it reaches 90%. The other way round, in the alkaline aqueous, the removal of Cr(VI) is bad or even no adsorption. As shown in Fig. 1, the adsorption of Cr(VI) decreases to 0.02% at pH 7.0. Because the pH of solution not only affects the adsorption sites of sorbent but also the ionic forms of the chromium solutions. The adsorption of Cr(VI) could be occurring via positively charged ligands [24]. The low pH leads to an increase in H+ ions on the carbon surface which results in strong electrostatic attraction between positively charged AC surface and chromium ions when the dominant form of Cr(VI)
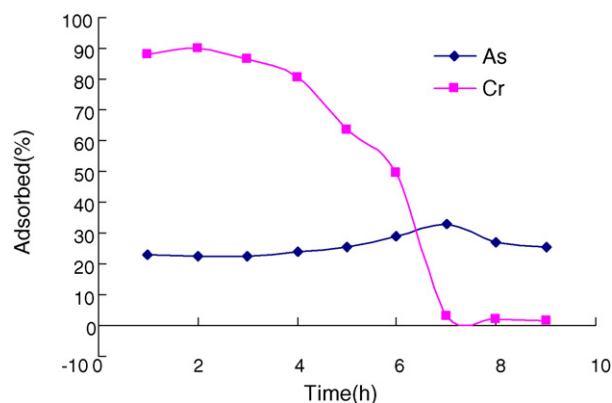
Fig. 1. Effect of pH of single Cr(VI) and As(III) while the initial concentration is 10 mg/L, respectively and $T = 30$ °C.
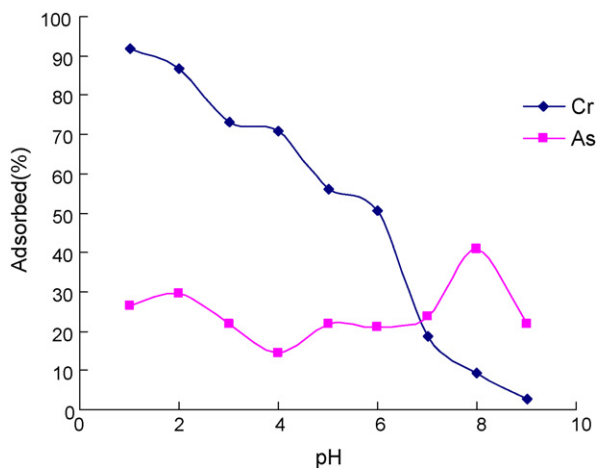
Fig. 2. Effect of pH of As(III) and Cr(VI) admixture while the initial concentration is 10 mg/L and $T = 30\,°C$.

is $HCrO_4^-$. So the adsorption of Cr(VI) is good at low pH. As the increase of pH, the dominant forms of Cr(VI) are CrO_4^{2-}, $Cr_2O_7^{2-}$ while the OH^- ions are increased too. The affinity of OH^- with AC is better than CrO_4^{2-}, $Cr_2O_7^{2-}$, so the adsorption sites are taken up by OH^- that the adsorption of Cr(VI) is invalidation [25].

The removal of As(III) increases as the pH of the system increases and reaches a maximum at about pH 7.0, followed by a decrease in the extent of adsorption up to 9.0. At pH 7.0, the adsorption could reach nearly 32.8%. That is because in the acidic medium the surface of AC is highly protoned and As(III) mostly exists in the form of neutral H_3AsO_3 species when the removal of As(III) is not favorable [26]. In the range of 4.0–9.0, neutral H_3AsO_3 is the dominant As(III) species in solution. Therefore, the adsorption of As(III) would be less strongly influenced by the charge repulsion forces. Given the adsorbing species are uncharged, adsorption will occur most readily on all surface sites [27].

Fig. 2 shows the effect of pH on the removal of Cr(VI) and As(III) admixture by AC. The initial concentration of Cr(VI) and As(III) admixture was 10 mg/L. Comparing to the single component adsorption of Cr(VI) and As(III) onto AC, the adsorption of admixture changed.

The adsorption of Cr(VI) decreases when the pH ranges from 1.0 to 9.0 and the maximum adsorption occurs at pH 1.0 that it reaches 92% while the maximum adsorption reaches 90% at pH 2.0 at the single adsorption experiment. At the pH 7.0, there still have adsorption that it reaches 18.67% which makes a difference with the single adsorption. Meanwhile, the adsorption of As(III) increases when the pH ranges from 1.0 to 3.0 which have overrun the single adsorption of As(III). The explanation is that it is influenced by the adsorption of Cr(VI) that chemical interaction maybe happened between Cr(VI) and As(III). The maximum adsorption of As(III) occurs at pH 8.0 in the binary metal mixture and the amount of adsorption reaches 40.9%. While, the maximum adsorption occurs at pH 7.0 in the single adsorption experiment and the amount of adsorption of As(III) reaches 32.8%. It can attain a conclusion that at the low pH the adsorption sites available relative to As(III) increases when the Cr(VI) exists.

3.2. Effect of concentration

The effect of initial concentration on the adsorption of Cr(VI) was studied in the range 5–75 mg/L with pH value fixed at 2.0 and the temperature at 30 °C. The amount of chromium adsorbed for different initial concentrations onto AC is shown in Fig. 3.
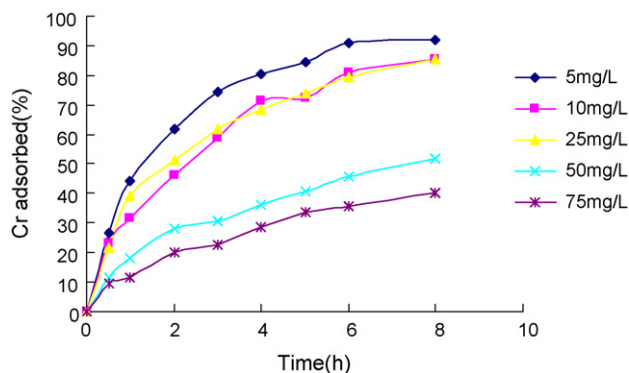

Fig. 3. Effect of initial concentration of Cr(VI) at $T = 30\,°C$.

The removal of Cr(VI) is strongly considered with the initial concentration. If the initial concentration is about 5 mg/L, the amount of Cr(VI) removed achieved 91.75%, whereas, if the initial concentration is 75 mg/L, the amount of Cr(VI) removed drops to 41.60% for a time period of 6 h. When the initial concentration of Cr(VI) increased, the rate of adsorption is decreased, but the amount of adsorption is increased. The adsorption of Cr(VI) is concentration dependant and it could be seen that the amount of adsorption enhanced with the increase of initial concentration. In the first stage, the speed of adsorption is rapid and then attains an equilibrium value. The equilibrium is achieved easily when the initial concentration is low. Because at the first stage the ratio of available surface of AC is large to the adsorption of Cr(VI) and as the contact time increases it gradually decreases until it attains an equilibrium [28].

Fig. 4 shows the effect of the time of treatment on the removal of As(III) by AC. The metal concentrations are 5 mg/L and 10 mg/L with pH 3.70 and 3.56, respectively and the temperature is at 30 °C.

It could be seen from Fig. 4 that the removal of As(III) increases with time and attains equilibrium in 6 h. Metal ion adsorption increases sharply for short time and slows gradually when the equilibrium approached. Because the available sites on the carbon surface decreased as the adsorption proceeds. If the initial concentration is about 5 mg/L, the amount of As(III) removed achieves 34%, whereas, if the initial concentration is 10 mg/L, the amount of As(III) removed drops to 11% for a period of 6 h. As some literature reported, the adsorption efficiency of As(III) decreases with the increase of initial concentration [29].

The adsorption data of Cr(VI) and As(III) from the binary mixtures are shown in Fig. 5. The initial concentration of Cr(VI) and As(III) admixture was 10 mg/L and 10 mg/L, respectively and the other one was 10 mg/L and 5 mg/L. In the figure, the letters repre-
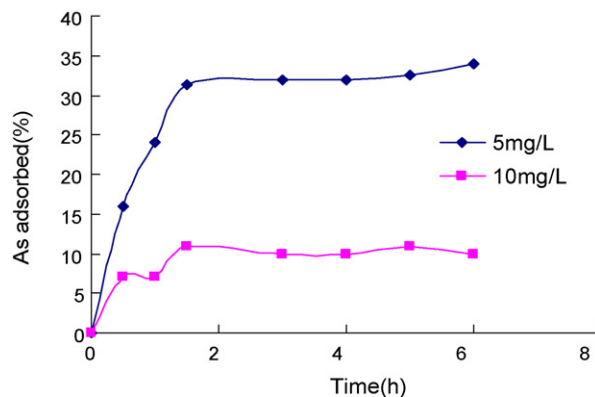

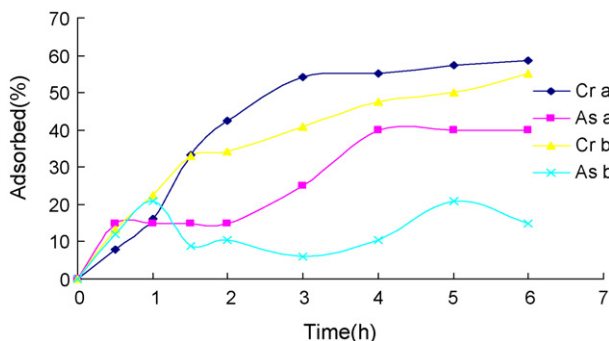**Fig. 4.** Effect of initial concentration of As(III) at $T = 30\,°C$.

Fig. 5. Effect of initial concentration of Cr(VI) and As(III) admixture at $T = 30\,^{\circ}C$.

Fig. 7. Effect of temperature of As(III) while the initial concentration is 10 mg/L.

sent as follows, Cr a is the Cr(VI) data while the initial concentration of Cr(VI) and As(III) admixture was 10 mg/L and 5 mg/L, respectively, As a is the As(III) data while the initial concentration of Cr(VI) and As(III) admixture was 10 mg/L and 5 mg/L, respectively, Cr b is the Cr(VI) data while the initial concentration of Cr(VI) and As(III) admixture was 10 mg/L and 10 mg/L, respectively, As b is the As(III) data while the initial concentration of Cr(VI) and As(III) admixture was 10 mg/L and 10 mg/L, respectively.

It could be seen from Fig. 5 that the maximum of adsorption rate of Cr(VI) is 58.62% when the concentration of As(III) is 5 mg/L, whereas, the maximum of adsorption rate of Cr(VI) is 55.26% when the concentration of As(III) is 10 mg/L. The trend that the adsorption of Cr(VI) decreased with the increase of initial ion concentration of As(III) seems reasonable since the relative number of binding sites is available to each metal. In the binary adsorption of As(III), the uptake of As(III) is generally higher than the single uptake. In the single adsorption the maximum adsorption rate of As(III) is 34% and in the binary metal mixtures the maximum adsorption rate of As(III) is 40% while the initial concentration is 5 mg/L. At binary metal mixtures the driving force is greater, forcing the adsorption of As(III) to get more. The possible explanation for the phenomena is that there are a variety of binding sites on the AC that are specific or partially specific for the metal ions or the total concentration change the ratio of adsorbent surface and metal solution [30,31].

3.3. Effect of temperature

The effect of temperature of adsorption was studied in the range 20–40 $^{\circ}C$. The initial concentration of Cr(VI) and As(III) is 10 mg/L, respectively. The results are shown in Figs. 6 and 7.

The conclusion could be attained from Fig. 6 that the adsorption capacity of Cr(VI) by AC is enhanced with the increase of temperature. The amount of adsorption also increases with contact time increases. This indicate that the temperature is higher the
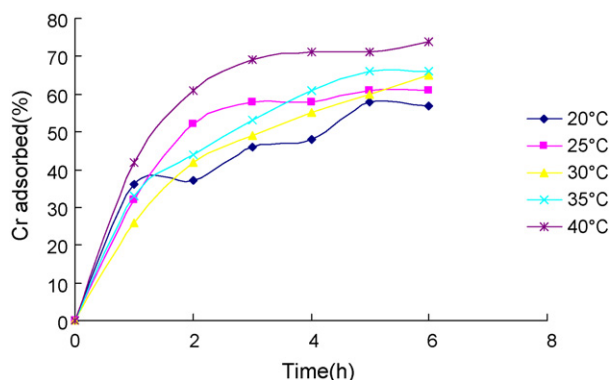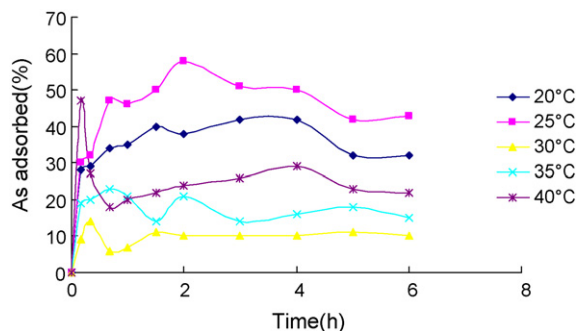
adsorption is more favorable. When the temperature is 40 $^{\circ}C$ the maximum amount of Cr(VI) reaches 74%. On the other hand, when the temperature is 20 $^{\circ}C$ the maximum amount of Cr(VI) reaches 57%. In the case of physical adsorption, the amount adsorbed increases as the temperature decreases [32], but the amount of Cr(VI) onto AC increases as the temperature increased. This phenomena may be due to the increase of the rate of intraparticle of Cr(VI) into the pores of the adsorbent at higher temperatures which indicates that the diffusion reactions play an important role in the process of adsorption [33,34].

Fig. 7 shows the effect of temperature of As(III) at the initial concentration of 10 mg/L at the nature pH. It could be seen from Fig. 7 that the amount of adsorption of As(III) increases at the first stage time and decreases as the contact time increases. When the temperature is 25 $^{\circ}C$, the amount of As(III) reaches 43%. On the other hand, when the temperature is 30 $^{\circ}C$ the amount of Cr(VI) reaches 10%. The adsorption is affected much that the volume increases rapidly at the first 1 h. But when the temperature is 40 $^{\circ}C$ and 30 $^{\circ}C$, the volume decreases then and attains equilibrium gradually. With the increases of temperature the stability of the bonds between the active sites of AC and arsenic decreases [22]. But the diffusion process increases when the temperature increases. Due to these reasons the effect of temperature is unfavorable.

From the data of Figs. 6 and 7, we can get the Table 2. As can be seen from the Table 2, the model parameters for the adsorption of single Cr(VI) and As(III) by Langmuir isotherm at different temperatures are represented. The correlation coefficient values ($R^2 > 0.9012$) indicates that it fits good to the Langmuir isotherm. Furthermore, Table 2 indicates that the Langmuir constant b is higher for Cr(VI) than for As(III) confirming that AC has the larger Cr(VI) energy of adsorption.

3.4. Recycling study

Recycling studies are helpful for the recovery of metal ions and reuse of the carbons. Two kinds of sludge were used for the recycling study and the volume was 600 ml each one. Then the sludge suspension was used for the desorption of AC. The adsorption of Cr(VI) and As(III) by recycling AC is shown in Fig. 8. In the figure,

Fig. 6. Effect of temperature of Cr(VI) while the initial concentration is 10 mg/L.

Table 2
Effect of temperature on the values of Langmuir isotherm constants

T ($^{\circ}C$)	Cr(VI)			As(III)		
	$K_L \times 10^{-3}$	b (L/mg) $\times 10^4$	R^2	$K_L \times 10^{-3}$	b (L/mg) $\times 10^4$	R^2
20	0.9652	3.6759	0.9834	1.6733	1.4182	0.9865
25	1.0495	3.4038	0.9855	1.2282	2.4170	0.9643
30	1.2550	2.5592	0.9327	26.109	0.0463	0.9208
35	0.9083	4.4634	0.9629	6.5104	0.2276	0.9838
40	0.6617	8.8852	0.9849	2.4295	0.8497	0.9012

T is temperature, K_L and b are the Langmuir constants, R^2 is the correlation coefficient.
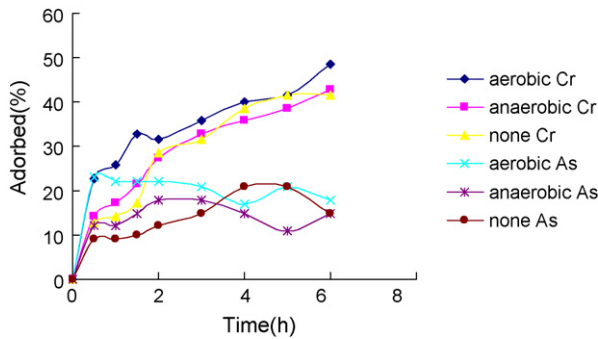
Fig. 8. Cr(VI) and As(III) adsorption by AC which had been desorbed and the initial concentration of Cr(VI) and As(III) is 10 mg/L and $T = 30\,^\circ$C.

the letters represent as follows, aerobic Cr is the Cr(VI) data while the AC had been added in aerobic sludge suspension for the adsorption of Cr(VI), anaerobic Cr is the Cr(VI) data while the AC had been added in anaerobic sludge suspension for the adsorption of Cr(VI), none Cr is the Cr(VI) data while the AC had not been desorbed for the adsorption of Cr(VI), aerobic As is the As(III) data while the AC had been added in aerobic sludge suspension for the adsorption of As(III), anaerobic As is the As(III) data while the AC had been added in aerobic sludge suspension for the adsorption of As(III), none As is the As(III) data while the AC had not been desorbed for the adsorption of As(III).

The initial concentration of Cr(VI) and As(III) is 10 mg/L and the adsorption temperature is 30 °C. As a whole, the maximum adsorption of Cr(VI) and As(III) occurs when the AC dip in aerobic sludge suspension for 48 h. The amount of adsorption of Cr(VI) reaches 48.57% and the amount of As(III) reaches 23%. It concludes that the Cr(VI) and As(III) desorbed by bacteria in aerobic sludge suspension better than the anaerobic sludge suspension. It shows that the biosorbent has a potential to adsorb the metal ions.

4. Conclusions

The AC can be used as a material for adsorption of chromium from aqueous solution. Temperature and pH are determinant factors for the removal of Cr(VI). This removal strongly increases in the lower pH and high temperature. The adsorption of As(III) is pH dependent and the pH adsorption shows that As(III) removal is optimum in the pH range of 7.0–8.0. It could be seen from the single adsorption experiment that the Cr(VI) is more affinitive with AC than As(III). In the binary mixture adsorption experiment, it could attain the conclusion that the total interactive effects of Cr(VI) and As(III) on AC can be thought to be synergistic with respect to the single As(III) ion situation.

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