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Removal of Cadmium from Aqueous Solutions by Activated Charcoal

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Abstract: A method based on adsorption phenomena has been established for the removal of cadmium from aqueous solution by activated charcoal. Experimental conditions were optimized for the maximum uptake of cadmium by charcoal in terms of shaking time, pH, amount of charcoal, cadmium ion concentration, and temperature. Maximum adsorption was observed at 4.0 pH within 20 min equilibrium time. Various adsorption isotherms, i.e., the Freundlich, Langmuir, and Dubinin-Radushkevich (D-R) have been applied to the adsorption data. The sorption mean free energy from the D-R isotherm was $3.07 \pm 0.05\,\mathrm{kJ\,mol^{-1}}$ indicating physical nature of adsorption. The adsorption of cadmium increased with the increase in temperature. Thermodynamic quantities ΔH, ΔS, and ΔG were calculated for the adsorption process. The method was tested by removing the cadmium from cadmium-spiked tap water using activated charcoal. It was observed that about 70–75% cadmium could be removed by activated charcoal in a single step.

Keywords: Cadmium, removal, adsorption, activated charcoal

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INTRODUCTION

Cadmium (Cd), a toxic element, when present even in a small amount in water poses hazards to human life. The harmful effects of cadmium on biosphere are well documented (1, 2). Cadmium enters into the aquatic system through mining and metallurgical processes, industrial effluents from cadmium-bearing alloys, soldering materials, electroplating, and Ni-Cd batteries. All these sources contribute in making the water toxic above 0.01 $mg\,L^{-1}$, the permissible limit for human consumption. It is, therefore, necessary to remove cadmium from aqueous industrial wastes before discharging it into the natural water stream. Hence there is a requirement for a suitable, economically viable method of preconcentration and separation of the metal from the discharge.

Various procedures have been reported for the removal/preconcentration of cadmium from aqueous media such as reductive precipitation (3, 4), solvent extraction (5, 6), ion exchangers (7, 8) and adsorption. The adsorbents used for cadmium include peat (9), bentonite (10), goethite (11), soil (12, 13), zeolites (14), metal oxides (15–18), chelating resin (19, 20), and activated carbon. Preconcentration of metal ions from aqueous solution by using active carbon is considered an effective method due to its well-known adsorptive characteristics. Adsorption of cadmium ions on activated carbon both virgin (21–26) as well as modified (27) has been reported in the literature. These studies have been investigated from different points of view. The present paper reports the results of a systematic study undertaken to optimize different physicochemical parameters to simulate the best conditions for the removal/preconcentration of cadmium ions from aqueous solution using charcoal which could be applied to the real system.

EXPERIMENTAL

Reagents

A commercial activated charcoal, BDH, Product No. 33032, having surface area 980 m^2/g; porosity 75.75%; pore volume 1.43 cm^3/g; particle size 3.7 ± 0.2 micron was used as such during this study. Cadmium nitrate tetrahydrate (Merck, Product No. 102019) was used to prepare stock solution of cadmium ions in deionized water.

Buffer solutions of pH 1 to 6, having ionic strength of 0.1 $mol\,L^{-1}$, were prepared by using appropriate volumes of solutions of KCl and HCl (pH 1–3), CH_3COONa, and CH_3COOH (pH 4–6). All pH measurements were made with a Metrohm 605 digital pH meter equipped with a combined glass/calomel electrode. All the reagents used were of analytical grade and used as such. Distilled and deionized water was used throughout.

Sorption Measurements

The uptake of cadmium ions by activated charcoal was studied using batch technique. Details of adsorption procedure employed were similar to those described elsewhere (28). The concentration of cadmium ions in solution was measured using X-ray fluorescence spectrometer (Siemens SRS200)/ atomic absorption spectrometer (Hitachi Z-8000). All experiments were performed at ambient temperature (23 ± 1°C) unless otherwise specified.

TREATMENT OF DATA

Adsorption of Metal

The distribution coefficient (K_d) and the amount of Cd adsorbed (q) on activated charcoal surface were calculated using the following equations:

$$K_d = \frac{C_o - C_f}{C_f} \times \frac{V}{M} \ (\mathrm{mL\,g^{-1}}) \quad (1)$$

$$q = \frac{(C_o - C_f)V}{M} \ (\mathrm{g\,g^{-1}}) \quad (2)$$

Where

C_o = initial concentration of metal in the solution (μg mL^{-1})

C_f = concentration of metal in solution after equilibrium (μg mL^{-1})

V = volume of adsorbate solution (mL)

M = amount of adsorbent (g)

Calculation of Thermodynamic Parameters

The values of ΔH and ΔS were calculated from the slope and intercept of the linear van't Hoff plot, respectively, using the relation:

$$\ln K_d = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \quad (3)$$

where

ΔS = entropy change for the process

ΔH = enthalpy change for the process

R = gas constant

T = absolute temperature

The changes in free energy (ΔG) for the specific adsorption have also been calculated using the equation

$$\Delta G = -RT \ln K_d \tag{4}$$

where the symbols have their usual significance.

RESULTS AND DISCUSSION

The adsorption of cadmium ions from aqueous solutions on activated charcoal was examined by optimizing various physicochemical parameters such as shaking time, pH, amount of adsorbent and concentration of adsorbate, using batch method. The effect of temperature on the adsorption of cadmium was also checked. The criterion for the optimization was the selection of parameters where maximum adsorption occurred. All the reported results are the average of at least triplicate independent measurements. The determined relative standard deviation was within $\pm 3.5\%$ unless otherwise specified.

Influence of Equilibration Time

Figure 1 shows the variation of distribution coefficient of cadmium ions onto activated charcoal with shaking time using deionized water as a sorptive medium. This was performed by shaking 10 mL of $0.1\,g\,L^{-1}$ cadmium ion

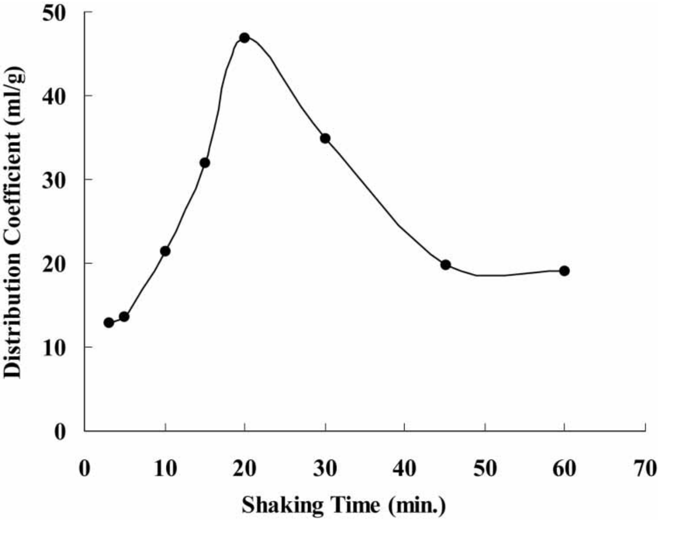

Figure 1. Variation of distribution coefficient with shaking time.

solution with 100 mg of activated charcoal for different intervals of time ranging from 3 to 60 min. This figure indicates that K_d values first increase with a rise in shaking time, attain maximum value around 20 min, and then start decreasing with a further rise in shaking time. A similar trend has been reported in the literature for the adsorption of cadmium ion onto manganese dioxide (15). Therefore, an optimum contact time of 20 min for maximum K_d was selected for subsequent adsorption studies.

Effect of Amount of Adsorbent

The influence of the amount of activated charcoal on the adsorption of cadmium ion was investigated by shaking 10 mL of 0.1 g L^{-1} cadmium ion solution for 20 min. The amount of charcoal was varied from 20 to 200 mg. The results are reproduced in Fig. 2, which shows that the K_d values increase with an increase in the amount of activated charcoal and attained maximum value at 100 mg of activated charcoal, which remains almost constant up to 200 mg of charcoal. Therefore, 100 mg of activated charcoal was used in all the subsequent studies.

Variation of pH

The adsorption of metal from aqueous solution is dependent on pH of the medium, which affects the surface charge of the adsorbent, degree of

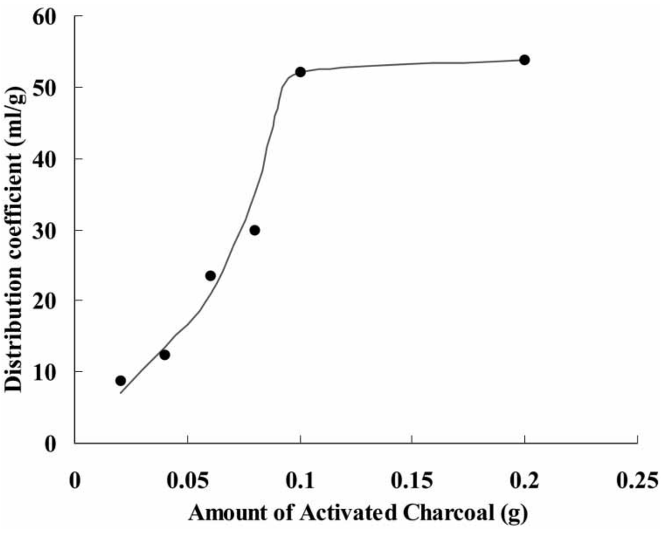

Figure 2. Variation of distribution coefficient with amount of activated charcoal.

ionization, and speciation of the adsorbate. Therefore, the effect of pH on the adsorption of cadmium ion was investigated in the pH range of 1–6 using 100 mg of activated charcoal. The variation of K_d with rise in pH of the solution is shown in Fig. 3. It is obvious from this figure that K_d value gradually increases up to pH 4 and then starts decreasing with a further rise in pH. Therefore, pH 4 was considered to be the optimum pH to remove cadmium ions from aqueous solution and was used for further investigations. The optimum pH values of 4.5 (25) and 4–6 (26), for maximum uptake of cadmium ions by activated carbon, have been reported, for aqueous solutions whereas a pH value of 5 has been quoted for solutions containing chelating agents like DTPA and o-phenanthroline (21).

Relatively lower adsorption of cadmium at lower pH could be explained on the basis that at low pH values electrostatic repulsion between metal ions and positively charged surface of charcoal will be operative. Whereas at higher pH values the extent of adsorption increases due to the replacement of hydrogen from charcoal surface by the metal ions.

Effect of Adsorbate Concentration

The effect of cadmium ion concentration on its own adsorption on activated charcoal has been studied at optimum conditions of shaking time 20 min,

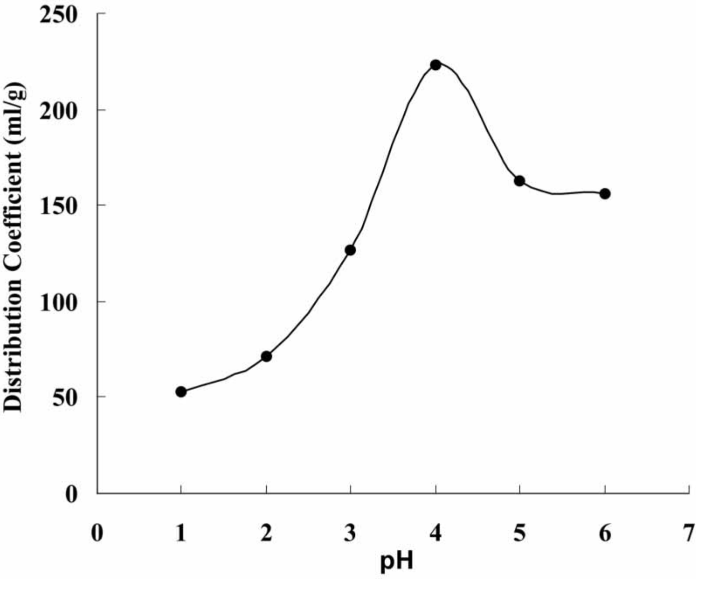

Figure 3. Variation of distribution coefficient with pH.

100 mg activated charcoal, and pH 4. The concentration of cadmium ions was varied from 0.5–3.5 $g\,L^{-1}$ and the results are shown in Fig. 4, which indicates that K_d decreases with the increase in cadmium ion concentration, indicating that energetically less favorable sites become involved in the process with increasing concentration. The data concerning the dependence of the extent of adsorption with the increase in cadmium ion concentration was subjected to examination of the Freundlich, Langmuir, and Dubinin-Radushkevich (D-R) isotherms equations.

The Freundlich equation was used in the form:

$$q = A\,C^{1/n} \tag{5}$$

Linearized form of the Freundlich equation is

$$\log q = \log A + 1/n \log C \tag{6}$$

where q is the amount of cadmium ions adsorbed per gram, C is the equilibrium concentration of cadmium ion in the solution, A and n are characteristic constants that can be related to the strength of the adsorptive bond and the adsorption bond distribution, respectively (30).

The Freundlich plot of log q vs. log C shown in Fig. 5 demonstrates the validity of the equation up to 2.5 $g\,L^{-1}$ of cadmium ions in solution. The numerical values of adsorption capacity (A) and intensity (1/n) were

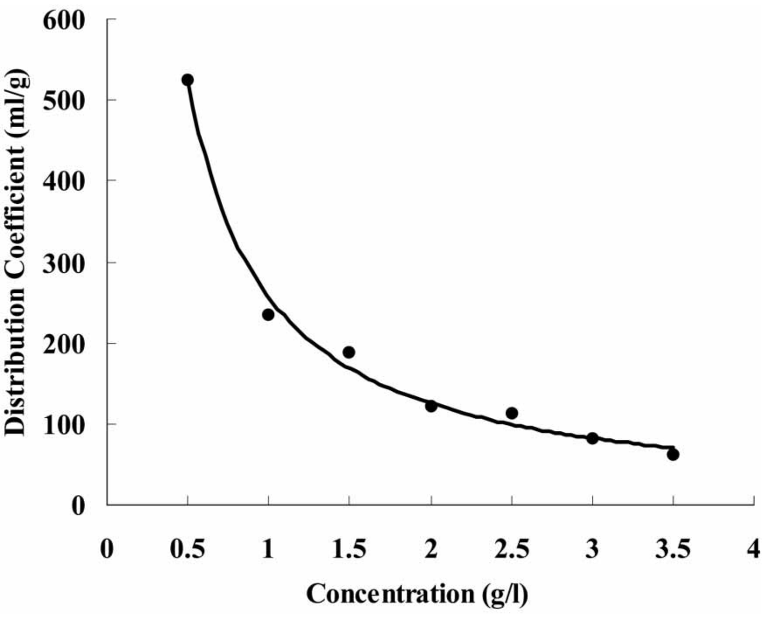

Figure 4. Variation of distribution coefficient with cadmium ion concentration.

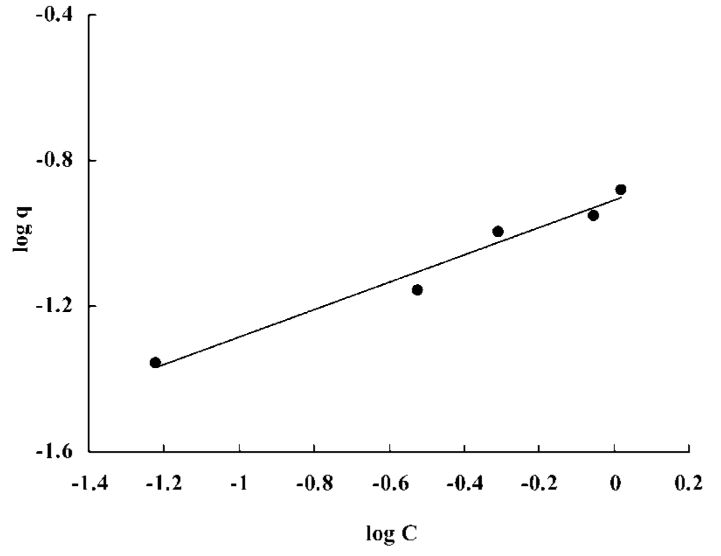

Figure 5. Freundlich plot for the adsorption of cadmium on charcoal.

calculated from the intercept and slope of the straight line using a least-square fit program and were found to be $123 \pm 2\,\mathrm{mg\,g^{-1}}$ and 0.34 ± 0.032, respectively.

The Langmuir equation was applied in the form:

$$C/q = 1/Kq_m + C/q_m \tag{7}$$

where q and C have been already defined, q_m is the measure of monolayer capacity, and K is the constant related to the heat of adsorption. A straight line was obtained by plotting C/q against C (Fig. 6), indicating the conformity of the data to the Langmuir equation in the entire concentration range studied. Values of the constants q_m and K calculated from the slope and intercept of the plot in Fig. 6 were $151 \pm 6\,\mathrm{mg\,g^{-1}}$ and $4.123 \pm 0.82\,\mathrm{g\,L^{-1}}$, respectively.

The linearized form of D-R isothermal equation used is

$$\ln q = \ln q_m - B\varepsilon^2 \tag{8}$$

where $\varepsilon = RT\ln(1 + 1/C)$, B is the constant related to the adsorption energy, R is the gas constant, and T is temperature in Kelvin. The quantities q, q_m, and C have their previously defined meanings. A straight line is obtained upon plotting ln q vs. ε^2 (Fig. 7), indicating that adsorption of cadmium ions onto activated charcoal also obeys the D-R isothermal equation in the entire concentration range studied. Values of q_m and B calculated from the intercept and the slope of the plot in Fig. 7 were $143 \pm 2.3\,\mathrm{mg\,g^{-1}}$ and $-5.7 \times 10^{-2} \pm 4.9 \times 10^{-3}\,\mathrm{kJ^2\,mol^{-2}}$, respectively. From the value of B it

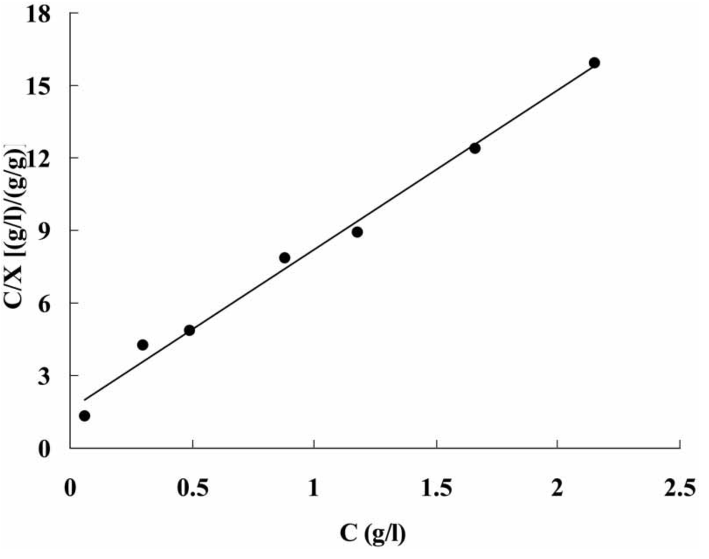

Figure 6. Langmuir plot for the adsorption of cadmium on charcoal.

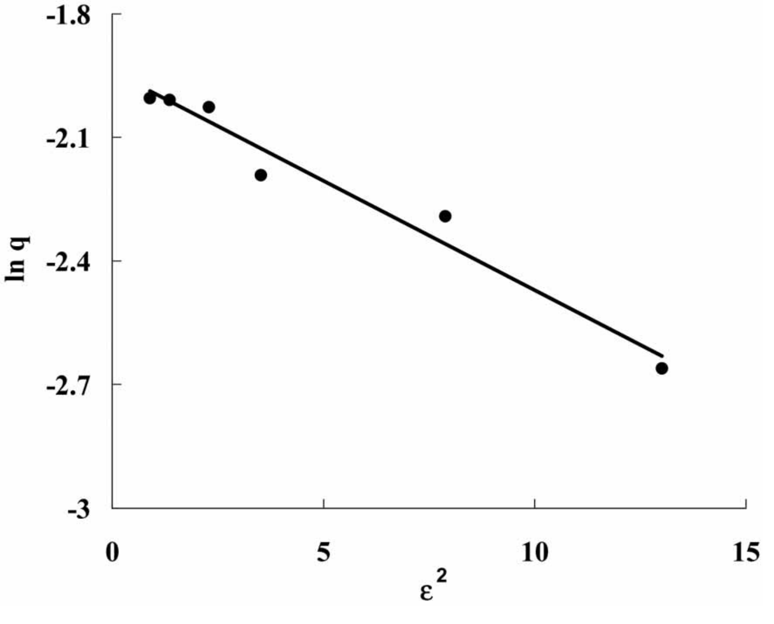

Figure 7. Dubinin-Radushkevich plot for the adsorption of cadmium on charcoal.

is possible to calculate the adsorption energy, E_a, using the following equation:

$$E_a = \frac{1}{\sqrt{-2B}} \quad (9)$$

which is the mean free energy of transfer of one mole of solute from infinity to the surface of activated charcoal. The calculated value of E_a from Eq. (9) was found to be $3.07 \pm 0.05\,kJ\,mol^{-1}$, which indicates that the cadmium adsorption onto activated charcoal is physical in nature (29).

Effect of Temperature

The dependence of K_d for cadmium ions adsorption on activated charcoal was also studied in the temperature range of 10 to 60°C in step of 10°C using the optimized conditions. The extent of cadmium adsorption on charcoal at various temperatures is given in Table 1 and Fig. 8, which show that K_d value increases with an increase in the temperature, which indicates that cadmium ions dehydrate considerably at higher temperature before adsorption and thus their size during adsorption is smaller, yielding a larger K_d value (30). Similar observations have also been reported in literature for the adsorption of cadmium ions on ceric oxide (17), zirconium oxide (18), wool (31), and activated charcoal (24).

The thermodynamic quantities such as ΔH, ΔS, ΔG of cadmium ions adsorption on activated charcoal were calculated from the K_d values using Eqs. (3) and (4). Values of ΔH and ΔS were computed from the slope and intercept of linear variation of $\ln K_d$ with the reciprocal of temperature (Fig. 9) and was found to be $26.69 \pm 0.53\,kJ\,mol^{-1}$ and $134 \pm 1.7\,JK^{-1}\,mol^{-1}$, respectively (Table 1). The values of free energy of

Table 1. Adsorption studies of cadmium ions on activated charcoal as a function of temperature.

Temp. (K)	1/T $(K^{-1}) \times 10^{-3}$	K_d $(mL\ g^{-1})$	$\ln K_d$	ΔH $(kJ \cdot mol^{-1})$	ΔS $(JK^{-1} \cdot mol^{-1})$	ΔG $(kJ \cdot mol^{-1})$
283	3.533	115	4.745			−11.16
293	3.413	170	5.136			−12.51
303	3.300	257	5.549	26.69	134	−13.98
313	3.195	351	5.861			−15.25
323	3.096	493	6.200			−16.65
333	3.003	614	6.420			−17.77

Amount of charcoal: 100 mg; Shaking time: 20.0 min.; Volume equilibrated: 10 mL; pH of the buffer solution: 4.0; Concentration of cadmium: $100\,mg\,L^{-1}$.

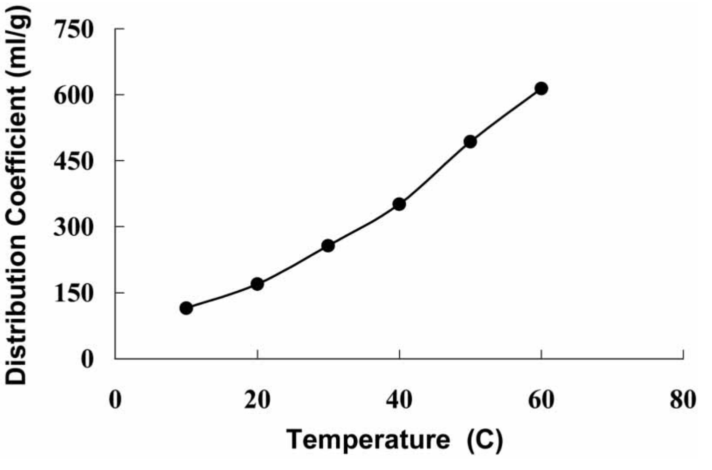

Figure 8. Variation of distribution coefficient with temperature.

specific adsorption, ΔG at various temperatures were calculated by using Eq. (4) and are listed in Table 1. The positive value of ΔH shows that the adsorption of cadmium ions on activated charcoal is an endothermic process that is in accordance with the reported results of cadmium adsorption on ceric oxide (17), zirconium oxide (18), and activated carbon (24) but contrary to those of activated carbon (23, 26). A possible explanation for the endothermicity of adsorption may be explained on the basis of the fact that Cd^{2+} ions are

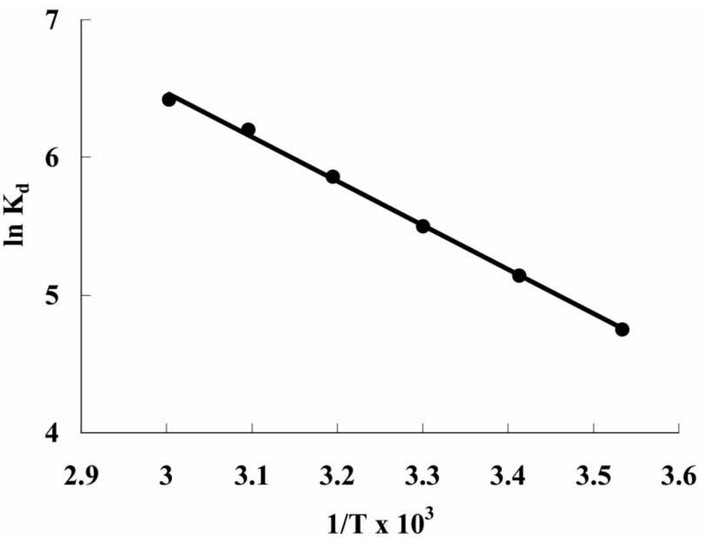

Figure 9. Plot of $\ln K_d$ vs. $1/T$.

well solvated in water. In order for these ions to adsorb, they are to some extent denuded of their hydration sheath. This dehydration process of ions requires energy. It is assumed that after adsorption the environment of the metal ions is less aqueous than it was in the solution state. The removal of water from ions is essentially an endothermic process, and it appears that the endothermicity of the desolvation process exceeds that of the heat of adsorption by a considerable extent. Similar arguments have been reported for the adsorption of various metals on activated charcoal (23). The values of free energy (ΔG) are negative as expected for a spontaneous process. The decrease in ΔG value with increasing temperature reveals that adsorption of cadmium ions on activated charcoal becomes favorable at higher temperature, because cadmium ions are more readily desolvated. The positive value of ΔS reveals the increase in randomness at solid-solution interface during the fixation of Cd ions on the active sites of the adsorbent.

Application of the Procedure

The adsorption efficiency of an activated charcoal is sometimes effected by the presence of other species in aqueous solution by their coadsorption or by negative adsorption along with the metal ions of interest. Therefore, using the optimized sorption parameters for the uptake of cadmium ions from aqueous solutions, the adsorption of spiked cadmium ($10.0\,mg\,L^{-1}$) in tap water was also studied. The determined composition of tap water is given in Table 2. Percentage recovery data for the spiked cadmium

Table 2. Determined composition of tap water

Cations/anions	Concentration ($mg\,L^{-1}$)
Ca	22.60
Mg	35.30
Na	85.60
K	5.80
Fe	0.22
Mn	0.02
Zn	0.18
Cu	0.01
Pb	52.90[a]
Cd	10.00[a]
CO_3^{-2}	0.96
HCO_3^{-2}	140.00
Cl^-	11.00
SO_4^{-2}	8.00

[a] $\mu g\,L^{-1}$.

Table 3. % Recovery of spiked cadmium from tap water by activated charcoal

Amount of Cd spiked ($mg\,L^{-1}$)	Medium of solution	% Recovery	Mean
10.0	Tap water as such	27.2	
	(pH = 6.5)	28.7	27.4
		26.3	
10.0	Tap water + Buffer solution	72.4	
	of pH = 4.0	70.4	71.2
		70.9	
20.0	Tap water + Buffer solution	72.3	
	of pH = 4.0	73.9	73.7
		74.9	

concentration in tap water is given in Table 3, which indicates that the presence of existing concentrations of cations and anions in the medium has insignificant effect on the adsorption efficiency of cadmium ions on activated charcoal; therefore, it could be used to combat the cadmium pollution in water.

CONCLUSION

On the basis of this study, it is concluded that activated charcoal can be used for the preconcentration/removal of cadmium ions from aqueous solutions within a short contact time of 20 min, without any extensive time-consuming adjustments. The method is simple, fast, and economical. It is, therefore, concluded that activated charcoal has great potential to be utilized for the removal of cadmium from aqueous media.

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