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Kinetic and Equilibrium Studies of Removal of Pb(II) and Cd(II) Ions from Aqueous Solution by Modified Duolite XAD-761 Resins

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Complete mixing of phenol-formaldehyde polymeric resin (Duolite XAD-761) with anionic surfactant-sodium dioctyl sulphosuccinate (SDOSS) and EDTA-disodium salt (chelating agent) in an aqueous solution led to the formation of a modified resin for the removal of Pb(II) and Cd(II) ions from aqueous solution. The effect of experimental parameters such as pH, contact time and modified resin dosage on the Pb(II) and Cd(II) ions removal was studied. The obtained data were fitted with the Langmuir and Freundlich equations to describe the equilibrium isotherms. The kinetic data were fitted with the pseudo-first-order and pseudo-second-order models. It was found that pH played a major role in the adsorption process. The maximum adsorption capacity for Pb(II) and Cd(II) on modified resin calculated from Langmuir isotherm was found to be 46.66 and 59.52 mg g^{-1}, respectively. The optimum pH range for the removal of Pb(II) and Cd(II) was found to be 4-7 and 4-8, respectively. Langmuir isotherm model was found to best describe the experimental data [$R^2 \geq 0.999$ for Pb(II) and $R^2 \geq 0.954$ for Cd(II)]. The kinetic rates were best fitted to the pseudo-second-order model [$R^2 \geq 0.999$ for Pb(II) and $R^2 = 1$ for Cd(II)]. Thermodynamic study showed the adsorption was a spontaneous process. The metal ions could be successfully recovered from the modified resin by treatment with 7-8 % NaCl. The modified resins thus regenerated could be used again to remove the heavy metal ions. Based on all results, it can also be concluded that it can be evaluated as an alternative adsorbent for removing Pb(II) and Cd(II) from aqueous solution.

Key Words: Pb(II) removal, Cd(II) removal, Duolite XAD-761, Sodium dioctyl sulphosuccinate, EDTA, Adsorption isotherms, Kinetics.

INTRODUCTION

Contamination of aqueous streams by toxic heavy metals through the release of industrial wastewater is a world wide environmental problem. Once heavy metal ions enter the environment, their chemical form largely determines their potential toxicity. It is well established that heavy metals obstruct with functional groups of essential enzymes even at lesser concentration[1]. The Pb(II) ions are discharged to aquatic bodies from many main industrial applications, such as storage battery, manufacturing, printing pigments, fuels, photographic materials and explosive manufacturing[2]. Lead(II) is one of the most toxic heavy metals may causing health problem such as behavioral anomaly, learning disabilities and seizures[3]. Cadmium(II) is also a harmful pollutant originating from metal plating, mining, metallurguical alloying and other industrial operations. Cadmium(II) may causing variety of syndromes and effects including hypertension, renal dysfunction, hepatic injury, teratogenic effects and lung damage[4]. Due to the extreme toxicity of Pb(II) and Cd(II) its selective determination and removal from the polluted sites is of particular important.

The development of chelating resins for heavy metals removal has been increased significantly. Their advantages include good selectivity, pre-concentration factor, binding energy and mechanical stability, easy regeneration for multiple sorption-desorption cycles and good reproducibility in the sorption characteristics[5]. XAD resins show good physical properties such as porosity, uniform high surface area, pore size distribution and chemically homogeneous non-ionic structure. Therefore, they have been used as supports for immobilization of chelating agents for metal removal[6,7].

Separation methods using surfactant micelles as separation media to remove heavy metals from water and wastewater have been extensively studied. The sorption of a surfactant on the external surface of a natural zeolite for heavy metal removal has been studied[8]. The most successful applications of surfactant loaded on XAD-4 for the collection of precious metals from water[9]. Several studies discovered that surfactants in combination with a complexing agent had an even greater capability of extracting heavy metals from contaminated soil[10]. The joint application of surfactants and EDTA improved the metal removal from municipal sludge[11] and purple soil[12]. EDTA can form complexes with heavy metals and thus substantially increase heavy metal removal from contaminated soil[13]. Based on the risk assessment report on EDTA approved by the Technical Meetings of European Union Member state representatives[14], EDTA was reported to have low aquatic toxicity and no bioaccumulation in living organisms through the food chain.

In the present work, attempts were made to prepare a novel modified Duolite XAD-761 resin with the use of non-hazardous sodium dioctyl sulphosuccinate (SDOSS)[15] and EDTA-disodium salt (chelating agent) for the separation of Pb(II) and Cd(II) ions from the aqueous solution. Effects of process variables such as contact time, pH and adsorbent dose on the adsorption capacity were studied. In order to understand the adsorption process by modified XAD-761 resin, adsorption kinetics and equilibrium isotherms were also studied.

EXPERIMENTAL

The AAS of Electronic Corporation India Ltd., (Hyderabad, India), model-SL 163, with air-acetylene burner was used for the determination of metal(II) ion concentration. The pH was measured with an Elico (Hyderabad, India) pH meter (model LI-120 digital). A thermally controlled mechanical shaker with a speed of 200 rpm was used for metal ion-chelating resin equilibration.

Reagents and solutions: All the reagents used in the present study were of analytical AR grade. The stock solution of Pb(II) and Cd(II) was prepared by dissolving an appropriate amount of $Pb(NO_3)_2$ and $CdSO_4.8H_2O$ (BDH, Mumbai, India) in distilled water, respectively. The working solution of metal ion was obtained by the dilution of the stock solution. The pH was adjusted with 0.1 M HNO_3 or 0.1 M HCl or 0.1 N NaOH. Duolite XAD-761 resin (surface area, 150-250 m^2 g^{-1} and mean size, 0.56-0.76 mm) was procured from Rohm Hass (Philadelphia, PA, USA) and their properties are given in Table-1. The sodium dioctyl sulphosuccinate (SDOSS) and EDTA-disodium salt (S.D. Fine Chem, Mumbai, India) were used as such for the preparation of chelating resin.

TABLE-1
PHYSICAL AND CHEMICAL PROPERTIES OF DUOLITE XAD-761 RESIN

Physical and chemical properties	Duolite XAD-761
Matrix	Cross linked phenol-formaldehyde polycondensate
Surface area (m^2/g)	150-250
Specific gravity	1.070-1.130
Particle size (mm)	0.560-0.760
Polarity	Polar (intermediate)
Porosity (mL/g)	0.95-1.18
Average pore diameter (Å)	600
Operating pH range	Up to 8

Source: Information provided by the manufacturer, Rohm and Hass Co., Philadelphia, Pa, USA.

Preparation of adsorbent: The resin duolite XAD-761 was purified with a 50 % ethanol-water solution containing 4 M HCl to remove inorganic impurities and monomeric material. After that, the resin was rinsed thoroughly with distilled water to eliminate chloride ions. For the impregnation purposes, slightly modified version of the dry method was followed[16]. 1 g of pure Duolite XAD-761 resin was well mixed with equal ratio (1:1) mixture of 20 mL of 66.6 % (v/v) water-ethanol mixture containing 2 g of SDOSS and 20 mL of water containing 2 g of EDTA (disodium salt) and beads were left in the mixture for 24 h. The polymeric beads were separated from the solution by filtration, then washed with water and dried at 50 ºC. The amount of impregnated resin was calculated from the material balance. The amount of ligand impregnated onto dry resin was found to be 156 mg g^{-1}.

Batch cxperiments: 100 mL of metal(II) ion solution with desired concentration adjusted to a desired pH were taken in polythene bottles of 300 mL capacity. A required amount of modified duolite XAD-761 resin was added to it. The solutions were agitated for a predetermined period at 30 ± 1 ºC and then resin particles were separated by filtration and the filtrate was analyzed by AAS for Pb(II) and Cd(II) ions. Adsorption isotherm and kinetic studies were carried out with different initial concentrations of Pb(II) and Cd(II) ions by maintaining the modified resin dosage at constant level. Metal ion per cent removal (%) was calculated using the following equation:

$$\text{Removal}\,(\%) = \frac{C_i - C_f}{C_i} \times 100 \quad (1)$$

where C_i and C_f are the initial and final metal ions concentrations, respectively.

RESULTS AND DISCUSSION

Effects on the adsorption capacity of modified Duolite XAD-761 resin: Both surfactant and EDTA-disodium salt (chelating agent) were individually loaded on Duolite XAD-761 resin and then tested for Pb(II) and Cd(II) ions adsorption. However, individually loaded resins could not remove the Pb(II) and Cd(II) ions from the solution effectively. The initial studies indicated that percentages of removals were found to be 75 % for surfactant loaded resin and 40 % for EDTA-disodium salt loaded resin for an initial Pb(II) and Cd(II) ions concentration of 10 mg/L and for a modified resin dosage of 0.2 g/100 mL, respectively. When the complex of surfactant and EDTA loaded on duolite XAD-761 resin, the results showed that both surfactant and EDTA could stimulate the adsorption of metal ions from aqueous solution. Hence the complex effect of surfactant and EDTA (disodium salt) on the polymer has been considered for the present study.

Effect of contact time: Fig. 1 shows the effect of contact time on the removal of Pb(II) and Cd(II) onto modified XAD-761 resin. It could be seen that the removal of Pb(II) and Cd(II) increases with rise in contact time and attains equilibrium at 180 min for both metal ions. Removal efficiency was found to be 99 % for both Pb(II) and Cd(II) ions by modified XAD-761 resin. This indicates that the contact time 180 min required for the quantitative removal of both Pb(II) and Cd(II) ions.

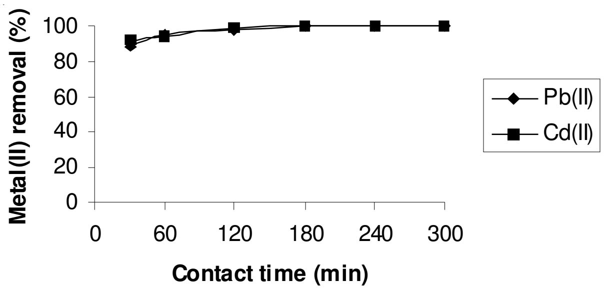


Fig. 1. Effect of contact time on the adsorption of Pb(II) and Cd(II) onto modified XAD-761 resin [metal (II) ion concentration = 10 mg/L, pH = 5, modified resin dosage = 200 mg/100 mL]

Effect of pH: The most important single parameter influencing the sorption capacity is the pH of adsorption medium[17]. The initial pH of adsorption medium is related to the adsorption mechanisms onto the adsorbent surface from water and reflects the nature of the physicochemical interaction of the species in solution and the adsorptive sites of adsorbents[18]. The effect of pH on the removal of Pb(II) and

Cd(II) onto modified XAD-761 resin was studied at pH 1-10 for initial metal ion concentration of 10 mg/L Pb(II) and 10 mg/L Cd(II) solution and shown in Fig. 2. It could be observed that the quantitative removal of Pb(II) and Cd(II) was achieved over the pH range of 4-7 and 4-8, respectively. At pH 2-3 range the adsorption is very low and rapidly increases between pH 4 and 5. This phenomenon can be explained by the surface charge of the modified resin and the H^+ ions present in the solution. At low pH the cations compete with the H^+ ions in the solution for the active sites and therefore lower adsorption. Moreover the stability of the SDOSS-EDTA complex which was adsorbed on the surface of the polymer matrix might be reduced with the result the adsorption of the metal ion decreases at low pH. When the pH values increased, adsorbent surfaces were more negatively charged which results higher attraction of cations. At high pH values the metal complex forms and results precipitation was observed because of the existence of OH^- ions in the adsorption medium at a pH greater than 8.0. Hence, the maximum removal of Pb(II) and Cd(II) ions onto modified XAD-761 resin is at optimum initial pH range 4-7 and 4-8, respectively.

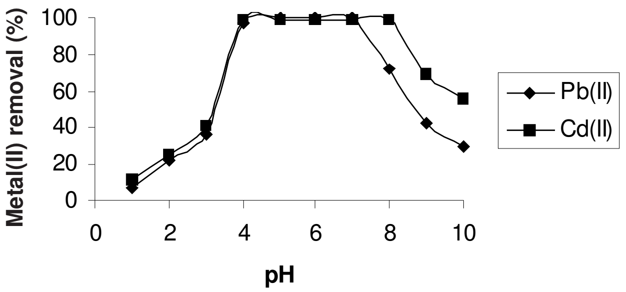


Fig. 2. Effect of pH on the adsorption of Pb(II) and Cd(II) onto modified XAD-761 resin [metal (II) ion concentration = 10 mg/L, equilibrated time = 3 h, modified resin dosage = 200 mg/100 mL]

Effect of modified resin dosage: Effect of modified resin dosage on the removal of Pb(II) and Cd(II) onto modified XAD-761 resin is shown in Fig. 3. A quantitative removal of Pb(II) and Cd(II) ions could be observed at an optimum modified resin dosage of 0.2 g/100 mL for an initial concentration of 10 mg/L of metal ion. Therefore, the maximum removal of both Pb(II) and Cd(II) ions has been achieved at the optimum modified resin dosage 0.2 g/100 mL.

Adsorption isotherms: The equilibrium adsorption isotherm is fundamental in describing the interactive behaviour between adsorbate and adsorbent and is important in the design of adsorption systems. Two well-known equilibrium models, Langmuir and Freundlich models were applied for this study.

Langmuir equation is given as[19,20]:

$$\frac{C_e}{q_e} = \frac{1}{q_0 b} + \frac{C_e}{q_0} \qquad (2)$$

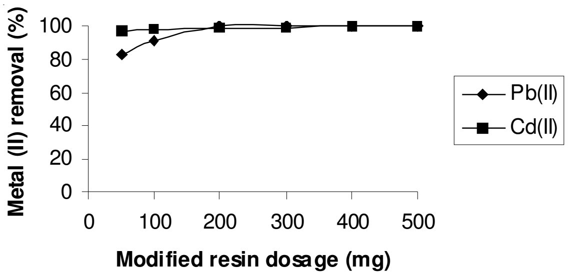


Fig. 3. Effect of modified resin dosage on the adsorption of Pb(II) and Cd(II) onto modified XAD-761 resin [metal (II) ion concentration = 10 mg/L, pH = 5, equilibrated time = 3 h]

where the constants q_0 is the adsorption capacity (mg g^{-1}) and b is related to the surface energy of adsorption (L mg^{-1}). A plot of C_e/q_e *versus* C_e as showed linear plot and values of Langmuir constant (q_0 and b) calculated from the slope and the intercept of the plot (Fig. 4) are presented in Table-2. The essential characteristics of Langmuir isotherm model can be explained in terms of dimensionless constant separation factor or equilibrium parameter R_L which is defined as:

$$R_L = \frac{1}{1 + bC_0} \quad (3)$$

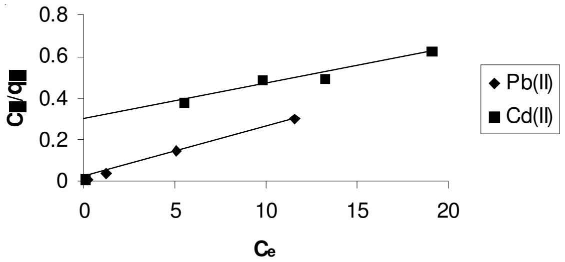


Fig. 4. Langmuir isotherm plots for the adsorption of Pb(II) and Cd(II) onto modified XAD-761 resin [modified resin dosage = 2 g/L, contact time = 24 h, pH = 5]

TABLE-2
LANGMUIR AND FREUNDLICH CONSTANTS FOR Pb(II) AND Cd(II) ADSORPTION

Metal ion	Langmuir model				Freundlich model		
	q_0 (mg g^{-1})	b (L mg^{-1})	R_L	R^2	K_F (mg g^{-1})	n (L mg^{-1})	R^2
Pb(II)	46.66	1.075	0.0398	0.999	14.12	6.50	0.952
Cd(II)	59.52	0.056	0.4077	0.954	3.23	1.60	0.875

where b is the Langmuir constant (L mg^{-1}) and C_0 is the initial concentration (mg L^{-1})[21]. Separation factor shows the nature of adsorption process and its value indicates the sorption process could be favorable, linear, unfavorable, when $0 < R_L < 1$, $R_L = 1$,

$R_L > 1$, respectively. R_L values for the modified XAD-761 resin were in the range of 0-1 (Table-2) and in addition the shape of curve (Fig. 4) confirm the favourable uptake of both Pb(II) and Cd(II) ions onto modified XAD-761 resin. It can be concluded that based on correlation coefficient values (R^2) values (Table-2), Langmuir model seems to be more agreeable indicating the formation of monolayer sorption.

The Freundlich isotherm is represented by the equation[22]

$$\log\frac{x}{m} = \log K_F + \frac{1}{n}\log C_e \tag{4}$$

where C_e is the equilibrium concentration (mg/L) and x/m is the amount of metal ion adsorbed per unit weight of adsorbent. The K_F is the Freundlich constant related to the adsorption capacity (mg g^{-1}) and n shows the adsorption intensity (L mg^{-1}). The linear plot of log (x/m) *versus* log C_e exhibits that the adsorption obeys the Freundlich isotherm and values of Freundlich constant (K_F and n) calculated from the intercept and slope of the plot (Fig. 5) are presented in Table-2. The value of $1 < n < 10$ indicates high probability of adsorption of both Pb(II) and Cd(II) ions onto modified XAD-761 resin. The R^2 values indicated that the Freundlich model does not well fit with the experimental data when compared with Langmuir model.

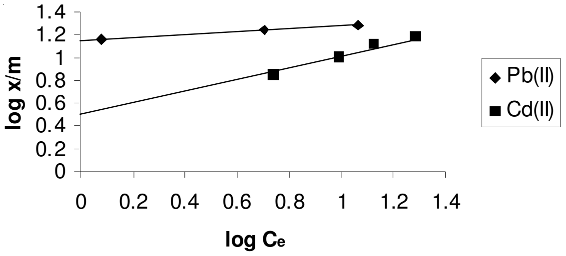


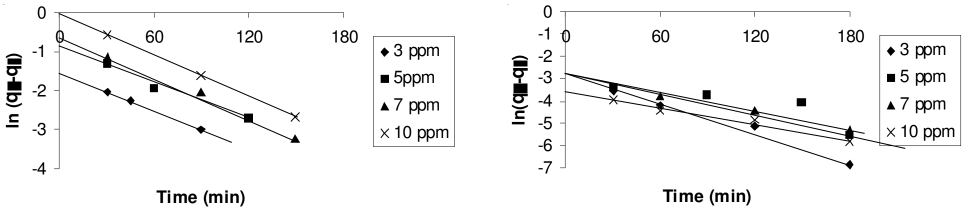
Fig. 5. Freundlich isotherm plots for the adsorption of Pb(II) and Cd(II) onto modified XAD-761 resin (modified resin dosage = 2 g/L, contact time = 24 h, pH = 5)

Adsorption kinetics: In order to clarify the adsorption kinetics of Pb(II) and Cd(II) ions onto modified XAD-761 resin two kinetic models, which are Lagergren's pseudo-first-order and pseudo-second-order model were applied to the experimental data.

The linearized form of the pseudo-first-order rate equation of Lagergren is given as[23],

$$\ln(q_e - q_t) = \ln q_e - k_1 t \tag{5}$$

where q_e and q_t are the amount of the metal ions adsorbed (mg/g) at equilibrium and t (min), respectively and k_1 is the pseudo-first-order equilibrium rate constant (1/min). A plot of $\ln(q_e - q_t)$ *versus* t gives straight line with slope of k_1 and an intercept of $\ln q_e$. It confirms the applicability of the pseudo-first-order rate equation (Fig. 6).



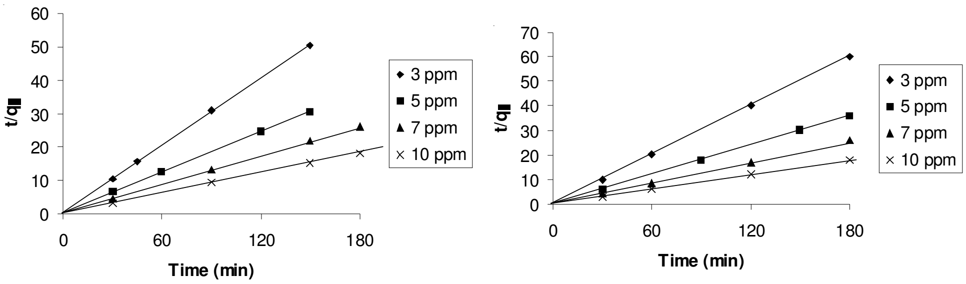
(a) (b)

Fig. 6. Pseudo-first-order kinetics for the adsorption of Pb(II) (a) and Cd(II) (b) onto modified XAD-761 resin (modified resin dosage = 2 g/L, pH = 5)

The pseudo-second-order model rate equation may be expressed as[24],

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{6}$$

where k_2 is the pseudo-second-order adsorption rate constant (g/mg min). A plot of (t/q_t) *versus* t produces straight line with slope of $1/q_e$ and intercept of $1/k_2q_e^2$. It indicates the applicability of pseudo-second-order model (Fig. 7).



(a) (b)

Fig. 7. Pseudo-second-order kinetics for the adsorption of Pb(II) (a) and Cd(II) (b) onto modified XAD-761 resin (modified resin dosage = 2 g/L, pH = 5)

In order to compare quantitatively the applicability of kinetic models in fitting to data, the per cent relative deviation (P), given by the following equation was calculated

$$P = \frac{100}{N} \sum \left\{ \frac{[q_{e(exp)} - q_{e(theo)}]}{q_{e(exp)}} \right\} \tag{7}$$

where $q_{e(exp)}$ is the experimental value of q_e at any value of C_e, $q_{e(theo)}$ the corresponding theoretical value of q_e and N is the number of observations. It is found that lower value of percentage deviation (P), better is the fit. It is generally accepted that when P value is less than 5, the fit is considered to be excellent[25]. The results have been analyzed using eqns. 5 and 6. The experimental data fitted well in both the equations. The values of $q_{e(theo)}$ calculated from these models are compared with experimental values $q_{e(exp)}$ in Table-3. It is found that values of $q_{e(theo)}$ calculated from the pseudo-first-order kinetic model differed appreciably from the experimental values $q_{e(exp)}$. The per cent deviation (P) is also very high. On the other hand, values of $q_{e(theo)}$ are found to be very close to $q_{e(exp)}$ when pseudo-second-order rate equation was applied. The per cent deviation (P) is well with in the range and also the values of correlation coefficients (R^2) are very high to pseudo-second-order kinetics compared with pseudo-first-order kinetics. These results indicated that the adsorption of both Pb(II) and Cd(II) ions onto modified XAD-761 resin follows well the pseudo-second-order kinetics.

TABLE-3
PSEUDO-FIRST-ORDER AND PSEUDO-SECOND-ORDER KINETIC CONSTANTS FOR THE ADSORPTION OF Pb(II) AND Cd(II) ON MODIFIED XAD-761 RESIN

Metal ion	Conc. (mg/L)	Pseudo-first-order kinetic					Pseudo-second-order kinetic				
		k_1 (g/mg min)	$q_{e(theo)}$ (mg/g)	$q_{e(exp)}$ (mg/g)	R^2	P	k_2 (g/mg min)	$q_{e(theo)}$ (mg/g)	$q_{e(exp)}$ (mg/g)	R^2	P
Pb(II)	3	0.0160	0.2163	2.98	0.987	92.74	0.1557	3.01	2.98	0.999	1.24
	5	0.0149	0.3902	4.94	0.979	92.10	0.0832	5.00	4.94	0.999	1.21
	7	0.0125	0.5749	6.92	0.994	91.69	0.0678	6.98	6.92	1.000	0.86
	10	0.0113	0.9430	9.90	0.989	90.47	0.0401	0.02	9.90	0.998	1.21
Cd(II)	3	0.0216	0.0750	2.99	0.977	97.49	0.9250	2.99	2.99	1.000	0
	5	0.0146	0.0580	4.99	0.841	98.83	0.7201	4.99	4.99	1.000	0
	7	0.0126	0.0501	6.98	0.995	99.28	0.6630	6.98	6.98	1.000	0
	10	0.0116	0.0267	9.95	0.970	99.73	0.4610	9.95	9.94	1.000	0.1

The free energy of adsorption (ΔG°) can be related with the equilibrium constant K (L mol^{-1}), corresponding to the reciprocal of the Langmuir constant, b, by the following equation[26,27],

$$\Delta G^o = -RT \ln b \tag{8}$$

where R is the universal gas constant (8.314 J mol^{-1} K^{-1}) and T is the absolute temperature (K). Gibbs free energy change (ΔG°) was calculated to be -30.50 kJ mol^{-1} for Pb(II) and -21.71 kJ mol^{-1} for Cd (II). Negative values of ΔG° indicated the feasibility of the process and spontaneous nature of the adsorption. However, the removal mechanism of metal (II) ions adsorption is not well understood. It may further be added that the modified XAD-761 resin extracts metal (II) ions from solution by ion exchange/chelation through electrostatic attraction.

Desorption studies: Desorption studies were conducted to recover the Pb(II) and Cd(II) ions from the modified XAD-761 resin. Attempts were made to desorb Pb(II) and Cd(II) ions from the spent resins using sodium chloride (1-10 %). The final Pb(II) and Cd(II) concentration in the aqueous phase was determined by using atomic absorption spectrophotometer. Results showed that 99 % of both Pb(II) and Cd (II) desorbed from the resin under optimum concentration of 7 and 8 % NaCl, respectively. After the extraction of Pb(II) and Cd(II) from modified resin, it was washed completely with distilled water. The adsorption capacity of the modified XAD-761 resin was again tested 5 cycles of operation. Results showed that adsorption capacity of NaCl regenerated modified XAD-761 was maintained over the range 99-92 % for both Pb(II) and Cd(II) even up to 5 cycles of operation.

Conclusion

This study focused on the removal of Pb(II) and Cd(II) ions onto modified XAD-761 resins from aqueous solution. Separation media was prepared by just mixing the resins in aqueous solution of anionic surfactant and EDTA. Thus, without chemical synthesis, chelating functionality was easily introduced on the solid surface. The operating parameters such as pH of the solution, modified resin dosage and contact time were effective on the sorption efficiency of Pb(II) and Cd(II). The adsorption capacity was found to be 46.66 for Pb(II) and 59.52 mg g^{-1} for Cd(II). The kinetic data signified that the adsorption of Pb(II) and Cd(II) onto modified resin followed well the pseudo-second-order kinetic model. The ΔG^o values showed the feasibility and spontaneous nature of the adsorption. The metal adsorbed modified resin could be regenerated by using inexpensive regenerate reagent such as 7-8 % NaCl. It has been found out that regenerated modified resins could be again and again used for at least 5 cycles of operation without major loss in adsorption capacity and loss of material. Furthermore, it can be evaluated as an alternative adsorbent for removing Pb(II) and Cd(II) from aqueous solution, since it has considerable high adsorption capacity.

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