



## Uranium Ions Removal Using Amberlite CG-400 Anion Exchanger Resin in the Presence of Sulfate Anions

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### A B S T R A C T

Uptake of uranium (VI) ions using an anion exchanger resin, namely Amberlite CG-400, was studied through this work in the presence of sulfate anions. Impact of various parameters including pH, contact time, initial concentration of uranium ions and intrusion with different anion contents (phosphate, chloride, sulfate, nitrate and fluoride) was examined on the sorption performance of the resin. Results showed that sorption of uranium ions on the resin occurs considerably at pH=4 and higher. Kinetics of the sorption process was very fast and uranium sorption percentage reached to its maximum at very short time. An estimation of sorption process was also performed using isotherm and kinetics equations, which led to conformity of Freundlich and pseudo-second-order models with experimental data, respectively. The maximum sorption capacity ( $q_{max}$ ) was found to be  $63.29 \text{ mg.g}^{-1}$ . Presence of various anions species in solution showed that uranium sorption using Amberlite CG-400 anion exchange resin could strongly be affected. Presence of sulfate ions in the solution led to an increase in the sorption of uranium content by resin. Phosphate ions did not interfere with uranium ions sorption, however, fluoride, nitrate and chloride existence in the medium caused a decline in the resin sorption's strength.

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## 1. INTRODUCTION

Environmental contamination caused by heavy metals including Pb(II), Hg(II), Co(II) etc have been concerned [1-3] due to their ease liberation into runoffs and ground water bodies where they have chemical interaction with living organisms followed by severe casualties for ecosystem.

Except aforementioned elements, advent of nuclear industry in the last century has emerged as another source of contamination in realm of heavy metals (i.e. uranium) [4]. Uranium propagation in the environment has been a major concern for two reasons: (1) its chemical reaction with other substances and in particular living things, and (2) its radioactivity impact [5]. The former should be more considered because of

its irretrievable effects. Getting exposed to uranium radioactivity (i.e. external contamination) has deterministic and stochastic effects including erythema, central nervous and GI syndromes. Moreover, accommodation of uranium fine particles in respiratory system causes another type of contamination (i.e. internal contamination) in which internal organs receive a huge amount of radiation energy through  $\alpha$ ,  $\beta$  or  $\gamma$  rays emitting from uranium and its daughter, radon [6, 7].

By now, many techniques have been exploited for removal of uranium from aqueous solution [8-11]. Out of those, adsorption deserves a particular position because of its advantages such as low cost of operation, operational simplicity, and use of different adsorbents. Surprisingly, using a variety of sorbents is a significant ability that converts the adsorption to a suitable technique for removal of hazardous materials [12-14].

Up to now, many researchers have examined uranium removal using different methods and even by

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different sorbents using adsorption technique [15-17]. Functionalized molecular sieves, mesoporous and nanoporous sorbents, strongly basic ionic exchangers, and impregnated nanocomposites are just some instances [18-20]. However, uranium adsorption in the presence of interfering anion contents whether single or binary has not been studied yet. As a matter of fact, it has been proved that presence of different species could adversely affect uranium removal using different adsorbents. Therefore, we focus on finding a new type of sorbent which is able to absorb uranium species from the solution, selectively. Effective materials that could improve uranium-bearing effluent's treatment for the final disposal into environment are required; such type of new sorbents should be traced in the realm of anionic exchanger resins.

The first part of this work is focused on investigation of impact of different operational parameters on uranium (VI) ions removal using an anionic exchanger resin, namely Amberlite CG-400. Contact time, initial concentrations of uranium, pH, and interference anions consist the operational parameters of this study. The last part of this work is devoted to mathematical description of sorption mechanisms in both isotherm and kinetics views.

## 2. EXPERIMENTAL

**2.1. Materials** Anion exchanger resin, namely Amberlite CG-400 (mesh size: 200-400) was provided from Rohm and Haas chemical Co. (France). Uranium nitrate,  $(\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O})$ , was used in the current study as source of uranium ions for sorption experiments. Sodium sulfate,  $(\text{Na}_2(\text{SO}_4)_2)$ , was also utilized as source of sulfate anions. Other minor reagents used in the present experiment were of analytical grade from Merck (Darmstadt, Germany).

**2.2. Sorption Experiments** Implementation of all adsorption experiments was initiated using 20 mg of Amberlite CG-400 through a batch method. In a typical experiment, 10 ml solution containing uranium (VI) ions of given concentration and the resin were introduced to a beaker. Additionally, sulfate anions with determined concentration of  $2655 \text{ mg.l}^{-1}$  existed in the solution to simulate the real effluent circumstances. The solution was shaken for a given time using a water bath shaker and then filtered. Subsequently, the amount of uranium ions in residual solution was measured using an inductively coupled plasma-atomic emission spectrometry (ICP-AES). Radical parameters for adsorption including adsorption capacity ( $q$ ,  $\text{mg.g}^{-1}$ ) and adsorption percentage (Ads. %) were calculated using following equations (Equations (1) and (2)):

$$q(\text{mg.g}^{-1}) = (C_i - C_f) * \frac{V}{m} \quad (1)$$

$$\% \text{Ads.} = \left( \frac{C_i - C_f}{C_i} \right) * 100 \quad (2)$$

in which  $C_i$  and  $C_f$  stand for initial and final concentrations of uranium ions before and after sorption ( $\text{mg.l}^{-1}$ ),  $V$  is the volume of uranium ions solution (l),  $m$  is the mass of resin (g), and  $q$  is the adsorption capacity of the Amberlite CG-400 ( $\text{mg.g}^{-1}$ ).

Since pH has an important role on sorption behavior, the pH effect on uranium ions removal was investigated at room temperature for 2 h. Typically, 10 ml uranium ions solution with 20 mg Amberlite CG-400 anion exchanger resin were used for pH tests over ranges from 2.3 to 7.9. Adjustment of the pH was performed using NaOH/HCl solution.

For investigating the contact time impact on Amberlite CG-400 performance, uranium uptake tests were conducted. On that account, uranium ions concentration of the treated solution was measured at different intervals up to 24 h.

One factor that leads to changing the sorbent efficacy is initial ion concentration of the sorbate. Accordingly, the initial uranium ion concentration impact on sorption behavior of Amberlite CG-400 anion exchanger resin was also evaluated. To do so, a concentration range from 2.24 to  $125 \text{ mg.l}^{-1}$  of uranium (VI) ions was employed.

Besides abovementioned factors, the effect of presence of different anion species in the solution on uranium sorption using Amberlite CG-400 anion exchanger resin was also investigated. Chloride, fluoride, nitrate, phosphate and sulfate anions with different concentrations were species which their interferences with uranium ions on sorption performance of the resin were tested.

In addition to investigation of the initial concentration and contact time effects on uranium sorption, these phenomena can mathematically be described for having a better estimation of the process. Isotherm analysis, which is carried out to identify the relation between adsorbed ions on the sorbate with that existing in the residual solution was performed with several binding models. The most used isotherm models, Langmuir and Freundlich isotherms, were exploited in this study. The Langmuir sorption isotherm model is based on the assumption that all the sorption sites are energetically identical and sorption occurs on a structurally homogeneous sorbent [21]. The Freundlich isotherm model depicts both multilayer sorption and sorption on heterogeneous surface.

The isotherms are represented by following the equations (Equations (3) and (4)):

Langmuir isotherm [22]:

$$\frac{C_e}{q_e} = \frac{1}{q_{\max}k_1} + \frac{C_e}{q_{\max}} \quad (3)$$

Freundlich isotherm [22]:

$$\log q_e = \log k_f + \frac{1}{n}C_e \quad (4)$$

where  $q_e$  ( $\text{mg.g}^{-1}$ ) and  $q_{\max}$  ( $\text{mg.g}^{-1}$ ) are the amount of uranium ions adsorbed per unit weight of the resin and the maximum sorption capacity, respectively.  $C_e$  ( $\text{mmol.l}^{-1}$ ) is the equilibrium concentration of the uranium ions in solution, and  $k_1$  ( $\text{l.mmol}^{-1}$ ) is Langmuir isotherm constant, which relates to the energy of sorption.  $k_f$  ( $\text{mg.g}^{-1}.\text{mmol}^{-1}$ ) is the measure of sorption capacity and also referred to as adsorption capacity, while  $n$  is the heterogeneity factor and related to adsorption intensity.

As for distinction of the mass transfer step in the sorption process, based on data acquired by contact time experiment, kinetics analysis using pseudo first-order and pseudo second-order models were performed. The kinetics equations are modeled as follows (Equation (5) and (6)):

Pseudo first-order model [23]:

$$\log \left( \frac{q_e}{q_e - q_t} \right) = \left( \frac{K_1}{2.303} \right) t \quad (5)$$

Pseudo second-order model [23]:

$$\frac{t}{q_t} = \frac{1}{K_2q_e^2} + \frac{t}{q_e} \quad (6)$$

where  $K_1$  ( $\text{l.min}^{-1}$ ) and  $K_2$  ( $\text{g.mg}^{-1}.\text{min}^{-1}$ ) stand for the rate constant of adsorption. Values of  $K_1$  and  $K_2$  were calculated from the straight line graph of  $\log(q_e - q_t)$  versus  $t$  and  $(t/q_t)$  versus  $t$  for different initial concentrations of the uranium (VI) ion, respectively.

### 3. RESULTS AND DISCUSSION

#### 3. 1. pH Effect

Figure 1 illustrates plot of pH versus adsorption efficiency in which the pH range changes from 2.3 to 7.9 along with the presence of sulfate anions. The pH effect on sorption performance of Amberlite CG-400 anion exchanger resin is remarkably considerable. Movement from acidic region toward alkaline zone improved sorption mechanism. Variation of pH amount could affect uranium species and convert them to a different form which is appropriate for the resin to absorb them. Also, the presence of sulfate anions in medium might take part in this process, nevertheless, the determined effect of  $\text{SO}_4^{2-}$  anions does not proved yet.

It is obvious that when pH is fixed at 4 or even higher values, the maximum sorption efficiency occurs. This shows that declining  $\text{H}^+$  ions concentration in medium possibly convokes the resin to engage with uranium species, effectively. Conceivably, presence of sulfate anions in medium and increasing the hydroxyl

anions cause all of the uranium species at  $\text{pH} < 2$  (i.e.  $\text{UO}_2^{2+}$ ) to convert to anionic species such as  $\text{UO}_2\text{SO}_4^{2-}$  and  $(\text{UO}_2)\text{OH}_4^{2-}$  at higher pHs whose affinity towards Amberlite CG-400 anion exchanger resin are great.

#### 3. 2. Isotherm Study

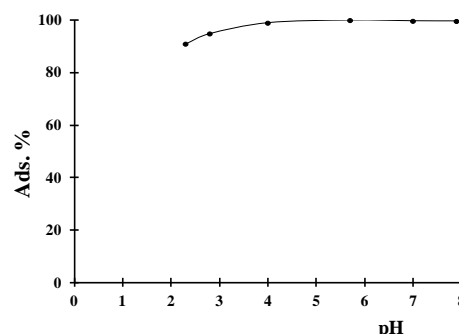
The result of changing initial concentration on sorption capacity is shown in Figure 2a. A striking increase in sorption capacity is achieved by increasing the amount of initial uranium ions in the solution and after a given concentration, it can be seen that the equilibrium condition dominates. Using the binding models, isotherm analysis result is given in Figures 2b and 2c and the constant parameters are listed in Table 1.

The Langmuir model excellently agreed with the experimental data compared to the Freundlich model. Thus, description of the sorption process relates to homogenous surfaces with identical binding sites. Eventually, calculated maximum sorption capacity ( $q_{\max}$ ) was  $63.29 \text{ mg.g}^{-1}$ .

#### 3. 3. Kinetics Study

Adsorption percentage of Amberlite CG-400 anion exchanger resin versus contact time is plotted in Figure 3a. The graph shows that at the early stage of the test, the sorbent efficiently adsorb uranium ions and the adsorption reach to an equilibrium situation.

Uranium ions sorption rate was analyzed using pseudo first-order and second-order equation to determine the rate-limiting step of the sorption process and the corresponding plots were also sketched (Figures 3b and 3c). The evaluation of experimental data gives the proof that pseudo second-order model has satisfactory estimation of kinetics data. Based on correlation coefficient ( $R^2$ ), compromising of pseudo second-order model with experimental data shows that the concentration of both adsorbate (uranium ions) and adsorbent (Amberlite CG-400) are involved in the rate-controlling step of the sorption mechanism. Constant parameters of both kinetics models are listed in Table 2.



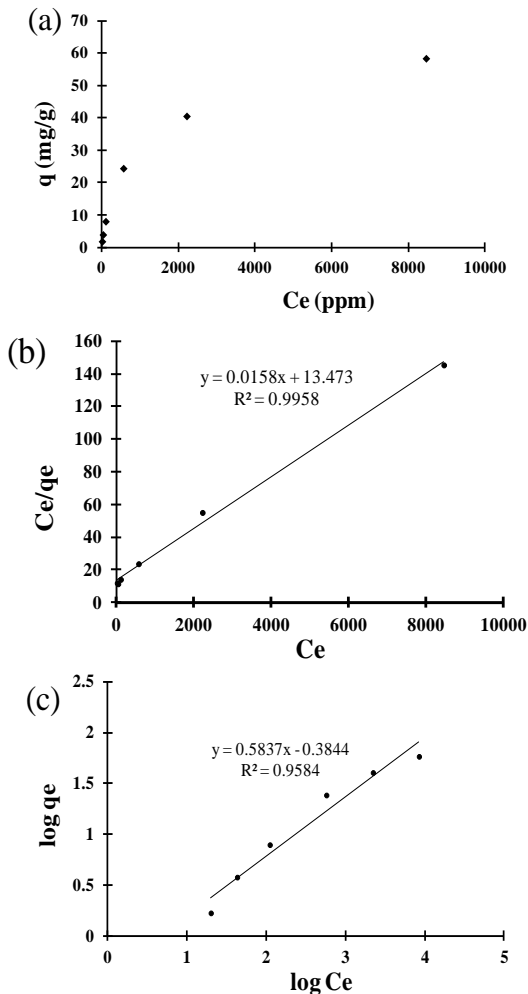
**Figure 1.** Plot of pH effect on uranium (VI) sorption using Amberlite CG-400 anion exchanger resin Conditions:  $C_0(\text{U})$ :  $5 \text{ mg.l}^{-1}$ , sulfate concentration:  $2655 \text{ mg.l}^{-1}$ , agitation time: 2 h, and temperature:  $25^\circ\text{C}$

**TABLE 1.** Isotherm parameters for uranium (VI) sorption onto Amberlite CG-400 anion exchanger resin

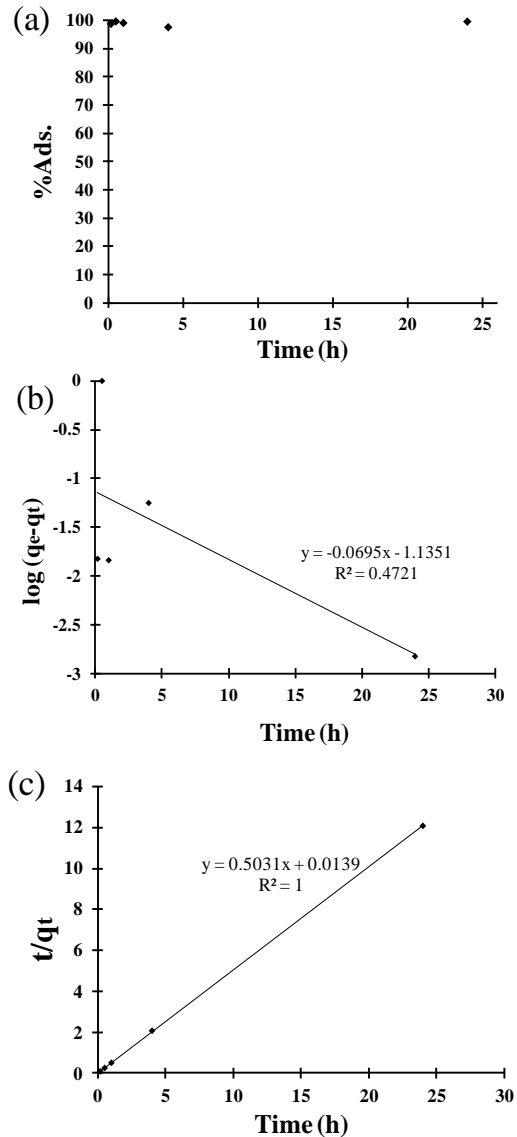
Isotherm		Parameters	
Langmuir equation	$q_{max}$ (mg.g <sup>-1</sup> )	$K_L$ (L.mM <sup>-1</sup> )	$R^2$
	63.29	0.001	0.996
Freundlich equation	$n$	$K_f$ (mg.g <sup>-1</sup> .mM <sup>-1</sup> )	$R^2$
	1.71	0.412	0.959

**TABLE 2.** Kinetics constants for uranium sorption using Amberlite CG-400 anion exchanger resin

Pseudo first-order	$k_1$ (min <sup>-1</sup> )	0.16
	$q_e$ (mg.g <sup>-1</sup> )	0.0732
	$R^2$	0.472
Pseudo second-order	$k_2$ (g.mg <sup>-1</sup> .min <sup>-1</sup> )	18.22
	$q_e$ (mg.g <sup>-1</sup> )	1.987
	$R^2$	1



**Figure 2.** Initial concentration versus sorption capacity graph for uranium removal using Amberlite CG-400: (a) linearized Langmuir, (b) Freundlich and (c) graphs. Conditions: initial pH value: 4 sulfate concentration= 2655 mg.l<sup>-1</sup>, agitation time: 2 h, and temperature= 25°C.



**Figure 3.** (a) Contact time versus adsorption efficiency of Amberlite CG-400 anion exchanger resin plot, (b) sorption kinetics plot of pseudo first-order model and (c) linearized plot for experimental data of pseudo second-order kinetics model

**3. 4. Adsorptive Behavior in the Presence of Various Anions on Uranium Sorption**

Different research groups studied the presence of different anionic species on uranium sorption system [24]. Interfering anions regularly showed negative effects on sorption behavior of many adsorbents. For revealing strength of the resin for uranium uptake in the presence of anionic species, sorption experiments were carried out using phosphate, sulfate, chloride, fluoride, and nitrate ions with a varying range of concentration.

As shown in Figure 4, existing the SO<sub>4</sub><sup>2-</sup> ions effectively improve uranium sorption using Amberlite

CG-400, while other anions including  $\text{Cl}^-$ ,  $\text{NO}_3^-$  and  $\text{F}^-$  cause a descending trend on uranium sorption. This shows that affinity of these ions are in compete with uranium ions on active sites of the resin. Nevertheless, it is clear from the graph that high concentration of  $\text{NO}_3^-$  and  $\text{F}^-$  just degenerate the sorption mechanism and at low concentration of all anions the uranium sorption performance is fairly appropriate. The figure also depicted the effect of phosphate anions,  $\text{PO}_4^-$ , on adsorption percentage and the result reveals that the anion has not any converse effect on uranium sorption.

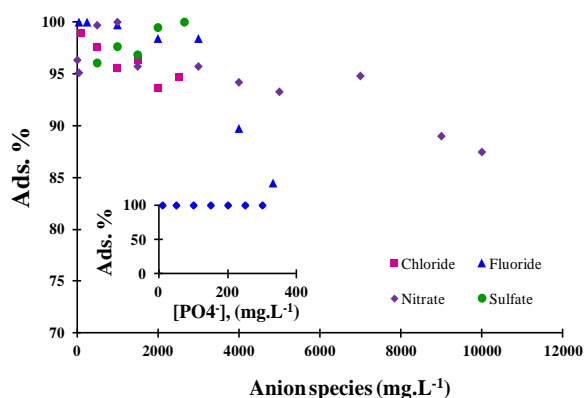
### 3. 5. Comparison Between Sorption Capacities of CG-400 Anion Exchanger Resin with Other Sorbents in Literature

In comparison between the sorption capacity, Table 3 provides a list of some sorbents with their sorption capacity for uranium sorption in the literature [25, 26] compared to that of our work.

As seen in Table 3, all of those strong base anionic sorbent had same strength in sorption of uranium content. Our sorbent also showed a remarkable amount of sorption capacity for uranium removal which lies between the range of that reported in the literature [25].

**TABLE 3.** Sorption capacity of different sorbents for uranium sorption from literature compared to the sorbent of current study

Sorbent	Sorption capacity ( $q_{\max}$ ) $\text{mg.g}^{-1}$	Ref.
IRA 910U	66 to 108	[26]
Dowex A	53 to 79	[26]
Amberlite CG-400	57.14 to 112.36	[25]
Amberlite CG-400	63.29	This work



**Figure 4.** Effect of anionic species on uranium (VI) removal [Sulfate (0-2655  $\text{mg.l}^{-1}$ ), nitrate (0-1000  $\text{mg.l}^{-1}$ ), chloride (0-2530  $\text{mg.l}^{-1}$ ), fluoride (0-4850  $\text{mg.l}^{-1}$ ), phosphate (0-300  $\text{mg.l}^{-1}$ )]

## 4. CONCLUSION

We concisely demonstrated here the sorption mechanism of uranium ions using Amberlite CG-400 anion exchanger resin in the presence of sulfate anions. It revealed that the adsorption could strongly occur at pH 4 and higher values. Amberlite CG-400 showed a maximum sorption capacity of  $63.29 \text{ mg.g}^{-1}$  for uranium sorption in the presence of sulfate ions. Isotherm modeling analysis revealed that the sorption process relates to homogenous surfaces with identical binding sites. Outcome of kinetics studies imply that the concentration of both adsorbate (uranium ions) and sorbent (Amberlite CG-400) are involved in the rate-controlling step of the sorption mechanism. In the batch experiment the effect of presence of high concentration of sulphate, nitrate, chloride, fluoride on uranium uptake was examined. It is concluded that sulphate and phosphate ions had no interference on uranium sorption which can be the result of co-ion effect, whereas existence of anionic species in the solution including  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{F}^-$  anions showed a significant decline in the uranium sorption performance.

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جذب یون‌های اورانیوم (VI) با استفاده از یک رزین تبادلگر آنیونی، Amberlite CG-400، در حضور آنیون های سولفات در اینجا مورد مطالعه قرار گرفته است. اثر پارامترهای مختلفی شامل pH، زمان تماس، غلظت اولیه یون های اورانیوم و مزاحمت محتوای آنیونی مختلف (فسفات، کلرید، سولفات، نیترات و فلئوئور) بر روی رفتار جذبی رزین آزمایش شده است. نتایج نشان داد که جذب اورانیوم بر روی رزین به صورت قابل توجهی در pH=4 و بالاتر رخ می دهد. سینتیک جذب یون های اورانیوم بسیار سریع بوده و درصد جذب اورانیوم در مدت زمان کوتاهی به بیشترین مقدار خود رسید. تخمین فرآیند جذب با استفاده از داده های ایزوترمال و سینتیک انجام گردید که به ترتیب به تطابق مدل های فروندلیچ و شبه درجه دوم با داده های آزمایشی منتج شد. بیشترین ظرفیت جذب ( $q_{max}$ ) برابر با  $63.29 \text{ mg.g}^{-1}$  بود. حضور گونه های آنیونی مختلف در محلول نشان می دهد که جذب اورانیوم با استفاده از رزین تبادلگر آنیونی Amberlite CG-400 می تواند تا حد زیادی تحت تاثیر قرار بگیرد. حضور یون های سولفات در محلول باعث افزایش جذب گونه ی اورانیوم توسط رزین می گردد. یون های فسفات در جذب یون های اورانیوم هیچ گونه تداخلی ایجاد نکرده، اما حضور یونهای فلئوئورید، نیترات و کلرید در محلول باعث کاهش قدرت جذب رزین می گردد.

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