



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

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### ABSTRACT

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. Result 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 was 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 mg g<sup>-1</sup>. Presence of various anions species in solution show that uranium sorption using Amberlite CG-400 anion exchange resin could strongly be affected. Presence of sulfate ions in the solution leads to an increase in the sorption of uranium content by resin. Phosphate ions have not interfered with uranium ions sorption, however fluoride, nitrate and chloride existence in the medium cause a decline in the resin sorption's strength.

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## 1. INTRODUCTION

Environmental contamination with heavy metals including Pb(II), Hg(II), Co(II) etc have been so far concerned [1-3] due to ease liberation of them into runoffs and ground water bodies of which having chemical interaction with living organisms followed 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 of 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 using 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 sorbent using adsorption technique [15-17]. Functionalized molecular sieves mesoporous and nanoporous, strongly basic ionic exchangers, and impregnated nanocomposites are just some instances [18-20]. However, uranium adsorption in presence of interference anions contents whether single or binary has been not studied yet. As a matter of fact, it was proved that presence of different species could adversely affect uranium removal using different adsorbent. Therefore, we focus on finding a new type of sorbent in which be able to absorb uranium species from the solution, selectively. Requiring to effective materials that could improve uranium-bearing effluent's treatment for the final disposal into environment, 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** An anion exchanger resin, namely Amberlite CG-400 (mesh size: 200-400), utilized in this experiment 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,  $(\text{Na}_2\text{SO}_4)$ , was also utilized as source of sulfate anions. Other minor reagents used in the present experiment were of analytical grade from E. 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 route, 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 due to simulation the real affluent circumstances. The solution was shaken for a given time using a water bath shaker and then was 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 concentration 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. and adjustment of the pH amount was executed 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 led 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.

Beside of abovementioned factors, the effect of presence of different anions 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 ranging concentration were species which their interferences with uranium ions on sorption performance of the resin were tested.

In addition to investigation 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 what relation had the amount of sorbed ions on the sorbate with that of exists in the residual solution, has performed with several binding models. The most used isotherm models, Langmuir and Freundlich isotherms, were exploited in this study as well. 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 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 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 used. 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]

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \quad (6)$$

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 change 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 of which is appropriate for the resin to absorb them. Also, the presence of sulfate anions in medium might took part in this process, nevertheless, the determined effect of  $\text{SO}_4^{2-}$  anions does not proved yet.

It is obvious that when pH fixed in 4 or even higher, the maximum sorption efficiency occurred. 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.  $\text{UO}_2^{2+}$ ) convert to anionic species such as  $\text{UO}_2\text{SO}_4^{2-}$  and  $(\text{UO}_2)\text{OH}_2^{2-}$  at higher pHs whose their affinity towards Amberlite CG-400 anion exchanger resin are great.

#### 3. 2. Isotherm Study

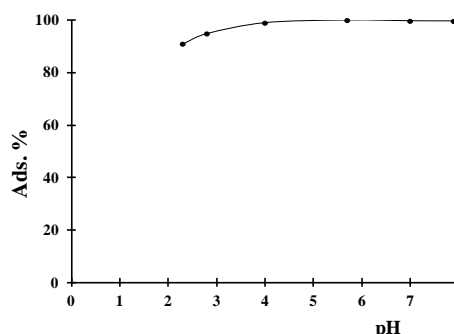
The result of changing initial concentration effects on sorption capacity is shown in Figure 2a. A striking increase at sorption capacity is achieved by increasing the amount of initial uranium ions in the solution and after a given concentration, it can be seen that the equilibrium condition dominates. 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 result excellently compromised with the experimental data than the Freundlich. 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 was plotted in Figure 3a. The graph showed that at the early of the test, the sorbent efficiently could absorb uranium ions and the adsorption reach to an equilibrium situation.

Uranium ions sorption rate was then analyzed using the 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 sorbent (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.



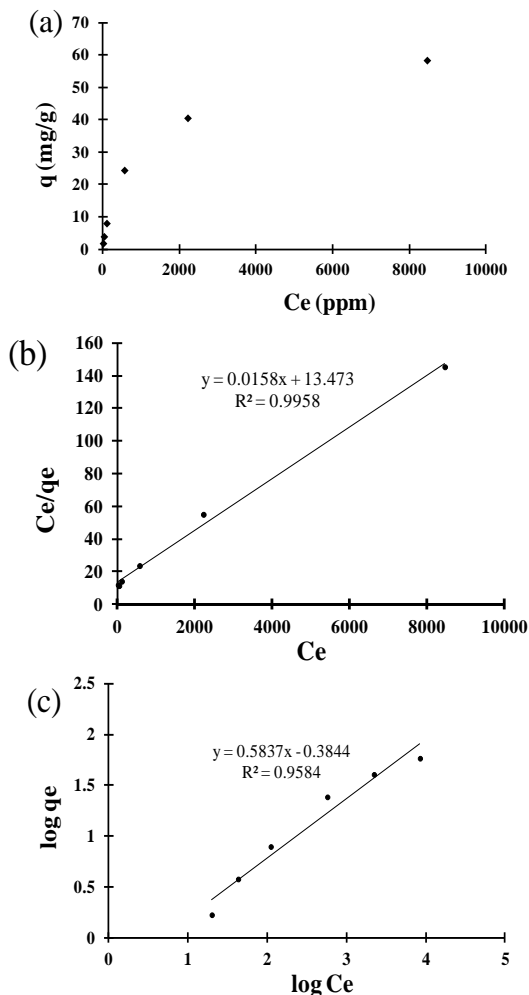
**Figure 1.** Plot of pH effect on uranium (VI) sorption using Amberlite CG-400 anion exchanger resin  
Conditions:  $C_0(\text{U})$ :  $5 \text{ mg L}^{-1}$ , sulfate concentration:  $2655 \text{ mg L}^{-1}$ , agitation time: 2 h, and temperature:  $25^\circ\text{C}$

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

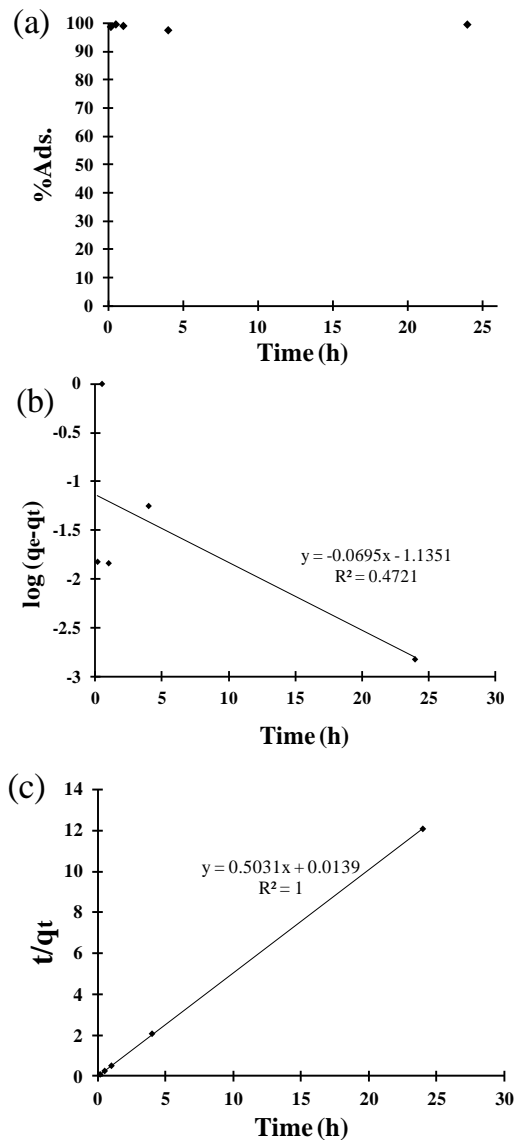
| Isotherm            |                                 | Parameters                                   |       |
|---------------------|---------------------------------|--|-------|
| Langmuir Equation   | $q_{max}$ (mg g <sup>-1</sup> ) | $K_L$ (L mM <sup>-1</sup> )                  | $R^2$ |
|                     | 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) and Freundlich (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 on U(VI) 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 have studied the presence of different anionic species on uranium sorption system [24]. Interference anions have 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 vary 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 by the resin. This shows that affinity of these ions are in compete with uranium ions on active sites of the resin. Nevertheless, it also 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 fairly is 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.

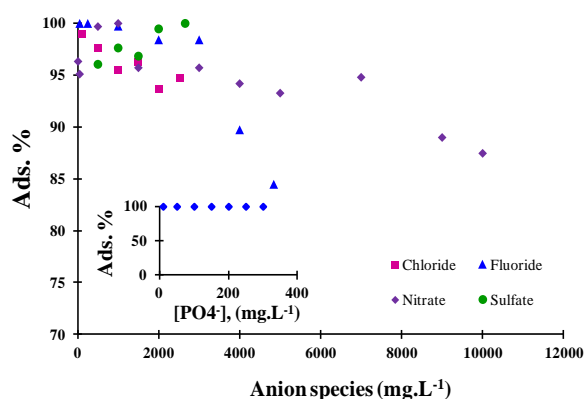
### 3. 5. Comparison Between Sorption Capacities of CG-400 Anion Exchanger Resin with Other Sorbents in Literature

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

As seen in Table 3, all of those strong base anionic sorbent have 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 of reported in [25].

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

| Sorbent          | Sorption capacity ( $q_{\max}$ ) | Ref.      |
|------------------|----------------------------------|-----------|
|                  | $\text{mg}\cdot\text{g}^{-1}$    |           |
| 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  $\text{mg}\cdot\text{L}^{-1}$ ), nitrate (0-1000  $\text{mg}\cdot\text{L}^{-1}$ ), chloride (0-2530  $\text{mg}\cdot\text{L}^{-1}$ ), fluoride (0-4850  $\text{mg}\cdot\text{L}^{-1}$ ), phosphate (0-300  $\text{mg}\cdot\text{L}^{-1}$ )]

## 4. CONCLUSION

We concisely demonstrate 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 the higher. Amberlite CG-400 showed a sorption capacity of 58.26  $\text{mg}/\text{g}$  for uranoium sorption in the presence of sulfate ions. 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. The results obtained by batch experiment examined the presence of high concentration of sulphate, nitrate, chloride, fluoride on uranium uptake. It is concluded that sulphate and phosphate ions had no interference on uranium sorption which can be 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 on the uranium sorption performance.

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Heavy Metals

جذب یون‌های اورانیوم (VI) با استفاده از یک رزین تبادلگر آنیونی، Amberlite CG-400، در حضور آنیون‌های سولفات در اینجا مورد مطالعه قرار گرفته است. اثر پارامترهای مختلفی شامل pH، زمان تماس، غلظت اولیه یون‌های 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 می‌دهد. سینتیک جذب یون‌های اورانیوم بسیار سریع بوده و درصد جذب اورانیوم در مدت زمان کوتاهی به بیشترین مقدار خود رسید. تخمین فرآیند جذب با استفاده از داده‌های ایزوترمال و سینتیک انجام گردید که به ترتیب به تطابق مدل‌های فروندلیچ و شبه درجه دوم با داده‌های آزمایشی منتج شد. بیشترین ظرفیت جذب ( $q_{max}$ ) برابر با  $63.29 \text{ mg g}^{-1}$  بود. حضور گونه‌های آنیونی مختلف در محلول نشان می‌دهد که جذب اورانیوم با استفاده از رزین تبادلگر آنیونی Amberlite CG-400 می‌تواند تا حد زیادی تحت تاثیر قرار بگیرد. حضور یون‌های سولفات در محلول باعث افزایش جذب گونه‌ی اورانیوم توسط رزین می‌گردد. یون‌های فسفات در جذب یون‌های اورانیوم هیچ گونه تداخلی ایجاد نکرده، اما حضور یونهای فلئوئورید، نیترات و کلرید در محلول باعث کاهش قدرت جذب رزین می‌گردد.

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