



Inorganic silica gel functionalized tris(2-aminoethyl)amine moiety for capturing aqueous uranium (VI) ion

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Abstract

The silica gel functionalized tris(2-aminoethyl)amine moiety (SG-TAEA-NH₂) has been successfully used for capturing aqueous uranium ion by batch sorption. Various sorption experiments are performed using several variables such as pH, initial concentration, contact time, and temperature. These variables enable us to study the thermodynamic and kinetic of sorption, which in turn, leads to know more about the interaction and behavior of the uranium ion on the surface. The equilibrium of sorption can be achieved within the first $t=5-10$ min upon the study conditions ($C_i=1$ mg L⁻¹, $T=25$ °C, 80 rpm, $pH_i=7$, and dosage = 2 g L⁻¹). The sorption of U(IV) ion onto the surface of SG-TAEA-NH₂ material. The sorption of U(VI) ion follows the Freundlich isotherm model ($R^2 > 0.999$). The motivation of sorption is due to the chemisorption of U(IV) ion onto an amino-active site forming a complex in the surface, which is proven through the values of (1) the Dubinin–Kaganer–Radushkevich sorption energy (ca. $E = -24$ to -36), (2) the pseudo-second-order kinetic model ($R^2 > 0.999$) and (3) the spectrum of the FTIR. The rate constant and sorption capacities are calculated. Based on these promising results, we recommend using SG-TAEA-NH₂ as an effective adsorbent and filter to remove uranium ion from the water up to 99%.

Keywords Silica gel · Tris(2-aminoethyl)amine · Uranium ion · Sorption · Freundlich isotherm · Pseudo-second order

Introduction

Jordanian governments tend to build nuclear reactors for peaceful purposes such as research purposes and to generate electricity. Uranium is considered the main source for the operation of nuclear plants, as a nuclear fuel in nuclear water, to generate energy. However, it is possible to produce amounts of the nuclear power reactor effluents containing uranium ion [1, 2]. When conditions are available and sufficient to oxidize uranium element turns into enriched uranyl ion (UO₂²⁺) [3–5]. Generally, uranium exists in solution as soluble UO₂²⁺ ion in the form of carbonate complexes (UO₂)₂CO₃(OH)₃⁻, UO₂CO₃⁰, UO₂(CO₃)₂²⁻, UO₂(CO₃)₃⁴⁻ and possibly (UO₂)₃(CO₃)₆⁶⁻ [6] and hydrolysis ion such as

[UO₂(OH)₂]₅²⁺ [5, 7, 8]. However, uranium and uranium ion can cause a threat for both the human body and environment because it has chemical toxicity and radioactivity. The presence of a high concentration of UO₂²⁺ ion in drinking water can accumulate in some human body organs such as kidney, liver, lung, and bone, causing serious risks like cancer disease and renal frailer (kidney damage) [9–14]. The EPA has set the level of safe contamination of uranium ion to be lower than 30 µg L⁻¹. Thus, the concentration of uranium ion in water (e.g. UO₂²⁺ ion) must be reduced to the recommended limit as well as selectively capturing within a safe framework structure. This research work can help us to remove the toxic uranium ions from nuclear water effluents as well as to re-concentrates it for another use.

Various techniques are used to remove UO₂²⁺ ion from water, including solvent extraction [15, 16], coagulation-filtration [17], lime softening [18], electrolytic reduction [19], ion exchange [20–22], chemical precipitation [23], reverse osmosis [24], ultra-filtration [25], membrane and electro-dialysis [26, 27], chromatographic extraction [28], flotation [29] and adsorption. Different adsorbents are widely utilized for uranium (VI) ion from water [30]. The importance of utilizing different type of adsorbents is due to the efficiently,

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flexibility, safely, economical visibility, simplicity and lower environmental health impact. There are pieces of examples such as mesoporous silica [31], nanoporous silica [32], synthetic resin [33], covalent organic framework [34], chitosan and cross-linked chitosan [35, 36], polypyrrole [37], composite adsorbents [38], silicon dioxide nanopowder [39], inorganic oxides nanosheets or nanofibers [40, 41], biomasses [42–44], hydroxyapatite [45, 46], activated carbon [47–49], carbon nanotubes [50], graphene oxide and its amine-functionalized composite [51], zero-valent iron [52, 53], polyamidoxime-functionalized colloidal particles [54], amino-functionalized urea–formaldehyde framework mesoporous silica [55], titania nanoparticles covalently functionalized with simple organic ligands [56], porous magnetic N-doped Fe/Fe₃C at carbon matrix and its highly efficient uranium(VI) remediation [57], and iron oxides [58–61], hematite [62] oxine functionalized magnetic Fe₃O₄ particles [63] and activated silica gel [64]. Uranium extraction by sulfonated mesoporous silica derived from blast furnace slag [65] montmorillonite, [66] hydroxide/graphene hybrid material [67] Manganese Oxide coated zeolite [68] amidoxime functionality within a mesoporous imprinted polymer material [69], Tendurek volcanic tuff [70].

These methods do not succeed in terms of selectivity and high cost, as well as are not easy to handle. From another direction, the natural adsorbents and their biomasses are economically effective and easy to handle. Nevertheless, in terms of selectivity, it is not. To achieve higher selectivity in capturing uranium ions from water, the silica gel functionalized organic or inorganic entities could be the best choice. Recently, the removal of uranium(VI) from water has been reviewed [71]. In terms of selectivity, the removal of uranium(VI) ion was investigated using amidoxime silica [72]. Other recent pieces of works on the market deal with utilizing silica gel functionalized organic or inorganic entities for capturing uranium ion from water. For example, silicate nanotubes [64, 73, 74], ethylene-di-amine-tri-acetate [75], amine-modified silica gel [76], murexide [77], silica gel or silica-gel-bound macrocycles [78, 79] and organic or inorganic polymeric ion exchangers [80–83].

Recently, Huang et al. have used tris(2-aminoethyl)amine ligand to modify the surface of silica gel. Wherein, it shows a high affinity for selective sorption Cr(III), Cd(II), and Pb(II) ions from water [84]. Besides, our group has found a highly efficient of such SG-TAEA-NH₂ material for capturing of a single ion phase of the ferric ion [85]. As the continuity of this recent work, we are still studying the efficiency of this adsorbent to capture a single phase of metal and heavy metal ions from water. It is important to indicate that we have not found any study related to the utilization of silica gel particles functionalized tris(2-aminoethyl)amine moiety (SG-TAEA-NH₂) for the sorption of aqueous UO₂²⁺ ion. Herein, we are still studying the efficiency of this

adsorbent to capture a single phase of U(VI) ion from water. The novelty of this study lies in understanding the mechanism of adsorption and the bonding of uranium ion into the surface of SG-TAEA-NH₂. This can provide us a complete knowledge and view regarding the selectivity capturing uranium ion from water, which can help us to understand the sorption mechanism in case of the binary ions phase of the next study. In this contribution, we use SG-TAEA-NH₂ solid particles for capturing and sorption the aqueous UO₂²⁺ ion from water. The distribution of uranium(VI) ion between the aqueous phase and the SG-TAEA-NH₂ solid phase can be investigated. The adsorption data of the various experiments can be analyzed by adsorption kinetics and isotherm models.

Materials and methods

Preparation of silica gel functionalized tris(2-aminoethyl)amine moiety (SG-TAEA-NH₂)

The silica gel functionalized tris(2-aminoethyl)amine moiety (SG-TAEA-NH₂) was prepared according to the reported literature [84]. A summary of the preparation method has been recently published [85]. The surface proposed structure of the tris(2-aminoethyl)amine-functionalized silica gel (SG-TAEA-NH₂) is demonstrated in Scheme 1.

Reagents

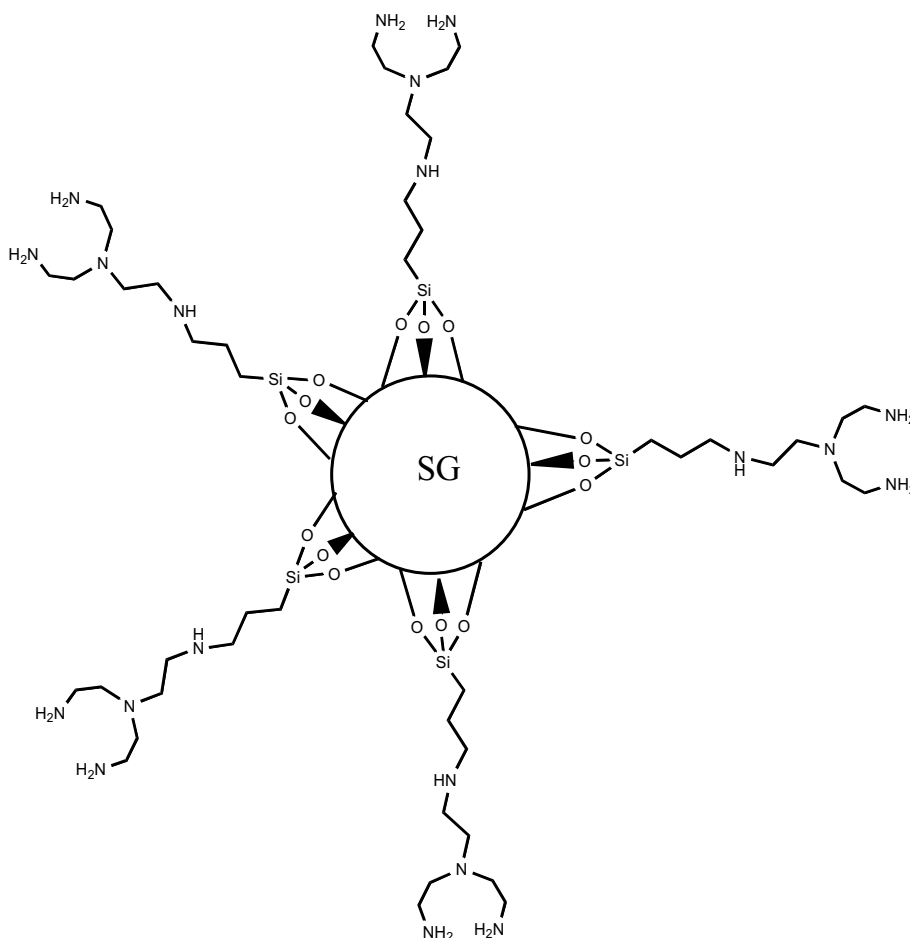
The analytical grade uranyl acetate (UO₂(CH₃COO)₂·2H₂O) was provided from Polysciences GmbH. Other reagents, the preparation of the stock solution (100 mg L⁻¹) of UO₂²⁺ ion, and the methodology of the experiments have been reported [85]. Arsenazo-III (3,6-bis[(2-arsenophenyl)azo]-4,5-dihydroxy-2,7-naphthalenedisulfonic acid) (Fluka Chemie, Buchs, Switzerland). Perchloric acid (70%) purchased from Sigma-Aldrich.

Apparatus and instruments

All absorbance measurements of the electronic spectra were made on a Shimadzu UV/Vis-1800 spectrophotometer (Shimadzu, Tokyo, Japan) with 1-cm quartz cells was used for measurement of the absorbance. The concentration of U was investigated by a spectrophotometric method using arsenazo(III) as a chromogenic reagent. Arsenazo III (Sigma-Aldrich): (0.07% w/v) solution was prepared in 3 M HClO₄.

Uranium standard solution (1000 mg L⁻¹ U (w/v)): This solution can be prepared by dissolving uranyl nitrate hexahydrate (Riedel-de-Haen, Switzerland) in 3 M HClO₄. In a 10 ml standard volumetric flask, 2 ml of the sample containing U(VI) and 3 ml of the 0.07% (w/v) arsenazo III solutions were mixed. The contents of the flask were made

Scheme 1 The proposed structure of the tris(2-aminoethyl) amine-functionalized silica gel (SG-TAEA-NH₂) [85]



up to the mark with 6 M HClO₄ solution to ensure a final concentration of 3 M for HClO₄. After shaking the sample, the absorption can be measured against the reagent blank at 651 nm [86].

Equilibrium studies

The adsorbed amount of the UO₂²⁺ ion onto the SG-TAEA-NH₂ at equilibrium and in a specific *t* is *q_e* and *q_t* (mg g⁻¹), which are calculated by Eqs. 1 and 2, respectively.

$$q_e = \frac{(C_i - C_e)}{S} \quad (1)$$

$$q_t = \frac{(C_i - C_t)}{S} \quad (2)$$

where *C_i*, *C_e*, and *C_t* are the initial, the equilibrium, and the final concentration at a specific time of UO₂²⁺ ion in the aqueous solution (mg L⁻¹), respectively.

The dosage (*S*) of SG-TAEA-NH₂ can be calculated by Eq. 3:

$$S = \frac{m}{v} \quad (3)$$

where *v* and *m* are the initial volume of UO₂²⁺ ion solution and the mass of SG-TAEA-NH₂ adsorbent, respectively.

The percentage removal of uranium ion can be calculated by Eq. 4:

$$\% \text{ Removal of U(VI) ion} = \frac{C_i - C_e}{C_i} \times 100\%. \quad (4)$$

The distribution ratio (*K_d*) can be clarified in Eq. 5:

$$K_d = \frac{\text{Amount of U(VI) ion in SG - TAEA - NH}_2}{\text{Amount of U(VI) ion in solution}} \times \frac{1}{S}. \quad (5)$$

wherein, the relationship between the adsorption percentages and *K_d* (L g⁻¹) is presented in Eq. 6:

$$\% \text{ of Adsorption} = \frac{1000K_d}{K_d + \frac{1}{S}}. \quad (6)$$

Batch sorption experiment

The sorption performance of SG-TAEA-NH₂ material toward the sorption of the uranium (VI) ion was tested by using the batch system at specific $T=35\text{ }^{\circ}\text{C}$ ($\pm 1\text{ }^{\circ}\text{C}$) with changing in C_i (1, 5, 10, and 20 mg L⁻¹) or at specific $C_i=10$ with changing T (25, 35, 45, and 55 °C). The closed sorption system containing 2 g L⁻¹ of SG-TAEA-NH₂ was shaken vigorously (*rpm* of the thermostatic mechanical shaker = 80) of up to 180 min. Afterward, the supernatant solution must be filtered by using filter paper (Whatman No. 41). The filtrate solutions can be analyzed by recording the average of at least triplicate measurements as mention in our reported experiment [85].

Result and discussion

FT-IR spectra

The silica gel functionalized tris(2-aminoethyl)amine moiety (SG-TAEA-NH₂) are mainly composed of silica gel and organic entities, which contain a variety of functional groups. The main functional groups in the FTIR spectrum are Si–O–Si and CH₂, primary and secondary NH₂, which have been recently discussed [85]. After heavy uranium (VI) ion sorption, some changes can be observed in the FTIR spectrum

of SG-TAEA-NH₂ material. Therefore, the FTIR spectra can be also used as evidence of the chemisorption and complex formation in the surface by controlling the shift in the stretching frequency of the primary and secondary NH₂. In particular, Fig. 8 shows the bending vibration of primary amine ($\delta_{\text{N-H}}^{\text{primary}}$) at 1594.4 cm⁻¹, which is shifted by $\Delta\delta=69\text{ cm}^{-1}$ in contrast to the SG-TAEA-NH₂ ($\delta_{\text{Nz-H}}^{\text{primary}}=1525\text{ cm}^{-1}$) before adsorption. The stretching ($\nu_{\text{N-H}}^{\text{primary}}$) is 3321 cm⁻¹, which is cannot be observed due to the weakness of peak transmittance. These changes and shifts in peak positions suggest the presence of the chemical interaction and a possible complexation reaction between the uranium(VI) ion and NH₂ moiety in the SG-TAEA-NH₂ surface [87]. These results are consistent with the mechanisms proposed for the removal of iron ion [85] (Fig. 1).

Effect of pH

Figure 2 shows that the sorption of uranium (VI) ion by the SG-TAEA-NH₂ matrix increase dramatically with increasing pH to the maximum 99% at pH = 7–8, as no further increase above this pH value. This result matches the reported one regarding the adsorption of uranium on the amidoxime resins [88], modified carbon nanotubes [89], and polyacrylyhydroxamic acid sorbent [90].

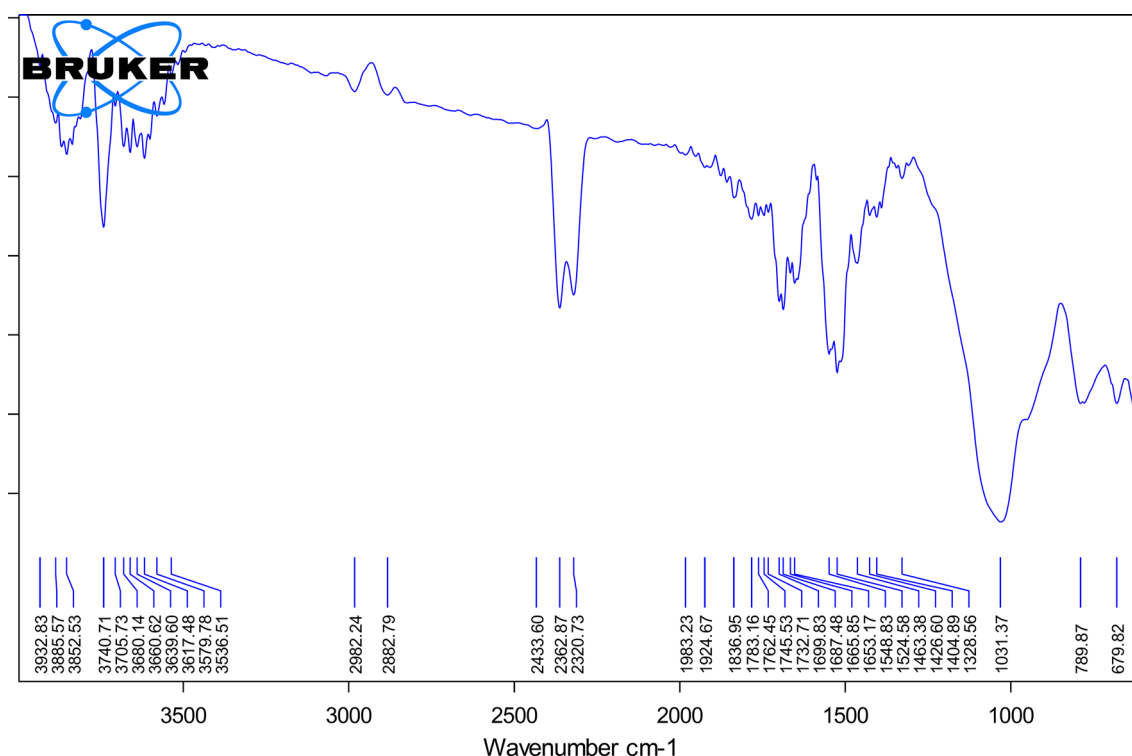
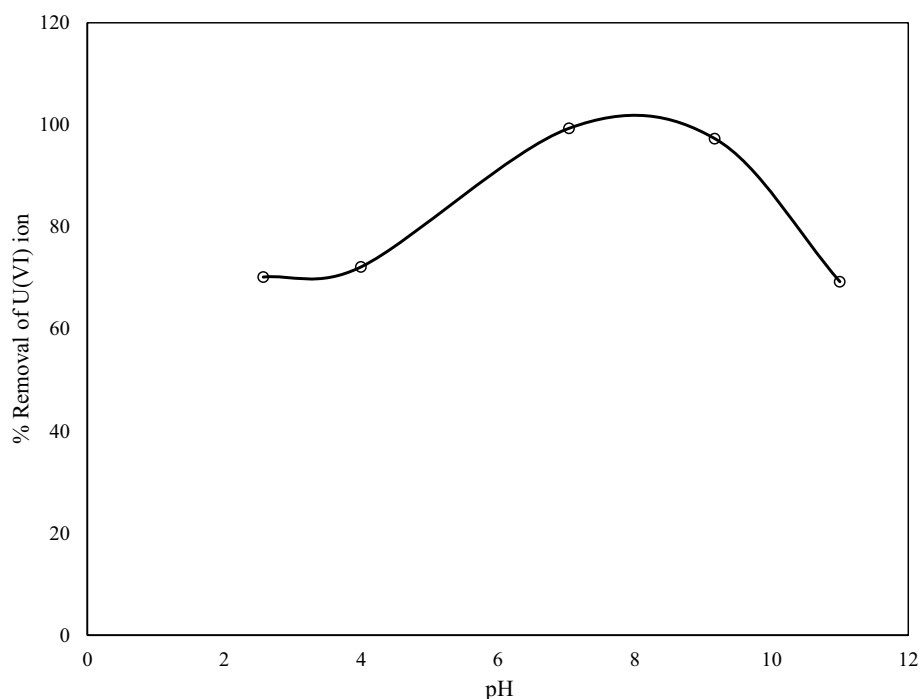


Fig. 1 ATR-IR spectrum for the SG-TAEA-NH₂-U(VI) after 20 min of sorption $C_i=10\text{ mg L}^{-1}$ at 25 °C

Fig. 2 The effect of pH on the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety, $T = 25\text{ }^{\circ}\text{C}$, dosage = 2 g L^{-1} , 80 rpm, and $C_i = 10\text{ mg L}^{-1}$, $t = 5\text{ min}$



The pH has a significant role in the sorption and binding of U(VI) ion with amino-entity in the inorganic silica gel of the SG-TAEA-NH₂ surface. Furthermore, the pH effects in both the stability of the U(VI) ion (complex species) and the SG-TAEA-NH₂ surface morphology (protonation of the amino-entity). Therefore, to understand the effect of pH on the sorption behavior of U(VI) ion onto the SG-TAEA-NH₂ surface, we have studied the sorption using the basic, neutral, and acidic medium.

In an acidic medium ($\text{pH} < 7$), there is a high concentration of H⁺ cation, which competes UO₂²⁺ ion ($\text{pH} < 5$) for binding in the amino-entity active sites. However, the functional amino-entity is protonated to be $-\text{NH}_3^+$. This can block and shield the attraction of di-cationic UO₂²⁺ ion toward the positively charged surface. However, the competition of proton (H⁺) cation for the amino-entity active sites decreases by increasing the pH ($1 > \text{pH} > 7$). Wherein, this can enhance the interaction and sorption of U(VI) ion onto the SG-TAEA-NH₂ surface forming SG-TAEA-NH₂... U complex.

In a basic medium ($\text{pH} > 7$), uranium (VI) ion gradually hydrolyzed to form a mono-cationic complex of UO₂(OH)⁺. The mono-cationic complex has less affinity of interaction with amino-entity on the surface than the di-cationic uranyl ion (UO₂²⁺; $\text{pH} < 5$). Therefore, the sorption affinity of UO₂(OH)⁺ cation with the SG-TAEA-NH₂ matrix at basic is less than acidic and neutral medium. This agrees with the reported work regarding the sorption of uranium (VI) ion from nuclear industrial effluent by using nanoporous silica adsorbent [91]. At $7 \leq \text{pH} \leq 11$, Uranium (VI) can be found

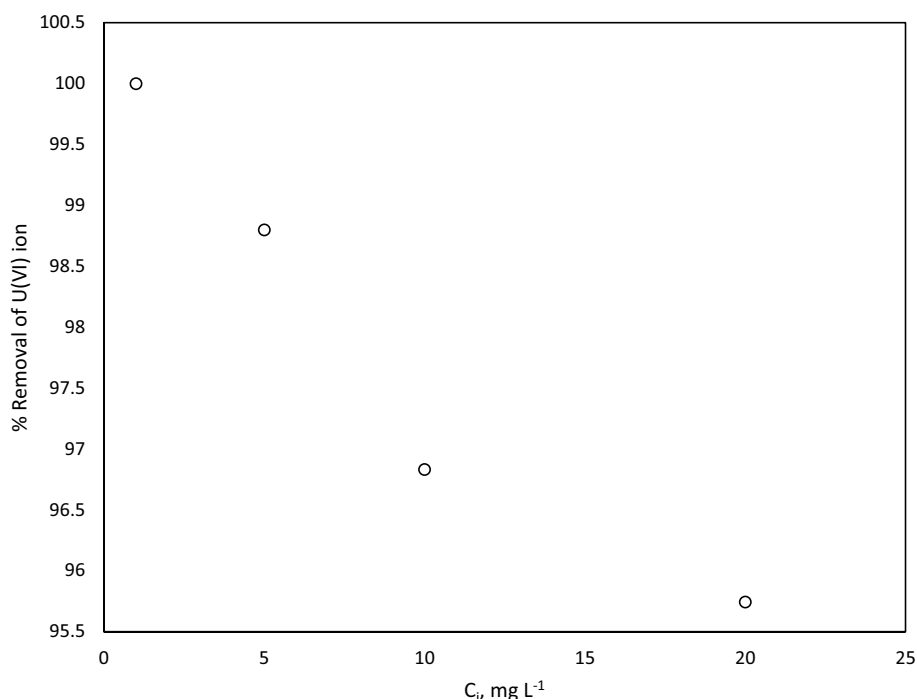
in the form of UO₂(OH)₂·H₂O neutral molecule [92], and wood powder and wheat straw [93].

The acidic medium can affect directly the SG-TAEA-NH₂ matrix forming a shield of positive charges against the sorption of di-cationic UO₂²⁺ ion. From another direction, the basic medium can decrease this positive shield on the SG-TAEA-NH₂ matrix, but forming a mono-cationic UO₂(OH)⁺. The acidic medium can affect directly complex. These two opposite factors guiding us to conclude that the capturing of uranium (VI) is not favorable in both acidic and basic medium to achieve the maximum removal. Therefore, we have chosen $\text{pH} = 7\text{--}8$ as an optimum pH for capturing of uranium (VI) onto the SG-TAEA-NH₂ matrix.

Effect of initial concentration

The batch sorption technique is used for all experiments at $35 (\pm 1\text{ }^{\circ}\text{C})$. Different initial concentrations of UO₂²⁺ ion ($C_i = 1, 5, 10, \text{ and } 20\text{ mg L}^{-1}$) are used of up to 3.0 h. All other sorption parameters are kept constant (e.g. $\text{pH}_i = 7$, $T = 25\text{ }^{\circ}\text{C}$ and 2 g L^{-1} dosage of SG-TAEA-NH₂ materials). Figure 3 shows the effect of initial concentration, wherein the percentage of capturing uranium (VI) ion from solution decreases with increasing the initial concentration of uranium (VI) ion in solution. For example, the maximum percentage of capturing uranium (VI) ion achieve 98% at $C_i = 1\text{ mg L}^{-1}$, while it is 93% at $C_i = 20\text{ mg L}^{-1}$. This is due to the availability the sufficient amino-active sites into the SG-TAEA-NH₂ matrix. For a given SG-TAEA-NH₂ particle dose, the total

Fig. 3 The effect of initial concentration (C_i) on the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety, $T=25$ °C, dosage = 2 g L^{-1} , 80 rpm, and $C_i = 1, 5, 10$ and 20 mg L^{-1} , $\text{pH}=7$, $t=5$ min

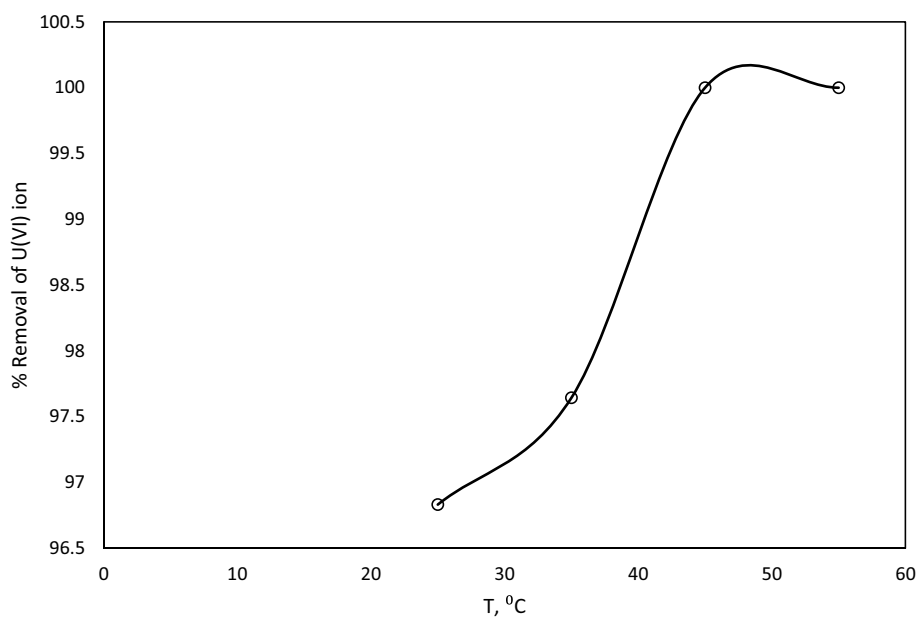


number of amino-active sites available is fixed; wherein, the fixed amount adsorb almost the equal amount of uranium (VI) ion. This results in a decrease in the removal of uranium (VI) ion inconsequent to an increase in the initial uranium (VI) ion concentration. Furthermore, due to the increasing of competing of U(VI) cation onto the active sites of the SG-TAEA-NH₂ matrix. This agrees with what reported previously regarding the utilization of wood powder and wheat straw [93].

Effect of temperature

The effect of temperature is studied over a variant $T=25, 35, 45,$ and 55 °C. Wherein, 50 mL of 10 mg L^{-1} of UO_2^{2+} ion and 2 g L^{-1} of TAEA-SiO₂ solid materials were shaken vigorously and controlled through a period of up to 3 h and $\text{pH}_i=7$. The percentage of capturing uranium (VI) ion increase with the increase of temperature as shown in Fig. 4. The maximum percentage of capturing uranium (VI) ion is 99% at $T=55$ °C, while it is 93% at $T=25$ °C. This is in line

Fig. 4 The effect of Temperatures on the capture of uranium (VI) ion from water by using Silica Gel particles functionalized tris(2-aminoethyl)amine moiety, $T=25, 35, 45,$ and 55 °C, dosage = 2 g L^{-1} , 80 rpm, and $C_i=10 \text{ mg L}^{-1}$, $t=5$ min, $\text{pH}_i=7.0$)



with what has been recently published regarding the sorption of uranium(VI) ion from aqueous solutions by using amidoxime-silica [72]. This indicates the endothermic sorption reaction and capturing process. We notice that no highly significant effect of temperature has appeared. Therefore, from the economic view, we use $T = 25\text{ }^{\circ}\text{C}$ for all experiment batches.

Thermodynamic isotherm

The sorption isotherms is a function of the uranium (VI) ion amount adsorbed into the SG-TAEA-NH₂ surface. To investigate the distribution of uranium(VI) ion between the aqueous phase and the SG-TAEA-NH₂ solid phase, the adsorption data were analyzed by using the Langmuir [94] and the Freundlich [95] adsorption isotherms. The isotherm experiments were conducted by using 1, 5, 10, and 20 mg L⁻¹ of UO₂²⁺ ion solutions. All other sorption parameters are kept constant for each experiment (e.g. $p\text{H}_i = 7$, $T = 25\text{ }^{\circ}\text{C}$ and 2 g L⁻¹ dosage of SG-TAEA-NH₂ material).

The Langmuir isotherm model of the uniform monolayer adsorption onto a surface can be linearized as given by Eq. 7:

$$\frac{C_e}{q_e} = \frac{1}{q_{\max}b} + \frac{1}{q_{\max}}C_e \quad (7)$$

where q_{\max} is the adsorption capacity and the maximum capturing of uranium (VI) ion per unit dosage of SG-TAEA-NH₂ material (mg g⁻¹); and the energy of adsorption b is Langmuir constant (L g⁻¹), which is exponentially proportional to the heat of adsorption and the affinity of binding sites. The linear plot of (C_e/q_e) versus the equilibrium concentration (C_e) gives a straight line. The slope of the plot gives $\frac{1}{q_{\max}}$, while the intercept of the plot gives $\frac{1}{q_{\max}b}$.

The Freundlich model represents the heterogeneous sorption of U(VI) ion on the surface of SG-TAEA-NH₂ material followed by a condensation effect resulting from strong U(VI)-U(VI) ions interaction. The linear form of the Freundlich model is also given by Eq. 8:

$$\ln q_e = \ln K_f + \left(\frac{1}{n}\right) \ln C_e \quad (8)$$

Where q_e represents the amount adsorbed U(VI) ion per amount of SG-TAEA-NH₂ material at the equilibrium (mg g⁻¹), C_e represents the equilibrium concentration (mg L⁻¹), K_f (mg g⁻¹) and n is the sorption capacity of the SG-TAEA-NH₂ material and the intensity of sorption, respectively. The plot of the $\ln q_e$ versus $\ln C_e$ gives the slope ($=\frac{1}{n}$) and intercept ($=\ln K_f$). The higher $\frac{1}{n}$ the value indicates the more favorable of the chemisorption. The Freundlich adsorption correlation coefficients (R^2), K_f and n are presented in Table 1.

Table 1 Freundlich constants of K_f and n

T (°C)	n	K_f	R^2
25	0.49766	45.7641	0.9999
35	1.05274	21.1640	0.9947
45	0.31931	65.1635	0.9963
55	0.46034	55.4954	0.9987

Figures 5 and 6 exhibit Freundlich and Langmuir plots, respectively, for the sorption of U(VI) cation into the SG-TAEA-NH₂ surface. The Langmuir sorption isotherm gives $R^2 < 0.800$, which measure a bad goodness-of-fit for the experimental results. While the results reveal that the Freundlich sorption isotherm is the best model ($R^2 > 0.99$). From the chemical point of view regarding the behavior of the complex reaction system on the surface, it should behave like a monolayer behavior following the Langmuir sorption isotherm model. Nevertheless, what we found is the opposite. The suggestion reason is that the SG-TAEA-NH₂ surface does not have complete coverage by TAEA-NH₂ entities. With this visualization, there is a small part of ($\equiv\text{Si-O-H}$) on the surface of silica gel still active, wherein most of the surface is covered with TAEA-NH₂ entities. Through the sorption process, part of the U(VI) ion is adsorbed by $\equiv\text{Si-O-H}$ entities and other bulk ions captured by TAEA-NH₂ entities. For this, we find that adsorption is heterogeneous and follows Freundlich sorption isotherm. Nearly similar results have been reported [85, 96]. Wherein, the heterogeneous layer of uranium (VI) ion is formed on the surface of powdery aerobic activated sludge including carboxyl ($-\text{COOH}$), Hydroxyl ($-\text{OH}$), Amino ($-\text{NH}_2$) achieving Freundlich isotherm [96]. Besides, our results are not in good qualitatively agreement with those found from adsorption of the iron ion with the SG-TAEA-NH₂ material [85]. The reason may be due to the interaction affinity of iron and uranium with SG-TAEA-NH₂ material and $\equiv\text{Si-O-H}$ entities in the acidic and neutral medium, respectively.

The thermal parameters ΔH and ΔS can be calculated by using Vant Hoff linear Eq. 9:

$$\ln k_c = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT} \quad (9)$$

The linear plot of the Vant-Hoff ($\ln k_c$ vs $\frac{1}{T}$) gives the slope $=\frac{-\Delta H^\circ}{R}$ and the intercept $=\frac{\Delta S^\circ}{R}$ (see Fig. 7 and Table 2).

The positive value of enthalpy change ($\Delta H = +48.71422\text{ kJ mol}^{-1}$) suggests the possibility of a strong binding between the UO₂²⁺ ion and the active site on the surface of the SG-TAEA-NH₂ as an endothermic process. That means, the positive value of ΔH° further confirms the endothermic nature of the sorption. The positive value of entropy change ($\Delta S = +0.1846623\text{ kJ mol}^{-1}\text{ K}^{-1}$)

Fig. 5 Frunedlich isotherm on the sorption of uranium (VI) ion by using Silica Gel particles functionalized tris(2-aminoethyl)amine moiety $T=25, 35, 45,$ and $55\text{ }^{\circ}\text{C}$, dosage $=2\text{ g L}^{-1}$, 80 rpm , and $C_i=1, 5, 10,$ and 20 mg L^{-1} , $t=10\text{ min}$, $\text{pH}=7.0$

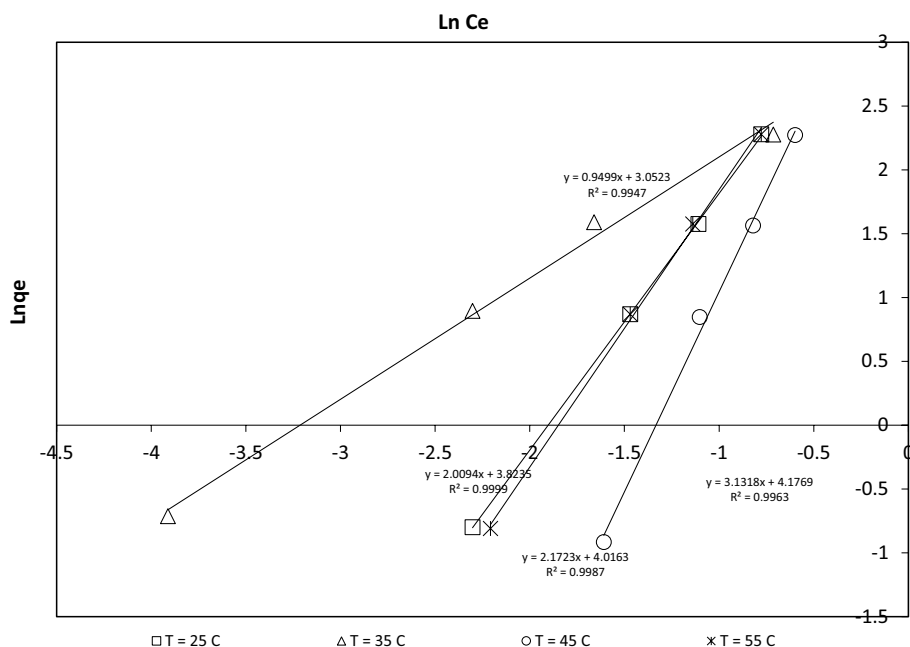
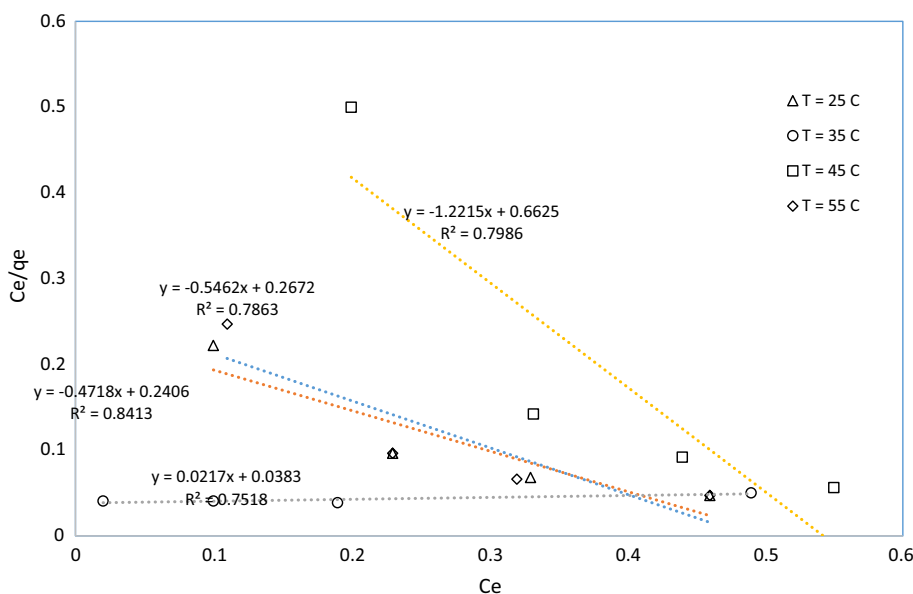


Fig. 6 Langmuir isotherm on the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety, $T=25, 35, 45,$ and $55\text{ }^{\circ}\text{C}$, dosage $=2\text{ g L}^{-1}$, 80 rpm , and $C_i=1, 5, 10,$ and 20 mg L^{-1} , $t=10\text{ min}$, $\text{pH}=7.0$



reflects a good affinity of uranium (VI) ion towards the SG-TAEA-NH₂ surface and increases the randomness at the solid–liquid interface during the sorption. At high temperature, the water molecules surrounded uranium (VI) ion decrease; this leads to increase the water molecules freedom. From other directions, less positive ΔS is due to the decreasing number of free molecules to add in on particle molecule of SG-TAEA-NH₂-U⁶⁺, they were two fragments and be one fragment.

The spontaneity of sorption can be measured by the Gibbs free energy (ΔG). Based on obtaining isotherm results, the best choice to calculate ΔG is Eq. 10:

$$G = \Delta H - T\Delta S \quad (10)$$

The calculated ΔG are found to be a negative in sign ($\Delta G < 0$) based on the changing in temperature values (25, 35, 45, and 55 °C, see Table 2). This indicates the spontaneous interaction of uranium ion into the SG-TAEA-NH₂ surface as shown in Fig. 8. Furthermore, ΔG° values are

Fig. 7 The plot of $\ln K_c$ versus $1/T$ regarding the capturing of uranium (VI) ion by using Silica Gel particles functionalized tris(2-aminoethyl)amine moiety, $t=2$ min, dosage = 2 g L^{-1} , 80 rpm, and $C_i=10 \text{ mg L}^{-1}$, $\text{pH}=7.0$

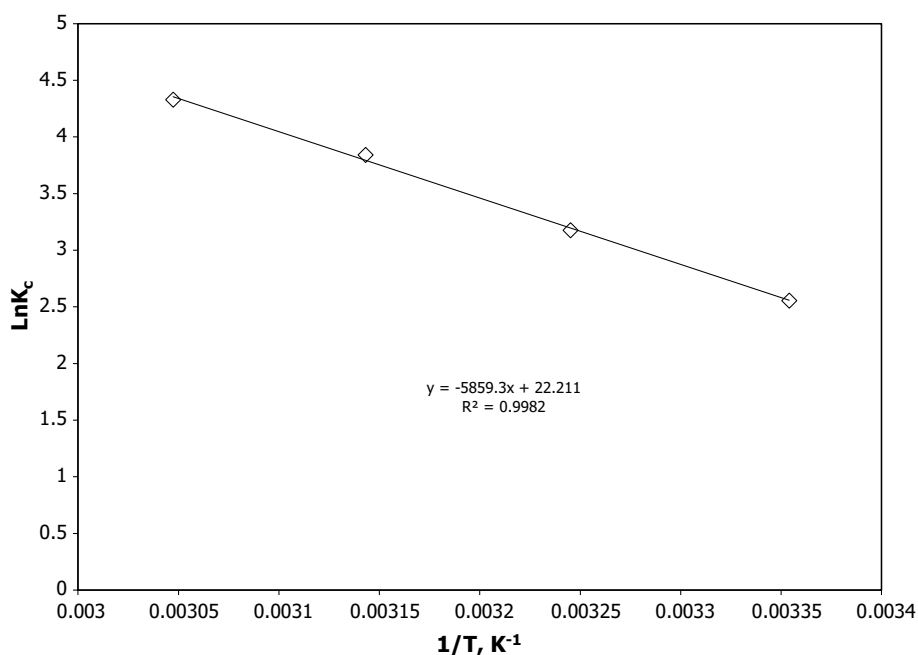


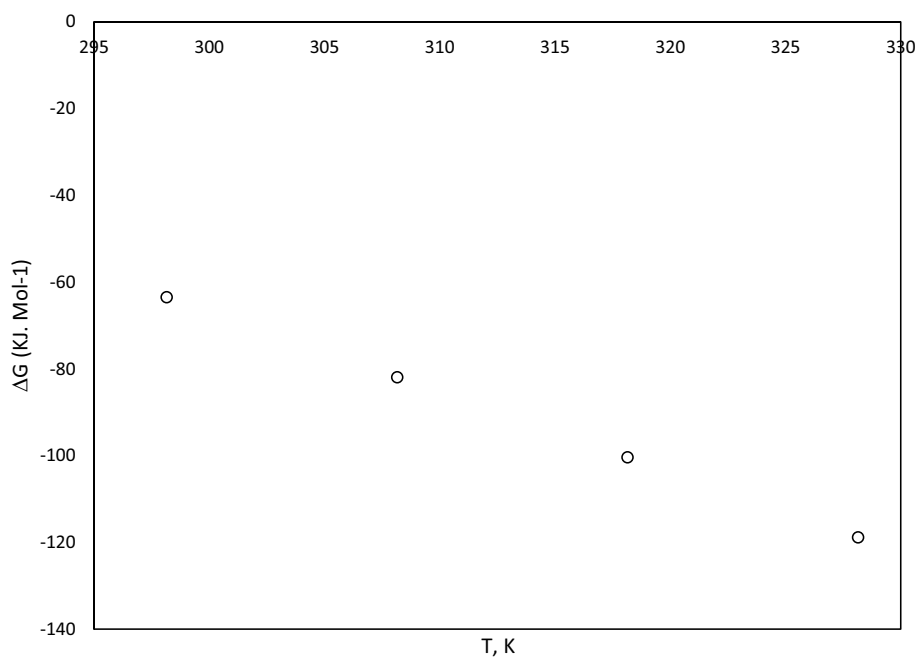
Table 2 Thermodynamic parameters of capturing U(VI) ion by SG-TAEA-NH₂ material

T (C)	ΔG (kJ mol ⁻¹)	ΔH (kJ mol ⁻¹)	ΔS (kJ mol ⁻¹ K ⁻¹)
25	-63.4283	+48.71422	+0.1846623
35	-81.8945		
45	-100.361		
55	-118.827		

found more negative and higher than -40 kJ/mol , which indicates a chemisorption process.

The DKR isotherm is reported to be more general than the Langmuir and Freundlich isotherms. It helps to determine the apparent energy of adsorption. The characteristic porosity of adsorbent toward the adsorbate and does not assume a homogenous surface or constant sorption potential [97]. The Dubinin–Kaganer–Radushkevich (DKR) model has the linear form as in Eq. 11:

Fig. 8 Plot of The value of standard Gibbs free energy change (G) versus temperatures ($T=25, 35, 45,$ and $55 \text{ }^\circ\text{C}$) for capturing of uranium (VI) ion from water by using the SG-TAEA-NH₂



$$\ln q_e = \ln q_{\max} - \beta \varepsilon^2 \quad (11)$$

wherein q_e is DKR monolayer capacity (mg g^{-1}), β is adsorption energy constant, q_{\max} is the amount of U(VI) ion adsorbed per unit weight of SG-TAEA-NH₂ material (mg g^{-1}), and ε is the Polanyi potential. The Polanyi potential can be calculated by using Eq. 12:

$$\varepsilon = RT \ln \left(1 + \frac{1}{C_e} \right) \quad (12)$$

wherein C_e is the equilibrium concentration of U(VI) ion in aqueous solution (mg L^{-1}), R is the gas constant, T is the temperature (K).

The sorption energy E can be calculated by Eq. 13:

$$E = -\frac{1}{\sqrt{-2\beta}} \quad (13)$$

wherein The Dubinin–Kaganer–Radushkevich sorption energy E can confirm the adsorption mechanism as follow [98]:

E (kJ mol^{-1})	Indication
–1 to –8	Physisorption
–8 to –16	Ion-exchange
–20 to –40	Chemisorption

The slope of the plot of $\ln q_e$ versus ε^2 gives β ($\text{mol}^2 \text{J}^{-2}$) and the intercept yields the sorption capacity, q_{\max} as shown in Fig. 9. The values of β and E , as a function of temperature,

Table 3 Dubinin–Kaganer–Radushkevich sorption parameters

T	β	E	R^2
25	–8.43	–24.3541	0.9809
35	–29.047	–13.12	0.9803
45	–3.7531	–36.4998	0.9945
55	–7.4985	–25.8225	0.9913

are listed in Table 3 with their corresponding value of the correlation coefficient, R^2 . The E value obtained shows that the sorption follows the chemisorption mechanism.

Adsorption kinetic

Effect of contact time

To study the kinetic models of such a sorption system, it is worth describing the sorption rate of aqueous U(VI) ion as a function of time-based (1) changing the initial concentration or (2) changing the Temperature.

Figure 10a–c show the effect of contact time onto the sorption process that controlled by changing initial concentration, temperatures, and pH_i . In all cases, we find that the removal rate of uranium (VI) ion is high in the first 5 min. This is due to the availability of the uncovered surface area of the SG-TAEA-NH₂ surface. During the next 5 min, the sorption equilibrium starts yielded a maximum removal of 99% (approx.). This finding is found better than what has been recently published regarding the removal of the U(IV)

Fig. 9 Dubinin–Kaganer–Radushkevich isotherm on the capture of uranium (VI) ion from water by using Silica Gel particles functionalized tris(2-aminoethyl)amine moiety, $T=25, 35, 45,$ and 55°C , dosage = 2 g L^{-1} , 80 rpm, and $C_i = 1, 5, 10,$ and 20 mg L^{-1} , $t = 10 \text{ min}$, $\text{pH} = 7.0$

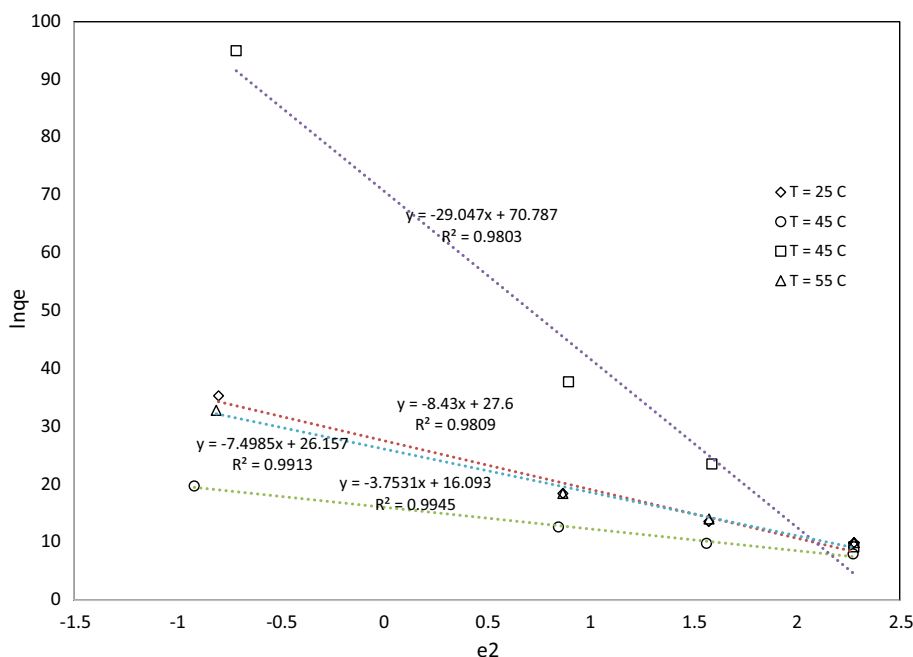
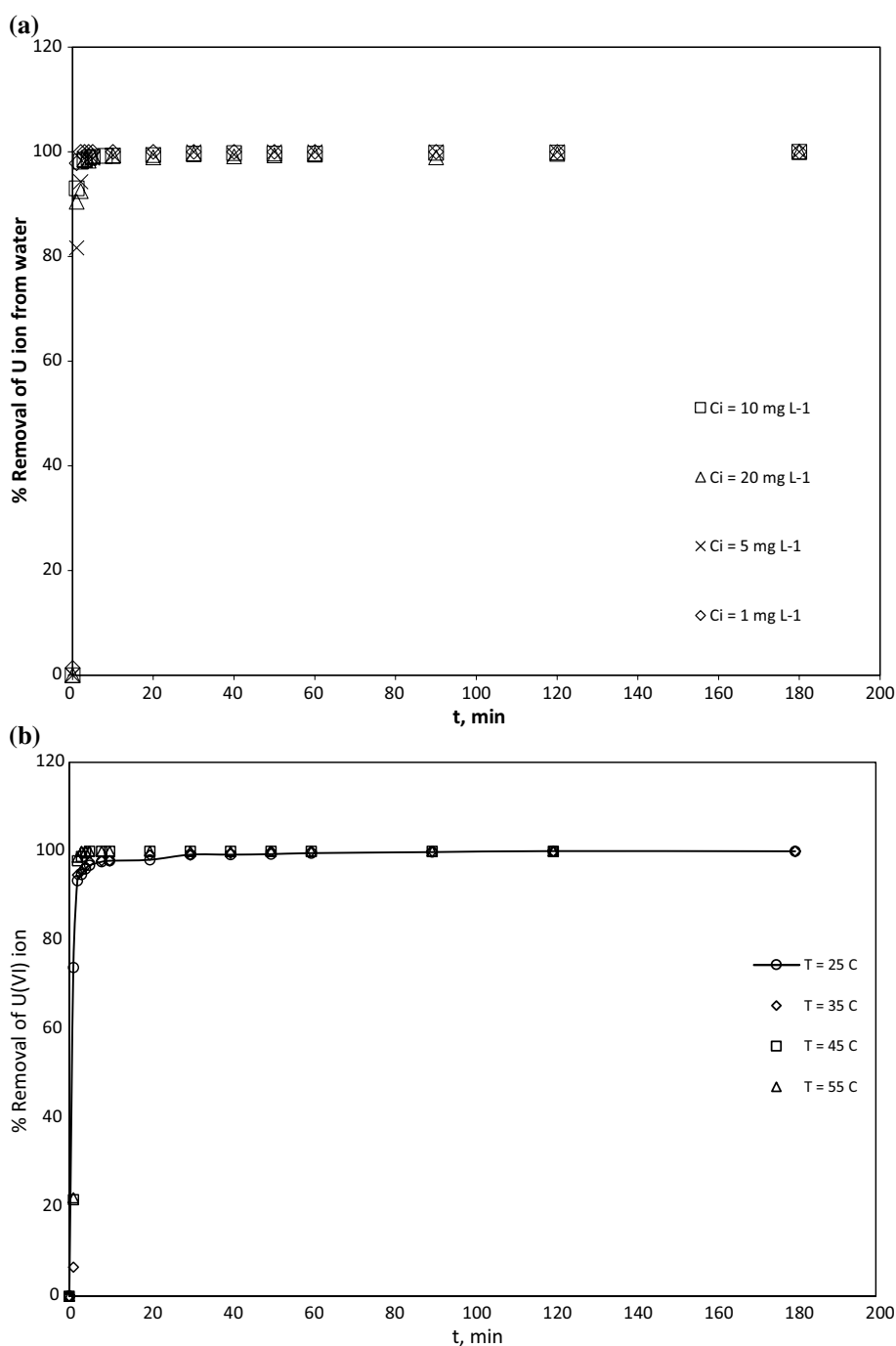


Fig. 10 a The effect of contact time controlled by changing initial concentration on the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety ($T=25\text{ }^{\circ}\text{C}$, dosage = 2 g L^{-1} , 80 rpm, and $C_i=20, 10, 5,$ and 1 mg L^{-1} , $\text{pH}=7$). **b** The effect of contact time controlled by changing temperatures on the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety ($T=25, 35, 45,$ and $55\text{ }^{\circ}\text{C}$, dosage = 2 g L^{-1} , 80 rpm, and $C_i=10\text{ mg L}^{-1}$, $\text{pH}=7$). **c** The effect of contact time controlled by changing pH on the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety ($T=25\text{ }^{\circ}\text{C}$, dosage = 2 g L^{-1} , 80 rpm, and $C_i=10\text{ mg L}^{-1}$)



ion by using an NH_2 -functionalized ordered silica [99] and amidoxime silica [72].

Sorption kinetic model

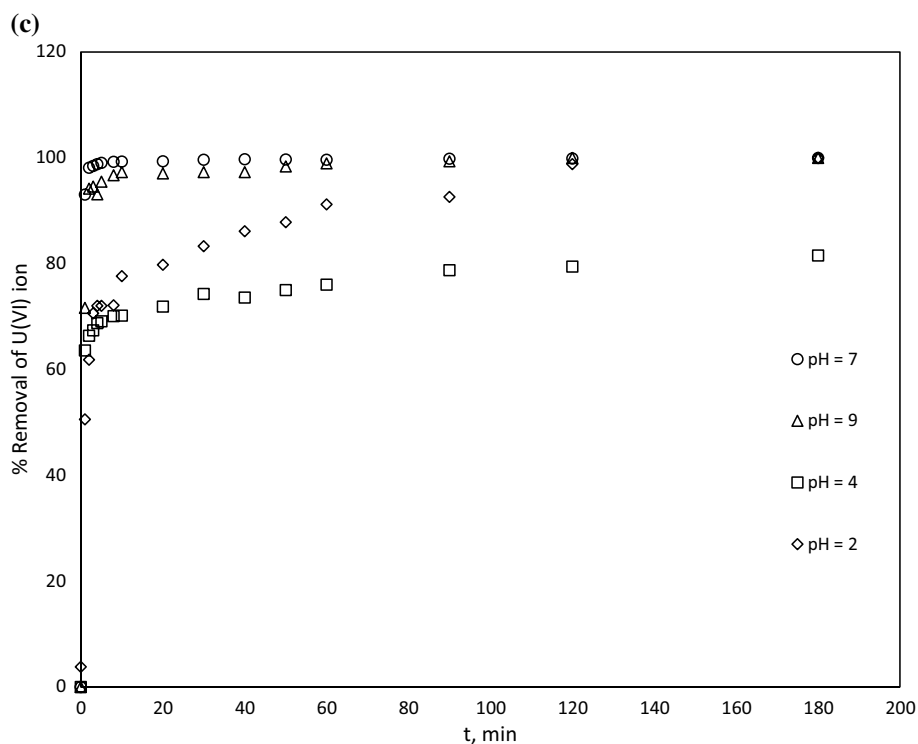
The rate constant and rate-determining step of the reaction can be measured by using the sorption kinetic models. In this respect, two commonly kinetic models of the sorption are used. Wherein, the pseudo-first-order kinetic model and its integral can be expressed by Eq. 14 [100, 101]:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (14)$$

where q_e and q_t (mg g^{-1}) are the amounts of adsorbed U(VI) ion at equilibrium and at the time (t), respectively, k_1 (min^{-1}) is the pseudo-first-order rate constant, and t (min) is contact time. The linear plot of $\ln(q_e - q_t)$ versus t provides the slope of the k_1 and the intercept of $\ln q_e$.

Besides, the pseudo-second-order kinetic model and its integral form are expressed by Eq. 15 [102, 103]:

Fig. 10 (continued)



$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (15)$$

where k_2 is the rate constant of the pseudo-second-order kinetic model ($\text{g mg}^{-1} \text{min}^{-1}$). The value of k_2 and q_e can be determined from the intercept and slope that can be generated by a plot of $\frac{t}{q_t}$ versus t .

Firstly, the kinetics sorption describes the removal rate of aqueous U(VI) ion based on the changing of the initial concentration ($C_i = 1, 5, 10, 20 \text{ mg L}^{-1}$) at constant other parameters ($\text{pH}_i = 7, 80 \text{ rpm}$, dosage = 2 g L^{-1} of SG-TAEA-NH₂ surface and $T = 25 \text{ }^\circ\text{C}$). Figures 11 and 12 show the pseudo-second-order kinetic and pseudo-first-order kinetic models, respectively. The pseudo-first-order kinetic model has the coefficient of determination value (R^2), which is less than

Fig. 11 Pseudo-second order model related to the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety, $T = 25 \text{ }^\circ\text{C}$, dosage = 2 g L^{-1} , 80 rpm, and $C_i = 20, 10, 5 \text{ mg L}^{-1}$, $\text{pH} = 7.0$

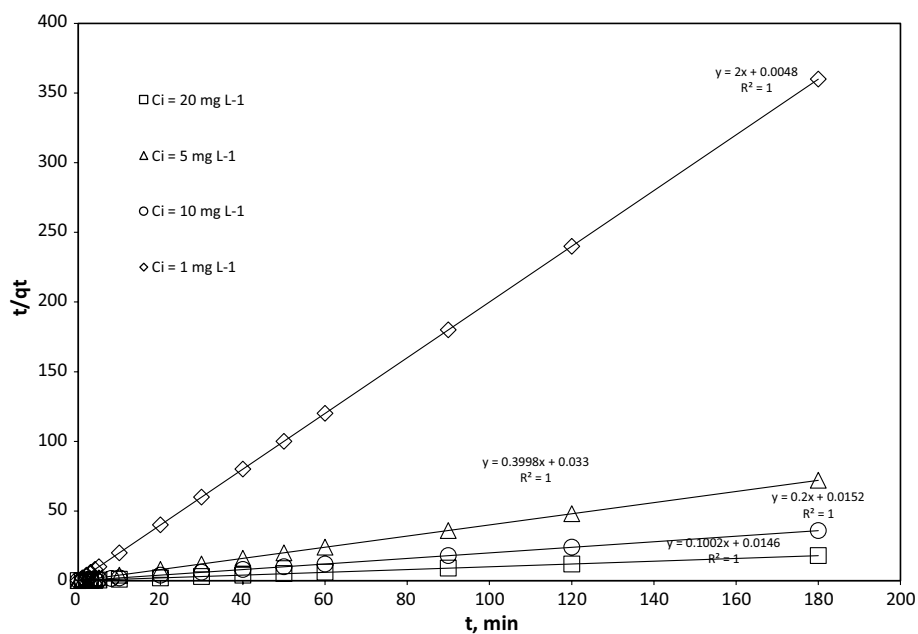
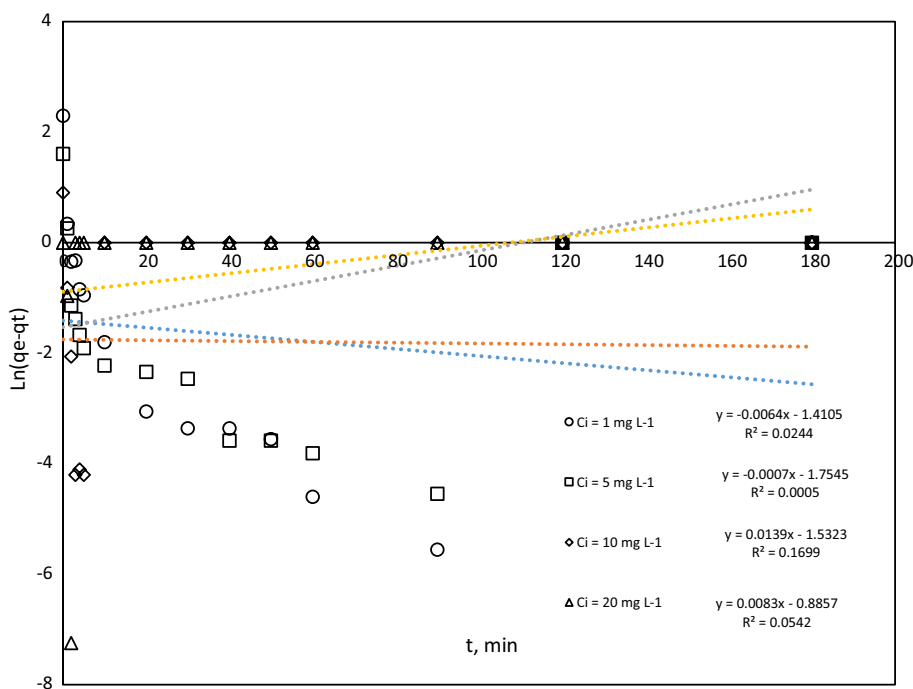


Fig. 12 Pseudo-first order model related to the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety, $T=25\text{ }^{\circ}\text{C}$, dosage = 2 g L^{-1} , 80 rpm, and $C_i=20, 10, 5\text{ mg L}^{-1}$, $\text{pH}=7.0$



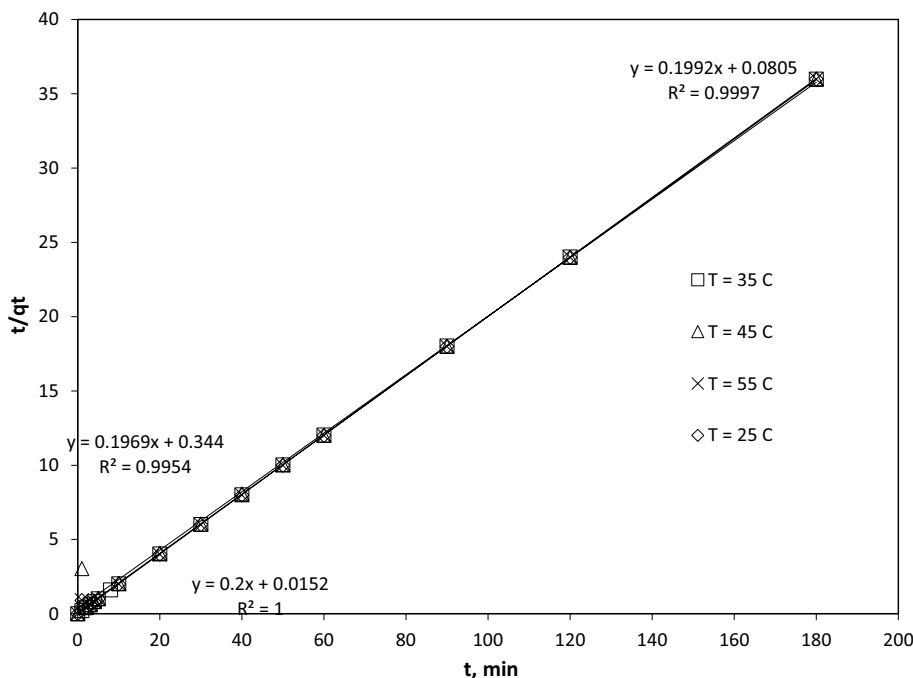
0.020. While, the pseudo-second-order kinetic model has a correlation coefficient $R^2=1.0$, which approve the results.

Secondly, the kinetic sorption describes the removal rate of aqueous U(VI) ion as a function of changing the temperatures ($T=25, 35, 45$ and $55\text{ }^{\circ}\text{C}$) at constant other parameters ($\text{pH}_i=7, 80\text{ rpm}$, dosage = 2 g L^{-1} of SG-TAEA-NH₂ surface and $C_i=10\text{ mg L}^{-1}$). The pseudo-second-order kinetic model has a linear plot with $R^2 > 0.995$ as shown

in Fig. 13. While the pseudo-first-order kinetic model has the coefficient of determination value $R^2 < 0.026$. Therefore, the kinetic adsorption data were satisfactorily fitted to the pseudo-second-order model.

Based on kinetic adsorption experiments as a function of changing the temperatures or initial concentration vs. contact time, the kinetic sorption data indicate that the pseudo-second-order kinetic model can perfectly describe the sorption

Fig. 13 Pseudo-second order model related to the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety, $T=25, 35, 45$ and $55\text{ }^{\circ}\text{C}$, dosage = 2 g L^{-1} , 80 rpm, and $C_i=10\text{ mg L}^{-1}$, $\text{pH}=7.0$



reaction of uranium ion onto SG-TAEA-NH₂ surface. This means that the U(VI) ion can be chemi-adsorbed by the sorption sites onto the SG-TAEA-NH₂ surface as shown in Schemes 2 and 3. We use two experiment functions to be further proof that adsorption exhibits chemisorption behavior. The good matching between the experimental and the calculated (q_{exp} and q_{cal}) support the finding results of kinetic models as listed in Tables 4 and 5. This result was found matching with the reported one by utilizing amidoxime silica [72] and adsorbing iron by SG-TAEA-NH₂ material [85].

Figure 14 shows the plot of the Arrhenius equation ($\ln k_2$ versus $\frac{1}{T}$ as in Eq. 16), which gives the slope ($= -\frac{E_a}{R}$) and intercept ($\ln A$). Wherein, the E_a and A is the activation energy and collision frequency, respectively.

$$\ln k_2 = \ln A - \frac{E_a}{RT} \quad (16)$$

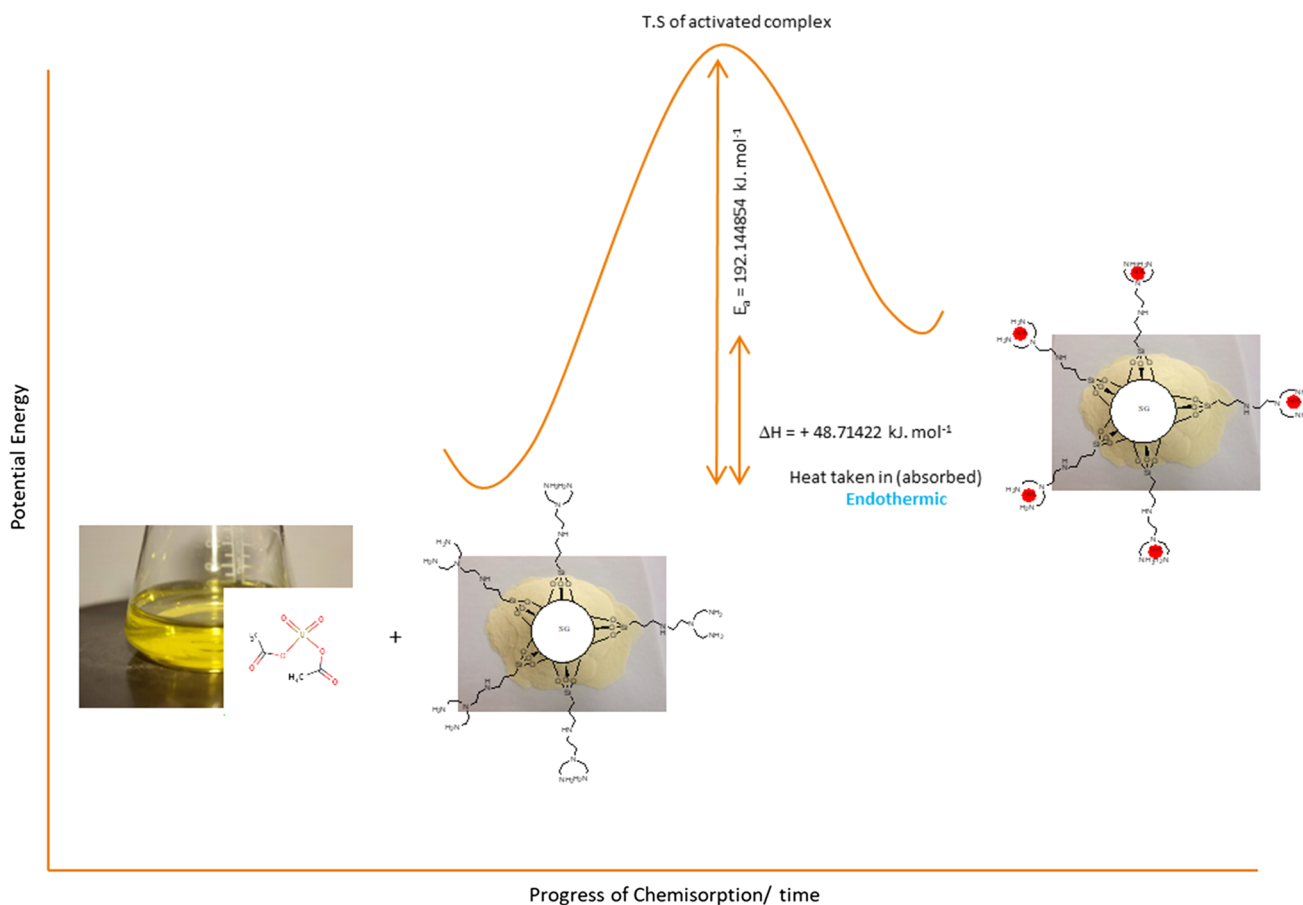
The calculated activation energy ($E_a = -192.144854 \text{ kJ mol}^{-1}$) has a negative sign. In this case, it is possible. There are attractive forces between the amino groups in the surface and uranium ion, there are no

barrier, or the barrier is submerged. This means that barrierless or there are complexes on the potential energy surfaces between the U(VI) ion and the surface of the adsorbent.

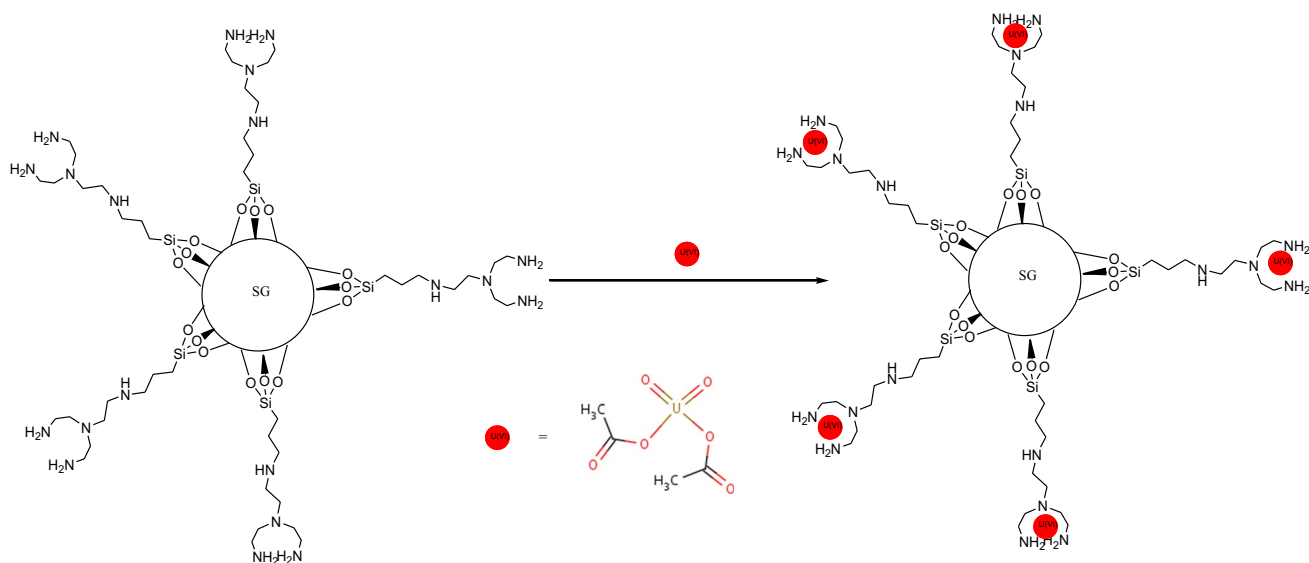
The Weber-Morris intraparticle diffusion model regarding the sorption of the U(VI) cation into the SG-TAEA-NH₂ surface can be discussed through the plot of the q_t vs. \sqrt{t} as mention in Eq. 17 [104]:

$$q_t = k_{\text{int}} \sqrt{t} + C \quad (17)$$

wherein k_{int} is the intraparticle diffusion rate constant ($\text{mg g}^{-1} \text{min}^{\frac{1}{2}}$). The straight line of the plot confirms intraparticle diffusion sorption. A plot presents multi-linearity, which indicates that three steps occur. These are the external surface adsorption (0–2 min), intraparticle diffusion (3 up to 20 min), and final equilibrium stage ($t > 20$ min). Herein, we could say that the intraparticle diffusion could be the rate-controlled ($k_{\text{int}} = 0.0479 \text{ mg g}^{-1} \text{min}^{\frac{1}{2}}$ and $R^2 = 0.8031$, see Fig. 15). This finding matches our recent study related to using SG-TAEA-NH₂ material for the sorption of iron ion from water [85].



Scheme 2 Reaction progress of sorption uranium (VI) ion onto the SG-TAEA-NH₂ surface



Scheme 3 Schematic representation of chemisorptions of U(VI) ion by the sorption sites of the SG-TAEA-NH₂ surface

Table 4 The parameters of the pseudo-Second order kinetic model

R^2	q_e , Calc (mg g ⁻¹)	q_e , Exp (mg g ⁻¹)	k_2 (g mg ⁻¹ min ⁻¹)	$C_1 = \text{mg L}^{-1}$
1.00	0.490	0.500	833.3333	1
1.00	2.485	2.501	4.844608	5
1.00	4.999	5.000	2.631579	10
1.00	9.960	9.980	0.687679	20

Table 5 The parameters of the pseudo-second order kinetic model

R^2	q_e , Calc (mg g ⁻¹)	q_e , Exp (mg g ⁻¹)	k_2 (g mg ⁻¹ min ⁻¹)	T (°C)
1.0000	4.995	5.000	2.631579	25
0.9954	4.980	5.079	0.11269	35
0.9997	4.980	5.020	0.492943	45
		5.018	0.514506	55

Conclusion

Capturing of uranium (VI) ion from water by the silica gel functionalized tris(2-aminoethyl)amine moiety (SG-TAEA-NH₂) is effective in the pH range of 7–9. The capturing percentage increases by increasing the temperature and decreasing the initial concentration. The maximum capturing is 99% based on the sorption parameters

of $T = 25$ °C, dosage = 2 g L⁻¹, 80 rpm, and low initial concentration, $t = 5$ min. The sorption equilibrium can be reached within 5–10 min in maximum. The obtained experimental data have excellent fits within the Freundlich isotherm ($R^2 > 0.999$) proving that the surface of SG-TAEA-NH₂ is un-uniform. The kinetic sorption data fits very well with the pseudo-second-order model indicating chemisorption behavior. Depending on the logic of coordination chemistry, sorption isotherm, FTIR, and kinetic model, the metal complex spheres can be formed spontaneously, favorable on the surface through the chemical interaction between the primary amine active sites (–NH₂) and U(VI) ion as heterogenous-layers. The new finding is the utilization of SG-TAEA-NH₂ as a good potential material for the removal of uranium ion from water.

Fig. 14 The Arrhenius plot related to the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety

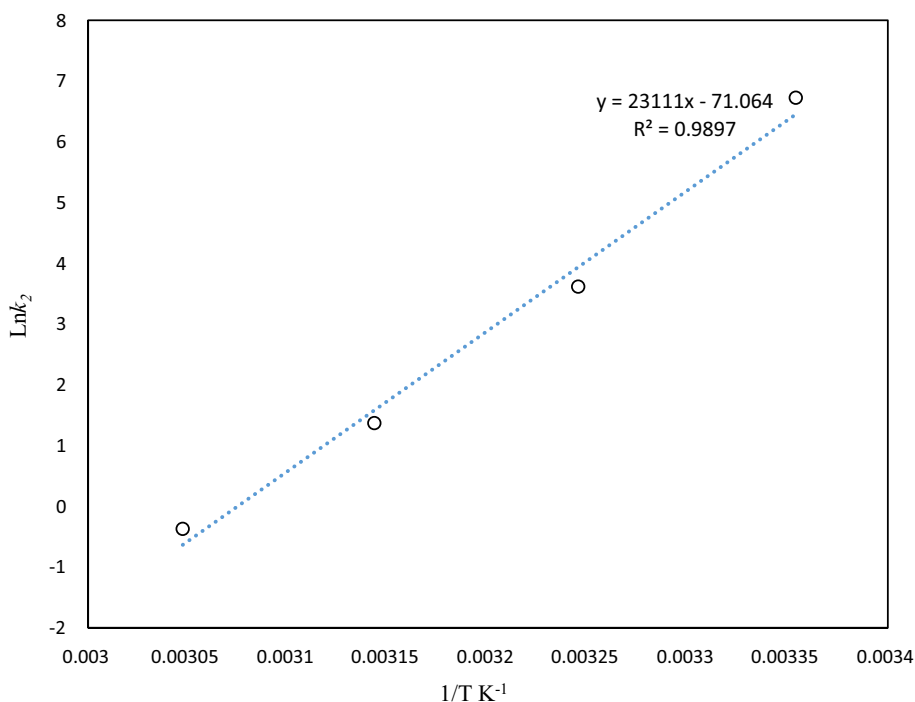
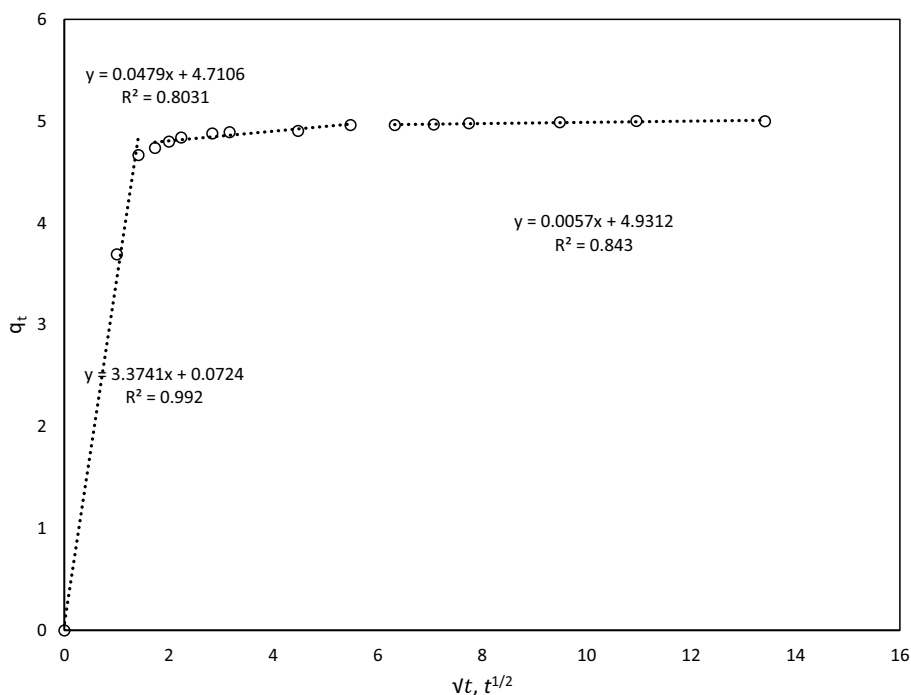


Fig. 15 Weber-Moris intra-particle diffusion model related to the capture of uranium (VI) ion from water by using Silica Gel functionalized tris(2-aminoethyl)amine moiety, $T = 25 \text{ }^\circ\text{C}$, dosage = 2 g L^{-1} , 80 rpm, and $C_i = 10 \text{ mg L}^{-1}$, $\text{pH} = 7.0$



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