



Research Article

Diallyl dimethyl ammonium chloride (DADMAC) and acrylic acid (AAc) embedded nonwoven irradiated polyethylene fabric as efficient adsorbent to separate U(VI) from aqueous solution

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ABSTRACT

Selective isolation of uranium (VI) from wastewater is now a subject of concern due to its damaging effect on living beings. In this study, the pre-irradiation technique was used to prepare grafted polymeric adsorbent by diallyl dimethyl ammonium chloride (DADMAC) and acrylic acid (AAc) onto nonwoven polyethylene fabric (PE) and the grafted adsorbent was applied for uranium (VI) adsorption from aqueous solution by batch method. After irradiation of the non-woven polyethylene fabrics with 50 kGy radiation dose, the grafting reaction was carried out at 80 °C with a monomer solution consisting of 20 g DADMAC and 20 g AAc to 110 mL deionized water. The prepared adsorbent was characterized by Fourier Transform Infrared (FTIR), Scanning Electron Microscopy (SEM), and Thermo-gravimetric Analysis (TGA). After treatment with NaOH solution, the adsorption study was analyzed by pH, initial metal ion concentrations, contact time, and temperature on the adsorption of U(VI). The highest graft yield was achieved at 598%. The maximum adsorption capacity achieved at 160 mg/g was found by treating with 0.1M NaOH for 4 minutes with an initial concentration of 1000 ppm, pH 3.3, and a contact time of 48 hours at room temperature (25 °C). Kinetic adsorption data fitted better with the pseudo-second-order equation and a good correlation of experimental data with the Langmuir isotherm model suggested monolayer adsorption. Langmuir equation showed that the maximum adsorption capacity for U(VI) was 333.333 mg/g. The study depicted good results on the desorption and reuse of the adsorbent.

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INTRODUCTION

Uranium (U) is a naturally occurring primordial radioactive element with atomic number 92. It has three major

isotopes such as ²³⁴U, ²³⁵U, and ²³⁸U with a natural abundance of 0.0050–0.0059%, 0.7198–0.7202% and, 99.2739–99.2752%, respectively. All these isotopes follow radioactive decay by releasing alpha particles along with weak gamma

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radiation and they exist ubiquitously in nature such as in soil, rock, and water with low concentrations [1]. However, due to anthropogenic activities, a significant portion of U is accumulated in the environment. First of all, in the past, several serious nuclear power plant accidents took place such as the Fukushima nuclear disaster (2011), the Chernobyl disaster (1986), the Three Mile Island accident (1979), and the SL-1 accident (1961) [2–5]. Secondly, various radiological incidents occurred in the world such as the Kyshtym disaster, the Windscale fire, the radiation accident in Morocco, the Goiania accident, the radiation accident in Mexico City, and the Mayapuri radiological accident in India [6–8]. Thirdly, some major nuclear submarine accidents happened including the K-19 (1961), K-11 (1965), K-27 (1968), K-140 (1968), K-429 (1970), K-222 (1980), K-431 (1985) accidents [9–14]. Finally, U-containing wastewater is discharged by the processing of mining and ore industries [15]. Due to all these phenomena, air, soil, and particularly water is prone to be a major source of U contamination. In radioactive wastewater, U is present with the principal valence states U(IV) and U(VI) depending on the redox environment [16]. However, U(VI), in the form of uranyl ion (UO_2^{2+}) and with multi complexes, is more soluble and widely present in radioactive wastewater and seawater for its highly migratory performance [17]. Thus, U(VI) can arrive at the top of the food chain where it is consumed by humans. Since all isotopes of U have mutagenic and carcinogenic properties, they are responsible for harmful effects on the human body such as liver damage, skin problem, kidney damage and even death [18–22]. Not only they are detrimental to human beings but also, they can cause significant damage to the aquatic environment. Thus, to protect humans and the biodiversity of the ecosystem, effective recovery of U by selective isolation of U(VI) from aqueous solution and sea water is of immense importance.

In the past, many approaches were applied to uptake the U(VI) from an aqueous solution, such as solvent extraction, chemical precipitation, flotation, ion exchange processes, biological treatment, and adsorption [23–34]. Among these techniques, adsorption is one of the most effective methods for U(VI) removal from wastewater and seawater because of its excellent efficiency, cost-effectiveness, and simple operation, and hence, the technique has gained much attention [35–40]. So far, several ligands such as imidazole, amidoxime, and phosphoryl groups were studied [41–46]. These organic ligands enhance adsorption selectivity and capacity towards uranyl ions which are tightly bonded or chemically grafted into adsorbents. However, the solid carriers are also vital parts of adsorbents besides these ligands because in unfavorable conditions like extreme pH, temperature, and radiation dose they must be survived entirely. Furthermore, these polymers should have suitable surface area and structure so that active groups could be covalently connected on their structures. In recent times, graft polymer becomes a

great blessing in the field of removal of heavy metals from aqueous solution [47–50]. In this case, the adsorbent has a higher value of adsorption capacity, and its ability to reuse reduces the probability of secondary pollution. In this method, functional monomers are covalently bonded onto the parent polymer chain. The reason for being most successful method of this is that this technique allows the blending of various functions of the grafted monomer to the parent polymer without disturbing the mechanical properties of the parent polymer [51–53]. Along with the other methods for initiating graft copolymerization like plasma treatment, decomposition of chemical initiators, ionizing radiation, ultraviolet light, and oxidation of polymers, the radiation-induced grafting technique is advantageous. To some extent, it is more suitable for its extensive penetration into the polymer chain and its rapid and uniform formation of radicals [54]. Available methods of radiation polymerization involve direct irradiation of fiber in monomer solution, vapor phase irradiation method, and pre-irradiation [50].

Many researchers have reported the elimination of toxic and heavy metals by radiation-grafted polyethylene covalently bonded with single or binary monomers (PE) [55–59]. Guo et al. [60] developed a facile route for the fabrication of polyethyleneimine-functionalized reduced graphene oxide/molybdenum disulfide composition aerogels (PEI-rGO/MoS₂ CAs) and applied them to adsorb U(VI) from aqueous solutions successfully. In another investigation, Das et al. [61] showed graft polymerization of acrylonitrile and vinyl-phosphonic acid onto polyethylene fiber is a good candidate to adsorb U(VI) from seawater. In the present study, the preparation of a new adsorbent to remove U(VI) ion based on nonwoven PE fabric was attempted by pre-irradiation grafting. Non-woven polyethylene (PE) fabric is known for its excellent mechanical and thermal properties and low cost. Grafting was carried out by binary monomers of acrylic acid (AAc) and diallyl dimethyl ammonium chloride (DADMAC). The grafted polymer was characterized by employing Fourier Transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy (SEM), and Thermogravimetric Analysis (TGA).

MATERIALS AND METHOD

Materials and Reagents

The mother polymer of non-woven polyethylene (PE) fabric used for preparing grafted adsorbent was collected from Kurashiki Textile Manufacturing Co. Ltd., AAc (99%) and DADMAC (65% in water) were procured from Sigma Aldrich (USA) and Fluka Chemie AG CH-9470 Buchs respectively utilized as monomers to graft onto PE. NaOH, NaCl, and HCl were supplied by Merck, Germany. Uranyl nitrate [$\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$] was supplied by BDH Chemicals Pvt. Ltd., England and Arsenago III was supplied by Fluka, Switzerland.

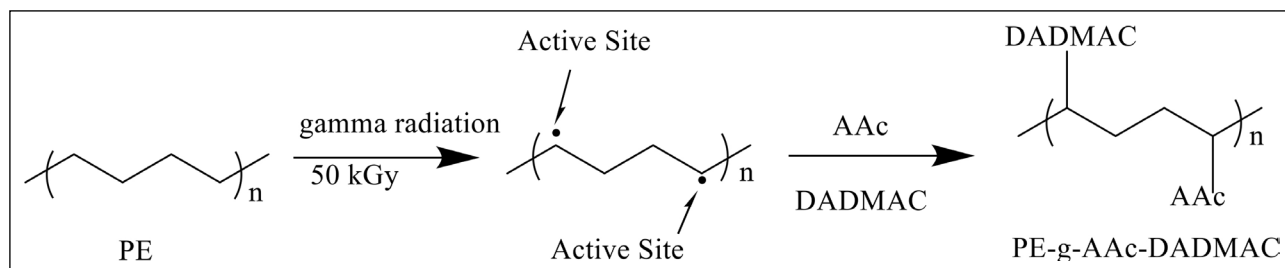


Figure 1. Reaction pathways for grafted PE-AAc-DADMAC formation.

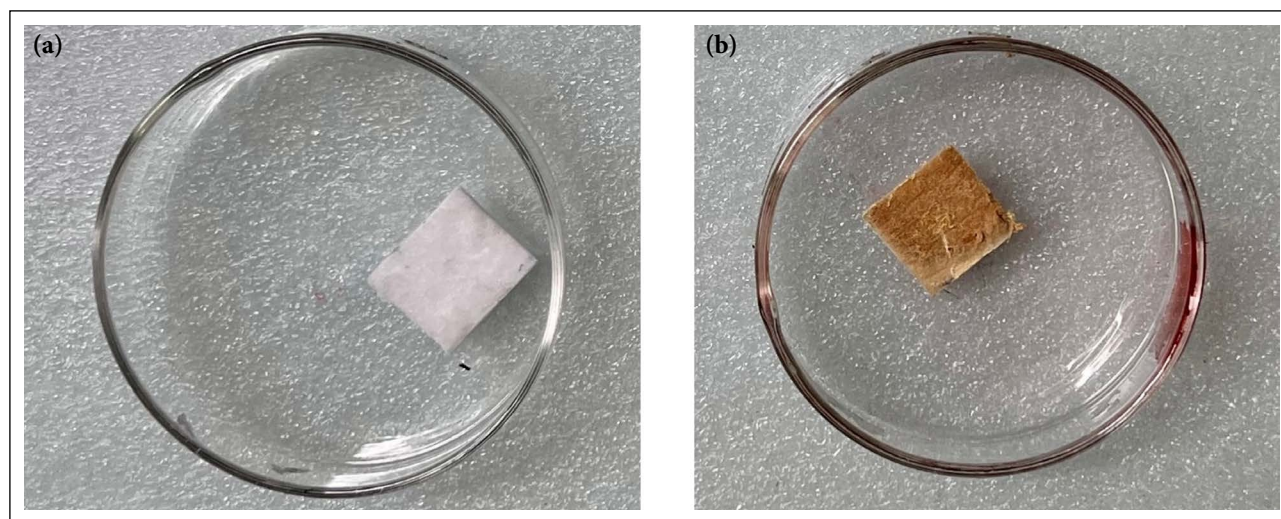


Figure 2. PE- g-AAc-DADMAC polymer before adsorption (a) and PE- g-AAc-DADMAC polymer after adsorption with U(VI) (b).

Instruments and Apparatus

The irradiation of PE was conducted by the Co-60 gamma-irradiator (Panoramic Irradiator of 90 kCi Batch Type collected from BRIT, India) from Institute of Food and Radiation Biology (IFRB), Atomic Energy Research Establishment, Savar, Dhaka, Bangladesh. IR Prestige-21, supplied by Shimadzu Corporation, Japan, was used to investigate FTIR-ATR in the wave number range of 700–4000 cm⁻¹. U(VI) ion concentration from the aqueous solution was analyzed by UV-VIS spectrophotometer (Model: UV2401PC from SHIMADZU, Japan). Scanning electron microscopy (Model JSM-6490LA, JEOL) was used to measure the change in surface morphology after grafting.

Preparation of Grafted Polymeric Adsorbent

In this study, non-woven polyethylene sheets were cut into pieces with 10 cm in length and 2 cm in width and put into airtight packets. Then these non-woven PE films were irradiated with 50 kGy radiation dose at ambient temperature. The irradiator activity was 50 kCi at the time of irradiation and the dose rate used was 13 kGy/h. As soon as the irradiation was completed, the irradiated PE films were preserved quickly under dry-ice condition until use. The monomer solution was prepared by the addition of 30 g DADMAC and 30 g AAc

to 150 mL distilled water. The monomer solution was taken into a beaker and heated at 70 °C on a hot plate for half an hour to make a clear solution. Then the solution was placed in the gas-passing bottle where argon gas was passed for 40 minutes. The monomer solution was bubbled with argon gas to remove dissolved oxygen. The previously preserved irradiated nonwoven PE films were taken into tubes and immediately the deaerated monomer solution was poured into the tubes. When the tube was fulfilled with monomer solution, it was securely closed with a lid to escape the invasion of oxygen from the air into the monomer solution as soon as possible. Afterward, the grafting reaction was carried out in a water bath at a temperature of 80 °C for 4 hours. Then, the grafted PE fabric was washed properly to remove the remaining monomers and homopolymers. Finally, upon washing and drying, it was ready to be used in the experiment. Schematic diagram is presented in the Scheme 1. The degree of grafting was determined by the following formula:

$$DG(\%) = \frac{W_1 - W_0}{W_0} \times 100 \dots \dots \dots (1)$$

Where, DG(%) = Degree of grafting
 W₁(g) = Weight of the PE fabric after grafting
 W₀(g) = Weight of the PE fabric before grafting

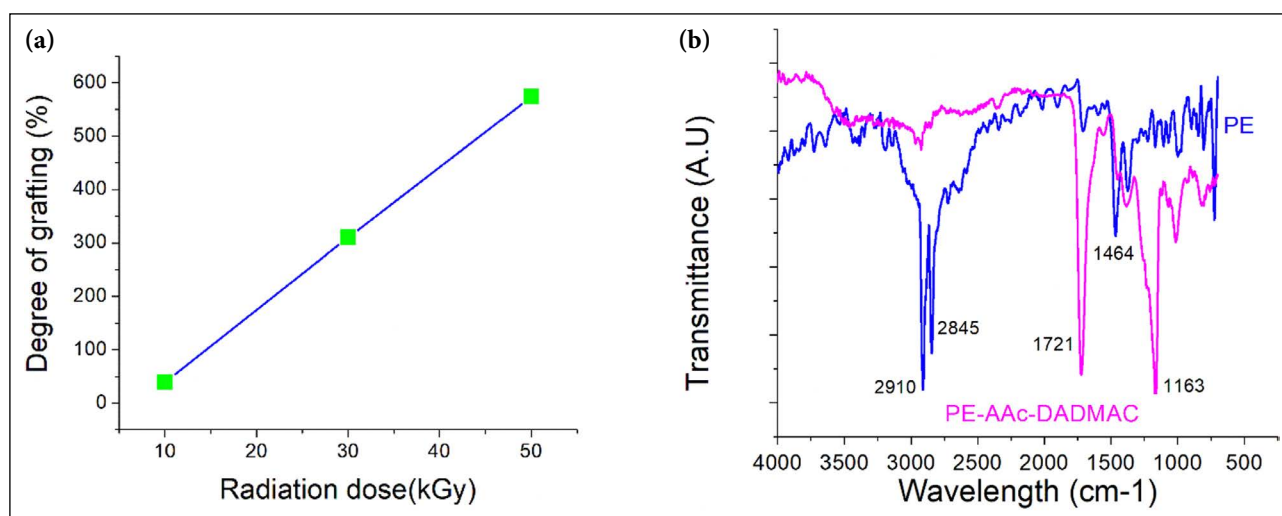


Figure 3. Radiation dose vs degree of grafting (a) and FTIR spectrum of PE and PE-g-AAc-DADMAC films (b).

U(VI) Adsorption by DADMAC and AAc onto Nonwoven Irradiated Polyethylene Fabric

Adsorption criteria were studied by changing contact time, concentration, pH, alkali treatment, and temperature of the U(VI) solution to notice the effect of the adsorption process. A constant weight of adsorbent was kept in a beaker inside the U(VI) solution. After adsorption, the white adsorbent depicted in Figure 1a turned yellow shown in Figure 1b and the U concentrations before and after adsorption were determined by the Arsenazo-III spectrophotometry method [62]. To measure U(VI) concentration, UV-Vis spectrophotometer color complex formation is vital. Thus, 4 mL of U(VI) solution was taken into a 50 mL volumetric flask and added 0.5 mL of 1M HCl, 5 mL of 0.01% Arsenazo and then filled the volumetric flask up to the mark with deionized water. Then sample concentration was determined by UV-Vis spectroscopy at the wavelength of 652 nm. Using the following formula adsorption capacity of the adsorbent was calculated,

$$Q_0 = \frac{C_1 - C_2}{W} \times V \dots \dots \dots (2)$$

Here,

Q_0 = Adsorption capacity of the adsorbent (mg/g)

W = Mass of dry adsorbent (g)

V = Volume of Metal ion solution (L)

C_1 = Initial concentration of metal ion before adsorption (mg/L)

C_2 = Final concentration of metal ion after adsorption (mg/L)

Desorption of U(VI) Ion and Reuse of the Adsorbent

U(VI) was desorbed from the adsorbent by treating it with HCl. After adsorption, the adsorbent was placed into 20 ml of 1M HCl solution in a beaker. The yellow color of the adsorbent turned into white quickly. After 5 hours the solution of U(VI) was separated from adsorbent. The formula used to calculate desorption percentage is given below:

$$\text{Percentage of desorption} = \frac{\text{Desorbed ions (mg)}}{\text{Adsorbed ions (mg)}} \times 100 \dots \dots \dots (3)$$

After desorption of the metal ion, the adsorbent was again capable for reuse to remove U(VI) by the same method.

RESULTS AND DISCUSSION

Degree of Grafting with Radiation Dose

There was a strong relationship between the degree of grafting of the adsorbent with radiation dose represented in Figure 2a. When the radiation dose increased from 10 kGy to 50 kGy, the degree of grafting climbed linearly from 50% to 598% which was the maximum degree of grafting. Hence, for the experimental works, 50 kGy grafted PE was selected to be used.

FTIR Analysis

Figure 2b shows the FTIR spectrum of PE and PE-g-AAc-DADMAC films. The FTIR pattern of PE showed the main characteristic peaks at about 2910 cm^{-1} , and 2845 cm^{-1} were resulted from C-H asymmetric and symmetric stretching vibrations from methylene groups, respectively. The bands at 1464 cm^{-1} and 1373 cm^{-1} were observed due to the symmetric and asymmetric bending vibrations of the C-H bond the methylene group. Besides, Figure 2b also demonstrated the band at 1721 cm^{-1} due to the carbonyl bond of AAc, and another peak at 1163 cm^{-1} was seen due to the C-N fundamental vibration of DADMAC. Hence, the above analysis confirms the addition of DADMAC and AAc on the PE sheet.

SEM Analysis of the Adsorbent

To relate the physical characteristic of nonwoven bare PE with PE-g-AAc-DADMAC scanning electron microscopy of nonwoven PE and grafted PE were analyzed. The SEM image of the bare nonwoven PE fabrics showed the trans-

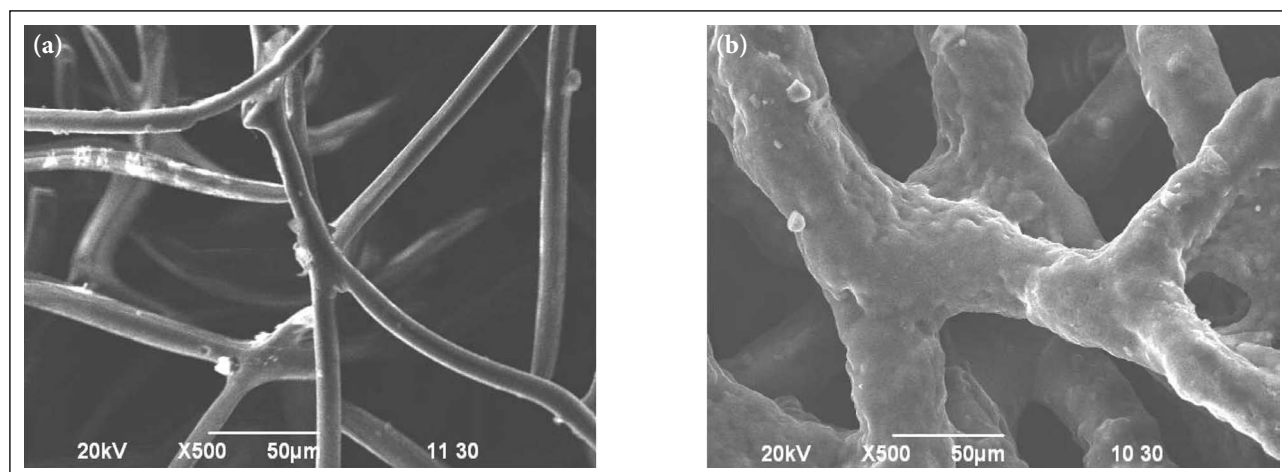


Figure 4. SEM images of PE (a) and grafted PE films (b).

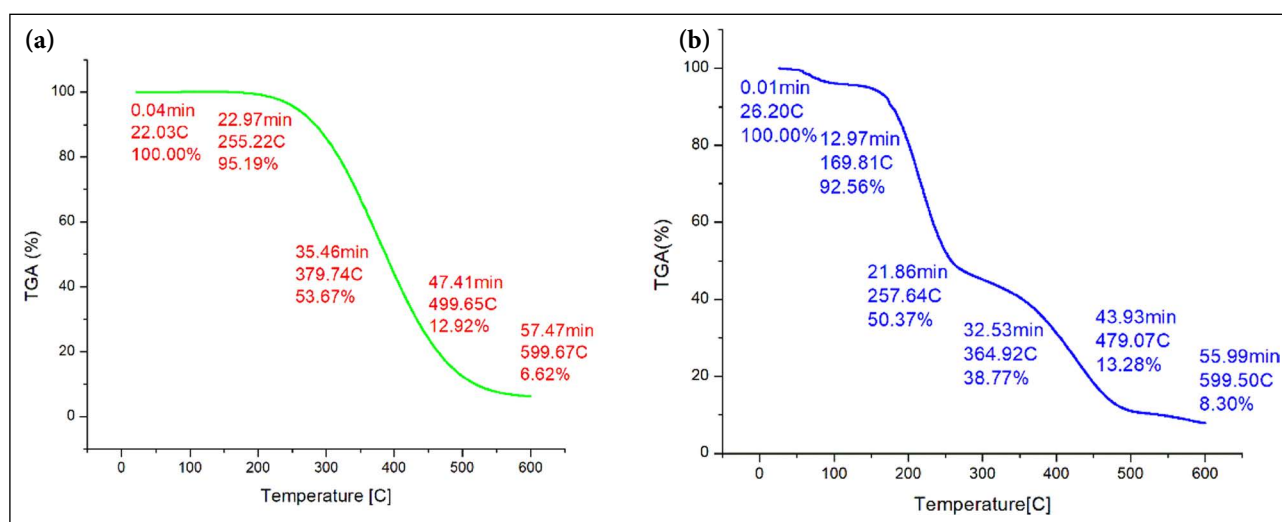


Figure 5. TGA of PE (a) and grafted PE films (b).

formation of the physical appearance after grafting with DADMAC and AAc. A layer of grafted chains covering the surface of PE film could observe after grafting with monomers, which was also seen in another study [63]. The SEM image of non-woven bare PE film is shown in Figure 3a and the SEM image of DADMAC and AAc grafted PE is shown in Figure 3b. The average diameter of fibers increases slightly and distributes more closely which indicates the additional surface due to grafting.

TGA Analysis of the Adsorbent

The thermogravimetric analysis (TGA) thermograms for PE film with DADMAC and AAc grafted PE adsorbent are shown in Figure 4 to understand thermal stability and decomposition and present clear differences in thermal stability between them. The original PE film in Figure 4a showed thermal stability up to 220 °C and above 220 °C it started to be decomposed. It showed two decomposition steps,

i.e., 220 °C–540 °C and above 540 °C. On the other hand, grafted PE film in Figure 4b decomposed more quickly than bare PE film and started weight loss (5%) at 50 °C due to the loss of moisture absorbed from the air. Then it followed four weight loss stages 50 °C–170 °C, 170–257 °C, 257–479 °C, and above 479 °C which were for degradation of grafted chain and degradation of the PE backbone. Above 479 °C the grafted PE film decomposed more than 90%. The thermal stability of DADMAC and AAc grafted PE film was lower than that of the original PE film might be due to the less thermal stability of monomers.

Effect of Treatment of the Adsorbents with NaOH Solution

The adsorbents were treated with NaOH solution to observe the effect of the adsorption capacity of grafted film of the PE-AAc-DADMAC sheet. The prepared adsorbents were sunk into 40 mL of NaOH solution with three different concentra-

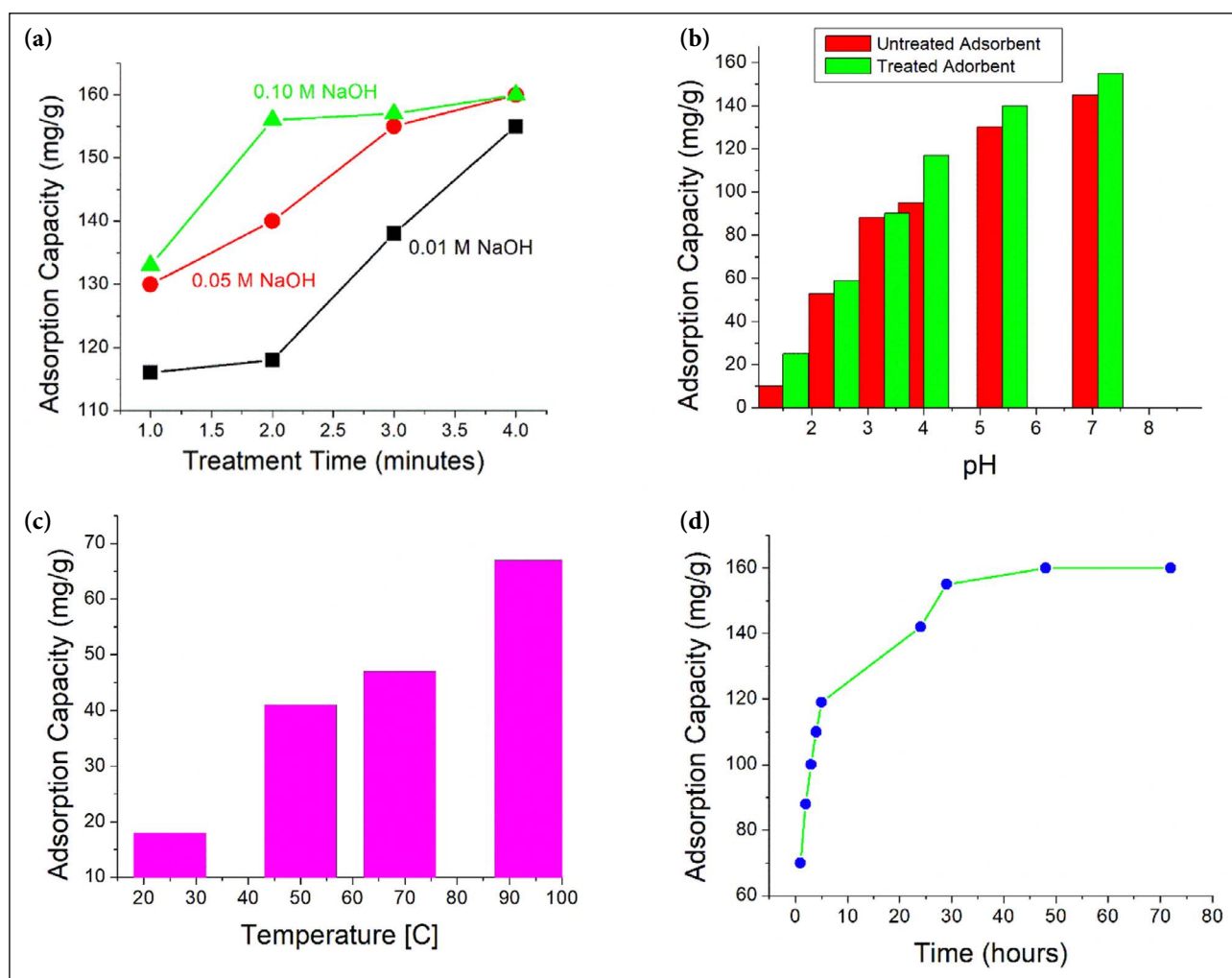


Figure 6. Treatment effect of the adsorbent (a); adsorption capacity vs pH (b); capability of adsorption vs temperature (c); Effect of contact time on U(VI) adsorption capacity with pH 3.2 and initial U(VI) concentration 1000 ppm (d).

tions such as 0.01M NaOH, 0.05M NaOH, and 0.1M NaOH. The adsorbents were dipped in each solution with constant stirring for 1, 2, 3 and 4 minutes respectively and washed immediately after the said times. These phenomena are graphically represented in Figure 5a. The adsorption capacity of the prepared adsorbents increased when treating with higher concentrations of NaOH solutions. However, above the concentration of 0.01M NaOH, the adsorbents swelled much, and degradation was observed. A 1000 ppm 10 mL solution of U(VI) was taken, and adsorption capacity was measured. It was found that the adsorption capacity was 100 mg/g for the untreated sample and 155 mg/g for the treated samples (treated with 0.01 M NaOH for 4 minutes stirring). Therefore, in this study for all experiments, the adsorbents were treated with 0.01M NaOH solution for 4 minutes.

U(VI) Adsorption Effect with pH of the Solution

The adsorption capacity of the prepared composite grafted film of PE-AAc-DADMAC sheet was studied with 800

ppm of U(VI) solution of pH 1.5 to 8.5 shown in Figure 5b. The pH of the U(VI) solution was adjusted by different concentrations of HCL and NaOH solutions. From the graph, it is shown that the effect of pH on U(VI) adsorption was significant for both samples of treated (with 0.01M NaOH solution) and untreated adsorbents. The pH of the mother solution showed acidic (pH=3.3) and in this condition, the adsorption capacity was found to be 80 mg/g and 90 mg/g for untreated and treated adsorbents respectively. However, for both the cases, adsorption capacity increased with increasing pH values from 1.5 to 7.1. Less adsorption capacity was measured below pH 3.3. However, when the pH of the solution increased, the uptake of metal ions enhanced and hence the higher adsorption capacity was observed. At low pH levels, protonated functional groups increased which repulsed the positively charged U(VI) ions resulting the reduction of adsorption of U(VI) cations. On the other hand, higher adsorption capacity was found at higher pH. At higher pH, number of hydroxyl ions increased. As a result,

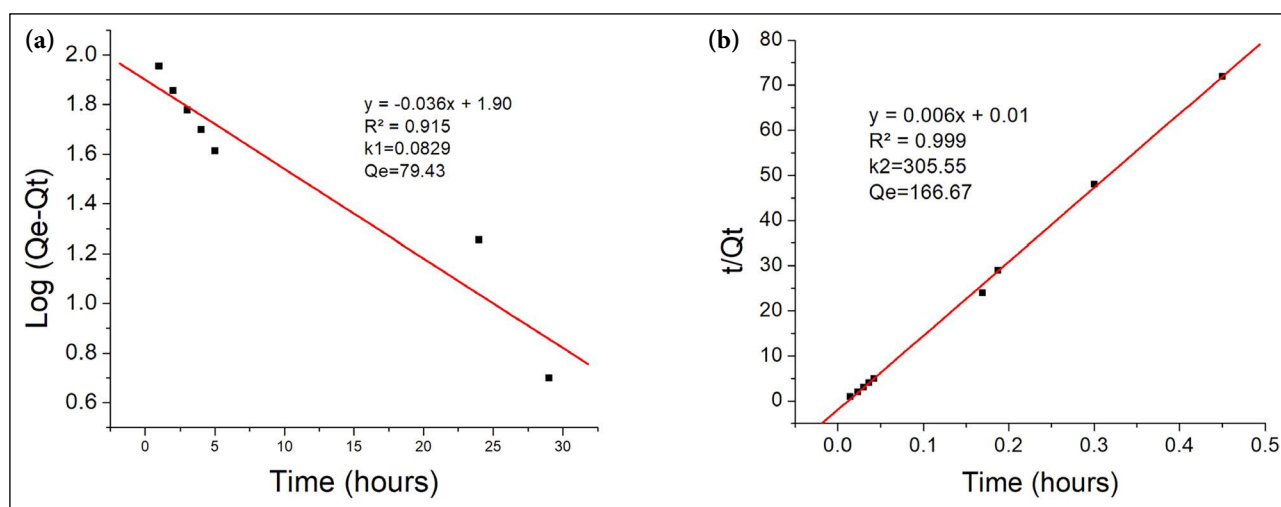


Figure 7. Adsorption kinetic model (a) Pseudo-first order (b) Pseudo-second order.

Table 1. The parameters of adsorption kinetics and adsorption isotherms

Pseudo	Kinetic models					Isotherm models			
	K_1	Q_e	K_2	R^2		Q_0	n	K_f	R^2
Order -1 st	0.083	79.430		0.915	Langmuir	333.333			0.993
Order -2 nd		166.660	305.550	0.999	Freundlich		1.46	2.328	0.983

the proton's competitive action on the metal ion decreased [31, 45]. In a previous study, Sun et al. [17] found that adsorption capacity increased when pH was increasing from 2 to 7. But at higher pH, U(VI) solutions became more turbid because of the precipitation formation of U complex [64]. pH 3.3 was set fixed for all the parameters of this study.

U(VI) Adsorption Effect by Temperature

To understand the effect of temperature, the adsorption capacity was studied with different temperatures such as 25 °C, 50 °C, 69 °C, and 94 °C with 200 ppm U(VI) solution. The solutions were heated for 4 hours except for the 25 °C while maintaining a constant pH. From the graph, it is seen that temperature plays an important role in the adsorption capacity of the adsorbent. This phenomenon is observed in Figure 5c where the adsorption capacity enhanced with increasing temperature. At 25 °C, adsorption capacity was only 18 mg/g, on the other hand, it raised to 67 mg/g at 94 °C. It suggested that the adsorption exhibited an endothermic behavior. It also indicated that the adsorption process is more effective at higher temperature because it needed sufficient energy to decompose U(VI) hydration [31]. The trend was similar with the other studies such as Yuan et al. [46].

U(VI) Adsorption by DADMAC and AAc Grafted PE Film with Standing Time (hr)

There is a relationship that has been established between contact time on U(VI) adsorption from aqueous solu-

tion which is depicted in Figure 5d. The grafted-PE films were treated with 40 mL of 0.1 N NaOH solution and kept into the aqueous solutions of U(VI) and maintained the solution with constant pH (pH 3.3) and initial metal ion concentration of 1000 ppm at room temperature (25 °C). The concentrations of U(VI) ions in aqueous solution were measured at a regular time intervals. From the process, it was seen that, at the initial stage, the U(VI) adsorption rate was rapid and after that the adsorption reached gradually toward equilibrium. After 29 hours, 96.87% adsorption was completed with the adsorption capacity of 155 mg/g, and after 48 hours, the adsorption capacity reached to an equilibrium and the value was 160 mg/g. This trend is similar to another study where 70–90% Cu (II) adsorption completed within 120 minutes and, after that, the adsorption rate became slower till equilibrium [65]. The experimental data are presented graphically in Figure 5d.

To get deeper understanding of U(VI) adsorption, the measured data of adsorption were analyzed using pseudo 1st and pseudo 2nd order kinetic model. The linear mathematical forms of these models are shown by equations (4) – (5), respectively

$$\log(Q_e - Q_t) = -\left(\frac{K_1}{2.303}\right) \times t + \log Q_e \dots \dots \dots (4)$$

$$\left(\frac{t}{Q_t}\right) = \left(\frac{1}{(K_2 \times Q_e)^2}\right) + \frac{t}{Q_e} \dots \dots \dots (5)$$

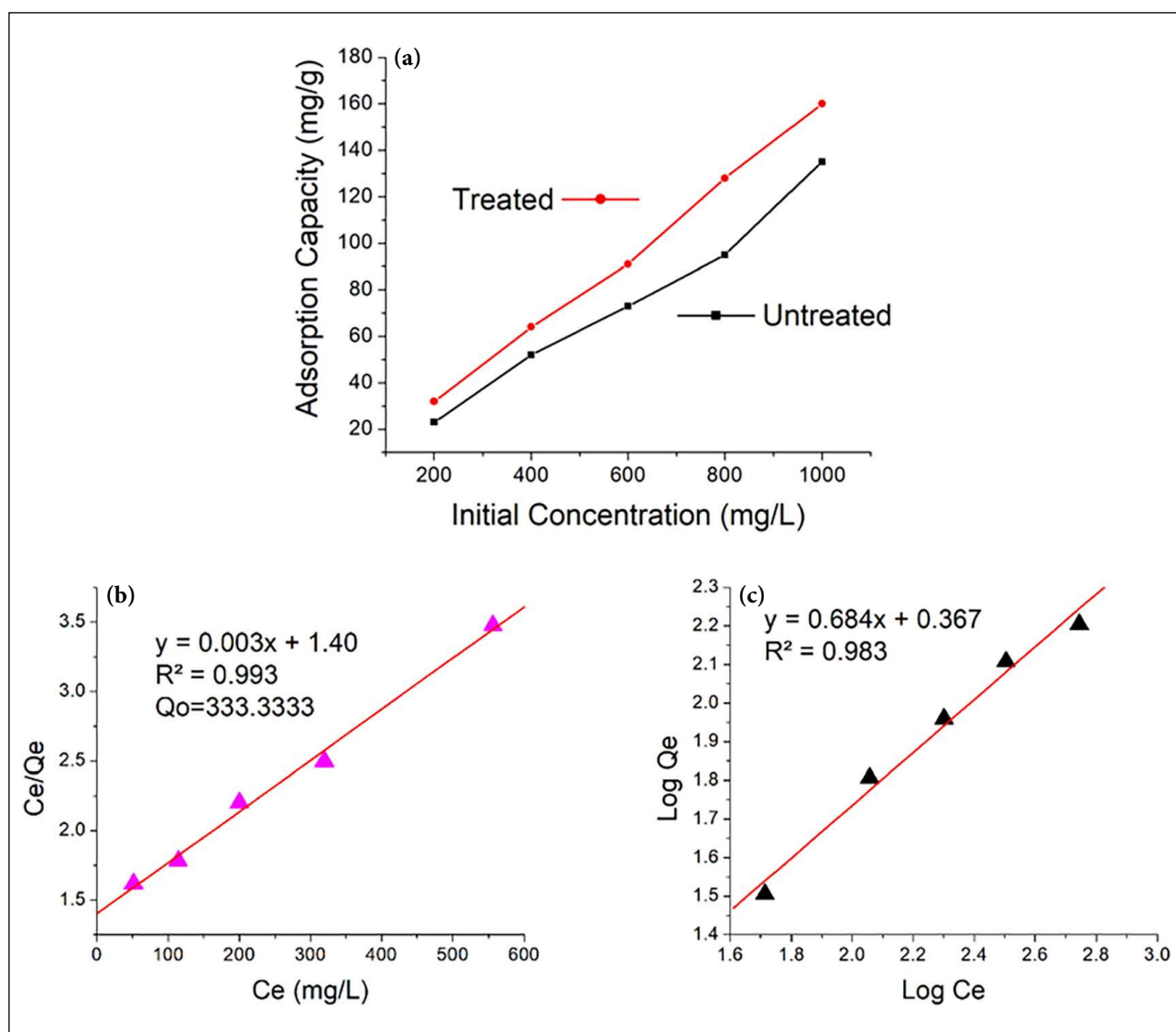


Figure 8. (a) Adsorption capacity variation with initial concentration of metal ions; adsorption isotherm model (b) the Langmuir (c) the Freundlich.

Where Q_e (mg/g) represents adsorption capacity at equilibrium and Q_t (mg/g) indicates adsorption capacity at time t (h), K_1 (/h) and K_2 (g/mg/h) denote the first and second-order kinetic constants, respectively. The rate constant for the pseudo first-order kinetic model could be determined by plotting $\log(Q_e - Q_t)$ versus t as shown in Figure 6a and the values of K_1 and Q_e can be calculated from the slope and intercept of the line graph. The values of K_1 , Q_e , R^2 (correlation coefficient) for U(VI) adsorption are presented in Table 1. Results revealed that the experimental Q_e value (160 mg/g) and Q_e value (79.43 mg/g) measured from the first-order kinetic model were different from each other.

Further, the pseudo-second-order rate constant can be determined from the graph of $\frac{t}{Q_e}$ against t shown in Figure 6b. The values of K^2 , Q_e , and R^2 are shown in Table 1. It is observed that the experimental Q_e value (160 mg/g) and the Q_e

value (166.67 mg/g) measured from the pseudo-2nd-order kinetic model match with each other and the R^2 value (0.99) was relatively higher and very close to unity compared to the pseudo-first-order kinetic model ($R^2 = 0.91$). Hence, the adsorption process of U(VI) uptake followed the pseudo 2nd order equation and might be regarded as chemisorption process with chelation and electron transfer between U(VI) and organic monomers which is similar with Li et al. [36]. In that study, pseudo-2nd-order kinetic model fitted better at low pH. Since the adsorption followed the pseudo 2nd order model, therefore, the intra-particle diffusion process might have happened in the rate-determining step [65]. Moreover, the maximum adsorption capacity for U(VI) removal of this study was compared with previous studies which are shown in Table 2.

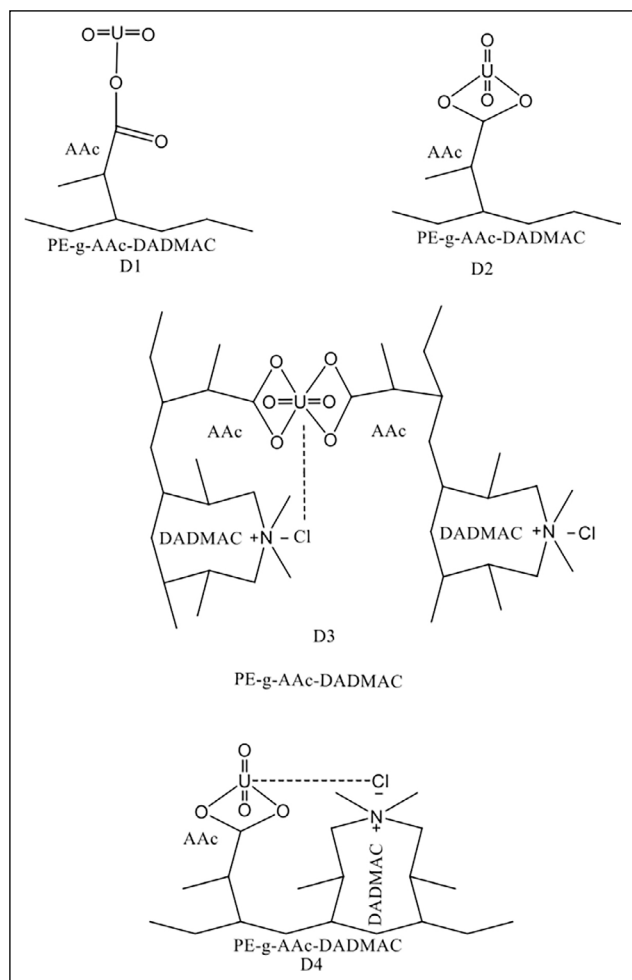


Figure 9. Proposed binding motifs of PE- g-AAc-DADMAC with uranyl summarized in the literatures. Solvent molecules adding sixth, seventh and/or eight coordination sites on the uranyl cation are not shown.

Adsorption Capability with Initial Concentration of Metal Ions

Figure 7a shows the relationship between the initial concentration of metal ions (200 mg/L–1000 mg/L) and the adsorption capacity. This figure depicts that the adsorption of metal ions increased with increasing initial metal ion concentrations up to 1000 mg/L for both the cases of treated with 0.01 N NaOH and untreated adsorbents. When the concentrations of U(VI) ions increased the probability of interaction between adsorbate and adsorbent enhanced. This interlinkage prompted adsorption capacity [35].

Adsorption Isotherm Models

The adsorption isotherm defines surface metal–ligand interaction processes. The Langmuir and the Freundlich adsorption isotherms are usually applied for better understanding of adsorption isotherm. The main difference between the Freundlich and the Langmuir adsorption iso-

Table 2. Adsorption capacity of different adsorbents for U (VI) ion removal

Adsorbents	Adsorption capacity (mg/g)	Ref.
Flax fiber	40.9*	[68]
Silicon dioxide nano-powder	10.15***	[69]
Nano-goethite powder	104.22*	[70]
Commercial ceramic	11.43**	[71]
Flower-goethite (TDFLG)	48.24**	[72]
Flower-goethite (MTDFLG)	112.35**	[72]
PE-g-AAc-DADMAC film [This study]	160*	
PE-g-AAc-DADMAC film [This study]	333.33**	

Ref: References; *: Maximum adsorption capacity from experimental value; **: Maximum adsorption capacity from Langmuir isotherm model; ***: Maximum adsorption capacity from Pseudo-second-order kinetic model.

therms is that Freundlich adsorption isotherm is empirical, whereas Langmuir adsorption isotherm is theoretical. The homogeneous method of Langmuir adsorption is demonstrated by the following linear equation:

$$\frac{C_e}{Q_e} = \frac{C_e}{Q_o} + \frac{1}{(Q_o b)} \dots \dots \dots (7)$$

Where C_e (mg/L) and Q_e (mg/g) are the concentration of the metal ion and capacity of the adsorption at equilibrium, Q_o (mg/g) represents the saturation adsorption capacity and b (L/mg) is the Langmuir adsorption constant. Q_o and b can be calculated by the slope and intercept of the plot of $\frac{C_e}{Q_e}$ vs C_e of the treated samples, which is shown in Figure 7b and summarized in Table 1. The value of maximum adsorption capacity, Q_o was found to be 333.33 mg/g, which is very similar to another study of U(VI) adsorption, where Q_o was 333.13 mg/g [66]. The dimensionless separation factor, R_L , is an important characteristic in Langmuir adsorption isotherm, can be expressed as,

$$R_L = \frac{1}{(1 + Q_o b)} \dots \dots \dots (8)$$

Where Q_o is the maximum adsorption capacity, b (L/mg) is the Langmuir adsorption constant. The value of R_L indicates the type of isotherm to be either unfavorable ($R_L > 1$), favorable ($0 < R_L < 1$), linear ($R_L = 1$) and irreversible ($R_L = 0.00$).

By following Langmuir equation, the values of Q_o [=333.333 (mg/g)], R_L (0.19) and R^2 (0.993) indicated that the adsorption data was well-fitted with the Langmuir adsorption isotherm model.

The Freundlich isotherm illustrates multilayer adsorption on heterogeneous surface and expressed by the linear Equation:

$$\log Q_e = \ln K_f + \frac{1}{n} (\log C_e) \dots \dots \dots (8)$$

Here, the Freundlich constants, which are represented by K_f (mg/g) and n (L/mg) calculated from the intercept and slope of the plot of $\log Q_e$ vs $\log C_e$ of the treated samples, are shown in Figure 7c and summarized in Table 1. n represents good, moderately difficult, and poor adsorption characteristics when the values of n are in the range of 2–10, 1–2, and less than 1 respectively. From the slope, the value of n was found to be 1.46. Thus, by analyzing the values of n and R^2 , it is understandable that the adsorption process is more compatible with the Langmuir adsorption isotherm model than the Freundlich adsorption isotherm model, indicating that PE-g-AAc-DADMAC implemented the specific homogenous sites and provided monolayer surface [46].

Adsorption Mechanism

Based on the results discussed above, mostly, the binding motifs of uranyl moiety with grafted PE-AAc-DADMAC in Scheme 2 were suggested where D1, D2, D3 and D4 binding motifs are presented. Binding motifs shown in D1 and D2 are U-O monodentate and bidentate covalent coordinate bond formation with coordination number three and four respectively [67]. Also, from the scheme, in D3 and D4, the electrostatic interactions between UO_2^+ and the DADMAC moiety and/or Cl⁻ ions stabilized the complex further. From the adsorption isotherm (pseudo-second order) and Langmuir adsorption isotherm model, the maximum adsorption capacity is determined to be 333.33 mg/g. This result indicated that both chelation and electrostatic interactions might be operating to reach a maximum adsorption due to the available binding sites. It should be mentioned that there are other probable binding motifs that may exist which could be responsible for the remaining adsorption. Although it is worthwhile to state that, understanding the interaction between uranyl moiety with grafted PE-AAc-DADMAC is uncertain [36].

Comparison of PE-g-AAc-DADMAC with Other Adsorbents

The U(VI) adsorption capacity (mg/g) of radiation-grafted adsorbent of DADMAC and AAc onto nonwoven PE fabric was compared with those of the other adsorbents reported in other studies, as shown in Table 2. In the present work, the maximum adsorption capacity was found to be 160 mg/g with an initial U(VI) concentration of 1000 ppm at room temperature and undisturbed pH of the solution. Also, according to Langmuir isotherm, the adsorption capacity for the grafted PE was 333.33 mg/g. From the experiment (Table 2), the adsorption capacity of flax fiber showed 40.9 (mg/g) where PE-g-AAc-DADMAC demonstrated 160 mg/g. Further, flower-goethite (MTDFLG) and nano-goethite powder had adsorption capacity 112.35 mg/g and 104.22 mg/g respectively from Langmuir isotherm model. However, adsorbent from this study showed 333.33 mg/g using the same model. Since the adsorption capacity was found to be higher in this experiment, it could be said that the adsorbent which were fabricated had a great affinity to adsorb U(VI) from aqueous solution.

Desorption and Reuse Study

After adsorption, adsorbents were sunk into 20 ml of 1M HCl solution for 5 hours with constant stirring for desorption. It was observed that the desorption was found to be 98%. Furthermore, after desorption, the adsorbents were used for recycling. The adsorbents were treated with 40 mL of 0.01M NaOH for 4 minutes and placed into 1000 ppm U(VI) solution to adsorb as the same process as earlier. In that condition, the maximum adsorption capacity was found to be 150 mg/g, which indicated that the adsorbents could be reused efficiently. Thus, this phenomenon demonstrated that the prepared adsorbent was suitable for recycling and reusing which are very beneficial to lower the cost of overall adsorption process.

CONCLUSIONS

Diallyl dimethyl-ammonium chloride (DADMAC) and acrylic acid (AAc) grafted PE adsorbent was fabricated effectively by the pre-irradiation technique. The prepared adsorbent was characterized by employing FTIR, TGA, and SEM techniques. The radiation dose was optimized to 50 kGy because in that dose, maximum degree of grafting (598%) was achieved. The prepared adsorbent was engaged in adsorbing U(VI) from various aqueous solutions. To complete the adsorption process, it took 48 hours while maintaining pH 3.3 as an optimum condition. The adsorption capacity was found to be 90 mg/g for the treated sample which was higher than the untreated counterpart (80 mg/g). At ambient temperature, the highest adsorption capacity was found to be 160 mg/g in 1000ppm metal solution. Adsorption capacity increased for both the initial metal ion concentrations and the temperature of the aqueous solution. Pseudo-second-order kinetic and Langmuir isotherm models fitted better with U(VI) adsorption. The maximum adsorption capacity was found to be 333.333 mg/g from the Langmuir isotherm model and 160 mg/g from the experiment. Desorption of metal ions and recycling of the adsorbents also showed promising results. To conclude, since the newly prepared grafted polymer was not only low costing but also had high adsorption capacity along with reusability characteristics, the grafted polymer can be considered as an excellent adsorbent for the U(VI) removal from wastewater.

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DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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