Removal and Recovery of Some From Simulated Nuclear Waste Radionuclides Using Amberlite XAD-16-Pyridylazo Resorcinol (AXAD-PR) Composite

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Abstract. In this study, Amberlite XAD-16-Pyridylazo resorcinol composite (AXAD-PR) were developed under laboratory conditions to detect the removals and recoveries of U(IV) and Am(III) from simulated nuclear wastes by adsorption and desorption processes. The effects of contacting time (5, 15, 25 and 200 min), of AXAD-PR concentrations (5, 25, 50 and 100 mg/l), of temperatures (15, 30 and 45°C), increasing U(IV) and Am(III) concentrations (1, 5, 10, 15, 20, 30 and 40) and pH levels (acidic, basic and neutral conditions) on the removals and recoveries of U(IV) and Am(III) radionuclides were investigated. The data obtained showed that the maximum adsorption yields of U(IV) and Am(III) were found to be 96% and 95%, respectively, after 25 min using 25 mg/l AXAD-PR at 30°C at an acidic pH of 5. The adsorption kinetic of radionuclides to AXAD-PR isotherm was in accordance with Langmuir. The AXAD-PR can be reused and can be used again. The regeneration of the composite is possible and can be attained a desorption efficiency of 99% using 0.4 M HNO₃ acid solution from 25 mg/l prepared AXAD-PR.

1. Introduction
Uranium (U-VI) and Americium (Am-III) are the most important strategic elements that have got wide applications which mainly include the nuclear industry and where its significant commercial uses, apart from manufacturing the nuclear fuel for energy production, include the electron microscopy and the glass and pottery glaze industries. However, uranium has a serious long-term potential environmental hazard because of its long half-life as well as its high radiological and biological toxicity [1]. In nature, uranium assays an average of about 4 mg/l in the earth’s crust; however, it forms several economic deposits in various rock types. Uranium is one of the most important radionuclide as nuclear fuel for energy production. Due to insufficient conventional energy sources in Turkey (coal, natural gas, thermal and sun and wind energies) the nuclear energy centrals has been developed. However, the radionuclides such as Uranium (U-IV) and Americium (Am-III) in the nuclear wastes are very toxic pollutants in the environment due to U(IV) and Am(III) have serious long-term potential environmental hazard because of theirs long half-life and high radiological and biological inhibitions. Extensive efforts have thus been recently focused on new solid phase extraction techniques using adsorbents, ion exchangers and other technologies for the separation and recovery of uranium (IV) from its aqueous solutions [2, 3]. In this regard, Amberlite XAD-16 resin series (polystyrene-divinely benzene) such as Amberlite XAD-1, XAD-2, XAD-4 and XAD-16 have a high hydrophobic character and have no ion-exchange capacity, however, these resins are quite important from both the economic and environmental points of view for solid phase extraction after their proper functionalization [4, 5]. Although several methods have been proposed in literature on techniques for removal of radionuclides; adsorption process appears to be the most applicable method for treatment and recovery of used composite adsorbent. The adsorption efficiency of AXAD-PR polymeric adsorbent on 2, 4, 6 trinitrofenol was investigated by Uslu and Demir (2012) [6]. 1.5 gr of AXAD-PR was found to be as the optimum those for maximum 2, 4, 6 trifeno adsorption. Borin and Pavko
(2009) investigated the adsorption of vancomycin antibiotic on AXAD-PR in a packed bed column with a maximum adsorption capacity of 53.76 Kg/m$^3$ at pH 7 [7]. Anaia et al. (2014) studied the adsorption of 1-(2-thiazolyazo)-2-naphtol on Amberlite XAD-7 [8]. Hosseini-Bandegharaei (2016) investigated the adsorption of Th(IV) on XAD-7 [9]. Hosseini-Bandegharaei et al., (2013) investigated separation of U(VI) from polluted environmental wastewaters using Amberlite XAD-2010 with a maximum adsorption capacity of 0.632 mmol/g [10]. Ghaedi et al. (2014) researched the 50 µg/l heavy metal adsorption on XAD-7 modified by chlorophenol and imidazole moieties with a yield of 80% [11]. Prabhakaran and Subramanian (2004) used AXAD-PR modified with hydroxyarsinoyl methyl osphonic acid to extract the U(VI), Th(IV) and La(III), concentrations varying between 18-23 ng/ml with a sorption yield of 80% from acidic wastewaters under 15 min contacting time [12].

Uranium is one of the most important radionuclide as nuclear fuel for energy production. Due to insufficient conventional energy sources in Turkey (coal, natural gas, thermal, sun and wind energies) the nuclear energy centrals has been developed. However, the radionuclides such as Uranium (U-IV) and Americium (Am-III) in the nuclear wastes are very toxic pollutants in the environment due to U(IV) and Am(III) have serious long-term potential environmental hazard because of theirs long half-life and high radiological and biological inhibitions [1].

In this study AXAD-PRs were developed under laboratory conditions to detect the removals and recoveries of U(IV) and Am(III) from simulated nuclear wastes by adsorption and desorption processes. The effects of contacting time (5, 15, 25, 200 min), of AXAD-PR concentrations (5, 25, 50 and 100 mg/l), of U(IV) and Am (III) concentrations (1, 5, 10, 15, 20, 30 and 40 mg/l), of temperatures (15, 30 and 45 °C), and pH levels (acidic, basic and neutral conditions) on the removals and recoveries of U(IV) and Am(III) radionuclides and the sorption isotherms were investigated.

2. Material and methods

2.1. Composition of synthetic nuclear wastewater composition

The synthetic composition of the nuclear wastewater used in this study is given in Table 1.

| Name of radionuclide | Concentration(mg/l) |
|----------------------|---------------------|
| Pu-239 | 19 |
| Sr-90 | 7 |
| Co-60 | 2.3 |
| Y-90 | 1.2 |
| Cs-134 | 12 |
| Cs-137 | 13 |
| Eu-154 | 9 |
| Eu-155 | 10 |
| Pm-147 | 17 |
| Pr-144 | 3.5 |
| C-14 | 25 |
| H-3 | 1.5 |
| S-35 | 2.5 |
| Np | 4.5 |
| U(IV) | 30 |
| Am(III) | 25 |
2.2. Batch sorption studies

Simulated synthetic nuclear power industry wastewater containing different U(IV) and Am(III) concentrations were mixed with increasing concentrations of AXAD-PR using an shaker at a shaking rate of 200 rpm. The adsorption studies were performed at batch wise using 400 ml glass quartz reactors. 100 ml of synthetic nuclear wastewater containing increasing U(IV) and Am (III) concentrations and increasing AXAD-PR were added to the glass quartz reactors. They were mixed using magnetic stirrers at a rpm of 200. The reactors were operated at different retention times, different pH levels and at different temperatures. The temperature was adjusted using water baths. The pH of the samples were adjusted by using weak acid and alkaline. The solid material was separated from the liquid after 15 min sedimentation. The radionuclide analysis was performed in the supernatant samples. The sorption efficiency, \( R (%) \), can be calculated by using the Eqs. 1 and 2.

\[
q_e = \frac{(C_i - C_f)V}{m}
\]

Sorption capacity, \( q_e \) (mg/g) was obtained using Equation 2

\[
A\% = \frac{(C_i - C_f) 100}{C_i}
\]

Where \( C_i \) and \( C_e \) are the initial and equilibrium concentrations of the U(IV) and Am(III) (mg /l), respectively. \( V \) is the volume of the testing solution (L), and \( m \) is the amount of the AXAD-PR (g) and A is the sorption efficiency (%).

2.3. Reusability of AXAD-PR

The regeneration–reusability property of AXAD-PR is important for an appropriate adsorbent at an optimum pH range, which implied that the loaded U(IV) and Am(III) radionuclides might be desorbed from spent sorbent. Therefore, the reusability experiments were performed using 1.0 mol/l HCl as desorbing agent.

2.4. Teflon coated quartz glass reactors for sorption processes

Sorption experiments were carried out in self-designed quartz glass reactors. The dimensions of the reactors were 38 and 3.5 cm. The experiments were performed at room temperature and the pH of the reaction mixture was adjusted from 2 to 8 using 1 mol/l of \( \text{H}_2\text{SO}_4 \) and NaOH solution.

2.5. Operational conditions in batch reactors

The effects of contacting time (5, 15, 25, 200 min), of AXAD-PR concentrations (5, 25, 50 and 100 mg/l), of U(IV) and Am(III) concentrations ( 1, 5, 10, 15, 20, 30 and 40 mg/l), of temperatures (15, 30 and 45 °C), and pH levels (acidic, basic and neutral conditions) on the removals and recoveries of U(IV) and Am(III) radionuclides and the sorption isotherms were investigated.

2.6. Analytical procedures

2.6.1. Measurements of U(IV) and Am (III)

The measurements of U(IV) and Am(III) were performed in Lawrence Berkeley Laboratory in Canada. Radionuclide analyze were performed using an ion chromatography device comprising a scintillation detector with a cell flow. This device is an CS5 separation column providing the separation of cation and anion in the waste water and the detector with a cell flow is scintillated from the glass bed activated with cerium. The studies were performed by taking 0.5 ml sample. The Chromatographic unit consisting from the ion exchanging column, from the gradient liquid pump and from an scintillation detector providing the wastes nonhazardous and from an computer. The chromatographic unit is an pre-concentration column and is consists from and cation exchange resin.
(ionex CC2) containing sulfonic acid and an analytical column (from an ionex CS5 containing both anion exchanging resin and both the sulfonic acid groups and alkyl quaternary’s amine groups). The radiation detector, consist from a chilled tube (solid scintillation apparatus) containing a glass bead activated with cerium. This detector is placed between two pairs of tubes increasing the rays. The system is run with the stages given in Table 2 and the sample volumes are taken from 0.5 ml up to 5 ml.

### Table 2. Examples of the radionuclide analysis stages

| Time (min) | Stage Description |
|------------|-------------------|
| 0.5        | The sample is rinsed with 4 ml of water |
| 5.17       | 0.09 M oxalic acid |
| 17.18      | Washing with water |
| 18.29      | 0.1M diglycolic acid |
| 29.30      | Washing with water |
| 30.50      | 1M nitric acid |
| 50.55      | Washing with water |
| 1.0       | Flow rate 1 ml/min |

After separation process, the actinides are sequentially going to the scintillation detector with a flow cell. The flow cell detector is calibrated with Am(III) and U(IV) at very low concentrations in National Institute of Standards and Technology. In order to determine the retention time of radionuclides the media was acidized by using 0.1 M HNO₃.

**Performed studies:** Fluent → sampling → passing from the pre-concentration column → passing from isolated column → passing from the separation column → passing from the detector cell flow.

**Calculation of Area:** It is shown in the Chromatograph given below (Figure 1). From here it is passing from the radionuclide concentration

![Chromatograph](image)

**Figure 1.** Studies for radionuclide measurements and standby time of radionuclide

### 2.6.2. Sorption Isotherm

#### Langmuir isotherm

The Langmuir isotherm is one of the most important monolayer sorption models, and it is based on the assumption that the sorption occurs on a perfect surface consisting of equivalent and independent active sites. According to this model, the sorption cannot proceed beyond monolayer coverage. The linear form of Langmuir model can be represented as follows:
Where \( b \) is the Langmuir constant relating to the sorption (l/mg) and \( q_{\text{max}} \) is the maximum sorption capacity (mg/mg). Where \( C_e \) (mg/l) is the equilibrium concentration of U(IV) or Am(III), \( q_e \) (mg/mg) is the sorption capacity at equilibrium, \( q_{\text{max}} \) (mg/mg) is the maximum sorption capacity and \( b \) (l/mg) is the Langmuir constant which is related to the sorption energy. A linear plot of \( C_e/q_e \) vs \( C_e \) was obtained.

### 2.6.3. Radionuclide Sorption Kinetics

#### First-Order Kinetics

\[
\log(q_e - q_t) = \log q_e = k_1 t / 2.303
\]

Where \( q_e \) and \( q_t \) (mg/mg) are the amounts of U(IV) or Am(III) sorbed at equilibrium and at time \( t \) (min), respectively, \( k_1 \) (1/min) is the rate constant of the pseudo-first order sorption and \( t \) (min) is the contact time. The adsorption rate constants \( (k_1) \) and the sorption capacity \( (q_e) \) for the U(IV) or Am(III) sorption by AXAD-PR were calculated from the slope and intercept of the plots of \( \log (q_e-q_t) \) against \( t \).

### 2.6.4. Synthesis of AXAD-16-PR resin

The AXAD-PR-pyridylazo resorcinol sorbent (AXAD-PR) is prepared by stirring 10 g of AXAD-PR powder 200 mesh size, with a nitration mixture (20 mL conc. HNO3 and 50 mL conc. H2SO4) at 60 °C in a water bath for 1 h. The reaction mixture is then poured into ice-water followed by filtration and washing for several times with distilled water until acid-free. The nitrated resin is afterwards reduced with a mixture of 40 g SnCl2, 45 mL conc. HCl and 50 mL ethanol and refluxed at 60 °C for 12 h. The product is filtered off, washed with water followed by washing with 2 M NaOH to obtain the free amino polymer. The latter is then treated with 2 M HCl and the excess of which is removed by water washing before it is suspended in 350 mL of ice-water. Subsequently, the reaction mixture is stirred with 1 mL aliquots of 1 M NaNO2 and 1 M HCl until a durable blue color is formed on the starch iodide paper as indicator. The polymer has thus been converted to the diazonium salt and which is washed with cold water followed by it treatment with 250 mL of 10% NaOH containing 5g pyridylazo resorcinol at 0–5 °C and keep at this temperature for 24 h until completion of the reaction. The obtained dark brown powder is filtered off and is finally washed with distilled water. The reactions sequence for the various preparation steps of the AXAD-PR resin functionalized with pyridylazo resorcinol.

### 3. Results

#### 3.1 Effect of increasing AXAD-PR concentrations on the sorption efficiencies of 25 mg/l U(IV) and Am(III)

As the AXAD-PR concentrations were increased from 5 mg/l up to 30 mg/l the sorption efficiency of 25 mg/l U(IV) increased from 57% to 99% in the U(IV) sorption process at pH =5.3 after 25 min contacting time at 29 °C temperature (Figure 2). Further increase of AXAD-PR concentration to 40 and 50 mg/l decreased the U(IV) sorption yield to 90% throughout 25 mg/l U(IV) sorption. As the AXAD-PR concentrations were increased from 5 mg/l up to 40 mg/l, the sorption efficiency of 25 mg/l Am(III) increased from 57% to 99% in the Am(III) sorption process at pH =5.6 after 25 min contacting time at 29 °C temperature. After 40 mg/l AXAD-PR concentration, the increasing of AXAD-PR to 50 mg/l decreased the 25 mg/l Am(III) sorption efficiency to 85% from 99%. U(IV) sorption reached to the maximum at low AXAD-PR concentration in the comparison to the sorption of Am(III) to AXAD-PR for the same operational conditions. Hosseini-Bandegharaei (2016) also found that by increasing the XAD-7 concentrations the removal percentage of Th(IV) increased while further increase of adsorbent dose from 80 to 140mg/l decrease the Th(IV) adsorption yield [9].
3.2. Effects of U(IV) and Am(III) concentrations on the sorption efficiencies on AXAD-PR

The maximum sorption efficiency of U(IV) (97%) was obtained at a U(IV) concentration of 20 mg/l while the maximum sorption efficiency of Am(III) (99%) was obtained at an Am(III) concentration of 30 mg/l after 25 min contacting time at 29°C temperature (Figure 3). The maximum sorption efficiency of U(IV) radionuclide was obtained at low U(IV) concentration compared to the Am(III) sorption. The maximum sorption efficiency of Am(III) was obtained (99%) at higher Am(III) concentration compared to U(IV) sorption. The reached sorption efficiency of Am(III) is higher compared to U(IV) sorption efficiency. As mentioned above at high U(IV) and Am(III) concentrations the adsorption yields of these radionuclides decreased. Similar results were found at Th(VI) adsorption on XAD-7 [9]. It should be mentioned that at higher radionuclide concentrations the lower adsorption yields can be attributed to the lower ratio of initial numbers of radionuclide concentrations to the available adsorbing sides existing on AXAD-PR surface and, also, the higher removal percentages at the low initial Am(III) and U(IV) concentrations.

Figure 2. Effect of AXAD-PR concentrations on the sorption of U(IV) and Am(III) sorption efficiencies at pH =5.3 after 25 min contacting time at 29°C temperature.

Figure 3. Effects of U(IV) and Am(III) concentrations on the sorption efficiencies on 30 mg/l AXAD-PR at pH =5.3 after 25 min contacting time at 29°C temperature.
3.3. Effect of sorption time on the sorption efficiency of U(IV)
The maximum sorption efficiency of 20 mg/l U(IV) was found to be 97% at a contacting time of 30 min at a pH of 5.2 at 29 °C temperature for 25 mg/l AXAD-PR concentration (Figure 4). Low sorption efficiencies was found for the lower AXAD–PR concentration: for example around 79% sorption efficiencies for 30 mg/l U(IV) was found for both 5 and 15 mg/l AXAD-PR concentrations. For higher AXAD-PR concentrations (>25 mg/l) the sorption removal of 30 mg/l U(IV) decreased to around to 45% and 25%. Demir and Uslu 2012 found that 120 min sorption time was the optimum duration for maximum adsorption of 14 gr/l 2, 4, 6 trinitrofenol using 1.5 g/l AXAD–PR [6]. In the comparison of the results it can be concluded that small amount of AXAD-PR is enough to remove the U(IV) from the nuclear wastes although the studied adsorbent and pollutant parameters are different. Hosseini-Bandegharaei et all., (2016) found that the contact time between the Th(VI) and XAD-7 can play important role in the adsorption process [9]. They found that the uptake of Th(VI) increased up to 60 min contacting time and then further increase of time did not affect the adsorption yield.

![Figure 4](image)

**Figure 4.** Effect of sorption time on the sorption efficiency of U(IV) at a contacting time of 30 min at a pH of 5.2 at 29 °C temperature for 25 mg/l AXAD-PR concentration.

3.4. Effect of increasing sorption time on the sorption efficiency of Am(III) versus increasing AXAD-PR concentrations.
The maximum sorption efficiency of 40 mg/l Am(III) was found to be 98% after 40 min contacting for sorption to 25 mg/l AXAD-PR concentration at pH=5.3 at 29 °C temperature (Figure 5). Low sorption efficiencies was found for the lower AXAD–PR concentration: for example around 40-55% sorption efficiencies of 40 mg/l Am(III) was found for both 5, 10 and 15 mg/l AXAD-PR concentrations. For higher AXAD-PR concentrations (>25 mg/l) for 40-50 mg/l AXAD-PR; the sorption removal of 40 mg/l Am(III) decreased to around to 50% and 40%, respectively. The maximum 10 µg/l U(VI) removal was obtained after 25 min and the loading half time was less than 6 min [10]. In the study also lower U(IV) removals was obtained at higher sorption time. This behavior is due to ionic strength of U(VI) is enhanced and the sorption process is inhibited.
3.5. Effect of increasing temperature on the sorption efficiency of U(IV)

As the temperature was increased from 20 °C to 30 °C, the sorption yield of 20 mg/l U(IV) increased from 49% up to 90% at 25 mg/l AXAD-PR concentration after 30 min sorption time (Figure 6) at pH=5.3. At temperatures > 30 °C the sorption yields of 30 mg/l U(IV) radionuclide to 25 mg/l AXAD-PR decreased from 75% to 60% for 25 mg/l AXAD-PR concentration. The maximum U(IV) sorption efficiency was obtained at 30 °C temperature. The sorption mechanism is dependent on the nature of adsorbent. On the type of pollutant as mentioned above, on the pH on the temperature. At high temperatures (60-80 °C) degrease the adsorption yields can be attributed to the increase of wastewater viscosity containing the U(IV) and Am(III) radionuclides. Under these situations the radionuclide ion mobility decreases. Hosseini-Bandegharaei et al. (2016) found different results between Th(VI) sorption and XAD-7. Thus, at Th(VI) concentration >50mg/l higher sorption capacities was observed due to higher ratio of the initial Th(VI) concentrations [9].

Figure 5. Effect of increasing sorption time on the sorption efficiency of Am(III) versus increasing AXAD-PR concentrations at pH=5.3 at 29 °C temperature.

Figure 6. Effect of increasing temperature on the sorption efficiency of U(IV) at 25 mg/l AXAD-PR concentration after 30 min sorption time.
3.6. Effect of increasing temperature on the sorption efficiency of Am(III)

As the temperature was increased from 20 °C to 30 °C and 40 °C the sorption yield of 30 mg/l Am(III) increased from 49% up to 94% at 25 mg/l AXAD-PR concentration after 40 min sorption time at a pH of 5.3 (Figure 7). At temperatures > 30 °C the sorption yields of 40 mg/l Am(III) radionuclide to 25 mg/l AXAD-PR decreased from 75% to 60% for 25 mg/l AXAD-PR concentration. The maximum Am(III) sorption efficiency was obtained at 40 °C temperature. As the temperature increased the raised diffusibility may contribute available Am(III) at the subsurface for more adsorption of radionuclide the decrease in adsorption efficiency at 50, 60, 70 and 80 °C temperature may be due to less favorable adsorption resulting in low adsorption yields of Am(III). Uslu and Demir (2012) found that the adsorption capacity of the XAD-16 is decreasing with increasing temperature from 24.85 °C and 44.85 °C [6].

![Figure 7. Effect of increasing temperature on the sorption efficiency of Am(III) at 25 mg/l AXAD-PR concentration after 40 min sorption time at a pH of 5.3](image)

3.7. Effect of pH on the sorption efficiency of Am(III) and U(IV)

As the pH of the process increased from 2 up to 5; The sorption efficiencies Am(III) and U(IV) increased from 39% and 35% up to 93% and 95 %, respectively (Figure 8). The increasing of pH from acidic (pH=6) up to alkaline (pH=8) level reduced the sorption efficiencies of both radionuclides to around 30% and 12%. The optimum pH for maximum Am(III) and U(IV) sorption yields was found to be 5. Hosseini-Bandegharaei (2016) observed that the degree adsorption of Th(IV) on XAD-7 was close to %90 at the pH range of 4-5 [9]. Therefore in this study and in our study adsorption was directly dependent to the pH. Hosseini-Bandegharaei et al., (2013) measured the pH effect on the sorption of U(VI) on XAD-2010, the sorption increases with increasing pH of sorptive simulated wastewater solution and attains a maximum value at pH 4.2-4.7 [10]. This indicated that the sorption process involves the release of H⁺ ions to allow the firm complexation of U(IV) and Am(III) ions to AXAD-PR at the optimum pH. At the low pH, the release of H⁺ ions was not efficient because of adsorbent could not be used efficiently for preconcentrations of U(IV) AND Am(III) in the low concentration range. Gaedi et al., (2014) studied the heavy metal adsorption at lower pH (pH > pHzp) [11]. In these pH values the XAD-7 has positive charge and at higher value it takes negative charge. Therefore, it seems that the pollutants are significantly and thermodynamically more fissile to accumulate on XAD at pH higher than pHzp. The approximate pHzp of modified AXAD-PR was found to be around 5. Therefore, it has neutral charge at this pH and confirms that the main mechanism for binding radionuclides and their enrichment is complexation with proposed method at pH >5 the competition of h⁺ ions with radionuclides for binding to reactive centers of XAD leads to significant reduction of adsorption of radionuclides. Therefor pH 5 was recommended to improve the sorption of radionuclides on XAD.
Figure 8. Effect of pH on the sorption efficiency of Am(III) and U(IV) at 25 mg/l AXAD-PR concentration after 40 min sorption time.

3.8. First order reaction kinetics of U(IV)
The sorption studies of 20 mg/l U(IV) to AXAD-PR showed a first order sorption kinetic with $R^2$ values of 0.99 and first order reaction kinetic constants ($K_1$) of $1 \times 10^{-4}$, $2 \times 10^{-4}$ and $3 \times 10^{-2}$ 1/min for 15, 30 and 50 mg/l AXAD-PR concentrations after 30 min sorption time at a temperature of 30 °C and a pH of 5.3 (Figure 9). Hosseini-Bandegharai et al., (2016) also found that the Th(VI) sorption on XAD-7 follows first order reaction with a $q_{max}$ level of 53.4 mg/g [9].

Figure 9. First order reaction kinetics of U(IV) at 20 mg/l U(IV); 15, 30 and 50 mg/l AXAD-PR concentrations after 30 min sorption time at a temperature of 30 °C and a pH of 5.3.

3.9. First order reaction kinetics of Am(III)
The sorption studies of 30 mg/l Am(III) to AXAD-PR showed a first order sorption kinetic with $R^2$ values of 0.99 and first order reaction kinetic constants ($K_1$) of $1.3 \times 10^{-4}$, $2.98 \times 10^{-4}$ and $3.67 \times 10^{-2}$ 1/min for 15, 30 and 50 mg/l AXAD-PR concentrations after 30 min sorption time at a temperature of 30 °C and a pH of 5.3 after 40 min contacting time (Figure 10). Uslu and Demir (2012) obtained that the adsorption of 2, 4, 6, trinitrofenol on XAD-16 fallows pseudo second order reaction kinetics.
The differences of adsorption kinetic can be attributed to the different wastewater type pollutant kind, to the pH and to the different environmental studied.

![Graph showing adsorption kinetics for Am(III) concentrations at 30 mg/l AXAD-PR at 30 °C and pH 5.3 after 40 min contacting time.](image)

**Figure 10.** First order reaction kinetics of Am(III) for 30 mg/l Am(III); 50, 15 and 30 mg/l AXAD-PR concentrations at a temperature of 30 °C and a pH of 5.3 after 40 min contacting time.

### 3.10. Langmuir sorption isotherms for U(IV) and Am(III)

The sorption data were used to evaluate the sorption kinetic of U(IV) and Am(III). Both radionuclides were sorbed according to Langmuir isotherm with high $R^2$ values and acceptable Langmuir Kinetic Constants ($K_L$) (Figures 11a and 11b). The $R^2$ values for U(IV) and Am(III) were calculated as 1.09 l/mg and 1.01 l/mg, respectively, while $q_{max}$ values were calculated as 19.8 mg/mg and 23.6 mg/mg, respectively. The Langmuir adsorption isotherm is based on the assumption that all adsorption sides are equivalent and that adsorption is an active side, is independent of whether the adjacent sides are occupied or not. The fact that the Langmuir isotherm fits the experimental data very well may be due to homogenous distribution of active chelating sides on the AXAD-PR since, the Langmuir equations assumes that the adsorbent surface is homogenous. Uslu and Demir (2012) also observed that the AXAD-PR exhibited Langmuir isotherm with a $K_L$ level of 0.0410 l/mg [6]. These results indicate that AXAD-PR shows similar responses to the adsorption process although the pollutants are quite different. Borin and Pavko (2009) also found that adsorption kinetic of vancomycin on AXAD-PR can be definite with Langmuir isotherm [7]. Anaia et al., (2014) found that 1-(2-thiazolylazo)-2-naphtol sorbet to Amberlite xad-7 by follow the kinetic proposed by Langmuir [8].
3.11. Recovery of U(IV) and Am(III)

The sorption efficiencies in both radionuclides were not reduced significantly, after four times sequential utilization of AXAD-PR (Table 3). Hosseini-Bandegharaei et al., (2015) also found higher Amberlite XAD-7 recoveries in the U(IV) and Th(VI) sorption [13] while Hosseini-Bandegharaei et all., (2016) obtained % 70 AXD-7 desorption-recovery yield in the sorption studies of Th(VI) [9].

Table 3. Sequential treatment of 20 mg/l U(IV) and 30 mg/l Am(III) in the presence of 30 mg/l AXAD-PR concentration for U(IV) and 40 mg/l AXAD-PR concentration for Am(III)

| Sorption efficiency (%) | Cycles   |
|-------------------------|----------|
|                         | First    | second   | third    | Fourth   |
| 20 mg/l U(IV)           | 96%      | 92%      | 88%      | 85%      |
| 30 mg/l Am(III)         | 98%      | 96%      | 90%      | 88%      |
| 50 mg/l U(IV)           | 75%      | 70%      | 65%      | 60%      |
| 50 mg/l Am(III)         | 78%      | 73%      | 70%      | 68%      |

4. Conclusions

Among the kinetic studies it was found that the adsorption kinetic was first order ($K_1 = 2 \times 10^{-3} \text{ min}^{-1}$) while the adsorption of radionuclides to AXAD-PR isotherm was in accordance with Langmuir. The AXAD-PR can be reused again with high yields after four sequential utilization times. The regeneration of the composite is possible and can be attained a desorption efficiencies of 85% and 88% for U(IV) and Am(III) removals using 0.4 M HNO$_3$ acid solution using 20 and 30 mg/l
prepared AXAD-PR. The maximum adsorption yields of U(IV) and Am(III) were found to be 96% and 95%, respectively, after 25 min contacting time using 30 and 40 mg/l AXAD-PR, respectively, at 30 °C at a pH of 5.

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