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Sorption studies of uranium and thorium on activated carbon prepared from olive stones: Kinetic and thermodynamic aspects

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ABSTRACT

Activated carbon prepared by the chemical activation of olive stone was examined for the sorption of uranium and thorium from aqueous solutions. Precursor/activating agent ($ZnCl_2$) ratio (1:2) and 500 °C carbonization temperature were used for the preparation of the sorbent. The total sorption capacities were found to be 0.171 and 0.087 mmol g^{-1} for uranium and thorium, respectively. The sorption of uranium and thorium was studied as a function of shaking time, pH, initial metal ion concentration, temperature and adsorbent concentration in a batch system. The sorption followed pseudo-second-order kinetics. ΔH° and ΔS° values for thorium and uranium sorption were calculated from the slope and intercept of plots of $\ln K_d$ versus $1/T$. The positive values of ΔH° indicate the endothermic nature of the process for both metals and decrease in the value of ΔG° with rise in temperature show that the sorption is more favorable at high temperature.

1. Introduction

Many activities dealing with radioactive materials produce low, intermediate and high level radioactive wastes that require advanced treatment [1–3]. Evaporation, precipitation, ion exchange, or adsorption may be used to concentrate wastes for further treatment or disposal. Among these separation techniques; adsorption is one of the most widely used one. Especially, activated carbon adsorption is often used in nuclear industry due to its selective adsorption, high adsorption capacity, high radiation stability and high purity [4].

Active carbons can be produced almost any carbonaceous material. Agricultural by-products represent an important and economic source for the production of carbon adsorbents, particularly where coal as a conventional source is not available. Many investigations have been devoted for the preparation and characterization of carbons and activated carbons from agricultural by-products; fruit stones, shells, seeds and olive stones [5–9].

The removal of radionuclide and toxic heavy metals such as uranium and thorium from aqueous waste solutions seems to be a significantly useful subject for environmental control [10]. The major use of uranium and thorium as nuclear fuel for the production of nuclear energy in a nuclear reactor is well known [11]. Nuclear power is derived from uranium, which has no significant commercial use other than as a fuel for electricity generation. Thorium can be used as an alternate nuclear fuel by converting into ^{233}U. Unlike uranium, thorium and its compounds and alloys find widespread use. Thorium oxide finds application as catalyst, high temperature ceramic and high quality lenses [12]. In view of the extensive usage of uranium and thorium for various industrial purposes and their toxicity, precise determination of these metals in environmental, metallurgical, geological materials is of greatest importance prior to pollution control measures or its use as alloys or in understanding the correlation between diseases of animals and aquatic organisms and soil chemistry.

Many researchers have been studying the removal of uranium and thorium using different types of activated carbons. Metaxas et al. [13] reported different types of adsorbents including activated carbons for the adsorption of thorium. Mahramanlioglu [14] tested the activated carbons from used tires adsorption of uranium. There are several research related to adsorption of uranium on activated carbons in the literature [15–19]. Furthermore activation processes by using gases for the production of activated carbons from olive stones has been investigated of many researchers [20–22].

In this study, olive stones were used as the starting material for the production of activated carbon owing to Aegean Region of Turkey is famous with olive trees. Therefore, there are considerable amount of olive stones as an agricultural by-product which can be widely used as a precursor for the preparation of activated carbon. Chemical activation was used for the preparation of activated carbon. Adsorption experiments were made by batch technique.

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2. Materials and methods

2.1. Chemicals and reagents

$UO_2(NO_3)_2.6H_2O$ and $Th(NO_3)_4.5H_2O$ were obtained from Merck Co. A stock solution of uranium and thorium (1 g l^{-1}) was prepared by dissolving $UO_2(NO_3)_2.6H_2O$ and $Th(NO_3)_4.5H_2O$ in deionized water and acidified with concentrated HNO_3 to prevent hydrolysis. Unless stated otherwise, all chemicals and reagents used in this study were of analytical reagent (AR) grade.

2.2. Preparation of the adsorbent and its characterization

Olive stone was used as the starting material for the preparation of activated carbon in this study. Olive stones were collected, washed with tap water, dried at 100 °C and crushed. Activated carbon was prepared by chemical activation of the starting material using $ZnCl_2$ as chemical agent. The preparation method has been described elsewhere [23]. The starting material was mixed by stirring with $ZnCl_2$ solution in the ratios of (starting material/activating agent) 1:1 and 1:2. The resulting chemical-loaded samples were then heated to varying carbonization temperatures from 500 to 700 °C.

The adsorption capacity of activated carbons is determined by their porous structure and surface area but is also strongly influenced by the presence of functional groups at the carbon surface. Activated carbons are known to contain a variety of heteroatoms such as oxygen, hydrogen, chlorine, and sulfur. Heteroatoms are either derived from the starting material, and become a part of the chemical structure as a result of imperfect carbonization, or chemically bonded to the carbon during activation or during subsequent additional treatments, such as oxidation. The most common are carboxyl, carbonyl, carbonyl (quinone type), phenolic hydroxyl, anhydride, ether-type, lactone, and lactal [24]. For this purpose FTIR spectroscopy was used to understand the surface characteristics of resulted activated carbon. The internal structure of the adsorbent was examined using a scanning electron microscopy.

Textural characteristics of the activated carbon were determined by nitrogen adsorption at 77.15 K using an automatic Micromeritics ASAP-2020 surface area analyzer. The Brunauer–Emmett–Teller (BET) surface area, micropore volume, and micropore surface area of the activated carbons were determined using BET method, using the software available with the instrument. The Barrett–Joyner–Halenda (BJH) method was used to calculate the pore size distribution.

2.3. Adsorption experiments

Batch adsorption experiments were performed by a thermostatically controlled shaker (GFL-1083 model). In the experiments, activated carbon was separately shaken with metal solutions at varying experimental conditions in 25 ml Erlenmeyer flasks at a speed of 100 rpm. The concentration of uranium and thorium was determined before and after equilibrium by the Perkin Elmer Optima 2000 DV Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES). All the experiments were carried out in duplicate and the mean value was used in all cases.

The amount of metal adsorbed by activated carbon was calculated using the following equation:

$$q_e = (C_0 - C_e) \times V/m \quad (\text{mg/g}) \tag{1}$$

where q_e is the metal uptake (mg g^{-1}), C_0 and C_e are the initial and equilibrium metal concentrations in the solution (mg l^{-1}), respectively, V is the solution volume (l), and m is the mass of adsorbent (g).

The distribution coefficient (K_d) of metal ions between the aqueous phase and the solid phase can be directly obtained using:

$$K_d = \frac{C_0 - C_e}{C_e} \times \frac{V}{m} \quad (\text{ml/g}) \tag{2}$$

where C_0 (g l^{-1}) and C_e (g l^{-1}) are initial and equilibrium uranium or thorium concentrations, respectively, V/m is the ratio of the volume of the metal solution (ml) to the amount of adsorbent (g) in a batch.

3. Results and discussion

3.1. Total sorption capacity

The total sorption capacity studies were carried out using the batch technique for uranium and thorium. The ratios of (starting material/activating agent) 1:1 and 1:2 and varying carbonization temperatures from 500 to 700 °C were examined. Adsorbent (0.1 g) was equilibrated with 200 mg l^{-1} metal ions for 3 h at 30 °C. All activated carbons have a particle size of <0.125 mm. The smaller particle sizes of a porous carbon have the greater rate of diffusion and adsorption. Therefore the smaller particles have more outer surface for contacting the adsorbate solution [23,24]. The amount of metal ions sorbed on the activated carbon was calculated from the difference in the metal ion concentration in the solution before and after sorption. Fig. 1 shows that the preparation conditions and total sorption capacities of the activated carbons. The results show that uranium sorption capacities are very similar for all types of adsorbents. But in the case of thorium, 500 °C carbonization temperature and starting material/activating agent ratio of 1:2 has the highest sorption capacity. The total sorption capacities of this adsorbent were found to be 0.171 and 0.087 mmol g^{-1} for uranium and thorium, respectively.

Uranium adsorption has been previously investigated by some researchers. For instance, Karadeniz et al. [25] have used commercially available activated carbon (Merck) for the uranium adsorption, and found the adsorption capacity of 0.120 mmol g^{-1} which is slightly lower than that of observed for olive stone. The similar result was found for Th adsorption worked by Metaxas et al. [13]. Activated carbon prepared by the activation of olive stone with a steam/nitrogen mixture, and K_d values found as 123.4 ml g^{-1} which is lower than the value obtained in current work (K_d = 170.0 ml g^{-1}). From this point of view starting material/activating agent ratio of 1:2 and 500 °C carbonization temperature were used for the preparation of the adsorbent and tested for the adsorption of uranium and thorium.

Fig. 2 shows the intra-structure of the prepared adsorbent obtained by scanning electron microscopy (SEM). From the picture different pore sizes may be observed.

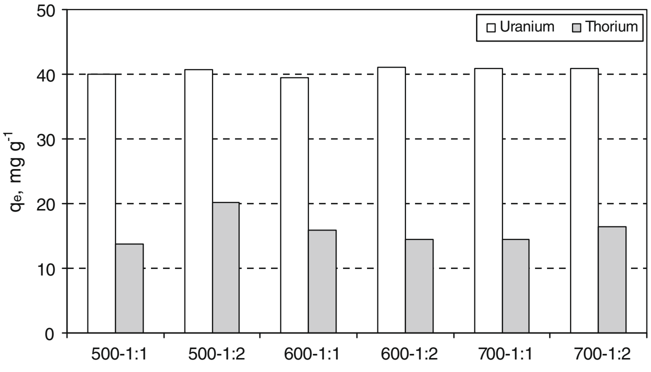

Fig. 1. Preparation conditions and adsorption capacities of the activated carbons.

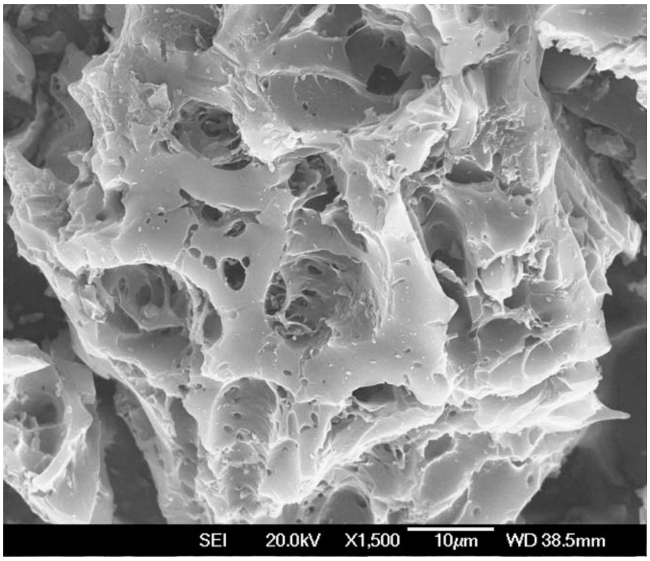

Fig. 2. SEM photograph of activated carbon.

The BET surface area of activated carbon is found as 464.68 m^2/g. BJH adsorption cumulative surface area of pores (17 Å < d < 3000 Å) is 0.521 m^3/g, whereas BJH adsorption cumulative volume of pores (17 Å < d < 3000 Å) is 0.020 cm^3/g. BJH adsorption average pore diameter is 1559.33 Å. According to t-plot analysis micropore volume is 0.11 cm^3/g and micropore area is 259.6 m^2/g.

FTIR spectroscopy was used to understand the surface characteristics of resulted activated carbon which has starting material/activating agent ratio of 1:2 and 500 °C carbonization temperature. As can be seen in Fig. 3, the absorption bands observed in the region 1000–1440 cm^{-1} was assigned to the C–O stretching and O–H bending modes [6]. The band in the region of 1800–1540 cm^{-1} was associated with the C=O stretching mode in carbonyls, carboxylic acids, and lactones [6,21,24].

3.2. Effect of contact time

The effect of contact time was investigated for the adsorption of U(VI) and Th(IV) on activated carbon as a function of shaking time varying from 5 to 240 min. Fig. 4 shows that increasing shaking time does not affect significantly the adsorption of U(VI) on activated carbon. But in case of thorium adsorption, the adsorption slightly increases with an increase of shaking time and reaches equilibrium within 30 min. Therefore, 5 min and 30 min shaking times were chosen as optimum contact times for uranium and thorium, respectively.

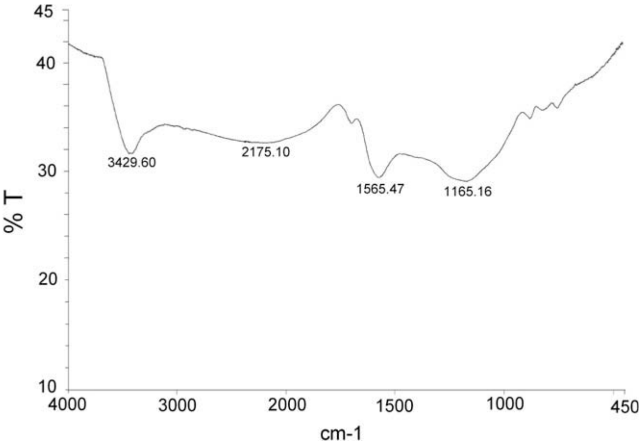

Fig. 3. FTIR spectroscopy of activated carbon.

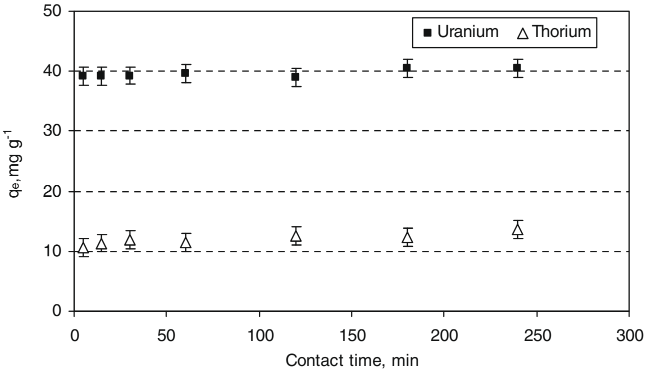

Fig. 4. Effect of shaking time on uranium and thorium adsorption (initial uranium and thorium concentrations: 200 mg l^{-1}; temperature: 30 °C; adsorbent amount: 0.1 g).

3.3. Kinetics of adsorption

In order to study the specific rate constant of uranium and thorium activated carbon system, a pseudo-second-order rate equation [26] was used to simulate the kinetic sorption of uranium and thorium on activated carbon.

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t \qquad (3)$$

where q_t (mg/g) is the amount of uranium or thorium adsorbed on activated carbon at time t, and q_e (mg/g) is the equilibrium adsorption capacity. K_2 (g mg^{-1} h^{-1}) is the pseudo-second-order rate constant of adsorption. The straight-line plots of t/q_t versus t (Fig. 5) indicate that the kinetic sorption of uranium and thorium on activated carbon is well described by the pseudo-second-order rate equation. The values of K_2 and q_e which are calculated from the intercept and slope of Eq. (3) were given in Table 1.

The correlation coefficients of the pseudo-second-order rate equation for the linear plot suggests that the kinetic adsorption can be described by the pseudo-second-order rate equation very well.

3.4. Effect of solution pH

The pH dependence of metal ion adsorption is a complex phenomenon and strongly influences the metal ion adsorption. Fig. 6 presents the effect of pH on the amount of metal ions adsorbed

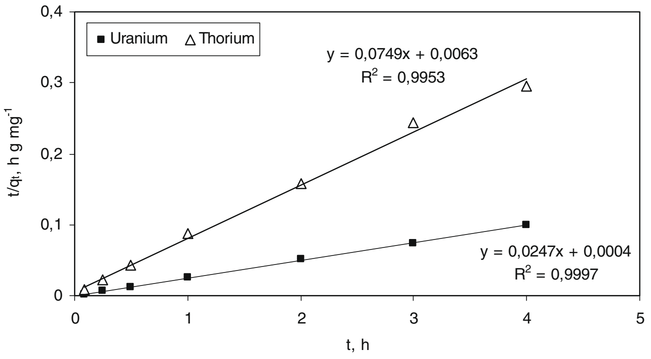

Fig. 5. Pseudo-second-order kinetic plots for the adsorption of uranium and thorium onto activated carbon.

Table 1
Kinetic parameters for the adsorption of uranium and thorium onto activated carbon.

	K_2 (g mg^{-1} h^{-1})	q_e (mg g^{-1})	R^2
Pseudo-second-order			
Uranium	1.53	40.49	0.9997
Thorium	0.89	13.35	0.9953

onto activated carbon with respect to q_e (mg g^{-1}). Uranium solution pH was adjusted using Na_2CO_3 and HNO_3. Carbonates can strongly coordinate uranyl ion. About pH 7, the uranyl ion forms very stable complexes with carbonate. As can be seen from the Fig. 6, adsorption slightly decreases as the pH higher than 6, this is because of the formation of $(UO_2(CO_3)_2)^{2-}$ and/or $(UO_2(CO_3)_3)^{4-}$ stable complexes [27].

Sodium acetate and acetic acid were used for adjusting thorium solution pH. As it was known that when pH is around 4.0, the predominant thorium complex ions would be $[ThCH_3COO]^{3+}$ and $[Th(CH_3COO)_2]^{2+}$, while the complexes $[Th(CH_3COO)_2]^{2+}$ and $[Th(CH_3COO)_3]^{+}$ are dominant when pH is around 5.0. So, it could be concluded from the experimental results that the size of Th(IV) complex ions is an important factor influencing the adsorption capacity on the adsorbent [28]. Hence, pH 6 for uranium and pH 4 for thorium were used all further studies.

3.5. Effect of initial metal ion concentration

The effect of initial metal ion concentration on the adsorption of uranium and thorium was studied in the range of 25–300 mg l^{-1}. The amount of uranium and thorium adsorbed on the adsorbent (mg g^{-1}) increased with the initial concentration of the metal ions. As can be seen from Fig. 7, an increase in the initial metal ion concentration from 25 to 300 mg l^{-1}, leads to an increase in the amount of metal ions from 6.1 to 52.2 mg g^{-1} for uranium and 4.5–18.3 mg g^{-1} for thorium adsorbed on activated carbon. The increase in initial metal ion concentration enhances the interaction between adsorbate and the adsorbent.

3.6. Effect of temperature

Fig. 8 shows that the effect of temperature on the adsorption of uranium and thorium on activated carbon. Uptake of uranium and thorium slightly increases with increasing temperature.

Adsorption enthalpy was measured using the method based on the Van't Hoff plot. The values of $\Delta H°$ and $\Delta S°$ are calculated from the slopes and intercepts of the linear variation of ln K_d with reciprocal temperature, $1/T$, Fig. 9, using the relation:

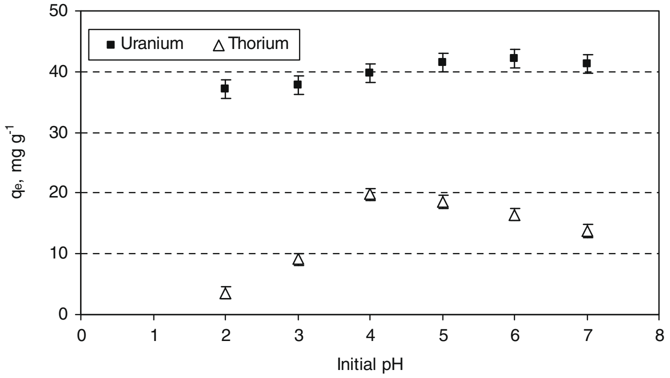

Fig. 6. Effect of initial pH on uranium and thorium adsorption on activated carbon (initial uranium and thorium concentrations: 200 mg l^{-1}; shaking time: 5 min for uranium, 30 min for thorium; temperature: 30 °C; adsorbent amount: 0.1 g).

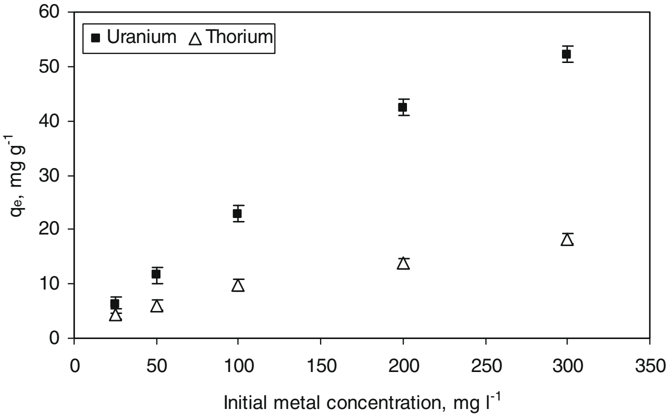

Fig. 7. Effect of initial uranium and thorium concentration of activated carbon adsorption (initial pH: 6 for uranium, pH: 4 for thorium; shaking time: 5 min for uranium, 30 min for thorium; temperature: 30 °C; adsorbent amount: 0.1 g).

$$\ln K_d = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT} \tag{4}$$

where $\Delta H°$ and $\Delta S°$ are the standard enthalpy and entropy changes of adsorption, respectively. The free energy of specific adsorption $\Delta G°$ is calculated using the equation:

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \tag{5}$$

$\Delta G°$ values are also given in Table 2. Positive values of $\Delta H°$ indicate the endothermic nature of the adsorption process for both metals and decrease in the value of $\Delta G°$ with rise in temperature show that the adsorption is more favorable at high temperature.

3.7. Effect of adsorbent concentration

The effect of the adsorbent concentration on the adsorption of uranium and thorium was studied. As seen from Fig. 10, the adsorption of uranium and thorium decreases with the rise of the concentration of adsorbent. Increasing number of adsorbent particles in the solution allows that more uranium and thorium ions to interact with the reaction sites.

3.8. Adsorption isotherms of uranium and thorium

In order to understand the adsorption capacity of the adsorbent, the equilibrium data were evaluated according to the Freundlich,

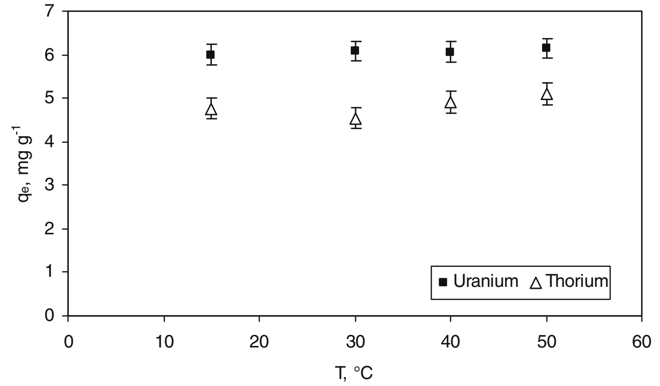

Fig. 8. Effect of temperature on the uranium and thorium adsorption (initial uranium and thorium concentration: 25 mg l^{-1}; initial pH: 6 for uranium, pH: 4 for thorium; shaking time: 5 min for uranium, 30 min for thorium; adsorbent amount: 0.1 g).

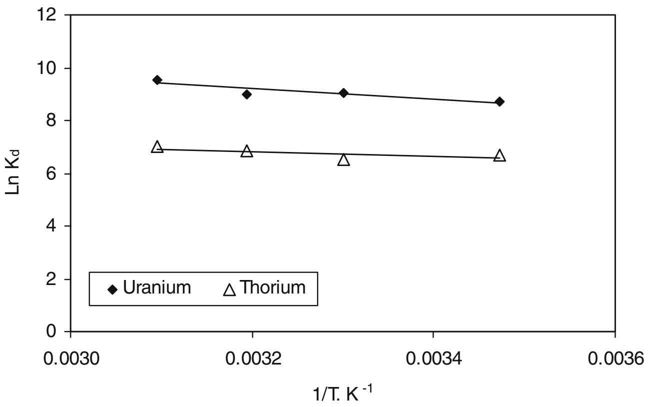

Fig. 9. Plots of ln K_d versus. $1/T$ for uranium and thorium adsorption on activated carbon.

Table 2
Thermodynamic parameters for uranium and thorium adsorption on activated carbon.

	ΔH° (kJ/mol)	ΔS°(kJ/molK)	ΔG° (kJ/mol)			
			288 K	303 K	313 K	323 K
Uranium	16.59	0.13	−20.76	−22.71	−24.00	−25.30
Thorium	7.42	0.08	−15.73	−16.94	−17.74	−18.54

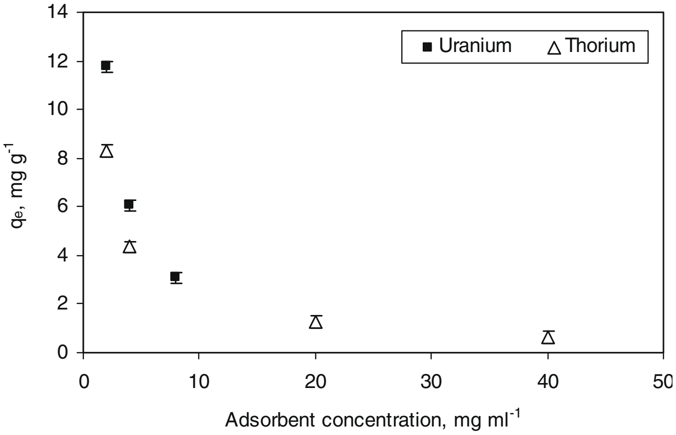

Fig. 10. Effect of adsorbent concentration on the uranium and thorium adsorption (Initial uranium and thorium concentration: 25 mg l^{-1}; Initial pH: 6 for uranium, pH: 4 for thorium; shaking time: 5 min for uranium, 30 min for thorium; temperature: 30 °C).

Langmuir and D–R isotherms. The Langmuir equation [29], which has been successfully applied to many adsorptions, is given by:

$$\frac{C_e}{q_e} = \frac{1}{bn_m} + \frac{C_e}{n_m} \tag{6}$$

where C_e is the equilibrium concentration (mg l^{-1}), q_e is the amount absorbed at equilibrium (mg g^{-1}), n_m and b is the Langmuir constants related to monolayer capacity and energy of adsorption. A linearised plot of C_e/q_e versus C_e is obtained for uranium and thorium adsorption on activated carbon as shown in Fig. 11.

The Freundlich equation, which was applied for the adsorption of uranium and thorium is given as,

$$q_e = KC_e^{1/n} \tag{7}$$

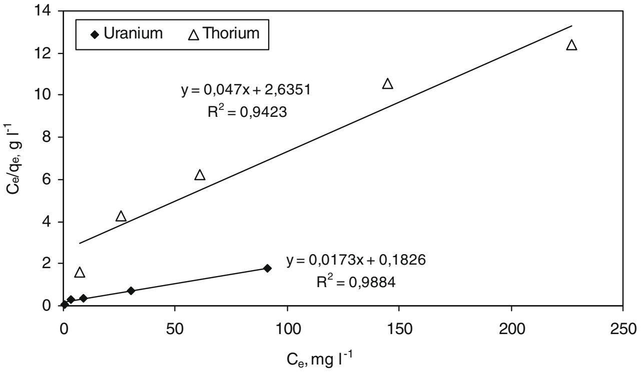

Fig. 11. Langmuir plots for the adsorption of uranium and thorium on activated carbon.

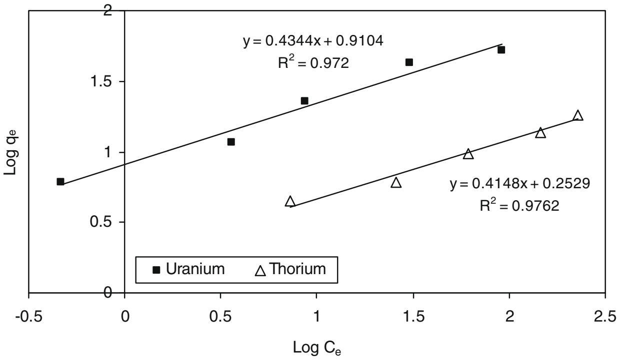

Fig. 12. Freundlich plots for the adsorption of uranium and thorium on activated carbon.

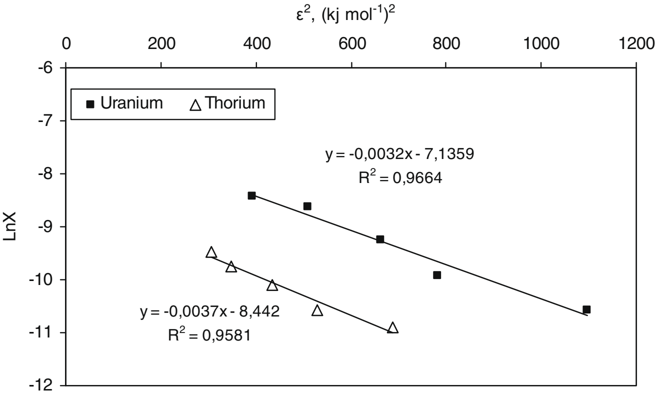

Fig. 13. D–R plots for the adsorption of uranium and thorium on activated carbon.

Eq. (7) can be rearranged to linear form,

$$\log q_e = \log K + \frac{1}{n}\log C_e \tag{8}$$

where q_e is the amount of solute adsorbed per mass unit adsorbent, C_e is the equilibrium concentration, K and n are the Freundlich constants characteristic of a particular adsorption isotherm and can be evaluated from the intercept and slope of the linear plot of $\log q_e$ versus $\log C_e$ (Fig. 12).

The experimental data were fitted using the Dubinin–Radushkevich (D–R) equation in order to understand the adsorption type.

Table 3
Adsorption isotherm constants for the adsorption of uranium and thorium on activated carbon.

	Freundlich			Langmuir			Dubinin-Radushkevich			
	R^2	n	K	R^2	n_m (mg/g)	b (l/mg)	R^2	K (mol^{2}kJ^{-2})	X_m (mol/kg)	E_{DR} (kJ/mol)
Uranium	0.9720	2.30	8.14	0.9884	57.80	0.095	0.9664	3.2×10^{-3}	7.9×10^{-4}	12.5
Thorium	0.9762	2.41	1.79	0.9423	21.28	0.018	0.9581	3.7×10^{-3}	2.2×10^{-4}	11.6

The linearised D–R equation can be written as,

$$\ln X = \ln X_m - K\varepsilon^2 \tag{9}$$

where ε is Polanyi potential and is equal to,

$$\varepsilon = RT \ln\left(1 + \frac{1}{C_e}\right) \tag{10}$$

X is the amount of metal adsorbed per unit mass of adsorbent, X_m is the theoretical adsorption capacity, C_e is the equilibrium concentration of metal, K is the constant related to adsorption energy, R is the universal gas constant and T is the temperature in Kelvin.

Fig. 13 shows the plot of $\ln X$ against ε^2. DR isotherm constants K and X_m were evaluated from the slope and intercept of the plot, respectively. The mean free energy of adsorption (E_{DR}) is the free energy change when one mole of a solute is transferred to the surface of the adsorbent from infinity in the solution, and that has been calculated by

$$E_{DR} = (-2K)^{-1/2} \tag{11}$$

The magnitude of E_{DR} (kJ mol^{-1}) is useful for estimating the type of adsorption reaction, and if it ranged between 8.0 and 16.0 the adsorption should be taken place by ion exchange reaction. The D–R parameters and mean free energies evaluated are shown in Table 3. In the present case, the E_{DR} value obtained (12.5 and 11.6 kJ mol^{-1}, for uranium and thorium, respectively), which indicates ion exchange mechanism.

The corresponding Freundlich, Langmuir and D–R parameters along with correlation coefficients are reported in Table 3. The correlation coefficients indicate that the Langmuir model fits better than the Freundlich model for the uranium adsorption on activated carbon (correlation coefficient is 0.99). n_m and b was found as 57.80 mg g^{-1} and 0.095 l mg^{-1}, respectively. In the case of thorium, the adsorption can be modelled by the Freundlich Isotherms. The K and n values, respectively, were found to be 1.79 and 2.41 (correlation coefficient is 0.98).

4. Conclusion

In the present study the preparation conditions of activated carbon from olive stones and its uranium and thorium adsorption properties are reported. Chemical activation process was used to prepare activated carbon from olive stones. $ZnCl_2$ was selected as an activating agent and 1:2 (starting material/activating agent) ratio and 500 °C carbonization temperature were determined as the optimum conditions to be used for the uranium and thorium adsorption. Adsorption experiments were conducted by batch technique. Uranium and thorium adsorption on activated carbon prepared from olive stone was pH dependent and the maximum adsorption occurred at pH 6 and pH 4, respectively. The kinetic data conformed better to the pseudo-second-order equation. Thermodynamic parameters $\Delta H°$, $\Delta S°$ and $\Delta G°$ were estimated and these parameters show that adsorption is endothermic. The experimental results indicated that the activated carbon from olive stone can be effectively applied to the removal of uranium and thorium from aqueous solutions.

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References

[1] M.-S. Yim, K.L. Murty, JOM 52 (2000) 26–29.
[2] K. Sakr, M.S. Sayed, M.B. Hafez, J. Radioanal. Nucl. Chem. 256 (2003) 179–184.
[3] T. Özdemir, A. Usanmaz, Prog. Nucl. Energy 51 (2009) 240–245.
[4] R. Qadeer, J. Hanif, M. Saleem, M. Afzal, J. Radioanal. Nucl. Chem. Lett. 165 (1992) 243–253.
[5] M.A. Hourieh, M.N. Alaya, J. Dabbagh, R. J. Aleppo Univ. Basic Sci. Ser. 14 (1992) 65–84.
[6] S. Ricordel, S. Taha, I. Cisse, G. Dorange, Sep. Purif. Technol. 24 (2001) 389–401.
[7] P. Galiatsatou, M. Metaxas, V. Kasselouri-Rigopoulou, J. Hazard. Mater. B91 (2002) 187–203.
[8] P.K. Malik, Dyes Pigments 56 (2003) 239–249.
[9] K.P. Singh, D. Mohan, S. Sinha, G.S. Tondon, D. Gosh, Ind. Eng. Chem. Res. 42 (2003) 1965–1976.
[10] T. Tsuruta, J. Alloys Compd. 408–412 (2006) 1312–1315.
[11] Y.A. El-Nadi, J.A. Daoud, J. Radioanal. Nucl. Chem. 265 (2005) 447–454.
[12] T. Prasada Rao, P. Metilda, J. Mary Gladis, Talanta 68 (2006) 1047–1064.
[13] M. Metaxas, V. Kasselouri-Rigopoulou, P. Galiatsatou, C. Konstantopoulou, D. Oikonomou, J. Hazard. Mater. B97 (2003) 71–82.
[14] M. Mahramanlioglu, J. Radioanal. Nucl. Chem. 256 (2003) 99–105.
[15] M. Saleem, M. Afzal, R. Qadeer, J. Hanif, Sep. Sci. Technol. 27 (1992) 239–253.
[16] W.A. Abbasi, M. Streat, Sep. Sci. Technol. 29 (1994) 1217–1230.
[17] G.I. Park, H.S. Park, S.I. Woo, Sep. Sci. Technol. 34 (1999) 833–854.
[18] S.J. Coleman, P.R. Coronado, R.S. Maxwell, J.G. Reynolds, Environ. Sci. Technol. 37 (2003) 2286–2290.
[19] A.M. Starvin, T. Prasada Rao, Talanta 63 (2004) 225–232.
[20] F. Rodriguez-Reinoso, M. Molina-Sabio, M.T. Gonzalez, Carbon 33 (1995) 15–23.
[21] C. Moreno-Castilla, F. Carrasco-Marín, F.J. Maldonado-Hódar, J. Rivera-Utrilla, Carbon 36 (1998) 145–151.
[22] M.A. Rodríguez-Valero, M. Martínez-Escandell, M. Molina-Sabio, F. Rodríguez-Reinoso, Carbon 39 (2001) 287–324.
[23] C. Kütahyali, M. Eral, Sep. Purif. Technol. 40 (2004) 109–114.
[24] M. Ahmedna, W.E. Marshall, R.M. Rao, Bull. Number 869 (2000).
[25] M. Karadeniz, C. Kütahyalı, M. Eral, Turk. J. Nucl. Sci. 29 (2004) 19–25.
[26] Y.S. Ho, G. McKay, Chem. Eng. J. 70 (1998) 115–124.
[27] T.J. Sorg, in: C.R. Cothern, P.A. Rebers (Eds.), Radon, Radium and Uranium in Drinking Water, Lewis, USA, 1991, pp. 173–191.
[28] X. Liao, L. Li, B. Shi, J. Radioanal. Nucl. Chem. 260 (2004) 619–625.
[29] I. Langmuir, J. Am. Chem. Soc. 40 (1918) 1361–1403.