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Evaluation of Uranium Removal from Aqueous Solution using Orange Peels in the Fixed Bed System

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Abstract

In the present work, removal of uranium (VI) using orange peels studied in a fixed-bed system. The parameters of bed heights, initial concentrations, and flow rates were studied, and comparison between experimental results and Thomas, Yoon-Nelson, and Adams-Bohart models was carried out. Thomas and Yoon-Nelson models are good agreement with experimental data than the Adams-Bohart model.

Keywords: Uranium; Orange peels; Adsorption; Kinetic; Column

Nomenclature

Ct: Effluent concentration (mg L^{-1}),

$C_{0:}$ Influent (Initial) concentration (mg L^{-1}),

F: Linear flow rate (L min^{-1}),

Cab: Adam-Bohart constant (L mg^{-1} min^{-1}),

Kit: Thomas rate constant (L min^{-1} mg^{-1}),

k_{YN}: Yoon-Nelson constant (L min^{-1}),

N_0: Saturation concentration (mg L^{-1}),

Q: Flow rate (ml/min),

q_e: Adsorption capacity at equilibrium, (mg of U (VI) /g adsorbate),

t: time (min),

τ: the time required for 50% adsorbate breakthrough (min),

x: Amount of adsorbent in the column (g),

Z: Bed depth of column (cm).

Introduction

Uranium is the important element in nuclear industries. So that the removal preconcentration and recovery of uranium are the main purpose of nuclear wastewater treatment. Various methods are carried out for the removal of uranium ions from nuclear wastewaters. Chemical precipitation, membrane processes, ion exchange, solvent extraction, photocatalysis and adsorption. Adsorption process is an effective method in the removal of low concentrations of uranium ion from a large volume of solution. Various sorbent materials such as activated carbon [1-3], synthetic [4-8] and natural zeolite [9,10], hematite [11], biotit [12], natural sepiolite [13], akaganeite [14], Orange peels [15], palm-shell [16], and biomasses [17] have been studied.

The Purpose of this work was to examine the removal of uranium ion by orange peels using a fixed bed column from aqueous solution. The parameters of flow rate, initial ion concentrations and bed height were studied. The evaluation of adsorption performance was carried out using Thomas, Adams-Bohart and Yoon-Nelson models.

Materials and Methods

Materials and reagents

Orange peels were prepared according to the procedures reported in my previous work [15]. 1000 mg/L of uranium solutions were prepared by dissolving appropriate amounts of $UO_2(NO_3)_2{\cdot}6H_2O$, Aldrich, USA, in distilled water. For experiments the required concentration were prepared by dilution. The concentrations of U (VI) in solution were determined spectrophotometrically employing Shimadzu UV-VIS-1601 spectrophotometer using arsenazo (III) as complexing reagent [18].

Experimental procedure

Fixed bed system studies were carried out using a glass column of 2 cm internal diameter and 50 cm height. Orange peels were packed in the column with a layer of glass wool at the bottom. Bed heights of 3, 6 and 9 cm were used. Three flow rates (1, 2 and 3 ml/min) were pumped in up flow mode using a peristaltic pump with initial ion concentrations of 50, 75 and 100 mg/L. The effluent samples were collected at regular intervals and analyzed for the residual U (VI) concentration. Column studies were terminated when the column reached exhaustion. In all experiments, the temperature was adjusted to 28°C. The column was run at a predetermined pH of 4.0 as established in my previous study [15]. Schematic of experimental set-up is shown in Figure 1.

Evaluation of break through curves in fixed-bed column

Thomas [19], Adams-Bohart [20] and Yoon-Nelson [21] models were used for kinetic studies.

Thomas model: Thomas model is one of the most widely used models in column performance studies. Thomas model is given in linear form by the following expression:

$$Ln\left(\frac{C_0}{C_t}-1\right)=\frac{K_{Th}q_eX}{Q}-K_{Th}C_0t$$

The parameters of Thomas model (k_{Th} and q_e) can be determined from a plot of $Ln\left(\frac{C_0}{C_t}-1\right)$ against time (t) at a given flow rate.

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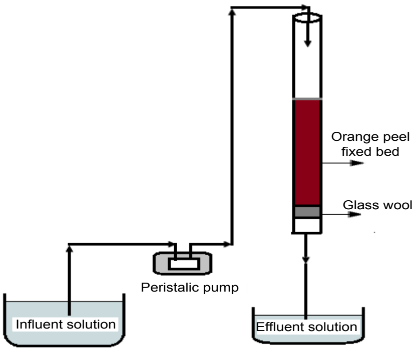

Figure 1: Schematic diagram of experimental system.

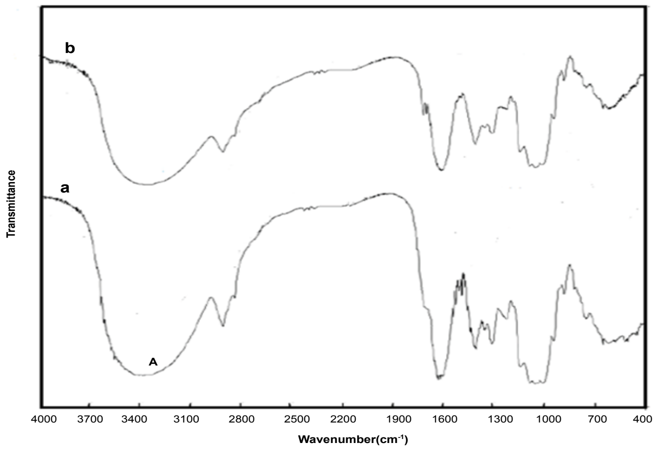

Figure 2: FTIR spectrum of OP and U (VI)-loaded OP.

Adams-bohart model: Adams-Bohart model is used for the description of the initial part of the breakthrough curve. The linear form of Adam-Bohart model is given by the following expression:

$$Ln\left(\frac{C_t}{C_0}\right) = k_{AB}C_0 t - k_{AB}N_0\frac{Z}{F}$$

The parameters k_{AB} and N_0 were determined from the intercept and slope of linear plot of *ln* (C_t/C_o) against time (t), respectively.

Yoon and nelson model: Yoon and Nelson model assumes that the decreasing rate of removal of adsorbate ion is proportional to the adsorbate breakthrough on the adsorbent material. The linear form is expressed by the following expression: $Ln\left(\frac{C_t}{C_0 - Ct}\right) = k_{YN}\tau - k_{YN}t$

The parameters k_{YN} and τ were determined from the intercept and slope of linear plot of *ln* [$C_t/(C_o-C_t)$] against time (t).

Results and Discussion

Characterization of adsorbent

The FTIR spectrum of OP before and after adsorption (Figure 2) displays a number of absorption peaks, indicating the complex nature of the adsorbent material. The absorption wavelengths of each peak and the corresponding functional groups are presented in Table 1.

From Table 1, after adsorption, shifts occurred in the wave numbers 3367, 2822, 1629 and 1425 cm^{-1} indicating an interaction of these functional groups with adsorbed U (VI). The appearance of wave number 1734 cm^{-1} in the U (VI) loaded spectra may indicate the interaction of this group with U (VI) ion.

Column adsorption

Effect of flow rate: The column adsorption study was carried out at different influent flow rates of 1, 2 and 3 L min^{-1} using initial U (VI) concentration of 50 mg L^{-1}, and bed height of 9 cm. Figure 3 shows that the faster breakthrough curve occurred at higher flow rate of 3 L min^{-1}. The faster breakthrough curve was attributed to faster movement of the adsorption zone along the bed and lower residence time of the influent in the column, thus reducing the contact time between U (VI) and the orange peels.

Effect of bed height: The effects of bed heights of 3, 6 and 9 cm were studied at influent concentration of 50 mg L^{-1} and 1 L min^{-1} flow rate. Figure 4 shows that the breakthrough time decreased with the

Wave number(cm^{-1})		Assignments
unloaded adsorbents	U(VI) loaded adsorbents	
3422	**3367**	**OH group**
2831	2822	C-H stretching
-	1734	C=O stretching
1655	1629	-COO- Carboxylic group
1549	-	-COO- Carboxylic group
1486	1425	C=O stretching
1019	1019	C-OH stretching

Table 1: FTIR data of OP and U (VI)-loaded OP.

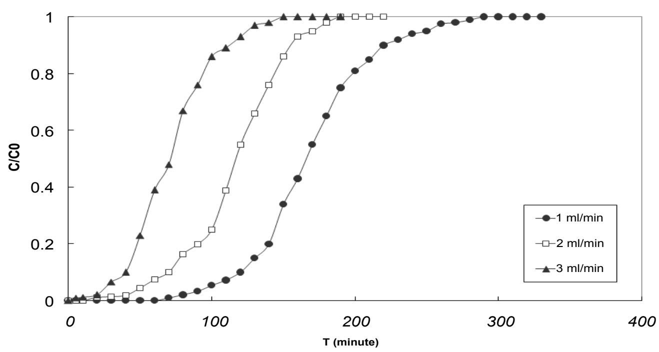

Figure 3: Breakthrough curves for U(VI) adsorption by OP at different flow rates.

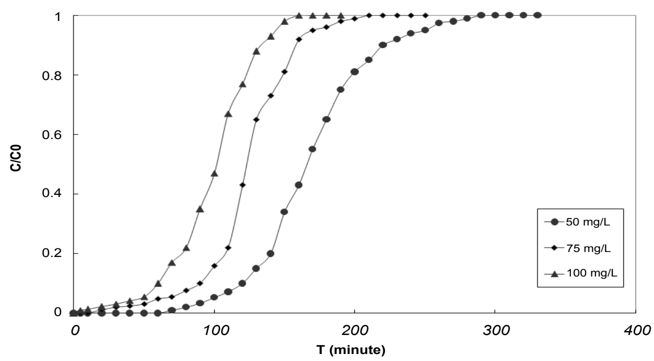

Figure 4: Breakthrough curves for U(VI) adsorption by OP at different initial U(VI) concentrations.

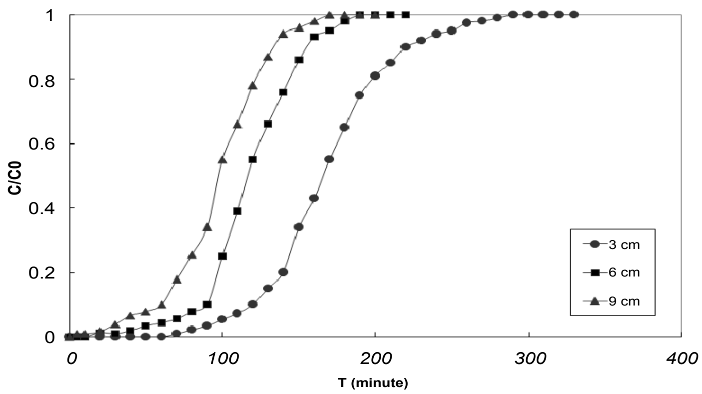

Figure 5: Breakthrough curves for U(VI) adsorption by OP at different bed heights.

Model type	Flow rate (ml/min)	Bed height (cm)	$C_{0\ (mg/L)}$	$q_{e,max\ (mg/g)}$	$k_{Th\ (mL/min.mg)}$	R^2
Thomas model	1	6	100	3.854	0. 410	1
	2	6	100	5.122	0. 511	0.99
	3	6	100	5.231	0. 630	0.99
	1	2	100	6.21	0. 352	0.99
	1	4	100	5.5	0. 381	0.98
	1	6	75	2.034	0. 650	0.97
	1	6	50	1.087	1.042	0.99
Yoon-Nelson model	Flow rate (ml/min)	Bed height (cm)	$C_{0\ (mg/L)}$	τ (min)	$k_{YN\ (1/min)}$	R^2
	1	6	100	173.4	0.041	1
	2	6	100	115.26	0.05	0.99
	3	6	100	74.55	0.063	0.99
	1	2	100	90.8	0.056	0.98
	1	4	100	118.41	0.053	0.99
	1	6	50	113.07	0.052	0.99
	1	6	75	121.46	0.049	0.97
Adams-Bohart model	Flow rate (ml/min)	Bed height (cm)	$C_{0\ (mg/L)}$	$N_{0\ (mg/L)}$	$k_{AB\ (L/min.mg)}$	R^2
	1	6	100	3.991	0. 200	0.84
	2	6	100	3.75	0.31	0.91
	3	6	100	2.88	0.781	0.85
	1	2	100	2.95	0.72	0.91
	1	4	100	3.11	0.426	0.93
	1	6	50	1.08	0.71	0.89
	1	6	75	2.022	0.4	0.91

Table 2: Thomas, Yoon-Nelson and Adams-Bohart model models parameters using linear regression analysis for U (VI) adsorption under various operating conditions.

bed height increasing. As the bed height increased, the U (VI) had more time to contact with orange peel that resulted in higher removal efficiency of U (VI) in the column.

Effect of initial concentration: The variations effect of initial U (VI) concentration from 50 to 100 mg/L at constant bed depth of 9 cm and flow rate of 1 mL/min in the breakthrough curves are shown in Figure 5. The higher the inlet concentration, the steeper is the slope of breakthrough curve. This is due to the increases of driving force and decreases in the adsorption zone length [22].

Evaluation of adsorption column models parameters

Models parameters were evaluated by linear regression analysis for U (VI) adsorption in different bed heights, initial concentrations, and flow rates. Linear regression results and values of R^2 of kinetic models are presented in Table 2. From Figures 6-8, it was observed that both Thomas and Yoon-Nelson models are appropriate models to describe fixed-bed system. But in the case of Adams-Bohart model, low correlation coefficient (R^2=0.925) is observed, which indicate that Adams-Bohart model is not as appropriate a predictor for the breakthrough curve.

Conclusions

From this study, adsorption rate strongly depends on flow rate, influent concentration, and bed height. The fixed-bed adsorption system was found to perform better with lower U (VI) inlet concentration, lower feed flow rate and higher bed height. Comparison between Thomas, Yoon-Nelson, and Adams-Bohart kinetic models with experimental data was performed, and model parameters were determined by linear regression analysis for U (VI) adsorption under various operating conditions. The column experimental data were fitted well with Thomas and Yoon-Nelson models, but the Adams-Bohart model predicted poor performance of fixed-bed column.

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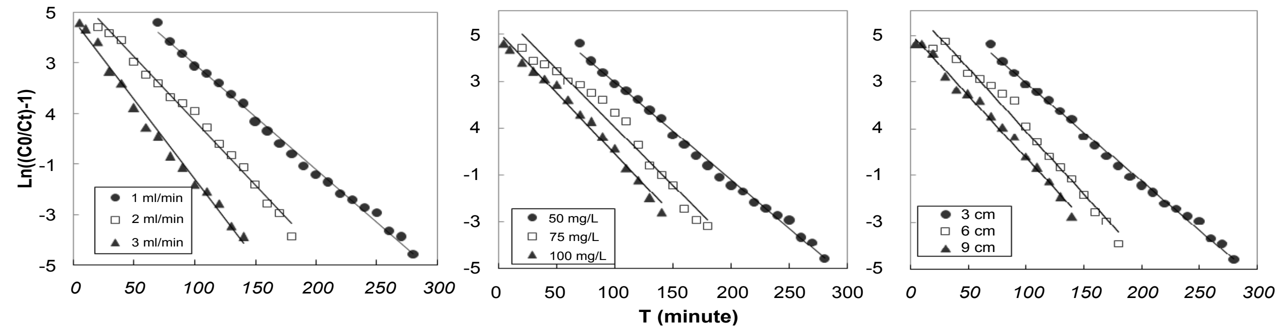

Figure 6: Linear plot of Thomas model with experimental data at different flow rates, initial U (VI) concentrations and bed heights.

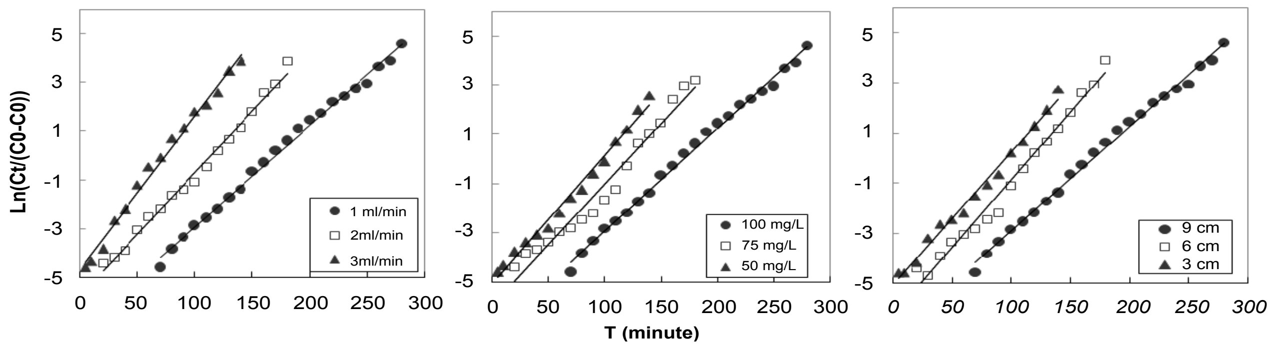

Figure 7: Linear plot of Yoon-Nelson model with experimental data at different flow rates, initial U (VI) concentrations and bed heights.

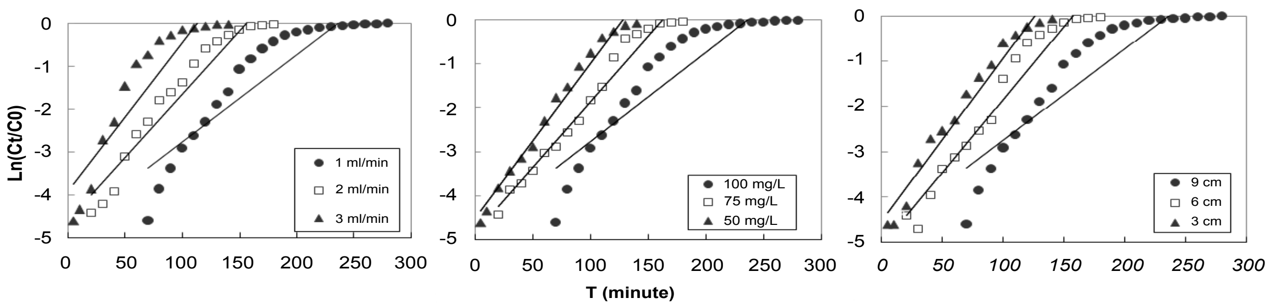

Figure 8: Linear plot of Adams-Bohart model with experimental data at different flow rates, initial U (VI) concentrations and bed heights.

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