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RESEARCH ARTICLE

Chemical Studies on Uranium Biosorption by using Non-Living Water Hyacinth Roots

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Abstract

Natural non-living water hyacinth roots (WHR) and the treated either with nitric acid (WHR-HNO_3) or sodium hydroxide (WHR-NaOH) are effectively adsorbed uranyl ions from its aqueous solution. At various pH values, adsorption equilibrium was reached within 10 min. and the adsorption isotherms were interpreted according to Langmuir and Freundlich models. Also, experimental adsorption data was analyzed by two kinetic models, namely; pseudo first order and pseudo second order. Among these models, pseudo second order model fits better with the experimental data of the treated non-living water hyacinth roots while pseudo first order model fits better with the data of the natural non-living water hyacinth roots. On the other hand, treatment of non-living water hyacinth roots by either nitric acid or sodium hydroxide was found to improve their interaction properties with uranium ions in adsorption process. The adsorption mechanism was investigated by Fourier transform infrared spectroscopy (FTIR), Scan electron microscope (SEM) and effect of pH. These analyses suggested that the adsorption mainly involved the ion exchange of UO_2^{2+} with cations and complex formation with functional groups on the surface of the roots. All the results showed that water hyacinth roots are an alternative low-cost adsorbent for the removal of uranium from aqueous media.

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1. INTRODUCTION

Mobilization of uranium in the environment due to the nuclear activities is of serious concern due to its toxicity in human and other forms of life [1]. The World Health Organization has determined that uranium is a human carcinogen and its concentration level in water should not exceed 50 mg/L. The U.S. Environmental Protection Agency has recommended a drinking water standard of 20 mg/L for ^{238}U [2]. Therefore, it is necessary to treat waste waters containing uranium in order to prevent radioactive contamination of the environment. Various techniques are employed for the removal of uranium ions from waste waters. Biosorption technologies in which living or dead biomass is used to accumulate uranium are methods that often replace conventional processes for remediation metal pollution in waste waters [3]. The major drawback of these microbial systems is the cost of growing a sufficient quantity of bacterial or algae biomass [4]. Alternatively, the removal of dissolved uranium by plant tissues has been studied using a variety of biomasses that represent byproducts from other commercial processes with little commercial value and thus representing good candidates for the development of inexpensive biosorption processes. Water hyacinth (Eichchornia crassipes) is a species of highly productive aquatic biomass in Egypt as well as in tropical and sub-tropical regions of the world, as might be expected from the fact that it grows in warm climates and has submerged roots and aerial leaves like reed swamp plants. It has been estimated that water hyacinth could be produced at rates up to about150 t/ (hm^2.y) if the plant were grown in a good climate [5]. The young plants are always predominant, and the water surface is always completely covered. Currently water hyacinth has no competitive uses and is considered to be an undesirable species on inland waterways which can render water ways,

reduce water flow and restrict commercial fishing. On the positive side, they have been shown to absorb and accumulate many of toxic elements [6-10]

Adsorption process of elements by water hyacinth occurs via the root system in the substrate water and leaves that are exposed to atmospheric dry-wet deposition. In either case the metals are distributed and bioaccumulated in various parts of the plant [11]. Bioaccumulation factors are higher in the roots than in the leaves [12,13]. Accordingly, the present study aimed to report the adsorption potential of dried water hyacinth roots either the natural or the protonated for the removal of uranium from waste water under various experimental conditions(e.g. uranium concentration, pH, temperature, and contact time).

2. EXPERIMENTAL

2.1. Chemicals

All chemicals and reagents of analytical grade, purchased from Aldrich, were used in the present work, double distilled water and filter paper (Whatman No. 40) was used in all experiments. The stock solutions containing uranium was prepared by dissolving its corresponding nitrate salt in double distilled water.

2.2. Roots preparation

Water hyacinth roots were collected in clean plastic bags from the River Nile at Zagazige City, Egypt. The roots were cleaned thoroughly with water to remove dirt and the unhealthy parts were discarded. The roots were then rinsed in double distilled water and dried at 60 °C, ground and passed through a 2 mm sieve. After the sieved sample material was homogenized to its chemical analysis.

2.3. Instruments

UV-Visible absorption spectrometric measurements were performed on a Schimadzu UV-VIS-160 double beam spectrophotometer using a 1- cm quartz cell. The pH values were measured using a pH-meter (Hanna- Instruments, 8519, Italy). Infrared (FTIR) analysis was recorded using a NICOLET FTIR MAGNA- 550 spectrophotometer with an instrumental resolution of 2 cm^{-1}. The scanning carried out by scanning electron microscope (SEM) using a PHILIPS-XL30.

2.4. Adsorption experiments

Batch adsorption experiments were carried out to investigate the quantitative uptake of uranium by water hyacinth roots. The analytical variables such as pH, time, temperature, and uranium initial concentration have been studied in detail for batch technique. Uranium was standardized and determined spectrophotometrically by Arsenazo III as reported elsewhere [14].

2.5. Adsorption isotherms

Batch adsorption studies of uranium were performed at room temperature (25 ± 2 °C) to obtain the equilibrium isotherms. For isotherm studies, a series of 50 ml test tubes were used. Each test tube was filled with 20 ml of uranium solution of varying concentrations (0.05 – 0.5 g / L) and a known amount of sorbent (1 g) was added into each test tube and agitated for various time periods. The uranium concentration retained in the sorbent phase (mg/g) was calculated by:

$$q_e = (C_i - C_e) V / m \quad (1)$$

where C_i and C_e are the initial and equilibrium concentrations of uranium (mg/L), respectively, V is the volume of the aqueous solutions (L), and m is the weight of the sorbent used (g). The distribution of uranium between the solid liquid interfaces at equilibrium has been studied by the Langmuir and Freundlich isotherm models. The Langmuir isotherm equation may be written as:

$$C_e / q_e = (1/ b\, q_o) + (C_e / q_o) \quad (2)$$

where q_e is the amount of solute sorbed per unit weight of adsorbent (mg/g), Ce is the equilibrium concentration of the solute in the bulk solution (mg/L), q_o is the monolayer adsorption capacity (mg/g) and b is the constant related to the free energy of adsorption.

The Freundlich equation may be written as:

$$\log q_e = \log K_f + 1/n \log C_e \quad (3)$$

where K_f is the constant indicative of the relative adsorption capacity of the adsorbent (mg/g) and 1/n is the constant indicative of the intensity of the adsorption.

2.6. Kinetic studies

Two kinetic models were used in this work to explain the adsorption process which are; the pseudo first-order and the pseudo second-order**.**

Pseudo first-order kinetics

The solute adsorption rate on the adsorbent is based on the adsorption capacity and follows the pseudo first-order equation:

$$\log (q_e - q_t) = \log q_e - [k_{ad}\, t / 2.303] \quad (4)$$

where; q_e and q_t are the amounts of the adsorbed metal ion (mg/g) at the equilibrium time and at any instant of time "t", respectively, and k_{ad} is the rate constant of the pseudo first-order adsorption operation (l/min). Plotting of log $(q_e - q_t)$ versus t gives a straight line for the pseudo first-order kinetics [15, 16].

Pseudo second-order kinetics

The applicability of the pseudo second-order kinetics has to be tested for q_e estimation with equation given by:

$$t / q_t = 1/h + (1/q_e)\, t \quad (5)$$

Where; $h = k_2 q_e^2$ that can be regarded as the initial sorption rate as $t \rightarrow 0$. Under such circumstances, the plot of t/q_t versus t should give a linear relationship which allows the computation of q_e and k_2.

3. RESULTS AND DISCUSSION

3.1. Chemical treatment of water hyacinth roots

Chemical treatments of water hyacinth roots take place by dilute solution of nitric acid or sodium hydroxide. The aim of treatment by dilute nitric acid is the oxidation of the sorbent surface to carboxylic and phenolic groups beside the protonation of the sorbent which eliminate metal ions like Na^+, K^+, Ca^{+2}, and Mg^{+2} etc, which bind to the acid functional groups of water hyacinth roots. At pH< 2, H^+ out compete other ions and strip off them from the sorbent ligands. Also the chemical treatment of water hyacinth roots using sodium hydroxide depend on 15 % of dry weight of water hyacinth roots is constituted by alginates. Alginates in water hyacinth roots are rich in carboxylic groups which responsible directly for the adsorption capacity of the roots. In the chemical alterations implied by the sodium hydroxide treatment, the sodium ions displaces protons from these binding sites making carboxylate and phenoxide groups making a binding site available. The results of the chemical treatment of the dried roots by nitric acid or sodium hydroxide as a function of weight loss are shown in figure 1. A slight loss in the roots was observed using 0.1 % and 1 % of nitric acid and sodium hydroxide, respectively. Difference in the adsorption capacity for treated and non treated roots was significant as shown in figure 2. The reason for these observations could be due to higher affinity of uranium towards binding sites as compared to the protons and the light metal ions. Therefore, for all the subsequent experiments roots in treated form by 0.1 % and 1 % of nitric acid and sodium hydroxide, respectively, was investigated.

3.2. Chemical and physical characterization of water hyacinth roots

Chemical composition of water hyacinth roots

From the analysis done with water hyacinth roots before and after chemical treatment as shown in tables 1 and 2, it has been found that all the percentage organic composition decreases with the chemical treatment.

Table 1: Organic composition of water hyacinth roots

Sorbents	Hemicellulose %	Cellulose %	Lignin %	Ash content %
WHR	16	28	15	17
WHR-HNO_3	13	27	14	09
WHR-NaOH	11	26	10	10

Table 2: Elemental analysis of water hyacinth roots

Element	WHR	WHR-NaOH	WHR-HNO_3
	Conc. %	Conc. %	Conc. %
C	60.0	54.0	55.0
O	30.0	33.0	42.0
Na	0.02	2.00	0.02
K	00.1	0.05	00.1
Ca	00.1	0.07	0.09
Mg	0.04	0.02	0.01
Si	12.0	14.0	12.0
Al	00.5	0.25	0.24
Fe	00.3	00.3	0.20
Cl	0.04	0.04	0.03

FTIR spectra

FTIR spectra of natural and treated water hyacinth roots showed some typical absorption bands as shown in table 3. The FTIR spectrum of water hyacinth roots is characterized by the following absorption bands:

Two sharp bands at 3428 cm^{-1} and 1083 cm^{-1}, mostly arises from OH and M-OH, respectively [17]. A band at 2369 cm^{-1}, indicating asymmetric stretching vibrations of a species M-O-CO. Here, the O-C-O group essentially has the structure of carboxylate group. Another spectral feature of examined samples is the appearance of a band at 1083 cm^{-1} ascribed to stretching vibrations of carboxylate surface groups. Two bands at 527 and 788 cm^{-1}, due to Si-O stretching bands which are complicated by variations in Al substitution for Si [18]. On the other hand, when H_2O is not fixed in a crystal lattice but is hydrogen-bonded to other water molecules, it results in a broad spectral feature centered near 3784 cm^{-1} due to O-H stretching vibrations and another near 1636 cm^{-1}, which is due to H-O-H bending vibrations. The effects of chemical treatment on the functional groups could be evaluated from the difference among the spectrums. The curves b and c in figure 1 shows the IR spectrum of treated water hyacinth roots either with nitric acid (WHR-HNO_3) or sodium hydroxide (WHR-NaOH) comparing these curves with curve a of natural roots there is no big difference except the shifting of some peaks to another values. It suggested certain changes have been happened to the surface of the roots. Furthermore, the IR peaks corresponding to C=O of the natural roots after uranium sorption as shown in curve d was shifted to lower values than that in the natural roots, indicating the bonding between this groups and uranium.

Table 3: FTIR peaks value of water hyacinth roots

n	WHR	WHR-HNO_3	WHR-NaOH	WHR-U
	Peak	Peak	Peak	Peak
1	3784	3720	3780	3424
2	3428	3600	3428	2930
3	2927	3440	2929	1618
4	2369	2900	2370	1545
5	1636	2330	1636	1462
6	1515	1600	1516	1234
7	1427	1520	1428	1032
8	1083	1400	1080	925.0
9	788.0	1020	788.0	677.0
10	527.0	800.0	530.0	611.0
11	462.0	500.0	463.0	555.0
12	-	420.0	-	533.0
13	-	-	-	463.0
14	-	-	-	441.0

Scanning Electron Microscope (SEM)

The above mentioned data of chemical analysis and FTIR spectra for the water hyacinth roots before and after chemical treatment are supported by electron micrographs presented in photos 1, 2 and 3. It has been noticed that the untreated roots is characterized by the cylindrical morphology and relatively heterogeneous structure which typify the roots. More over; the electron micrographs reveal that the chemical treatment of the roots leads to partial degradation of the water hyacinth roots cell structure and increasing on the pores structures. Also the photographing of the roots after adsorption of uranium in the electron microscope as shown in photos 4, 5 and 6, showed the good adsorption characteristic of the roots in a very clear manner. For instance, the photos showed that the adsorption of uranium takes place in interlayer spaces as well as on external surface of the roots where the bright parts indicate for uranium.

3.3. Effect of pH on the uranium adsorption

The effect of pH on the uranium adsorption onto WHR, WHR-HNO_3 and WHR-NaOH was investigated using 20 ml of uranium solution, 50 mg/L for a pH range of 1.0–8.0 at 25 $^{\circ}$C for 10 min. The results are shown in figure 4. The uranium adsorption capacity (q_e) by WHR, WHR-HNO_3 and WHR-NaOH was strongly depended on variations of the solution pH, the adsorption capacity increased with increasing pH up to pH 5.0 and then declines slowly with further increase in pH. In strong acidic solutions, more protons will be available to protonate phenolic and carboxylic groups, reducing the number of binding sites for the adsorption of UO_2^{2+}, therefore, the adsorption capacity of uranium is lower in acidic solutions (pH < 5.0). However, the increasing of pH value beyond 5.0; hydrolysis precipitation starts due to the formation of complexes in aqueous solution [19]. The hydrolysis of uranyl ions results in decline of adsorption capacity of uranium (VI) [20]. Also, it was noticed that the treated water hyacinth roots either with HNO_3 or NaOH have higher adsorption capacity than the untreated one due to it is believed that the adsorption occur through interactions with functional groups that are found in the lipids, proteins, and carboxylates that make up the cell wall of the roots[21]. The treatment of the roots by HNO_3 increases the phenolic and the carboxylic groups; also the treatments of the roots by NaOH, the degree of protonation of the functional groups were changed and hence, influence the sorption process.

3.4. Effect of contact time on the uranium adsorption

Under the conditions of 0.1 g sorbent amount, pH 5.0, temperature, 25 $^{\circ}$C and 50 mg/L uranium , the adsorption experiments were carried out for contact times ranging from 1 to 30 min. The results are shown in figure 5. It is clear that the adsorption capacity of uranium onto the roots increases with an increase of contact time and reaches adsorption equilibrium within 10 min. Rapid uranium adsorption which is similar to the adsorption of other metals[22] was contributed significantly to equilibrium uptake, which is interpreted to be the instantaneous adsorption stage or external surface adsorption. After 10 min, there are no notable effects on the uranium adsorption capacities so, 10 min was considered as the adsorption equilibrium time for the further work.

3.5. Effect of uranium initial concentration and adsorption isotherm

The effect of the initial uranium concentration on the adsorption capacity of the roots was studied using a range of initial uranium concentrations, 0.05 to 0.5 g/L. The results are shown in figure 6. It is clear that the adsorption capacity of uranium increased with increasing the initial uranium concentration in the aqueous solution, this is because more mass of uranium is put into the system with increasing the initial uranium concentration in the aqueous solution, but the same amount of adsorbent [23]. The adsorption data was subjected to analysis according to the Langmuir isotherm by plotting C_e/q_e vs C_e as shown in figure 7 which gives a straight line with intercept and slope values equal to $1/bq_o$ and $1/q_o$, respectively. Also Freundlich isotherm was applied from the adsorption data by plotting log q_e vs log C_e giving a straight line with slope and intercept values equal to 1/n and log K_f , respectively, as shown in figure 8. The isotherm data for the uranium adsorption was calculated from the slopes and intercepts of figures 7 and 8 and reported in table 4. The values of b for the treated roots are higher than that for the natural roots. Higher b values refer to stronger interaction between uranium and the active sites of the sorbent [24].

Table 4: Isotherm data of uranium adsorption

Sorbents	Isotherm data			
	Langmuir		Freundlich	
	q_o (mg/g)	b	1/n	log K_f
WHR	48	0.0025	0.69	0.4
WHR-HNO_3	67	0.0070	0.29	0.9
WHR-NaOH	87	0.0043	0.43	0.6

Comparison of uranium adsorption results obtained in this work to those in other relevant studies is summarized in table 5 which indicates that the treated water hyacinth roots have higher affinity and sorption capacity for uranium than the other sorbents reported in literatures.

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Table 5: Comparison of the uranium adsorption capacities on different studies

Sorbent	q_o (mg/g)	Reference
WHR	23	This study
WHR-HNO_3	45	This study
WHR-NaOH	50	This study
WHR	5.15	25
Olive cake	23	26
Aspergillus fumigatus beads	34	27
Activated carbon	28	28

3.6. Effect of temperature on the uranium adsorption

The effect of temperature on the adsorption of uranium was studied from 30 to 70 ^oC. The results are shown in figure 9. It can be observed that there is no change in the adsorption capacity of uranium, with changing temperature indicating that the process is spontaneous in nature

3.7. Adsorption kinetics

Kinetics of adsorption describing the solute uptake rate, which in turn governs the contact time, is one of the important characteristics defining efficiency of adsorption. The study of the equilibrium established in any liquid–solid system is important in determining distribution of the solute between the solid and liquid phases and determining feasibility and capacity of the sorbent for metal sorption [29]. In order to determine kinetic parameters and to explain the mechanism of the adsorption processes, lots of researchers have used first and pseudo-second-order rate expressions [30]. The uranium adsorption data was subjected to analysis according to the pseudo first order equation by plotting log ($q_e - q_t$) versus t as shown in figure 10. The adsorption rate constants (k_1) can be determined experimentally from the obtained straight line. Table 6 shows the rate constants and r^2 values of the kinetic models. The experimental data gives only a good fit for natural roots (r^2=0.96) indicating that the pseudo first order model is applicable for only the natural roots. Also pseudo second order model was applied from the adsorption data by plotting t/q_t versus t as shown in figure 11. The rate constant (k_2), and q_e are given from the figure and reported in table 6. Based on the obtained correlation coefficients (r^2), the experimental data only of the treated roots conformed better to the pseudo-second-order equation, evidencing chemical sorption involving valence forces through the sharing or exchange of electrons between adsorbent and adsorbate [31] as rate-limiting step of adsorption mechanism [32].

Table 6: Kinetic data of uranium adsorption

Sorbents	Kinetic data					
	Pseudo first order			Pseudo second order		
	k_1	log q_e	r^2	k_2	q_e	r^2
WHR	0.038	0.86	0.96	0.189	23.0	0.79
WHR-HNO_3	00.03	0.98	0.82	0.037	45.0	99.0
WHR-NaOH	0.022	0.98	0.86	0.029	50.0	0.98

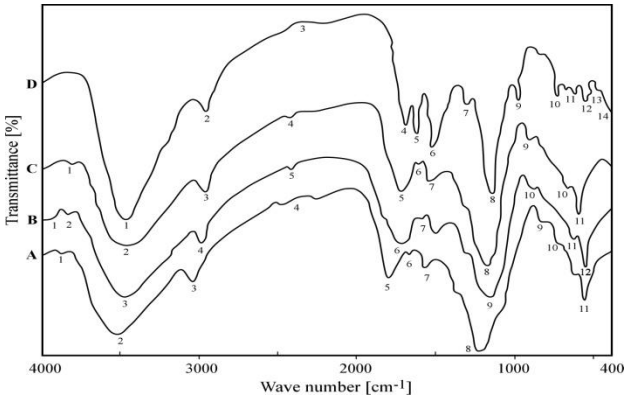

Figure 1. FTIR of WHR, WHR-HNO_3, WHR-NaOH and WHR-Uranium
(A- WHR, B- WHR-HNO_3, C- WHR-NaOH and D- WHR-Uranium)

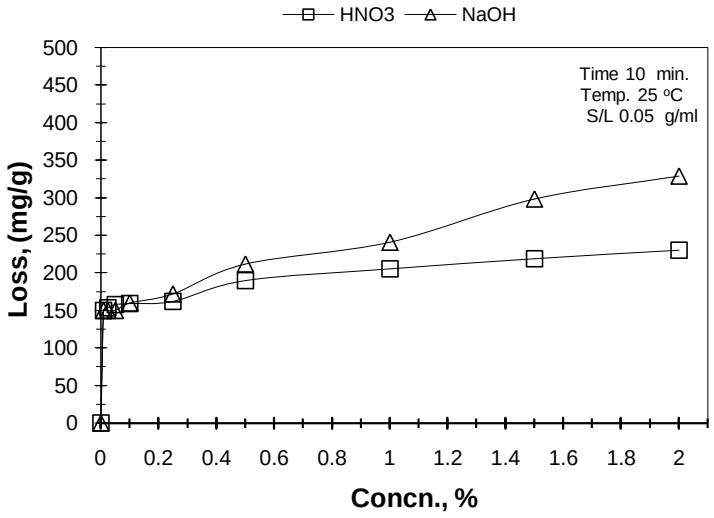

Figure 2. Effect of HNO_3 or NaOH concentration on the weight loss of WHR

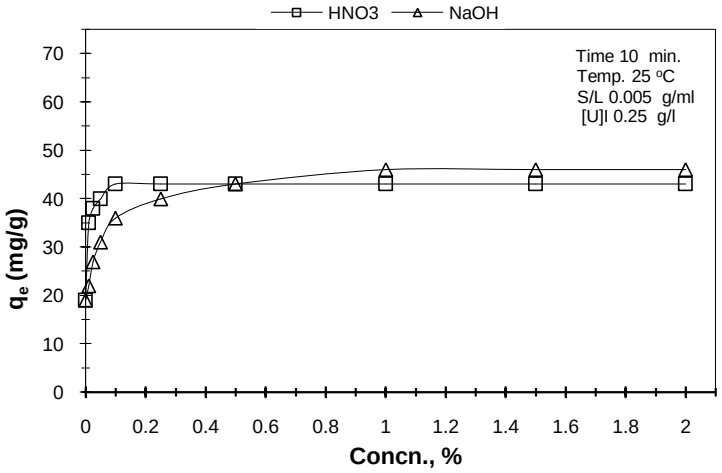

Figure 3. Effect of HNO_3 or NaOH concentration on the uranium adsorption

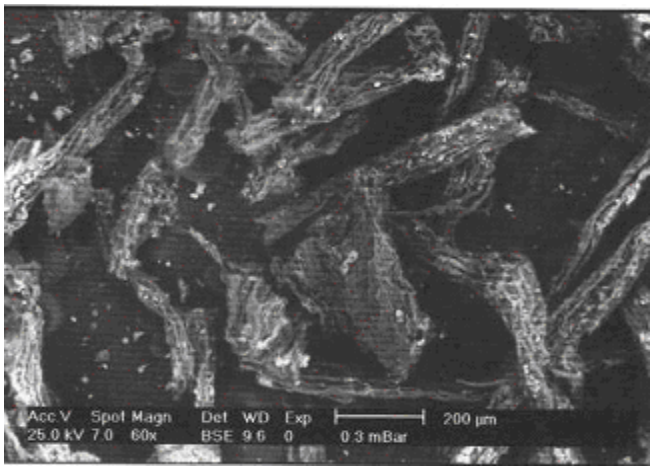

Photo 1. SEM of natural water hyacinth roots (WHR)

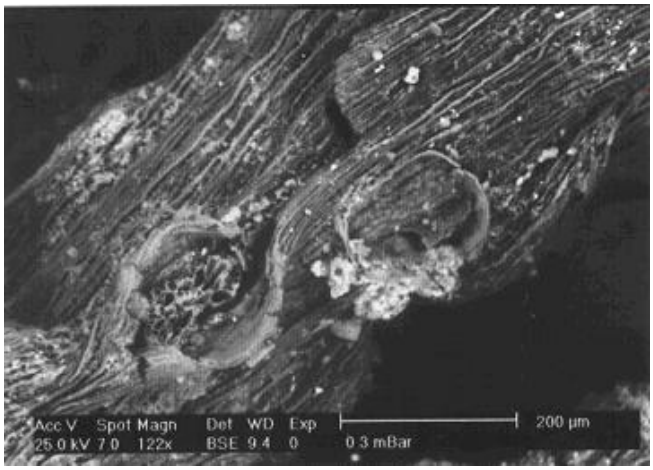

Photo 2. SEM of water hyacinth roots after nitric treatment (WHR-HNO_3)

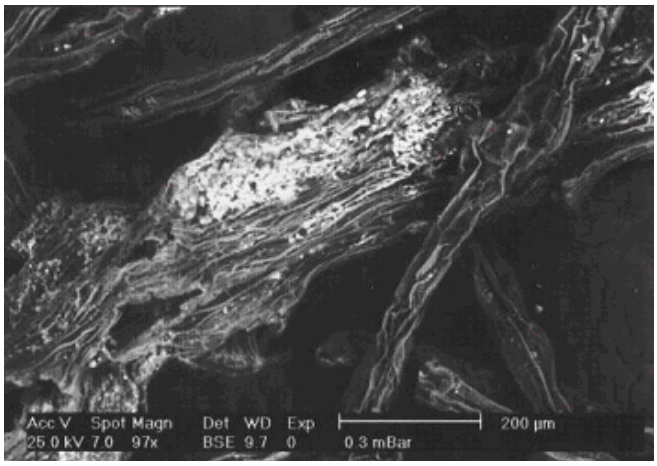

Photo 3. SEM of water hyacinth roots after sodium hydroxide treatment (WHR-NaOH)

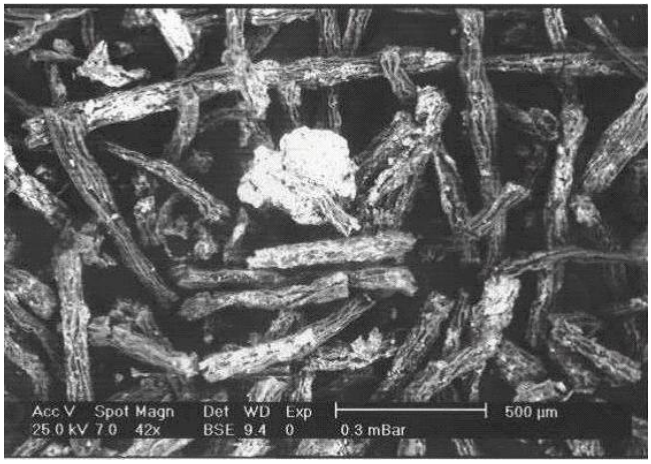

Photo 4. SEM of WHR after uranium adsorption

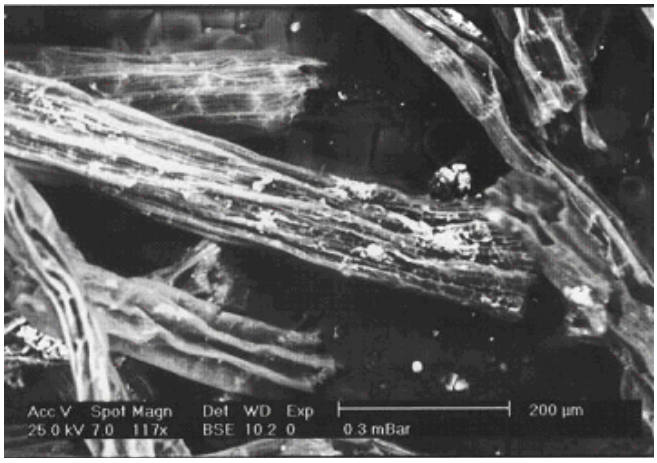

Photo 5. SEM of WHR-HNO_3 after uranium adsorption

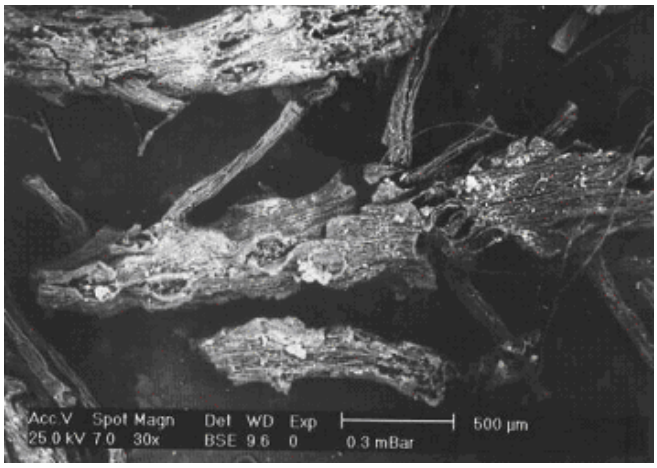

Photo 6. SEM of WHR-NaOH after uranium adsorption

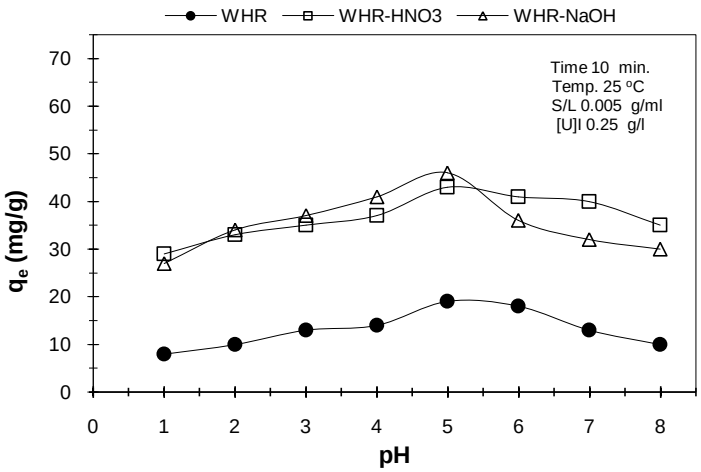

Figure 4. Effect of pH on the uranium adsorption

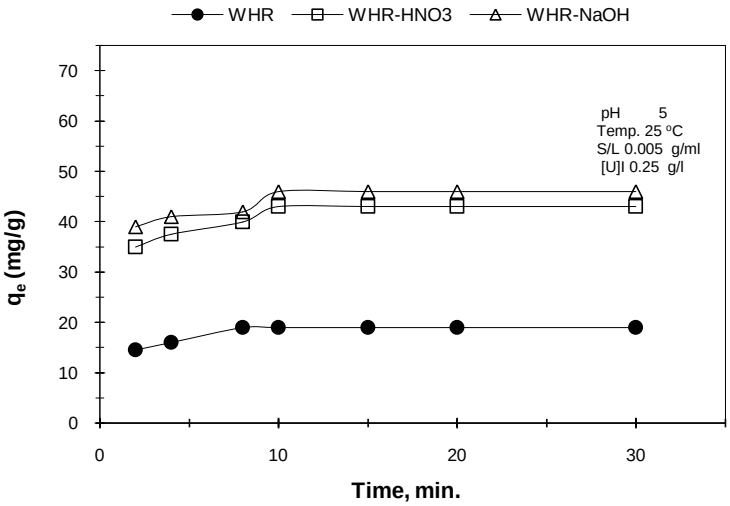

Figure 5. Effect of contact time on the uranium adsorption

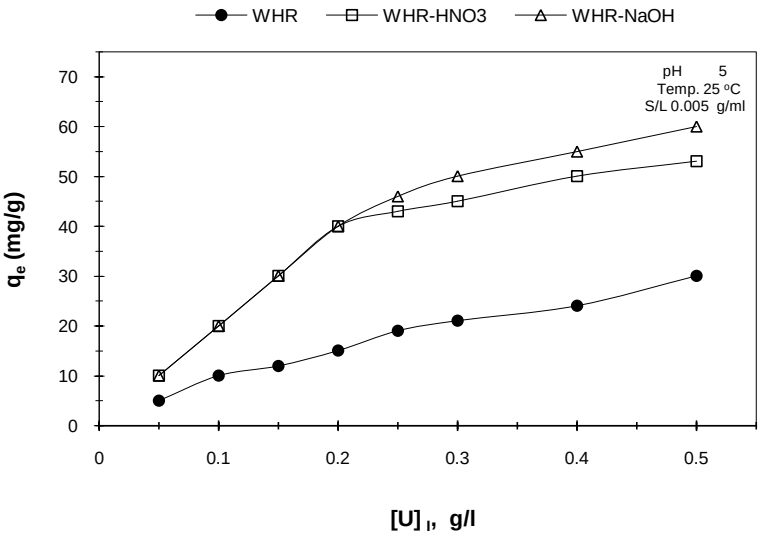

Figure 6. Effect of uranium initial concentration on the adsorption

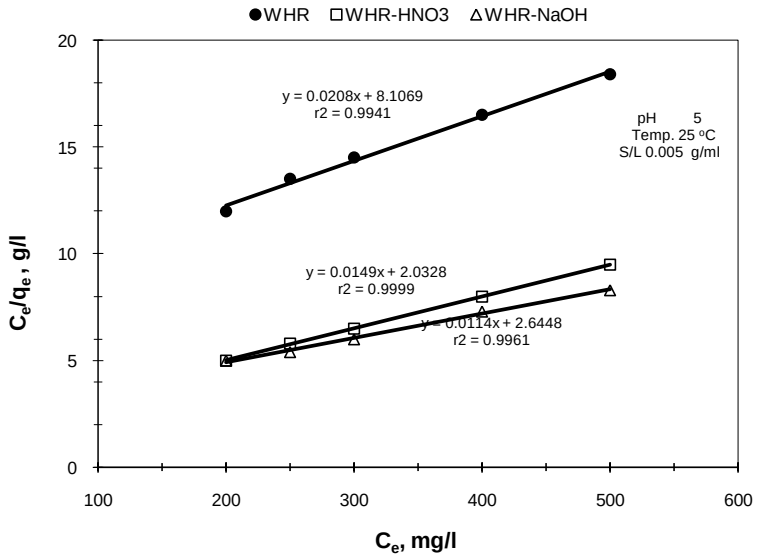

Figure 7 . Langmuir isotherm of the uranium adsorption

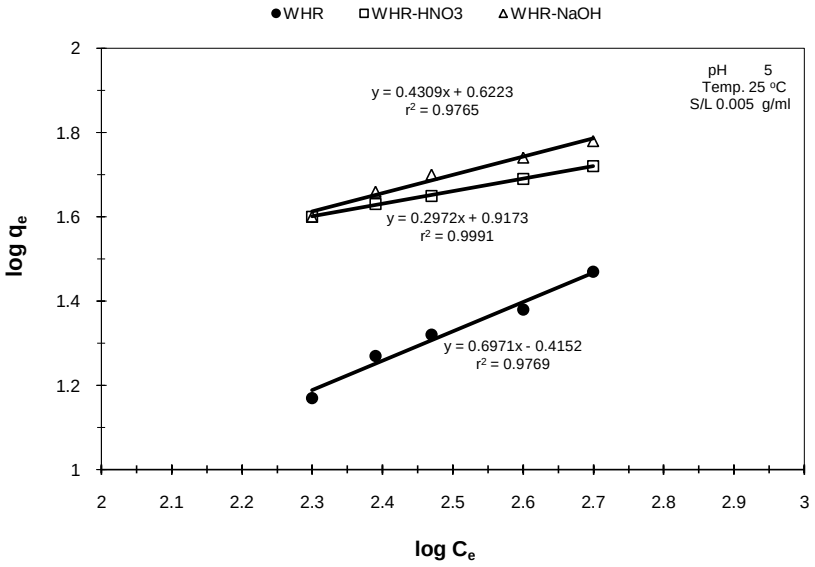

Figure 8 . Freundlich isotherm of the uranium adsorption

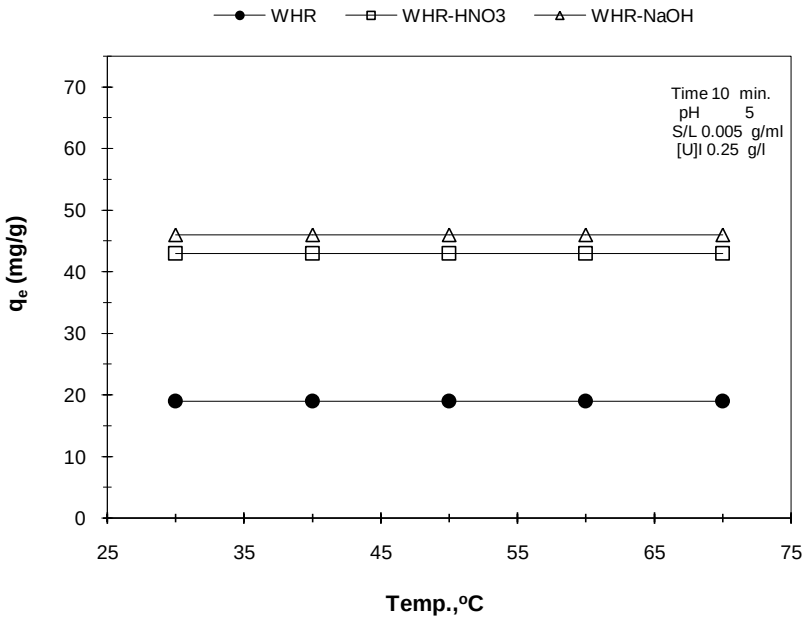

Figure 9. Effect of temperature on the uranium adsorption

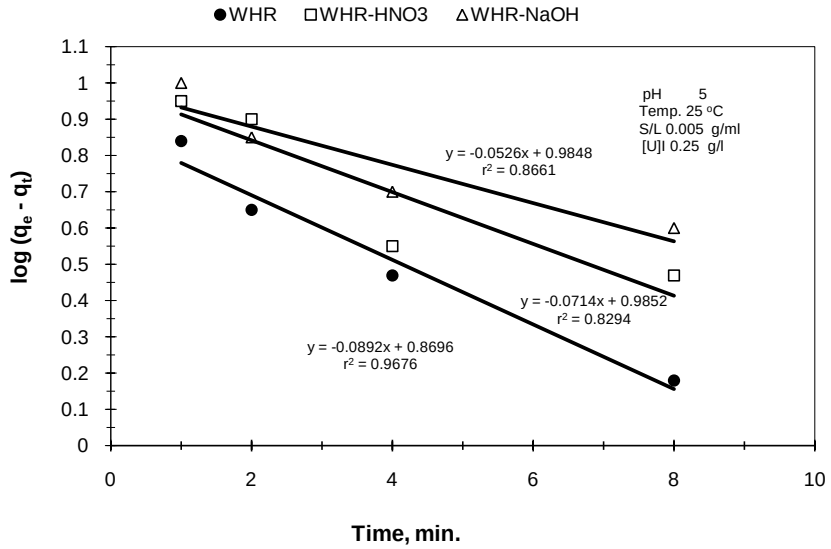

Figure 10. Pseudo first order model for the uranium adsorption

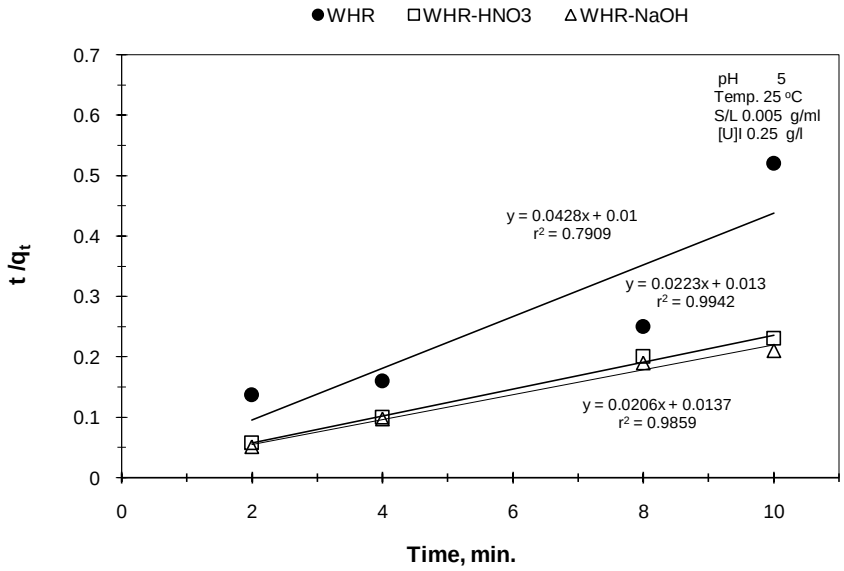

Figure 11. Pseudo second order model for the uranium adsorption

4. CONCLUSIONS

The results obtained from this study lead to the following conclusions: (1) the optimum pH for uranium adsorption by water hyacinth roots is pH 5, (2) the experimental data can fitted satisfying both Langmuir and Freundlich model, (3) the experimental data of WHR can fitted satisfying first order model, (4) the experimental data of both WHR-HNO_3 and WHR-NaOH can fitted satisfying pseudo second order model , (5) the treatment of the water hyacinth roots with either nitric acid or sodium hydroxide increase the adsorption capacity and (6) the adsorption capacity is found to be 23, 45 and 50 mg g^{-1} for WHR, WHR-HNO_3 and WHR-NaOH, respectively

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