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Influence of pH on the Adsorption of Uranium Ions by Oxidized Activated Carbon and Chitosan

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ABSTRACT

The adsorption characteristics of uranyl ions on surface-oxidized carbon were compared with those of powdered chitosan over a wide pH range. In particular, an extensive analysis was made on solution pH variation during the adsorption process or after adsorption equilibrium. Uranium adsorption on the two adsorbents was revealed to be strongly dependent on the initial pH of the solution. A quantitative comparison of the adsorption capacities of the two adsorbents was made, based on the isotherm data obtained at initial pH 3, 4, and 5. In order to analyze the adsorption kinetics incorporated with pH effects, batch experiments at various initial pH values were carried out, and solution pH profiles with the adsorption time were also evaluated. The breakthrough behavior in a column packed with oxidized carbon was also characterized with respect to the variation of effluent pH. Based on these experimental results, the practical applicability of oxidized carbon for uranium removal from acidic radioactive liquid waste was suggested.

INTRODUCTION

Liquid effluents generated from the nuclear fuel cycle contain various kinds of radiotoxic nuclides. They are typically fission products, uranium and transuranic elements (1–3). In particular, because of its long half-life and high radiological toxicity, uranium is considered to be a serious long-term envi-

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ronmental hazard. A very low concentration of uranium is allowed in the effluents from nuclear facilities (4, 5). Therefore, uranium should be removed to below its tolerance limit before the effluents are released into the environment.

Commonly available methods for the removal of uranium from an aqueous solution include sorption using synthetic polymer resin, activated carbon, or biosorbent. Many researchers have extensively studied the recovery of uranium from seawater by using metal oxides or polymer resins (6, 7). Activated carbon as an inorganic adsorbent has been commonly used to remove the radionuclides from liquid effluents because of its high chemical, radiological and thermal stability (8). Numerous experimental studies have also been conducted on the removal of residual uranium in radioactive liquid wastes by using activated carbon (9–11). In acid solution, however, activated carbon can adsorb only a small quantity of uranium. Many attempts have been made to enhance the adsorption capacity of activated carbon. Some recent R&D results showed that its oxidative treatment with nitric acid or hydrogen peroxide solution effectively increases the adsorption capacity for polar molecules and uranium (12, 13). In addition, chitosan, a natural biopolymer derived from chitin which is a principal component of the shells of crustacean organisms, has a high affinity for transition metal cations due to complexation of the metal with the amine (NH_2) of the chitosan molecule (14). Recently, many researchers have carried out experimental studies on the adsorption characteristics of chitosan powder or modified chitosan beads (15–17). However, the adsorption behavior of uranium on these adsorbents has not been characterized in detail.

It has been reported that the fractional distribution of uranyl ion species depends on the solution pH and the total uranium concentration (9–11, 17). From this point of view, pH should be considered as the important parameter that affects uranium adsorption from an aqueous solution. Therefore, in this study, experimental work was performed to characterize the pH-dependent adsorption behaviors of uranium in aqueous solution on chitosan powder and oxidized carbon treated with nitric acid. This experiment includes equilibrium and kinetic studies as well as fixed-bed adsorption in terms of the initial solution pH. The analysis of pH variation during the adsorption process was also investigated.

EXPERIMENTAL

Materials

Coconut-based activated carbon, provided by Han-Il Green Tech. Co., and chitosan powder (average MW = 100,000, degree of deacetylation = 87%), supplied by Acros Organic Co., was used as the adsorbent. The granules of activated carbon were ground, sieved to give a particle size of 16–30 mesh. The

sieved particles were used after being washed free of dust with distilled water and dried at 110°C in a vacuum drying oven. Pre-treated activated carbons were oxidized by heating in 7 N nitric acid solution at a temperature of 80–90°C for 10 hours in a volumetric flask (12, 13). They were then exhaustively washed with distilled water and dried in a vacuum oven at 100°C. On the other hand, chitosan powder was sieved to a size of 70 mesh (0.27 mm). The physical properties of activated carbon and oxidized carbon were measured by BET-N_2 analysis. Only a small amount of the surface area and pore volume were decreased by the wet oxidation of activated carbon, as shown in Table 1 and Fig. 1. Uranium solutions were prepared by dissolving the depleted GR-graded uranyl nitrate hexahydrate, $UO_2(NO_3)_2 \cdot 6H_2O$, in distilled water. The uranium-235 content of this salt was determined to be 0.33%. The ionic strength of the uranium solution was not controlled in this study.

Analysis

A spectrophotometric technique was used to determine the uranium concentration in solution (18). Arsenazo III was used as the color-developing agent. The chelate complexes formed by U^{+6} with Arsenazo III have molecular absorptivities, ε, of 1.27×10^5 L/mol·cm at $\lambda = 656$ nm and $\varepsilon = 1.15 \times 10^5$ L/mol·cm at $\lambda = 655$ nm. An UV/Vis spectrophotometer (Spectronic 1201, Milton Roy) was used for the quantitative determination of the uranium concentration. The solution pH was measured by a pH-meter (Model 920A, Orion).

Procedures

The adsorption experiments of uranium were conducted at various initial concentrations of uranium, ranged from 10 to 500 mg/L in the concentration of U^{6+}. Adsorption equilibrium data were obtained at initial pH values of 3,

TABLE 1
Physical Properties of Activated Carbon and Oxidized Carbon

Physical properties	Units	Activated carbon	Oxidized carbon
Particle density	kg/m^3	880	860
Total pore volume	m^3/kg	7.37×10^{-4}	7.07×10^{-4}
Micropore volume	m^3/kg	6.41×10^{-4}	5.88×10^{-4}
Average pore radius	Å	19.295	20.75
BET surface area	m^2/kg	1.53×10^6	1.36×10^6
Micropore area	m^2/kg	1.46×10^6	1.28×10^6
Average particle radius	m	4.25×10^{-4}	4.25×10^{-4}

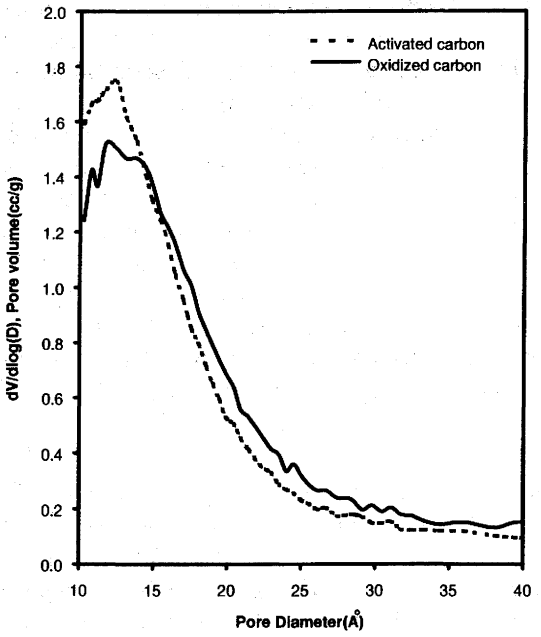


FIG. 1 Pore size distribution for activated carbon and oxidized carbon.

4, and 5. The pH of the uranium solution was adjusted with GR-graded HCl and NaOH. The solution pH, however, was not controlled during the adsorption process. Precipitation of uranium ions due to hydrolysis was never observed below a uranium concentration of 500 mg/L (pH about 7). The uranium solution was poured into a glass bottle containing the weighed adsorbent. The solid-to-solution ratio (g/L) in the isotherm experiment was 1.25 for chitosan and 2.5 for activated carbon. After equilibrium, the solution was filtered by using a 0.45-μm membrane filter.

Kinetic data of uranium adsorption were obtained in a Carberry-type batch adsorber with four baffles (19). The rotor speed was approximately 500 rpm, so the external mass transfer resistance would be nearly negligible. Samples were taken periodically with a micromembrane pipet, and the uranium concentration of the sample solution was measured. The solid-to-solution ratio was 2.5 for all runs. The variations of solution pH during adsorption were also obtained.

The fixed-bed runs were carried out using a glass column of 0.01 m inside diameter and 0.3 m in length. A micrometering pump was used to maintain a constant down flow of 2 mL/min. Effluent from the column was collected pe-

riodically with a fraction collector. Since powder-type chitosan was used in this experiment as an adsorbent, the data from the fixed-bed runs were limited to oxidized activated carbon.

All the experiments were conducted at 30°C except where otherwise specified.

RESULTS AND DISCUSSION

The Effect of pH

Numerous works (9–18) have shown that the adsorption of metal cations onto adsorbents is strongly dependent on the solution pH. In order to make a thorough investigation of the influence of pH, preliminary experiments of uranium adsorption on activated carbon, oxidized carbon, and chitosan were carried out in an initial pH range of 2–10. The solid-to-solution ratio was fixed to 1.25 for three adsorbents. Figures 2(a) and 2(b) show the removal efficiency of uranium at 50 and 300 mg/L, respectively. The corresponding final pH is also represented in Fig. 3. It was observed that the final pH at equilibrium was considerably influenced by the adsorbents used. At low pH ranges (less than pH 3), significant variations of the solution pH were not observed. On the other hand, in the 3–8 pH range the final pH increased when chitosan and activated carbon were used as the adsorbents. In contrast, for oxidized carbon the final pH decreased when compared with the initial pH. These pH variations are closely related to the surface characteristics of the adsorbents. The pH_{pzc} (point of zero charge) of chitosan prepared from crab shell is about 6.3 (14), and around 8 for activated carbon (20). The pH after the addition of an adsorbent into a solution, in principle, tends to vary toward the direction of the sorbent's pzc (21). It was also reported (12) that coconut-based activated carbon oxidized with 6.6 N nitric acid shows a large increase in its total acidity and surface functional groups when compared with untreated activated carbon. These surface characteristics lead to a decrease of the solution pH. The removal efficiency of chitosan at an initial pH of 3 or below was similar to that of oxidized carbon. Oxidized carbon showed good removal efficiency for uranium in the high pH range. In particular, the adsorption capacity of oxidized carbon was not decreased even at higher pH values, at which that of chitosan was greatly decreased.

The pH effect on uranium adsorption can be explained by the solution chemistry of uranium as well as by the surface characteristics of the adsorbents. As the pH of a uranium solution increases, the uranyl ions are easily hydrolyzed, and these hydrolysis products are also polymerized (10, 18, 22). Figures 4(a) and 4(b) show four types of hydrolyzed uranyl species, UO_2^{2+}, $UO_2(OH)^+$, $(UO_2)_2(OH)_2^{2+}$ dimer, and $(UO_2)_3(OH)_5^+$ trimer, which are predicted by the MINTEQ2 code (23) in pH ranging from 3 to 6. It has been ex-

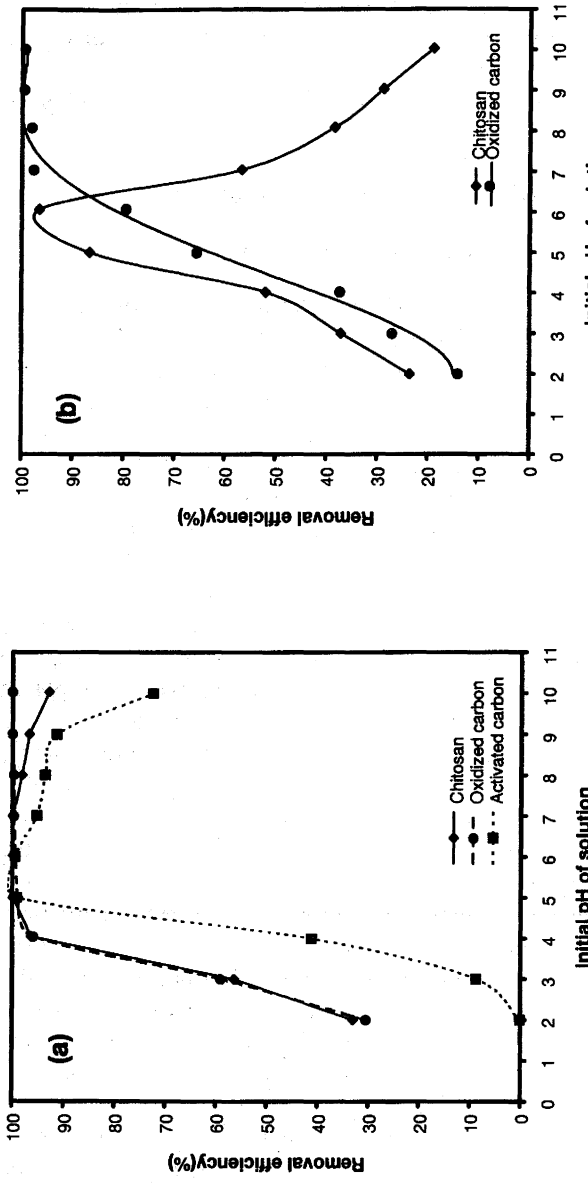


FIG. 2 Variations in removal efficiency of uranium with the initial pH of solution at (a) 50 mg/L and (b) 300 mg/L of uranium concentrations, respectively.

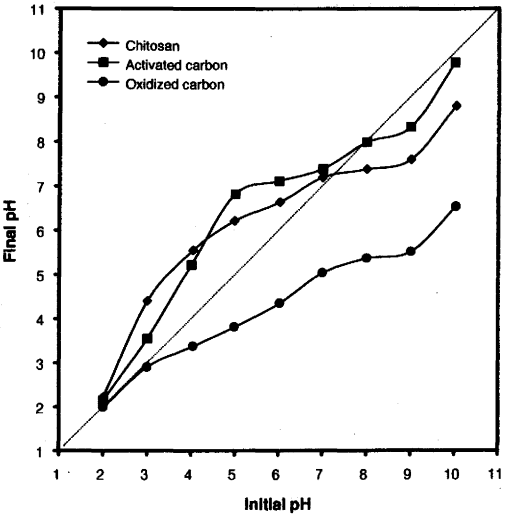


FIG. 3 Comparison of initial and final pH of the solution containing each adsorbent at adsorption equilibrium.

perimentally shown that an anionic hydrolyzed species such as $(UO_2)_3(OH)_7$ exists at a relatively high pH (22). However, it is easy to calculate the fraction of these species if their accurate equilibrium constants for the hydrolysis reaction are known. It has been reported (15–17) that only cation metals are adsorbed by their complex reaction with the amine group (NH_2) on chitosan. Therefore, it is thought that at a high pH the decrease in the removal efficiency of uranium on chitosan is due to an increase of the fraction of negative hydrolysis products in solution. On the other hand, it was observed that the adsorption capacity of oxidized carbon does not decrease even at higher initial pH ranges. This phenomenon can be explained by the following two facts: 1) only cation forms of uranyl ions exist over wide pH ranges due to the high surface acidity of oxidized carbon, and 2) larger amounts of surface oxides, such as the carboxyl and phenolic hydroxyl groups, lead to an increase of the adsorption capacity of uranium.

Figures 5(a) and 5(b) show the influence of the amount of adsorbent on the removal efficiency of uranium at initial solution pH values of 3 and 5, respectively. Overall, chitosan has a higher removal efficiency than oxidized carbon, except at pH 3 and at a uranium concentration of 100 mg/L.

Equilibrium Adsorption Isotherm

The radioactive liquid effluent generated from the nuclear fuel cycle is generally considered to be acidic (2, 3). Based on this fact, more detailed experi-

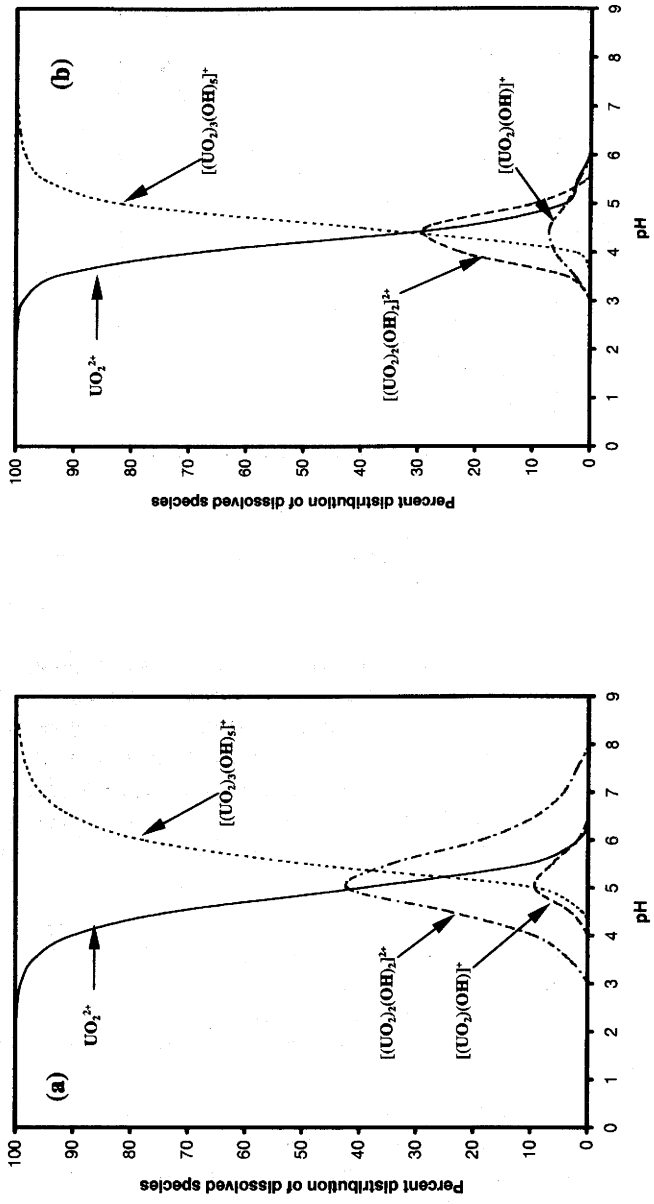
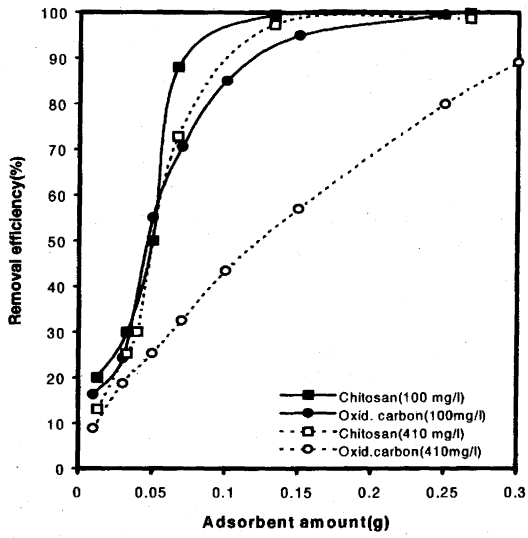
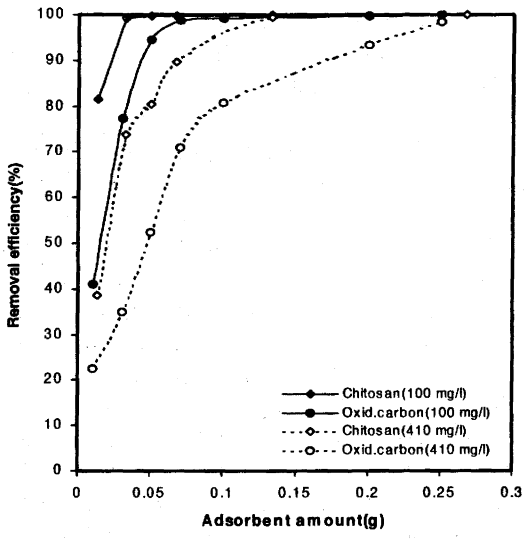


FIG. 4 Distribution of hydrolyzed uranyl ions vs pH in pure water at 30°C and uranium concentration of (a) 10^{-5} mol/L and (b) 10^{-3} mol/L.



(a) pH=3



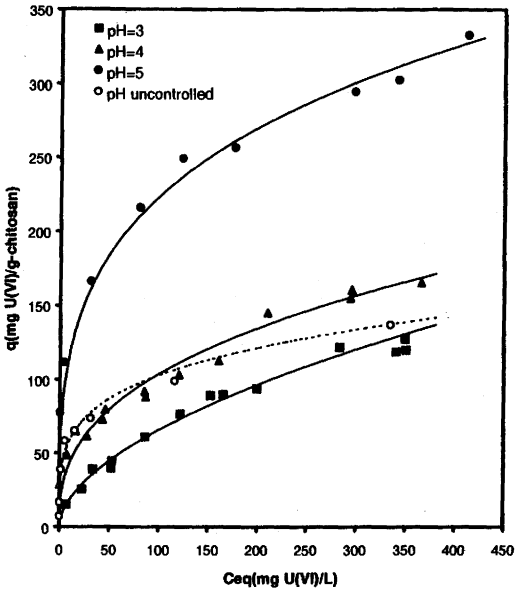
(b) pH=5

FIG. 5 Influence of adsorbent amount on the adsorption of uranium at pH 3 and 5 and the uranium concentration of 100 and 410 mg/L, respectively.

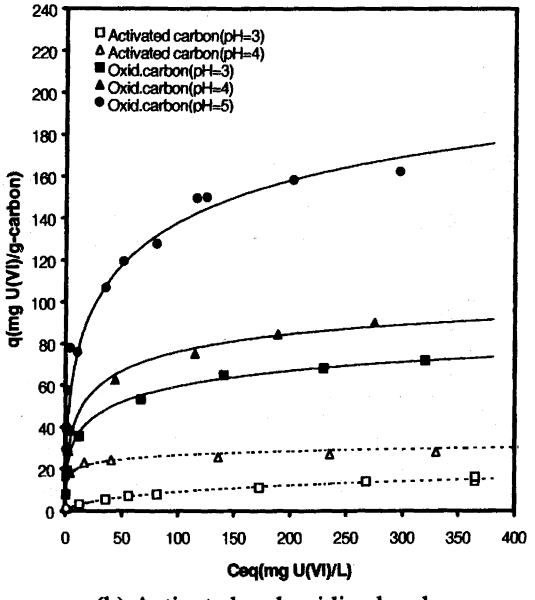
ments for uranium adsorption were carried out within an initial pH range of 3–5. The adsorption amount of uranium per unit mass of adsorbent was obtained by the following relationship:

$$q = \frac{(C_0 - C_{eq})V}{m} = \frac{F_0 C_0}{m/V} \quad (1)$$

The adsorption isotherms of uranium on chitosan, activated carbon, and oxidized carbon are shown in Figs. 6(a) and 6(b). These isotherms, which are correlated by the Sips isotherm equation, were represented by the total concentration of uranium. The effect of the pH of the initial solution on the equilibrium adsorption amount of uranium was clearly found in all three adsorbents. At an initial pH 5, the maximum adsorption amount on chitosan was about 350 mg/g, which is almost twice as much as that on oxidized carbon. In addition, the adsorption amount on oxidized carbon was much higher than that on activated carbon. Interestingly, when the uranium concentration and initial pH were lower, there were no discernible differences in equilibrium adsorption amounts on either chitosan or oxidized carbon, specifically in the concentration range of $C_{eq} < 50$ mg/L at pH 3. Considering that the uranium adsorption capacity of chitosan is much higher than that of oxidized carbon at an initial pH 5, it can be interpreted that at low pH the competitive adsorption between hydronium and uranium ions on chitosan increased in comparison with oxidized carbon. It is thought that this inference can also be explained by the solution pH variations after adsorption equilibrium. Figures 7(a) and 7(b) represent the variations of final pH, namely equilibrium pH, on chitosan and oxidized carbon. At initial pH 5, the equilibrium pH on chitosan was slightly increased, except at low equilibrium concentration ranges. No significant difference in equilibrium pH at $C_{eq} > 50$ mg/L was observed even though the initial pH was fixed at 3 and 4. The adsorption amount at pH 4 was, however, slightly higher than that at pH 3, as shown in Fig. 6(a). Eiden et al. (24) observed a pH increase during Cr(III) adsorption on chitosan. Also, Rorrer et al. (15) identified the competitive adsorption between cadmium and the hydronium ions on chitosan from pH measurements during the adsorption process. These behaviors are clearly different from those in the uranium-oxidized carbon adsorption system. The equilibrium pH at an initial pH 3 leveled off at 2.98–2.8 over a whole equilibrium concentration ranges, and leveled off at 3.2 and 3.8 from initial pH 4 and 5, respectively. Abbasi et al. (13) investigated the adsorption of uranium in near-neutral aqueous solutions on oxidized carbon, and proposed an ion-exchange mechanism for uranium sorption. Carboxyl groups with a pK value of 4–5 or phenolic group are largely responsible for the ion exchange of divalent uranyl ions or other hydrolyzed ionic species from solution. It is thought that the decrease in pH reflects uranium removal by an ion-exchange mechanism. That is, hydronium ions on ion-exchange

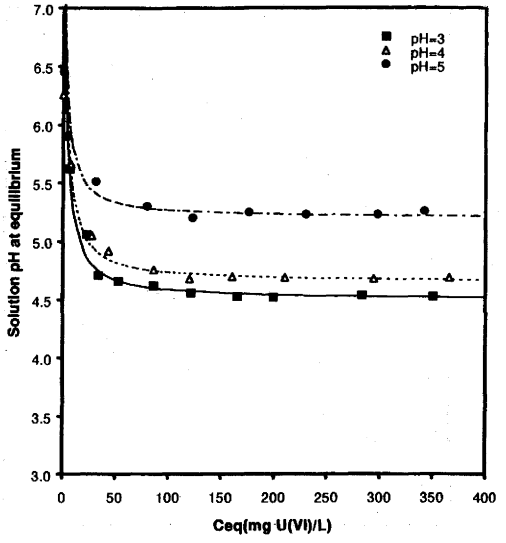


(a) Chitosan

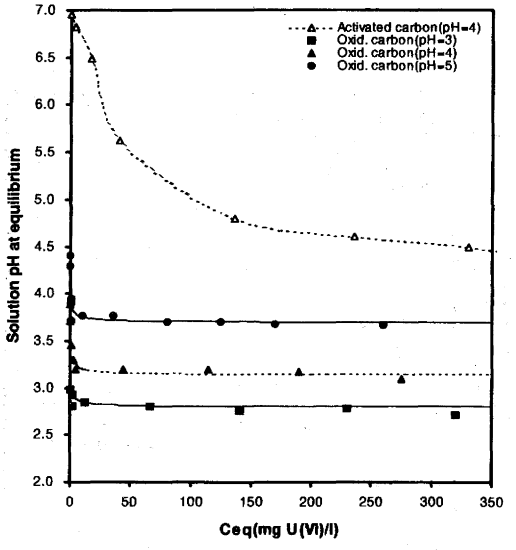


(b) Activated and oxidized carbons

FIG. 6 Adsorption isotherms of uranium on chitosan and carbons at 30°C.



(a) Chitosan



(b) Activated and oxidized carbons

FIG. 7 Variation of solution pH at equilibrium on chitosan and carbons at 30°C.

sites, mainly surface functional groups, are displaced as the uranium ions adsorb onto this site, leading to a pH decrease with adsorption time. Actually, oxidized carbon has a large amount of surface functional groups. Among these groups, the weakly or strongly acidic carboxyl group at relatively low pH is available in the adsorption of uranium because the pK values of the phenolic group are about 10–12 (25).

On the other hand, when the initial pH of a uranium solution was not adjusted with HCl or NaOH, the stock solution pH was 5.3 to 3.5 with an increase of uranium concentration due to chemical forms of this reagent. As predicted through the above results, the isotherm curves of the adsorption experiment performed at uncontrolled initial pH values showed a typical profile at a fixed initial pH 5 in low concentration ranges and at pH 3 in high concentration ranges.

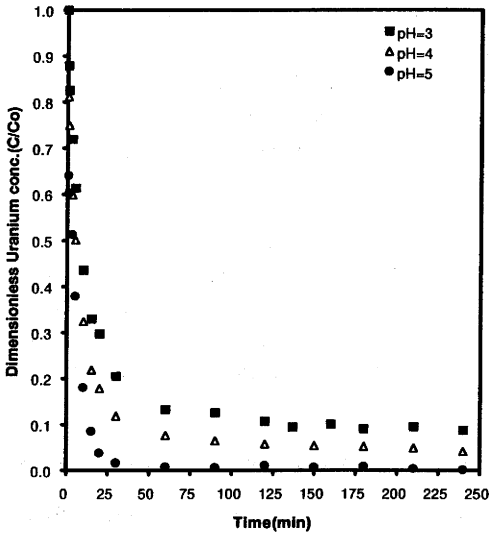
Adsorption Kinetics in a Batch Reactor

In order to obtain information on the mass transfer and sorption kinetics incorporated with the pH effect, batch experiments using chitosan and oxidized carbon were carried out in the range of initial pH 3–5. The uptake curves of uranium ions at concentrations of 50 and 200 mg/L are presented in Figs. 8 and 9, respectively. The pH effect on the adsorption rate of two adsorbents was not clearly observed. However, the adsorption rate on chitosan was clearly much greater than that on oxidized carbon. This may indicate that the adsorption rate of uranium on oxidized carbon was diffusion-controlled through the micro- and mesopores (26). In general, it is thought that the majority of the functional groups on oxidized carbon are located near the edges of hexagonal carbon rings (27). The radius of a simple uranyl ion is 3.84 Å (28). At high solution pH, the polymerization of uranyl hydrolysis products results in bigger hydrolyzed uranyl ions in the solution (10, 18, 22). Their complexes, which are formed with surface oxides at the pore entrances of oxidized carbon, may affect the adsorption rate of uranyl ions through pore diffusion.

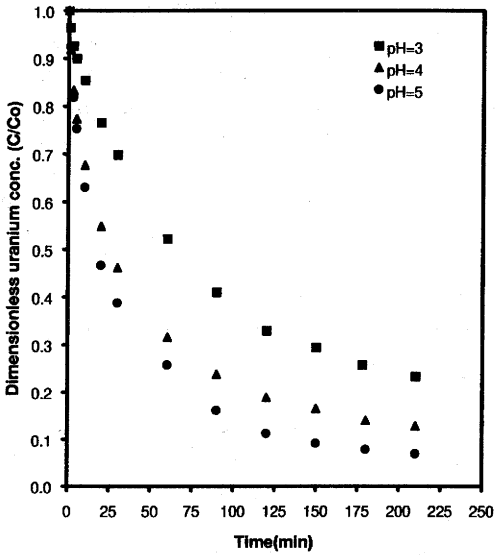
Figures 10(a) and 10(b) show pH variations during batch experiments at a uranium concentration of 200 mg/L. The pH of a uranium solution contacting chitosan increased very rapidly as the adsorption proceeded. In the case of oxidized carbon, the solution pH sharply decreased in the earlier adsorption stage and leveled off to the equilibrium pH. Similar phenomena were also observed in adsorption isotherm experiments.

Adsorption in a Fixed Bed

Breakthrough curves of uranium ions at an influent pH of 3–5 using oxidized carbon were obtained at uranium concentrations of 50 and 200 mg/L, as

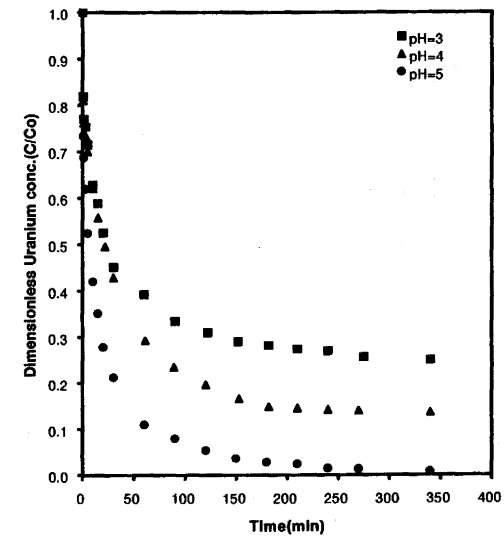


(a) Chitosan

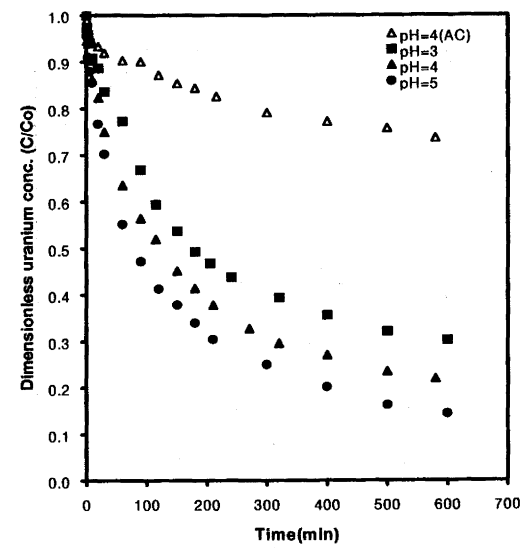


(b) Oxidized carbon

FIG. 8 Uptake curves of uranium on chitosan and oxidized carbon at uranium concentration of 50 mg/L.

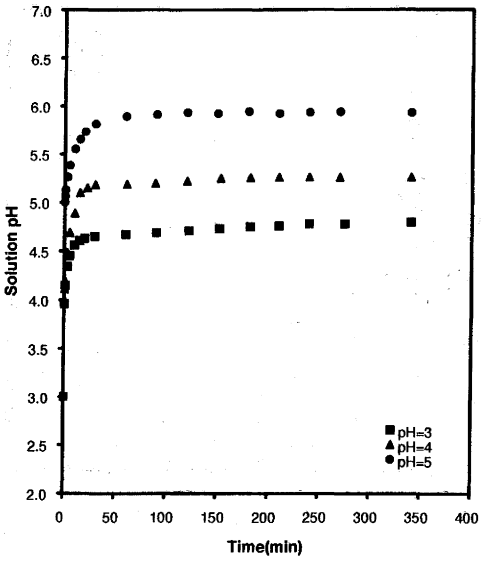


(a) Chitosan

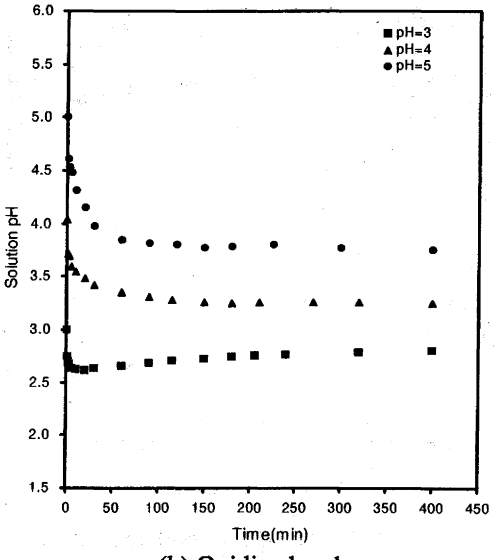


(b) Activated and oxidized carbons

FIG. 9 Uptake curves of uranium on chitosan and carbons at uranium concentration of 200 mg/L.

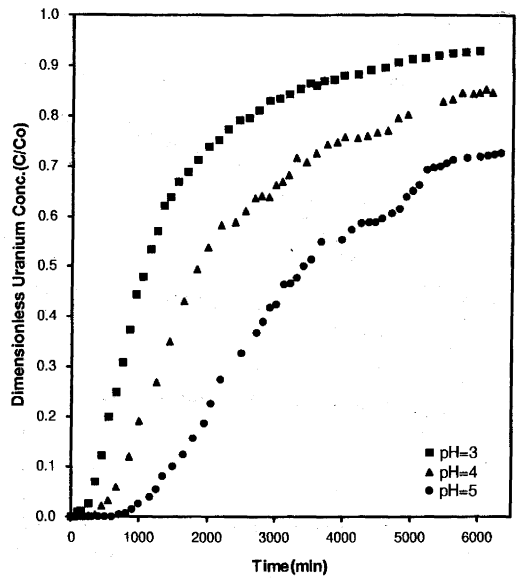


(a) Chitosan

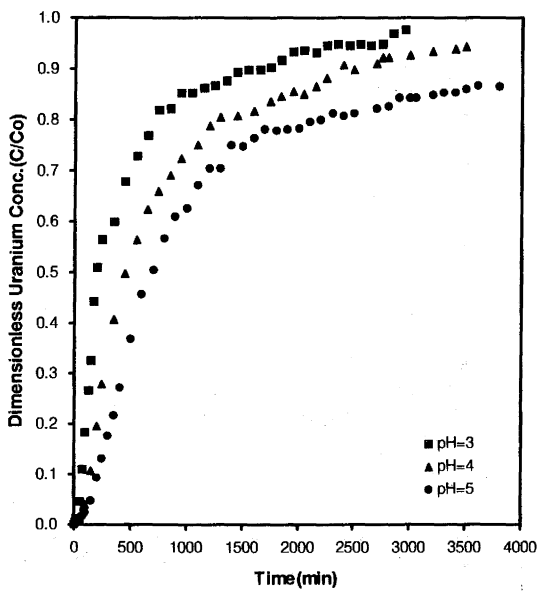


(b) Oxidized carbon

FIG. 10 Profiles of pH variation in batch experiment of uranium adsorption on chitosan and oxidized carbon.

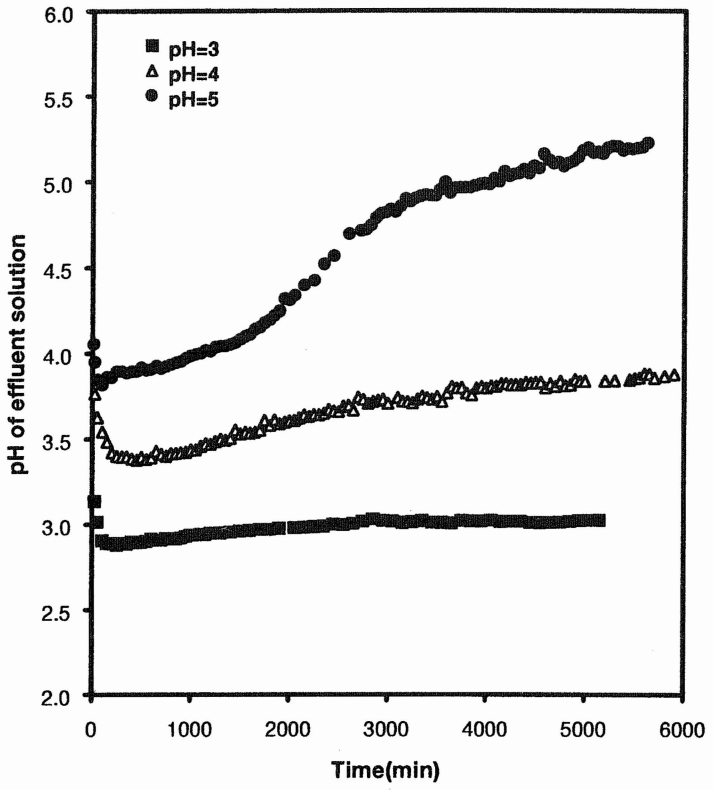


(a) Co=50 mg/l

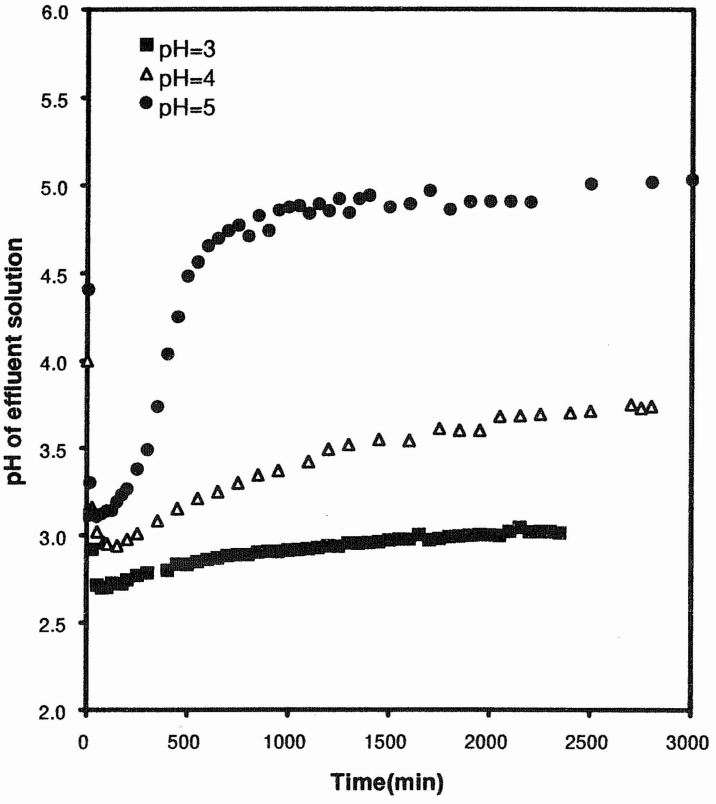


(b) Co=200 mg/l

FIG. 11 Effect of pH on the breakthrough curves of uranium adsorption on oxidized carbon at uranium concentrations of 50 and 200 mg/L (weight of carbon = 3 g).



(a) Co=50 mg/l



(b) Co=200 mg/l

FIG. 12 pH variation of effluent solution during the adsorption of uranium on oxidized carbon in fixed bed at pH 3–5 and C_0 = 50 and 200 mg/L.

shown in Figs. 11(a) and 11(b), respectively. At the influent pH 3, the breakthrough curve shows the common S-shape typical for single species adsorption on activated carbon. However, when the influent pH becomes higher, there are unusual breakthrough behaviors, such as trailing and plateau regions, which is normally represented by a multicomponent system with competitive adsorption (29–31).

The effluent pH decreased in the earlier adsorption stage, and then increased to influent pH as adsorption proceeded, as represented in Figs. 12(a) and 12(b). At pH 5 and a uranium concentration of 200 mg/L, the effluent pH sharply decreased up to about 3 and then rapidly increased compared with the pH variations at a concentration of 50 mg/L. As discussed previously, the rapid decrease of effluent pH in the earlier adsorption stage also implies that large amounts of uranium were removed by an ion-exchange mechanism with hydronium ions on the surface functional group (13).

It is thought that the trailing behaviors in the breakthrough profiles are closely related to differences in adsorption capacity with pH variation of a uranium solution. The rapid decrease of pH in the earlier adsorption stage leads to a decrease of the adsorption amount of uranium, mainly due to the pH effect. As adsorption proceeds, the solution pH in a fixed bed increases and the adsorption amount becomes higher. Therefore, in order to predict the adsorption characteristics of uranium in a fixed bed, the pH influence must be considered.

In one theoretical method the single-species isotherm parameters for each ionic form are extracted from all sets of adsorption equilibrium data at various pH values, and then these data are incorporated with the pH variation in a fixed bed.

CONCLUSION

Experimental studies were carried out to characterize the influence of pH on the adsorption of uranyl ions in aqueous solution using oxidized carbon and chitosan at various initial pH ranges. The variations of solution pH through the adsorption time in batch and fixed-bed experiments were also investigated.

The adsorption of uranium on both oxidized carbon and chitosan was strongly dependent on the initial solution pH. Although chitosan has a high affinity for uranium adsorption in the neutral pH range, its adsorption capacity at a relatively higher pH is greatly decreased due to an increase of the negative hydrolyzed uranyl species in the solution. However, based on isotherm data obtained at initial pH 3, 4, and 5, the adsorption capacity of oxidized carbon was comparable to that of chitosan when the pH and uranium concentration were lower. In addition, it was observed that surface-oxidized carbon has an adsorption capacity superior to that of untreated activated carbon. From this

point of view it is suggested that the use of oxidized carbon is practical for uranium removal from radioactive liquid waste which is generally acidic.

Kinetic data from batch experiments showed that chitosan has a relatively higher adsorption rate than oxidized carbon. The trends of solution pH profiles with adsorption time were a decrease for oxidized carbon and a rapid increase for chitosan. These pH variations provide further support for identifying the removal mechanism of uranium proposed in previous works.

Breakthrough curves of uranium in a column packed with oxidized carbon at influent pH 3, 4, and 5 were obtained. The effluent pH sharply decreased in the earlier adsorption stage, and such breakthrough behaviors as a plateau region and trailing in the later stage were observed. These adsorption characteristics can be interpreted by incorporating the equilibrium isotherms of each ionic species with pH variations during the adsorption process. Further works should be carried out to simulate the characterization of breakthrough behaviors in a fixed bed.

NOMENCLATURE

C	uranium concentration in solution (mg/L)
C_0	initial uranium concentration in solution (mg/L)
C_{eq}	equilibrium uranium concentration in solution (mg/L)
F	$1 - C_{eq}/C_0$ (—)
m	mass of adsorbent (mg)
pH_{pzc}	pH at which the net surface charge is zero
q	equilibrium amount adsorbed on adsorbent (mg/g)
q_m	maximum adsorption capacity of adsorbent (mg/g)
V	solution volume (mL)

Greek Letters

ε	molecular absorptivity of Arsenazo III
λ	wavelength

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