IMPROVEMENTS IN URANIUM ADSORPTION ABILITY OF A CIRCULATING FLUIDIZED BED ADSORBER

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We have developed a new type of fluidized-bed called a circulating fluidized-bed adsorber (CFBA) that brings adsorbent particles into contact with a large quantity of water at high efficiency. In the present study, effects of outer scale and internal structure of the CFBA on the uranium uptake from natural seawater were investigated, and some improvements in the adsorption ability of the CFBA were achieved.

By improving the internal structure of the CFBA, the total amount of uranium recovered from natural seawater became 1.6 times larger than that in an adsorber developed at the beginning of the study. The uranium recovery efficiency of the CFBA was compared with that of a conventional fluidized-bed adsorber. The amount of uranium adsorbed per unit volume of the CFBA was evaluated to be 20 % larger than that of the fluidized-bed adsorber.

Operational stability of the CFBA was examined for eighteen days. In spite of using unfiltered seawater, clogging did not occur during the adsorption period. The CFBA ran very stably.

Introduction

The world's total terrestrial uranium resources are estimated to be about five million tons. The total amount of uranium contained in the world's oceans is, however, estimated at about four billion tons, even though its concentration is extremely low at 3 ppb. For recovering uranium from seawater, Japan has many favorable features. The islands of Japan are surrounded by the sea and are washed by the Kuroshio Current. The Kuroshio Current can supply fresh uranium-bearing seawater to a uranium recovery plant, and the high seawater temperature of the current is advantageous for uranium recovery.

Among several methods for recovering uranium from seawater, adsorption methods using organic and inorganic adsorbents are considered to be among the most promising methods because of their high selectivity, ease of handling and safety to the environment[5]. In recent years, many efforts have been made to develop organic adsorbents which have high selectivity and high adsorption capacity[1, 4, 11]. To recover a significant amount of uranium by the adsorption method, however, an adsorbent must be contacted with a huge volume of seawater. Even if we assume 100 % recovery efficiency, 3×10^{11} m³/year of seawater must be processed in an adsorption plant for recovering one thousand tons of uranium per year.

We have developed a new type of fluidized-bed adsorber, called a circulating fluidized-bed adsorber (CFBA) that brings particulate adsorbent into contact with a large quantity of water at high efficiency[7]. In our previous study, the adsorption characteristics of the CFBA were investigated by experiments on the recovery of uranium from natural seawater[2]. A numerical model simulating the adsorption kinetics in the CFBA was also developed[3, 8]. The numerical simulation showed that the uranium was adsorbed mainly in a contacting section. This result suggested that the net contacting time between seawater and adsorbent in the contacting section should be increased to improve its adsorption ability.

In the present study, effects of outer scale and internal structure of the CFBA on its uranium adsorption performance were investigated according to the predictions of numerical simulation. To increase the net contacting time of adsorbent with seawater in the contacting section, the volume fraction of adsorbent in the contacting section was increased by modifying the internal structure. To evaluate the adsorption ability of the CFBA, the uranium recovery efficiency of the CFBA was compared with that of a conventional fluidized-bed adsorber.
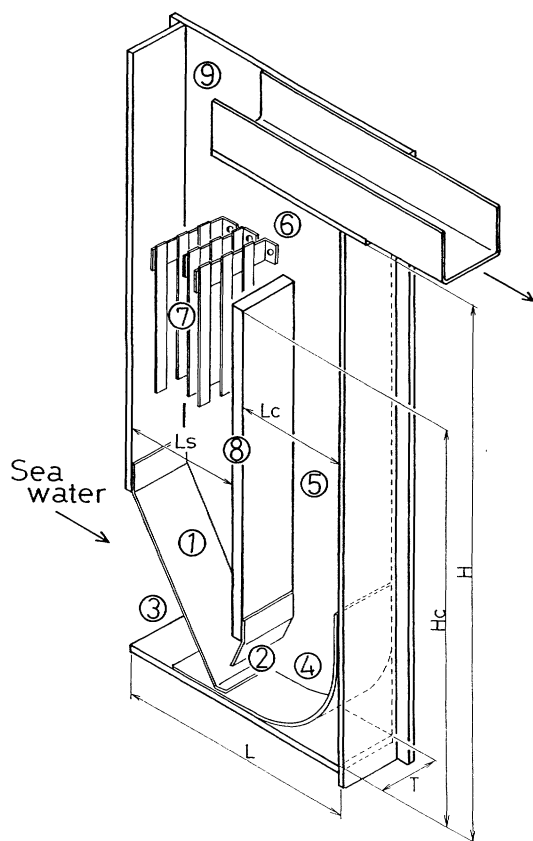
1. Experimental

1.1 CFBA

Figure 1 shows the CFBA used in the experiment. The movement of adsorbent particles and water in the CFBA is as follows. Water fed into the vessel from the nozzle (3 in the figure) develops a jet along the horizontal bottom wall and mixes with the adsorbent falling through a slit (2) from the packing section (1). The jet of

Fig. 1 Circulating fluidized-bed adsorber
1 packing section, 2 particle nozzle, 3 water inlet,
4 mixing section, 5 contacting section, 6 sedimentation
section, 7 baffle plates, 8 partition wall, 9 water outlet

Table 1. Properties of PAN-HTO adsorbent

Mesh	d_p (cm)	ρ_a (g/cm^3) dry wet	ρ_t (g/cm^3)	ε_p (-)	u_s (cm/s)
14/24	0.075	0.55 1.34	2.64	0.79	3.5

Table 2. Dimensions and adsorbent inventory of CFBAs

Model	H (cm)	H_c (cm)	L (cm).	L_c (cm)	L_s (cm)	W_{ad} (cm^3)
CFBA S	12.5	7.5	5	2	2.5	69
CFBA M	25	15	10	4	5.5	275
CFBA L	50	30	20	8	11	1100
CFBA 1	50	30	20	8	11	1100
CFBA 2	50	40	20	8	11	1710
CFBA 3	50	40	20	13	6.5	1700
CFBA 4	50	40	20	11	8.5	1800

T was fixed at 5 cm for all models.

the mixture turns its direction upward and ascends in the contacting section (5). At the top of this section, the mixture overflows the partition wall (8) and flows down toward the packing section. In the upper part of the packing section, the mixture collides with the surface of the packed adsorbents. Then the particles deposit on the surface, gradually move downward in the packing section and drop through the slit to mix again with fresh water, while the depleted water flows out of the opening on the top wall (9). The adsorbent particles circulate well in the CFBA without deposition and carry over in a certain water-velocity region[7].

1.2 Adsorption experiment

Experiments on the adsorption of uranium from natural seawater were carried out at the Sado Marine Biological Station of Niigata University, located on the northern coast of Sado Island in the Japan Sea. Natural seawater was pumped up in a 60 l overflow tank and then supplied to the adsorbers.

Hydrous titanium oxide granulated with polyacrylonitrile (PAN-HTO) was used as an adsorbent. Physical properties of PAN-HTO adsorbent are listed in **Table 1**[9].

At each prescribed time in operation, 2 cm^3 of the adsorbent particles was sampled from the CFBA and the amount of uranium adsorbed was determined as a function of adsorption time. Details of the analytical procedure for determining the uranium amount were reported previously[3].
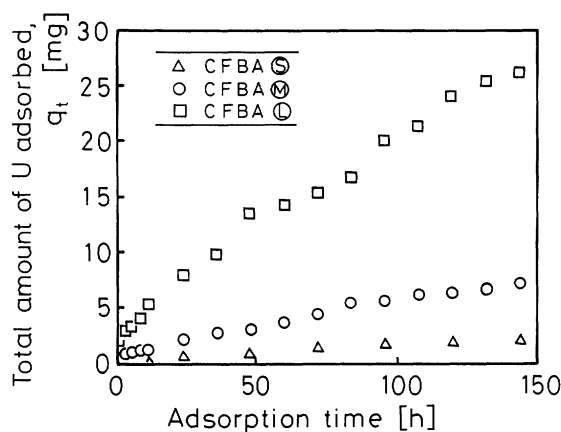
Table 2 shows the dimensions of the adsorbers used in the adsorption experiments. CFBA S, CFBA M and CFBA L were used to examine the effect of bed scale on uranium uptake. The three adsorbers were geometrically similar, with a scale factor of two or four except that the bed thickness T was fixed at 5 cm for all models. The volume ratio of these three adsorbers, therefore, was 1 : 4 : 16. The adsorbent inventory W_{ad} was kept to 24 % of each bed volume.

CFBA 1, CFBA 2 and CFBA 3 were used to examine the effect of internal structure of the adsorber on uranium uptake. The outer dimensions of the three adsorbers (H, L and T) were identical. The inner dimensions of CFBA 1 were the same as for CFBA L, which was developed at the beginning of the study. In CFBA 2, the contacting section height H_c was 10 cm larger than that of CFBA 1. In CFBA 3, the contacting section width L_c as well as H_c were larger, while the packing section width L_s was smaller. Thus the contacting section volume became larger in the order of CFBA 1 < CFBA 2 < CFBA 3. In CFBA 1 and CFBA 2, a small part of the adsorbent located at a corner of the packing section did not circulate smoothly and, therefore, a small dead space was formed in that section. In CFBA 3, however, the adsorbent moved downward smoothly and quickly in the packing section.

To compare the adsorption ability of CFBA with that of a conventional fluidized-bed adsorber, CFBA 4, which had dimensions between those of CFBA 2 and CFBA 3 was made. The fluidized-bed adsorber used in this experiment was made of 2.4 cm inner-diameter acrylic pipe. PAN-HTO (81 cm^3) was loaded in the fluidized-bed adsorber, and seawater flowed upward at a constant velocity of 0.5 cm/s.

Fig. 2 Effect of bed scale on uranium uptake. Seawater temperature is 26-28°C. u: CFBA S; 1.1 cm/s, CFBA M; 0.9 cm/s, CFBA L; 1.4 cm/s

2. Results

2.1 Scale of CFBA

The effect of bed scale on uranium uptake is shown in **Fig. 2**. Seawater temperature ranged from 26 to 28°C during the experiment. The amount q of uranium adsorbed per unit weight of adsorbent decreased slightly with increase in bed scale, but the degree of decrease was very small. Thus, the total amount q_t of recovered uranium, which is equal to q times mass of the loaded adsorbent, increased proportionally to bed scale as shown in Fig. 2. At the end of an adsorption period of 144 hours, the total amounts of recovered uranium were 1.9, 7.2 and 26.7 mg respectively for CFBA S, CFBA M, CFBA L.

2.2 Volume fraction α of adsorbent

According to predictions from previous numerical simulation, the uranium uptake increases with increasing net contacting time between seawater and adsorbent in the contacting section[3]. This is achieved by increasing the volume fraction of adsorbent in the contacting section and shortening the deposition time of adsorbent in the packing section. The volume fraction α is the ratio of the bulk volume of adsorbent particles suspended in the contacting section to the volume of that section.

Figure 3 shows the effect of volume fraction α of adsorbent in the contacting section on uranium uptake. These curves were obtained by numerical calculation. Material valances in CFBA for the numerical calculation are as follows.
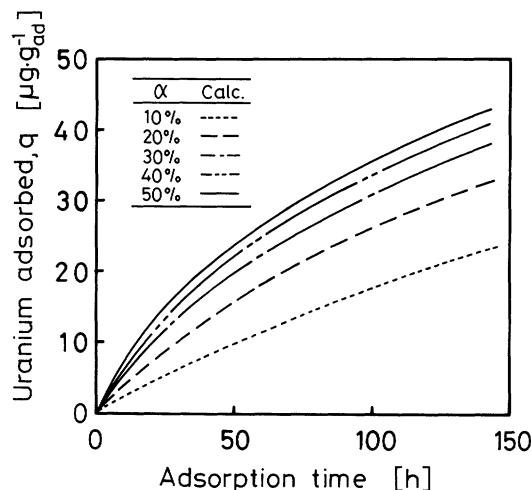
Material balance in the contacting section:

$$u_c \frac{\partial C}{\partial h} + k_f a_v (C - c_s) = \varepsilon_b D_b \frac{\partial^2 C}{\partial h^2} \tag{1}$$

Diffusion in a particle:

$$D_e \left(\frac{\partial^2 c}{\partial r^2} + \frac{2}{r} \frac{\partial c}{\partial r} \right) = \rho_a \frac{\partial q}{\partial t} \tag{2}$$

Boundary condition:

Fig. 3 Numerical calculation of influence of α on uranium uptake for CFBA M. Parameters used in numerical calculation are tabulated in Table 3

Table 3. List of parameters used in numerical calculation

d_p (cm)	D_e^* (cm²/s)	n^{**} (-)	q_o^{**} (μg/g)	H_c (cm)	W_{ad} (cm³)	u (cm/s)
0.075	1.4×10^{-6}	1.5	103	15	275	1.3

*, **) Data taken from our previous studies[3, 9].

$$\left(\frac{\partial c}{\partial r} \right)_{r=r_p} = \frac{\int_{Z_{in}}^{Z_{out}} k_f (C - c_s) \, dh}{D_e} \tag{3}$$
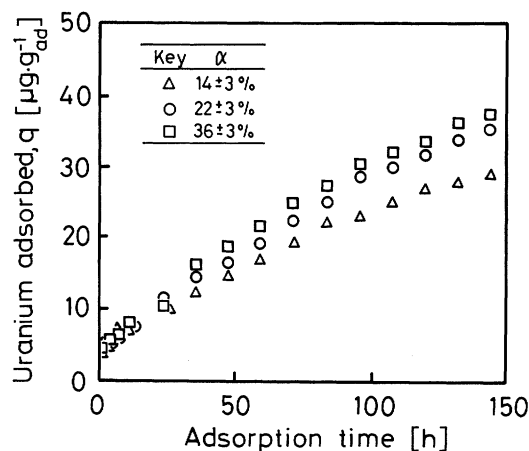
Initial condition:

$$q = c = 0 \quad (t = 0, \ 0 \le r \le r_p) \tag{4}$$
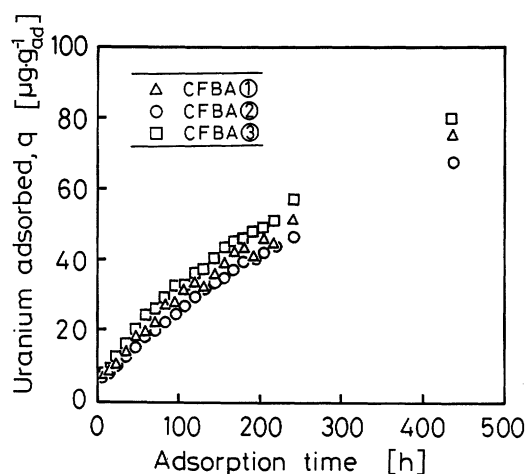
Freundlich-type adsorption isotherm:

$$\frac{q}{q_0} = \left(\frac{c}{C_0} \right)^{\frac{1}{n}} \tag{5}$$

Time dependence of the amount of uranium adsorbed in the CFBA is calculated by solving Eqs. (1) to (5) numerically by the Crank-Nicolson method. Details of the numerical simulation were reported previously[3]. The CFBA used in the numerical calculation is that of medium size in Table 2, i.e., CFBA M. The parameters used in this calculation are tabulated in **Table 3**. These values are based on the results of the adsorption experiment. The adsorbent inventory is kept to 275 cm³ for each α. Thus, when α increases, the amount of adsorbent deposited in the packing section decreases, and the net contacting time between seawater and adsorbent in the contacting section increases. As shown in this figure, q increases with increase in α: when α becomes large, the increase rate of q becomes small.

This relation between α and q was confirmed experimentally as shown in **Fig. 4**. The value of α was adjusted by changing the width of particle nozzle 2 in Fig. 1. The bulk volume of adsorbent particles sus-

Fig. 4 Influence of α on uranium uptake obtained experimentally for CFBA M. Seawater temperature is 14-16°C. $u = 1.3$ cm/s, $W_{ad} = 275$ cm³
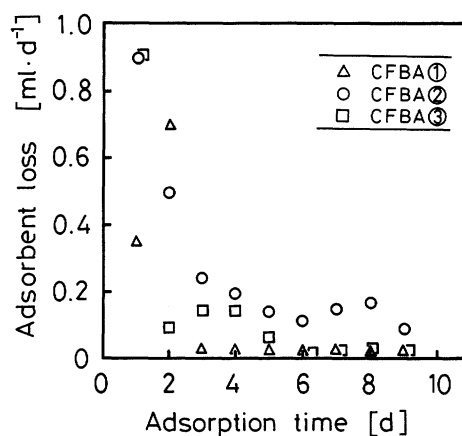

Fig. 5 Time profiles of uranium uptake in CFBA 1, CFBA 2 and CFBA 3. Seawater temperature is 18-21°C. $u = 1.1$ cm/s

pended in the contacting section was evaluated by subtracting the amount of adsorbent in the packing section, which was evaluated by a side-view photograph, from the adsorbent inventory. This range of α, i.e., $\alpha = 14$ to 36 %, is an adjustable range in the CFBA. An increase of α over 36 % is difficult because of the pressure balance between the contacting section and the packing section in the CFBA[7]. The uranium uptake q increased greatly when α increased from 14 to 22 %, while the increase from $\alpha = 22$ to 36 % was only a few percent. These time profiles of uranium uptake as a function of α are in good agreement with the simulation results shown in Fig. 3. These results confirm the reliability of the adsorption kinetic model.

2.3 Internal structure of the CFBA

Another approach to increase the uranium uptake is enlargement of the contacting section volume V_c (= $H_c \times L_c \times T$). To demonstrate the effect, we conducted an adsorption experiment using three CFBA models with different V_c values, i.e., CFBA 1, CFBA 2 and CFBA 3 in Table 2. The adsorbent inventories in these adsorbers

Fig. 6 Amount of carried-over adsorbent particles per day during adsorption experiment. $u = 1.1$ cm/s

were 1100, 1710 and 1700 cm³ respectively.

Figure 5 shows the time profiles of uranium uptake in these three adsorbers. The values of q in CFBA 2 and CFBA 3 were comparable to that in CFBA 1 in spite of larger adsorbent inventory. Therefore, the total amounts of recovered uranium q_t ($\propto q \times W_{ad}$) were 44.0, 60.7 and 72.1 mg in the order of CFBA 1, CFBA 2 and CFBA 3 at the end of 436 hours of operation. The total uranium uptake became 1.6 times larger by improving the internal structure of the adsorber and loading a large quantity of adsorbent.

2.4 Operational stability

The amounts of adsorbent carried over per day from CFBAs are plotted as a function of adsorption time in **Fig. 6**. The mean linear velocity u of seawater was 1.1 cm/s in each adsorber. The value of u was derived from dividing the seawater flow rate F by the cross-section of the whole equipment $L \times T$, i.e., $u = F/(L \times T)$. The seawater velocity in the contacting section was 2.5 times larger than u in CFBA 1 and CFBA 2. In CFBA 3, it was 1.5 times larger than u.

A small amount of the adsorbent particles was carried over throughout the adsorption experiment. The carryover rate decreased with increase in adsorption time, and most of the particles carried over were of very small size. These fine particles were contained in as-supplied adsorbent and had not been removed in spite of grading. At the end of 216 hours operation, the ratios of the total adsorbent loss to the initial inventory were 0.06, 0.15 and 0.09 % in the order of CFBA 1, CFBA 2 and CFBA 3. In a test plant at the Nio Institute on uranium recovery from seawater, three types of fluidized beds were used as the adsorption apparatus[10]. It was reported that the adsorbent loss was about 0.1 % of the initial inventory for 480 hours operation in these test plants[6]. Thus the adsorbent losses in CFBAs were of comparable magnitude with the actual fluidized-bed apparatus.

The variations of mean linear velocity in the three adsorbers were very small through the experimental period, as shown in **Fig. 7**. In this experiment, u was set
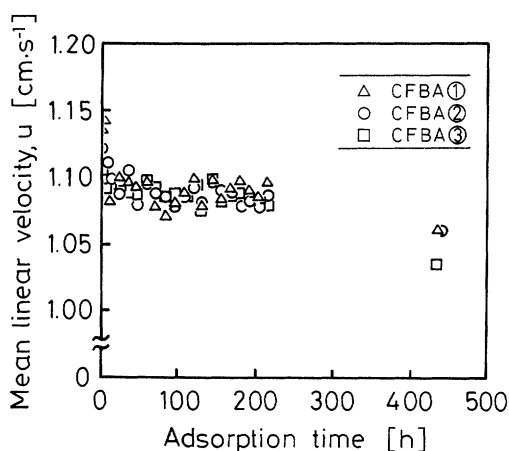
Fig. 7 Change of mean linear velocity of seawater during adsorption experiment. Set value of u is 1.1 cm/s

Fig. 8 Influence of contacting section height on uranium recovery ratio obtained by a numerical calculation

to be 1.1 cm/s. At the end of 436 hours operation, the change of u in CFBA 1 was 4 % of the set value. In CFBA 2 and CFBA 3, the changes were 4 and 6 % respectively. Even though unfiltered seawater was supplied to the CFBA in the experiment, u changed only slightly. This result shows that clogging by contamination such as suspended solids and seaweed in natural seawater does not occur in the CFBA at any time.

3. Discussion

When supplying fresh seawater into the CFBA, the adsorbent particles mix with the seawater and ascend in the contacting section. Adsorption of uranium mainly occurs in this section. Therefore, the concentration of uranium in the seawater gradually decreases while ascending in this section. The degree of concentration decrease becomes large with increase in bed scale, so that the total uranium uptake will not necessarily increase in proportion to bed scale.

In this experiment, however, the total amount q_t of recovered uranium increased proportionally with bed scale, as shown in Fig. 2. Hence we examined the effect of contacting section height on the total uranium amount by numerical calculation. The result is shown in **Fig. 8**. In this figure, the vertical axis represents the uranium recovery ratio R, the ratio of the amount of uranium adsorbed to that available in the seawater. The parameters used in this calculation is as follows: $d_p = 0.075$ cm, $D_e = 2.2 \times 10^{-6}$ cm^2/s, $n = 1.5$, $q_o = 149$ μ g/g, $u = 1.5$ cm/s and $\alpha = 30$ %. These values are based on the results of an adsorption experiment using PAN-HTO adsorbent[3, 9]. We also assume that the seawater temperature is 25°C and that the adsorption period is 20 days. The uranium recovery ratio increases linearly with bed scale when the height is less than 1 m. This is in agreement with the experimental data mentioned above. When it is higher than 1 m, however, the ratio becomes smaller than the value expected by linear extrapolation.

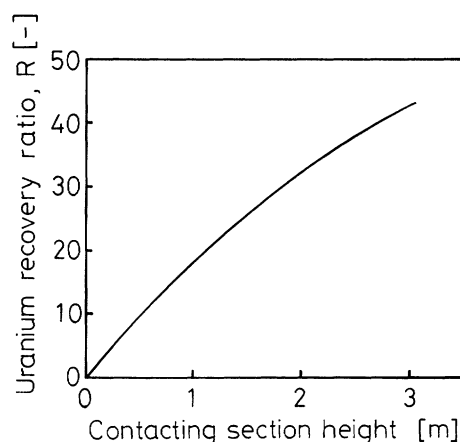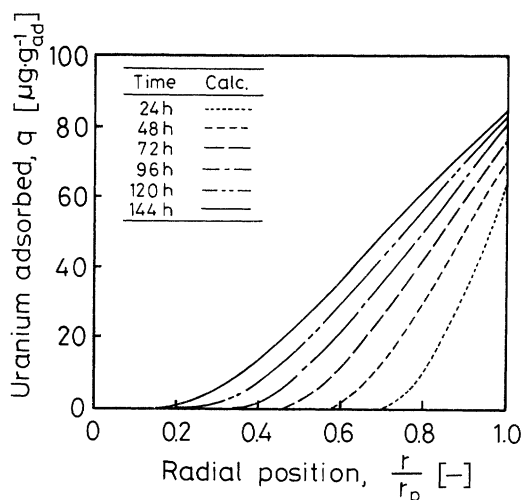The limit of scale-up of CFBA could be evaluated

from the energy consumption in supplying seawater. The major portion of the total pressure drop through the CFBA occurs in the contacting section, where the pressure drop is proportional to the amount of adsorbent suspended in that section. In CFBA L used in this experiment, the pressure drop is about 1.8 cm H$_2$O at a mean linear velocity of 1.5 cm/s when PAN-HTO is used as the adsorbent[7]. We plan to utilize the Kuroshio current, which flows near the islands of Japan, as a means to supply seawater into the CFBA. The velocity range of the Kuroshio current is 125 to 225 cm/s. A current velocity of 125 cm/s corresponds to a water-column pressure of 8 cm H$_2$O. Therefore, CFBA L could scale up more than four times; i.e., its contacting section height is over 1.6 m, assuming that the ocean current is used directly and PAN-HTO is used as the adsorbent. In this condition, a uranium recovery ratio of 27 % is expected from Fig. 8.

When the volume fraction α of adsorbent in the contacting section increases, the amount of adsorbent in the packing section decreases with α. Thus, the deposition time of adsorbent in that section becomes short. This implies that the net contacting time between seawater and adsorbent in the contacting section increases with α. The uranium amount q, therefore, increases with increase of α as shown in Figs. 3 and 4, even though the increase rate of q becomes smaller at higher α. The reason for the decrease in the increase rate of q at higher α is clarified by numerical calculation. **Figure 9** shows the distribution of uranium adsorbed in an adsorbent particle. These curves are obtained by numerical calculation. Such calculation is done for the experimental condition of $\alpha = 22$ % in Fig. 4. Other parameters used in the calculation are the same as for the case of Fig. 3. The diffusion rate of uranium into the particle is small— 1.4×10^{-6} cm^2/s[3]—so the adsorbed uranium concentrates on the surface of the adsorbent particle. The concentration of uranium on the surface becomes higher with increase in α. Thus, the increase rate of q becomes smaller at higher α, as shown in Figs. 3 and 4, because
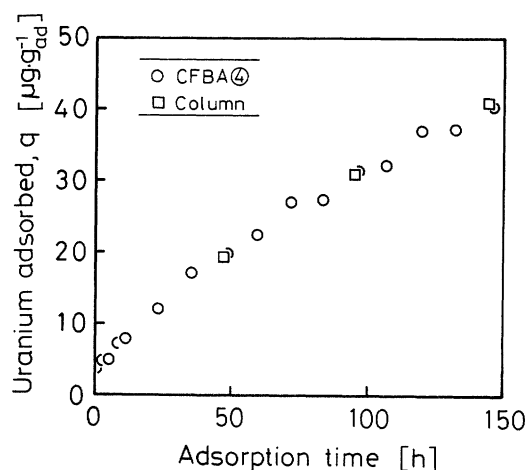
Fig. 9 Numerical calculation of distribution of uranium adsorbed in an adsorbent particle based on experimental conditions for α =22 % in Figure 4. Parameters used in the numerical calculation are shown in Table 3

Fig. 10 Comparison of uranium uptakes in CFBA 4 and a fluidized-bed adsorber. Seawater temperature is 18-19°C

the uranium adsorption rate from the liquid phase to the particle surface is proportional to the concentration gradient across the liquid-film layer. The adjustable range of α is 14 to 36 % in CFBA as mentioned previously. Hence, we consider that the optimum value of α is 36 % in the present conditions.

Considering the experimental results presented so far, the proper design of a CFBA using PAN-HTO adsorbent is CFBA 2 or CFBA 3 in Table 2. An improved model, CFBA 4, was made and its adsorption ability was compared with that of a fluidized-bed adsorber. To compare the adsorption characteristics of the adsorbers, the adsorbent and seawater conditions must be equal. We carried out, therefore, an experiment with a fluidized-bed adsorber simultaneously with the experiment on the CFBA. The uranium uptakes q in the two adsorbers are shown in **Fig. 10**. This result shows that q in CFBA 4 is nearly equal to that in the fluidized-bed adsorber throughout the adsorption period. A ratio of the adsorbent inventory to the total bed volume is 36 % in CFBA 4, while the ratio for the fluidized-bed adsorber is estimated to be 25-30 % from the results of the test plant at the Nio Institute, where total bed height was twice its fluidization height[10]. Using this value in estimating the bed volume of the fluidized-bed adsorber, the amount of uranium adsorbed per unit volume of CFBA 4 becomes larger than that of the fluidized bed adsorber by a factor of 1.2-1.5.

Conclusions

Experiments on the recovery of uranium from natural seawater were carried out using a CFBA. Hydrous titanium oxide granulated with polyacrylonitrile was used as the adsorbent. The effects of scale-up and the internal structure of the CFBA on uranium uptake were investigated in order to improve adsorption ability.

Uranium uptake increased with increase in bed scale. By improving the internal structure of the CFBA, the total amount of uranium recovered from natural seawater became 1.6 times larger than that in the adsorber developed at the beginning of the study. The uranium recovery efficiency of the CFBA was compared with that of a conventional fluidized-bed adsorber. The amount of uranium adsorbed per unit volume of the CFBA was evaluated to be 20 % larger than that of the fluidized-bed adsorber. The adsorption ability of the CFBA is expected to be superior to that of the conventional fluidized-bed adsorber.

Operational stability of CFBA was proved for a period of eighteen days. Clogging did not occur during the adsorption period, even though unfiltered seawater was supplied directly. The CFBA ran very stably.

The CFBA developed here is applicable to other applications in water-treatment engineering, where trace contaminants are to be recovered or removed from a large volume of water.

Acknowledgment

Nomenclature

a_v	=	particle surface area per unit volume	
		of contacting section	[cm²/cm³]
C	=	concentration of uranium in liquid phase	[μg/l]
C_o	=	uranium concentration in seawater	[μg/l]
c	=	concentration of uranium with radial position r	
		in particle	[μg/l]
c_s	=	concentration of uranium on particle surface	[μg/l]
D_b	=	axial dispersion coefficient	[cm²/s]
D_e	=	intraparticle diffusion coefficient	[cm²/s]
d_p	=	average particle size	[cm]

F	=	seawater flow rate	[cm³/s]
H	=	bed height	[cm]
H_c	=	contacting section height	[cm]
h	=	axial position in contacting section	[cm]
k_f	=	liquid-film mass transfer coefficient	[cm/s]
L	=	bed width	[cm]
L_c	=	contacting section width	[cm]
L_s	=	packing section width	[cm]
n	=	Freundlich constant	[-]
q	=	amount of uranium adsorbed at radial position r in particle	[μg/g]
q_t	=	total amount of uranium adsorbed ($\propto q \cdot W_{ad}$)	[mg]
q_o	=	amount of uranium adsorbed in equilibrium with C_o	[μg/g]
R	=	uranium recovery ratio	[%]
r	=	radial position in particle	[cm]
r_p	=	particle radius	[cm]
T	=	bed thickness	[cm]
t	=	adsorption time	[h]
u	=	mean linear velocity of seawater = $F/(L \cdot T)$	[cm/s]
u_c	=	seawater velocity in contacting section = $F/(L_c \cdot T)$	[cm/s]
u_s	=	settling velocity of an adsorbent particle	[cm/s]
w_{ad}	=	bulk volume of adsorbent inventory	[cm³]
z	=	adsorbent layer in contacting section	[-]
α	=	volume fraction of adsorbent in contacting section	[-]
ε_b	=	void fraction in contacting section	[-]
ε_p	=	adsorbent porosity	[-]
ρ_a	=	apparent density of adsorbent particle	[g/cm³]
ρ_t	=	true density of adsorbent particle	[g/cm³]

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