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Chelating Polymers for Recovery of Uranium from Seawater*

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ABSTRACT

Despite the low concentration of uranium in seawater (3.3 ppb), a special emphasis has been placed on its recovery. Although the concentration is low, it has been estimated that the world's oceans contain about 4×10^9 tons of uranium—theoretically an unlimited supply of nuclear fuel. Adsorption has been considered to be a technically feasible procedure for a uranium recovery process with regard to economic and environmental impacts. The present paper restricts its coverage to those applications using chelating polymeric resins containing amidoxime groups as the most promising adsorbent.

INTRODUCTION

Considering the anticipated exhaustion of terresterial uranium reserves in the near future, the extraction of uranium from seawater has become of interest during the past three decades (1–8).

The first work was reported by Davies et al. (7). They tested many different organic and inorganic adsorbents for their ability to recover ura-

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* Dedicated to Prof. Iwao Tabushi for his endeavors in the field of "recovery of uranium from seawater."

nium from seawater. Among the resins containing chelating functional groups (e.g., mono- and dibasic phosphoric and phosphonic acids, 8-hydroxyquinoline, kojic acid, salicylic acid, amino-carboxylic acid, and amino-phosphoric acid), a condensation product of formaldehyde and resorcinol arsonic acid was shown to have a high uranium uptake (9–12).

Several inorganic adsorbents had uptakes comparable to the arsonic acid resin, and a hydrous titanium oxide was selected as the most promising (13).

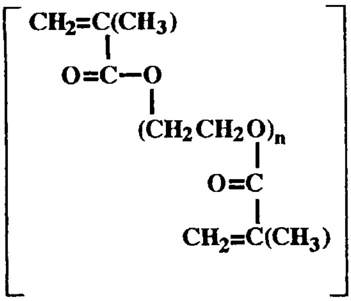
However, the development of chelating polymeric resins demonstrated that this type of adsorbent was much more suitable than hydrous titanium oxide due to its higher capacity, higher adsorption rate, recycling ability, and better mechanical stability.

SPHERICAL AMIDOXIME-RESINS

Egawa et al. synthesized a number of chelating resins containing the amidoxime group (RNH) according to the reactions illustrated in Scheme 1. It was found that these RNHs exhibit much superior adsorption characteristics than the previously developed hydrous titanium oxide (14–17). Several research groups continued this line of work, developing the various kinds of adsorbents containing amidoxime groups (18–21).

Egawa et al. reported that important factors governing improvement of the uptake rate for uranium include the optimization of the porosity and the specific surface area in the matrix polymer. Several attempts have been made to obtain chelating resins with an effective pore structure and a high mechanical stability by Egawa et al. (14–17).

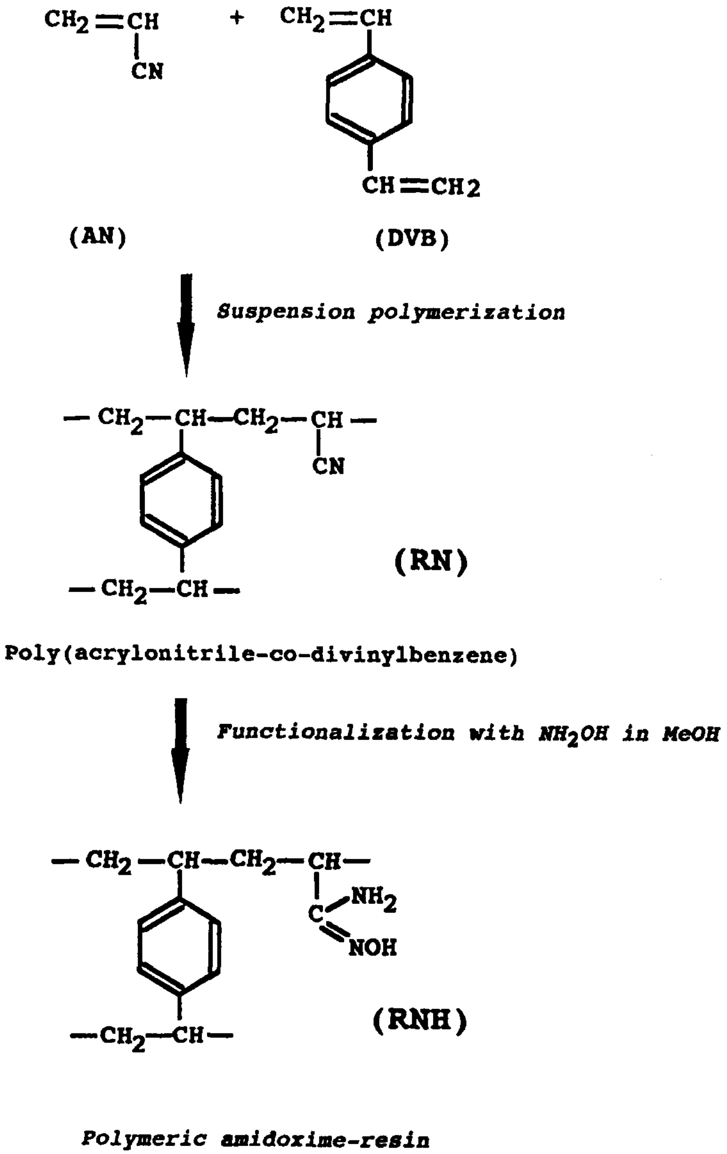
A number of macroreticular chelating resins containing amidoxime groups with various degrees of crosslinking were synthesized by using various amounts of ethyleneglycol dimethacrylate (1G), diethyleneglycol dimethacrylate (2G), triethyleneglycol dimethacrylate (3G), tetraethyleneglycol dimethacrylate (4G), and nanoethyleneglycol dimethacrylate (9G) as the hydrophilic crosslinking reagents (**1**).



$$\left[\begin{array}{l} CH_2{=}C(CH_3) \\ \quad | \\ O{=}C{-}O \\ \qquad | \\ \qquad (CH_2CH_2O)_n \\ \qquad\qquad | \\ \qquad\quad O{=}C \\ \qquad\qquad | \\ \qquad CH_2{=}C(CH_3) \end{array}\right]$$

n=1 1G
n=2 2G
n=3 3G
n=4 4G
n=9 9G

(1)

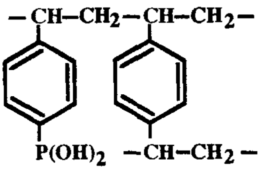


SCHEME 1 Preparation of chelating resins (RNH) containing amidoxime group.

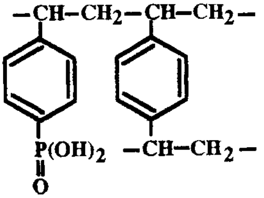
These resins showed a high adsorption ability for uranium because of the formation of a favorable pore structure (micropore) based on the swelling caused by alkaline treatment. However, these resins exhibited low physical and chemical stability (22).

On the other hand, the chelating resins prepared by the simultaneous use of etyleneglycol dimethacrylate and divinylbenzene had an effective macropore and micropore structure for recovery of uranium from seawater (23). In similar work, resins based on poly(acrylonitrile-*co*-divinylbenzene-*co*-methylacrylate) exhibited a high adsorption rate for uranium (i.e., 100 mg of U/L of resin/10 days or 260 mg of U/kg of resin/10 days) (24).

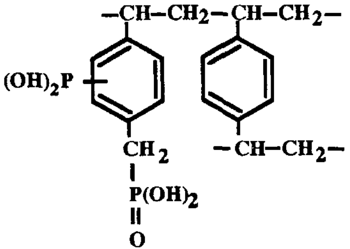
Macroreticular chelating resins [RSP (**2**), RSPO (**3**), RCSP (**4**) and RCSPO (**5**)] containing dihydroxyphosphino and/or -phosphano groups were synthesized elsewhere, and their sorption capacity for UO_2^{2+} and the recovery of uranium from seawater were investigated by Egawa et al. (25, 26).



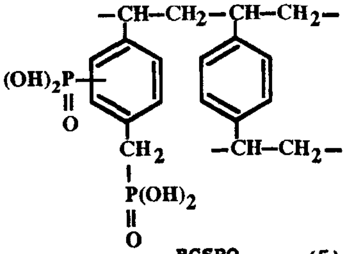
RSP *(2)*



RSPO *(3)*



RCSP *(4)*



RCSPO *(5)*

In addition, these resins were used for the purification and concentration of uranium eluted from RNH with acidic solutions (27, 28).

Egawa et al. recently reported that the polymeric amidoxime-resins derived from macroreticular acrylonitrile-divinylbenzene (DVB 10 mol%) copolymer beads which were synthesized by varying the amount of polymerization initiator exhibited different adsorption abilities for uranium, although they are similar in chemical properties and physical pore structure. It was concluded that the pore structure in the swollen state is important for the improvement of the adsorption rate (29). We recently synthesized the lightly crosslinked (DVB 5 mol%) amidoxime-resins using benzoyl peroxide (BPO 40 or 180 mmol/L-monomer mixture) or azobisisobutyronitrile (AIBN 180 mmol/L-monomer mixture) as a polymerization initiator. Despite having superficially similar physical and chemical properties, the chelating resins based on the lightly crosslinked acrylonitrile–divinylbenzene copolymer beads which were synthesized using AIBN (180 mmol/L-monomer mixture) showed a remarkably enhanced adsorption rate for uranium from seawater (30). The enhancement in adsorption rate was attributed to increased micropores in the swollen state on the basis of data obtained by gel permeation chromatography (shown in Fig. 1).

We have also described the importance of alkaline treatment on lightly crosslinked chelating resins containing amidoxime groups for the enhancement in uranium uptake from seawater (30). The diffusion of uranium was remarkably hastened by alkaline treatment through micropore formation and chemical structural changes as well. As shown in Fig. 2, alkaline

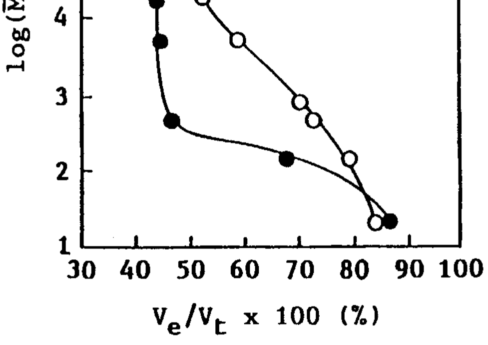


FIG. 1 Calibration curves of GPC for chelating resins (RNH) synthesized using various polymerization initiators during suspension copolymerization. Eluant: deionized water. Flow rate: 0.5 $cm^3 \cdot min^{-1}$. Column: 30 cm × 5 mm i.d. (●) BPO, 40 $mmol \cdot dm^{-3}$ monomer. (○) AIBN, 180 $mmol \cdot dm^{-3}$ monomer.

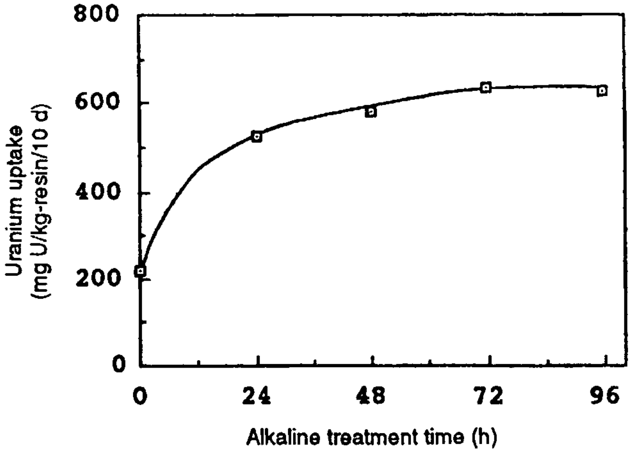


FIG. 2 Effect of alkaline treatment on uranium uptake from seawater.

treatment at 30°C for 72 hours is required for optimum performance. In addition, gel permeation chromatography gave some information for the analysis of pore structure in the swollen state. Figure 3 graphically summarizes the changes in the pore structure by alkaline treatment. The micropore formation caused by swelling led to permeation of the larger molecules. Thus, uranium could easily attach to the fixed donating atoms through diffusion into micropores due to the high swelling (30, 31).

On the other hand, solid-state (CP-MAS) ^{13}C-NMR spectroscopy gave some information on the resulting chemical structure of alkali-treated polyacrylamidoxime (31, 43). It was shown that the amidoxime group seemed to be partially converted to cyclic imidedioxime and carboxylic acid after alkaline treatment. It was speculated that both pore structure in the swollen state and the chemical structure contributed to the overall performance by alkaline treatment (31).

Most recently, we synthesized lightly crosslinked, highly porous chelating resins containing amidoxime groups by using solvating diluents such as chloroform, dichloroethane, and tetrachloroethane during suspension polymerization (31). These resins provided a more favorable pore structure for a rapid rate of diffusion of uranium within the resin structure. These resins exhibited remarkably enhanced adsorption rates for uranium due to their modified pore structures. Figure 4 shows time plots of uranium adsorption during 30 days of large-scale performance tests carried out with highly porous chelating resins (DCE, dichloroethane; CH, chloroform; TCE, tetrachloroethane). Elsewhere, the kinetic behavior of these

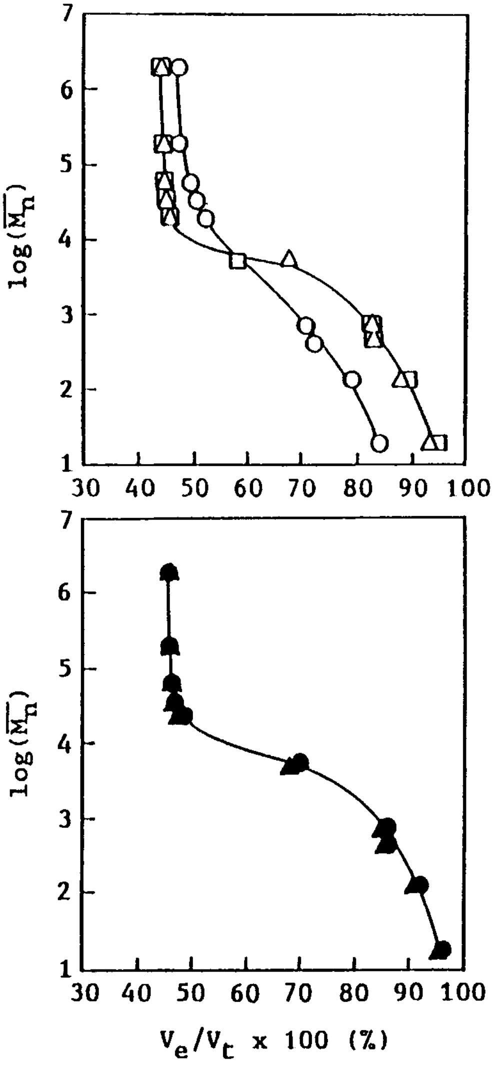


FIG. 3 Effect of alkaline treatment period on the calibration curves of GPC for RNH in aqueous medium. Alkaline treatment: 1.0 mol·dm^{-3} NaOH, 30°C. (○) 0 h, (△) 24 h, (□) 48 h, (●) 72 h, (▲) 96 h.

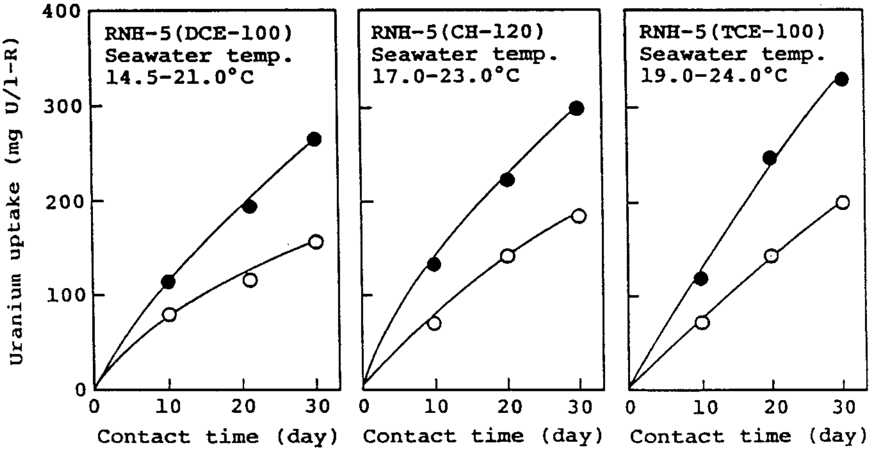


FIG. 4 Plots of adsorption rates during the large-scale performance tests. Adsorption: Columnar (upflow); column, 60 mmϕ × 1 m; resin, 50 cm^3; SV, 640 $hour^{-1}$. Elution: Resin, 1 cm^3, H_2SO_4 (0.5 mol·dm^{-3}), 10 BV; SV, 3 $hour^{-1}$. (○) Nontreated resin. (●) Alkali-treated resin.

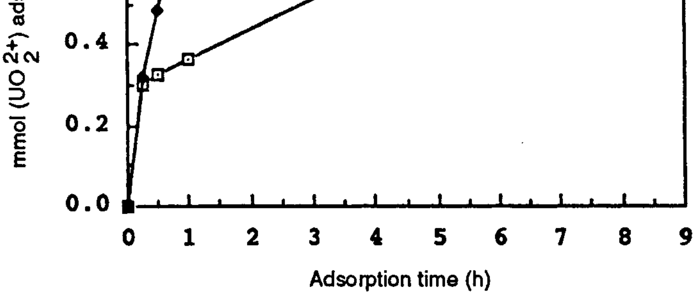


FIG. 5 Effect of alkaline treatment on the loading of chelating resins RNH-5 (CH-120) using 0.01 mol UO_2^{2+} ·dm^{-3} at pH 3.25 and 30°C as a function of time. (□) Nontreated. (■) Alkali-treated resin.

resins for batchwise adsorption of UO_2^{2+} was examined (32). An increase in porogen content resulted in a marked increase in the adsorption of UO_2^{2+}. As shown in Fig. 5, alkaline treatment allowed high swelling and hence rapid accessibility of UO_2^{2+} to the ligands.

In addition, the lightly crosslinked, highly porous chelating resins have been used in large-scale column adsorption/elution cycles to examine the resin performance as well as the efficacy of eluant (33). The adsorbed metal ions were rapidly and more quantitatively eluted from the resins with acid eluants. Figures 6 and 7 show the elution curves obtained by the employment of H_2SO_4 or HCl as the eluting agent. Uranium recovery was maintained at a nearly constant value by the employment of bicarbonate eluants. However, it was clarified that elution with 0.25 M H_2SO_4 was better than with bicarbonate eluants in view of the efficient and selective stripping of uranium as well as from an economic viewpoint. The lightly crosslinked chelating resins showed an affinity for not only uranium but also for other metal ions. As shown in Table 1, the concentration factors for Zn(II), Ni(II) and V(V) were as large as that of uranium among the metal ions adsorbed (33). A high reproducibility was confirmed with repeated adsorption/elution cycles. For comparison, the values obtained after cycle 2 were also summarized in Table 1.

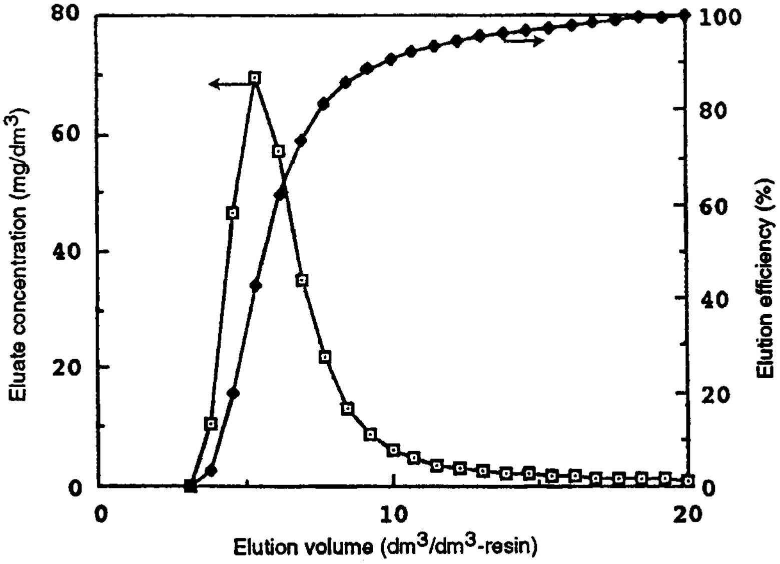


FIG. 6 (□) Elution curve of uranium. (■) Elution efficiency of uranium. Resin, 13 cm^3; SV, 3 $hour^{-1}$; eluting agent, 0.25 $mol \cdot dm^{-3}$ H_2SO_4.

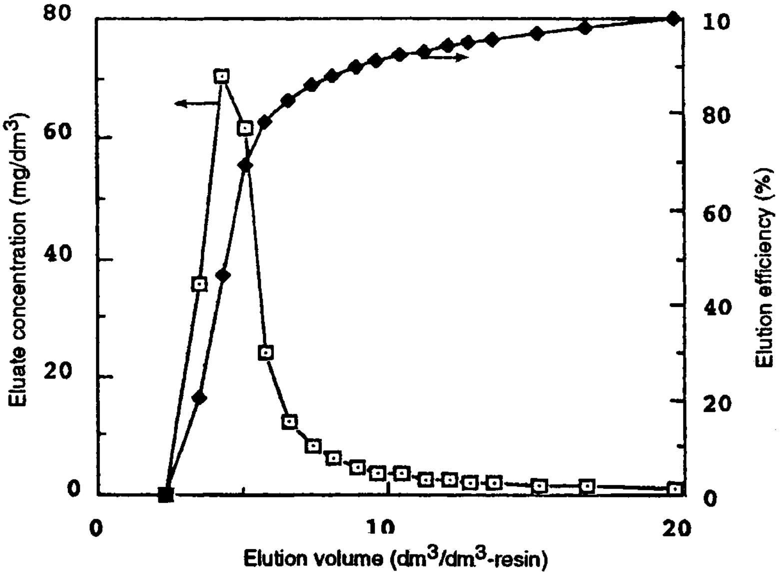


FIG. 7 (□) Elution curve of uranium. (■) Elution efficiency of uranium. Resin, 13 cm^3; SV, 3 hour^{-1}; eluting agent, 0.5 mol·dm^{-3} HCl.

TABLE 1
Concentration Factors for Various Metal Ions Adsorbed on Chelating Resin RNH-5(CH-120)AT[a]

Metal	Concentration[b] in seawater (mg·dm^{-3})	Cycle 1		Cycle 2	
		Metal ion adsorbed (mg/kg-resin)	Concentration factor (CF)[c]	Metal ion adsorbed (mg/kg-resin)	Concentration factor (CF)
Cu	0.003	196	6.5×10^4	115	3.8×10^4
Fe	0.01	191	1.9×10^4	296	2.9×10^4
Ni	0.002	495	2.5×10^5	596	3×10^5
V	0.002	137	6.8×10^4	240	1.2×10^5
Zn	0.01	6,621	6.0×10^5	9,277	9.3×10^5
Ca	400	6,145	15	9,727	23.7
Mg	1,350	10,281	7.62	17,108	12.7
Sr	8	35	4.32	64	7.9
Al	0.002	51.2	5.1×10^3	160	1.6×10^4
Ba	0.02	0.1	6.4	0.3	13.2
Mn	0.0002	9.1	4.5×10^3	18.6	9.3×10^3
U	0.003	845	2.8×10^5	705	2.3×10^5

[a] Adsorption: Resin 50 cm^3, columnar (upflow). Elution: 0.5 mol·dm^{-3} H_2SO_4 as an eluant.

[b] T. Shigematsu, Kaisuishi, *21*, 221 (1968).

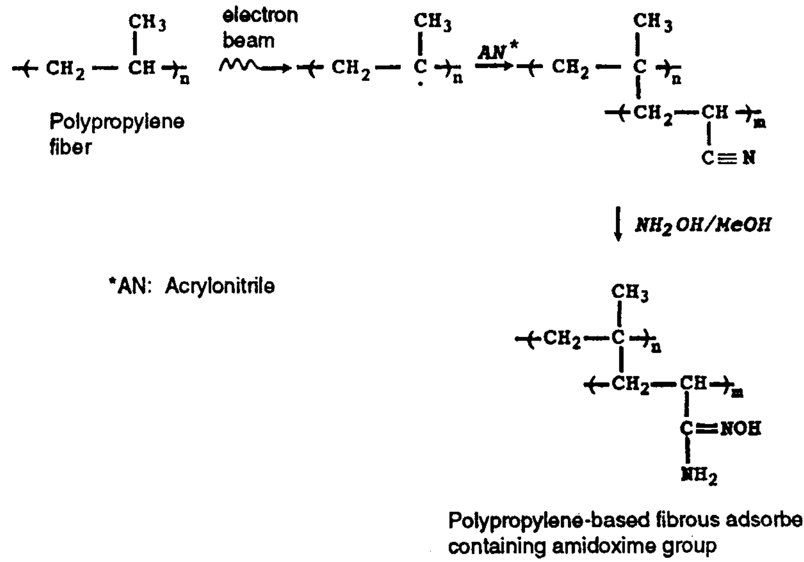
[c] Concentration factor (CF) = metal ion adsorbed/concentration of metal ion in seawater [(mg/kg-resin)/(mg/dm^3)].

FIBROUS AMIDOXIME-ADSORBENTS

The fibrous amidoxime-adsorbents prepared from commercial poly(acrylonitrile) fiber were found to be promising because of their large exchange surface and high uranium uptake (21, 34). The fibrous adsorbents, however, exhibited poor mechanical stability when they had contact with alkali. Fibrous adsorbents containing amidoxime group were recently synthesized by radiation-induced grafting to obtain a high mechanical strength. A number of fibrous adsorbents have been prepared by radiation-induced graft copolymerization of acrylonitrile onto polyethylene or polypropylene fiber (35–39). As an example, the preparation route of a polypropylene-based fibrous adsorbent containing amidoxime group by radiation-induced grafting is shown in Scheme 2.

In addition, Saito et al. (40–42) suggested a novel adsorption system using hollow-fiber adsorbents containing amidoxime groups by radiation-induced grafting of acrylonitrile onto polyethylene hollow-fiber. These adsorbents are proposed for application in uranium recovery from seawater since they exhibited a good performance of an adsorption bed with a low flow resistance.

We prepared hollow-fiber adsorbents from polyacrylonitrile hollow-fiber. On the basis of data obtained by solid-state (CP-MAS) ^{13}C-NMR



SCHEME 2 Preparation of fibrous adsorbent containing amidoxime group.

spectroscopy, we tried to elucidate the chemical structure of hollow-fiber adsorbents (43).

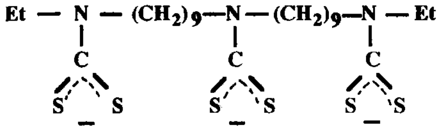
Kobuke et al. reported on a composite fiber adsorbent with superior adsorption characteristics (44, 45). Katoh et al. (46) prepared ball-type adsorbents by entangling chopped fibers during amidoximation.

OTHER ADSORBENT SEARCH AND DEVELOPMENT ENDEAVORS

Tabushi et al. (47–49) reported on several "uranophiles" (**6–9**) having extraordinarily large formation constants as well as large selectivities with uranyl ion.

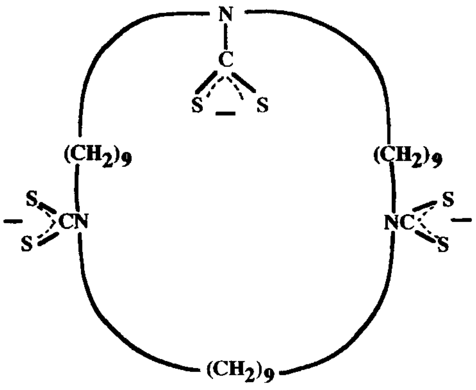
Cyclic $\left[\!-(CH_2)_5-COCH_2CO-\!\right]_3$ *(6)*

Cyclic $\left[\!-(CH_2)_8-C(CO_2H)_2-\!\right]_3$ *(7)*



$$\mathrm{Et}-\underset{\substack{|\\ C\\ S\;\;S^{-}}}{N}-(CH_2)_9-\underset{\substack{|\\ C\\ S\;\;S^{-}}}{N}-(CH_2)_9-\underset{\substack{|\\ C\\ S\;\;S^{-}}}{N}-\mathrm{Et}$$

(8)



(9)

Polymer adsorbents functionalized toward these uranophiles were demonstrated to recover uranium successfully from seawater (50, 51).

Some attempts have been made to recover uranium from seawater by using a variety of biomass and such biological substances as chlorella and streptomyces which showed high adsorption abilities (52–56). Accordingly, some synthesized and immobilized tannin compounds were found to recover uranium from seawater with high efficiency (57, 58). In Refs. 52–58 the same authors investigated the adsorption abilities of various plant wastes for uranium recovery (59). Ueda et al. (60) synthesized a novel adsorbent containing oligo-peptides for efficient recovery of uranium. Muzarelli et al. (61) reported on the synthesis of chitosan and its ability for the recovery of uranium from seawater.

SYSTEM STUDIES FOR THE UTILIZATION OF NATURAL SEAWATER MOTIONS

A number of processes using solid adsorbents were tested for the recovery of uranium from seawater. Earlier efforts focused on actively pumped systems. However, the energy consumed for pumping seawater accounts for the major part of the total cost of yellow cake production from seawater. Attention has recently been paid to passive contactor systems utilizing natural seawater motions (62–69). Nobukawa proposed towing and/or mooring systems for amidoxime fiber balls, and he reported that the results of trials with these systems were promising (70).

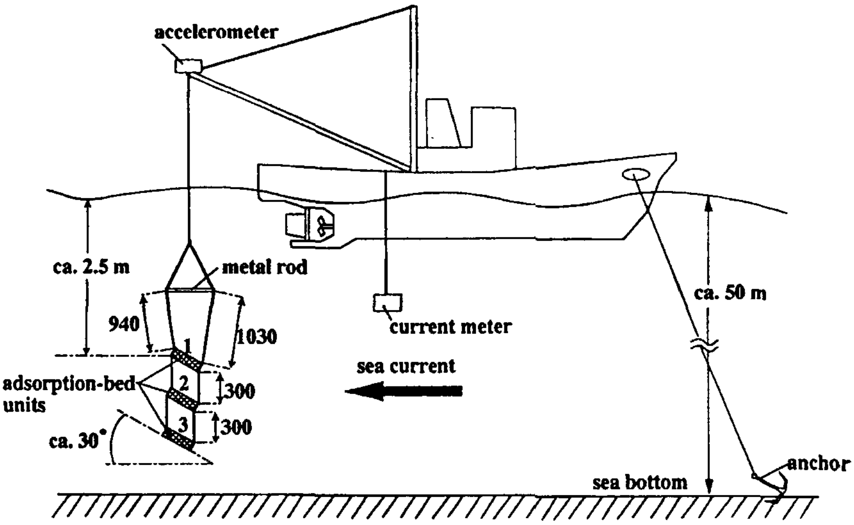


FIG. 8 Mooring operation utilizing natural sea current and wave motions.

Most recently, we have tested the performance of lightly crosslinked highly porous amidoxime-resins in uranium adsorption systems utilizing natural seawater motions (71). Figure 8 shows the assembled adsorption-bed units immersed into the sea current from a ship. We reported that the efficiency of uranium uptake became higher with a decrease in the thickness of the packing layers, indicating the important role of fluidization of the resin particles. We concluded that spherical amidoxime resins can be successfully applied to the recovery of uranium from seawater by mooring operations in sea currents and/or wave motions.

CONCLUSIONS

This paper presents efforts for the development of chelating polymers to be employed in "uranium recovery from seawater" systems. Adsorption capacity, adsorption rate, and durability are the most important parameters in the analysis of these systems. Polymeric amidoxime-resins are shown to take up uranium from seawater rapidly and to have a stronger preference for uranium and several other metal ions than for alkali and alkaline-earth metal ions. Other advantages include cheapness, good physical and chemical stability, and high adsorption capacity.

An adsorption system utilizing natural sea current and wave motion has been identified as being important in reducing system capital and operating costs for uranium recovery from seawater.

In conclusion, efforts should be continued to produce more powerful chelating polymers with high adsorption rates and better physical strengths for a successful "recovery of uranium from seawater" technology (72).

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