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V. Ramachandhran[a]; Shaji C. Kumar[a]; M. Sudarsanan[a]
[a] Desalination Division, Bhabha Atomic Research Center, Mumbai, India

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PREPARATION, CHARACTERIZATION, AND PERFORMANCE EVALUATION OF STYRENE-ACRYLONITRILE-AMIDOXIME SORBENT FOR URANIUM RECOVERY FROM DILUTE SOLUTIONS

V. Ramachandhran,[1,*] Shaji C. Kumar,[2] and M. Sudarsanan[2]

[1]Desalination Division and [2]Analytical Chemistry Division, Bhabha
Atomic Research Center, Mumbai 400 085, India

ABSTRACT

Styrene-acrylonitrile copolymer was synthesized by emulsion polymerization using persulphate as the radical initiator. The polymer obtained was functionalized by reacting with hydroxylamine to convert the nitrile group into amidoxime. The amidoxime sorbent was characterized in terms of moisture uptake elemental composition, IR spectra, thermal stability and exchange capacity. The sorbent, obtained in particulate form, was investigated for its sorption properties with respect to uranium from uranyl nitrate solution in batch as well as in column conditions. This paper will present the preparation, characterization and performance evaluation with respect to uranium sorption as a function of contact time, uranium concentration, and temperature. The effect of excess sodium chloride and free dissolved chlorine on uranium uptake are also included. The potential of the sorbent for uranium recovery and other heavy metal from sea water is ascertained.

Key Words: Uranium; Separation, Amidoxime; Polymer; Sea water

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*Corresponding author.

INTRODUCTION

Over the past three decades, the extraction of uranium from sea water has attracted attention the world over in view of the anticipated depletion of uranium resources in the near future. This is evident from the exhaustive survey, made on the processes, their feasibility and economics [1-10], and a few international meetings held on this topic. Even though the reported concentration of uranium in sea water is very low, it constitutes a significant source due to the large quantity of sea water available. A prominent dissolved form of uranium in sea water is a stable tetra valent uranyl carbonate anionic complex [11]. The adsorption method using solid adsorbents is promising for uranium recovery from sea water with regard to economic and environmental impact. Among the inorganic sorbents, hydrous titanium oxide (HTO) has shown high selectivity for uranium [2]. Inorganic, though inexpensive and easily available, have poor mechanical strength. When made stronger with binders, the loading capacity decreases. They also have an disadvantage in that they cannot be prepared in any form, at times not even as granules. On the other hand, organic sorbents, though relatively more expensive, have the advantage in that they can be prepared in any form like, granules, sheets, fiber, etc. Polymeric materials having polyfunctional groups such as carboxylic, amide, nitrile, iminodiacetic acid, amidoxime, and ammonium groups, etc., not only possess good hydrophilic properties, but also have good ion exchange properties which make them suitable for metal recovery from aqueous solutions [12, 13]. Efforts to develop chelating polymers for uranium recovery from seawater is reported in detail [14]. Various polymeric adsorbents have been designed and successfully demonstrated for selective sorption of uranium. Resins based on polyamines polyethyleneimine and polyallyl amine are reported for uranium recovery [15-17]. Chelating resins functionalised with dithiocarbomate is reported for uranium recovery [18]. A calixarene moity immobilized on a polyethyleneimine support was recently reported for uranium recovery [19]. Complexation of uranyl ions in aqueous solution was reported using porous polymers containing primary amino groups [20]. Polyhydroxamic acid also shows chelating properties towards heavy metal ions like iron and uranium. Resins, hollow fibers, and non-woven fabrics containing amidoxime groups were extensively studied for accumulating uranium from seawater [21-28]. Water insoluble polymers with amidoxime ($—C(NH_2) = N(OH)$) functional group are known to accumulate uranyl ions without carbonate ions by ligand displacement reaction. This paper describes the preparation, characterization and uranium uptake performance from dilute solutions, for amidoxime sorbent anchored on styrene acrylonitrile polymer, by chemical route.

EXPERIMENTAL

Synthesis of Styrene–Acrylonitrile Copolymer

Styrene stabilized with 4-t-butylcatechol and acrylonitrile stabilized with 4-methoxy phenol were locally procured and were purified by solvent extraction using 10% NaOH solution twice, at a volume ratio of 5:1 (monomer to caustic). Purified monomers were used within two weeks of purification. Polymerization reaction was carried out in emulsion mode using potassium persulphate as initiater and sodium lauryl sulphate as emulsifier under nitrogen atmosphere at 60°C. After polymerization, the emulsion was broken by addition of 2 g of NaCl followed by acidification by H_2SO_4. The coagulated polymer (SAN) was filtered, washed in hexane and vacuum dried.

Amidoxime Conversion

The SAN polymer sample was reacted with hydroxylamine at 80°C for 6 hours to convert the nitrile group into amidoxime group. For carrying out the reaction, hydroxylamine hydrochloride in 1:1 methanol–water mixture was taken and the pH was adjusted to 7 by adding KOH solution. The amidoxime derivative (SANAO) obtained was filtered, washed with demineralized water and vacuum dried. The reaction scheme of SANAO synthesis is given in Figure 1. The extent of conversion of nitrile group into amidoxime group was limited to around 55%. The conversion increases with increase in reaction time and it was found that the conversion reaches a maximum of 55% in 6 hours time.

Characterization

IR spectra of styrene-acrylonitrile polymer (SAN), styrene-acrylonitrile-amidoxime polymer (SANAO) and styrene-acrylonitrile-amidoxime polymer (SANAO-U) with sorbed uranium are taken using KBr pelletization . DTA, TGA spectra of samples were recorded in air at a heating rate of 10°C/min. The anion exchange capacity by equilibrating a known weight of the adsorbent with 0.1 N HCl for 24 hours and estimating the chloride released, when the acid treated sample was treated with subsequently, alkali. The details of the procedure are given elsewhere [14].

Uranium Sorption Studies

Batch and column experiments were carried out using uranyl nitrate solution to study the extent of uranium sorption as a function of monomer ratio, time of contact, uranium concentration, and temperature. In batch experiments, a known weight of the sorbent was equilibrated with a definite volume of uranyl nitrate solution in passive contact without any mechanical agitation. The sorbent, after a specified period of contact, were washed thoroughly and eluted using 1N HCl solution. Column experiments were carried out under identical conditions for a single flow rate. Uranium estimation was carried out spectrophotometrically using Arzenoazo III reagent at 662 nm.

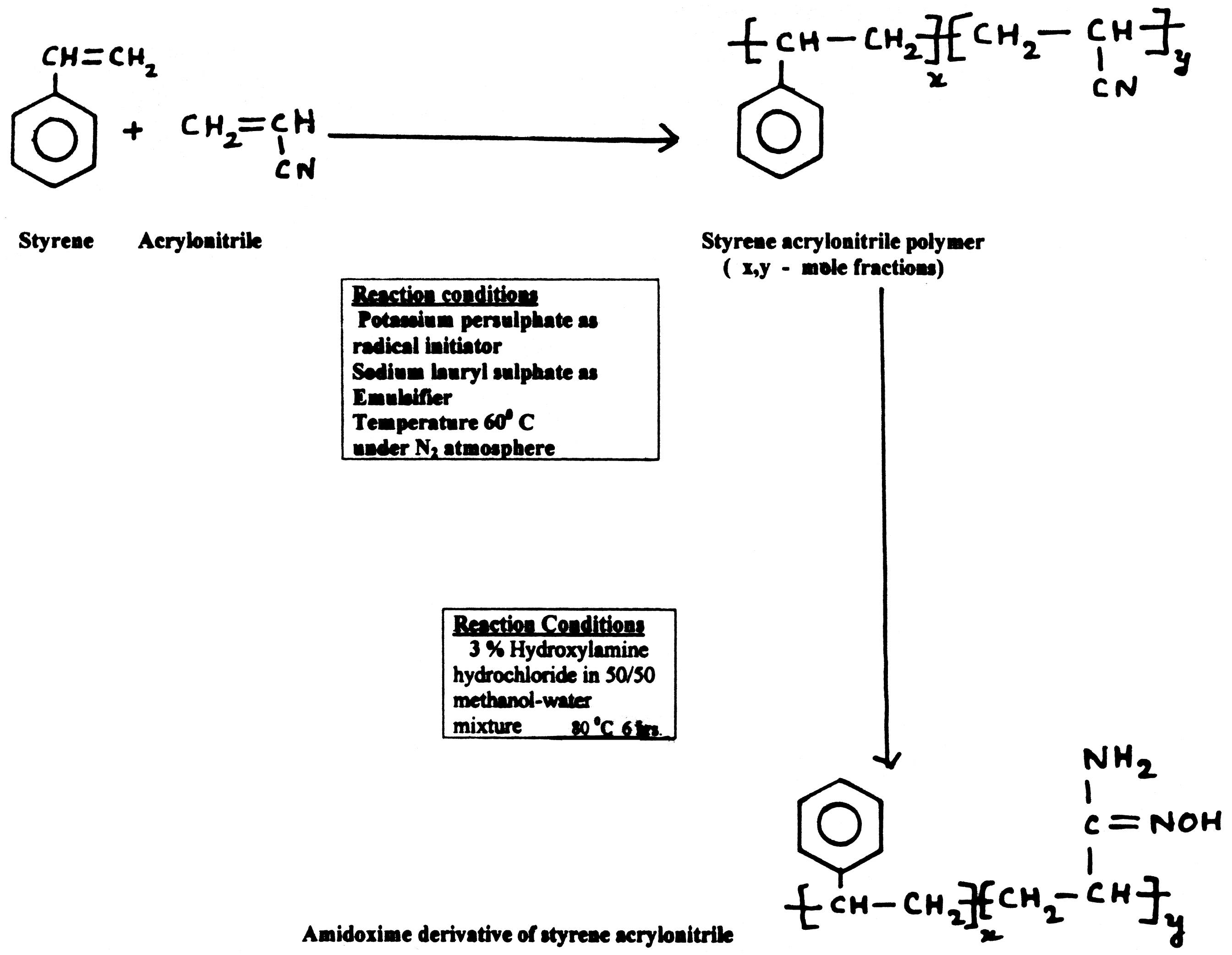

Figure 1. Reaction scheme for synthesis of amidoxime sorbent.

RESULTS AND DISCUSSION

Characterization

Three different polymer samples with varying styrene-acrylonitrile mole ratios, namely, 0.4/0.6, 0.3/0.7, and 0.2/0.8 were synthesized, funtionalized, and designated as SANAO 4060, SANAO 3070, and SANAO 2080, respectively. The characterization data for a typical sample (SANAO 3070) in terms of % moisture uptake, anion exchange capacity and elemental composition are given in Table 1. The % moisture uptake data for the amidoxime derivative has been found to be higher as compared to the base polymer indicating enhanced hydrophilic character due to incorporation of amino and hydroxyl groups. The experimentally observed elemental composition data for both the polymer and amidoxime derivative agree satisfactorily with respect to carbon and hydrogen. However the observed values are higher with respect to nitrogen and oxygen for both the samples, which could be due to the random nature of the copolymer, as well as partial oxidation of the polymer, forming carboxyl groups.

The IR spectra recorded for both the samples are given in Figure 2. In addition to the characteristic adsorption peaks, the amidoxime sample also has additional peaks at 3110-3400 cm^{-1} characteristic of NH and NOH stretching, 1640 cm^{-1} characteristic of double bond CN stretching, 1600 cm^{-1} characteristic of NH bending and 930 cm^{-1} characteristic of NO stretching vibrations. A prominent peak at 2234 cm^{-1} characteristic of triple bond CN stretching is found in the SAN sample which diminishes in intensity in the amidoxime derivative. It also indicates the partial conversion of nitrile group. The amidoxime groups are known to func-

Table 1. Physicochemical Properties of Styrene-Acrylonitrile-Amidoxime Sorbent

S. No.	Sample	Property		
1		*Elemental Composition (wt.)*		
		Element	*Obsd.*	*Calc.*
	SAN	Carbon	83.09	84.03
		Hydrogen	7.11	7.05
		Nitrogen	9.7	8.89
		Oxygen	—	—
2		Moisture uptake (at 85% RH, &20 °C)	0.82	
3		Carbon	70.3	71.93
		Hydrogen	7.5	7.36
	SANAO 3070	Nitrogen	11.8	9.4
		Oxygen	10.4	11.31
4		Moisture uptake (at 85% RH & 20°C)	2.94	
5		Anion exchange capacity (m.equi. / g)	1.17	

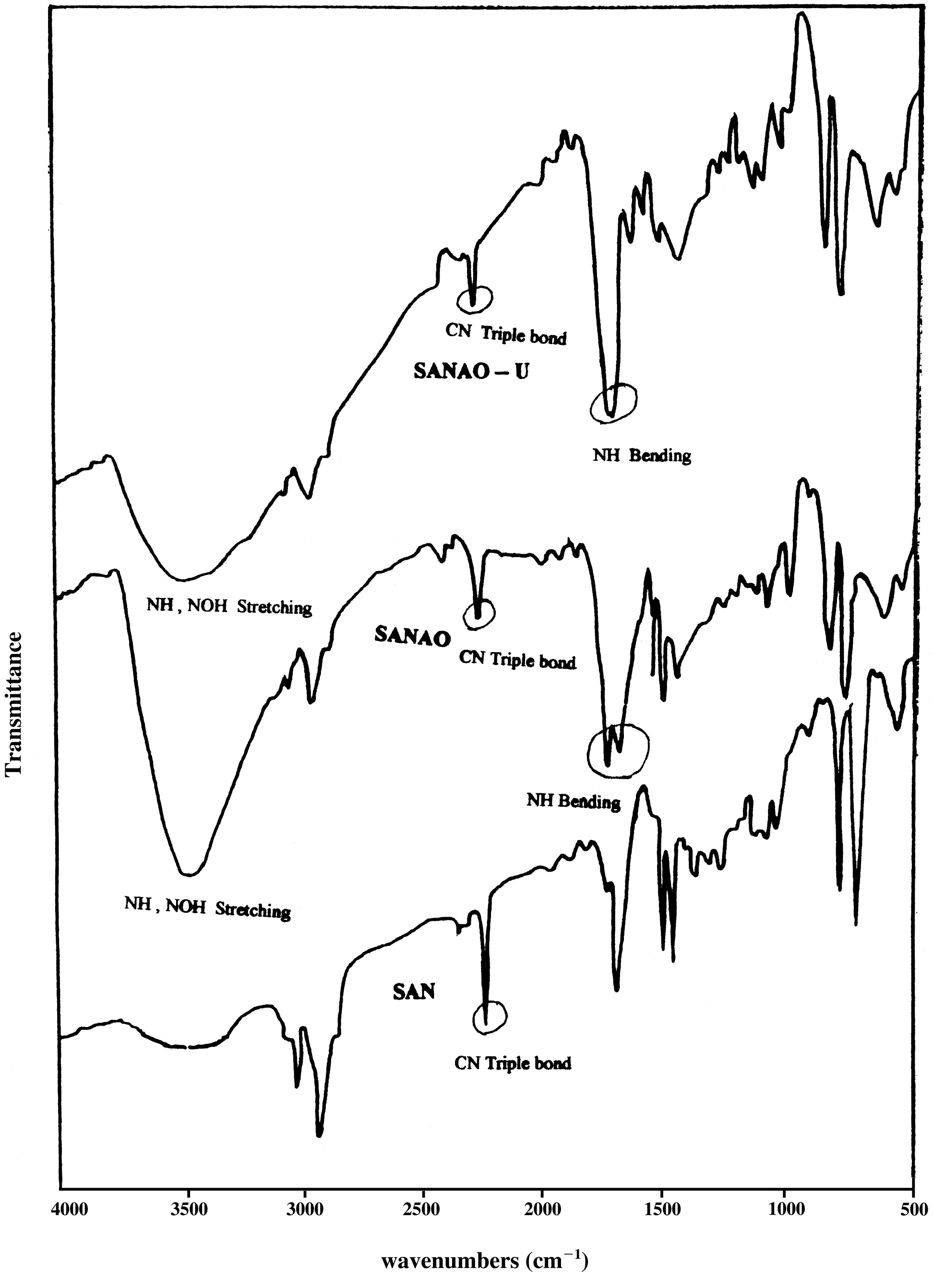

Figure 2. IR spectra of SAN and SANAO samples.

tion as monodentate nonchelating neutral ligands and bind uranium through oxime. Four amidoxime groups coordinate with uranium. Nitrogen atoms of the amide groups are not involved in the bonding.

The DTA and TGA spectra of SAN and SANAO samples are given in Figures 3 and 4, respectively. Both the samples show an endothermic peak at around 300 to 350°C. The peak appears at a lower temperature of around 300°C, whereas it appears at around 350°C for the funtionalized polymer. The % weight loss also shows a similar trend with SAN sample registering a relatively higher weight loss as compared to the SANAO sample in the temperature range measured. The relative higher thermal stability of the funtionalized sample may be due to the polar character and intermolecular and intramolecular hydrogen bonding.

Uranium Sorption Studies

Monomer Mole Ratio

Uranium sorption characteristics were evaluated for three adsorbent samples, namely, SANAO 2080, SANAO 3070, and SANAO 4060 using uranyl

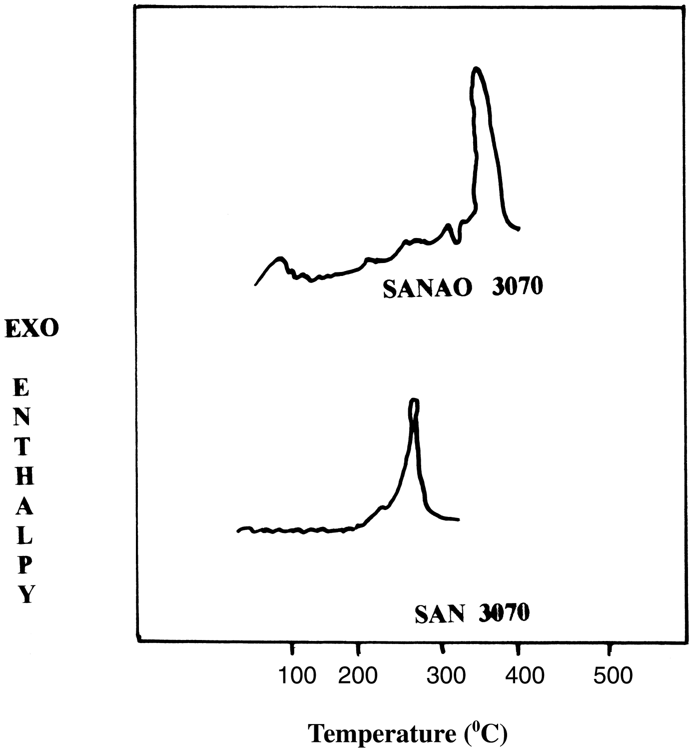

Figure 3. DTA spectra of SAN and SANAO polymers.

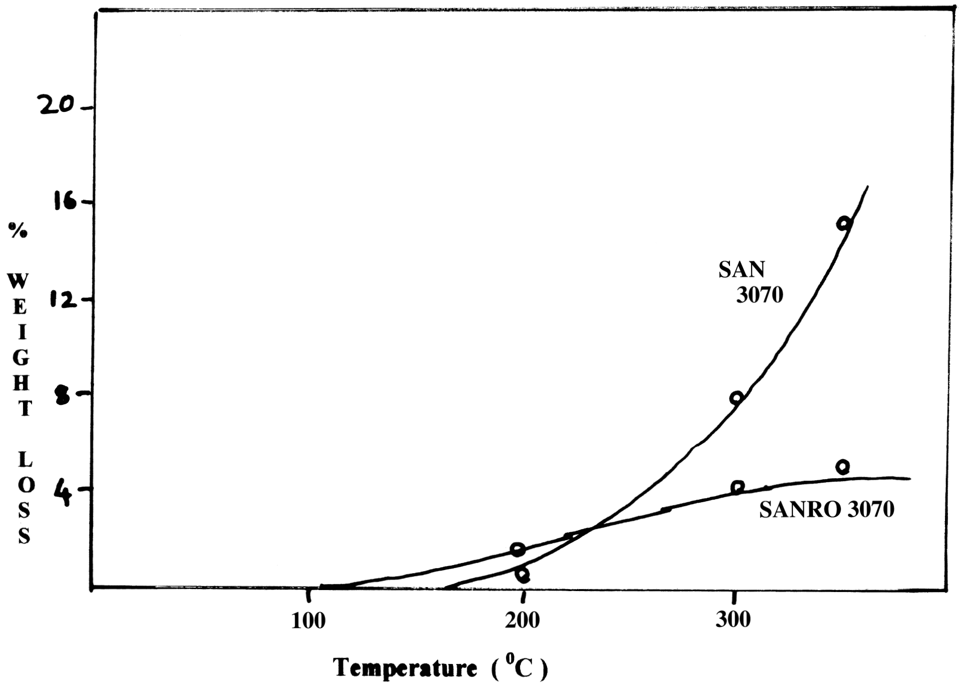

Figure 4. TGA spectra of polymer samples.

nitrate solutions (0.193 mM) under batch conditions. The uranium uptake (expressed as µg/g) is given in Table 2. The pH of the equilibrating uranyl nitrate solution was 2.18 and the pH of the uranyl nitrate solution after equilibration was found to 2.11 which indicates the release of H^+ ions inexchange for uranyl ion. The uranium uptake data and the uranium concentration in the aqueous phase at equilibrium is also given in Table 2. It can be seen that the uranium uptake decreases with a decrease in acrylonitrile molefraction in the polymer, which is as expected.

Table 2. Uranium Sorption Data for Varying Mole Ratio

	Mole Ratio		
S. No.	Styrene	Acrylonitrile	Uranium Uptake (mg/g)
1	0.2	0.8	482.4
2	0.3	0.7	405.6
3	0.4	0.6	309.4

Weight of the adsorbent: 0.5 g.
Adsorbate: 50 mL of 0.193 M uranyl nitrate.
Contact time: 24 hrs.

Effect of Time on Uranium Extration

The effect of time on the uranium uptake for the SANAO sample for two different concentrations, namely, 0.193 mM, and 0.0193 mM, under batch conditions is presented in Figure 5. The uranium uptake is higher (852 μg/g) for higher uranium concentration and the uptake improves with time. The rate of uptake for the two different concentrations is found to be nearly identical to the ratio of uranyl nitrate concentrations employed. From the sorption data, equilibrium appears to have reached in about 6 hours time for the higher concentration, whereas it tends towards equilibrium for the lower concentration during the same time interval. The slower rate of attaining equilibrium may be attributed to the slow diffusion rate of uranyl ions to the chelating sites because of the reduced concentration gradient. It is worthwhile comparing the uranium uptake data obtained in this work with the data reported for other organic and inorganic sorbents. Hydrous titanium oxide prepared different methods are reported [29] to give uranium uptake values in the range of 760 to 1550 μg U/g Ti in natural seawater conditions. Most of the commercially available organic ion exchange resins having amidoxime functional groups are reported [1] to give uranium uptake values up to 1000 μg/g resin.

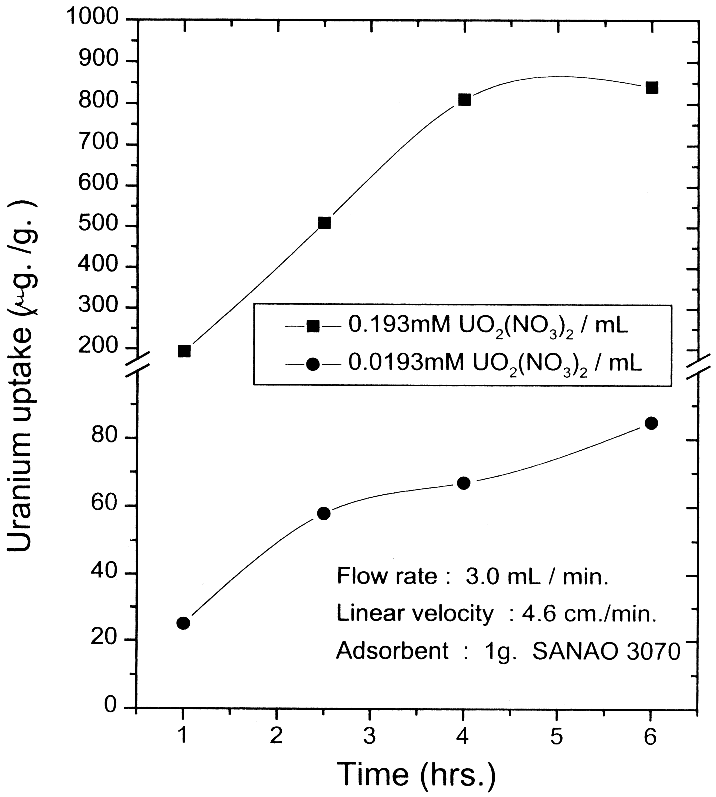

Figure 5. Uranium uptake as a function of time (column).

Resorsinol arsenic acid resin is reported [2] to give a uranium uptake value of 1010 μg/g resin. But, this resin was found to undergo slow hydrolytic attack on the carbon-arsenic bond resulting in a steady loss in capacity.

Comparison Between Column and Batch Experiments

Uranium uptake by the SANAO 3070 sample under batch and column conditions are compared in Figure 6, where uranium sorption data is plotted as a function of time for two different uranium concentrations, namely, 0.193 mM and 0.0193 mM (as uranyl nitrate) under batch and column conditions. It can be seen that uranium uptake is higher under column conditions during the time of contact studied for both the concentrations. This is understandable due to the continuous shifting of equilibrium, faster diffusion of uranyl ions to the chelating sites because of thinner boundary layers, and due to the higher concentration gradient under column conditions.

Effect of Temperature

The effect of the temperature of uranyl nitrate solution on the uranium uptake by SANAO 3070 sample was evaluated in the temperature range of 17°C to 50°C for 0.193 mM uranyl nitrate solution and for a specified period of contact.

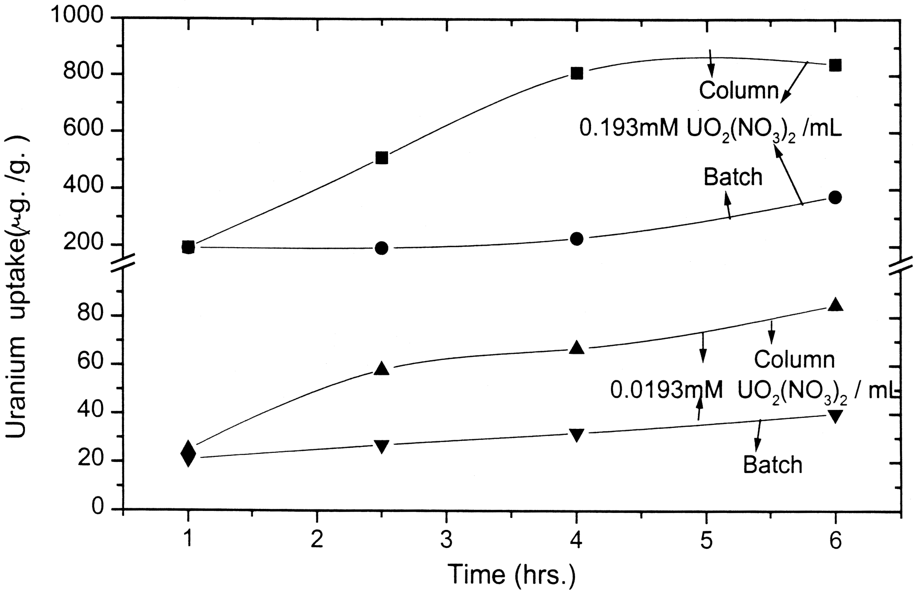

Figure 6. Comparison of uranium uptake between batch and column experiments.

Table 3. Uranium Sorption Data as a Function of Temperature

S. No.	Temperature (°C)	Uranium Uptake (μg/g)
1	17.5	272
2	30	419
3	41.2	491
4	50.8	579

Weight of the adsorbent : 0.5 g SANAO 3070.
Adsorbate: 50 mL of 0.193 M uranyl nitrate solution.
Contact time: 24 hrs.

The results are given in Table 3. It can be seen that the uranium uptake increases with a rise in temperaure from 272 μg/g. to 579 μg/g of adsorbent when the temperature is raised from 17.5°C to 50.8°C. The activation energy of uranium sorption was computed using Arrhenius equation from the above data and it works out to 5.6 K cals/mol indicating chemisorption as the principal mode of sorption as already indicated in the literature (15).

Effect of Presence of Other Metals

The effect of the presence of excess sodium chloride on the uranium uptake by the SANAO 3070 sample was evaluated under batch conditions. The uranium uptake data is presented in Figure 7, as a function of time of contact for two different concentrations of uranyl nitrate, namely, 0.193 mM and 0.0193 mM containing 0.598 M sodium chloride each. The uranium uptake data in the absence of sodium chloride is also shown in the figure for the sake of comparison. It can be seen that the presence of excess sodium chloride reduces the uranium uptake during the time of contact studied for both the uranyl nitrate concentrations. The possibility of the excess sodium leaching out the sorbed uranium from the sorbent or sodium ions competing with uranyl ion for sorption was explored separately, and it was found that neither excess sodium elutes the sorbed uranium nor sodium competes with uranium for sorption. The observed reduction in sorption was attributable to either osmotic shrinking of the adsorbent particles or due to ionic strength effect. The impact of the presence of other metals, like cobalt, chromium and copper on the sorption of uranium was studied. Equimolar solutions of uranyl nitrate with the above salt solutions are equilibrated for 24 hours. The uranium sorption data are given in Table 4. It can be seen that uranium sorption reduces almost in a similar manner The uptake of cobalt, chromium and copper is also given in Table 4. It can be seen that the uptake of copper and cobalt are that of uranium where as the uptake of chromium is lower than that of uranium.

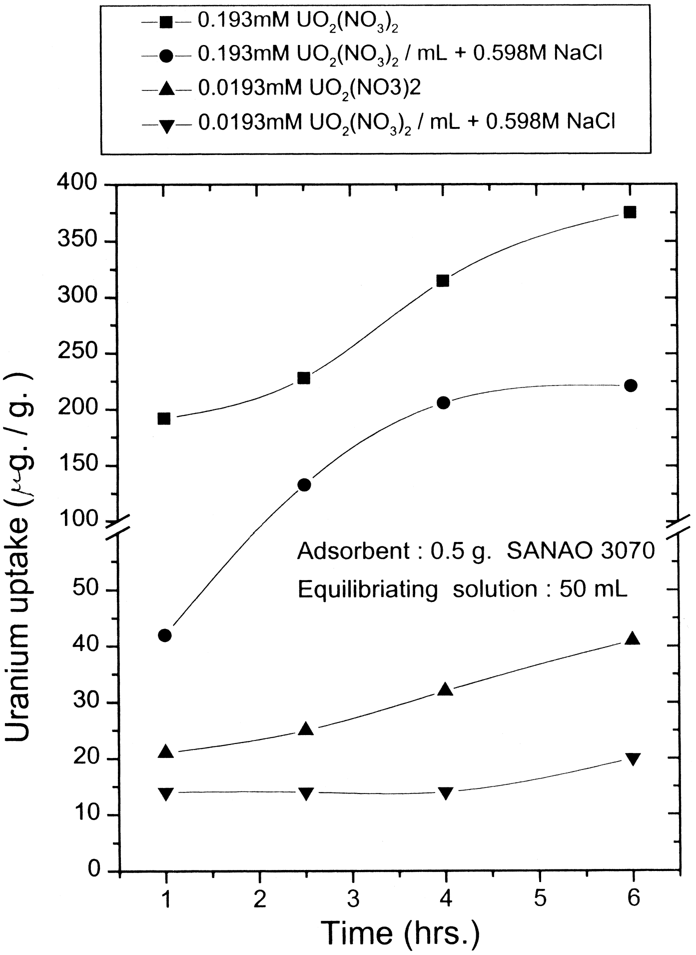

Figure 7. Uranium uptake as a function of time in presence of NaCl (batch).

Table 4. Uranium Sorption Data in Presence of Other Competing Metals

S. No.	Equilibrating Solution	Urianium Uptake (µg/g)	Metal Uptake (µg/g)
1	0.193 mM uranyl nitrate + 0.2 mM cobalt chloride	232.9	262.2
2	0.193 mM uranyl nitrate + 0.2 mM chromium nitrate	172.4	160.9
3	0.193 mM uranyl nitrate + 0.2 mM copper chloride	234.9	356

Weight of the sorbent: 0.5 g.
Volume of the equilibrating solution: 50 mL.
Contact time: 24 hrs.

Effect of the Presence of Dissolved Free Chlorine

The effect of free dissolved chlorine, which is normally dosed in sea water coolant circuits, to hinder biological growth, on the uranium uptake was studied for SANAO 3070 sample. The adsorbent sample was kept in contact with deionized water containing 1.4 ppm of free dissolved chlorine for varying periods of time. The uranium uptake for the exposed sample was evaluated using 0.193 mM uanyl nitrate solution which also contained free residual chlorine. The results are given in Table 5. It can be seen that uranium uptake has declined from 384 μg/g to 164 μg/g after 85 days of exposure to free residual chlorine indicating the damage to chelating groups on prolonged contact. No significant change in uranium uptake was however noticed up to 25 days of contact. The susceptibility of polymers containing nitrogen to free chlorine is well known. The performance decline is generally attributed to N-chlorination and subsequent cleavage of the chelating groups from the polymer backbone.

Repeated Use of Sorbent

The repeated usability of the sorbent was evaluated by collecting uranium uptake data with the same adsorbent sample SANAO 3070 under identical conditions by repeating sorption-elution cycles. The results are given in Figure 8. It can be seen that uranium uptake has declined from 447 μg/g to 327 μg/g after 10 cycles of sorption-elution. This accounts for 26% loss in the uptake capacity. The loss in sorption capacity in repeated use is attributed to the instability of cyclic imidoxime groups with repeated use of acid. For better uranium uptake the presence of cyclic imidoxime groups are found to be essential. However, it is reported that it is difficult to stabilize these groups by suitable physical or chemical modification of the polymer matrix.

Recovery of Uranium and Other Trace Metals from Sea Water

Recovery of uranium and other trace heavy metals from sea water by sorbent SANAO 3070 was evaluated under column conditions using gravity flow. A total

Table 5. Uranium Uptake in Presence of Trace Residual Chlorine

S. No.	No. of Days of Exposure	Uranium Uptake (μg/g)
1	0	384
2	25	374
3	85	165

Sorbent: 0.5 g SANAO 3070.
Adsorbate: 50 mL 0.193 mM uranyl nitrate solution containing 1.4 ppm residual chlorine.
Contact time: 24 hrs.

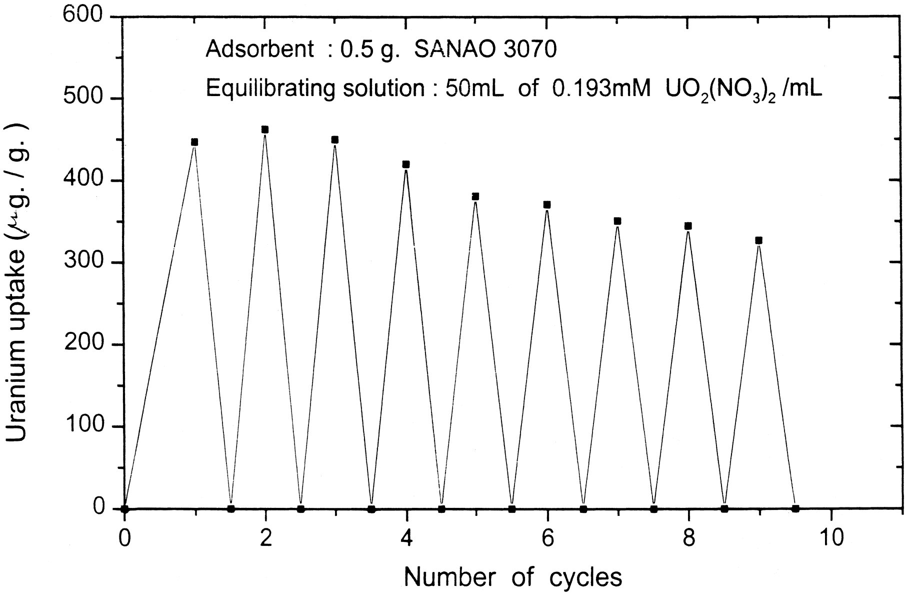

Figure 8. Uranium uptake under repeated use of adsorbent.

of 35.8 liters of filtered sea water was passed through a cylindrical glass column containing 2 g of adsorbent. The sorbed heavy metals, namely, Ti, V, U, Co, and Mo were evaluated and analyzed be ICP AES. The uptake and concentration factor obtained are given in Table 6. The concentration factor (CF in L/Kg) was computed from the ratio of concentration of heavy metals sorbed, to that present in sea water. It can be seen that the sorbent exhibits high selectivity not only for uranium but also for cobalt and vanadium as well. It has been reported [30]. That the uranium concentration factor is higher than alkaline earth metals, but lower than the concentration factors for other transition metals for amidoxime sorbents. The selectivity with respect to molybdenum and titanium are found to be poor. The

Table 6. Recovery of Uranium and Other Trace Heavy Metals from Sea Water

S. No.	Element	Conc. in Sea Water (μg/L)	Uptake (μg/g)	Concentration Factor (L/Kg.)
1.	Ti	1	0.15	150
2	U	3.3	14.55	4409
3.	V	1.9	4.8	2526
4.	Co	0.4	4.5	11250
5.	Mo	10	0.9	90

Sorbent: 2g SANAO 3070.
Adsorbate: 35.8 litres of natural sea water.
Average flow rate: 3.3 mL/min.

material is not very specific for sorbing uranium from seawater. The sorption of vanadium and cobalt can be considered as a valuable byproduct if amidoxime sorbents are considered for extracting uranium from sea water.

CONCLUSION

Styrene–acrylonitrile–amidoxime sorbent in particulate form was synthesized in the laboratory using potassium persulphate as radical initiator. The uranium uptake characteristics of this sorbent was evaluated for dilute uranyl nitrate solutions as a function of monomer mole ratio, time of contact, temperature, and concentration of uranyl nitrate solution used. The uranium uptake is higher for higher concentration though the rate of uptake for two different concentrations are nearly identical to the ratio of concentration of uranyl nitrate. A higher temperature of the equilibrating solution improves the uptake for a given contact time. The equilibrium appears to have been reached earlier for higher concentration of uranyl nitrate. The presence of excess sodium chloride hinders the uranium uptake. The presence of dissolved free chlorine is detrimental to the sorbent under prolonged exposures. The sorbent is reusable but repeated use reduces the sorption capacity. The sorbent selectively removes uranium, vanadium, and cobalt from natural sea water. The selectivity for titanium and molybdenum are found to be poor.

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