Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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B. Venkataramani[a]; S. B. Karweer[a]; R. K. Iyer[a]; G. M. Phatak[a]; R. M. Iyer[a]
[a] CHEMISTRY DIVISION, BHABHA ATOMIC RESEARCH CENTRE, BOMBAY, INDIA

To cite this Article Venkataramani, B. , Karweer, S. B. , Iyer, R. K. , Phatak, G. M. and Iyer, R. M.(1988) 'Removal of Aluminum(III)-Based Turbidity in Water Using Hydrous Titanium Oxide Dispersed in Ion-Exchange Resins', Separation Science and Technology, 23: 4, 473 — 487

To link to this Article: DOI: 10.1080/01496398808060717

URL: http://dx.doi.org/10.1080/01496398808060717

SEPARATION SCIENCE AND TECHNOLOGY, 23(4 & 5), pp. 473-487, 1988

Removal of Aluminum(III)-Based Turbidity in Water Using Hydrous Titanium Oxide Dispersed in Ion-Exchange Resins

B. VENKATARAMANI,* S. B. KARWEER, R. K. IYER,
G. M. PHATAK, and R. M. IYER

CHEMISTRY DIVISION
BHABHA ATOMIC RESEARCH CENTRE
TROMBAY, BOMBAY 400085, INDIA

Abstract

An adsorber consisting of hydrous titanium oxide (HTiO) dispersed in a Dowex-type ion-exchange resin matrix (designated RT resins) has been developed which is capable of removing Al(III)-based colloidal dispersions in the neutral pH condition. The effect of resin crosslinking, particle size, HTiO loading, turbidity level, and flow rate on the turbidity removal efficiency of RT resins has been studied. It is demonstrated that a train of columns comprising RT resin, H^+, and OH^- form of resins could be used for large-scale purification operations at high flow rates. These columns, apart from removing turbidity and associated radioactivity, can effectively remove dissolved uranium present in ppb levels when used for water purification in nuclear reactors.

INTRODUCTION

Stability of colloidal dispersions is due to the electric charge on the particles because of which they experience repulsive forces among themselves and also to the chemical nature of the surface of the colloid itself (*1*, *2*). In industrial water treatment plants the aqueous suspensions are destabilized and coagulated by the addition of hydrolyzable metal ions like Al(III). Excess of these hydrolyzable metal ions poses problems when treated water is used for ion-exchange purification or in boilers.

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*To whom correspondence should be addressed.

A similar but more serious problem of colloidal Al(III)-oxide-based suspensions has been experienced in nuclear reactors that use aluminum as a cladding material (*3*, *4*). Under the effect of nonisothermal conditions and dissolved oxygen, corrosion of aluminum led to the formation of stable colloidal oxide of Al(III), appearing as turbidity in the cooling water. The colloidal suspension has been found to contain recrystallized gibbsite particles (*3*, *4*). The colloidal oxide, due to its ion exchange and adsorption properties (*5*), acts as a carrier for radioactive nuclides, like uranium and fission products, present in water. This poses problems of activity transport in coolant water and consequent man-rem problems.

Thus clarification of Al(III)-based turbidity in water is of importance in industrial operations and in particular nuclear reactor technology (*3*, *4*). Since the colloidal suspensions cannot be filtered easily, a method to overcome this problem is to coagulate the colloidal particles by addition of destabilizing agents and filtering them. Hydrolyzed aluminum ion is known to adsorb on a variety of hydrophobic and hydrophilic colloidal surfaces, including mineral particles, oxides, etc. (*6*). Many of these processes are batch techniques that cannot be coupled to flowing systems and are not suited for reactor coolant systems. Ion-exchange purification, which in principele exposes electrically charged surface (polyelectrolyte) to colloidal hydrolyzed Al(III), does not destabilize these colloidal suspensions (*3*, *4*). Physical techniques like distillation and centrifugation, which are energy intensive, could mitigate this problem (*3*, *4*).

With a view to developing a system capable of destabilizing, coagulating, and adsorbing Al(III)-based turbidity on a solid matrix under flow conditions, we have studied the clarification properties of hydrous titanium oxide (HTiO) loaded onto ion-exchange resins. HTiO was chosen as the destabilizer because titanium oxide is known to coagulate Al(III)-based turbidity or Al_2O_3 itself (*6*–*10*). Moreover, HTiO is known to be a good adsorber for uranium in trace level concentrations (*11*) and other radioactive nuclides (*12*). We have taken advantage of the fact that inorganic precipitates, like HTiO, can be incorporated in an ion-exchange resin matrix (*13*, *14*) without change in their surface properties, like ion adsorption. As the main thrust of the study was to develop a system that could be coupled to a flow system, the performance of HTiO dispersed in ion-exchange resins in clarifying Al(III)-based turbidity under various hydrodynamic conditions is reported in this paper.

EXPERIMENTAL

Preparation of the Adsorber

Hydrous Titanium Oxide

Pure HTiO was prepared synthetically by the addition of NaOH to an acidified Ti sulfate solution. After aging the precipitate for 2 days, it was filtered, washed, and air dried (sample designated T1). Another sample of HTiO used for this purpose was the commercial variety marketed by Applied Research, Sprl, Belgium (designated T2). Both these samples contained Na^+, as a result of which the water in contact with them was alkaline (pH ~ 9). Incorporated Na^+ from both T1 and T2 was leached out using dilute $HClO_4$, until the water of suspension of the oxide had a neutral pH (samples designated T1T and T2T, respectively).

HTiO Loaded Ion-Exchange Resins

The H^+ form of the Dowex-type strong ion-exchange resin was converted to the Ti(IV) form by treating with acidic $TiCl_4$ solution. After washing the resin free of excess acid and Ti(IV) with water, it was treated with 1 *M* NaOH which precipitated HTiO in the resin matrix, and the resin was converted to the Na^+ form (single-step loading). The HTiO-loaded resin could again be treated with $TiCl_4$ solution to convert the resin to the Ti(IV) form, and HTiO was precipitated in the matrix using NaOH (multistep loading). HTiO loading was done in a batch process by contacting $TiCl_4$ solution (about 20 mg Ti per g resin) for a minimum of 1 h and precipitating the HTiO using 1 *M* NaOH (contact time about 1 h). The resins thus prepared are designated RT resins. In the present work, both single-step loaded and two-step loaded RT resins were used.

As in the case of pure HTiO samples (for example, T1 and T2), the RT resins were treated with dilute $HClO_4$ until the pH of the aqueous solution in contact with RT resin was 6–7.

The following resins were used for preparing RT resins: Dowex 50WX4, 50-100 BSS mesh; Dowex 50WX8, 50-100 BSS mesh; Indian Resin (Dowex type), 8% crosslinking and 20-50 BSS mesh.

The amount of Ti(IV) loaded on the resin was estimated either by repeated leaching of the Ti(IV) from RT resin using 2 *M* H_2SO_4 or incinerating the resin and estimating the Ti(IV) present in the residue. Ti(IV) was estimated colorimetrically using H_2O_2 at 410 nm (*15*). Values of Ti(IV) determined by both techniques agreed within the limits of experimental error. Ti(IV) content is expressed as % Ti present in resin dried at 100°C to constant weight.

Al(III)-Based Trubid Solutions and Estimation

Colloidal hydrolyzed Al(III) suspensions were prepared by neutralizing a solution of $AlCl_3$ to pH 6.1 to 6.3 using NH_3. Although this approach gave solutions with permanent turbidity, it was not satisfactory in large-scale preparations and is not representative of the true situation since it contains an additional electrolyte concentration.

Turbidity was generated on a large scale by striking an arc between two aluminum electrodes dipped in dimineralized water at ~60 V and passing about 30–40 A current. The turbid solution was later aged at 60°C for 48 h. The pH of the aqueous suspension was ~6.

The performance of RT resins was also tested with turbid reactor coolant water and in the presence of traces of uranium and other radioactive nuclides. The conductivity of the turbid solution was <1 μs/cm and pH ~6.

Turbidity was estimated using a turbidity meter with standard silica solutions or spectrophotometrically at 400 nm with a known concentration of Al(III)-based turbid solutions as standards. Turbidity values are expressed as ppm on a silica scale.

Turbidity removal efficiency was calculated as the percent of the initial turbidity. From a knowledge of the total volume of the turbid solution used and the amount of turbidity removed, the capacity of the RT resin column was calculated.

The performance of RT resins was tested in a recirculation system in which the turbid solution from a reservoir was pumped through the column using a peristaltic pump. By using a mercury manometer, the pressure drop across the adsorber column was monitored during the run. The adsorber column consisted of RT resin, H^+, and OH^- form of the resin packed in that sequence from the top.

RESULTS

Adsorption of Al(III)-based turbidity by HTiO alone is given in Table 1. Notable features of the results were that a slight alkaline condition

TABLE 1
Adsorption of Al(III)-Based Turbidity on HTiO Oxide (0.4 g; solution, 25 mL; time, 2h; initial pH, 6.2)

Oxide	Mesh size	Final pH	% Adsorption
T1	+60 BSS	8.2	96
T1T	+60 BSS	6.3	26
T1T	−100 BSS	6.7	51
T2	30–150	7.9	98
T2T	30–150	6.2	31

improved the adsorption property, and reducing the particle size also enhanced the adsorption. Because a column containing fine HTiO particles would lead to problems like pressure drop, choking, etc., the idea of dispersing HTiO inside ion-exchange resins was resorted to.

When a Al(III)-based turbid solution at pH ~6 was passed through a column of RT resin, 20% of the turbidity was removed and the pH of the outlet solution was ~9. Passing this effluent through a column of the H^+ and OH^- form of the resin clarified the solution completely, and the pH of the solution was near neutral.

After passing Al(III)-based turbid solution containing various fission products and other radioactive nuclides through a 3-tier column of RT resin, H^+, and OH^- form of the resin, the activity distribution in the 3 columns was checked. 65% of the total radioactivity was found on the H^+ form of the resin, 29% on the RT resin, and 7% on the OH^- form of the resin. It appears that RT resin alone cannot clarify the Al(III)-based turbidity but it does modify the turbid solution which is subsequently adsorbed on the H^+ form of the resin. In addition, the H^+ and OH^- forms of the resins also help to maintain the neutral pH of the solution.

When the columns of the H^+ and OH^- forms of the resins are used as one column of mixed bed, turbidity started appearing in the effluent in the course of time. However, no such breakthrough of turbidity occurred when the H^+ and OH^- forms of the resins are used as an independent column, with the OH^- form succeeding the H^+ form. This could be related to the random distribution of H^+ and OH^- forms of the resin in the mixed bed, as a result of which the turbidity was adsorbed to the bottom of the column, unlike the independent column.

A column of the H^+ and OH^- forms of the resin alone (without the RT resin column), either in the sequential bed mode or the mixed bed mode, did not remove turbidity. A mixed bed of RT resin, H^+, and OH^- form of the resin also did not remove any turbidity.

All the results reported in this section were done with a 3-tier column of

RT resin, H^+, and OH^- form of the resin packed in that sequence from the top of a column.

The effect of HTiO loading on the turbidity removal efficiency and the capacity of RT resins are shown in Figs. 1A and 1B, respectively. 2–3% Ti loading appeared to be the optimum loading. Turbidity removal efficiency and capacity were better if the same amount of Ti was loaded in 2 steps instead of as a single step.

The effects of mesh size and crosslinking of the RT resins on turbidity removal efficiency are shown in Table 2. The efficiency increases with finer mesh-size RT resins and less crosslinked resins.

The performance of 20-50 BSS mesh RT resin at two turbidity levels as a function of time is given in Fig. 2. Under the same conditions, the turbidity removal efficiency was higher at a lower turbidity level than at a higher turbidity level. In terms of long-term operation, this trend is advantageous in that RT resins function with better efficiency as the purification process proceeds and the turbidity level decreases.

The effect of flow rate on turbidity removal efficiency is shown in Fig. 3. The performance of finer mesh RT resin was better than that of coarser RT resin. The efficiency, however, decreased with an increase in flow rate, and was more pronounced with finer mesh-size RT resins.

The height of RT resin column had a marked effect on the turbidity removal efficiency; the deeper the bed, the more is its capacity to remove turbidity (Fig. 4).

The efficiency of the RT resin column, when operated intermittantly and when turbidity levels were changed, was also studied. Typical results of one such run are given in Fig. 5.

A notable feature of the experiment depicted in Fig. 5 was that whenever the column was started afresh, or when fresh turbid solution was introduced, the turbidity already held in the column was not washed down and the column behaved as if it was a new one. This indicated that the turbidity already adsorbed did not migrate down the column on standing or when operated again.

TABLE 2
Effect of Mesh Size and Crosslinking of RT Resins on the Turbidity Removal Efficiency

% Ti	Mesh size	% Crosslinking	Flow rate (mL/min)	Capacity (ppm/mL)
5	30–50	8	12	7.4×10^4
5	100–120	8	12	8.4×10^4
4	50–100	4	25	9.5×10^4
4	50–100	8	25	7.1×10^4

The various resins were loaded on the column in the following sequence from the top: 2 cm 20-50 BSS mesh RT, 6 cm 1:1 mixture of 20-50 and 50-100 BSS mesh RT, 2 cm 50-100 BSS mesh RT (all 8% crosslinking), followed by 9 cm each of H^+ and OH^- forms of the resin (20-50 BSS mesh). The radiation field distribution on the column from the top to bottom was in the ratio 3:5:5:2:1. Although the magnitude of the radiation field increased at various sections of the column with time, the ratio of the radiation field was maintained. This indicated that the turbidity and radioactivity adsorbed at various levels did not migrate down on standing or during operation. The radiation field ratio also indicated that the performance of finer mesh RT resin was better than that of coarser mesh resin.

RT resin had good chemical stability. This was tested by contacting various RT resins with demineralized water at 60°C for 3 weeks and analyzing the Ti(IV) leached out. Less than 0.5 ppm Ti(IV) (below the detection limit in AAS) was found in the supernatant solutions.

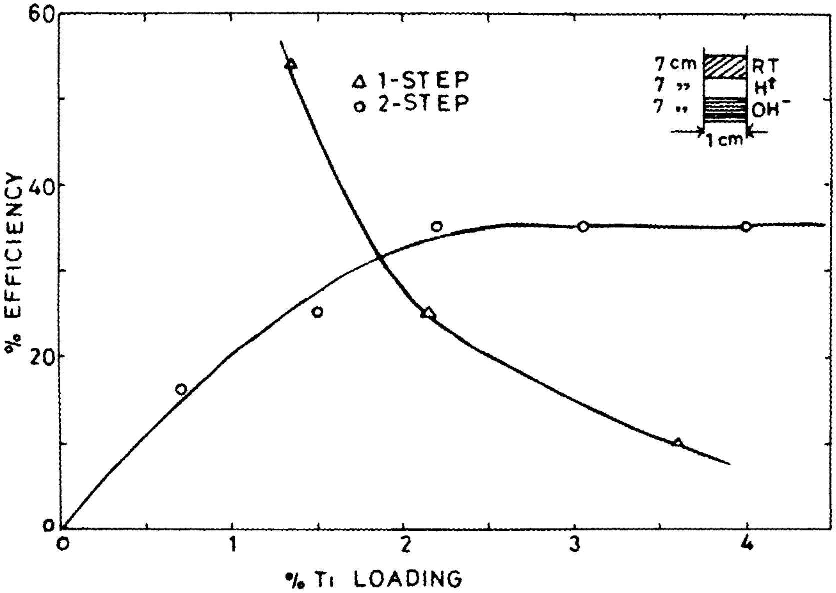
The performance of RT resins in large-scale operations was evaluated by preparing 50 L of RT resin using both 20-50 and 50-100 BSS mesh resins and loading 4% Ti (in two steps). The prepared resins were loaded in a column of 42 cm diameter to a height of 82 cm in the following sequence from the top: 15 cm RT resin 20-50 BSS mesh, 17 cm RT 50-100 BSS mesh, 15 cm RT 20-50 BSS mesh, and 15 cm (20-50 mesh) each of H^+ and OH^- forms of the resin. The column was operated at a flow rate of 40–45 L/min (hold up, 60 tons). This RT resin column removed 50% of both turbidity and radioactivity. A notable feature was the removal of dissolved uranium present in trace amounts (15 ppb). Figure 6 shows the performance of the RT resin column for 50 days of operation. The average uranium removal efficiency was 40–45%.

The salient features of the RT resins can be summarized thus: RT resins are easy to prepare and any type of ion-exchange resins can be used as the starting material. The resins are chemically stable. Resins of finer mesh size and of lower crosslinking have better turbidity removal efficiency. They can be operated at higher flow rates without much loss in their efficiency, and the efficiency was independent of the turbidity concentration. The turbidity and radioactivity retained on the column do not migrate down on standing or during operation. Although data have not been presented in this paper, it was found that the use of RT resin did not pose pressure drop problems.

DISCUSSION

Varieties of hydrolyzed species of Al(III) are known to exist at different pH's of the aqueous solution (*16*). In the pH region 6–7, the predominant

A


60
40
20
0
% EFFICIENCY
0
1
2
3
4
% Ti LOADING
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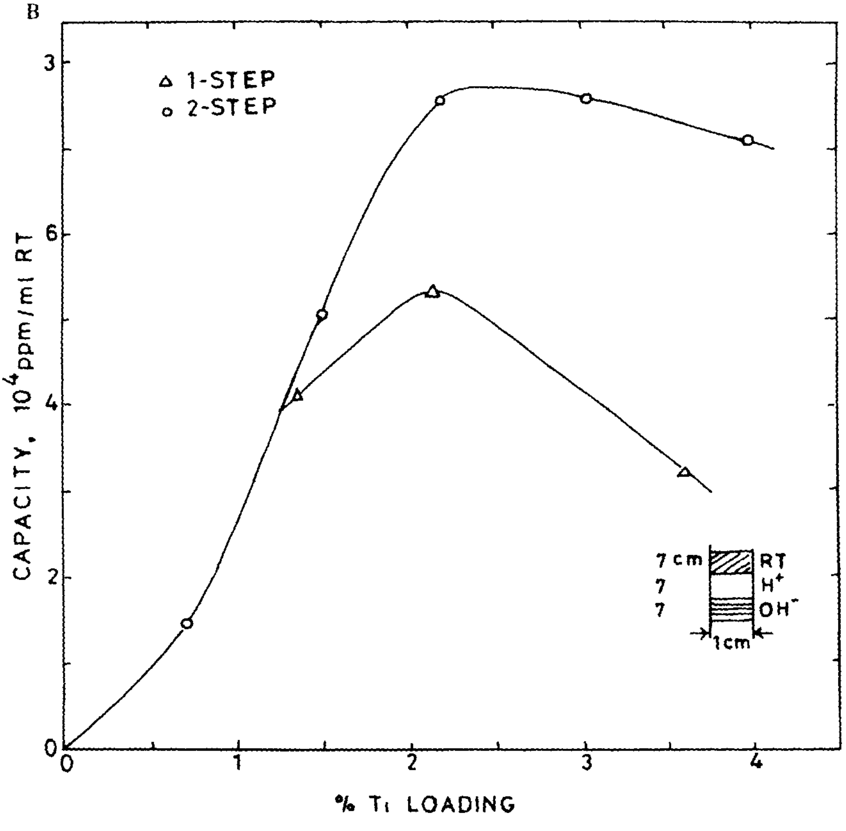
7 cm RT
7 ” H+
7 ” OH−
1 cm

B


3
6
4
2
0
CAPACITY, 10⁴ ppm/ml RT
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3
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7 H+
7 OH−
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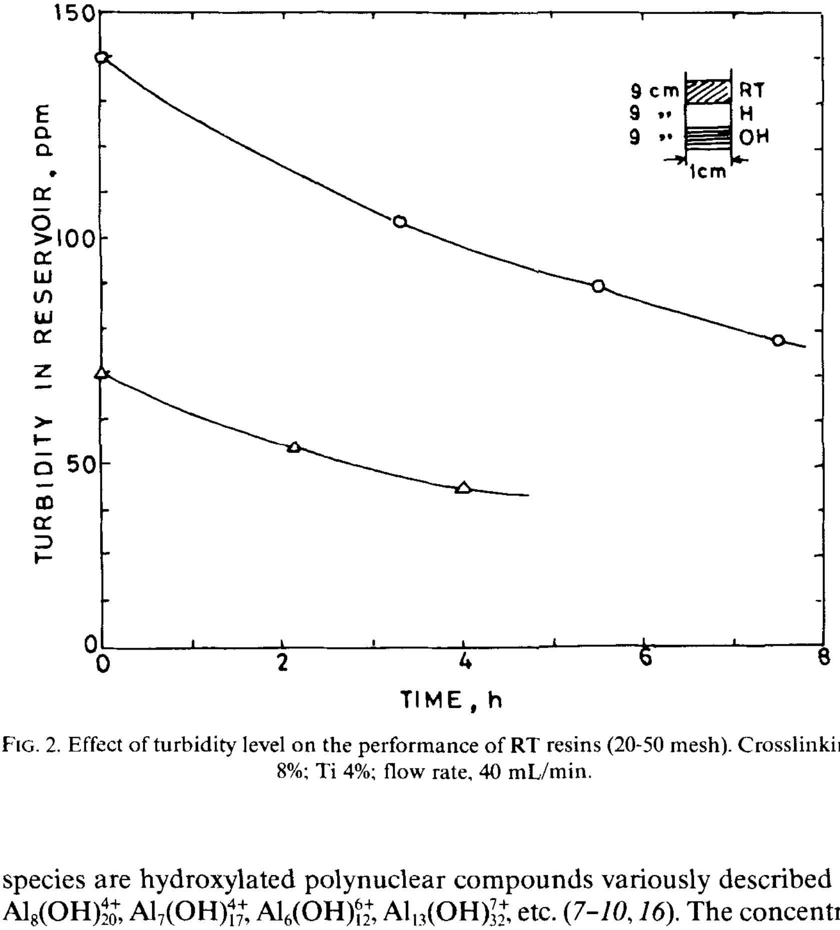

FIG. 2. Effect of turbidity level on the performance of RT resins (20-50 mesh). Crosslinking, 8%; Ti 4%; flow rate, 40 mL/min.

species are hydroxylated polynuclear compounds variously described as $Al_8(OH)_{20}^{4+}$, $Al_7(OH)_{17}^{4+}$, $Al_6(OH)_{12}^{6+}$, $Al_{13}(OH)_{32}^{7+}$, etc. (*7–10, 16*). The concentration of these polynuclear species will be high near the precipitation value of Al(III), which is close to pH 6 (*9*). Thus, the particles that cause turbidity in nuclear reactors have been identified as recrystallized gibbsite, $Al_2O_3 \cdot 3H_2O$ (*3, 4*).

Adsorption of Al(III) on oxides under near neutral pH conditions is

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FIG. 1A. Effect of HTiO loading on the turbidity removal efficiency of RT resins (50-100 mesh). Crosslinking, 4%; flow rate, 25 mL/min; initial turbidity, 200 ppm.

FIG. 1B. Effect of HTiO loading on the capacity of RT resins (50-100 mesh). Crosslinking, 4%; flow rate, 25 mL/min; initial turbidity, 200 ppm.

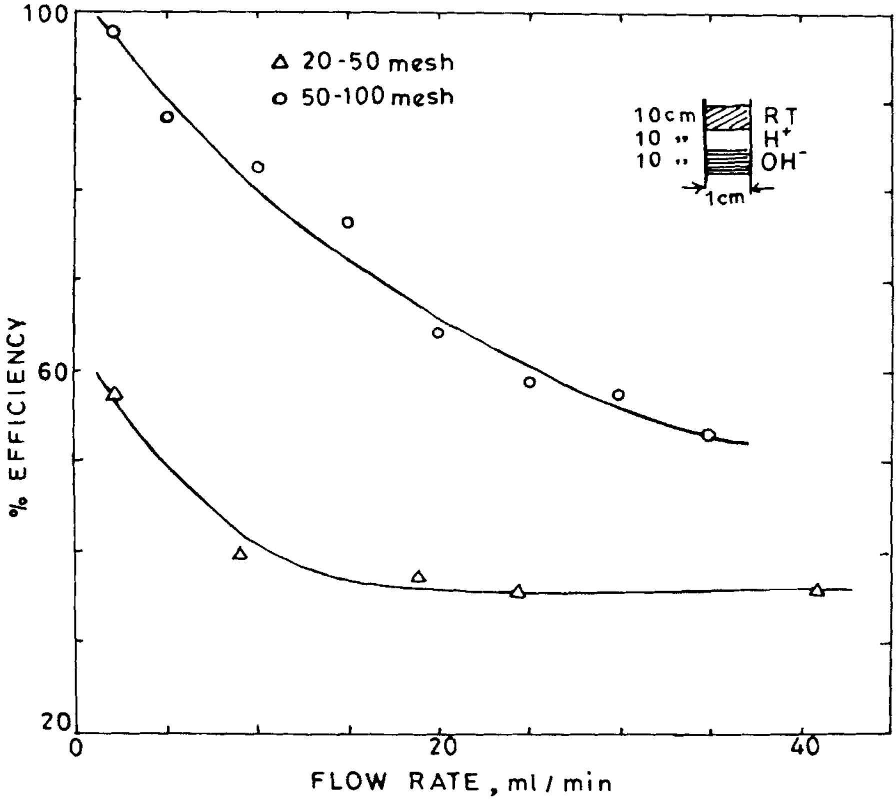

FIG. 3. Effect of flow rate on the turbidity removal efficiency of RT resins. Crosslinking, 8%; Ti, 4%; initial turbidity, 170 ppm.

also thought to involve polymerized species. Some investigators claim that monomeric species like $Al(OH)^{2+}$ are involved in the adsorption process (*6–10*). However, the overall adsorption and retention mechanism appears to be precipitation of Al(III) hydroxide on the surface of oxides, initiated by a surface nucleation or precipitation process (*6*). Electrophoretic mobility studies on the adsorption of Al(III) on TiO_2 indicate (*6–9*) that as a result of adsorption, the TiO_2 surface acquires the character of aluminum oxide.

Studies on the coagulation behavior of Al(III) in the presence of TiO_2 indicate that the overall process is heterocoagulation of Al_2O_3 and TiO_2, in which the Al(III) species derived from the dissolution of Al_2O_3 colloid adsorbs on the TiO_2 surface (*7–10*). Heterocoagulation of Al(III) on TiO_2

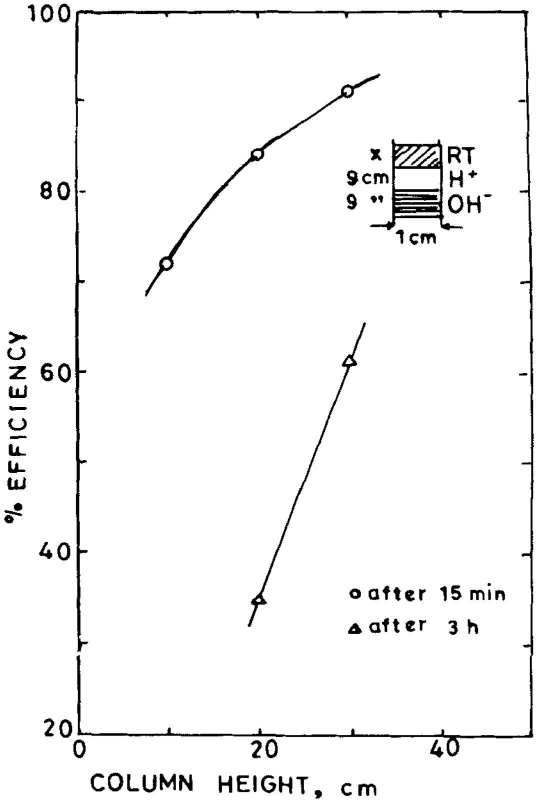

FIG. 4. Effect of column height on the turbidity removal efficiency of RT resins (50-100 mesh). Crosslinking, 8%; Ti, 4%; flow rate, 25 mL/min; initial turbidity, 110 ppm.

seem to occur without adsorption or electrostatic interaction (*10*). However, it is also reported that in the pH range 5-8, H^+ ion interacts strongly with TiO_2 solution, leading to adsorptive coagulation (*10*). The presence of NaOH is also known to promote coagulation of oxides (*2*, *10*). The coagulation behavior also depends on the surface area of the destabilizing agent, the efficiency increasing with increasing surface area (*1*, *8*).

Here we will attempt to rationalize the behavior of the RT resins reported in the earlier section based on the above discussion.

The fact that charge neutralization of the colloidal dispersion alone is not enough, and that a chemical binding medium is also essential, has been demonstrated by the failure of a pure ion-exchange resin column to

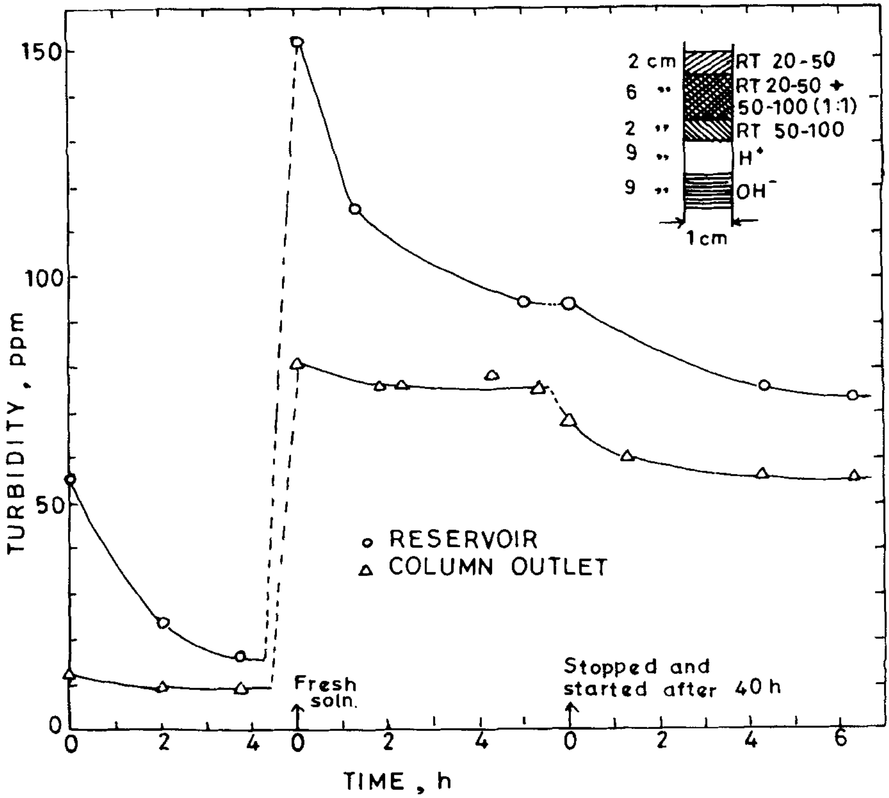

FIG. 5. Turbidity removal efficiency of RT resins with intermittant column operation. Crosslinking, 8%; Ti, 4%; flow rate, 25 mL/min; reservoir volume, 5 L.

clarify Al(III)-based turbidity. This has been observed by us and by other investigators (*3*, *4*), and by the effectiveness of RT resins for the same process. The mechanism by which turbidity is removed by a RT resin does not appear to be just physical but involves some sort of chemical binding. This is evident from the fact that the adsorbed turbidity did not migrate down (Fig. 5) on standing or during the operation, and also the absence of any pressure drop effects.

Though the exact mechanism by which colloidal Al(III) is adsorbed on HTiO is not clear from the literature (*6–9*), the usefulness of HTiO as a sorbent for colloidal Al(III) is evident from the lowering of turbidity by both pure HTiO (T1T, T2T) (Table 1) and RT resin samples. It is surmised that the effectiveness of RT resin should be due to the presence

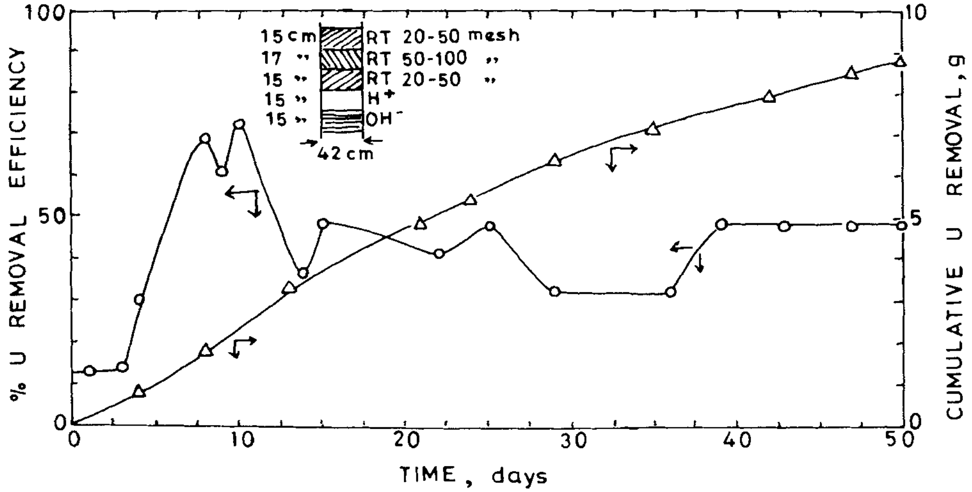

FIG. 6. Removal of uranium by RT resins in large-scale operation. Crosslinking, 8%; Ti, 4%; flow rate, 42 L/min; average U concentration, 15 ppb; reservoir volume, 60 tons.

of HTiO in finely dispersed form inside the resin phase by offering a large surface area for colloidal Al(III) to be adsorbed. Evidence for the above assumption is provided by the work of Michas et al. (*17*) who, based on various surface analysis techniques, concluded that uranium and ruthenium oxide precipitated in the resin phase by using alkali were present as fine particles and were concentrated near the surface of the perfluorinated ion exchange (Nafion) membranes.

As the adsorption of colloidal Al(III) on HTiO presumably involves surface precipitation (*6*), the 4% crosslinked resin offers more pore volume for the above process than 8% crosslinked RT resins (Table 2). Thus, the reduced performance of 8% crosslinked resin should be regarded as a steric effect.

The effect of Ti(IV) content in the RT resin on the turbidity removal efficiency (Figs. 1A and 1B) can also be explained in terms of the steric effect. As the loading of Ti(IV) in the resin increases, the effective pore volume inside the resin phase decreases, and hence the "adsorption-precipitation" also decreases.

When the concentration of the metal ion is low, the particle size of the resulting colloidal oxide formed is also small. When Ti(IV) is loaded on the ion-exchange resin in 2 steps, the concentration of Ti(IV) used in each step is low and hence the particle size of the HTiO formed is also small. A higher concentration of Ti(IV) is used in single step loading of HTiO, and the resulting larger particle size of HTiO reduces the effective pore volume. Thus, the performance of RT resin prepared by a 2-step process is better than for one prepared by a single-step process (Figs. 1A and 1B).

A minimum of 15 s has been found to be necessary before the heterocoagulation process reaches equilibrium (*8*). At the flow rates employed in all the column operations reported in this paper, the contact time between the colloidal Al(III) and RT resin could not have been sufficient for equilibrium to be attained. Hence it is concluded that the efficiency of RT resins decreases at higher flow rates (Fig. 3).

A possible reason why RT resins alone were not able to completely remove the Al(III)-based turbidity has to be considered. It could be that part of the colloid on contact with HTiO is electrically or chemically modified so that subsequently the H^+ form of the resin could hold it. That the colloidal dispersion is modified chemically on passing through a bed of RT resins is evident from the change in the pH of the effluent coming out of a RT resin bed. The high turbidity pickup efficiency of the H^+ form of the resin in conjunction with RT could possibly be due to the role of the H^+ ion in the "adsorptive coagulation process" (*10*).

The main objective of the present study was to develop an adsorber capable of purifying Al(III)-based turbidity under high flow rate conditions. The versatility of the RT resins—HTiO dispersed in ion-exchange resins—is evident from the results reported. Their performance could be analyzed based on the data available in the literature. Future research will be focused on elucidating the basic mechanism of adsorption properties of RT resins, so that this adsorbent will find wider industrial application.

Acknowledgments

The authors wish to thank Head, Analytical Chemistry Division, and Head, Reactor Chemistry Section, Reactor Operations and Maintenance Group, for providing facilities and helping in chemical analysis during the course of the investigation.

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Received by editor July 14, 1987