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CLEANUP OF SAVANNAH RIVER PLANT SOLVENT USING SOLID SORBENTS

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ABSTRACT

The degradation products produced in Purex solvent by exposure to nitric acid and radiation can be divided into two groups: those which are removed by scrubbing with sodium carbonate solutions and those which are not; these latter materials are called secondary degradation products. This study investigated the use of solid sorbents for removal of the secondary degradation products from first-cycle Savannah River Plant solvent that had been previously washed with sodium carbonate solution. Silica gel, activated charcoal, macroporous resin, attapulgite clay and activated alumina were the sorbents investigated in preliminary testing.

Activated alumina was found to be most effective for improving phase separation of the solvent from sodium carbonate solutions and for increasing the interfacial tension. The activated alumina was also the sorbent most useful for removing complexants which retain plutonium at low acidity, but it was less effective in removing anionic surfactants and ruthenium. We found that the capacity of the activated alumina was greatly improved by drying the solvent before treatment.

1. INTRODUCTION

All existing and planned reactor fuel reprocessing plants use the Purex solvent extraction process for recovery of the fissionable materials from the spent fuel elements.¹ The head-end of the Purex process involves dissolution of the oxide fuel in nitric acid to yield a solution containing ~200 g of heavy metals per liter, in 3 to 4 M HNO₃. This solution is then contacted with a 20 to 30 vol % solution of tri-n-butylphosphate (TBP) in a normal paraffin hydrocarbon (NPH) diluent. The uranium and plutonium are preferentially extracted into the organic phase, leaving the bulk of the fission products in the aqueous raffinate. The extract is then scrubbed with nitric acid to increase the separation from fission products, and the heavy metals are stripped from the solvent.

The TBP-NPH used in these processes is degraded by radiation damage and by hydrolytic and dealkylation reactions with the nitric acid. A variety of methods have been proposed for cleanup of the used Purex process solvent to minimize the problems encountered when the degradation products accumulate. In an earlier study, we found that scrubbing with sodium carbonate solutions (the current practice in Purex plants) was the preferred primary solvent cleanup treatment, unless sodium in the effluent must be restricted.

The secondary degradation products (i.e., those remaining after the primary cleanup) can cause problems with phase separation and retention of cations. Since the identities and chemical properties of the important contaminants are unknown, secondary cleanup studies must be conducted using actual recycle solvent. Our studies have used solvent obtained from the Savannah River Plant (SRP) for testing a number of solid sorbents including activated charcoal, silica gel (as-received and treated with NaOH), activated alumina, attapulgite clay, and macroreticular anion-exchange resin.

2. EXPERIMENTAL STUDIES

2.1 MATERIALS

The materials in this study included the SRP Purex solvent and a variety of solid sorbents.

2.1.1 SRP Solvent

After each solvent extraction cycle, the SRP solvent is scrubbed with sodium carbonate, dilute acid, and again with sodium carbonate. No treatment to remove the secondary degradation products is attempted in normal process operation. During periods when the plant is inactive, the solvent scrubbing continues; this means the solvent is cleaner immediately after periods of plant shutdown.

During our studies, we used three batches of solvent. The first batch had been stored at Oak Ridge National Laboratory (ORNL) for several

years before the time of use. The later two batches were fresh and were used within a period of several months after they were obtained. The solvents were scrubbed with 0.2 volume of 0.25 M sodium carbonate—0.02 M sodium tartrate solution and filtered through Whatman #40 paper before use. The initial properties of the three solvents varied, as shown in Table 1. A later section of this report discusses the methods for determining solvent properties.

Table 1. Initial properties of Savannah River Plant Purex solvent

Solvent batch	Separation time (s)	Interfacial tension (dynes/cm)	Anionic surfactants ^a	Plutonium retention (M)	Zirconium retention (M)
First	160	6.2	0.075	4.2×10^{-5}	1.5×10^{-7}
Second	105	7.0	0.177	2.9×10^{-5}	7.0×10^{-8}
Third	50	6.6	0.141	---	---

^aAbsorbance of anionic surfactants at 653 nm by the methylene blue test (see Ref. 3).

2.1.2 Solid Sorbents

The solid sorbents tested were activated charcoal, silica gel (as-received and treated with NaOH), activated alumina, attapulgite, and macroreticular resin. The activated charcoal (Barnebey-Cheney, Columbus, Ohio) was about 12–42 mesh and was washed several times with acetone (total wash volume about 15× the charcoal volume) to remove most of the materials in the as-received activated charcoal that might cause discoloration. The 60–120-mesh silica gel (BDH Laboratories Ltd., Poole, England) was generally used as received. In one test, the fines (<100-mesh) were removed to examine the effects on pressure drop and effectiveness. For another test, Grade 35, 12–42-mesh silica gel (Davison Chemical, Baltimore, MD) was used. The activated alumina was Alcoa F-1 type, 60–120-mesh (Aluminum Co. of America, Pittsburgh, PA). Attapulgite clay Florex AA-LVM, 45–60 mesh, from Floridin Company, Pittsburgh, PA, was used as received.

This material is a type of fuller's earth that has been extruded and calcined for activation. Amberlyst A-26 resin, 14-50 mesh (Rohm and Haas, Philadelphia, PA) was converted to the hydroxide form before use.

2.2 APPARATUS

The test columns were made of 8-mm-OD, 6-mm-ID glass tubing, except for the charcoal test, in which the glass column was of about 10-mm OD and 8-mm ID. A typical experimental setup is shown in Fig. 1. The solvent flowed down through the beds in all cases, and the flow rate through the packed beds was 1 mL/min (except for the tests using charcoal and 12-42-mesh silica gel, where the flow rate was 2 mL/min). In the tests using 60-120-mesh activated alumina, the bed depth was ~5.25 cm and the solvent head required to maintain the 1 mL/min flow rate was ~50 cm. Most tests were at room temperature (~22°C). In a few tests, the temperature of the bed was increased by immersing it in a beaker of heated water.

2.3 ANALYSIS METHODS

The effluent was saved in 5-mL samples, and a number of these were examined for interfacial tension and phase-separation time from sodium carbonate/tartrate solution. Anionic surfactant content was determined by the methylene blue (MB) technique.³ Plutonium retention and ruthenium removal were also assessed. The zirconium retention of the solvent⁴ was so low ($\sim 7 \times 10^{-8}$ M) that it was considered to be of no significance and was not routinely determined. The test methods used on the organic samples are discussed in the following sections.

2.3.1 Interfacial Tension

The organic solvent sample to be tested was first equilibrated with the sodium carbonate/tartrate solution. A small quantity of the separated organic phase was drawn into a 0.2-mL micrometer syringe (Gilmont Instruments, Inc., Great Neck, NY) equipped with a 0.0636-cm-OD,

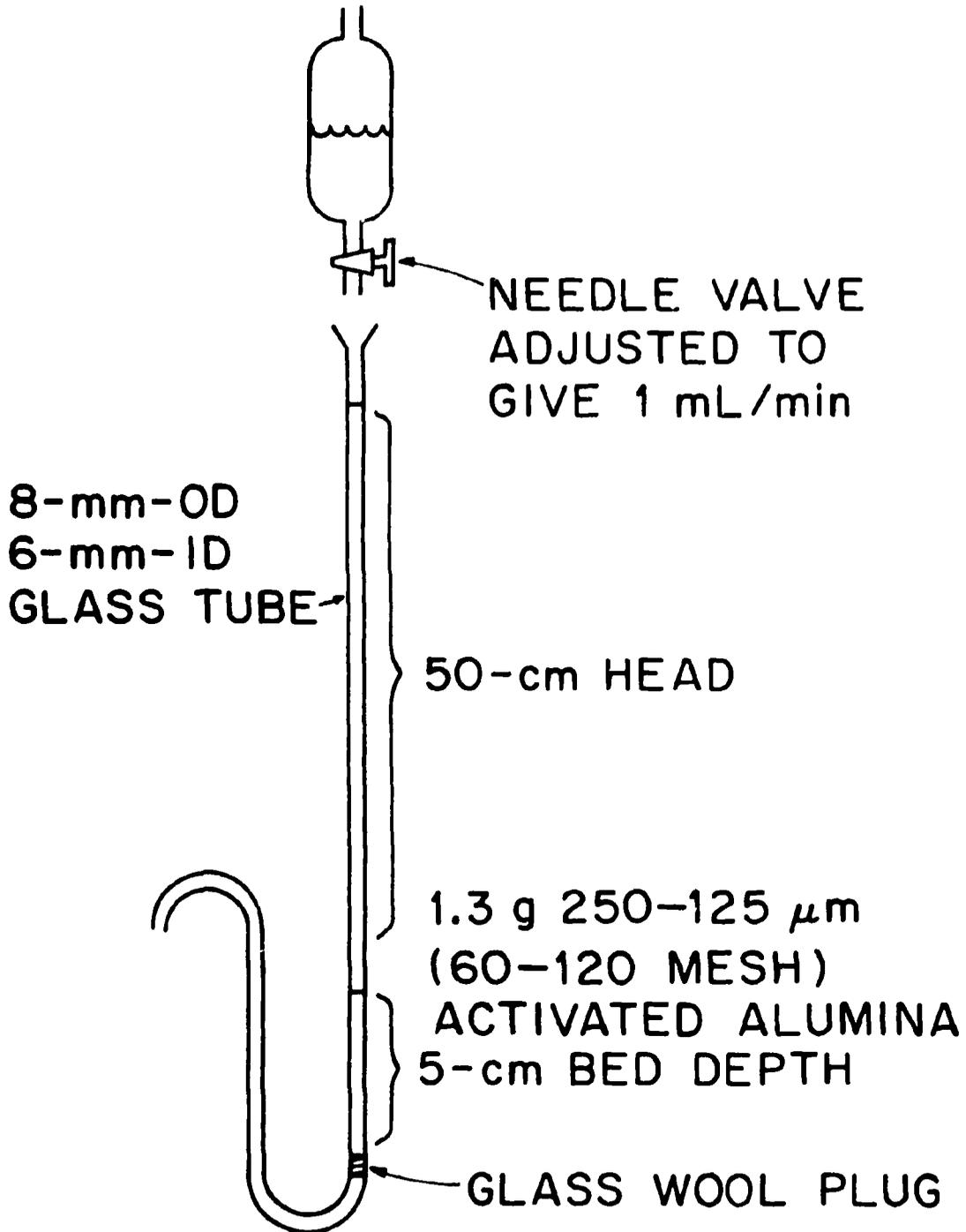


Fig. 1. Typical column used for secondary cleanup tests on Savannah River Plant Purex solvent.

flat-ended, stainless steel needle. This was then mounted with the tip below the surface of the pre-equilibrated aqueous phase. Drops of organic solvent were slowly dispensed, and the volume reading of the micrometer dial was recorded as each drop fell. The interfacial tension was calculated for each drop and the average determined for several drops.⁵ The interfacial tension for freshly prepared 30% TBP-NPH was found by this technique to be ~10-11 dynes/cm.

2.3.2 Phase-Separation Time

The phase-separation time was determined by placing 2 mL of the organic solvent and 2 mL of the sodium carbonate/tartrate solution in a glass vial of ~1.2-cm diam and mixing by slow inversion several times. The separation time was taken to be the time for the emulsion to collapse to one layer of drops at the interface. All these determinations were done by one individual for maximum consistency in results. Even so, this must be considered a relatively inexact method. It is a necessary measurement, however, since phase separation from sodium carbonate solutions is one of the problem areas for degraded solvents. The phase separation time for unused 30% TBP-NPH (freshly prepared from TBP and NPH and scrubbed with sodium carbonate solution) is 20-25 s.

2.3.3 Anionic Surfactants

The MB test was performed according to the method of Neace which is summarized here.³ Methylene blue (0.045 g) is added to 1 L of 0.5 M acetic acid and the pH adjusted to 5.0 by adding NaOH. To a 150-mL beaker should be added 50 mL of water, 10 mL of the MB reagent, 10 mL of chloroform, and 1 mL of the organic solvent to be tested. This is stirred vigorously on a magnetic stirrer for 60 s and the aqueous phase discarded. The organic phase is filtered through a Whatman #40 paper filter and placed in a 10-mm-path spectral cell. The absorbance at 653 nm vs absorbance of a chloroform standard gives an indication of the concentration of anionic surfactants. Freshly prepared 30% TBP-NPH gave no absorbance by this test.

2.3.4 Plutonium Retention

The solvent (2 to 5 mL) was contacted with ~0.05 mL (one drop) of 2.2 M HNO_3 containing ~40 mg Pu/mL. This was scrubbed three times with equal volumes of water. A sample that had been filtered or allowed to settle overnight was then submitted for gross-alpha determination; from this count, the plutonium retention can be estimated in molar units (assuming 10^8 cpm/mg Pu). Freshly prepared 30% TBP-NPH gave a plutonium retention of 2.8×10^{-6} M by this method.

3. EXPERIMENTAL RESULTS

An initial series of short tests using the first batch of SRP solvent investigated the ability of the various sorbents to improve the solvent's quality. Improvement was largely defined on the basis of changes in interfacial tension and phase-separation time. These preliminary tests did not examine ruthenium removal, since the ruthenium in this solvent had decayed to low levels. The tests used the as-received sorbent solids. After the initial evaluations, more lengthy tests investigated the preferred operating conditions for activated alumina, identified as the best sorbent in the initial tests. Centrifugation was also tested as a means for removal of suspended particulates in the solvent.

3.1 PRELIMINARY TESTING

3.1.1 Silica Gel

The changes in interfacial tension and phase-separation time for the solvent treated with silica gel (0.5 g of 60-120-mesh BDH) are shown in Fig. 2. The interfacial tension was only slightly improved over that of the feed. However, the phase-separation time was significantly improved over the entire test. This is currently thought to be due to filtration of particulates from the solvent. Significant removal of anionic surfactants (MB test) was observed, but no improvement was noted in plutonium retention. In a separate test using 1.4 g of 12-42-mesh silica gel, no improvement was found in interfacial tension, phase-separation time,

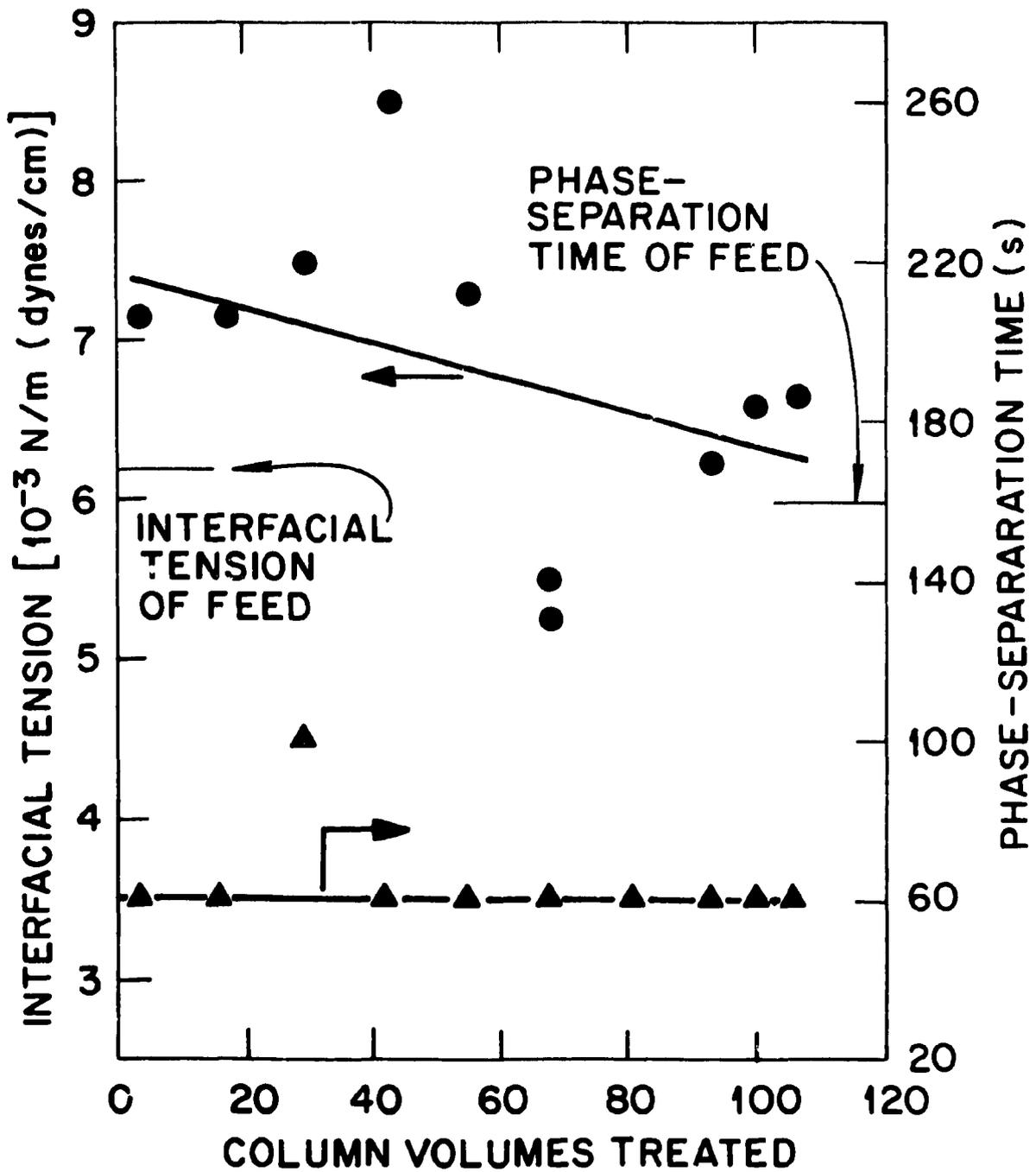


Fig. 2. Interfacial tension and phase-separation time for SRP solvent after passage through 0.5 g of silica gel (60-120 mesh) at 22°C.

anionic surfactant content, or plutonium retention. A test using 1.4 g of 60-120-mesh silica gel, which had been treated with sodium hydroxide to obtain a very basic surface, gave no improvement in the measured indices of solvent quality.⁶ None of the tests using silica gel showed dramatic improvement in solvent quality.

3.1.2 Activated Charcoal

The interfacial tension and phase-separation results for the test using acetone-washed, activated charcoal are given in Fig. 3. The interfacial tension increased slightly with time, indicating that initially some material that decreased the interfacial tension was being leached from the charcoal. The phase-separation time also showed some improvement with time. The content of anionic surfactants was high initially, declined during the run, but was never as low as the feed content. Evidently deleterious compounds were leached from the charcoal by the TBP. This leaching effect was also evidenced by an increase in the yellow coloration of the solvent after passage through the charcoal bed. The plutonium retention showed some improvement, but this improvement declined with time. Overall, the charcoal treatment showed no significant benefits for solvent improvement.

3.1.3 Macroreticular Resin

The results for the test using 2 g of hydroxide-form, A-26 macroreticular resin are shown in Fig. 4. The interfacial tension was slightly improved, and the phase-separation time was significantly shortened, perhaps as a result of filtration of particulates. The final anionic surfactant content of the solvent was higher than that of the feed; this may have been caused by leaching of organics from the resin, as previously observed.⁷ With use of the resin, plutonium retention was improved over that of the feed, and the improvement increased with time. This suggests that the resin needed preconditioning with the solvent. The overall improvement in solvent properties was not sufficiently great to justify further testing of this sorbent.

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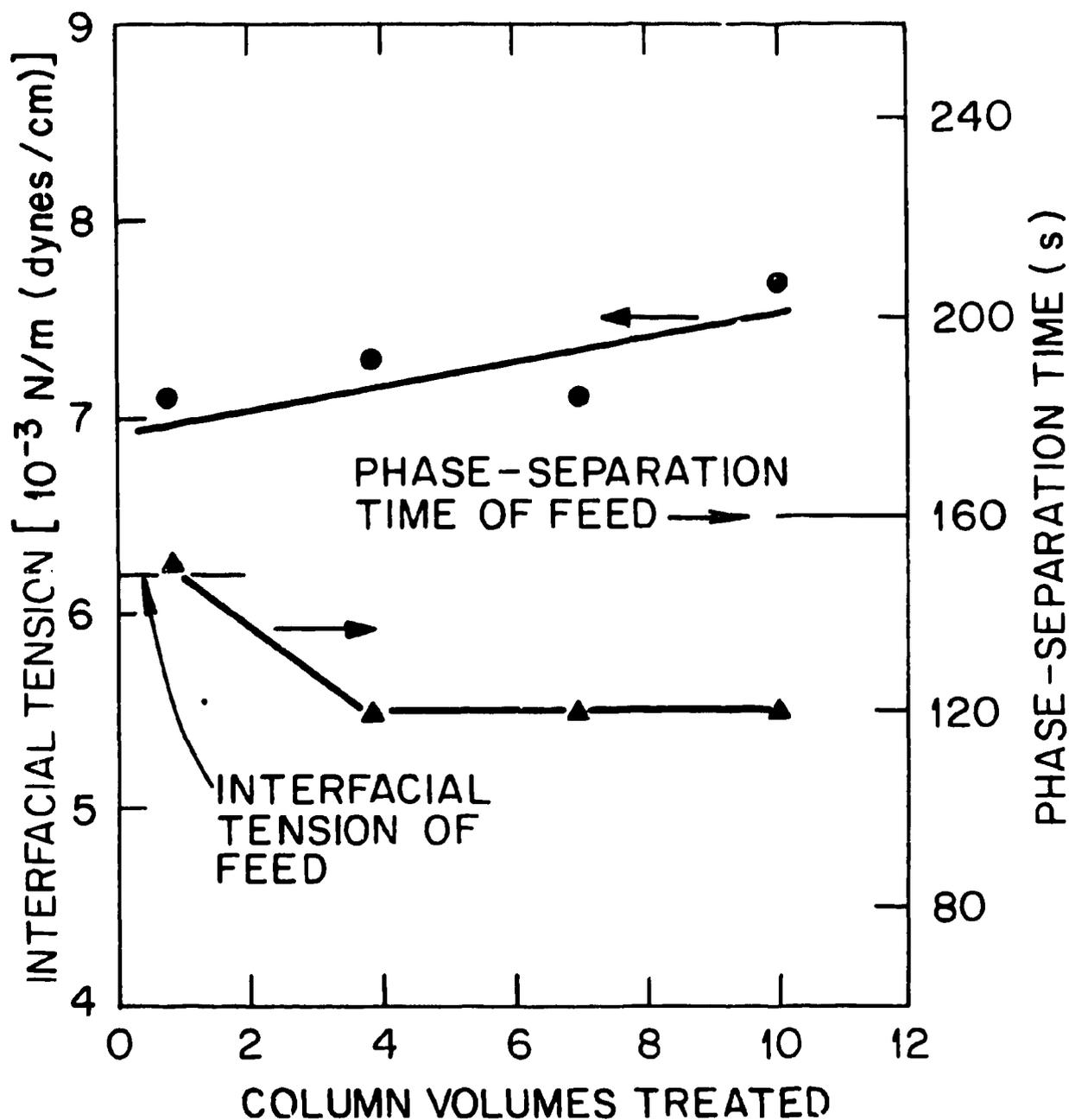


Fig. 3. Interfacial tension and phase-separation time for solvent after passage through 3.4 g of activated charcoal (12-42 mesh) at 22°C.

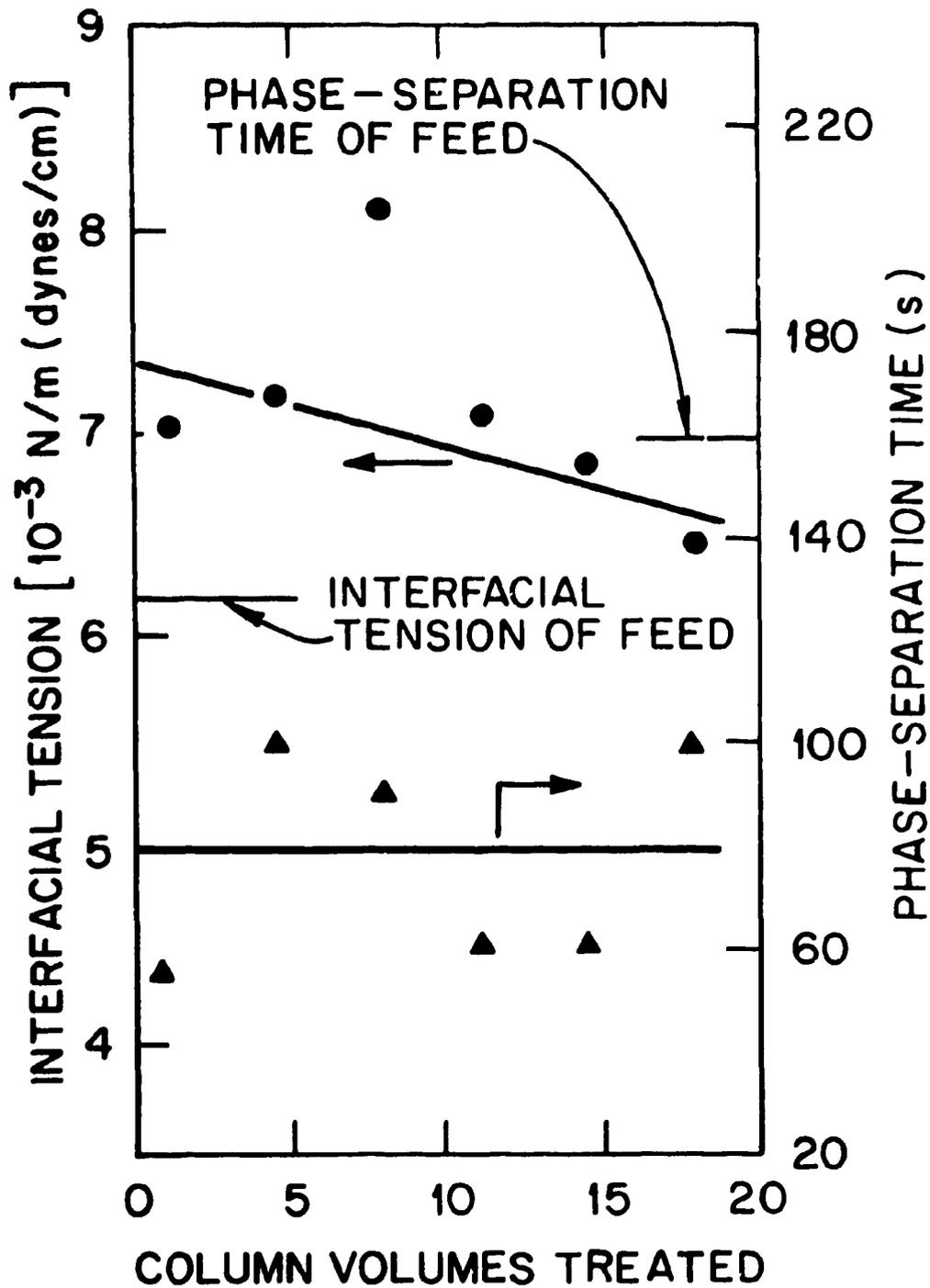


Fig. 4. Interfacial tension and phase-separation time for SRP solvent after passage through 2 g of hydroxyl-form A-26 macroreticular resin at 22°C.

3.1.4 Attapulгите

The results for the test using 1.4 g of attapulгите clay are shown in Fig. 5. The interfacial tension was not significantly affected by passage of the solvent through attapulгите. The phase-separation times were improved, but the improvement rapidly declined. The anionic surfactant content was also reduced by the attapulгите treatment. Plutonium retention was not determined for the solvent used in this test. The attapulгите treatment showed promise of solvent improvement, but it was not pursued because of the superior results found with activated alumina.

3.1.5 Activated Alumina

Three tests were done using ~1.3 g of 60-120-mesh activated alumina. Results are given in Figs. 6-9. The initial interfacial tension of the solvent leaving the column in the first test was very high, indicating that the TBP had been removed by the activated alumina (Fig. 6). In subsequent tests, the activated alumina was preconditioned by treatment with ~10 mL of clean 30% TBP-NPH to saturate the surface with TBP. The interfacial tension of the first ~15 column-volumes of solvent in each of the three tests was increased to the level expected for fresh solvent. After this, the interfacial tension decreased rapidly. A similar trend was observed for the phase-separation time (Fig. 7), with the initial low separation times showing a sharp increase at about the same treated volume for which the rapid decrease in interfacial tension was seen. The phase-separation times remained lower than those of the feed, due to filtration of particulates. Anionic surfactant levels (Fig. 8) were somewhat improved over those in the initial ~15 column-volumes of solvent treated. Plutonium retention (Fig. 9) showed significant improvement over a much larger treated volume.

3.1.6 Centrifugation

The second batch of SRP solvent was centrifuged to test for the removal of impurities that cause increased phase-separation times from sodium carbonate/tartrate solution. In the tests, the radius to the sample in the centrifuge bowl was 12.7 cm and rotation rates were 1000,

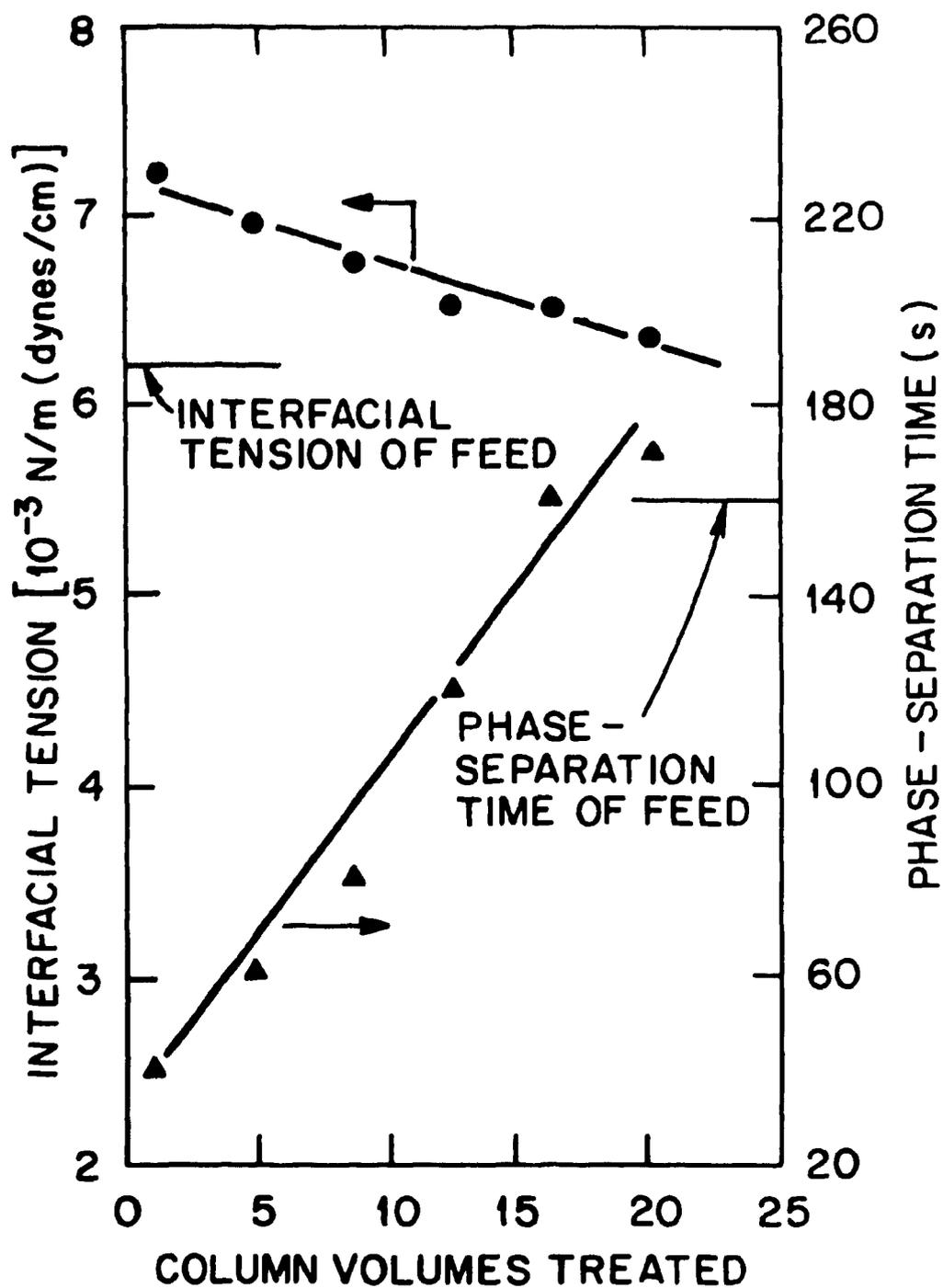


Fig. 5. Interfacial tension and phase separation time for solvent passed through 1.4 g of 45-60 mesh attapulgite.

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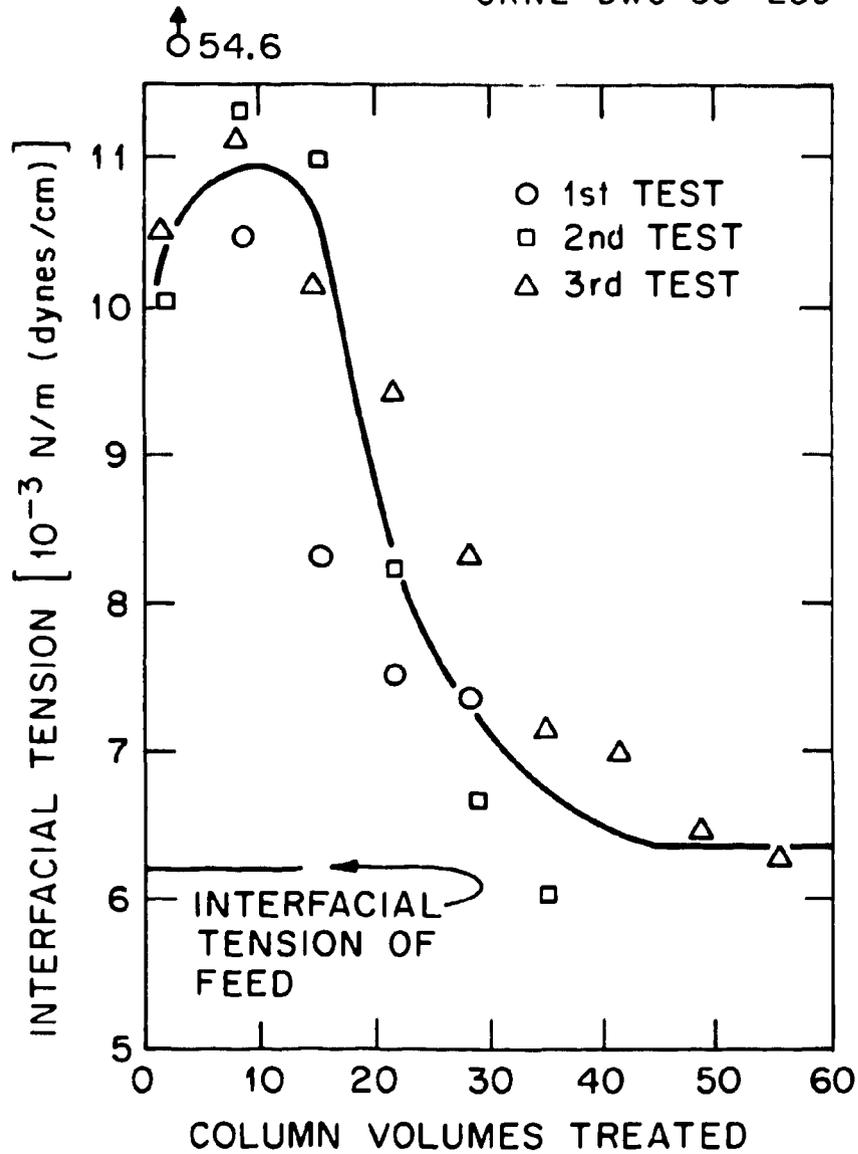


Fig. 6. Interfacial tension of undried solvent passed through 1.3 g of 60-120 mesh activated alumina.

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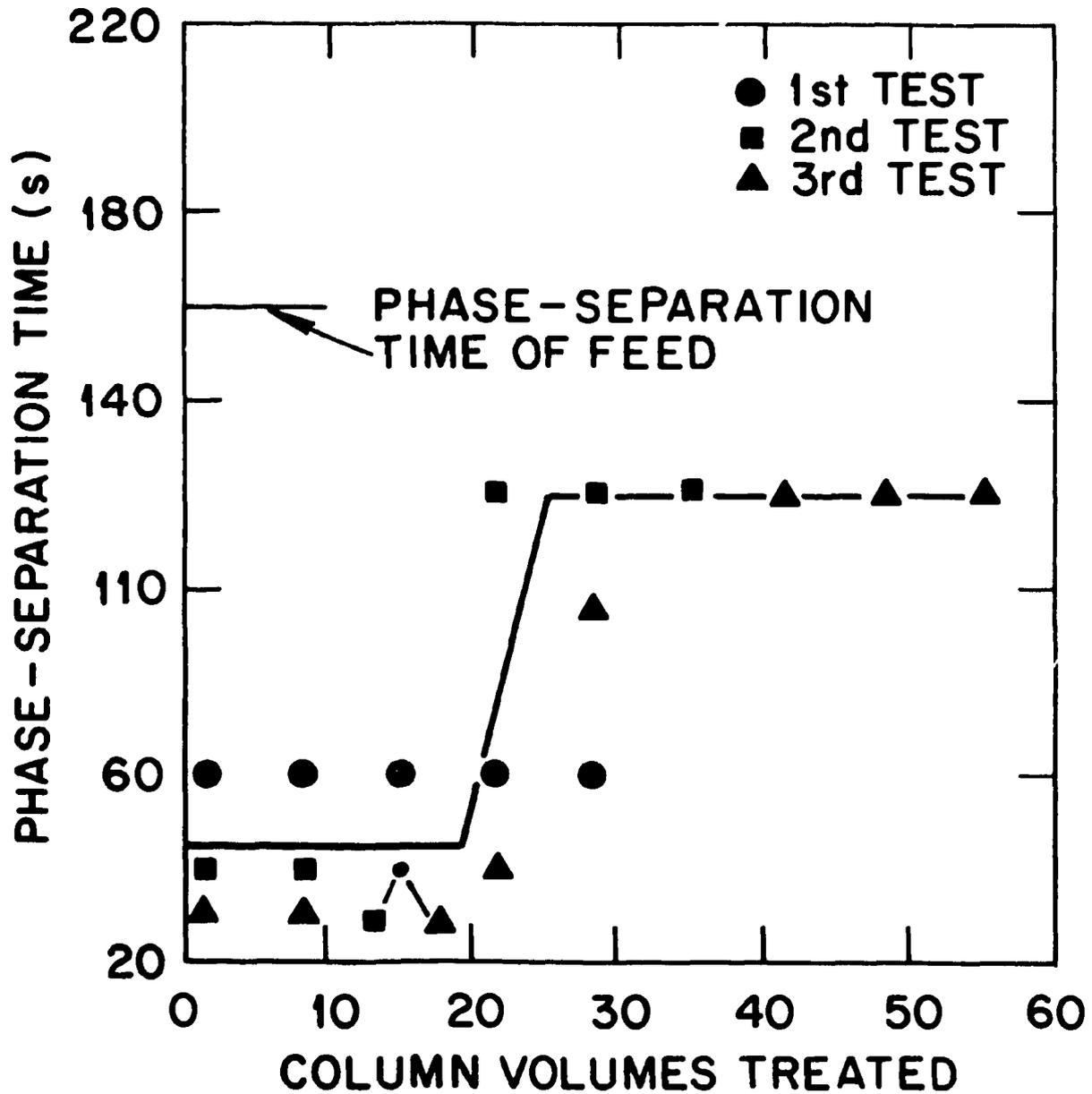


Fig. 7. Phase-separation times for undried SRP solvent after passage through 1.3 g of activated alumina (60-120 mesh) at 22°C.

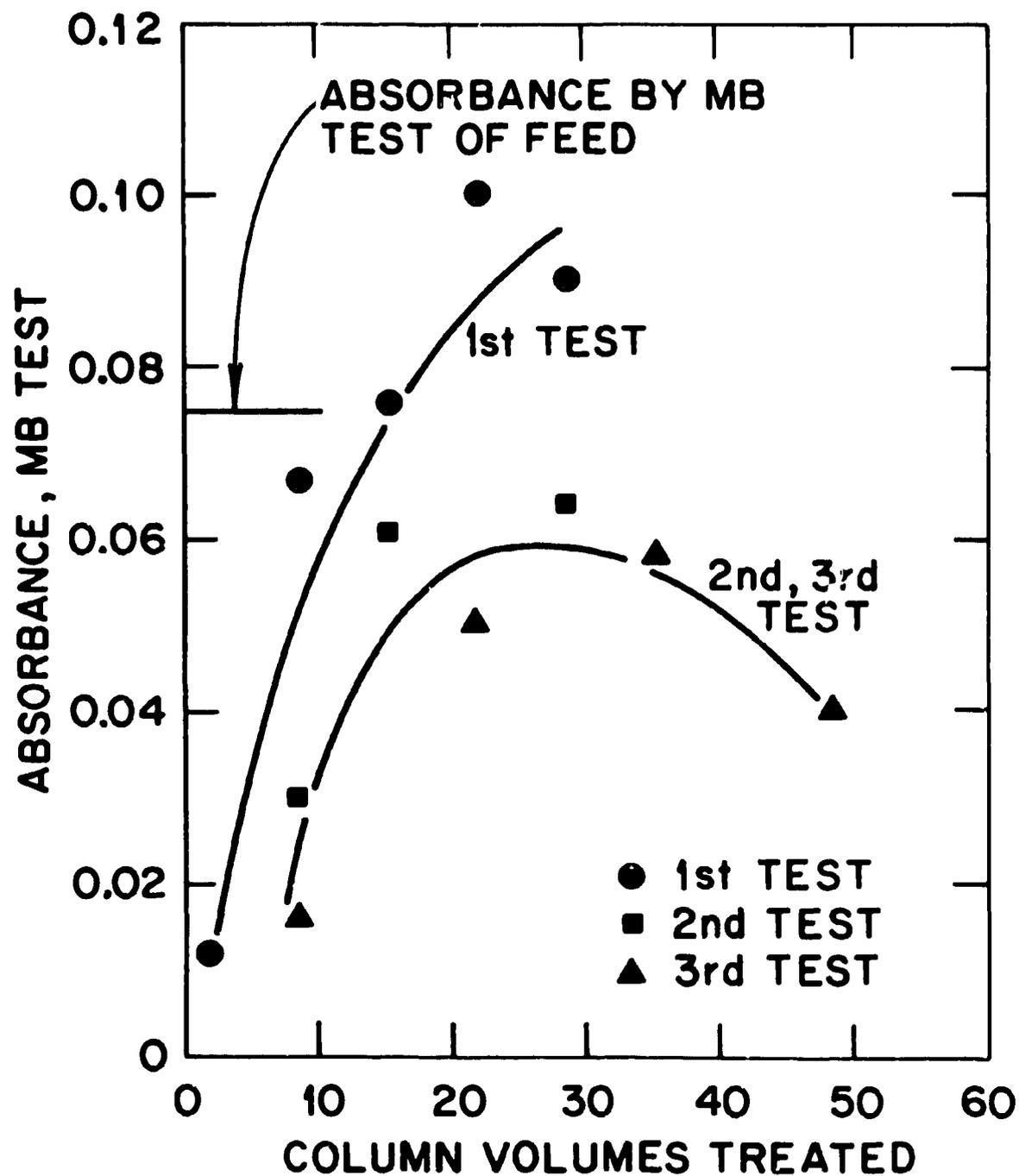


Fig. 8. Absorbance of anionic surfactants in undried SRP solvent after passage through 1.3 g of activated alumina (60-120 mesh). [Determined by the methylene blue (MB) method, with absorbance at 653 nm.]

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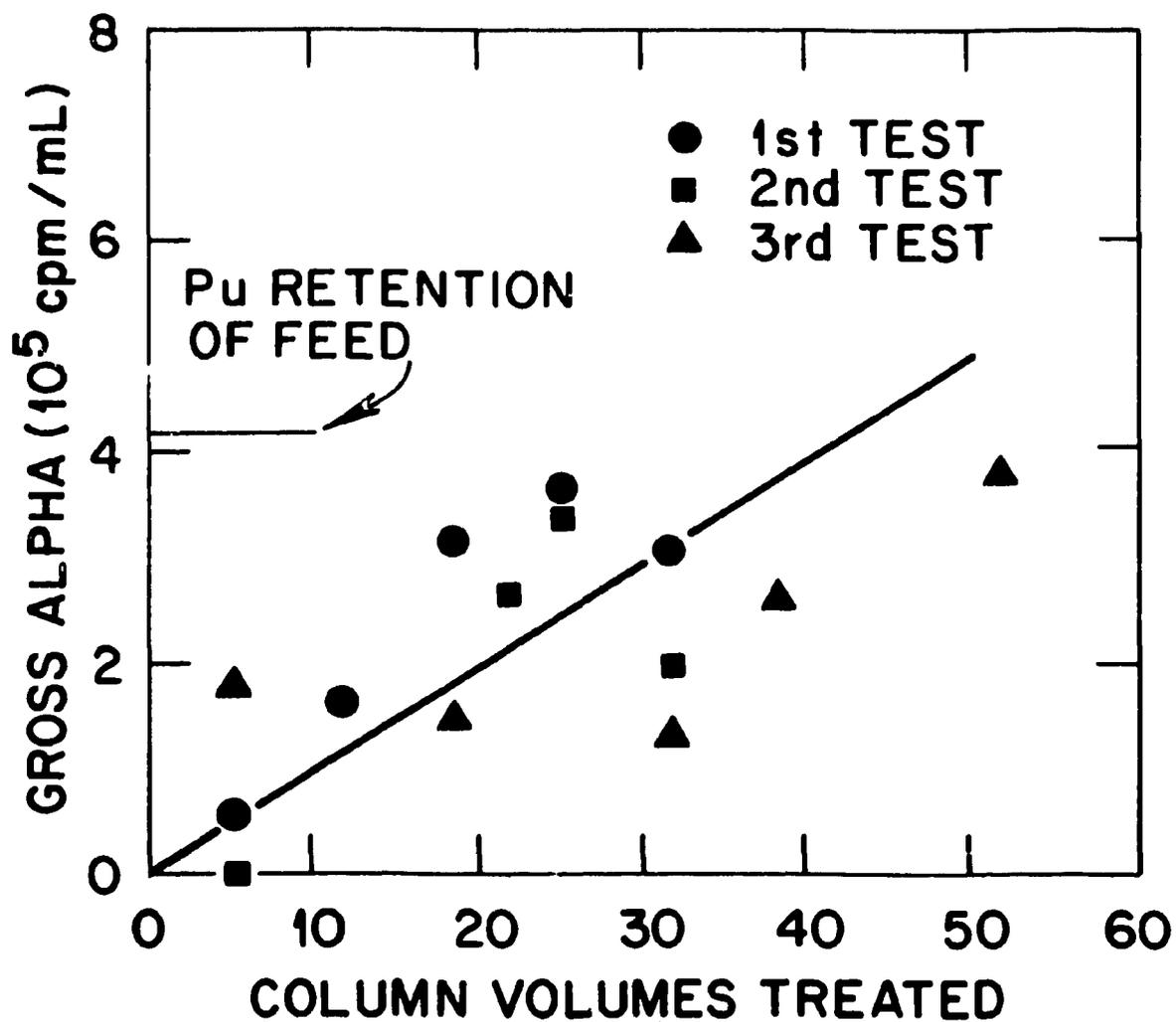


Fig. 9. Plutonium retention as measured by gross-alpha determination on undried SRP solvent after passage through 1.3 g of activated alumina (60-120 mesh).

1245, and 2900 rpm. Centrifugation time was 1 min for all samples. At 1245 rpm, the force was equivalent to ~240 times gravity (g). This approximates the centrifuge gravitational field planned for solvent cleanup in the Breeder Reprocessing Engineering Test (BRET). In all of our tests, the phase-separation times were decreased from 100 s (without centrifugation) to ~50 s. Thus, a significant portion of the phase-separation problem appears to be associated with interfacially active particulates that can be removed by centrifugation. The ruthenium activity of the solvent decreased only ~5%, indicating that the major fraction of the ruthenium is not associated with the particulates.

A 15-mL sample of the third as-received SRP solvent was centrifuged and the zirconium, niobium, and ruthenium counted in the lower 0.5 mL and in the remaining 14.5 mL. If no separation of the radionuclides had occurred, the fraction of radionuclides in the bottom 0.5 mL should have been 0.033. The fractions actually determined for zirconium, niobium, and ruthenium were (respectively) 0.096, 0.108, and 0.035. Thus, some of the zirconium and niobium, but none of the ruthenium, must be associated with the centrifugable material. This is in agreement with our experience using simulated degraded solvent, where we found the zirconium has a tendency to form suspended material when contacted with sodium carbonate solutions. Since we have found that this centrifugable material is associated with problems of phase separation of solvent from sodium carbonate solutions, it follows that the zirconium and niobium compounds may be interfacially active.

3.2 CLEANUP OF DRIED SOLVENT WITH ACTIVATED ALUMINA

3.2.1 Room-Temperature Tests

The data from the activated alumina test suggested that the capacity of the beds was lower than desired and that this might be due to competition for the active sites on the adsorbent by water in the solvent. Drying the solvent in a Purex plant will require a simple process that does not create waste problems; a likely approach is to contact the solvent with a dry gas stream at a modest temperature. In our test we

sparged 200 mL of SRP solvent, which was saturated with 6.85 g H₂O/L, for 6 h with 200 mL (STP) of dry air per min at 60°C. Figure 10 shows graphically the water content of the solvent as a function of time. The initial rate of water removal corresponds to ~50% saturation of the air. Complete saturation of the air is not possible, since the water is not at unit activity. The apparently slow water removal after 2 h of sparging may be due partially to water absorbed by the samples before they could be analyzed. Treatment at 60°C is safe, since this is below the flash point of the solvent. Under these conditions, only ~0.5% of the diluent would be volatilized, and even this could be recovered by condensing the vapors and separating the diluent from the water. Interestingly, the phase-separation time of the sparged solvent from the sodium carbonate/tartrate solution was decreased from 105 to 75 s, indicating that some volatile components contribute to phase-separation problems.

In our tests, the solvent was then passed through the activated alumina column, with the results given in Figs. 11 and 12. For the first 200 column-volumes treated, the phase-separation time (15 s) and the interfacial tension (>10.5 dynes/cm) were better than those measured for freshly prepared 30% TBP-NPH (Fig. 11). The treatment capacity of the column (determined as the mid-point of the upturn in the phase-separation time) was ~270 column-volumes. Figure 12 shows the percentage of the initial ruthenium and anionic surfactants in the feed solvent remaining after passage through the column. Stopping the solvent flow and restarting with a newly dried batch of solvent gave improved solvent cleaning for a short period of time. This may have been due to the lower flow rates during the start-up period with the new solvent. This phenomenon was less noticeable in later tests, where a larger batch of solvent was dried initially to limit the times of shutdown. The close agreement between the values for ruthenium and anionic surfactants removal supports the suggestion by Neace that retention of ruthenium is related to anionic surfactants.⁸ The plutonium retention of the solvent was reduced to an average of 30% of the initial value. The lowest plutonium retention values were 2% (after 200 column-volumes) and 9% (after 400 column-volumes), suggesting nearly complete removal of the plutonium complexes.

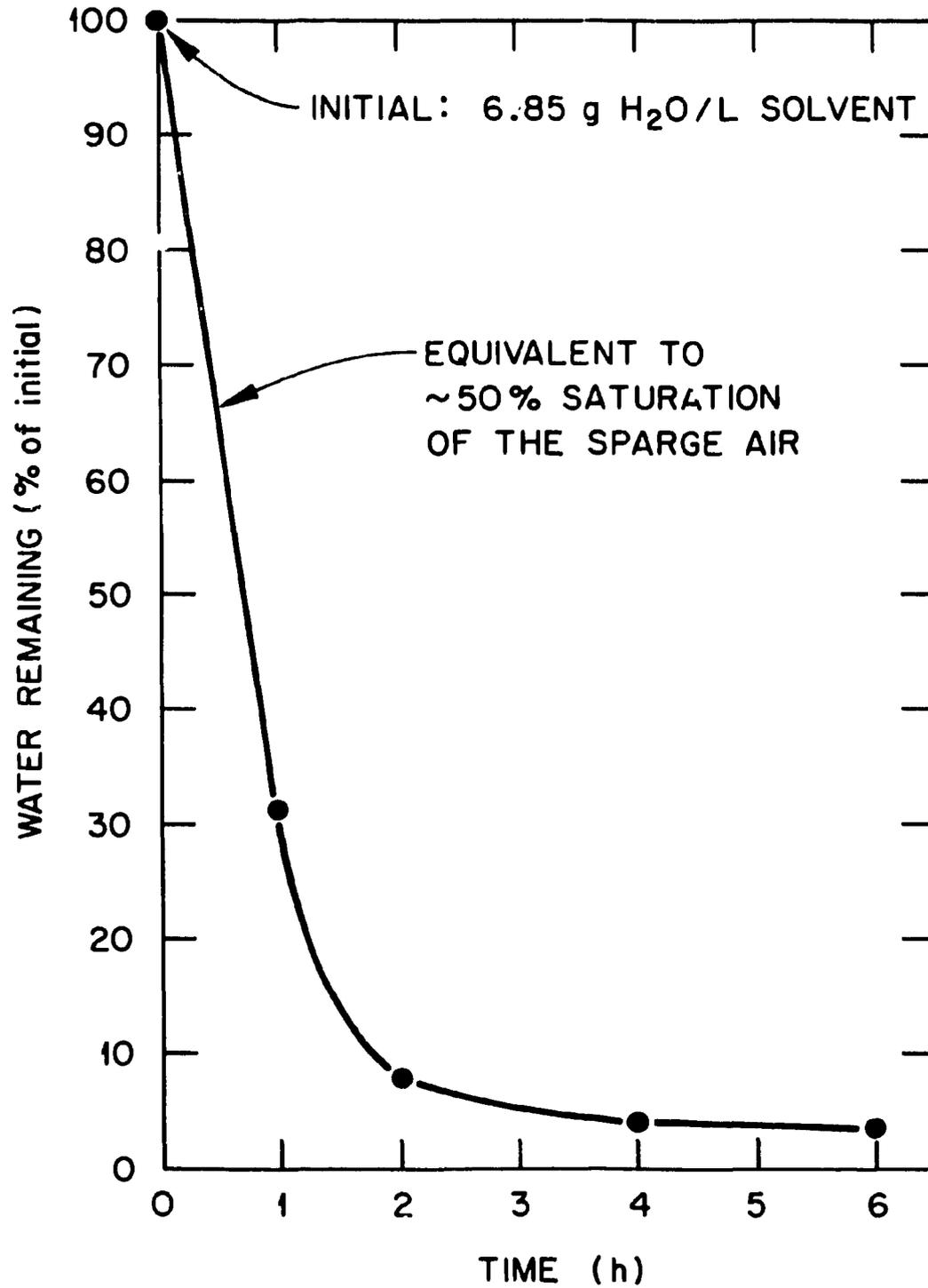


Fig. 10. Drying of SRP solvent by air-sparging at 60°C (200 mL solvent; 200 mL/min dry air).

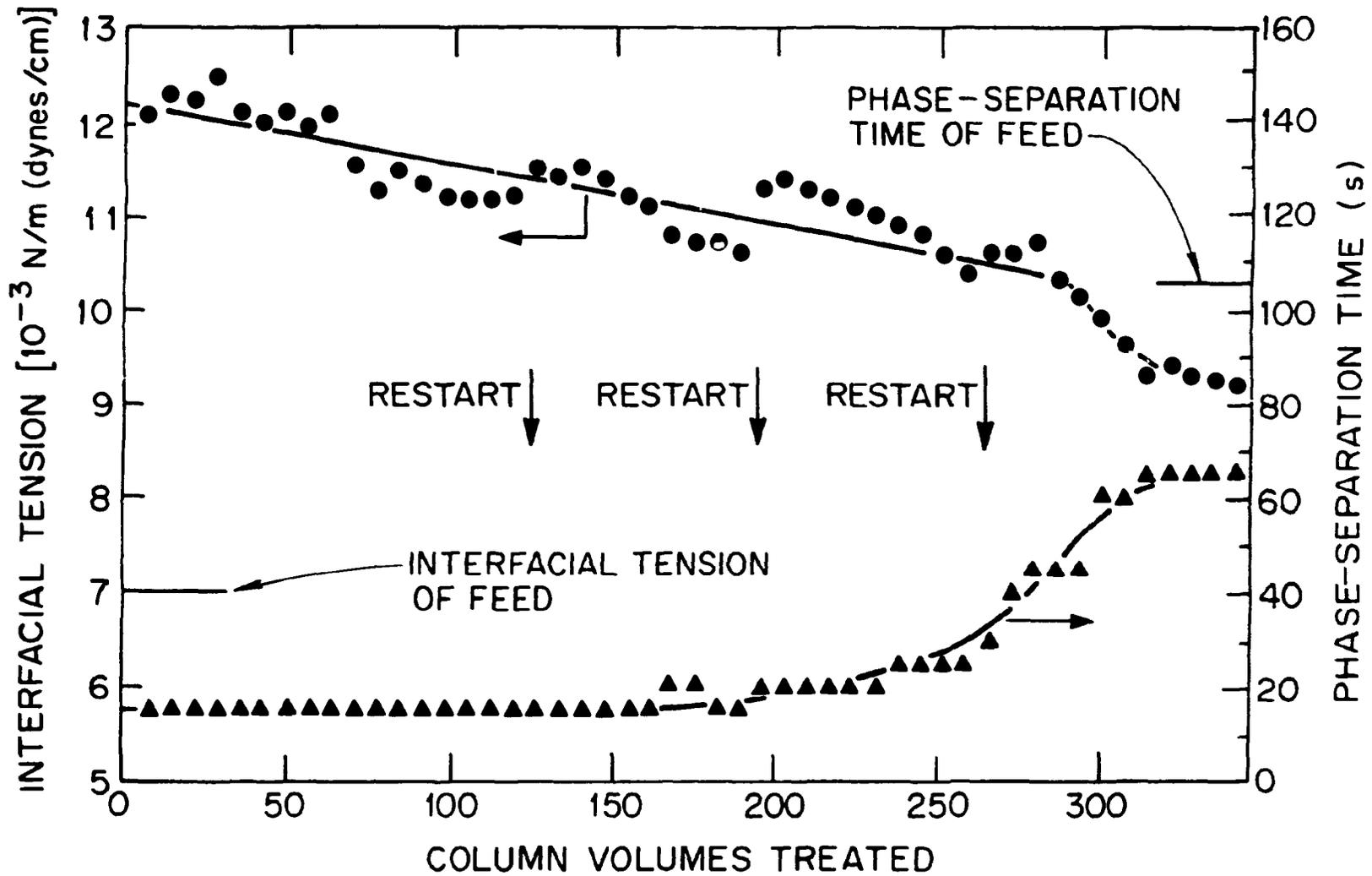
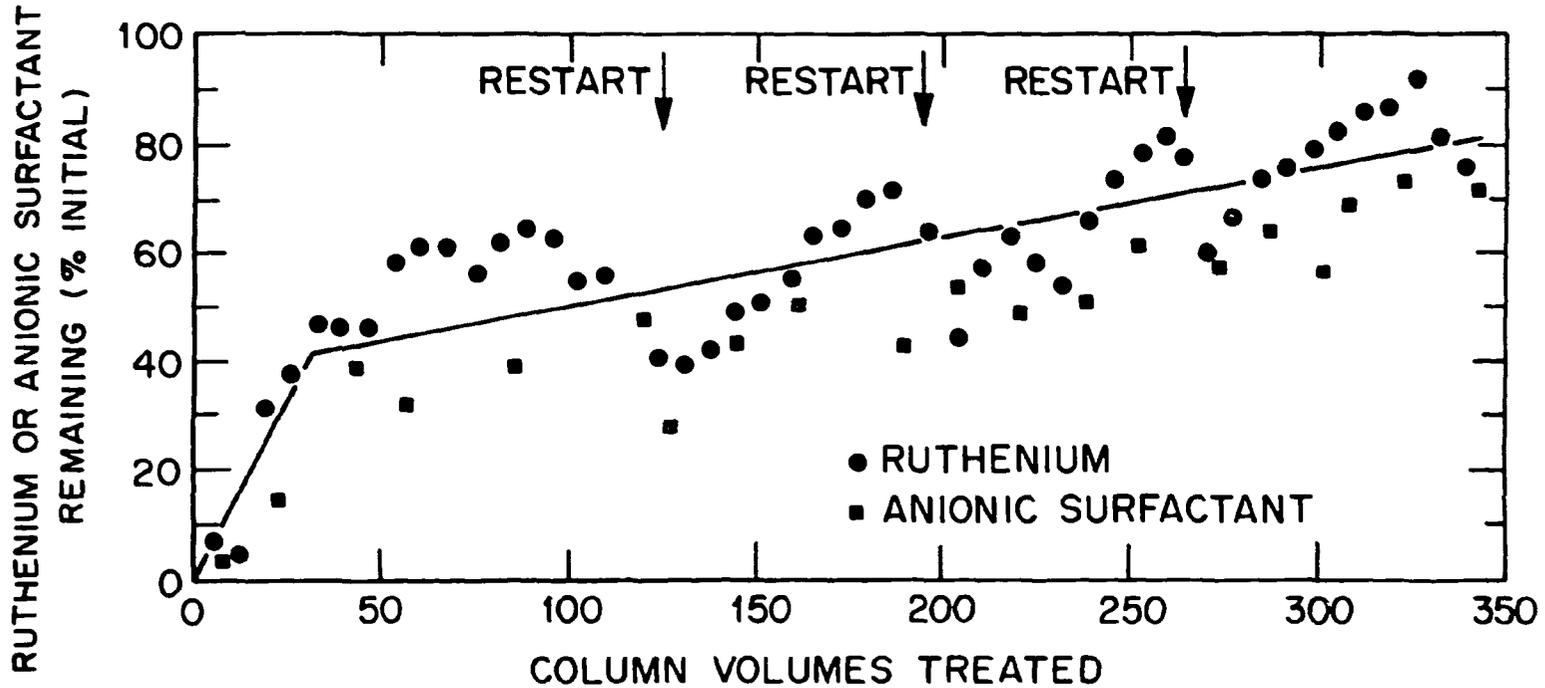


Fig. 11. Interfacial tension and phase-separation time for dried SRP solvent after passage through 1.3 g of activated alumina (60-120 mesh) at 22°C.



RUTHENIUM AND ANIONIC SURFACTANTS (BY MB TEST)
 REMAINING IN TREATED SOLVENT. LINE IS FOR
 RUTHENIUM DATA.

Fig. 12. Ruthenium and anionic surfactant remaining in dried SRP solvent after passage through 1.3 g of activated alumina (60-120 mesh). [Determined at 653 nm by the methylene blue test.]

The lack of any trend in plutonium retention with volume of solvent treated suggests that the higher values measured may be due to entrainment.

The alumina column was washed with hexane to remove TRP and any other loosely held species and then was redried at 290°C. After regeneration, the column capacity was only ~75 column-volumes. Additional regeneration tests are discussed later in this report.

3.2.2 Tests Using Third SRP Solvent

The third batch of SRP solvent was used for the most extensive series of tests with activated alumina. This solvent was similar to the second batch of SRP solvent except that the phase-separation time of the solvent after scrubbing with sodium carbonate/tartrate and filtering through Whatman #40 paper was significantly shorter.

Figure 13 shows a graph of the interfacial tension and phase-separation time vs sodium carbonate/tartrate for our series of tests with the activated alumina bed at 22 and 45°C. The capacity of the bed was slightly higher at 22°C than at 45°C, although the difference (~30%) could be due to normal variability between tests. Figures 14 and 15 show the values for ruthenium and anionic surfactant removal for the tests at 22 and 45°C; the differences between results of the two tests were minimal. As in the tests with the second batch of SRP solvent, the values for removal of ruthenium and anionic surfactants were nearly identical.

The columns from the two runs using the new solvent were tested for regeneration using two procedures. The column bed used in the first test at 22°C was treated with 16 M HNO₃ at 100°C, attempting to decompose or desorb the material adsorbed on the activated alumina. The column was then washed with water and dried at 300°C. The solvent treated by passage through this column had a phase-separation time longer than that for untreated solvent and a slightly improved interfacial tension over less than 20 column-volumes. The column bed that had been used at 45°C was tested for regeneration by contacting with 3 M NaOH to determine whether the NaOH would decompose or desorb some of the adsorbed material and etch the surface of the activated alumina. The column was then

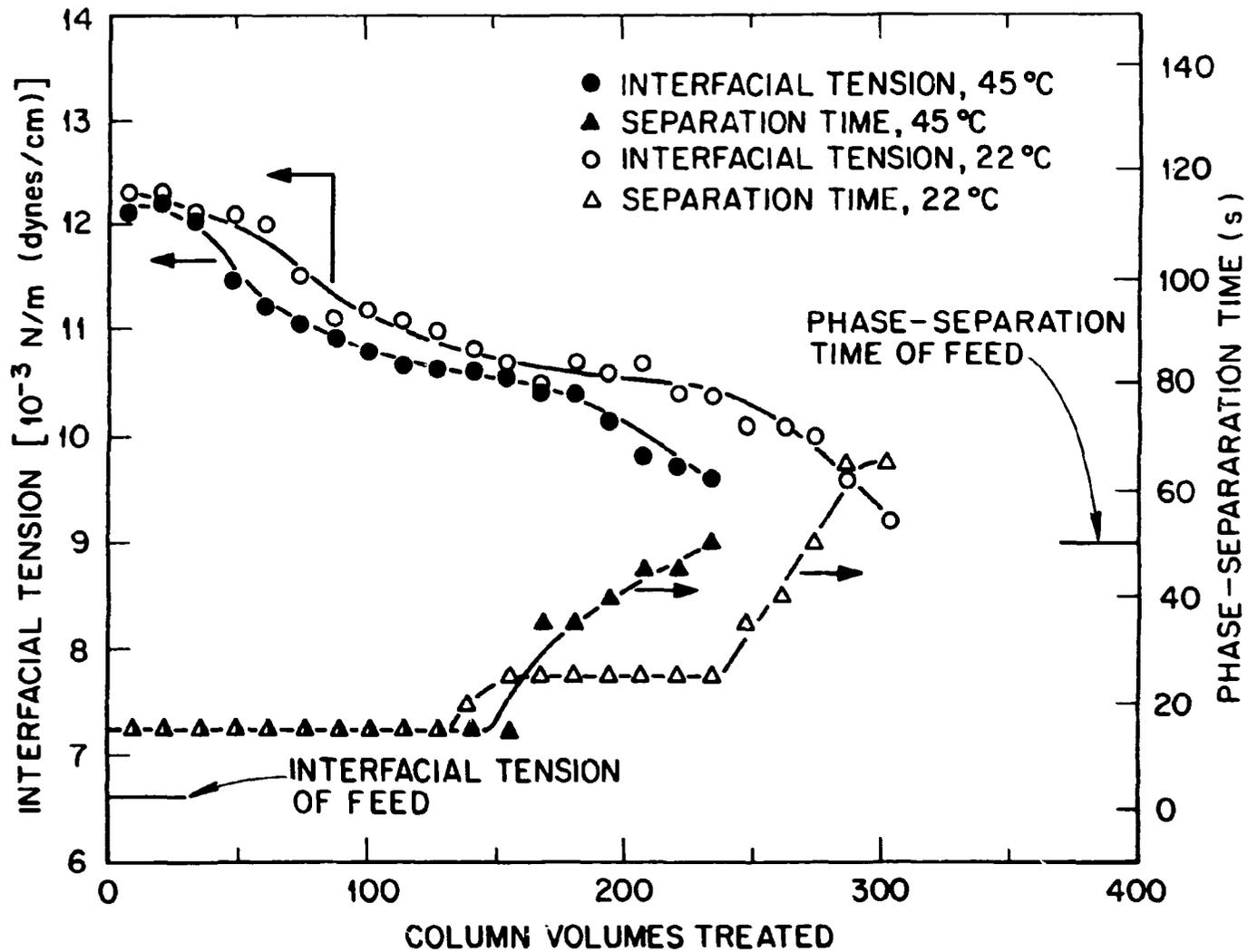


Fig. 13. Interfacial tension and phase-separation times for dried SRP solvent after passage through activated alumina (60-120 mesh).

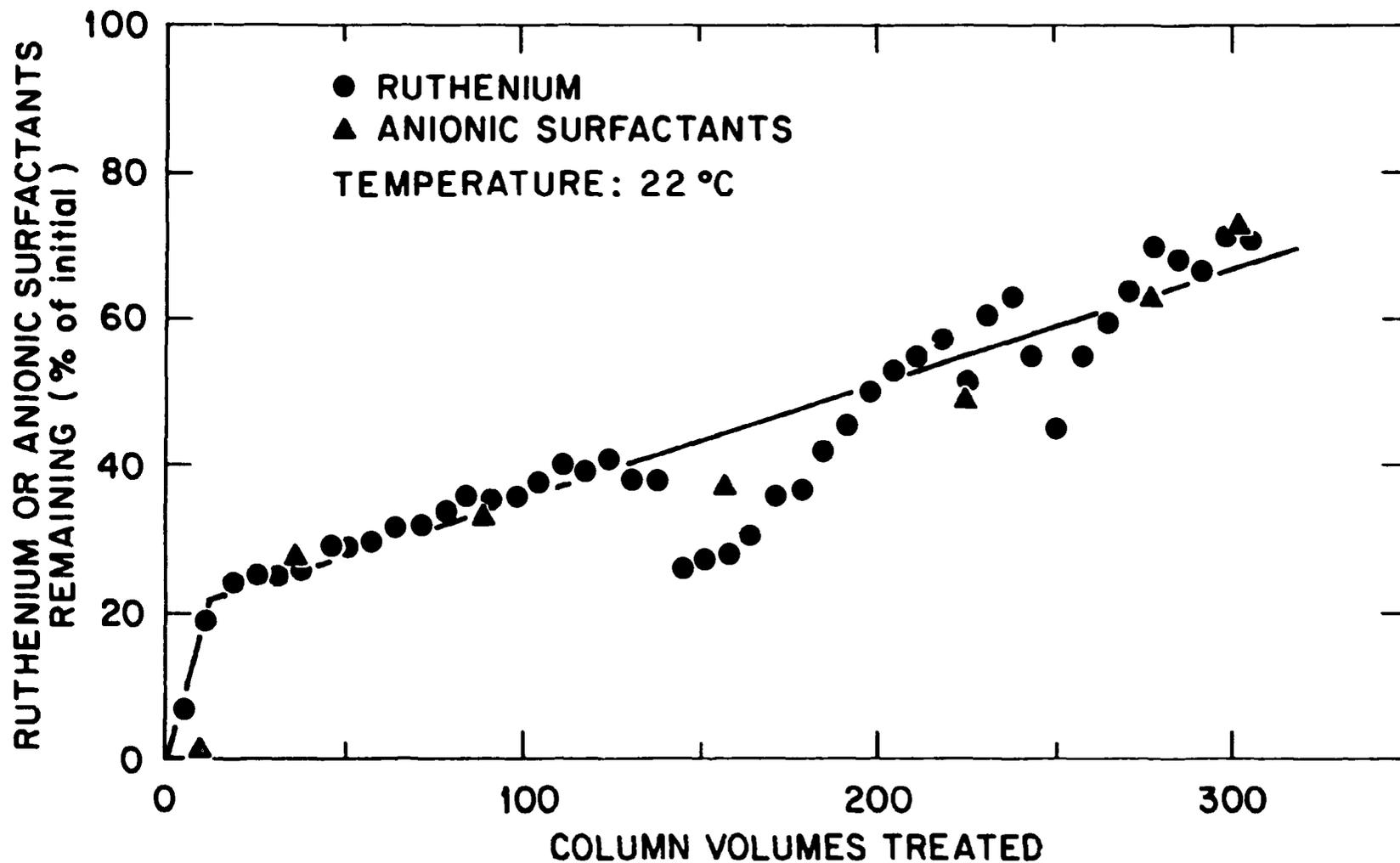


Fig. 14. Ruthenium or anionic surfactants remaining in dried SRP solvent after passage through activated alumina (60-120 mesh) at 22°C.

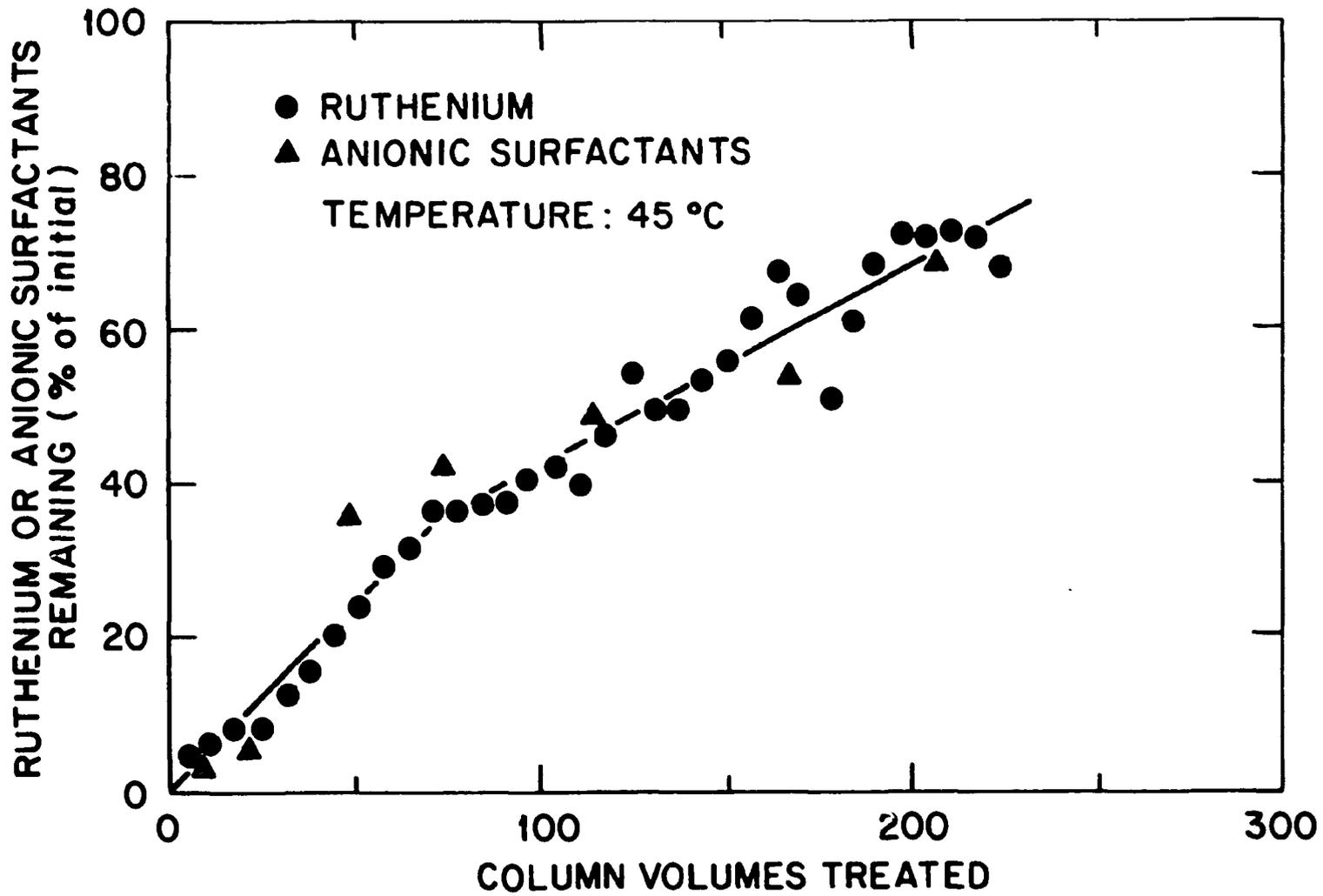


Fig. 15. Ruthenium or anionic surfactants remaining in SRP solvent after passage through activated alumina (60-120 mesh) at 45°C.

washed with water, with dilute HNO_3 , again with water, and dried at 300°C . This column, when subsequently tested with SRP solvent at 22°C , showed no improvement in phase-separation time and very little improvement in interfacial tension.

Since centrifugation to remove particulates improved the phase-separation time of the SRP solvent, a portion of the solvent was centrifuged before being dried and was then tested with an activated alumina column at 22°C . During the centrifugation, no visible material was removed. Figure 16 shows the interfacial tension and phase-separation times vs sodium carbonate/tartrate. The capacity of the column, about 240 column-volumes, was intermediate between the capacities found for treatment of uncentrifuged solvent at 22 and 45°C . Thus, the centrifugation did not result in an improvement in column capacity. The percentages of ruthenium and anionic surfactants removed in this experiment are shown in Fig. 17. These percentages are similar to those found in other tests using activated alumina, and the values for ruthenium and anionic surfactant removal are nearly the same.

4. CONCLUSIONS

The use of activated alumina after drying of the solvent with warm air appears to be a feasible secondary cleanup method for the Purex system. Operation at 22 and 45°C gave similar results. The degree of ruthenium removal was sensitive to minor changes in the operating conditions, suggesting that improvement can be made, perhaps by increasing the solvent residence time. This method has been tested only with solvent which was contacted with base; if the final contact was with acid, the compounds in the solvent might have different properties and lead to different results. Efforts to regenerate the activated alumina had limited success; the best technique was found to be washing with hexane, followed by drying. The activated alumina system should be tested in conjunction with an operating Purex system to confirm the improvements observed in these small-scale tests.

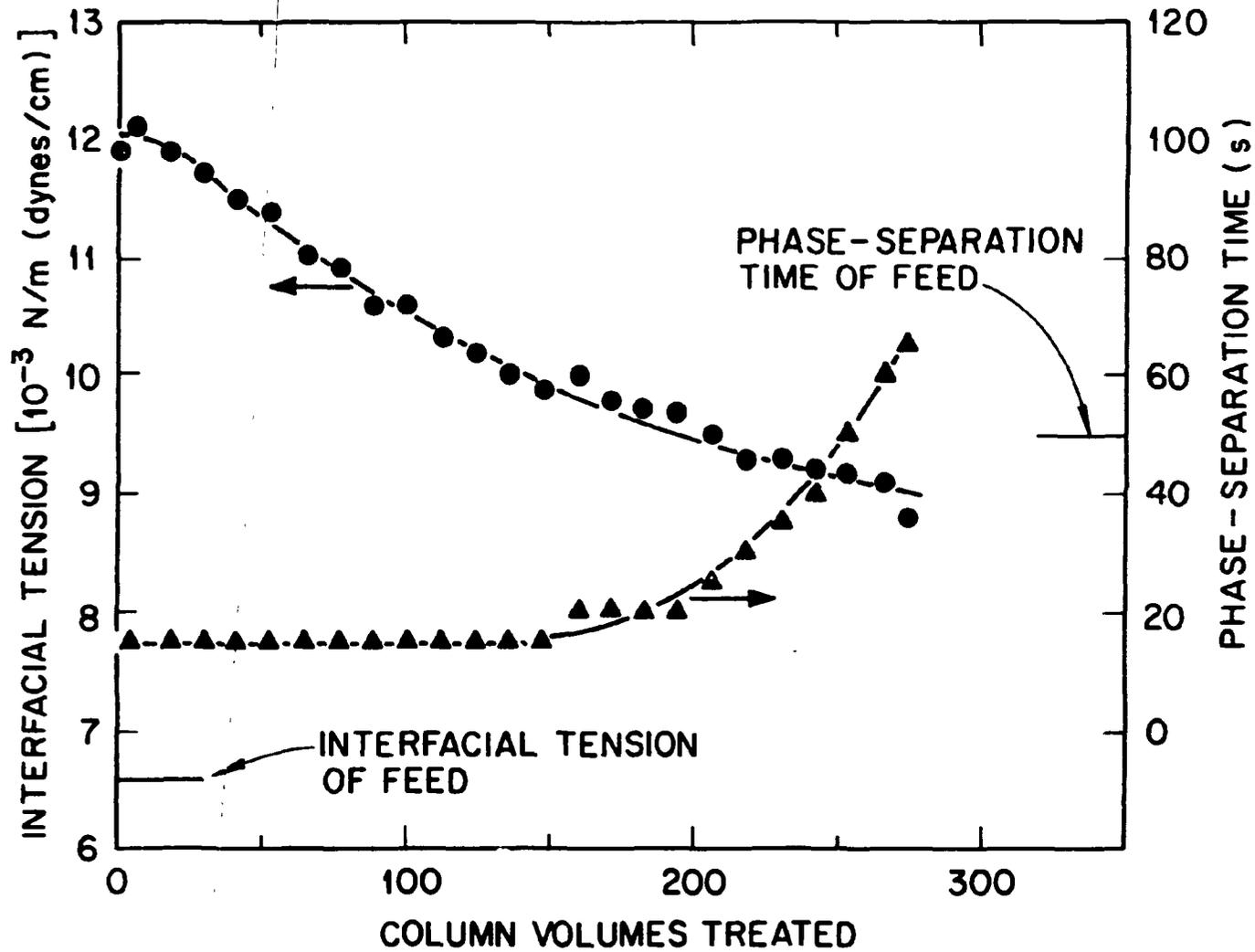


Fig. 16. Interfacial tension and phase-separation time for centrifuged, dried SRP solvent after passage through activated alumina (60-120 mesh) at 22°C.

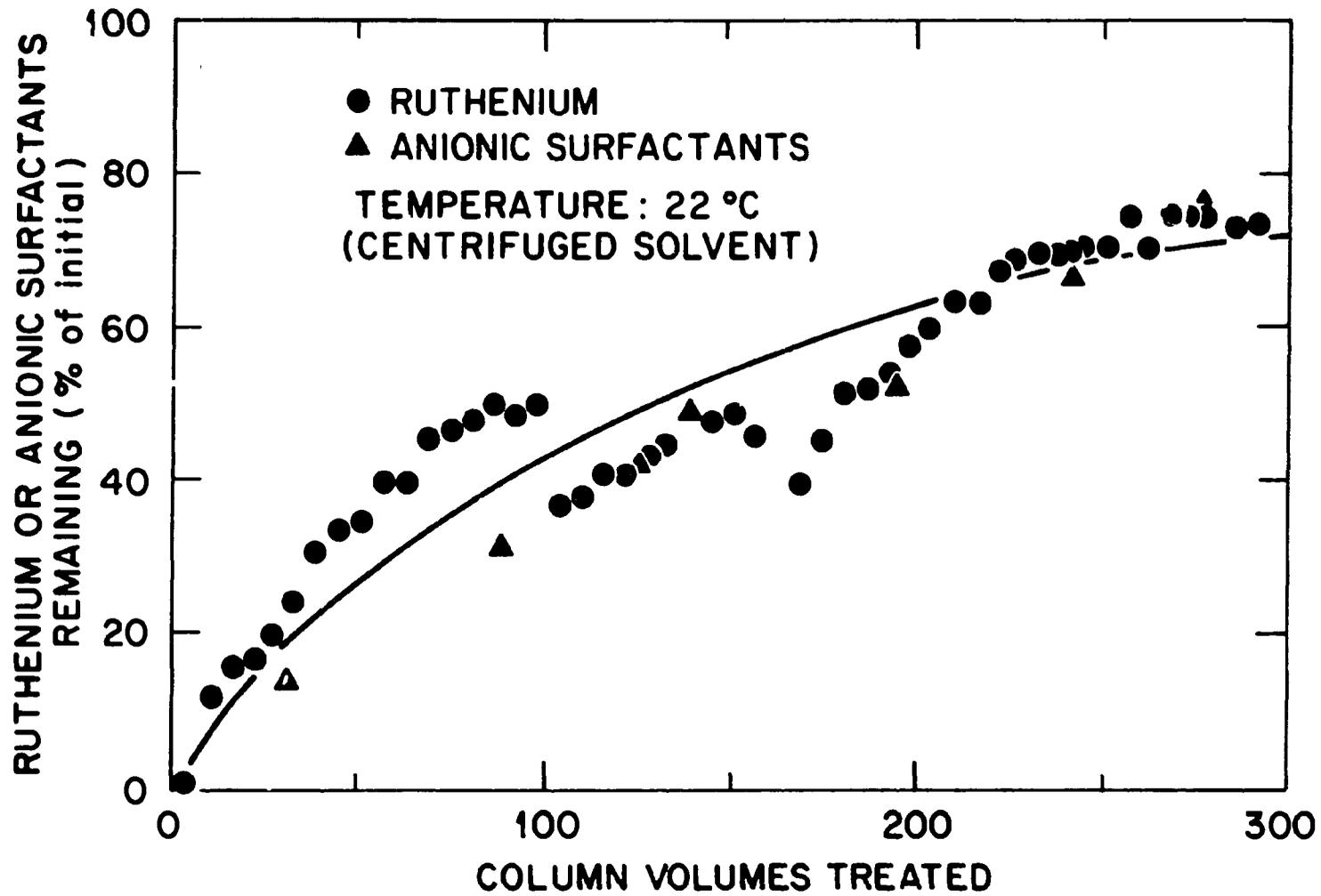


Fig. 17. Ruthenium or anionic surfactant removal from centrifuged, dried SRP solvent after passage through activated alumina (60-120 mesh) at 22°C.

Our study was unable to examine a number of important factors in this system, and we did not investigate others in sufficient detail. Important aspects of any future study should include the effects of residence time, the properties of other types of activated alumina, the effects of acidic or basic contaminants, and additional testing of regeneration procedures.

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