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SEPARATION OF PROTACTINIUM FROM THORIUM IN NITRIC ACID SOLUTIONS BY SOLVENT EXTRACTION WITH TRIBUTYL PHOSPHATE OR BY ADSORPTION ON PULVERIZED UNFIRED VYCOR GLASS OR SILICA GEL

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ABSTRACT

Two methods are being investigated for the separation and recovery of protactinium from short-decayed thorium fuel in nitric acid solutions. The Pa^{233}, thorium, and uranium may be coextracted from highly acidic feed solutions with 30% tributyl phosphate, or the protactinium may be preferentially adsorbed on pulverized unfired Vycor glass or silica gel. Major effort has been on the adsorption method.

Adsorption experiments with tracer concentrations of Pa^{233} in nitric acid solutions showed distribution coefficient maxima for protactinium of about 1000, 325, and 175 from 6 to 10 $\underline{M}$ HNO_3 for laboratory-prepared silica gel, unfired Vycor, and commercial silica gel, respectively. Unfired Vycor, a commercial, leached borosilicate glass containing 96% SiO_2 and about 3% B_2O_3, was used for most of these preliminary studies. Fired Vycor glass adsorbed little or no protactinium.

The adsorption coefficient of protactinium by unfired Vycor glass from nitric acid solutions increases as the contact time increases or as the particle size of the glass decreases and is dependent on the concentration of salt or nitric acid in the solution. The adsorbed protactinium may be eluted with oxalic or tartaric acids. In column experiments, as much as 10 mg of Pa^{231} per gram of unfired Vycor was adsorbed from 0.50 $\underline{M}$ Th, 11 $\underline{M}$ HNO_3, 0.1 $\underline{M}$ Al, 0.10 g/liter Pa^{231}, and <0.1 $\underline{M}$ F. With a column loading of 2 mg Pa/g glass, more than 99% of the protactinium was adsorbed from a similar feed solution containing 0.05 g/liter of Pa^{231}. On increasing the loading to about 5 mg Pa/g glass, a total of about 95% of the protactinium had been adsorbed. Greater than 99% was eluted with 0.5 $\underline{M}$ oxalic acid, 98% of which was eluted at a concentration of 4.1 mg/ml Pa^{231}. The protactinium concentration in the feed solutions were stable, showing no change other than the ±10% analytical variation after standing one month at room temperature in plastic containers. Although the optimum conditions for column operations have not been determined, decontamination factors of protactinium from thorium, uranium, ruthenium, zirconium-niobium, and total rare earths of 6×10^3, 1.6×10^4, 4×10^3, 3, and 5.8×10^5, respectively, have been obtained in tracer experiments.

Batch countercurrent scouting experiments with tracer protactinium showed that about 90% of the protactinium was extracted with the thorium and uranium from 5 $\underline{M}$ HNO_3--1 $\underline{M}$ $Al(NO_3)_3$ solutions, with a decontamination factor of 70 from ruthenium and about 10^5 from rare earths. About half of the zirconium-niobium extracted also. A partitioning strip or a second solvent extraction cycle may be provided to separate protactinium from the uranium and thorium and to increase decontamination from fission products.

INTRODUCTION

The Consolidated Edison Thorium Reactor fuel is to be irradiated to an average of 18,000 Mwd/metric ton in a 2-year cycle. At the time of discharge, the fuel will contain about 66 kg of uranium, 1 kg of protactinium, and 18 kg of fission products per metric ton of thorium. The protactinium in the feed represents only about 1.5% of the potential fissionable material, but it contributes about half of the activity of the fuel solution. Removal of protactinium from the dissolved fuel prior to extraction of the uranium and thorium not only provides a means of recovering the protactinium but also decreases difficulty in the solvent extraction cycle. Adsorption of protactinium on unfired Vycor glass or silica gel is being investigated for this application. An alternative to this is the coextraction of uranium, thorium, and protactinium from dilute aqueous solutions with 30% tributyl phosphate (TBP). Laboratory experiments with simulated feed solutions have indicated satisfactory separation of products from the high-cross-section fission products by both these methods, but neither method has been sufficiently investigated that it may be recommended for processing actual reactor fuels. This report will be concerned primarily with the adsorption of protactinium on unfired, pulverized Vycor. In addition, some preliminary silica gel adsorption and solvent extraction data will be presented.

ADSORPTION OF PROTACTINIUM ON UNFIRED VYCOR GLASS

The successful adsorption of Nb^{95} on pulverized, unfired Vycor glass from zirconium nitrate-nitric acid solutions containing Zr-Nb tracer[1] led to the study of the adsorption properties of this glass for protactinium from nitrate solutions. Other workers had shown that quartz glass, plastic, and metal surfaces will adsorb protactinium from nitrate solutions,[2] and that silica gel will remove protactinium from dilute[3] and 6 $\underline{M}$ HNO_3.[4]

Unfired Vycor is a leached borosilicate glass manufactured by Corning Glass Works of Corning, New York. This material, which is sometimes referred to as "thirsty" glass, is the porous, unshrunk form of Corning's basic 7900 Vycor glass. The pores average about 4 mμ in diameter, and the surface area ranges from about 150 to 200 meter^2/g. Chemically it is 96% silica with the balance mainly boric oxide (Table 1).

Table 1. Specification of Unfired Vycor Glass No. 7930

Apparent dry density, g/cc	1.45
Average pore diameter, A	4
Surface area, m^2/g	150-200
Void space, %	28
Approximate composition (anhydrous):	
SiO_2, %	96
B_2O_3, %	3
R_2O_3, %	0.4
Alkali	trace
Arsenic	trace

The distribution coefficient of unfired Vycor for protactinium, that is the counts per minute per gram of glass divided by the counts per minute per milliliter of aqueous after contact, varies directly with the nitric acid concentration and reaches a maximum of about 3000 in 6 M HNO_3 (Fig. 1). These data were obtained in 60-min batch equilibrations of nitric acid solutions containing about 5×10^5 counts min^{-1}ml^{-1} of Pa^{233} with 10 g/liter of 100 to 200 mesh unfired Vycor.

The adsorption of protactinium is greatly influenced by the presence of other nitrate salts in the solution. This is shown by the triangular marks in Fig. 1 and are the results of similar batch equilibrations using aqueous solutions containing 0.1 N HNO_3 and a total of 6 N nitrate, with the designated cation supplying the nitrate. The maximum coefficient was observed with nitric acid alone, and the minimum value, 20, with thorium nitrate. Intermediate values, in descending order, were obtained with aluminum, sodium, ammonium, calcium, and lithium nitrate, respectively.

The effectiveness of thorium in decreasing the adsorption of protactinium from nitric acid varies inversely with the nitric acid in the system. In batch tests 10 g/liter 100 to 200 mesh adsorbent and 60-min

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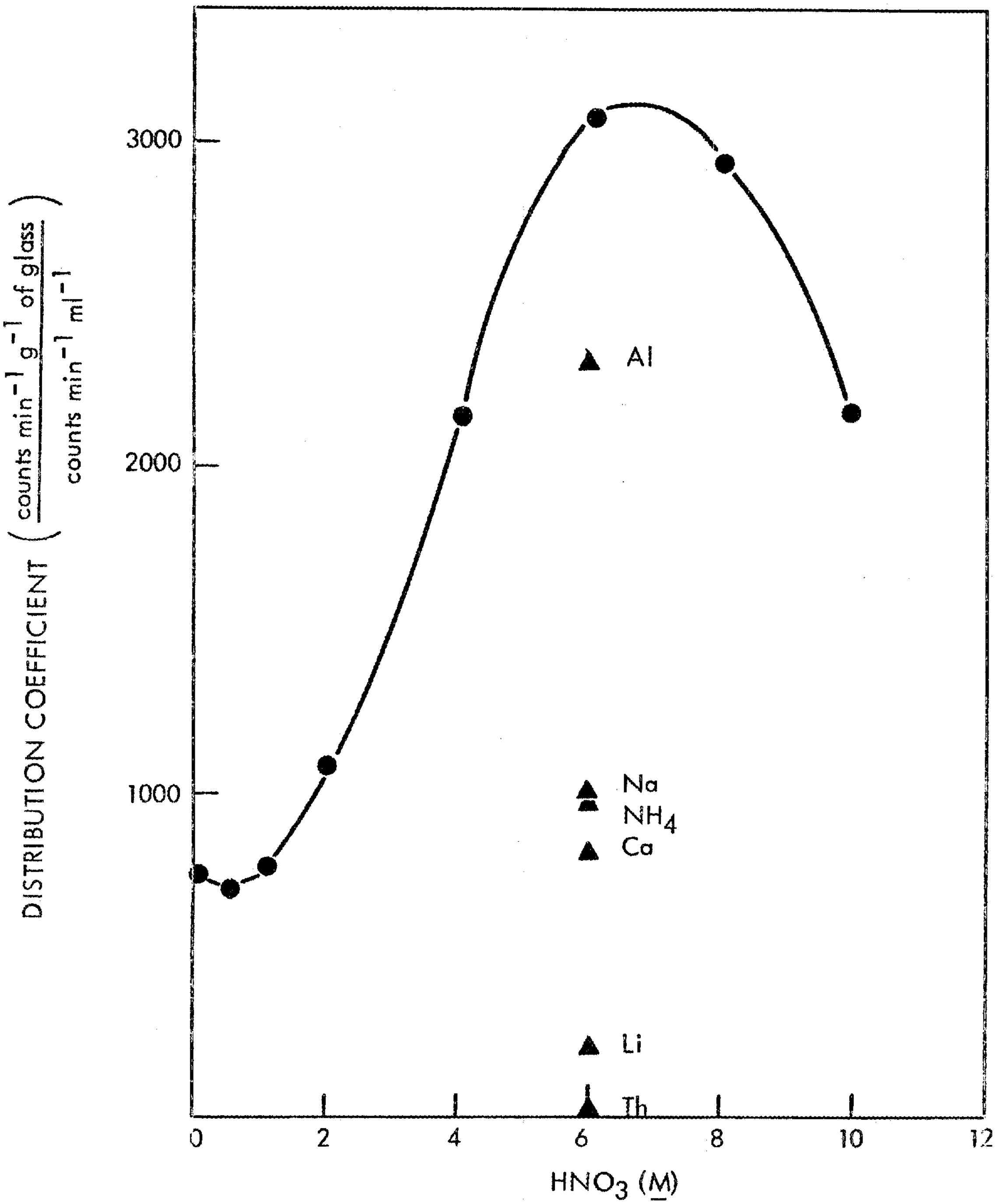

Fig. 1. Adsorption of Pa^{233} on 100 to 200 Mesh Unfired Vycor.

equilibrations, 0.1 M HNO_3 required only about 0.03 M $Th(NO_3)_4$ to decrease the coefficient by a factor of 2, whereas 2 M HNO_3 solutions required about 0.3 M Th to produce the same effect. After this initial decrease, the effect of additional thorium was approximately the same for both acidities (Fig. 2).

A decrease in the particle size of unfired Vycor decreases the time required to reach equilibrium in the adsorption of protactinium from nitric acid. A series of batch equilibrations were made by contacting 10 g/liter of unfired Vycor of different mesh size with 6.1 M HNO_3 containing about 9×10^5 c/m.ml Pa^{233} for 5 to 300 min. The resulting distribution coefficients increased as the contact time increased, or the particle size decreased (Fig. 3).

An increase in adsorption with time was observed in all the nitric acid solutions examined, 0.1 to 10 M HNO_3. An increase in the distribution coefficients obtained in experiments using 10 g/liter of 100 to 200 mesh Vycor and 0.1 to 10 M HNO_3 continued to be observed after 4 hr of equilibration time (Fig. 4).

In the separation and recovery of protactinium from short-decayed thorium fuel dissolved in nitric acid, a major apprehension has been the stability of the thorium-protactinium-nitrate solutions that would be encountered. Stable solutions have been prepared simulating those which would be produced in acid thorex processing[5] short-cooled 18,000 Mwd/metric ton Th Consolidated Edison Thorium Reactor Fuel. A synthetic dissolver solution equivalent to one that would be obtained in the dissolution of about 10-day-cooled fuel was prepared containing 116 g/liter Th, about 70 mg/liter Pa^{231}, 11 M HNO_3, 0.1 M Al, and 0.03-0.1 M F. In addition, a solvent extraction feed solution having more than twice the maximum concentration of protactinium obtainable with freshly discharged fuel, was made containing 40 g/liter of Th, 90 mg/liter of Pa^{231}, 5 M HNO_3, 1 M Al, and 0.03-0.1 M F. These solutions were allowed to stand one month at room temperature in plastic containers and analysed periodically for protactinium. There was no change other than a ±10% scatter in the analytical values over the entire period. Furthermore, these solutions showed no change in protactinium concentrations after con-

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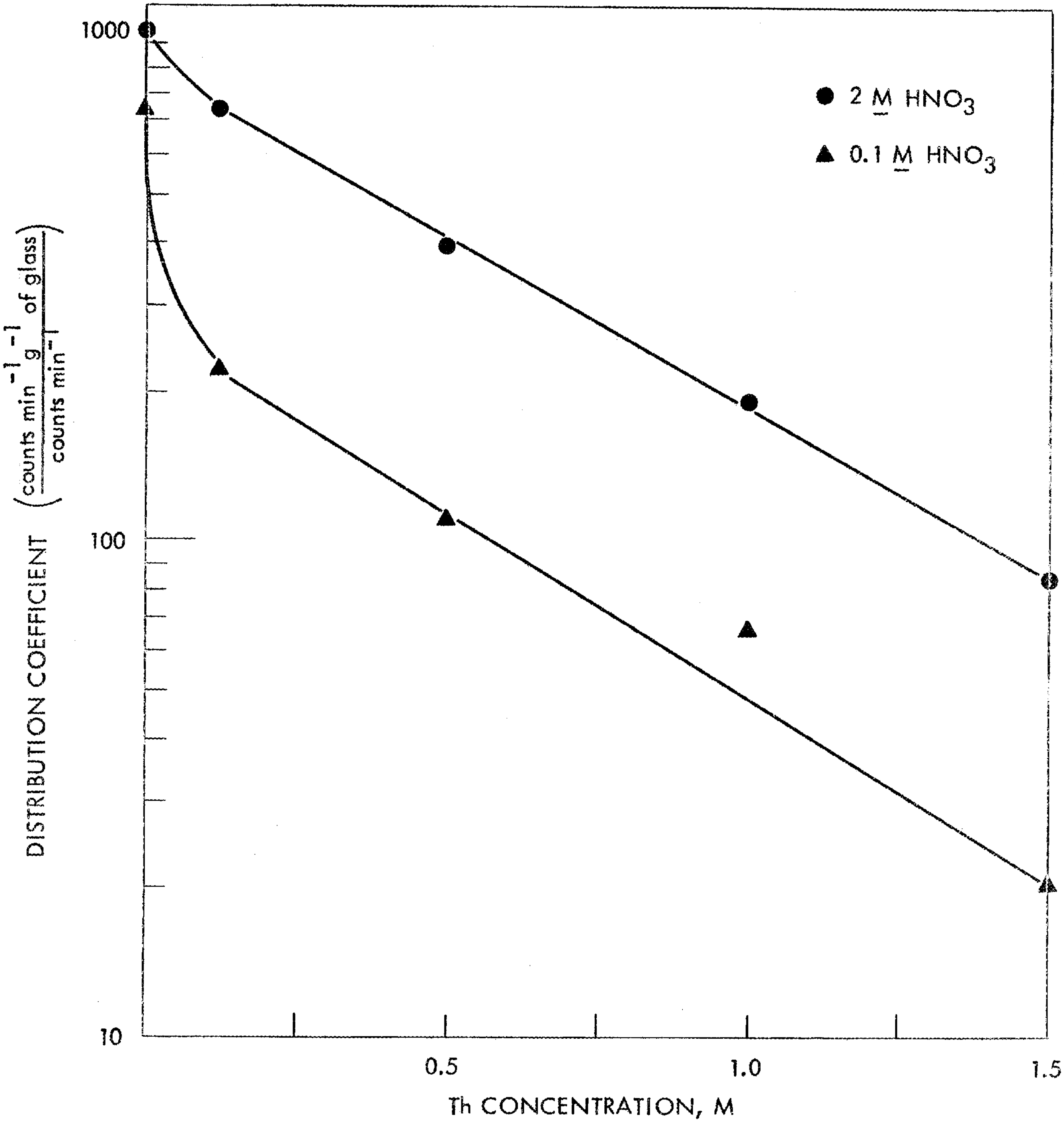

Fig. 2. Effect of Thorium Nitrate on Adsorption of Pa^{233} on Unfired Vycor from Nitric Acid.

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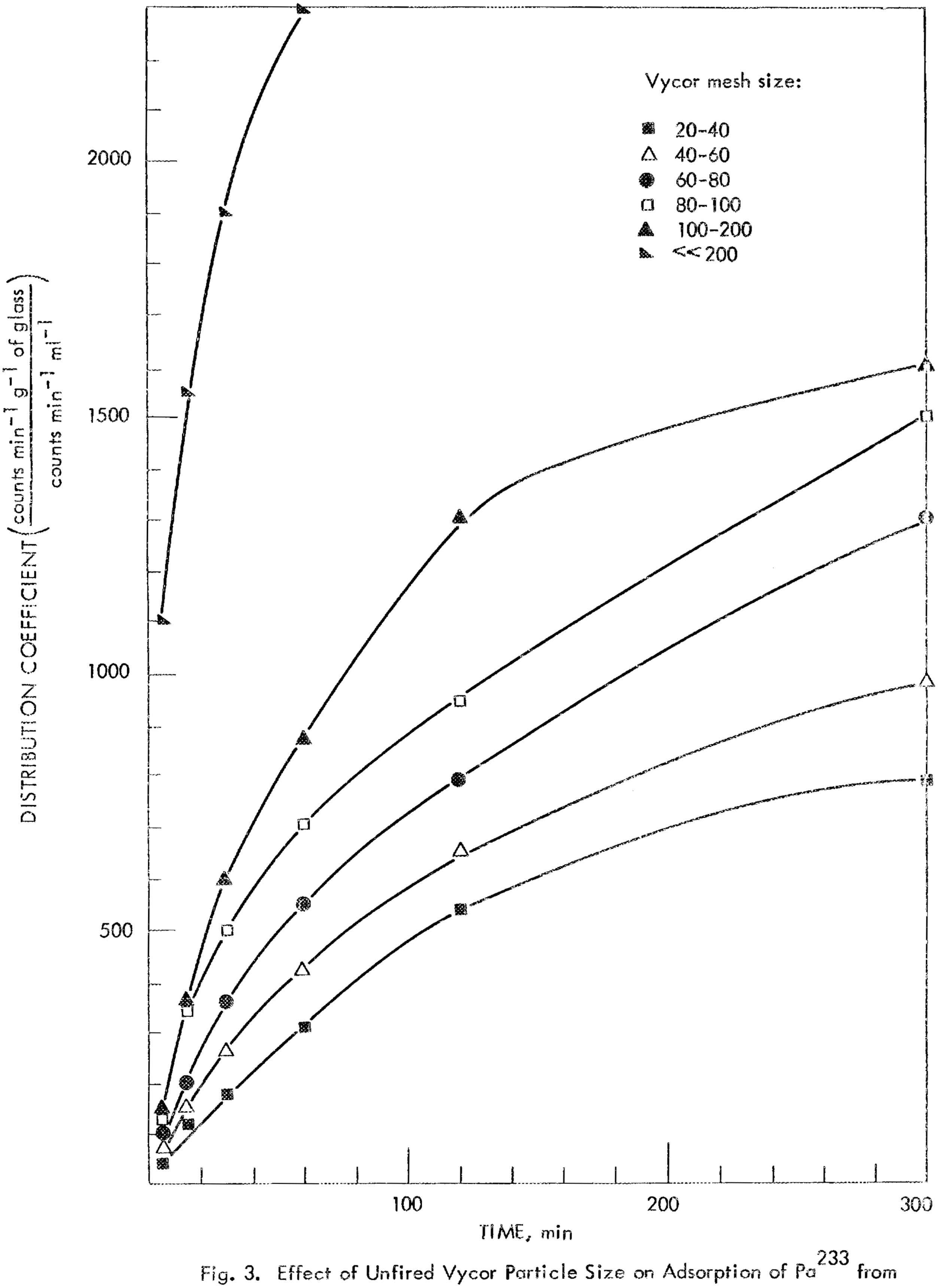

Fig. 3. Effect of Unfired Vycor Particle Size on Adsorption of Pa^{233} from 6 $\underline{M}$ HNO_3.

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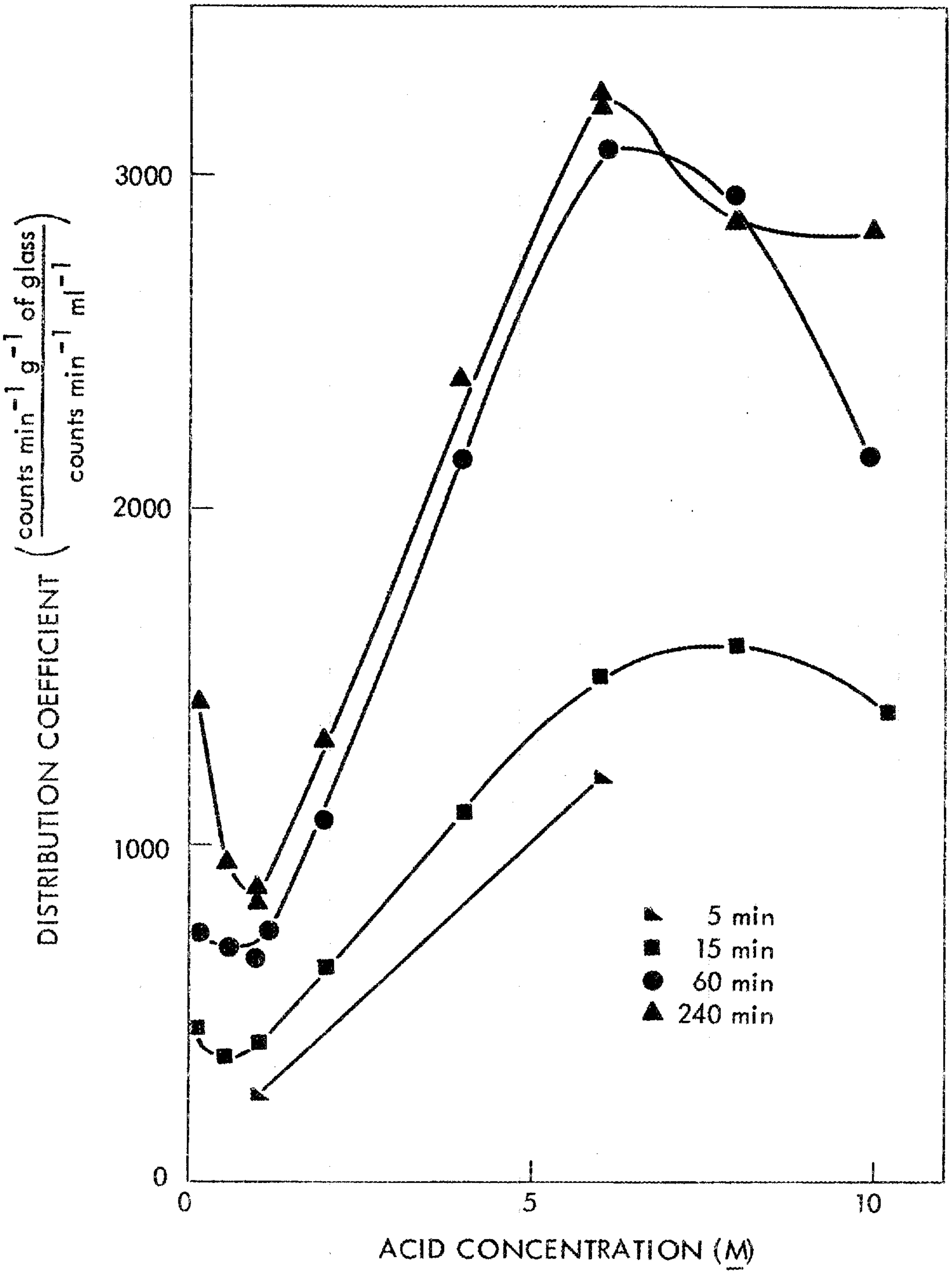

Fig. 4. Effect of Nitric Acid Concentration on Adsorption of Pa^{233} by Unfired Vycor After Varying Contact Times.

tacting with type 347 stainless steel surfaces with and without welds, for one week at room temperature. The Pa^{231} concentration of these solutions were ±6% of the original after contacting 0 and 2 cm^2 stainless steel/ml for one week.

Column runs using simulated short-cooled Consolidated Edison dissolver solutions have shown that the capacity of unfired Vycor is greater than 10 mg Pa/gm of glass. A column 0.4 cm diam and about 9 cm long containing 1 gm of 60 to 80 mesh unfired Vycor adsorbed 10 mg of Pa^{231} from a solution containing 116 g/liter of Th, 11 $\underline{M}$ HNO_3, 0.1 $\underline{M}$ Al, 100 mg/liter of Pa^{231} and less than 0.1 $\underline{M}$ F. A total of 66% of the protactinium was adsorbed. The average flow rate for the entire run was 1.8 $ml/min.cm^2$. Greater than 99% of the protactinium adsorbed on the column was recovered at a concentration equivalent to 12 times the feed concentration by eluting with 0.5 $\underline{M}$ oxalic acid (Fig. 5). Subsequent qualitative tests have shown that tartaric acid is probably equally as effective as an eluting agent. The thorium decontamination factor was 6300 and an alpha analysis of the product showed that it was greater than 99% Pa^{231}. In the adsorption portion of the run, the data show that the adsorption ranged from about 90% initially to about 50% at the end of the run. The initial loss will be discussed later in this report.

The initial Pa^{231} concentration in the raffinate from a larger column using a slower flow rate was about 4% of the feed concentration and remained fairly constant until the glass was loaded to about 4 mg Pa^{231}/g of glass. The column was 1 cm in diameter, 8.64 cm long, and contained 5 gm of 80 to 100 mesh unfired Vycor. The feed solution contained 0.5 $\underline{M}$ Th, 11.1 $\underline{M}$ HNO_3, 0.074 $\underline{M}$ Al, about 0.03 $\underline{M}$ F, and 54 mg/liter of Pa^{231} and was passed through the column at a flow rate of 0.22 $ml/cm^2.min$. At about 4 mg/g of glass, the protactinium concentration in the raffinate slowly increased to about 46% of the initial feed concentration. At this point, 85% of the protactinium had been adsorbed, or about 7.8 mg Pa^{231}/g of glass. The loaded column was then washed with 80 ml of 11 $\underline{M}$ HNO_3 , which removed 13% of the adsorbed protactinium, and then the column was eluted with 0.5 $\underline{M}$ oxalic acid. The first 5 ml of oxalic acid removed 95% of the remaining protactinium, yielding a solution containing

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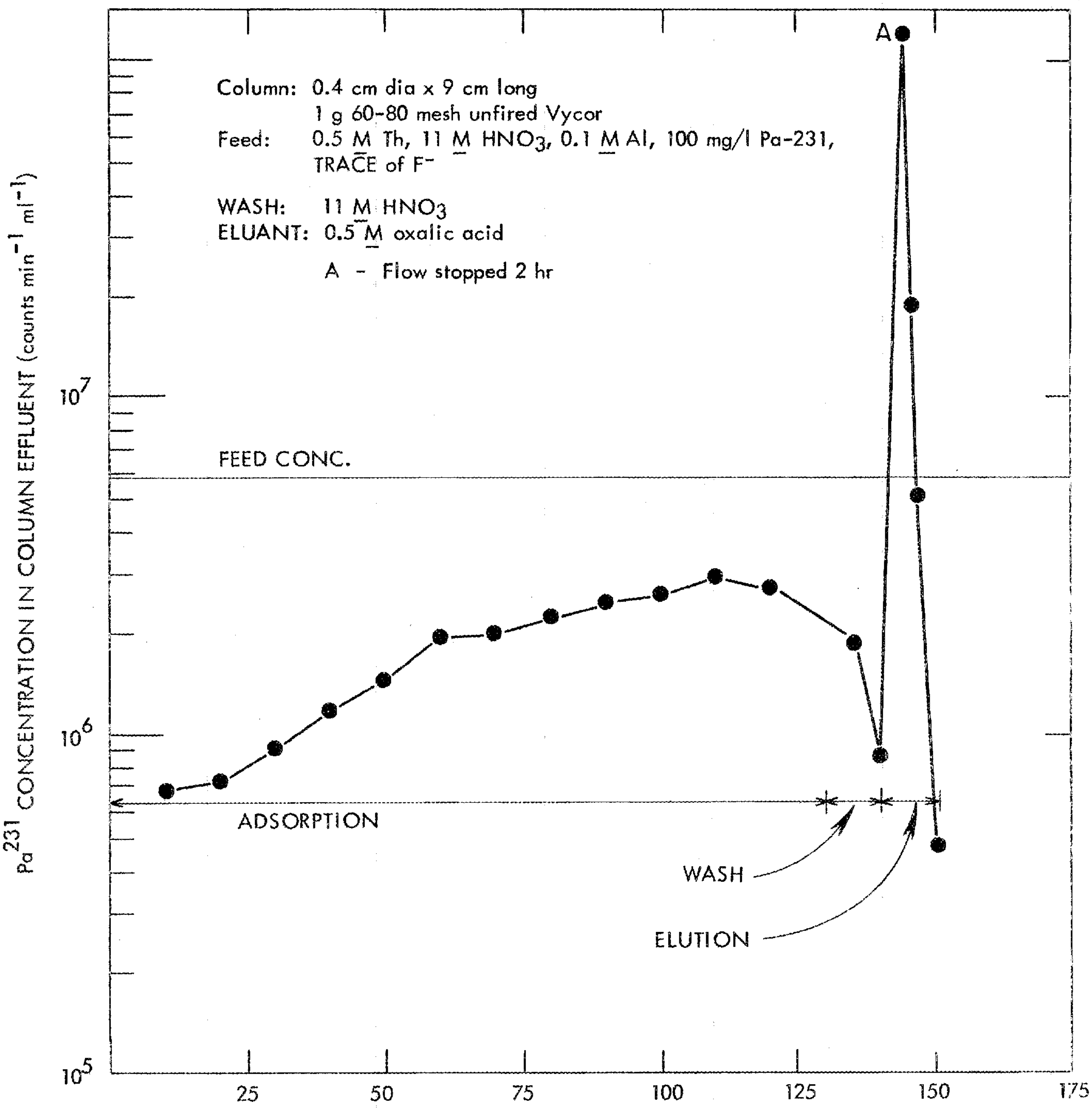

Fig. 5. Sorption of Protactinium with Unfired Vycor.

6.4 mg/ml of Pa^{231}, or about 120 times the concentration of the Pa^{231} in the feed solution. Greater than 99% of the protactinium had been removed after a total of 30 ml of strip solution had passed through the column (Fig. 6).

The initial loss observed in this and the preceding column is due to the fact that part of the protactinium in the feed solution was in an unadsorbable form. This was verified by taking the first 400 ml of the raffinate from the above mentioned column and passing it through an additional column 1 cm in diameter and 8.64 cm long. None of the protactinium adsorbed. However, more than 99% of the protactinium was initially adsorbed from a fresh feed solution prepared from the protactinium contained in the strip solution of this run. The 0.5 $\underline{M}$ oxalic acid solution was boiled down several times with concentrated nitric acid. Then 0.8 $\underline{M}$ HF, 2 $\underline{M}$ $Al(NO_3)$, and a thorium-nitric acid solution was added to produce a solution containing 0.5 $\underline{M}$ Th, 10 $\underline{M}$ HNO_3, 0.04 $\underline{M}$ F, 0.1 $\underline{M}$ Al, and 56 mg/liter of Pa^{231}. This solution was passed through a column of unfired Vycor similar to the last one. Up to a column loading of 2 mg Pa/g of glass, more than 99% of the protactinium was adsorbed from the solution. At 5 mg Pa/g of glass, a total of 95% of the protactinium had been adsorbed. About half way through the run it was necessary to increase the flowrate from about 0.2 ml/cm^2 to about 1.1 $ml\ cm^{-2} min^{-1}$, and this may have caused a premature protactinium breakthrough. However the experiment did show that it is possible to achieve essentially complete adsorption. An investigation is in progress to determine methods for ensuring that all the protactinium is in an adsorbable form.

Although the optimum conditions for column operations have not been determined, decontamination factors of protactinium from thorium, uranium, ruthenium, zirconium-niobium, and total rare earths of 6×10^3, 1.6×10^4, 4×10^3, 3, and 5.8×10^5, respectively, have been obtained in a variety of tracer experiments. These should be regarded as minimum values since no effort has been made to ensure maximum decontamination through appropriate washing solutions and techniques.

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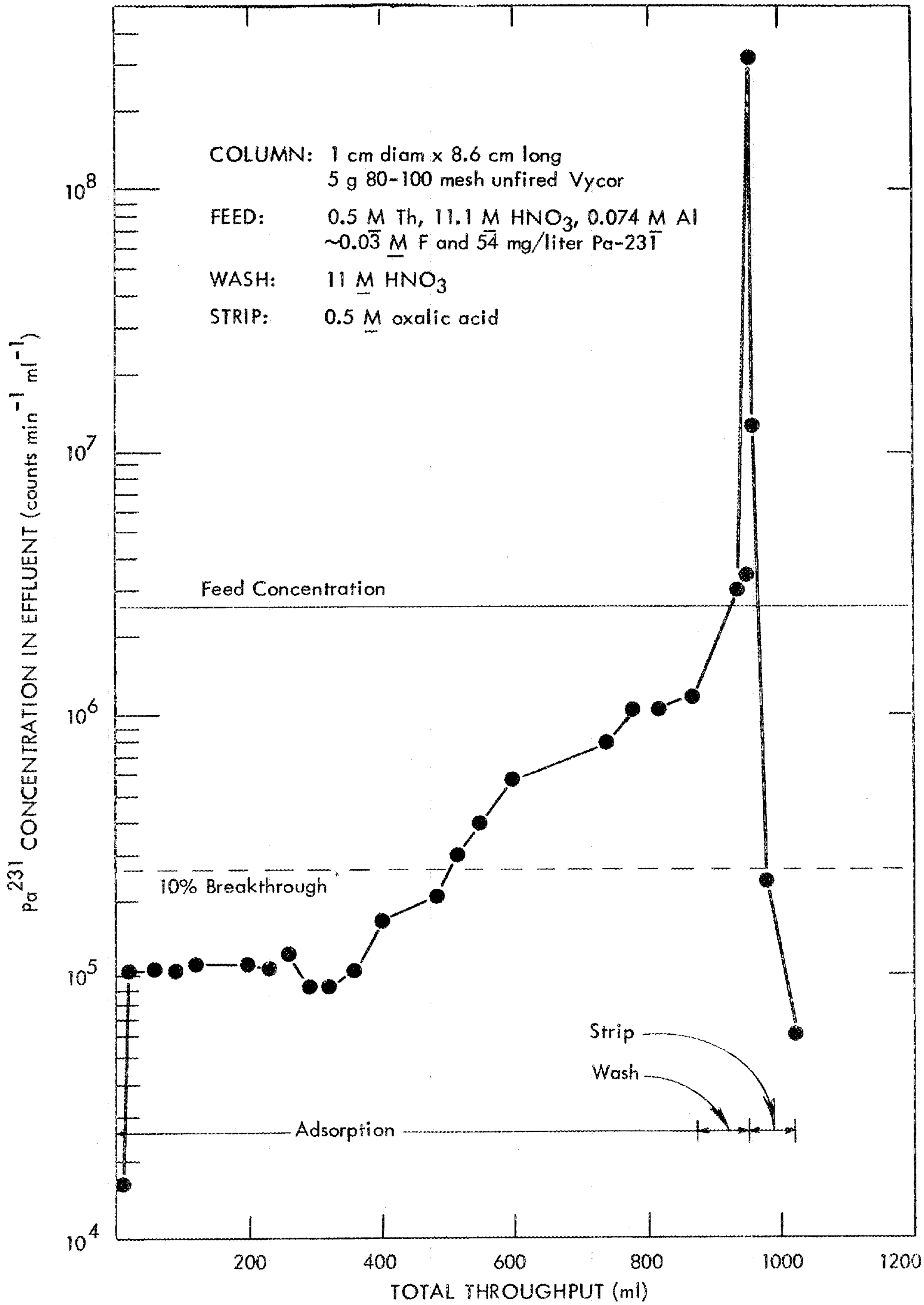

Fig. 6. Sorption of Protactinium with Unfired Vycor.

The present proposal for the dissolution of Consolidated Edison Thorium reactor fuel requires the use of 13 M HNO_3 containing about 0.04 M NaF as a catalyst and 0.04 to 0.1 M $Al(NO_3)_3$ to decrease the rate of corrosion of plant equipment by the fluoride ions. The question immediately arises as to the effect of the fluoride on the corrosion of the finely divided unfired Vycor glass, and the effect of the aluminum concentration on the adsorption of protactinium from fluoride-nitrate systems.

The corrosion of unfired Vycor by fluoride was decreased by a factor of 50 by the presence of 2-1/2 moles of Al/mole F. Solutions of 0.05 M HF, 11 M HNO_3 with and without 0.12 M $Al(NO_3)_3$ present were passed through columns 4 mm diam and 9 cm long containing 1 g of 60 to 80 mesh unfired Vycor at a flow rate of about 1 ml/min.cm^2. After approximately 200 ml had passed through the columns, the merging raffinate with aluminum contained only 0.006 mg/ml Si, whereas the solution without aluminum contained 0.32 mg/ml Si (Fig. 7).

Fluoride decreases the adsorption coefficient of unfired Vycor glass for protactinium. The distribution coefficient was about 1400 in a 24-hr batch equilibration between 10 M HNO_3, about 6 g/liter Th, 1.7×10^6 c/m.ml Pa-233 and 20 g/liter of 60 to 80 mesh unfired Vycor. This coefficient was decreased by factors of 10, 100 and 1000, respectively when 0.01, 0.02 and 0.03 M HF was present. With solutions containing 112 g/liter Th, the distribution coefficient was reduced a factor of 3 by the presence of 0.04 M F(Fig. 8).

About 7-1/2 moles of aluminum per mole of fluoride were required to completely complex the fluoride, so the distribution was not affected in solutions containing only 6 g/liter Th. This mole ratio of aluminum to fluoride also produced the maximum coefficients with feed solutions containing 112 g/liter Th and 0.04 M F^-, however, the coefficient was still about 30% less than the value obtained without fluoride. The coefficient decreased from 1200 with no fluoride present to 345 with a fluoride concentration to 0.04 M. The presence of 7-1/2 moles of aluminum/mole of fluoride increased the coefficient to about 800 (Fig. 9).

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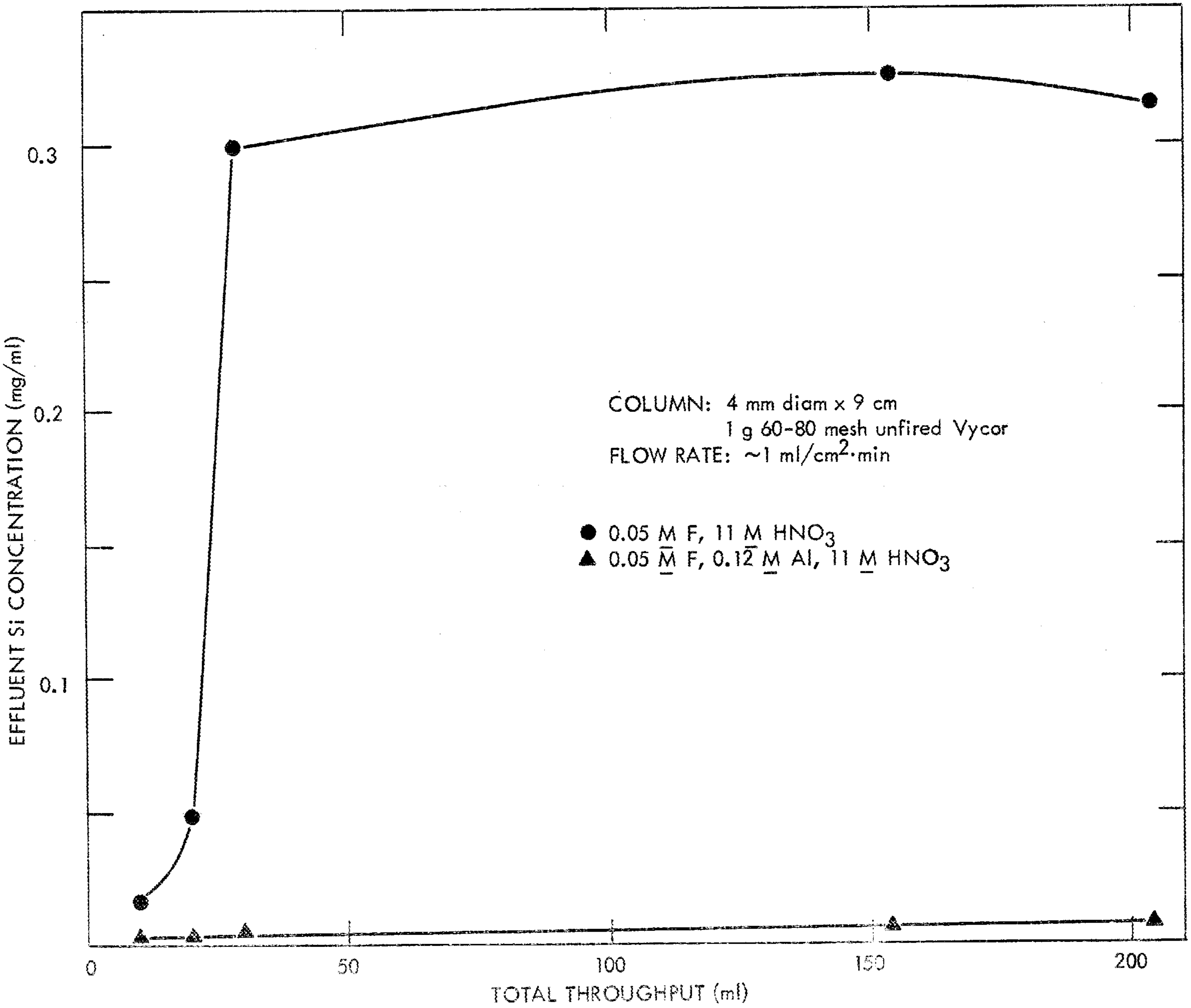

Fig. 7. Corrosion of Unfired Vycor by Fluoride.

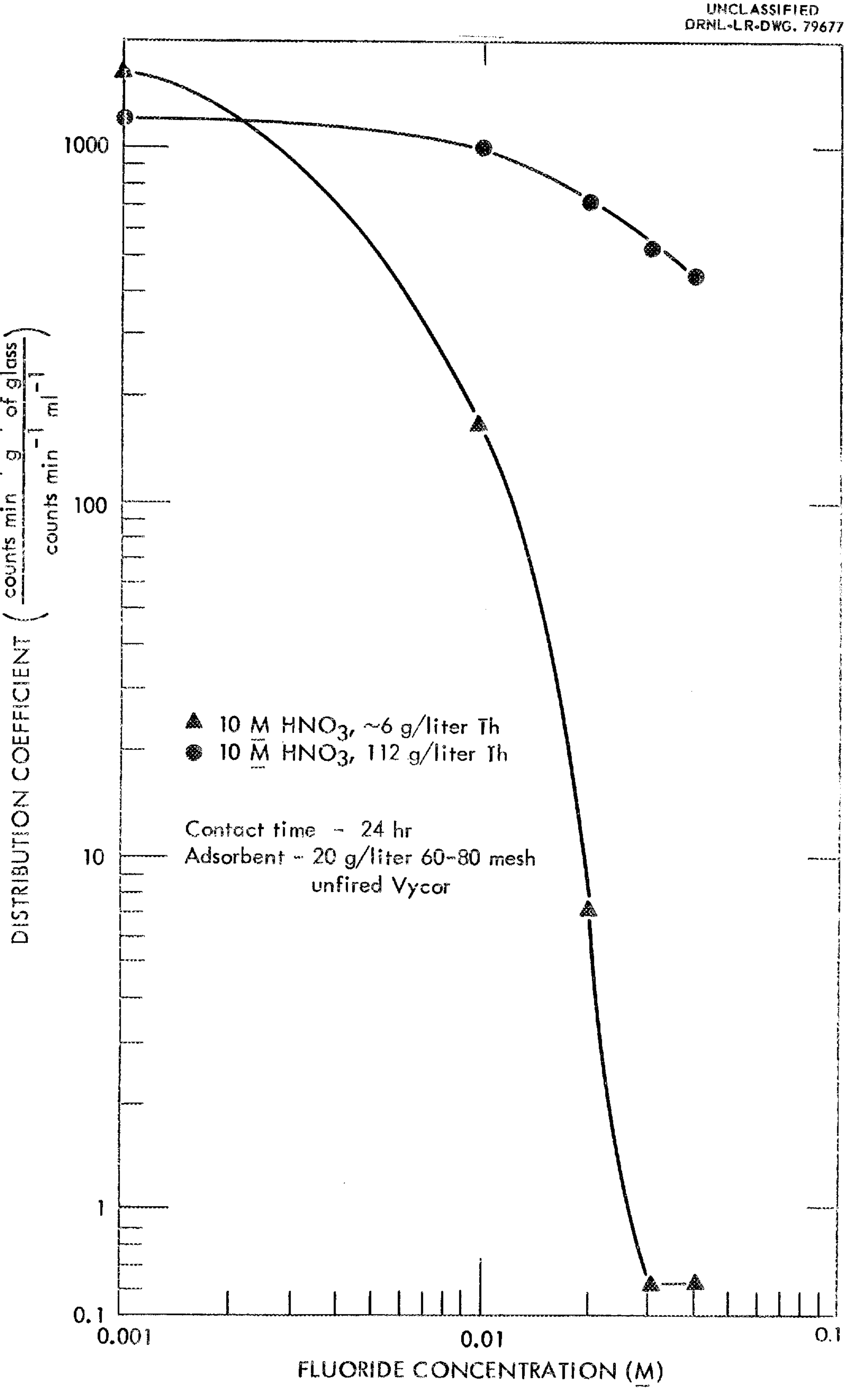

Fig. 8. Effect of Fluoride on Protactinium Adsorption.

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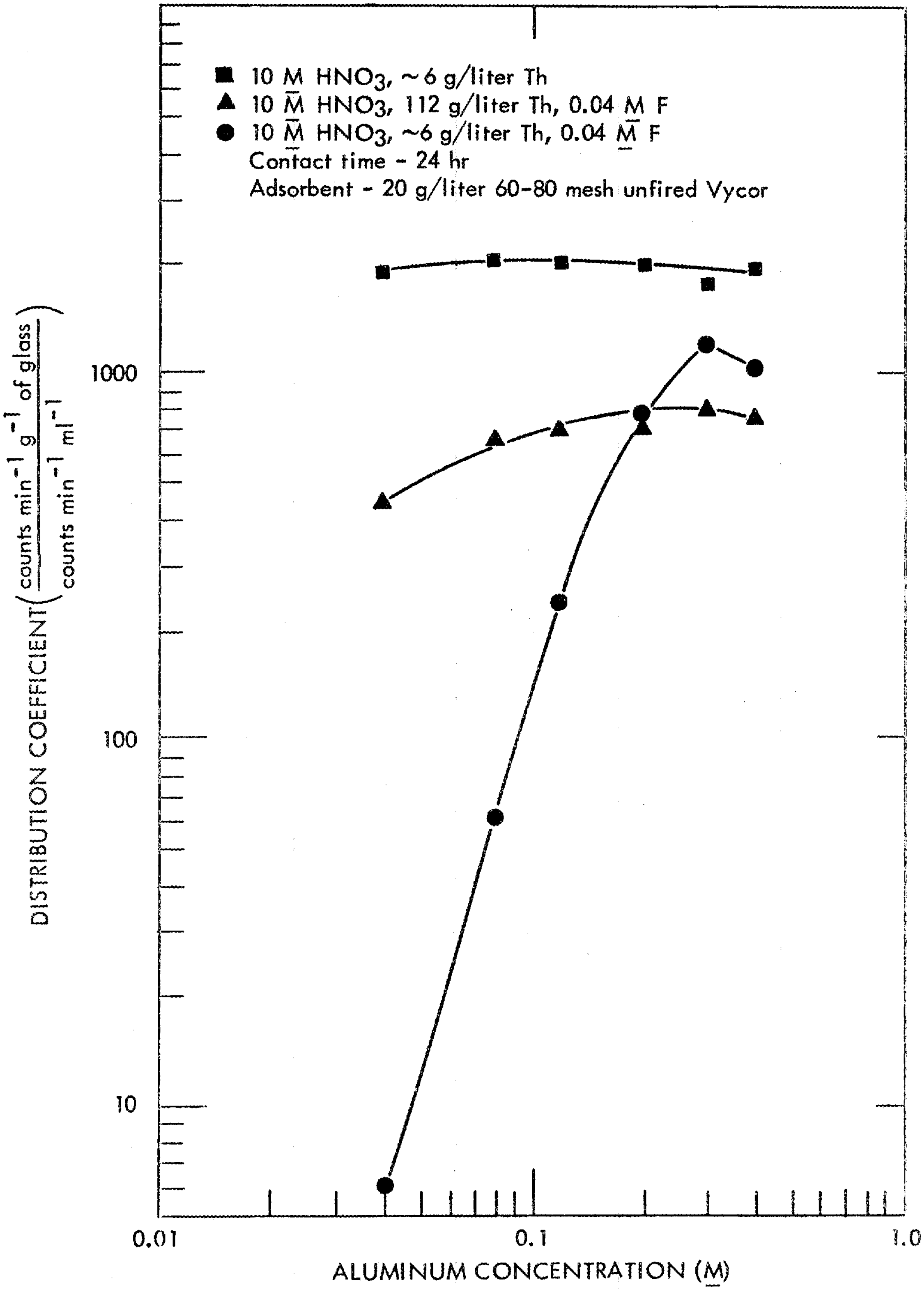

Fig. 9. Effect of Fluoride and Aluminum on Protactinium Adsorption.

ADSORPTION OF PROTACTINIUM BY SILICA GEL

The distribution coefficient of laboratory-prepared silica gel is about 3 times the coefficient obtained with unfired Vycor and more than 5 times the value obtained with commercial silica gel. There is little or no adsorption of protactinium by fired Vycor. These results were obtained by contacting 0.3 g of 20 to 40 mesh adsorbent for 1 hr with 30 ml of nitric acid solution containing about 5×10^5 c/m.ml Pa^{233} (Fig. 10).

Large differences were observed in the surface area, hydroxyl content, and sodium content of the adsorbents. The surface area of commercial silica gel was about 3 times that of the laboratory prepared silica gel or unfired Vycor. The hydroxyl and sodium content, however, were much lower in the commercial gel than in the laboratory prepared material (Table 2).

Column runs indicated that the capacity of the laboratory prepared gel is higher than that of the commercial material. On passing a solution containing 116 g/liter Th, 70 mg/liter Pa^{231}, 11 $\underline{M}$ HNO_3, 0.1 $\underline{M}$ Al, and a trace of F through a column 0.4 cm in diameter and 14 cm long containing 1 gm of 20 to 40 mesh silica gel, 95% of the protactinium was adsorbed. This was equal to a loading of about 3 mg/g (Fig. 11). Under similar conditions, 1 g of commercial silica gel removed only 57% of the protactinium. The amount adsorbed on the commercial silica gel ranged from 88% at the beginning of the run to 48% at the end. About 1.9 mg/g of silica gel was adsorbed (Fig. 12). In both experiments the flow rate was about 1 ml/min.cm^2.

Table 2. Pertinent Physical and Chemical Properties of Vycors and Silica Gels

Adsorbent	Surface Area (m^2/g)	OH^- (Meq/g)	Na (Wt %)
Fired Vycor glass	0.18	---	---
Unfired Vycor glass	144.2	---	---
Laboratory-prepared silica gel	196.0	0.12	0.55
Commercial silica gel	567.5	0.005	0.07

The fired Vycor glass was 100-200 mesh; the remaining adsorbents, 20-40 mesh.

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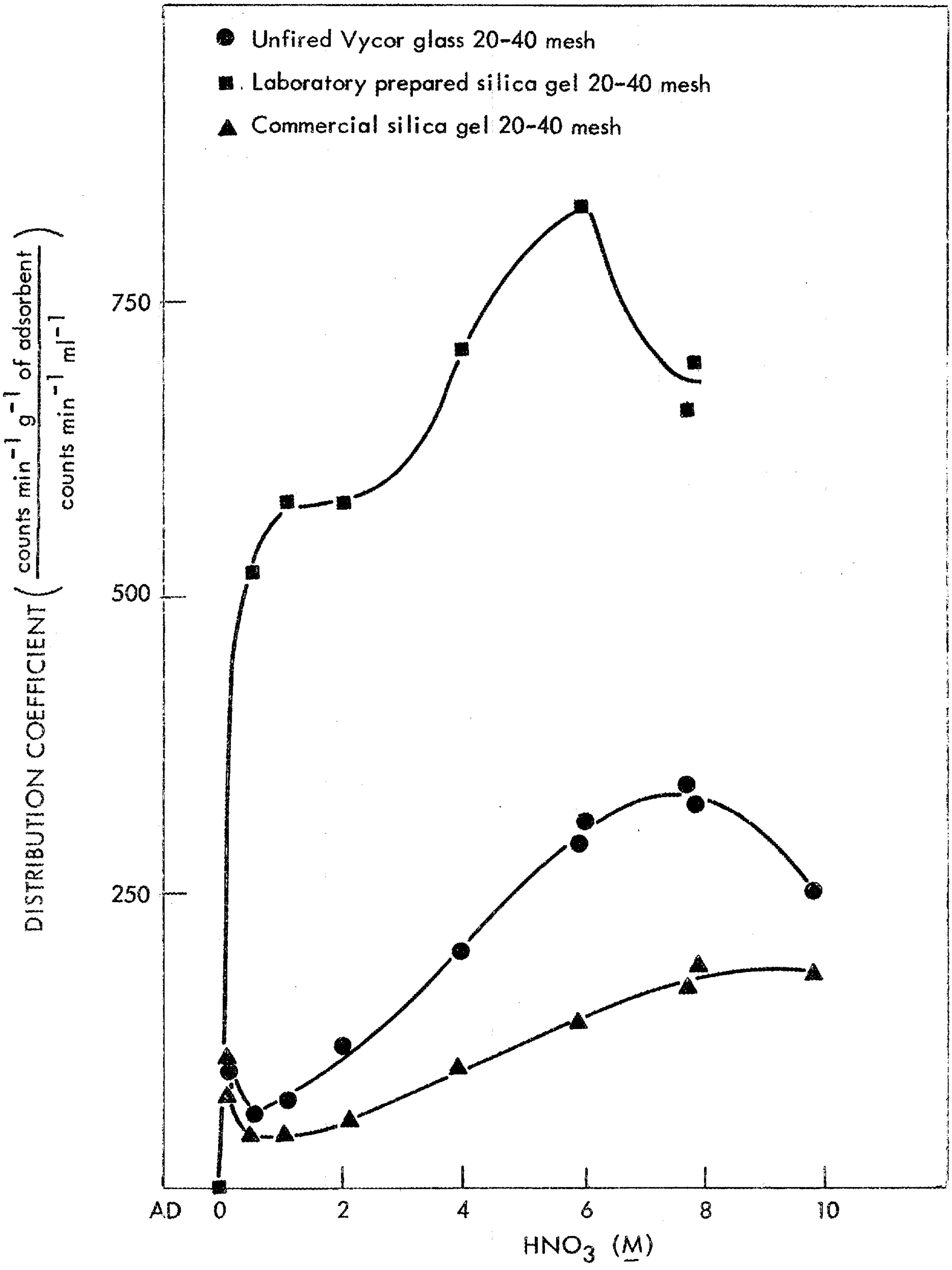

Fig. 10. Adsorption of Pa^{233} from Nitric Acid.

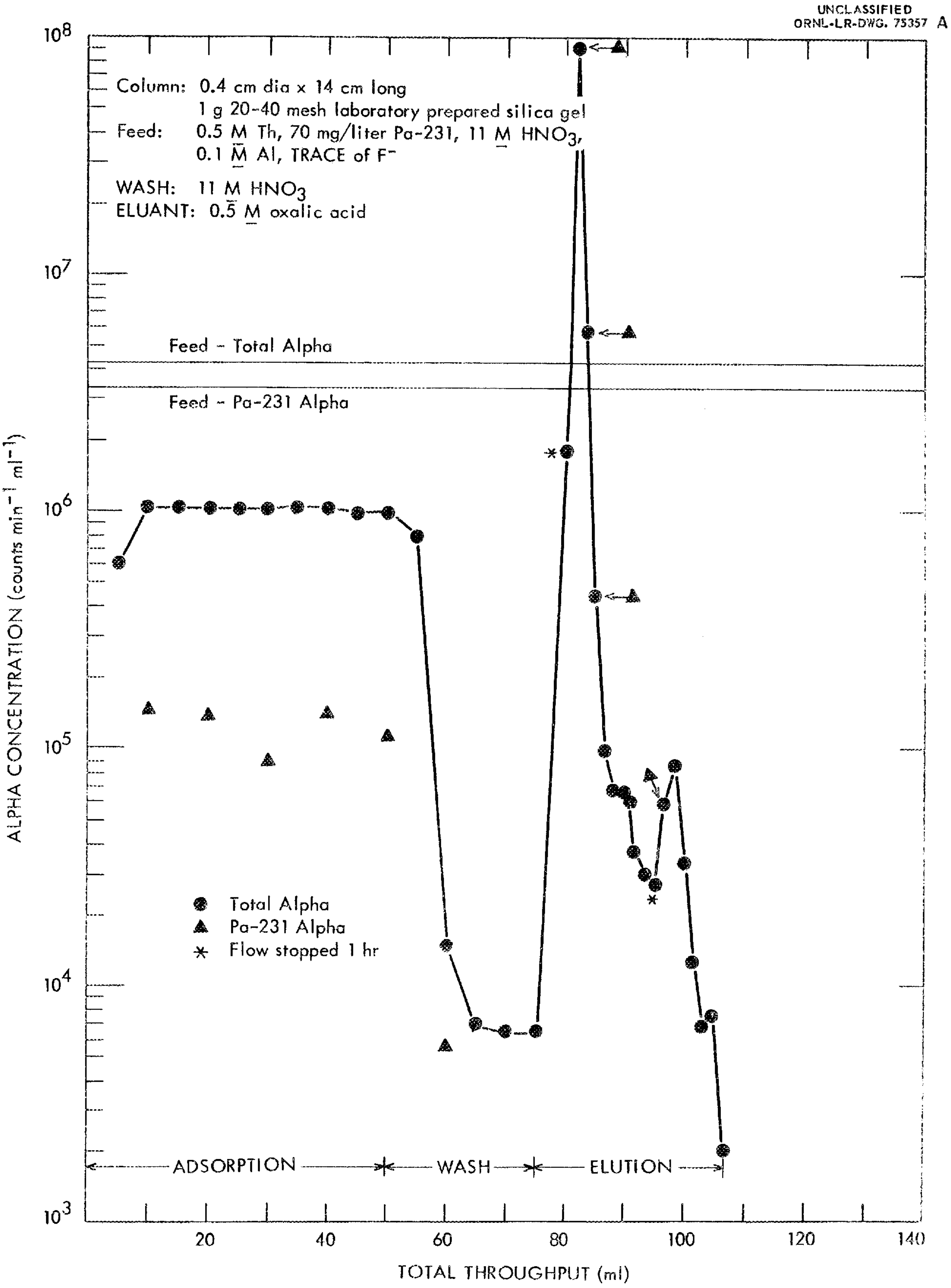

Fig. 11. Sorption of Protactinium with Laboratory-Prepared Silica Gel.

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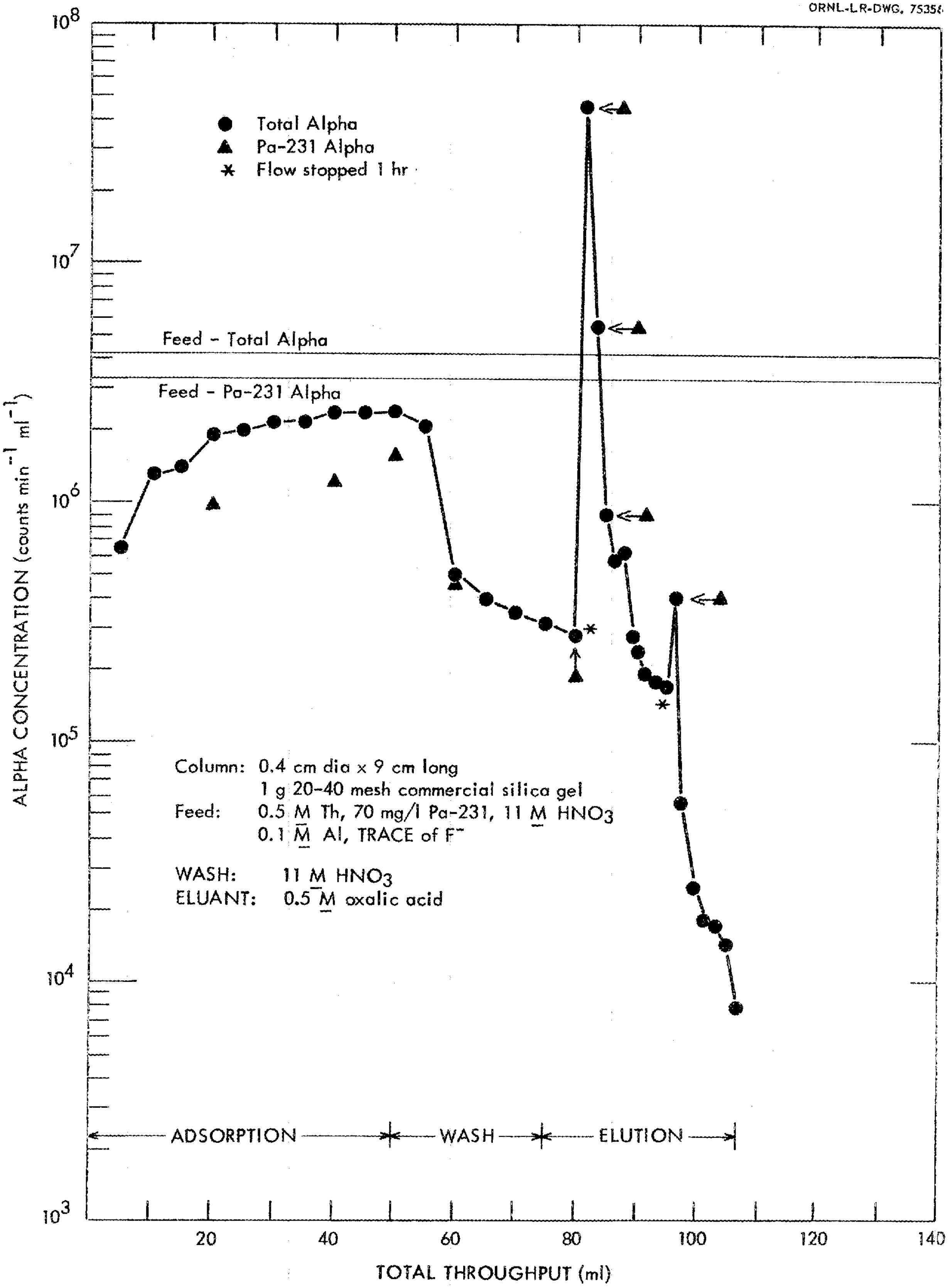

Fig. 12. Sorption of Protactinium with Commercial Silica Gel.

COEXTRACTION OF URANIUM, THORIUM, AND PROTACTINIUM WITH TBP

The extraction of protactinium nitrate with TBP has been extensively studied at Harwell,[4] ORNL,[3] and other laboratories. The data was used to design a coextraction flowsheet for high-burnup thorium breeder fuels. Since the recycled thorium from such fuels must be processed in shielded equipment because of the rapid growth of the U^{232} and Th^{228} daughters, it will only be necessary to separate the uranium, thorium, and protactinium from the high-cross-section fission products prior to recycling.

The coextraction process when tested at tracer levels of protactinium in laboratory batch countercurrent experiments resulted in the extraction of over 99% of the uranium and thorium and about 90% of the protactinium. The remaining 10% of the protactinium was essentially unextractable and similar to the adsorption studies, is likely due to the methods of preparing the feed solutions. Decontamination factors of 70 and about 10^5 were obtained for the long half-life and high-cross-section ruthenium and rare earth elements. About half of the zirconium-niobium extracted with the product, but this would be of small concern due to its low cross section and 65-day half-life. The principal disadvantage to this flowsheet is the large amount of aluminum in the high level aqueous waste (Fig. 13).

Of particular concern in the extraction of protactinium from thorium fuels is the complexing of the protactinium by fluoride. As previously shown, protactinium is strongly complexed by the fluoride to give a complex which is not extracted by TBP or adsorbed by Vycor.[4] As indicated by multiple extraction and Martin technique[6] experiments, in the absence of fluoride the protactinium is completely extractable from 5 $\underline{M}$ HNO_3 and has a distribution coefficient of about 4 (Fig. 14). The addition of a small amount of fluoride, however, not only decreases the amount of protactinium which is extractable but also greatly decreases the distribution coefficient of the extractable species. In the presence of 5 $\underline{M}$ HNO_3 containing 0.02 $\underline{M}$ HF, about 94% of the protactinium is unextractable, and the extractable species has a distribution coefficient of about 0.07 (Fig. 15). In these experiments the amount of protactinium which is unextractable is indicated by the intercept of the extraction line with the aqueous coordinate and the distribution coefficient of the extractable species by the slope of the line.

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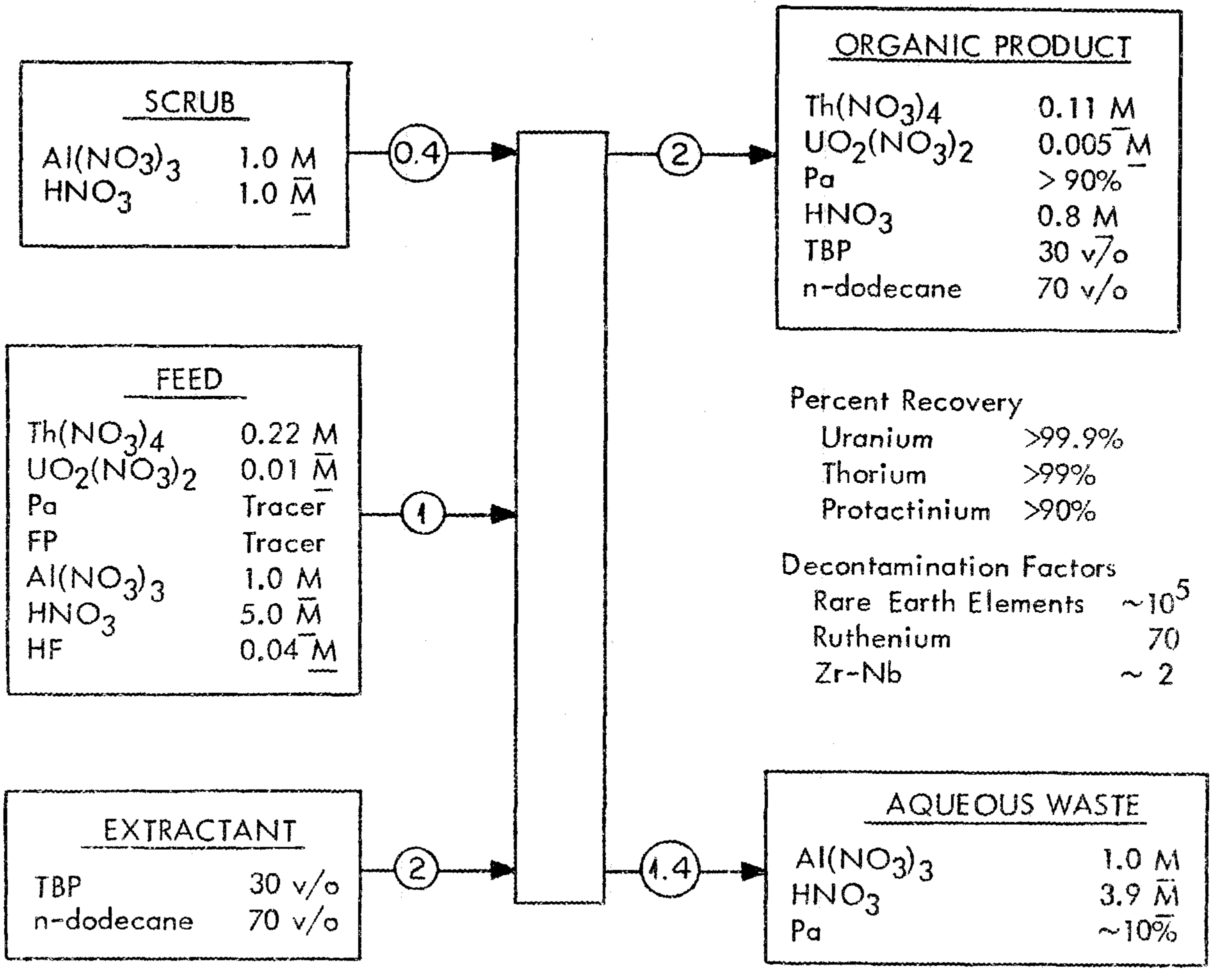

Fig. 13. Co-extraction of Uranium, Thorium, and Protactinium with Tributyl Phosphate.

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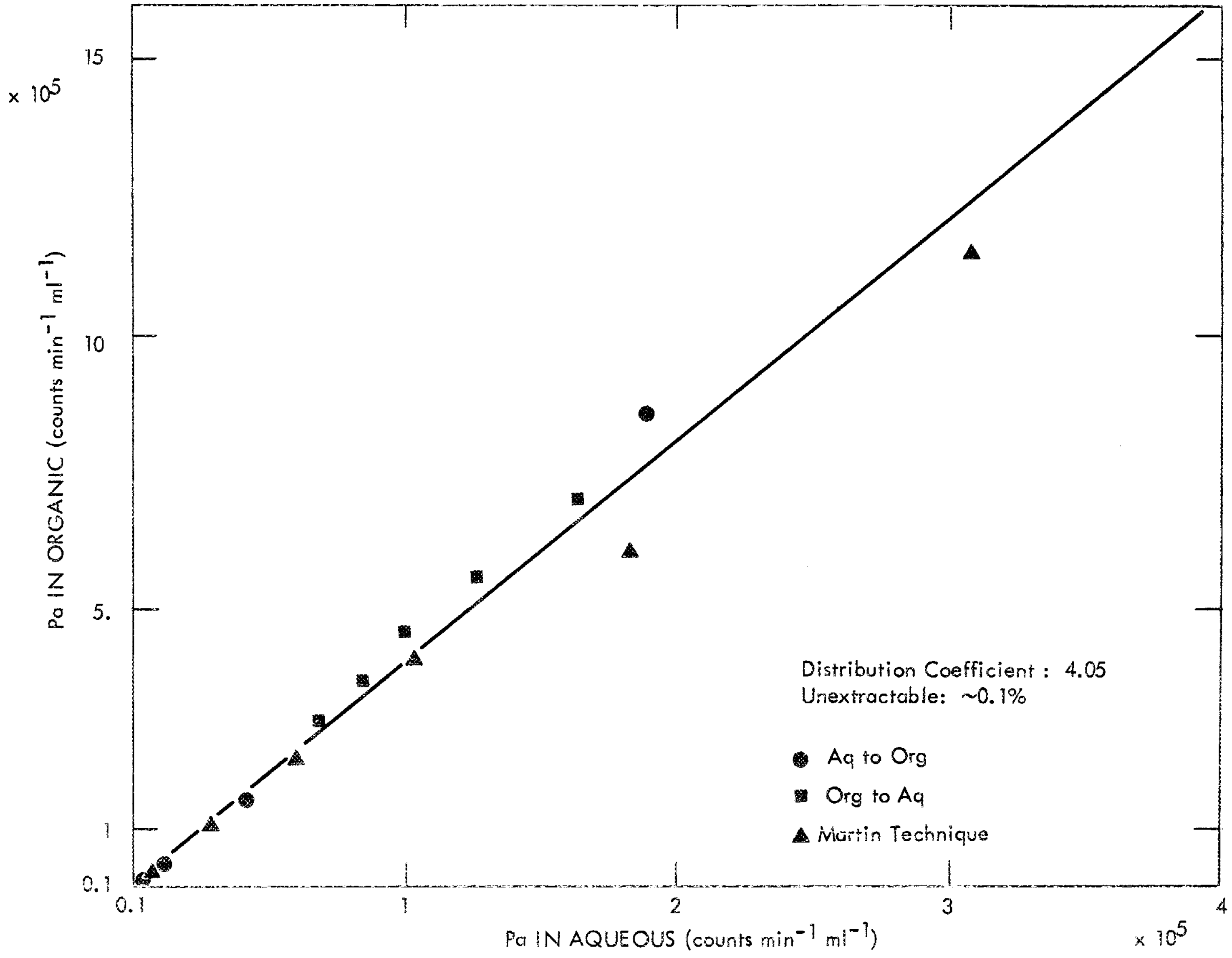

Fig. 14. Extraction of Protactinium from 5 M HNO_3 with 30 v/o Tributyl Phosphate in n-dodecane.

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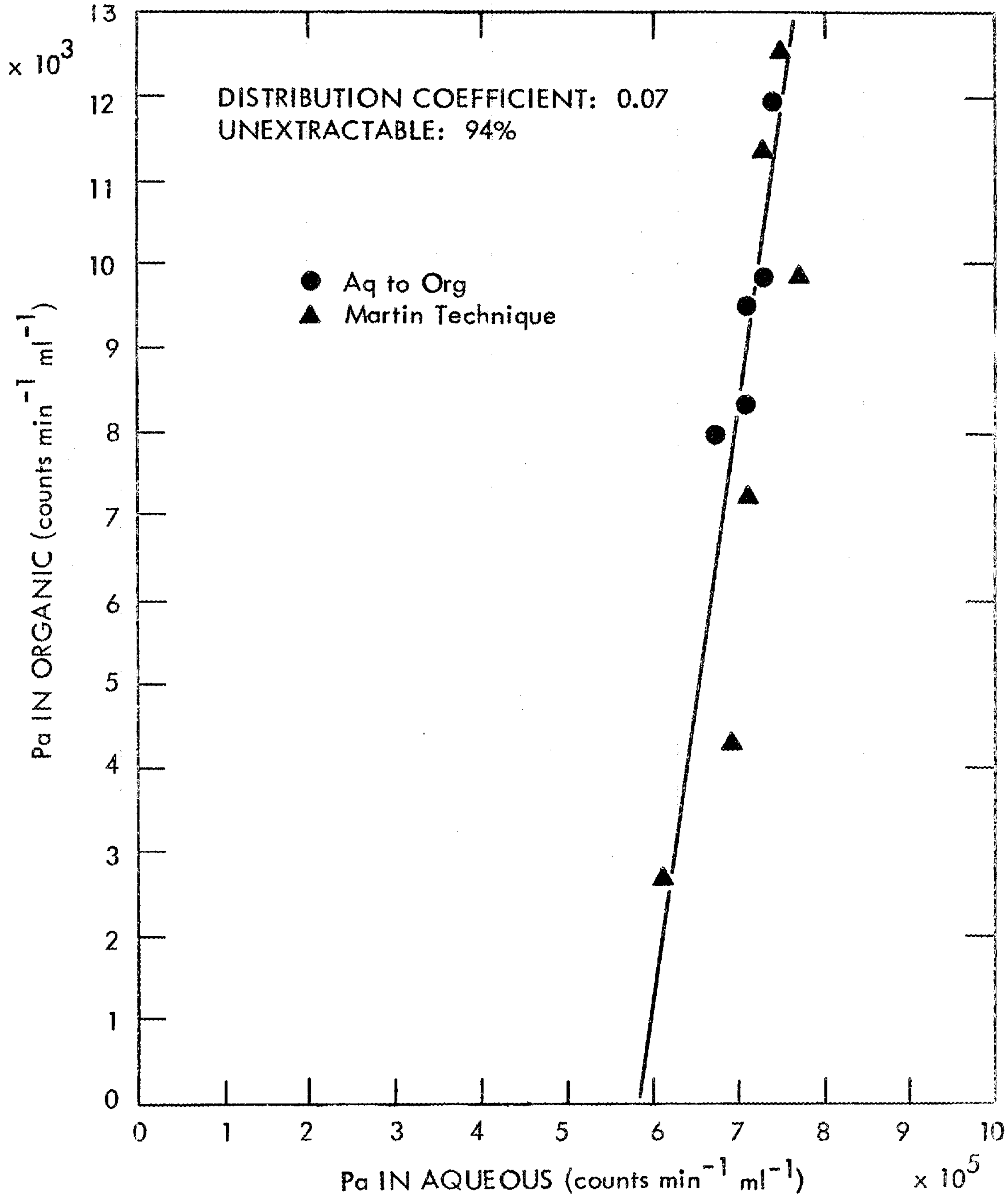

Fig. 15. Extraction of Protactinium from 5 N HNO_3 Containing 0.02 N HF with 30% TBP in n-dodecane.

CONCLUSIONS

Although there are many unanswered questions concerning the adsorption of protactinium on unfired Vycor, the results of this investigation are encouraging. It was shown that unfired Vycor will preferentially adsorb protactinium from nitrate systems and has a usable capacity of at least 5 mg Pa/g glass. The protactinium may be easily eluted from the glass with oxalic acid solutions, yielding a product decontaminated from all contaminants except zirconium-niobium by factors greater than 10^3.

Since all experiments were made with simulated feed solutions, the amount of adsorbable protactinium that would be in solution under actual fuel processing conditions is unknown. Future plans call for small column experiments with high-burnup short-cooled thorium solutions, which should define better the process potentialities as well as limitations. In addition, laboratory work will continue on column conditions necessary for maximum effect, and the adsorption properties of unfired Vycor and silica gel for protactinium from other acidic solutions will be investigated.

The countercurrent batch extraction experiments with TBP show that in the presence of thorium and a large excess of aluminum, protactinium can be satisfactorily extracted. Laboratory experiments are continuing to determine the minimum amount of aluminum that will be required for the process.

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93. J. A. Swartout
94. J. W. Ullmann
95. M. E. Whatley
96. R. G. Wymer
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103. E. L. Anderson, Washington, AEC
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