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STUDY OF A HALF-INCH PULSE COLUMN WITH A TBP SYSTEM

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1.0 Abstract

A glass pulse column 0.5 in. diam. by 36 in. high was studied with uranium solutions and tributyl phosphate - hydrocarbon mixed solvent. Variables were: (1) plate spacing, (2) plated design, (3) pulse amplitude, (4) solvent/aqueous flow ratio, (5) nitric acid concentration.

[redacted]

2.0 Introduction

A device with movable perforated plates in a vertical column to agitate intimately two immiscible liquids has been described by Van Dijck [1]. The same patent proposes stationary plates with moving or pulsed liquid phase, which has been described as the "pulse column" [2]. The pulse column is a vertical column with stationary perforated plates with the agitation of the two phases obtained by a cyclic pulse applied to the stream entering the bottom of the column. The organic phase feed is usually pulsed because this is the "cool" or non-radioactive stream. The pulse forces the liquid (non-radioactive phase) through the small perforated plates, the light phase rising in tiny droplets and the aqueous phase falling to the plate below on the "pull" of the "push" "pull" pulse. The two phases move countercurrently by gravity through the column, depending on the viscosity and difference in density of the two phases being used in the column.

The principal advantage offered by the pulse column as compared with conventional packed columns is the marked reduction of column height obtainable. In most uranium extraction systems, the pulse column height required to equal the effectiveness of a packed column was lower by 1/3 to 1/2. Such height reductions are of considerable significance in radiochemical operations because of the consequent large reduction in shielding requirements, i.e., such as cell structures.

3.0 Summary

Pulse column studies in a 0.5 in. diameter unit show:

1. That equilibrium conditions are obtainable in about 3.5 to 4.0 hours or 8-10 volume changes using a UNH feed solution 2.4 N in nitric acid and 40 g/l U using an organic mixture 15% TBP - 85% Varsol, with a total through put rate of 9.4 cc/min, a pulse amplitude of 1/16 in., and a pulse frequency of 60 cycles/min. in the simple extraction column.
2. That uranium loss was less with 1" plate spacings than with 2" plate spacings. Furthermore throughput rate variations apparently did not noticeably affect uranium loss when using one inch plate spacings. For the two inch spacings, higher rates produced lower loss values, due to the agitation caused by the turbulence of flow.
3. That varying the pulse amplitude from 1/16 inches to 3/32 inches increased the uranium loss by a factor of three. Beyond this point an increase in amplitude showed a decrease in the uranium loss.
4. The uranium loss increases with an increase of the hole diameter in the perforated plates.
5. An increase in the pulse frequency gave a decrease in uranium losses (from 30 RPM to 60 RPM).

[redacted] [redacted]

4.0 Objectives

The determination of uranium loss from a one-half inch laboratory pulse column as pulse amplitude and plate spacing varied was the primary objective of these experiments. The time necessary for the column to attain dynamic equilibrium was also investigated. Other variables which are important, particularly for pulse columns to be used in plant operations, include:

1. The ratio of continuous to discontinuous phase.
2. Column length and its effect on HTU.
3. Column diameter with respect to: channeling, coalesence, and flows.
4. Plate design, and spacing related to HTU.
5. Possible variation in plate spacing at different positions in the column.
6. Pulse frequency as related to the plate spacing.
7. Pulse duration or retention time in the column.
8. Column capacity or flooding data.
9. Effects of the physical properties of the fluids on previously mentioned variables.

5.0 Apparatus

1. Column - The column was made from a 36 inch section of 1/2 inch I. D. pyrex tubing with 6 inches of 1 inch I. D. flanged pyrex pipe attached to each end. The end fittings are typical column fittings to permit flow to and from the column. The feed inlet extends the entire length of the one inch section to a point near the first plate (Figure 7). The bottom fixture contains (a) the raffinate outlet whereby the raffinate leaves the column through a jackleg and (b) the organic inlet which extends the length of the one inch section at the bottom of the column and ends near the bottom plate.

   A plate assembly rod 1/16 inch in diameter and 42 inches long is attached to the center of the top plate. Stainless steel plate spacers 3/32 inches O. D. were cut to the desired lengths for plate supports and spacing. The extraction section was 36" in length. (Figure 8).

2. Plates - The plates for the column consisted of 18 and 22 gauge stainless steel sheets (Figures 9-10) 0.50 inches in diameter. The 22 gauge plates had 32 holes of 0.039 inch diameter drilled on an equilateral triangular pattern whereas the 18 gauge plate had 56 holes on the same pattern. These plates suspended on the 1/16 inch stainless steel rod by means of the spacers (Figure 11). The open area for the 22 gauge plates is 20-21% of the plate area.

[redacted]

3. Pump - A bellows pump (Figure 12) attached directly to the organic feed inlet line was used to disperse the phases through the plates of the column. The pump used a 1/8 HP motor and pulsed at the rate of 60 pulses per minute. The "push-pull" effect from the pump upon the two phases in the column is controlled by raising or lowering the bar below the bellows. This bar is contacted by the cam attached to the motor and furnishes the pulse for forcing the organic phase through the plates and drawing the aqueous phase downward. The "pull" is obtained by removing the check valve near the pump and allowing part of the column solutions to enter the section of organic feed line between the column and pump.

4. Miscellaneous - The aqueous feed and organic tanks are pressurized with dried filtered air to the desired pressure of approximately 5 pounds. No pump is needed for the aqueous feed since the 5 pounds of air pressure is sufficient to push the feed into the column and the organic pump is used only to obtain the pulse in the column.

   The disengaging interface between phases is maintained at the top of the column and is principally controlled by varying the volume of the dispersed liquid by means of a mechanical "jack leg".

5. Screw Turns on Pump vs. Pulse Amplitude - One run was made to check the pulse amplitude in the 1/2" Pyrex glass column as the cam regulating screw was turned. The column run was started with as small a pulse as possible and the amplitude in the column checked. The screw was

Screw Turns on Pump vs. Pulse Amplitude (continued)

turned one complete turn and the amplitude checked for each complete turn. One complete turn gives a pulse amplitude in the column of 1/16 inch and sixteen turns gave an amplitude of 14/16 inch the largest used in the investigation. Figure V, the number of turns you make on the screw, will tell the amplitude you can expect to obtain in the column.

6.0 Experimental Results

6.1 Preliminary Considerations

In some preliminary investigations, it was noted that the number of holes affects the plate efficiency, but no experiments were carried out to ascertain the magnitude of this effect. A large number of holes with very close spacing might result in increased coalescence and lower extraction rates.

The thickness of the plate to be used in the column is of some slight significance and in general it would be desirable to keep the thickness to the minimum mechanically feasible. A thick plate will require a greater "push-pull" on the column phases and probably increase the necessary overall column pressure drop in approximately direct proportion to the thickness.

The use of a pulse rate in the vicinity of 50 pulses per minute was found by Hanford[2] to be the most efficient for hexone extraction of UNH. The work reported herein was done at a pulse rate of 60 per minute which was found to improve operation when compared to a rate of 30 pulses per minute using TBP-Varsol mixture as the [redacted]

Preliminary Considerations (continued)

The organic to aqueous ratio is somewhat of an economic consideration and its investigation was not of immediate importance in this work.

6.2 Equilibrium Time in the 1/2" Pulse Column

A. Simple Extraction Column

To determine the time necessary for the 1/2" simple extraction pulse column to reach a constant uranium loss, a run of 8.5 hours duration was made using a 15% TBP - 85% Varsol mixture as the organic feed with a 2.5/1.0 O/A ratio and the aqueous feed solution of 2.40 N in nitric acid and containing 35.50 g/l of uranium. The column was sampled at half hour intervals and the results indicate the uranium loss to be essentially constant at the end of three hours of operation and definitely constant after operating for a period of five hours - (Fig. 1) which gives 2.5 to 3.0 aqueous throughputs.

B. Compound (Extraction and Scrub) Column

The 1/2" simple pulse column was converted to a compound column having a 36 inch extraction and a 24 inch scrub section to investigate the time required to obtain equilibrium in the column using Purex conditions. Equilibrium was obtained in 3.5 to 4.0 hours using a feed 314 g/l uranium and 2.0 normal nitric acid, organic 30% TBP - 70% Varsol mixture and a scrub of 3.0 normal nitric acid. The flow rates being 10-3-2 organic - feed - scrub, respectively.

Compound (Extraction and Scrub) Column (continued)

The uranium loss does not gradually rise to the equilibrium condition as was predicted, but increased rapidly for two hours and then declined to reach equilibrium in four hours of column operation (Figure II). This "hump" is probably due to the fact that about this time (2.0 hours after start up), the organic is extracting large amounts of nitric acid from the aqueous feed allowing the uranium to come farther down the column before being extracted.

6.3 Plate Spacing

Plate spacings of one and two inches were investigated at total flow rates of 9.4 ml/min. and 18.2 ml/min. using a feed containing 80.0 g/l of uranium at a nitric acid concentration of 4.85 N. The organic phase was 15% TBP - 85% Varsol mixture.

With plate spacings of one inch, the uranium loss was virtually constant irrespective of the throughput, varying from an average of 0.14 percent at the low flow rate to 0.16 percent at the high flow rate. When the plate spacings were increased to two inches, it was found that with a flow rate of 9.4 ml/min. the uranium loss was approximately 2.25 percent although a constant loss was never attained and the loss was increasing at the termination of the run. At the two inch spacings with a throughput of 18.2 ml/min. the uranium loss was 1.50 percent. This decrease in loss is probably partly due to the added turbulence caused by the increased throughput. Other plates and spacers have been [redacted] closer plate spacing, i.e., one-half inch, will be investigated.

6.4 Pulse Amplitude vs. Uranium Loss at Constant Throughput

Using the simple extraction column 1/2" I.D. and starting with the lowest pulse amplitude without flooding the column, 4/64" the uranium loss was 0.04%. As the amplitude was increased to 7/64", the uranium loss increased to 0.10%. Increasing the pulse amplitude beyond this value shows decreasing uranium losses with each increase of pulse amplitude. The total throughput for the 1/2" column being slightly more than 100 gal/hr/sq. ft.

Data were obtained for pulse amplitudes up to one-eight of an inch in the column and are recorded in Table I. The work at Hanford had shown a decreasing uranium loss with increasing amplitude, however, we find a situation wherein the losses increase with increasing amplitude and probably go through a maximum somewhere in the vicinity of a pulse amplitude of one-eighth inch. Figure III. A possible explanation of this situation may be obtained by considering mechanism of the pulsing action. With a pulse amplitude of about 1/16 in., one finds the droplets formed by the pulse to be of small size and consequently large area to volume ratios which thereby permit an increased rate of mass transfer. As the amplitude increases to 1/8 in. the size of the droplets increases and the efficiency of the extraction decreases. Above 1/8 in. amplitude, it is possible that the greater amplitude creates added turbulence which tends to offset the larger apparent droplet size and therfore increase the efficiency of extraction.

[redacted]

[redacted]

Pulse Amplitude vs. Uranium Loss at Constant Throughput (continued)

It is well to note that when the column is put into operation, the organic phase is generally dispersed as very small droplets, but as the run proceeds the small droplets are found only in the lower section of the column. This might be attributed to the nitric acid or uranium coming further down the column as the dynamic equilibrium is approached. Further work will be performed to ascertain the cause of this condition.

6.5 The Effect of Nitric Acid Concentration upon Uranium Extraction

To investigate the effect of nitric acid upon the extraction of uranium from a UNH feed having 35.5 g/l of U, 6 runs were made with the nitric acid content varying from 0.3 N to 2.4 N as shown. (Fig. IV). As the HNO_3 content in the feed increased from 0.30 N to 1.9 N, the uranium loss decreased from 34% to 0.03%. Further increase of nitric acid showed very small effect in uranium loss.

7.0 Bibliography

1. U. S. Patent 2,011,186 (1935) Van Dijck

2. HW-14728, The Design and Operation of the Pulse Column, W. A. Burns, C. Groot, and C. M. Slansky, October 12, 1949.

8.0 Acknowledgements

The author acknowledges his appreciation to L. T. Corbin and associates of the Analytical Chemistry Division for the necessary analysis in conjunction with this work.

The author acknowledges the guidance in this work of J. I. Stevens, Research Participant, during the summer of 1950 at ORNL and Assistant Professor of Chemical Engineering at Vanderbilt University.

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Table I

Further Effect of Varying the Pulse Amplitude upon Uranium Losses Starting With the Smallest Pulse Not to Flood the Column - to Full Pulse

Feed
- Source UNH
- U - 35.5 mg/ml
- HNO_3 - 2.1 N

Column
- 36" Pyrex Pipe 1/2" I.D.

Pulse
- Frequency - 60 RPM
- Amplitude - as shown

Organic
- 15% TBP
- 85% Varsol

Plates
- 22 Gauge
- 32 Holes per plate
- 0.039" diameter

O/A Ratio
- 2.5/1

Total Throughput
- 16.5 ml/min.

Run No.	Pulse (in) Amplitude	U - Loss / Sample (%)								U in Org. (mg/ml) Average
		*3:20	*3:40	*4:00	*4:20	*5:00	*5:20	*5:40	*6:00	
2	4/64	0.40	0.08	0.04	0.05	0.05	0.04	0.04	0.04	15.0
3	5/64	0.29	0.13	0.05	0.05	0.05	0.05	0.06	0.05	14.4
4	6/64	0.16	0.23	0.20	0.14	0.12	0.09	0.09	0.09	15.3
5	7/64	0.16	0.12	0.17	0.07	0.11	0.10	0.16	0.10	13.9
6	8/64	0.08	0.08	0.07	0.07	0.07	0.07	0.07	0.07	12.5
7	10/64	0.10	0.05	0.06	0.05	0.06	0.06	0.06	0.06	13.5

* Time of column operation before sample was taken (hrs.)

[redacted]

Figure I

The Effect of Varying the Pulse Amplitude vs. Uranium Loss

Feed:
Source - UNH
U - 35.50 g/l
HNO_3 - 2.40
Organic:
15% TBP.
85% Varsol

Column:
Simple Extraction
36" Pyrex pipe 1/2" I.D.
Plates:
22 gauge s.s.
32 Holes 0.039" diameter

Pulse Amplitude:
P-2 = 0.063"
P-3 = 0.081"
P-4 = 0.100"
P-5 = 0.120"
P-6 = 0.137"
P-7 = 0.156"

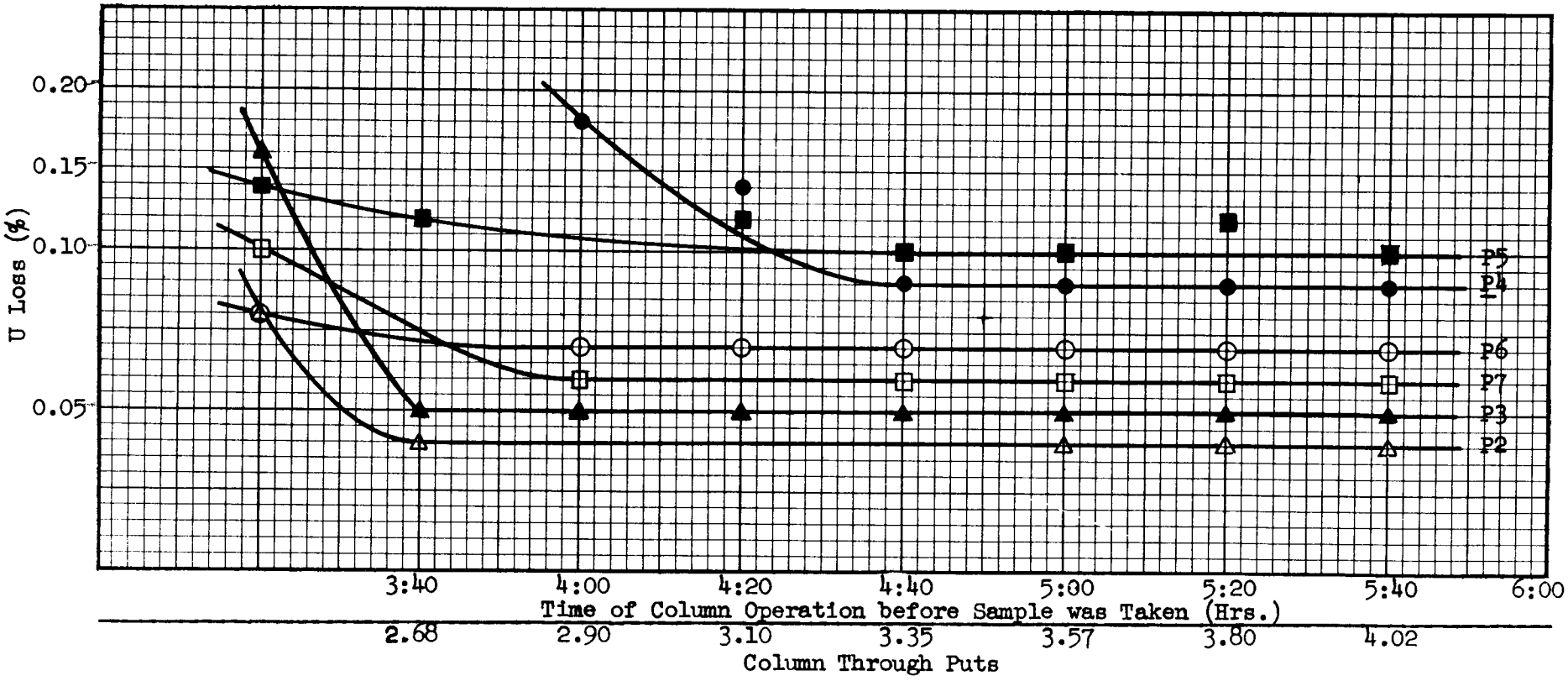

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Figure II

Time Required to Obtain Equilibrium in the 1/2" Compound Column Using a 36 Inch Extraction and a 24 Inch Scrub Section

Feed:
U - 314.0 g/l
HNO_3 - 2.0 N

Organic:
30% TBP
70% Amsco

Scrub:
3.0 N HNO_3

Flows: (Ratio)
Organic - 10
Feed - 3
Scrub - 2

Column:
Extraction - 36"
Scrub - 24"

Plates:
1" apart

Holes:
0.039" Diameter

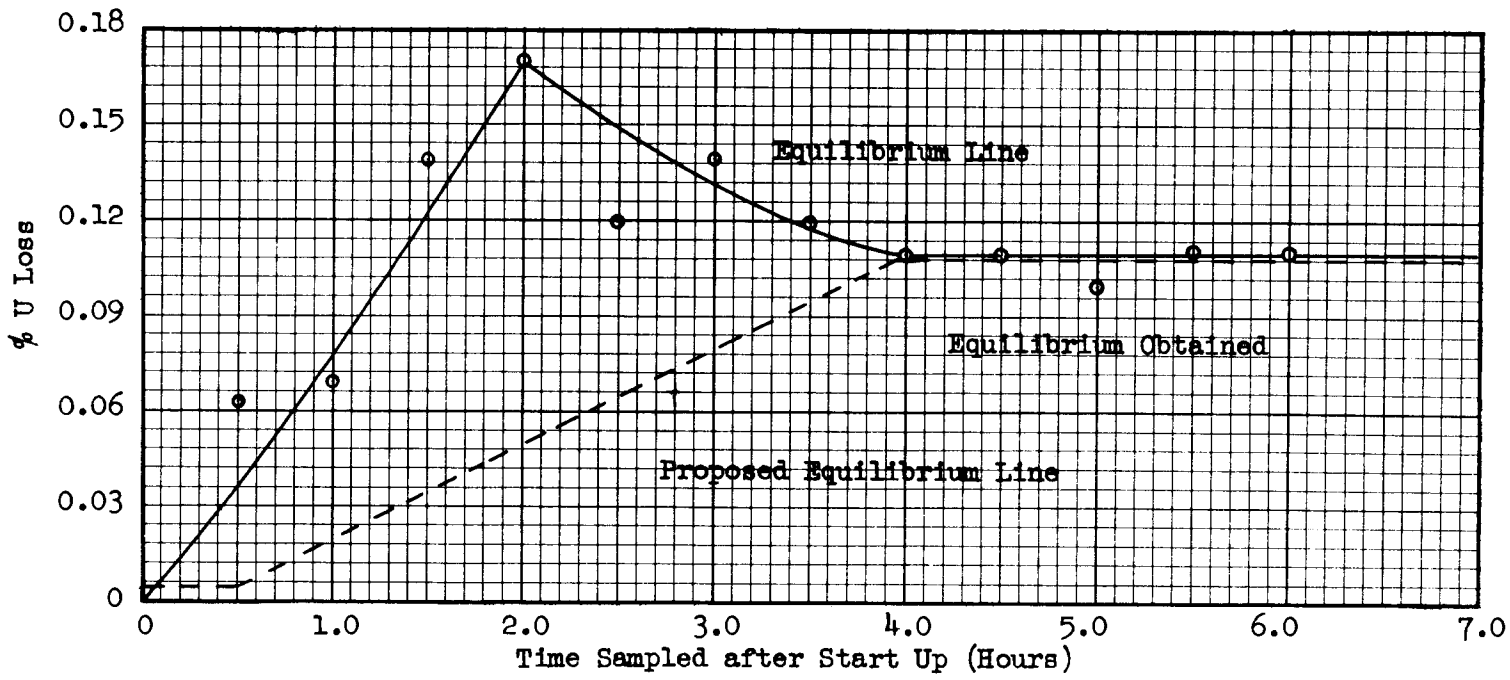

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FIGURE III

The Effect of Varying the Pulse Amplitude vs. Uranium Loss

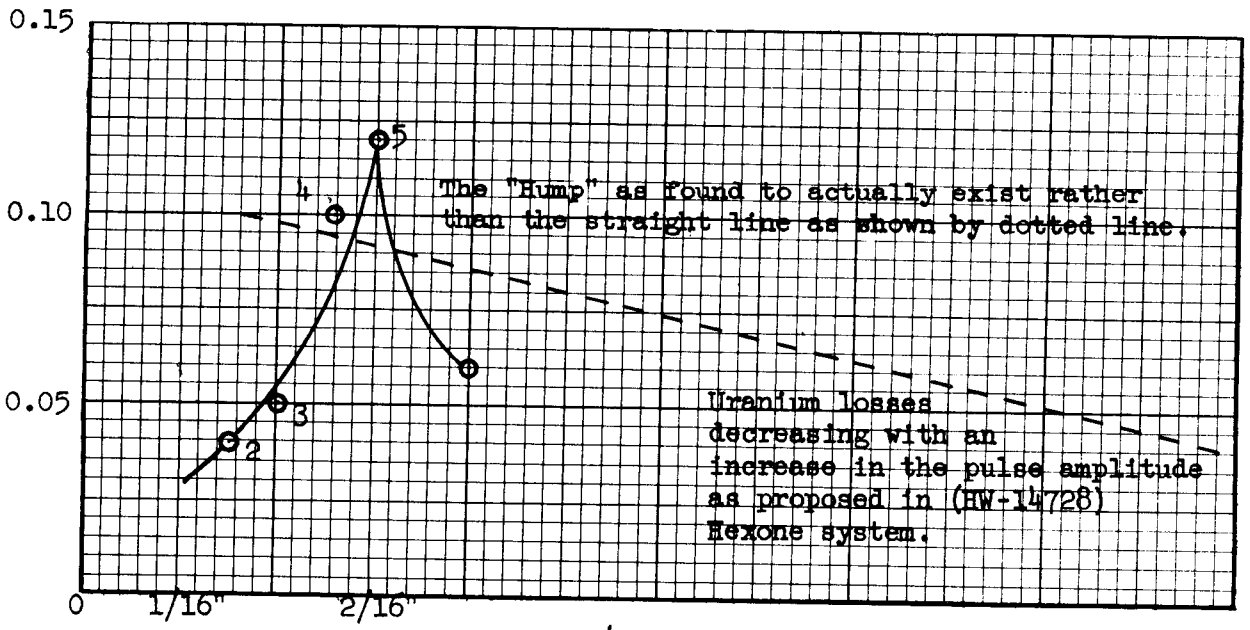
Feed: UNH
U - 35.5 g/l
HNO_3 - 2.1 N

Org.
15% TBP
85% Varsol

Column:
36" Long 1/2" I.D.

Plates: 22 gauge s.s.
32 holes 0.039" diam.

O/A - Ratio 2.5/1.0



Amplitude Increase/Turns on Pump Screw (Fig. II)

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Figure IV

The Effect of Varying The HNO_3 in The Aqueous Feed Upon

Uranium Extraction (%Loss) Keeping All Other Variables Constant

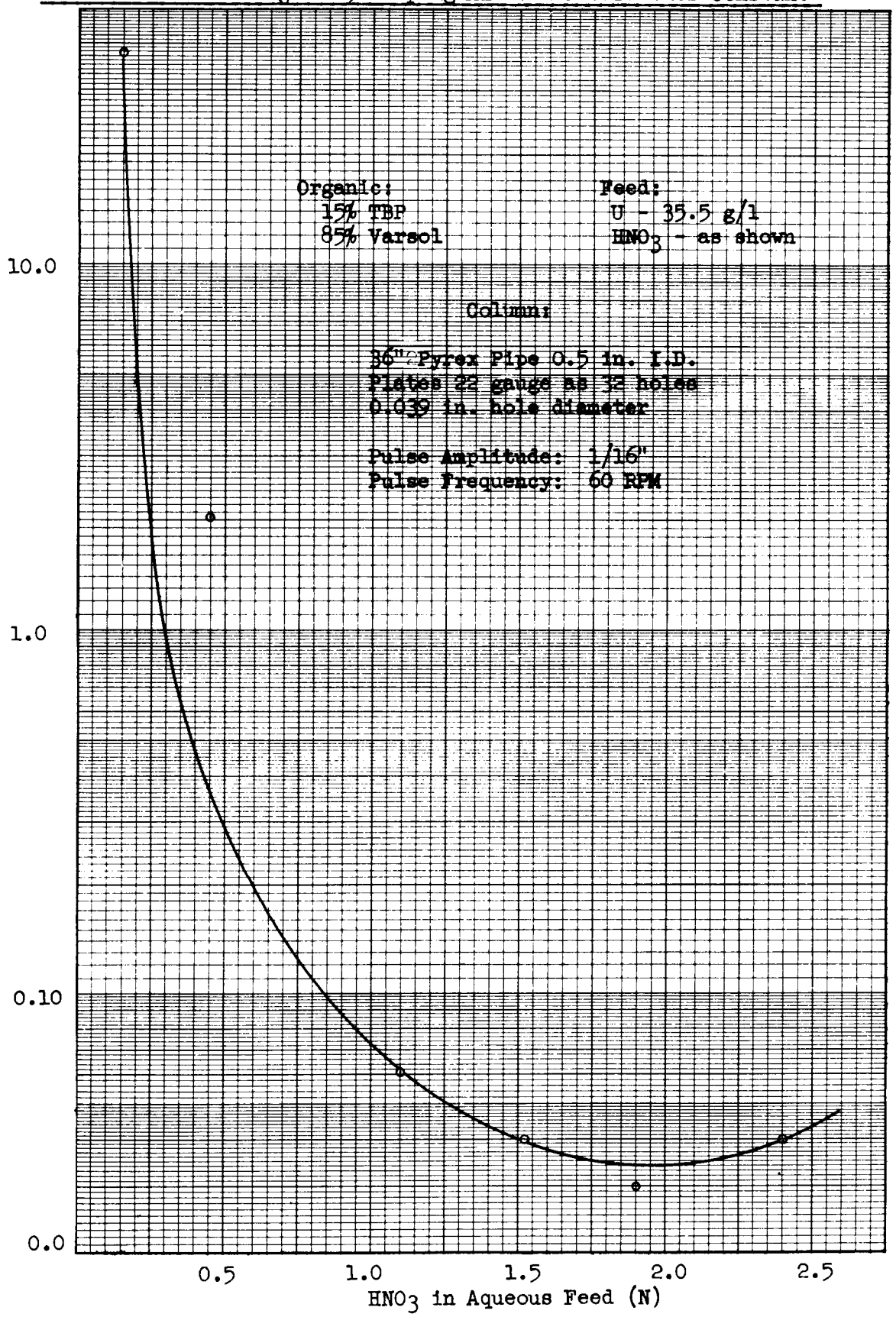

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Figure V

The Number of Screw Turns on the Pulse Pump vs.

Pulse Amplitude

Feed:
U - 35.5 g/l
HNO_3 - 2.8 N

Org:
TBP - 15%
Varsol - 85%

Column:
36" Long, 1/2" I.D.
Plates: 22 gauge ss.
32 Holes: 0.039 in diameter spaced 1" apart

Time of Run before measuring - 5.0 hours
Air pressure on feed tanks = 5#

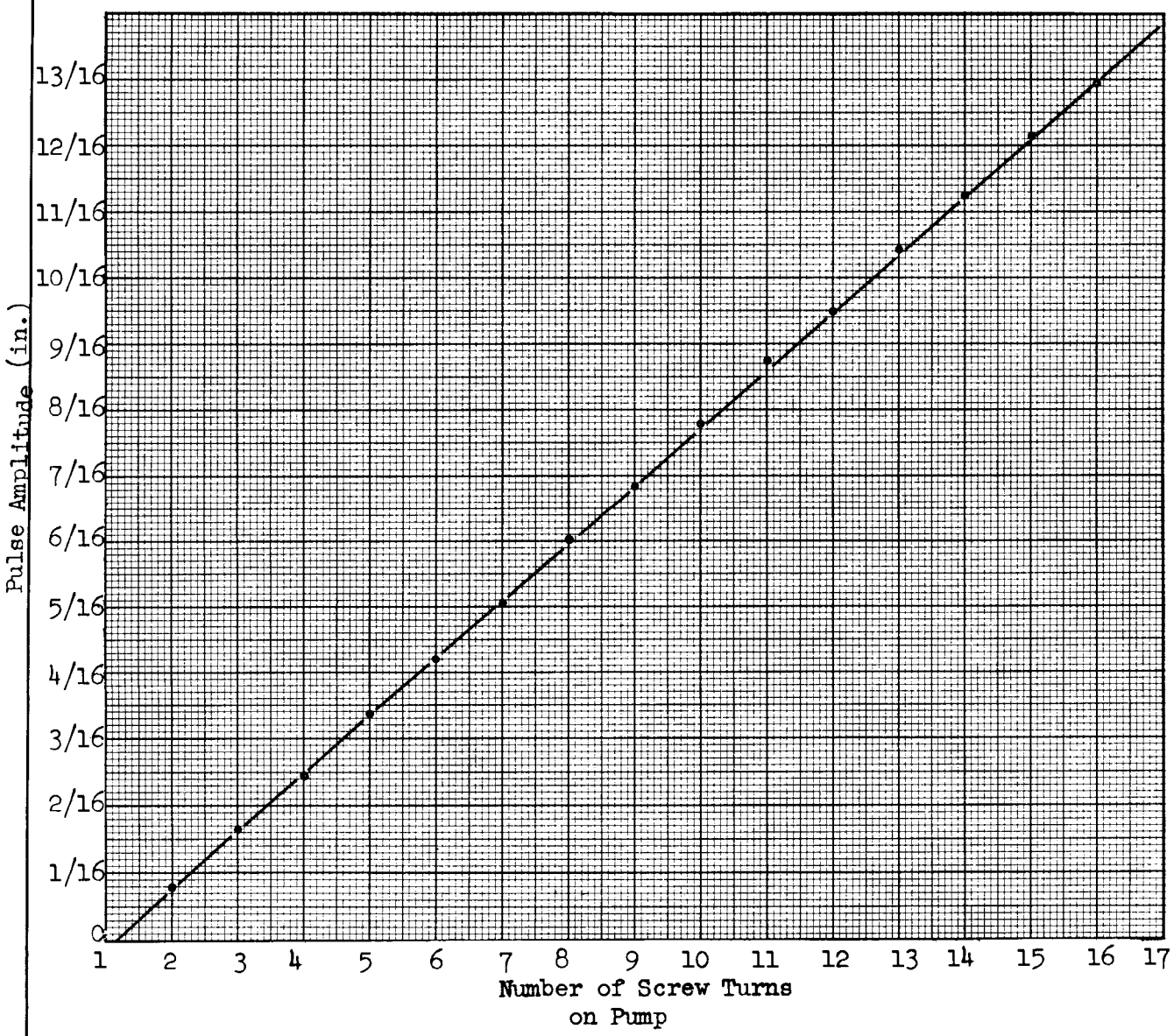

Figure 7

Top section of the 1/2" Pulse Column showing feed inlet at left and plate support rod attached to top of column in center. Feed inlet has the end plugged and hole in side of the tube.

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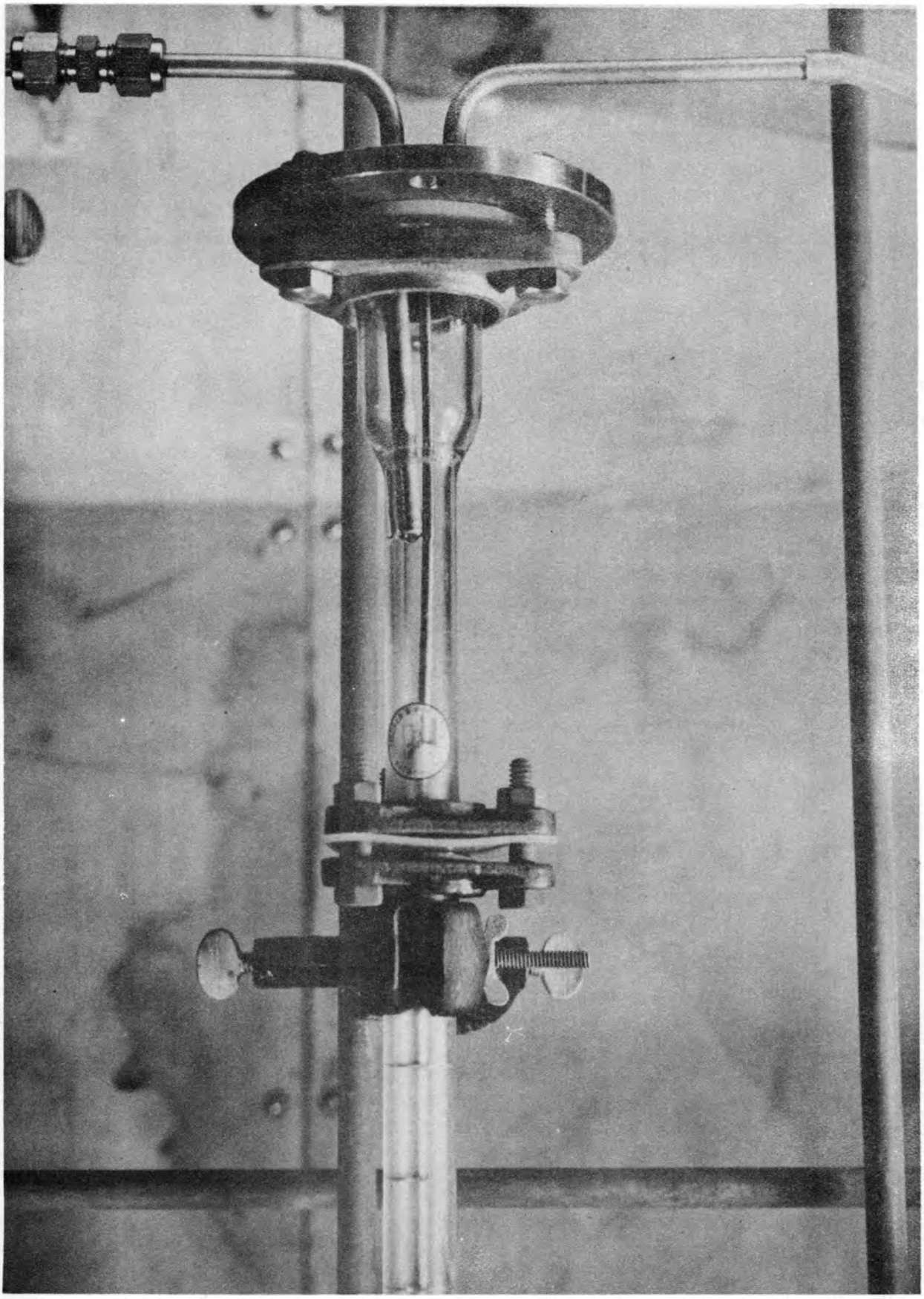

FIG. 7

Figure 8

The entire 1/2" pulse column unit, showing column, rotameters, pump and tanks. Both feed and organic tanks were connected to a filtered air line.

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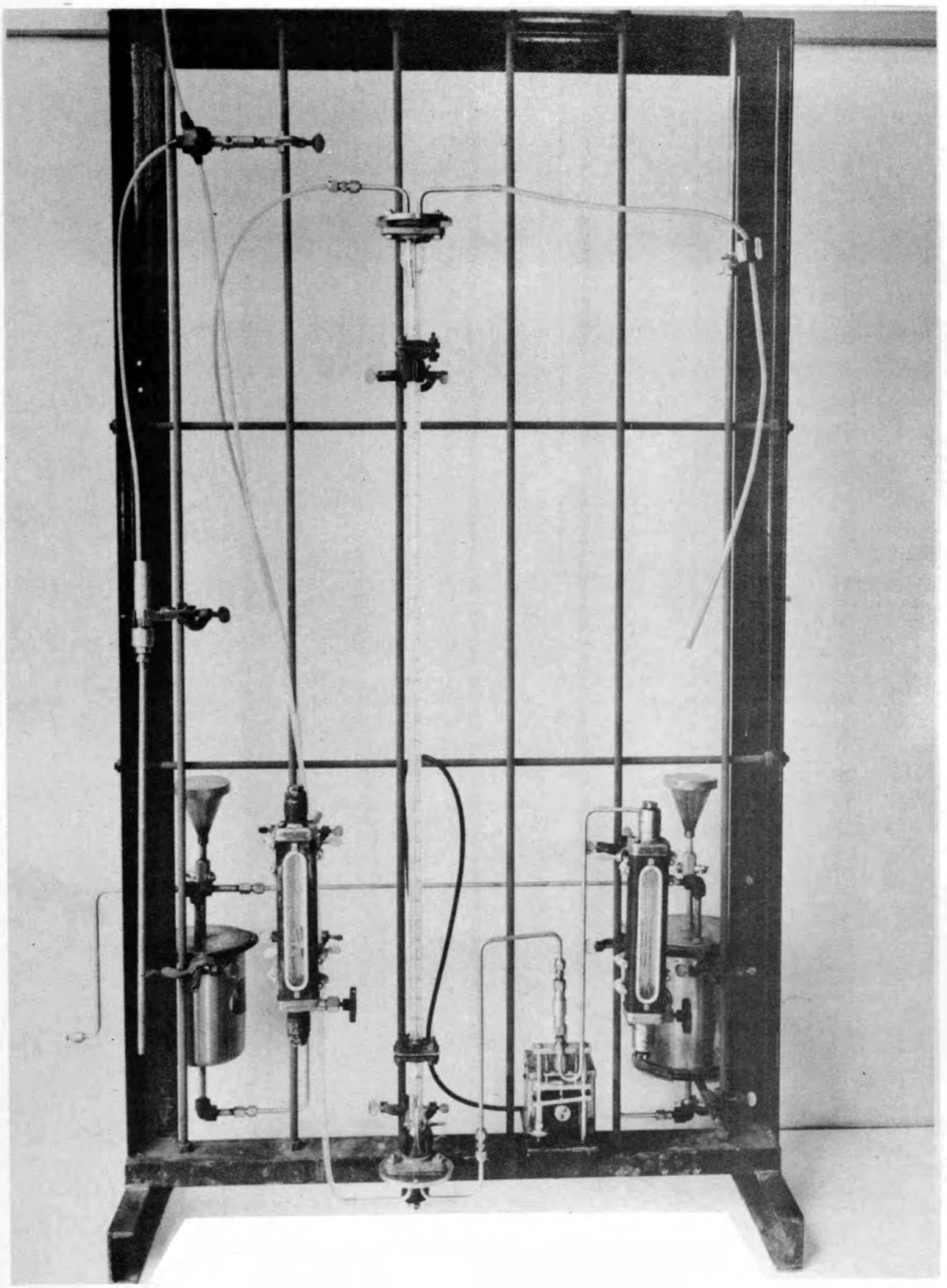

FIG. 8

Figure 9

Plate for the 1/2" pulse column made from 22 gauge s.s. having 32 holes 0.039" in diameter showing area and thickness.

[redacted]

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Neg. P-108(A)

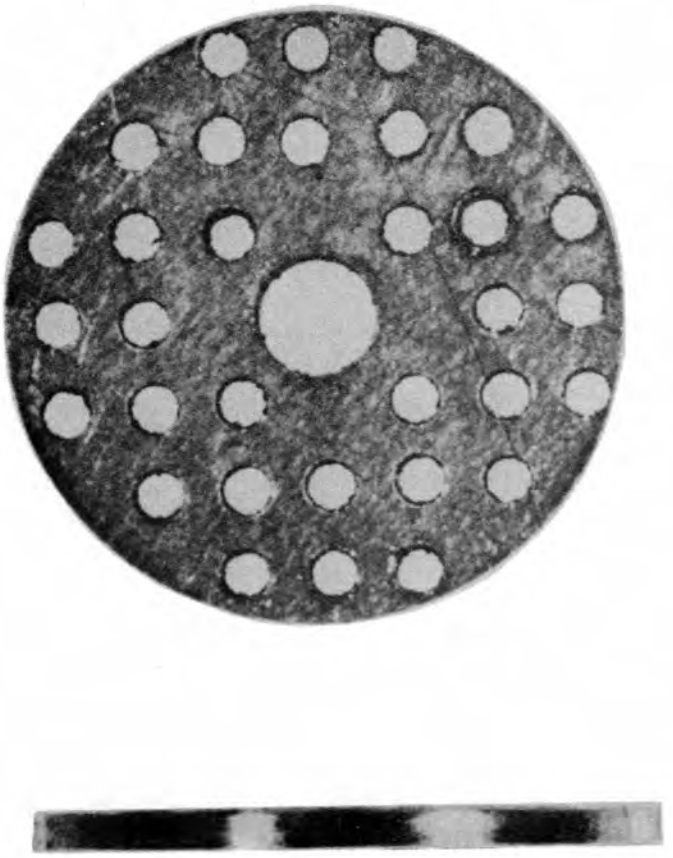

FIG. 9

Figure 10

Plate for the 1/2" pulse column made from 18 guage s.s. having 56 holes, 0.039" in diameter showing area and thickness.

[redacted]

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Neg. P-107

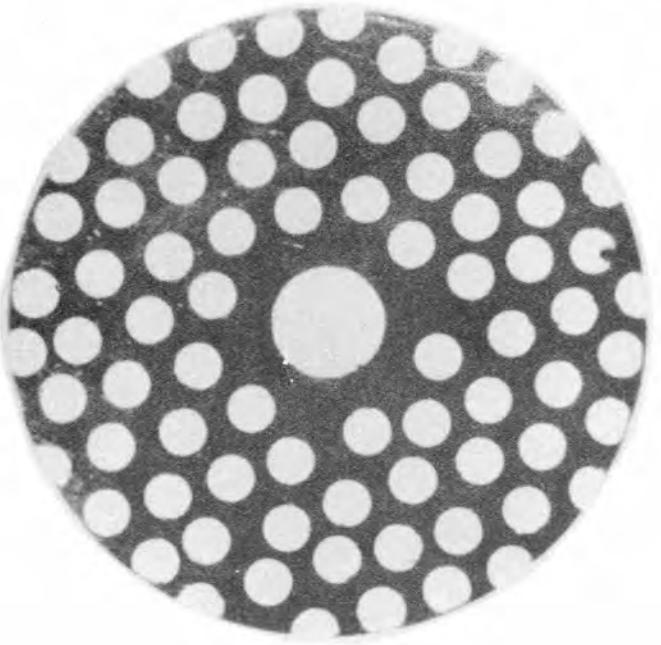

FIG. 10

Figure 11

Plate spacer for the 1/2" pulse column 3/32" I.D. 1" long. and made from s.s. tubing.

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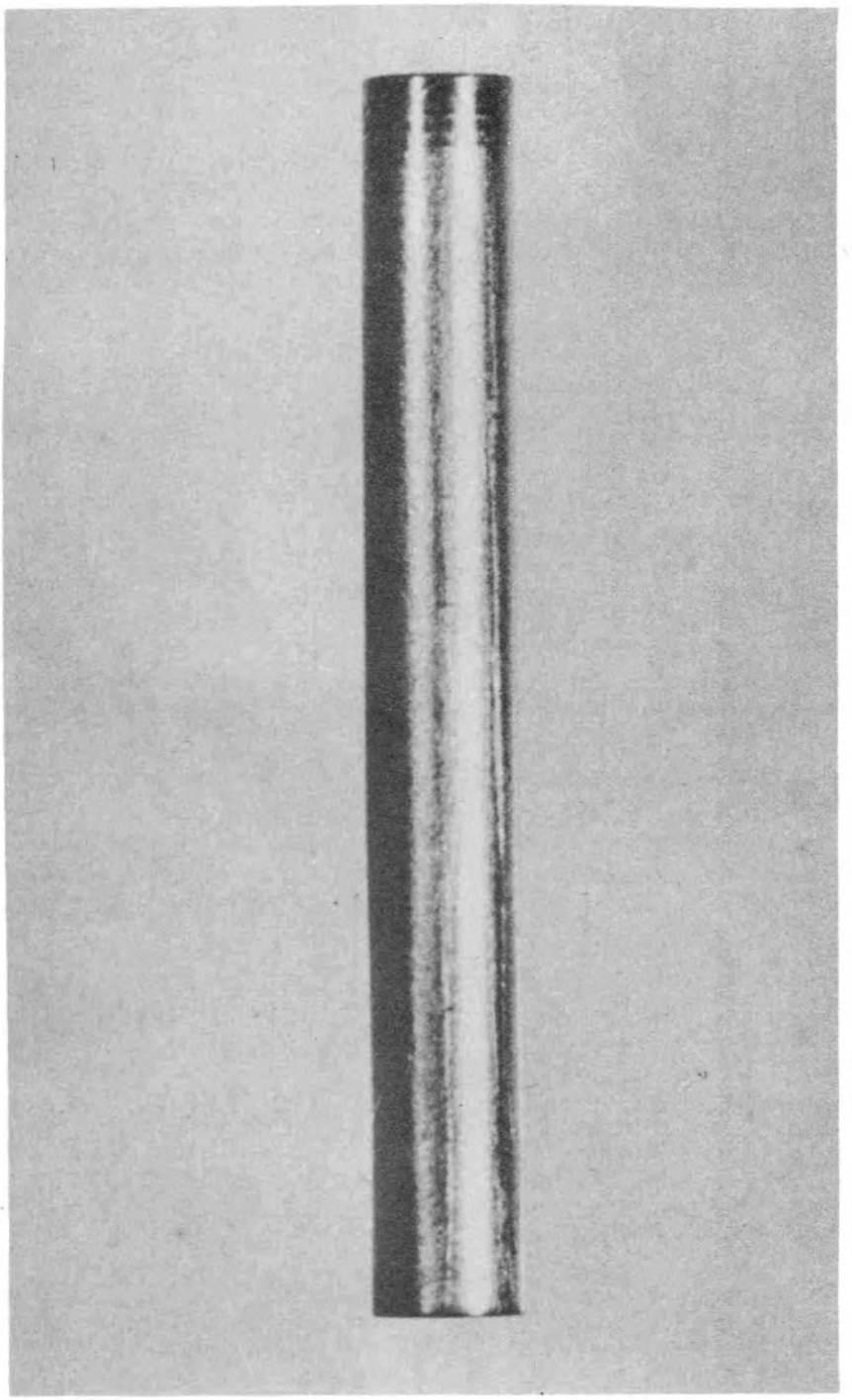

FIG. 11

Figure 12

Bellows pump for the 1/2" pulse column showing check valve assembly at top, bellows, cam and the screw for regulating the pulse amplitude by raising or lowering the bar above cam. Capacity 30-2000 cc/hr.

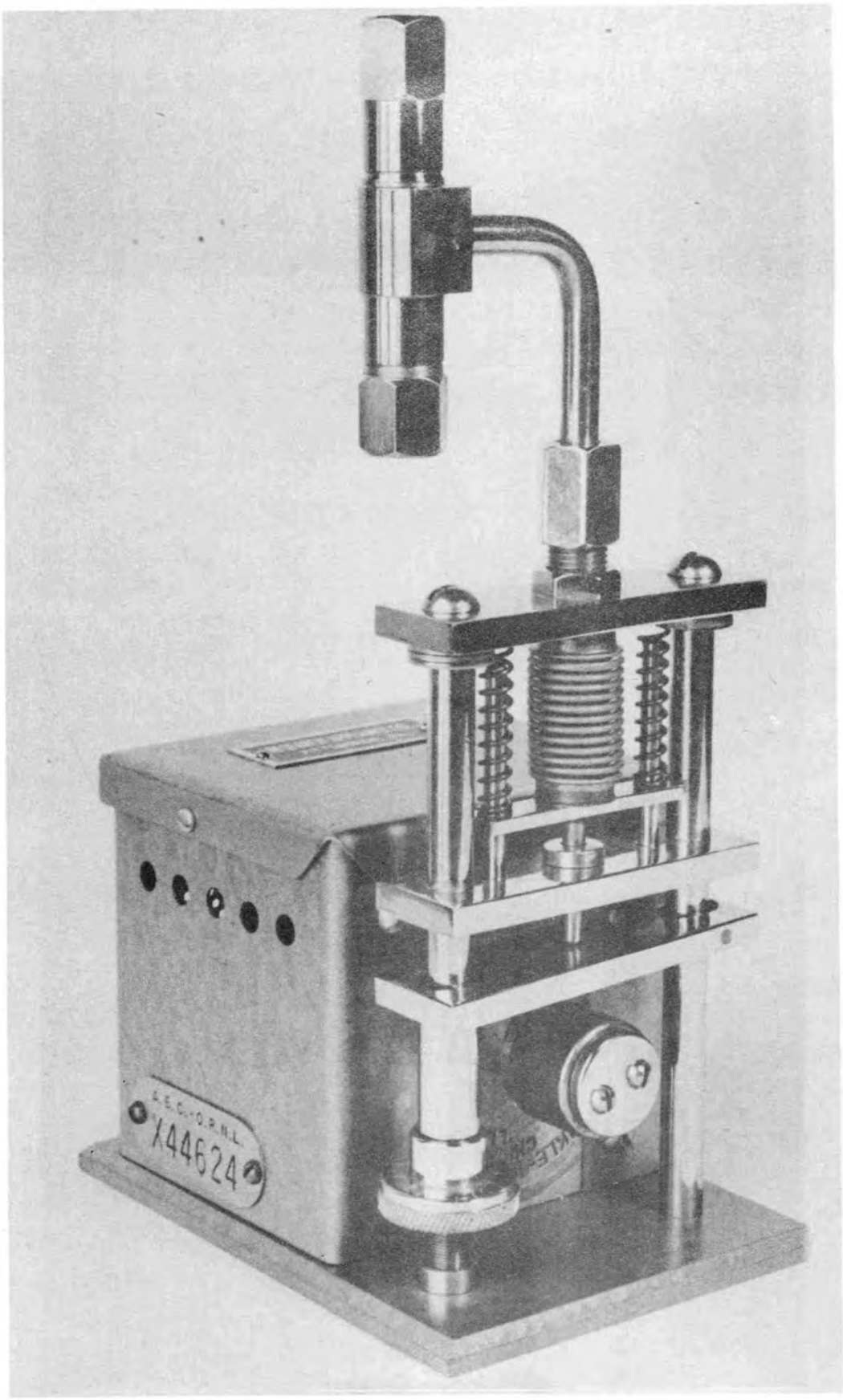

FIG. 12