SECRET

ORNL-96 2A

CHEMISTRY-SEPARATIONS PROCESSES FOR URANIUM

TECHNICAL DIVISION

LIMITING FLOW RATES IN CONTINUOUS COUNTERCURRENT EXTRACTION COLUMNS

OAK RIDGE NATIONAL LABORATORY

DIVISION OF
CARBIDE AND CARBON CHEMICALS CORPORATION
POST OFFICE BOX P
OAK RIDGE, TENNESSEE

SECRET

[redacted]

ORNL-96
July 15, 1948

OAK RIDGE NATIONAL LABORATORY

Technical Division

This Documents Consists
pages.
Copy 2 of 86 Series
ISSUED: 706-A.

Chemical Process Development Section

****

Final Report

****

LIMITING FLOW RATES IN CONTINUOUS COUNTERCURRENT EXTRACTION COLUMNS

By J. B. Ruch

Period Covered: September 1946 to January 1947

Previous Reports: None

Notebook: CL-1204

Experimental Work by: J. B. Ruch

Date Received: 7/21/48 Date Issued: AUG 9 1948

[redacted]

CONTENTS

Page No.

1.0 Abstract

Limiting flow (flooding) rates in continuous liquid-liquid countercurrent extraction columns were investigated for hexone and dibutyl cellosolve solvents, with aqueous aluminum nitrate concentrations of 1.0 and 2.0M.

2.0 Introduction

The limiting flow (flooding) rates in liquid-liquid countercurrent extraction systems depend upon the type of equipment and solution conditions. Since Data were not available from other sources, it was necessary to determine these limiting flow rates for the "25" solvent extraction process so that safe operating ranges could be established. The specific data herein apply only to certain conditions and equipment proposed for this process. However, the values obtained should serve as good approximations for operations in similar equipment using the same packing material and solvent with comparable aqueous solutions, but are not recommended as authentic values except in cases where the equipment and solution conditions are identical. No further work on determining limiting flows is planned for the present "25" process.

3.0 Summary

Limiting flow rates for each phase in hexone systems proposed for the "25" solvent extraction process, using a one to one aqueous to organic flow ratio, were found to be: 570 gal/hr./sq.ft. with 3/8"x3/8" Raschig rings, 465 gal/hr./sq.ft. with 1/4"x3/8" Raschig rings and 370 gal/hr./sq.ft. with 3/16" helices packing. For these investigations the continuous phase was 1.0M Aluminum nitrate. The curves in Figure 2 through 6 show limiting flow rates at various flow ratios. Figure 2 is for hexone-aqueous stripping solutions with 3/16" helices packing in 1", 2" and 3" columns. Figures 3 through 6 are for dibutyl cellosolve with both aqueous extraction and stripping solutions using 3/8"x3/8" Raschig rings, 1/4"x3/8" Raschig rings and 3/16" helices packings in 1-1/2" or 3" columns.

The general conclusions are:

1. From the standpoint of limiting flow considerations, both hexone and dibutyl cellosolve proved to be satisfactory solvents for the continuous liquid-liquid countercurrent extraction process.
2. Within the range studied (1-1/2" to 3"), column diameter has no appreciable affect upon the limiting flow rates.
3. The limiting flow rates decrease with a decrease in the size of packing materials.
4. Limiting flow rates are higher for hexone systems than for dibutyl cellosolve systems.

4.0 Experimental Work

4.1 Equipment

Limiting flow investigations were made in 1-1/2", 2", and 3" glass columns. The stainless steel packing material used were: 3/8"x3/8" Raschig rings, 1/4"x3/8" Raschig rings and 3/16" helices retained by a conical stainless steel support (See Figure 1).

4.2 Solution Properties

Concentrations and properties are shown in Table No. 1 of the solutions for two processes.

Table No. 1

Solution Concentrations and Properties

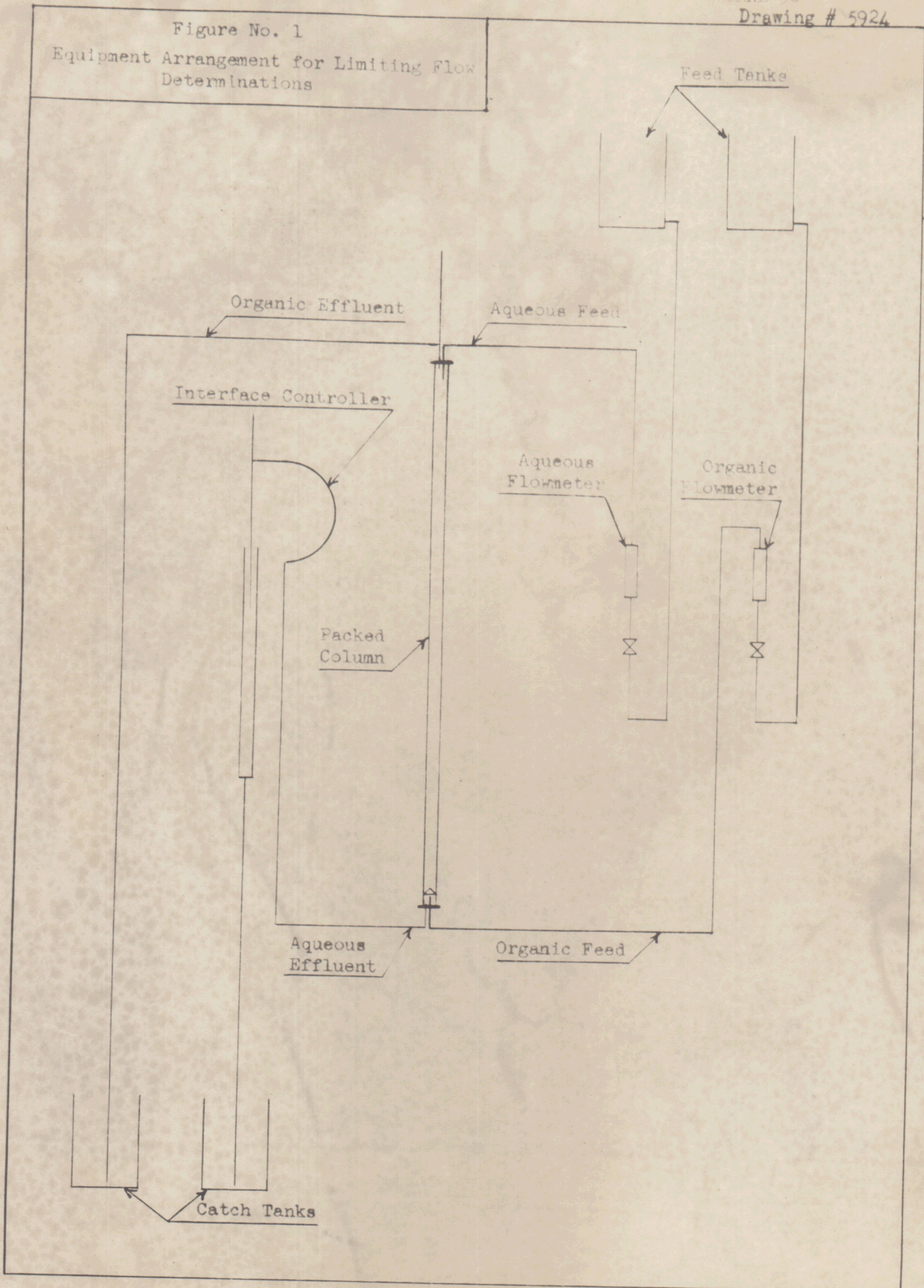
OPERATION	AQUEOUS Chemical Composition	AQUEOUS Sp. G.	AQUEOUS Vis.* (centp.)	ORGANIC Chemical Composition	ORGANIC Sp. G.	ORGANIC Vis.* (centp.)	INTERFACIAL TENSION*
Hexone Systems Extraction	1.0M $Al(NO_3)_3$ 0.6N HNO_3	1.165	1.97	0.5M HNO_3	0.804	0.59	9.7 dynes/cm
Stripping	0.1M HNO_3	1.000	0.896	0.5M HNO_3	0.804	0.59	11.0 dynes/cm (approximately)
Dibutyl Cellosolve Extraction	2.0M $Al(NO_3)_3$ 0.1M HNO_3	1.305	5.57	0.17M HNO_3	0.837	1.34	19 dynes/cm. (Approximately)
Stripping	0.1M HNO_3	1.000	0.896	0.17M HNO_3	0.837	1.34	19 dynes/cm (Approximately)

*These data were obtained from E. H. Turk of the Chemistry Division.

Drawing # 5924

Figure No. 1

Equipment Arrangement for Limiting Flow Determinations



4.3 Procedure

In all systems investigated the aqueous was the continuous phase and had the higher specific gravity. To start a run the column was filled with aqueous phase, the organic flow started at a low rate, and an interface was established. The aqueous flow rate was held constant at a point where the limiting flow was to be determined. To obtain a first approximation, the organic flow rate was increased rapidly while the aqueous jackleg was adjusted to hold the interface constant. When the interface could no longer be held constant and/or it was possible to observe organic "build-up" beneath the packing support, the limiting flows had been exceeded. Then the organic flow rate was decreased approximately 30% and an interface again established. To determine the limiting flows more accurately, the organic was increased very slowly in increments of 10-15 gal./hr./sq.ft. every five minutes, and the aqueous flow held constant. When the interface again became uncontrollable and/or an organic "build up" occurred beneath the packing support, these rates were considered the limiting flows. This procedure was repeated with different settings of the aqueous flow until enough points were determined to cover the desired range for the limiting flow curves.

4.4 Results

4.41 Hexone Systems

Limiting flows for hexone systems were determined only for one to one aqueous to organic flow ratios, with aqueous extraction solutions. Table 2 shows that for these conditions in 3" columns the limiting flows decrease with the size of packing material. Using the 3/8"x3/8" Raschig rings as the basis for comparison, the limiting flows were only 81% as high for 1/4"x3/8" Raschig rings and 65" as high for 3/16" helices. The limiting rates for extraction solutions containing 0.10M sodium dichromate were identical

Table No. 2

Limiting Flows
Hexone-Aqueous Extraction Solutions

Conditions:

1. 3" glass column
2. Packed height 31"
3. 1:1 Aq. to Org. flow ratios
4. Aqueous continuous phase

Packing Material	Limiting Flows in gal./hr./sq.ft.*
3/8"x3/8" Raschig rings	572 : 572
1/4"x3/8" Raschig rings	464 : 464
3/16" helices	371 : 371

*Based on cross-sectional area of unpacked column

with those with no dichromate.

Data for hexone with aqueous stripping solutions were obtained for limiting flow curves, using 3/16" helices packing in 1-1/2", 2", and 3" glass columns and are plotted in Figure 2. The curves are identical except for deviations caused by experimental error, which is ± 30 gal/hr./sq.ft.

for each phase. Some inconsistency can be seen by grouping the first three and last three points of the 1-1/2" column curve (points marked X). Apparently this was caused by flowmeter calibration errors since it was necessary to use different flowmeters for high and low ranges.

4.42 Dibutyl Cellosolve Systems

Dibutyl cellosolve systems using both aqueous extraction and stripping solutions were investigated with the various packing materials in 3" glass columns, and with 3/16" helices packing in 1-1/2" glass columns. The curves in Figure 3 show that with extraction solutions, the total column throughputs for both phases at the limiting flows are approximately: 950 gal/hr./sq.ft. with 3/8"x3/8" Raschig rings, 750 gal/hr./sq.ft. with Raschig rings and 600 gal/hr./sq.f.t with 3/16" helices. When 3/8"x3/8" Raschig rings are used as the basis for comparison the limiting flows are about 79% as high for 1/4"x3/8" Raschig rings and 63% as high for 3/16" helices. The curves for stripping solutions shown in Figure 4 are similar to those for extraction solutions; however, limiting flows are approximately 26% lower for stripping solutions as shown in Figure 5.

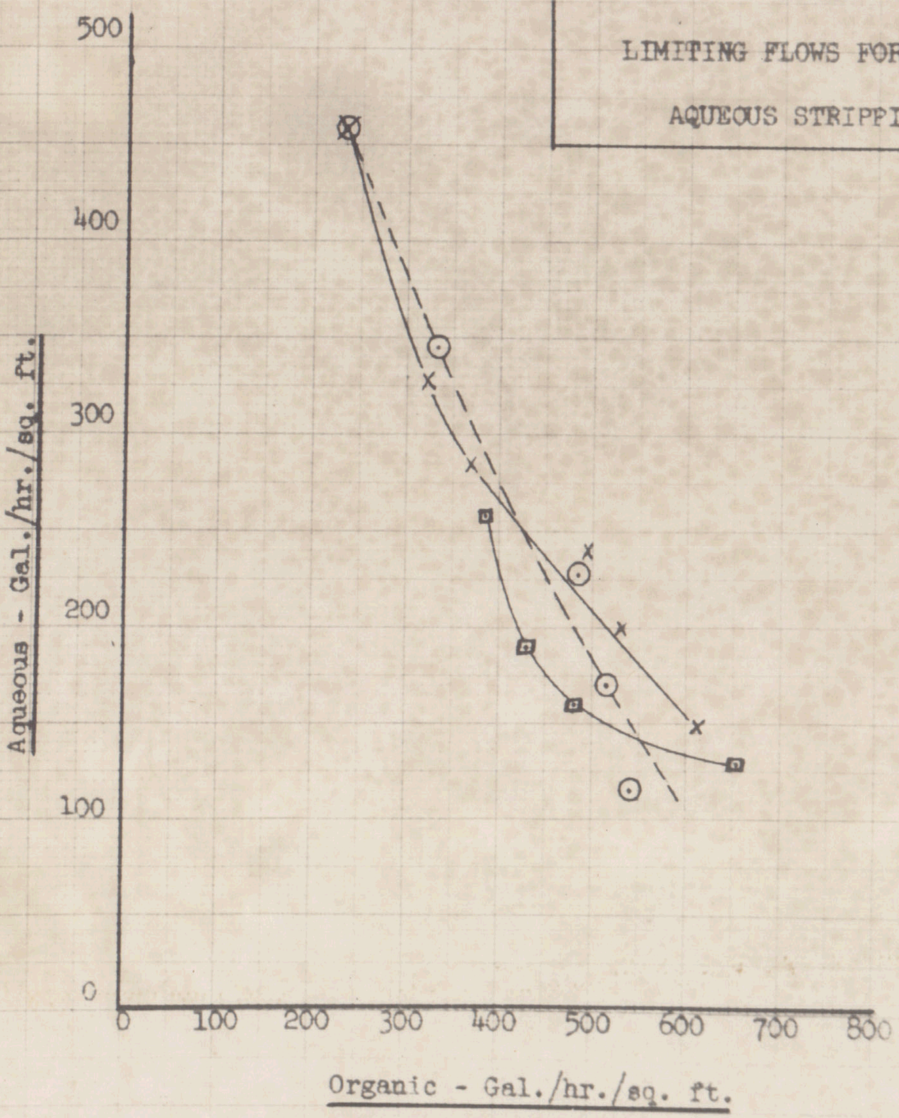
4.43 Comparison of Solvent Systems

For stripping solution conditions in 1-1/2" columns with 3/16" helices packing the limiting flows were approximately 50% higher in hexone systems (See Figure 6). From Table 1 and Figure 3 it can be seen that the limiting flows are approximately 22% higher in hexone systems with extraction solution conditions using 3/16" helices packing.

FIGURE NO. 2

LIMITING FLOWS FOR HEXONE SYSTEMS

AQUEOUS STRIPPING SOLUTIONS



GLASS COLUMN DIAMETER

⊙ - 3"
⊡ - 2"
× - 1 1/2"

SOLUTIONS

Aqueous: 0.1 N HNO_3
Organic: 0.4 N HNO_3

PACKING MATERIAL

3/16" Helices

PACKED HEIGHT

31"

AQUEOUS PHASE CONTINUOUS

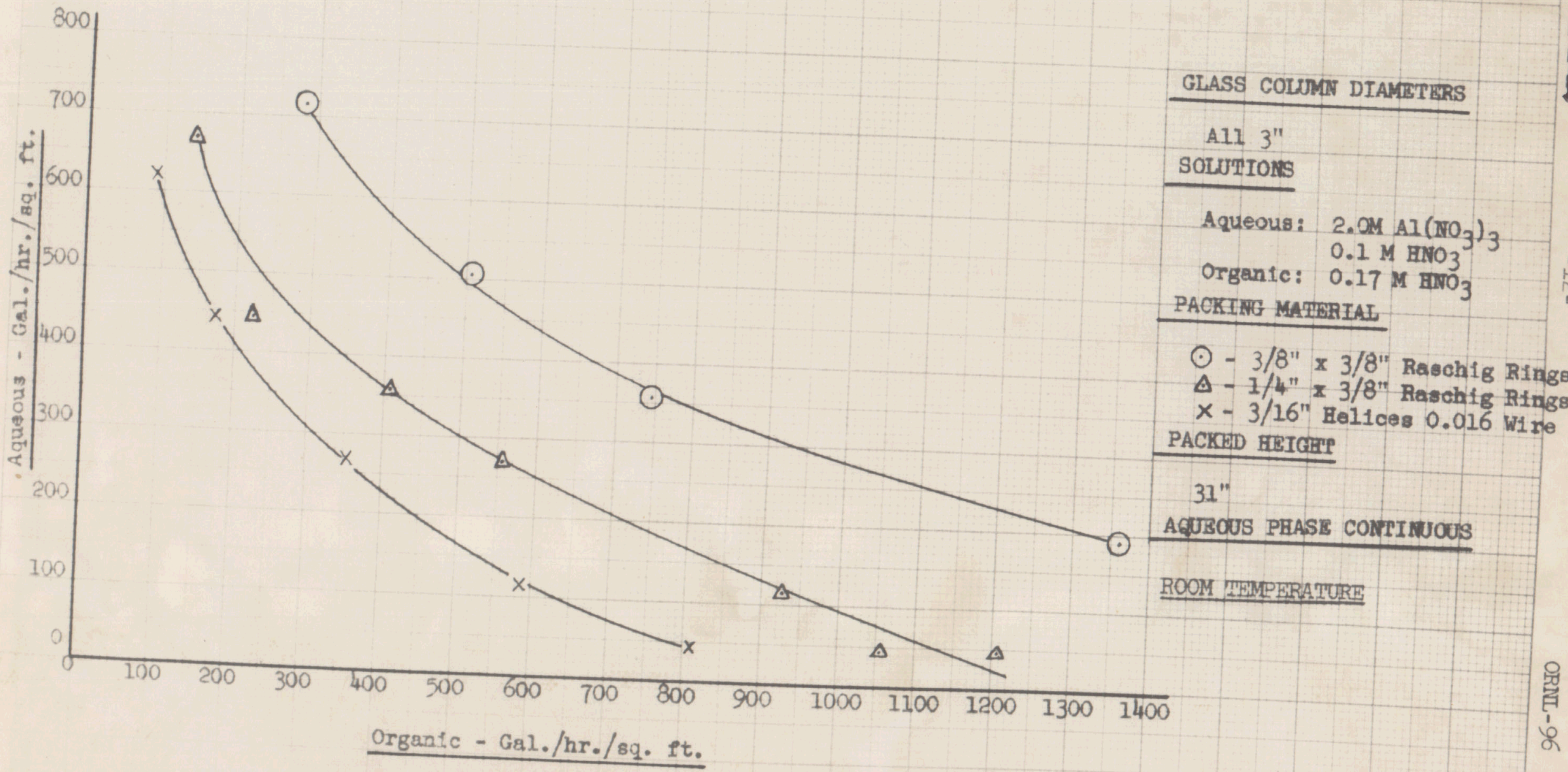
ROOM TEMPERATURE

ORNL-96 Drawing # 592

FIGURE NO. 3

LIMITING FLOWS FOR DIBUTYL CELLOSOLVE SYSTEMS

AQUEOUS EXTRACTION SOLUTIONS



-12-

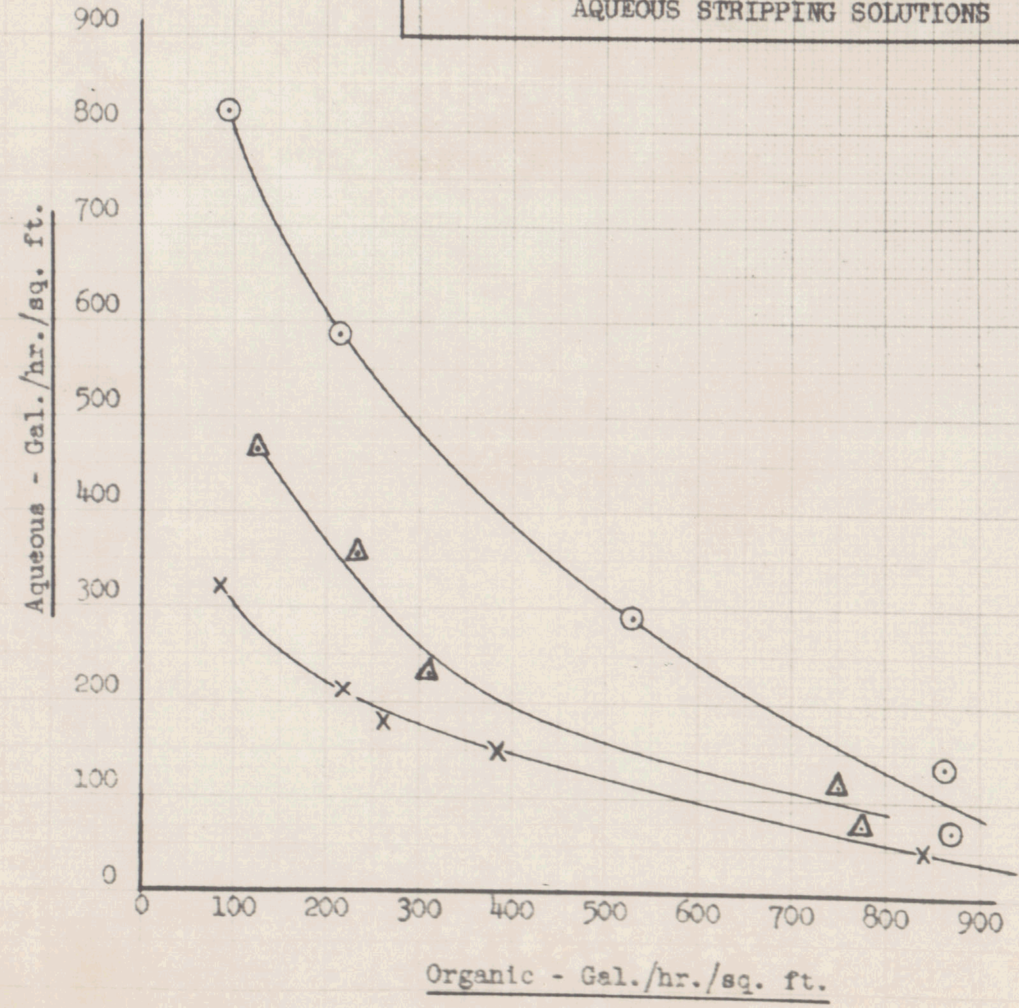
ORNL-96

Dwg.# 59

FIGURE NO. 4

LIMITING FLOWS FOR DIBUTYL CELLOSOLVE SYSTEMS

AQUEOUS STRIPPING SOLUTIONS



GLASS COLUMN DIAMETERS

3" For Raschig Ring Packings
1 1/2" For Helices Packings

SOLUTIONS

Aqueous: 0.1 N HNO_3
Organic: 0.17 N HNO_3

PACKING MATERIALS

⊙ - 3/8" x 3/8" Raschig Rings
Δ - 1/4" x 3/8" Raschig Rings
X - 3/16" Raschig Rings

PACKED HEIGHT

31"

AQUEOUS PHASE CONTINUOUS

ROOM TEMPERATURE

SECRET

ORNL-96 Dwg. # 5927

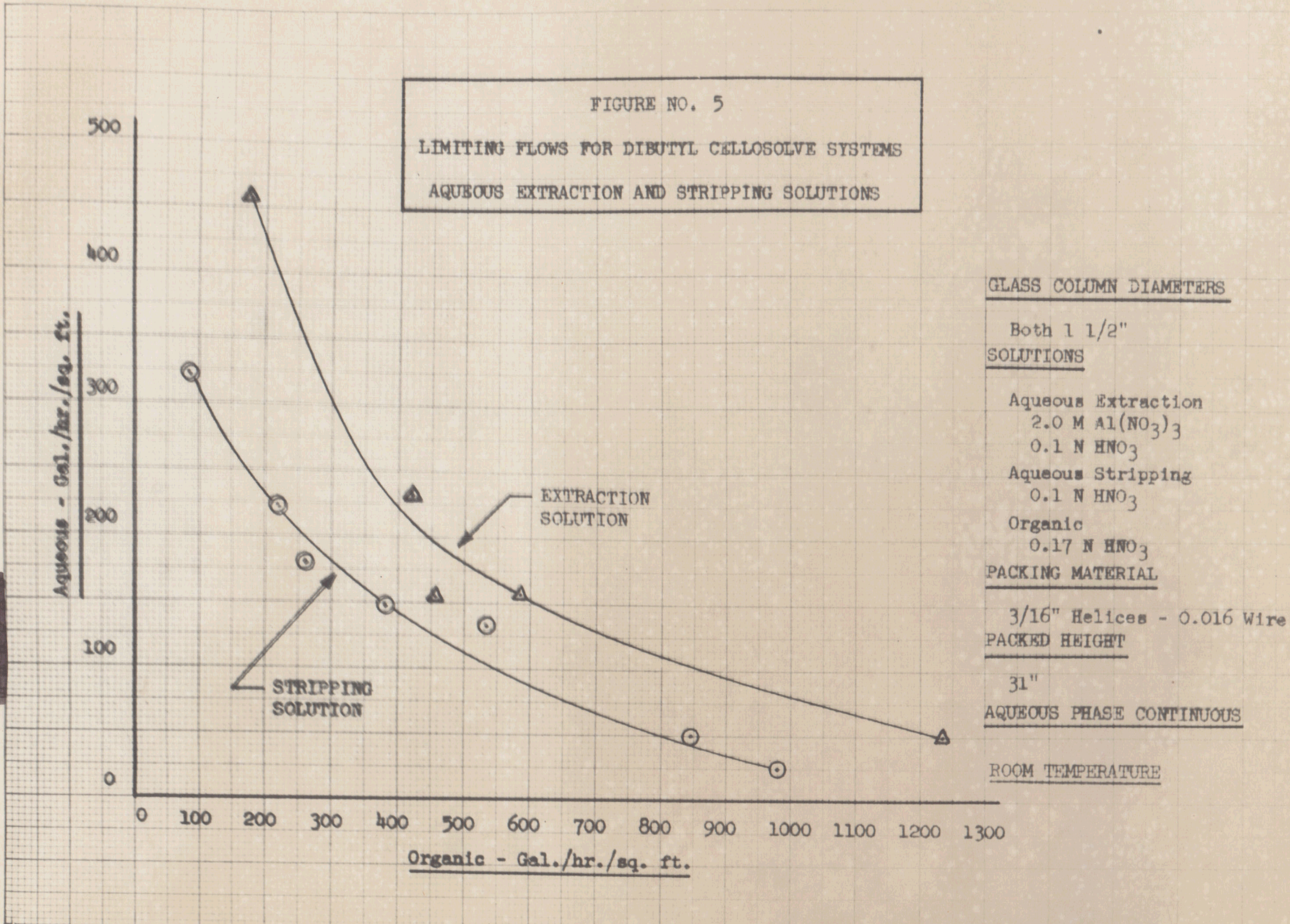


FIGURE NO. 5

LIMITING FLOWS FOR DIBUTYL CELLOSOLVE SYSTEMS

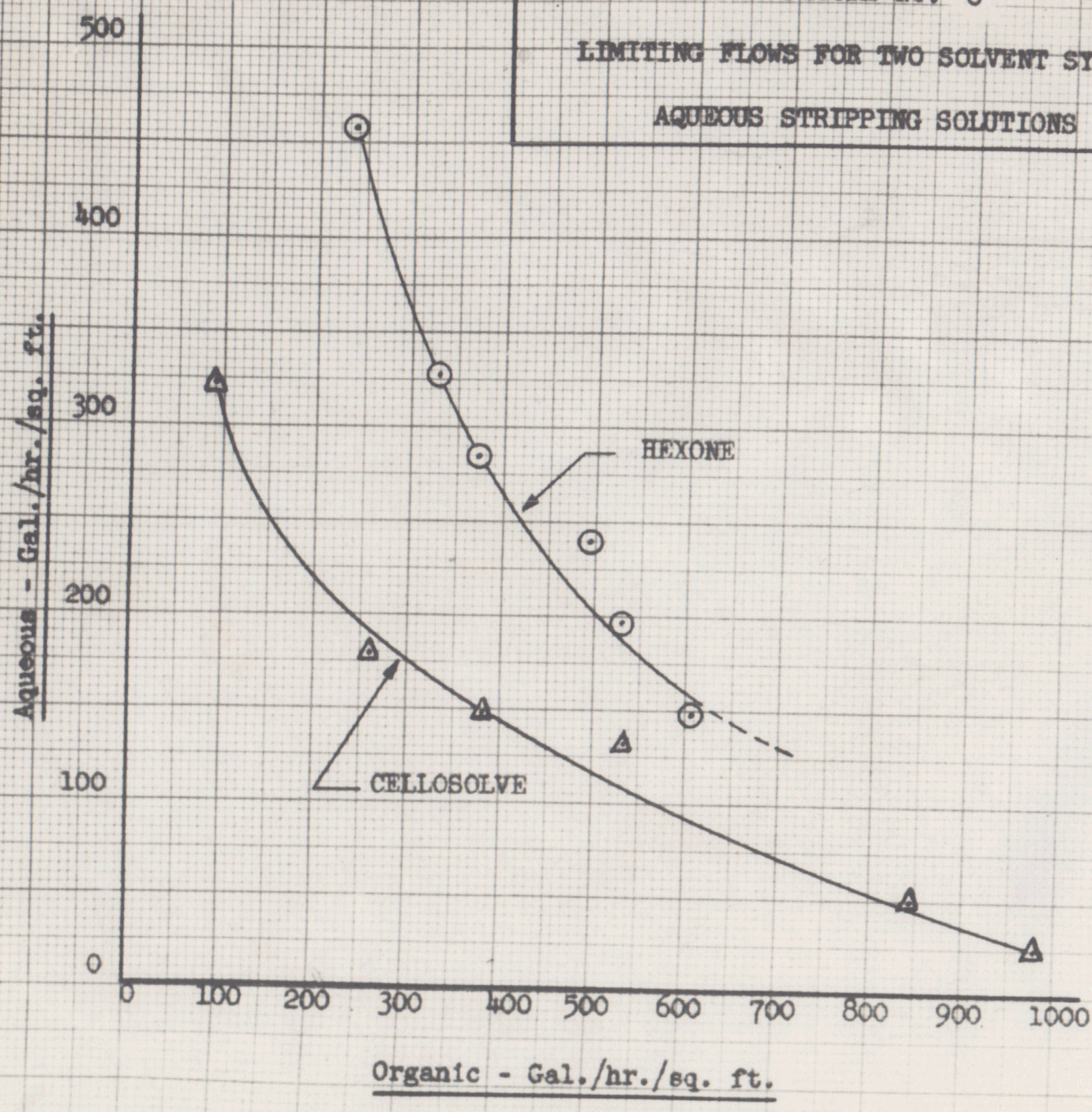
AQUEOUS EXTRACTION AND STRIPPING SOLUTIONS

ORNL-96 Drawing# 5928

FIGURE NO. 6

LIMITING FLOWS FOR TWO SOLVENT SYSTEMS

AQUEOUS STRIPPING SOLUTIONS



GLASS COLUMN DIAMETERS

Both 1 1/2"

SOLUTIONS

Aqueous: 0.1 N HNO_3
Hexone: 0.4 N HNO_3
Dibutyl Cellosolve: 0.17 N HNO_3

PACKING MATERIALS

3/16" Helices - 0.016 Wire

PACKED HEIGHT

31"

AQUEOUS PHASE CONTINUOUS

ORNL-96
Drawing # 5929

5.0 Appendix

5.1 Equipment Details

Both feeds entered the columns through 1/2" saran tubing. The organic feed tube extended through the bottom to the base of the packing support and was centered in the cross section of the column. The aqueous feed tube extended through the top approximately 6" and was also centered in the cross section of the column. The effluents were removed from the extremities of the column and the aqueous effluent was sent through the jackleg for interface control

The packing materials were stainless steel Raschig rings and helices, the dimensions of which are shown in Table 3.

Table No. 3

Packing Dimensions

Type	O.D.	I.D.	Length	Wall Thickness
Raschig split ring	3/8"	1/4"	3/8"	1/16"
Raschig split ring	1/4"	3/16"	3/8"	1/32"
Helices (Fenske)	3/16"	-	-	.016 Wire

All packing was made of stainless steel

The conical stainless steel packing supports used in this work had an altitude equal to the diameter of the base and the surface area was filled with 3/8" holes. The open area through a support was approximately 60% the cross sectional area of the unpacked column.

5.2 Column Packing Methods

All columns were packed 31" high. For packing Raschig rings, the column was filled with water and the rings were dropped in a hand full at a time. The helices (Fenske) were dropped into a dry column in slugs of about a cubic inch and tamped down with a wooden rod after each slug.

5.3 Solution Materials and Preparation

The solutions were prepared from technical grade aluminum nitrate, nitric acid, hexone and dibutyl cellosolve. No pretreatment of the solvents or filtration of aqueous solutions was performed. Uranium and sodium dichromate were not present in the aqueous solutions because test runs showed that at their low concentrations they did not affect the limiting flows. Without these ingredients the extraction and scrubbing solutions were obviously the same.