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CHALK RIVER PROJECT

RESEARCH AND DEVELOPMENT

PULSED AMPLITUDE AND FREQUENCY EFFECTS IN A PULSED PACKED COLUMN

CEI-69

BY

S.H. RUSSELL

CHALK RIVER, ONTARIO

APRIL, 1954

ATOMIC ENERGY OF CANADA LIMITED
Research and Development

PULSE AMPLITUDE AND FREQUENCY EFFECTS IN A PULSED PACKED COLUMN

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SUMMARY

A study has been made of the effect on the efficiency and capacity of applying pulses of varying amplitude and frequency to a packed column.

In the efficiency studies, the maximum efficiency was obtained with a pulse having an amplitude of 3/8" and a frequency of 140 cycles per minute. Under these conditions, the column was about five times as efficient as a simple packed column.

Two general types of results were obtained in the capacity studies. Under certain conditions, the capacity increased over that of a simple packed column, but under others, it decreased. Some of the factors causing this were investigated but the fundamental reasons were not determined due to a lack of personnel for the necessary experiments.

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PULSE AMPLITUDE AND FREQUENCY EFFECTS IN A PULSED PACKED COLUMN

1.0 INTRODUCTION

When it was first decided to build a pilot plant to study the TBP process, the final selection of the contactor to be used was left open. Packed columns were considered to be unsatisfactory due to the length of the columns that would be required. Pulsed packed columns and airlift contactors were thought to be more satisfactory but an extensive development program was needed to determine the best design and performance characteristics of each before a final choice could be made. The development of the airlift contactor is dealt with in another report (1). This report describes the studies made on a pulsed packed column.

The studies can be divided into two sections. In the first section, studies on the efficiency of the column are described. The second section deals with the studies on the capacity of the column.

2.0 EFFICIENCY STUDIES

2.1 Theory

In this study, the efficiency of the column was determined from the extraction rates that were obtained for the various combinations of pulse amplitude and frequency. The extraction rates were interpreted in terms of the height of a transfer unit (HTU).

The HTU is obtained from the equation

$$HTU = \frac{Z}{NTU}$$

where Z is the length of a column and NTU is the number of transfer units contained in that length. In differential form

$$HTU = \frac{dZ}{d(NTU)} \qquad (1)$$

The system used here (the back-washing of uranium from a 20% TBP-Turpolene solution by water) involved dilute solutions and the principal resistance to the transfer of the uranium lay in the solvent phase. Under these conditions,

$$NTU_{oo} = \int_{W_{o_2}}^{W_{o_1}} \frac{dW_o}{W_o - W_o^{\bigstar}}$$

where W = uranium concentration, weight uranium /weight nonsolute
subscript o = solvent phase
subscript 1 = that end of the column where solutions are concentrated
subscript 2 = that end of the column where solutions are dilute
superscript ★ = uranium concentration that would exist if the system were at equilibrium (see Fig. 1)

In differential form, this becomes

$$d(NTU)_{oo} = \frac{dW_o}{W_o - W_o^{\bigstar}} \qquad (2)$$

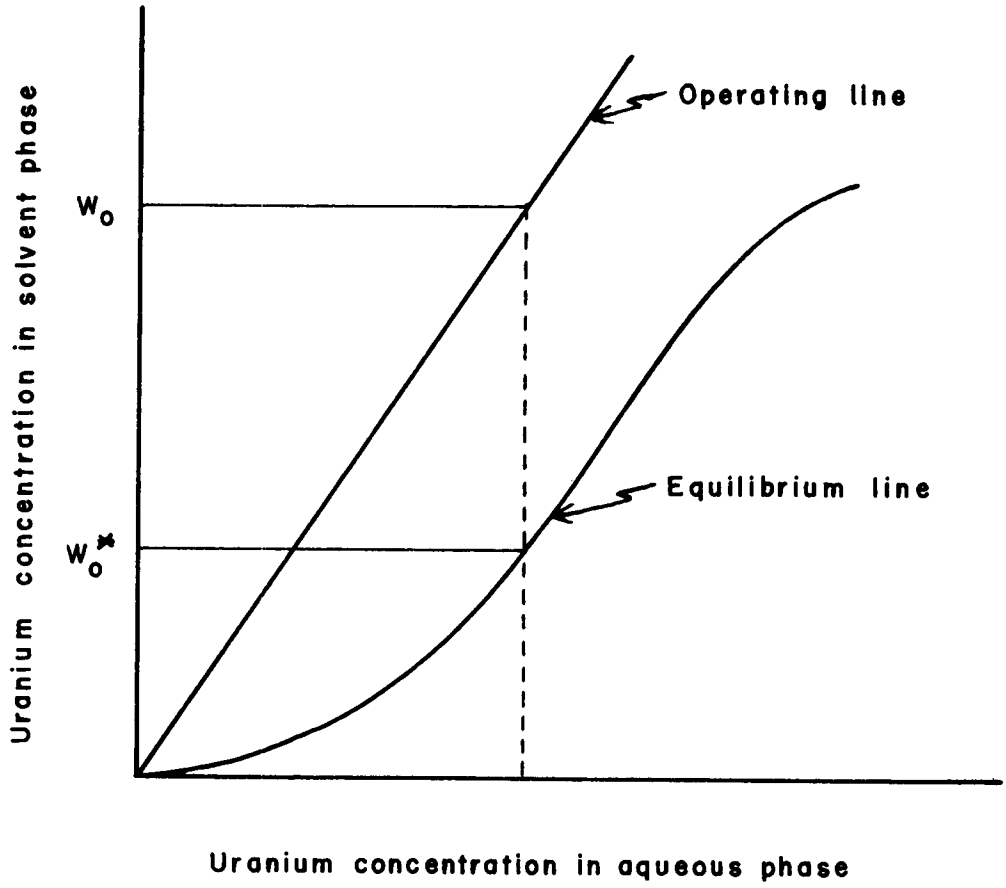

Figure 1 Typical plot for obtaining values of $W_o - W_o^*$

By substitution in equation (1)

$$HTU_{OO} = \frac{dZ}{dW_O / W_O - W_O^*}$$

or, by rearrangement,

$$HTU_{OO} = \frac{W_O - W_O^*}{dW_O / dZ} \qquad (3)$$

This expression allows values of HTU_{OO} to be calculated for various values of W_O since $W_O - W_O^*$ can be determined from the operating and equilibrium lines for the system and dW_O / dZ can be obtained from a graph showing the variation of the uranium concentration with length of column.

Thus, for any particular combination of pulse amplitude and frequency, a graph showing the variation of HTU_{OO} with uranium concentration in the solvent phase can be obtained. The optimum combination can then be determined by comparing the HTU's for any particular uranium concentration.

2.2 Apparatus

The apparatus used is illustrated in Figure 2. The column was built from 4-inch Pyrex glass sections and a stainless steel section 2 ft. long. Sampling lines entered the column through the steel section, these lines permitting samples of continuous phase to be withdrawn as required. Only the steel section was packed with ½-inch ceramic Raschig rings.

Pulses were generated mechanically through a flexible Teflon diaphragm attached to the bottom of the column. The

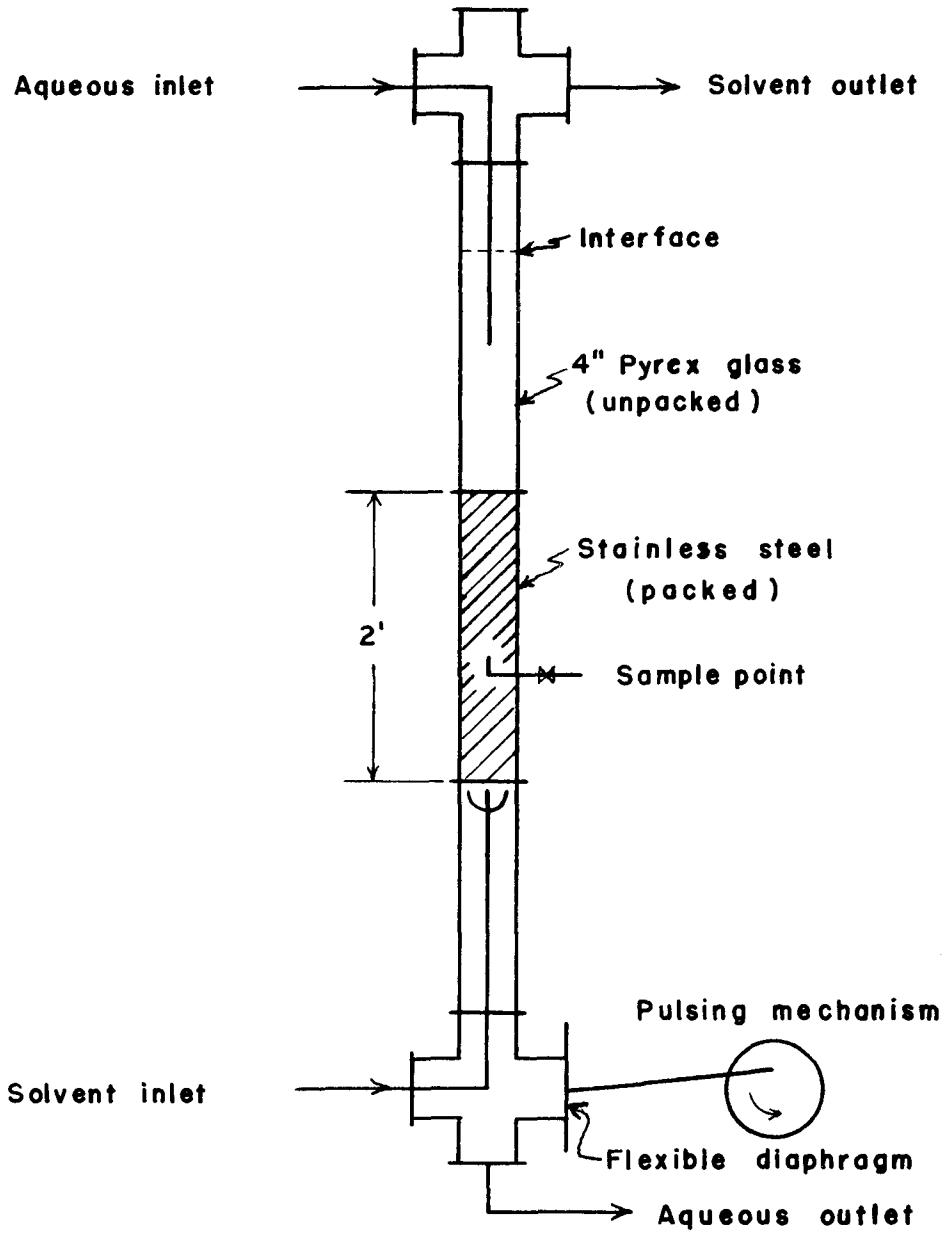

Figure 2 Diagram of column used for efficiency studies.

diaphragm was actuated by an arm attached eccentrically to a drive wheel rotated by an electric motor. Pulse amplitudes were varied by changing the degree of eccentricity of the arm on the drive wheel. The amplitude was determined by measuring the total excursion of the aqueous-solvent interface as the pulsing mechanism turned through one complete cycle. The pulse frequency was varied either by changing the gearing in the gear chain between the motor and drive wheel or by altering the line voltage to the motor by means of a Variac.

The flow rates of the streams entering the column were kept constant by means of calibrated rotameters, and were checked with calibrated burettes on the outlet lines.

Uranium concentrations in the various samples were determined colourimetrically by the peroxide method.

2.3 Procedure

For each experiment, the pulse amplitude and frequency were adjusted to the desired values. The solvent and aqueous inlet streams were then admitted to the column. The flow rates of these streams were checked several times during an experiment by means of the calibrated burettes. After sufficient time had elapsed for the column to reach equilibrium, samples were taken of the solvent feed stream, of the solvent and aqueous outlet streams, and of the aqueous phase at each of the sample points along the column. (In all of these experiments, the aqueous phase formed the continuous phase in the column). The uranium concentration in each of these

samples was then determined. The material balance across the column was then checked from the uranium concentrations in the inlet and outlet streams and the flow rates of each. If satisfactory (i.e. within $\pm$ 5%), the variation of the uranium concentration of the solvent phase along the column was calculated by means of a material balance from the measured variation of the uranium concentration in the aqueous phase. A graph was then drawn showing this variation with column length. From the slope of the curve so obtained, values of dW_o / dZ were obtained for particular values of W_o. The values of $W_o - W_o^*$ for these same values of W_o were then obtained from another graph containing the equilibrium curve for this system and the operating line for the experiment. Values of HTU_{oo} for these particular values of W_o were then calculated from equation (3), and graphs were drawn showing the variation of HTU_{oo} with uranium concentration in the solvent phase.

The experiment was then repeated with another combination of pulse amplitude and frequency until all combinations in the range of pulse amplitudes from 1/8" to 1/2" and of frequencies from 40 to 200 cycles per minute had been studied. The results are given in Table I.

From these results, graphs were drawn showing the variation of HTU_{oo} with frequency and amplitude for two different uranium concentrations in the solvent phase. These graphs are shown in Figure 3.

TABLE I

VARIATION OF HTU WITH PULSE AMPLITUDE, PULSE FREQUENCY AND URANIUM CONCENTRATION IN THE SOLVENT PHASE

Pulse amplitude	Pulse frequency	Uranium concentration in solvent	HTU_{oo}
inches	cycles per minute	mgms.U/ml.	inches
1/8	80	48.0	29.3
		41.6	25.9
		35.2	22.5
		28.8	20.4
1/8	80	48.0	36.6
		41.6	30.2
		35.2	26.2
		28.8	23.1
1/8	140	54.4	36.5
		48.0	28.5
		41.6	26.8
		35.2	24.0
		28.8	21.1
1/8	140	54.4	25.3
		48.0	25.9
		41.6	24.6
		35.2	22.2
		28.8	19.9
1/8	190	48.0	30.6
		41.6	27.8
		35.2	25.4
		28.8	22.0
1/8	260	41.6	20.4
		35.2	18.7
		28.8	16.6
		22.4	14.6
1/4	40	41.6	24.1
		35.2	24.0
		28.8	21.7
		22.4	18.0
1/4	40	44.8	32.7
		38.4	27.0
		32.0	27.2
		25.6	25.2
1/4	80	54.4	22.9
		48.0	20.3
		41.6	17.8
		35.2	15.5
		28.8	13.0
		22.4	10.3
1/4	80	51.2	24.8
		44.8	21.8
		38.4	19.5

TABLE I (continued)

Pulse amplitude	Pulse frequency	Uranium concentration in solvent	HTU_{oo}
inches	cycles per minute	mgms.U/ml.	inches
1/4	80	32.0	16.6
		25.6	13.6
1/4	145	48.0	12.6
		40.0	9.3
		32.0	8.2
		24.0	7.7
		16.0	6.9
		8.0	5.3
1/4	145	48.0	12.8
		40.0	9.8
		32.0	8.7
		24.0	7.9
		16.0	7.1
		8.0	5.2
1/4	190	48.0	14.0
		41.6	11.8
		35.2	10.7
		28.8	9.7
		22.4	8.6
		16.0	7.7
		9.6	6.7
1/2	40	44.8	23.0
		38.4	21.5
		32.0	19.6
		25.6	18.6
		19.2	20.9
1/2	80	51.2	12.0
		44.8	12.5
		38.4	12.8
		32.0	12.8
		25.6	12.2
		19.2	11.6
1/2	145	48.0	17.2
		41.6	12.5
		35.2	11.1
		28.8	9.9
		22.4	8.9
		16.0	7.6
		9.6	6.2
1/2	200	48.0	16.1
		41.6	15.3
		35.2	14.3
		28.8	14.4
		22.4	18.3

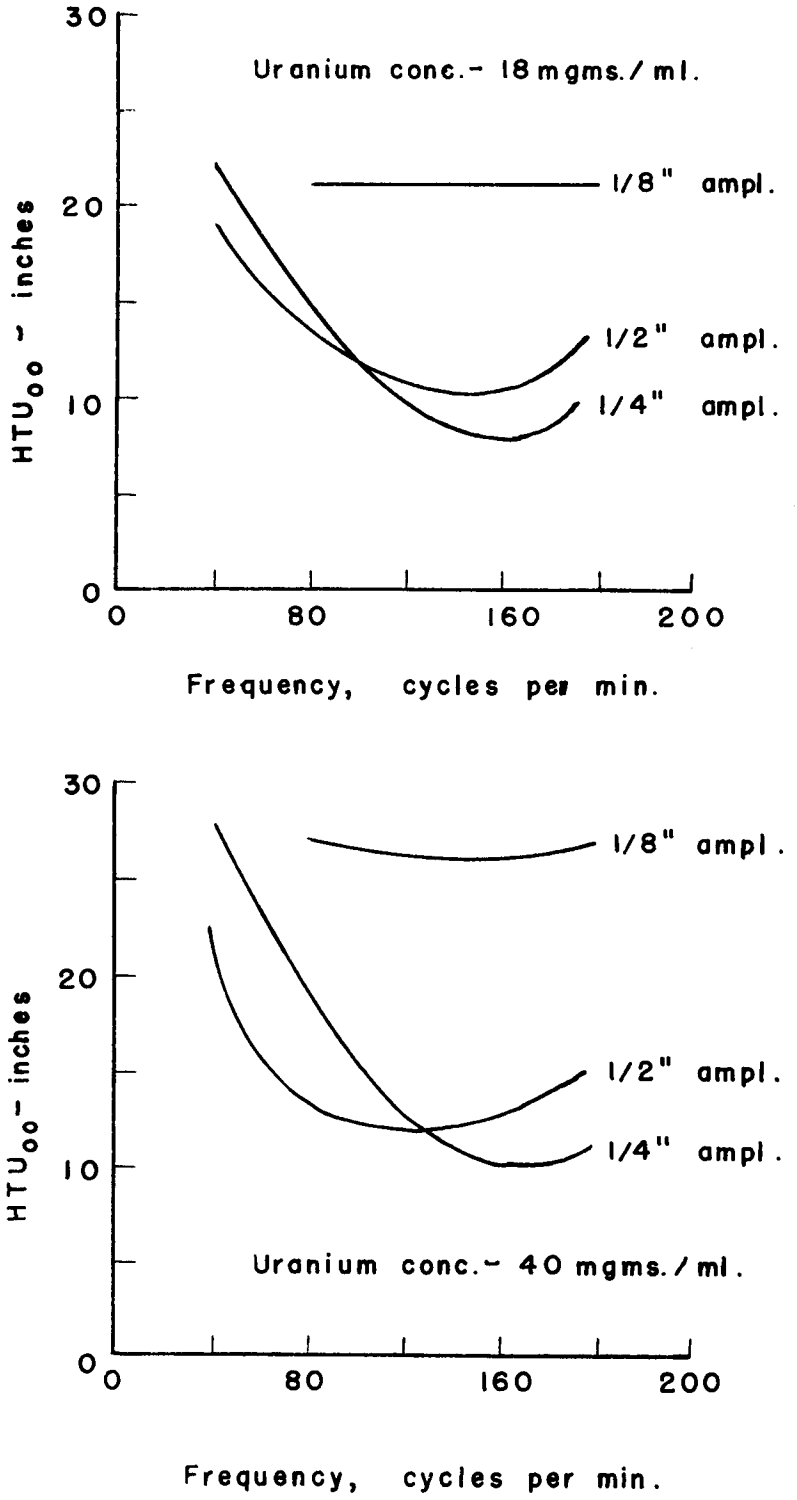

Figure 3 Variation of HTU_{oo} with frequency, amplitude, and uranium concentration in solvent phase.

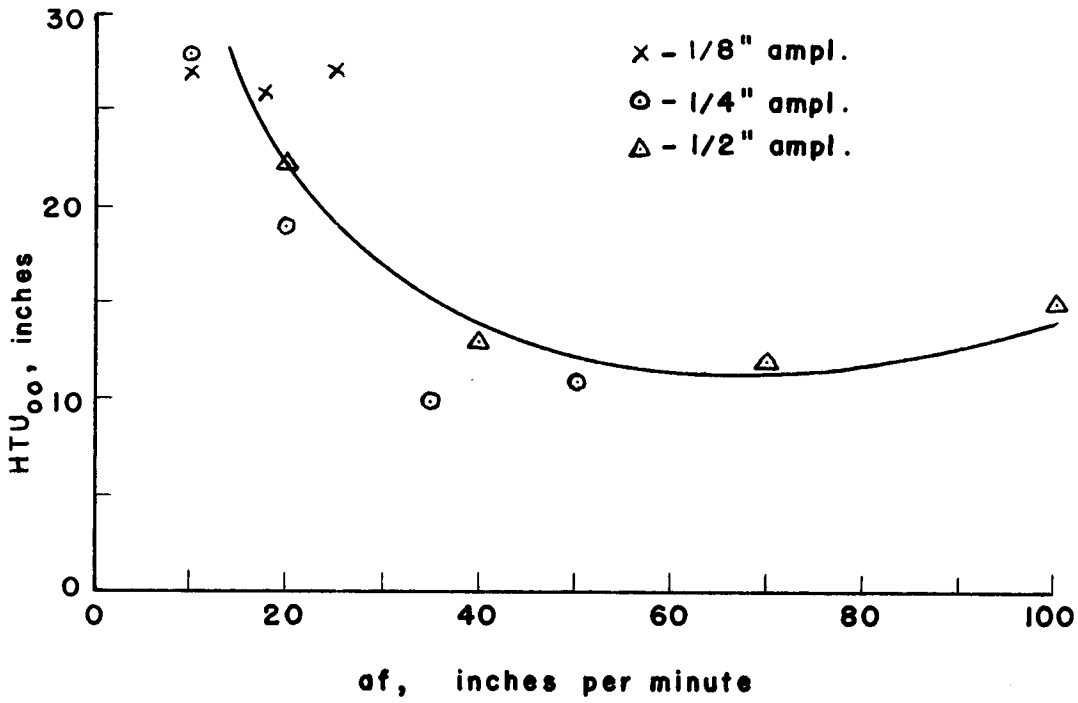

Figure 4 Variation of HTU_{oo} with product of amplitude x frequency

Another graph, Figure 4, was drawn showing the variation of HTU_{oo} with the product of amplitude times frequency.

2.4 Discussion

Referring to Figure 3, it will be seen that the minimum HTU is obtained when the pulse amplitude is between 1/4" and 1/2" and the pulse frequency between 120 and 160 cycles per minute. For design purposes, therefore, a pulse having an amplitude of 3/8" and a frequency of 140 cycles per minimum would give safe operating conditions and minimum HTU's.

The HTU's obtained under these conditions are about 20% of the HTU's obtained in an unpulsed, packed column operating with the same system.

Most of this improvement can probably be attributed to the smaller drop size obtained in the pulsed column. There might also be some increased efficiency from the greater deformation of the drops as they are drawn back and forth in contact with the packing.

It will be noted in Figure 4 that minimum HTU's are obtained when the product of amplitude times frequency is about 60 inches per minute. This value is approximately the same as the optimum amplitude-frequency product for operation of the pulse plate column with a TBP system (2). The pulse plate column is operated with an amplitude of 1" and a frequency of about 60 cycles per minute.

3.0 CAPACITY STUDIES

3.1 Theory

The purpose of this part of the study was to determine the variation in throughput of the column when pulses of different amplitudes and frequencies were applied.

The system studied was the countercurrent flow of water and 20% TBP in Turpolene. The reasons for this choice were that this system has the lowest flooding capacity of any of those encountered in the TBP Process and, secondly, the reagents were easily prepared and recycled as required.

Since the purpose of the study was only to compare the variations encountered in the throughput, the capacity was expressed as the sum of the throughputs of each phase at the flooding point. To allow for the normal variations in capacity encountered when studying unpulsed columns, the through-put of the unpulsed column was checked at least once a day, or oftener if any change was made in the conditions of the experiment. The capacity of the pulsed column was expressed as a percentage of that of the unpulsed column.

3.2 Apparatus

The column was built from 4" Pyrex glass packed for a length of 5 feet with ½" stainless steel Raschig rings. The pulsing mechanism and flow controllers were the same as described in the first part of this report.

Since the flow rates of each phase were expected to be high, arrangements were made to pump the solvent and water

back up to their respective head tanks after passing through the column. This decreased the amount of solution required for any one experiment.

3.3 Procedure

In each experiment, the pulsing mechanism was first adjusted to give the desired amplitude and frequency. The solvent and aqueous streams were then turned on and gradually increased until the column flooded. An effort was made to keep the flow rates of the two streams equal. Flooding was defined as that point when a separate solvent phase started to form below the level of the packing. When this happened, the aqueous inlet flow rate and the main aqueous-solvent interface above the packing were maintained constant. The solvent inlet flow rate was adjusted to maintain a constant amount of the separate solvent phase below the packing. The exit aqueous and solvent flow rates were then measured with the calibrated burettes. For purposes of this report, the capacity of the column with this combination of pulse amplitude and frequency was defined as the sum of these two flow rates. This capacity was then expressed as a percentage of the capacity of an unpulsed column. The results were plotted on graphs showing the variation of the relative capacity with frequency, the amplitude being a parameter.

3.4 Results and Discussion

In plotting the results, it soon became apparent that two general types of curves were obtained as shown in Figure 5.

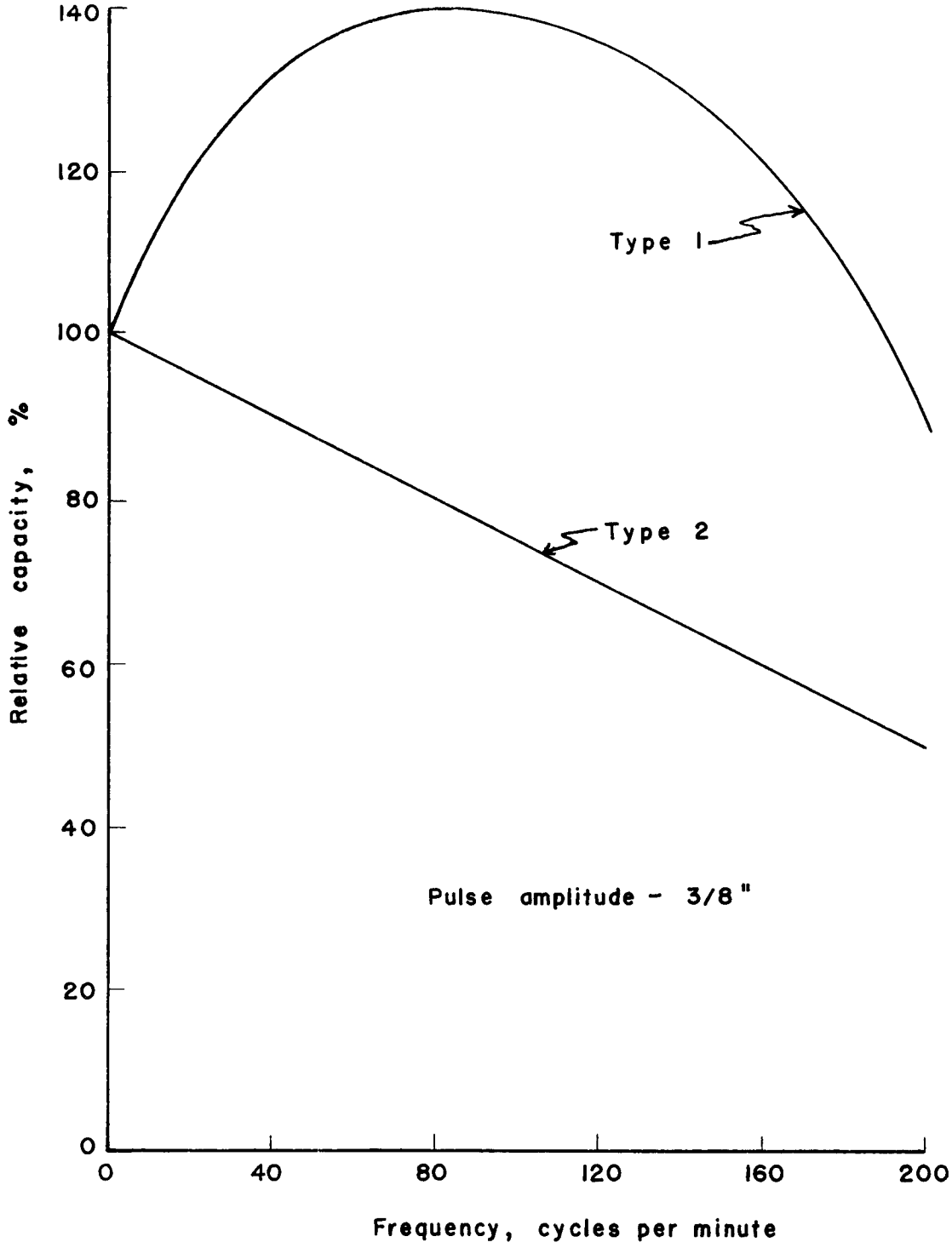

Figure 5 Variation of relative capacity of pulsed column with frequency

In the first type, the relative capacity increased as the frequency of the pulse was increased, reaching a maximum of about 140% when the frequency was 80 cycles per minute and decreasing steadily as the frequency was increased above this value. In the second type, the relative capacity decreased almost linearly with increasing frequency.

In trying to account for this behaviour, the following variables were studied:

1) The effect of using fresh, unwashed 20% TBP - Turpolene instead of solvent that had been washed and recycled many times.

2) The effect of dismantling the column and repacking it with the same packing.

3) The effect of replacing the packing that had been used for many experiments with new stainless steel Raschig rings that had been washed with carbon tetrachloride.

4) The effect of using tap water instead of distilled water.

These changes affected the absolute magnitude of the results to a slight extent, but the curves showing the variation of the relative capacity with frequency were all similar to that described above as type one.

With the following variables, however, curves having type two characteristics were obtained:

1) The use of unwashed stainless steel Raschig rings instead of washed rings;

2) The use of ceramic Raschig rings instead of stainless steel rings;

3) The use of water that was passed through the column only once instead of recycled water.

The increase in capacity of the pulsed packed column with certain combinations of amplitude and frequency can probably be attributed to two factors. First, the average drop size under pulsing conditions was very much smaller than obtained with no pulsing. It was also apparent that more of the cross-sectional area of the column was being actively employed than was normally found with a simple packed column.

The fact that the use of unwashed stainless steel rings or ceramic rings caused a different type of behaviour might be attributed to different wetting characteristics. The difference between the use of recycled water and water used only once is more difficult to explain. The most easily observed difference was that when the water was recycled many times, it became milky in colour as though some solvent had been emulsified into it. However, the connection between this fact and the observed difference in behaviour is not clear. It would probably require an extensive laboratory program to find the answer.

Since the laboratory personnel for this investigation were not available, and the pilot plant personnel were required for other work, this study on pulsed columns had to end at this point.

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E. E. Winter and S. H. Russell

2. HW 28900 - Pulse Column Variables. Solvent Extraction of Uranyl Nitrate with TBP in a 3-inch Diameter Pulse Column.
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