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THE DESIGN AND OPERATION OF PUREX PROCESS PULSE COLUMNS

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THE DESIGN AND OPERATION OF PUREX PROCESS PULSE COLUMNS

I. INTRODUCTION

The nuclear separations industry was one of the first to use solvent extraction for the purification of massive amounts of metallic elements. The use of this method for recovering and purifying uranium and plutonium from irradiated uranium was investigated in the early days of the Manhattan project; however it wasn't until the construction of the Redox Plant at Hanford in 1950, followed closely by the construction of the TBP-Plant in 1951, that solvent extraction achieved its first large scale application in nuclear separations processing. The Redox process, employing hexone as the solvent, was readily adapted to the use of conventional packed columns. Height requirements for packed columns in the TBP process, which used diluted tributyl phosphate to extract and recover uranium from stored Bismuth Phosphate process waste, proved to be excessive; however the use of pulse columns, first investigated by General Electric at Hanford in 1948(1), provided sufficient height reduction to permit installation of this process in an existing building. Development of pulse columns proceeded both at Hanford and other AEC sites and led to their installation in 1955 as the contactors for the Hanford Purex Plant. Much of the early Hanford Pilot-plant work was published by Stevenson and Bradley (2), Sege and Woodfield(3), and Geier(4,5). The intent of this paper is to bring this work up to date, to indicate qualitatively the performance attainable with improved cartridge designs, and to show some of the factors involved in scaling up pilot plant results to full plant scale.

The pulse column as used at Hanford consists of a vertical cylinder containing either fixed, spaced, horizontal perforated plates or conventional Raschig ring packing. The plates are assembled in cartridges which can be inserted or removed by either direct or remote methods. A piston or bellows connected to the bottom of the column superimposes a sinusoidal reciprocating motion to the counter-currently flowing liquids in the column. This motion provides intimate mixing of the two phases, the extent depending on the amount of energy supplied. A schematic sketch of the pulse column is shown in Figure 1.

II. THE PUREX PROCESS

The Hanford Purex Plant was originally scoped and built as a three-cycle plant employing a solvent-extraction cycle for co-decontamination of both uranium and plutonium, a partition cycle for the separation of the two, and a final decontamination cycle for each product.(11) The pulse columns were designed to produce separated uranium and plutonium products decontaminated by at least a factor of 10^7 from fission products, with average uranium and plutonium losses of less than 0.2 percent per column and an overall yield of at least 99 percent of each product.

Since startup, however, improved pulse column technology, increased process "know-how", improved solvent treatment methods, and the addition of tail-end cleanup facilities (ion exchange for plutonium and silica gel for uranium) which provide up to 10-fold decontamination for out-of-specification product have permitted the above design goals to be exceeded with only two cycles of solvent extraction. The following performance has been demonstrated with the present column design, using a flowsheet similar to that presented in Figure 2.(6)

Separation of U from Pu:	10^7
Separation of Pu from U;	10^6
Decontamination of Pu from fission products:	4×10^7
First cycle:	2×10^4
Second cycle:	2×10^3
Decontamination of U from fission products:	10^7
First cycle:	2×10^4
Second cycle:	5×10^2
Plutonium and Uranium Recovery	99.9%

In this flowsheet, the irradiated uranium metal containing plutonium and fission products is dissolved in nitric acid and fed to the midpoint of the dual-purpose 1A Column. In the lower section of this column the uranium and plutonium are extracted by 30 volume percent TBP in a refined kerosene diluent. The extract then passes countercurrently to a nitric acid scrub stream in the upper section of the 1A Column and through the 1S Column for removal of the essentially inextractable fission products. The extract enters the 1BX Column where the plutonium is reduced to the less extractable trivalent state and stripped into the aqueous phase. The solvent, still containing uranium, then enters the 1C Column where the uranium is stripped into a very dilute nitric acid solution. After steam stripping (to remove TBP) and concentration, the uranium is sent through the 2D and 2E Columns, which function like the 1A and 1C Columns, for additional fission product and plutonium removal.

The plutonium stream from the 1BX Column is processed through the 1BS Column to remove uranium by re-extraction and subsequently, after reoxidation to the tetravalent state, through the 2A-2B Column extraction cycle for further fission product decontamination.

The spent solvent from the 1C Column is reconditioned by a three-step contacting procedure which includes the IO Column. The solvent leaving the 2E Column is purified by washing it in the 20 Column with a sodium carbonate solution.

Several streams are recycled to improve process efficiency. For instance, the solvent leaving the 2B Column is added to the solvent feed entering the 1BX Column and the solvent leaving the 1BS Column is added just above the feed point of the 1A Column for recovery of plutonium. A similar backcycling procedure is used for the aqueous wastes from the 2A and 2D Columns. These wastes are combined, steam stripped, concentrated, and added to the 1A Column just below the feed point.

The chemical flowsheets of the extraction sections of the 1A and 2D Columns are similar enough to permit the columns to be designed alike. This is also true of the 1A and 2D Column scrub sections and the 1S Column, the 1C and 2E Columns, and the IO and 20 Columns. Thus the plant consists of 8 column types.

Salient features of the columns recommended for current Purex service are given in Table I(6). Maximum desirable operating capacities of these columns range from about 800 to 2000 gal./(hr)(sq ft), sum of flows. H.T.U.'s for uranium extraction range from one to two feet. H.T.U.'s for plutonium extraction are believed to be comparable, but those for fission product scrubbing are believed to be more in the range of two to four feet. Fission product decontamination factors are not believed to be limited by a lack of transfer units in the columns but to the presence of a very small fraction of the fission products complexed in the solvent in an unscrubbable form.

The primary development work for the Purex columns was carried out in three and four-inch diameter columns made of Pyrex glass pipe.(3) Further scale-up of some of the more promising cartridges were carried out in 8, 16, 24, and 27-inch-diameter columns. The knowledge gained in these studies plus actual large scale service led to the development of the above column cartridges.

The following items were some of the more important factors considered in choosing a specific cartridge:

1. For most of the columns it was considered of great importance to place the interface as far as possible from the product outlet to avoid entrainment of highly radioactive interfacial scums (i.e., at the bottom of the 1A, 1S, 2A, and 2D Columns and at the top in the 1BX, 1BS, 1C, 2B, and 2E Columns).

2. Those columns containing plutonium undiluted with uranium had to be made geometrically "safe" from a nuclear criticality standpoint (7-inch-diameter maximum).

3. For design simplicity, no more than two pulse generator sizes were desired; one for the large diameter columns and one for the smaller, critically-safe columns. Amplitudes between 0.5 and 1.2 inches are generally considered optimum; this restriction limits the ratio of the maximum-to-minimum column diameters to about 1.5 for a given pulser size.

4. Cartridge, and hence column, heights were sized to permit gravity cascade flow of the product streams from the 1A to the 1S Column, the 1BX to the 1BS and 1C Columns, the 2D to the 2E Column, and the 2A to the 2B Column. The maximum height available was fixed by the depth of the cells in which the columns are installed. This is approximately 40 feet.

5. In replacing an old cartridge in an existing column with a new, improved type of cartridge, it is essential that the new cartridge be compatible with the process with respect to efficiency, capacity, and general operability using the old column shell and pulse generator.

III. PULSE COLUMN BEHAVIOR

A. General Performance Characteristics

Five distinct types of behavior have been observed in pulse columns as a function of throughput rate and pulsing conditions. These are shown in Figure 3, a typical plot of the total volume velocity versus pulse energy input, i.e. the product of the amplitude and frequency.

The first zone is flooding due to insufficient pulse. With sieve plate cartridges, there is essentially no countercurrent flow without the pumping action of the pulser. The volumetric capacity of the pulse "pump" is termed the pulsed volume velocity and is equal to twice the amplitude-frequency product.

As the frequency is increased beyond this pulsed volume velocity threshold, a mixer-settler type of operation occurs, characterized by relatively coarse drops of the dispersed phase and pronounced settling of the two phases into discrete layers during the quiescent portions of the pulse cycle. Although very stable, this type of operation is rather inefficient unless very small plate perforations are used.

Emulsion-type operation occurs at still higher energy input. This highly efficient type of operation is characterized by small drop size and fairly uniform dispersion throughout the pulse cycle.

As the throughput rate or frequency is increased still further, complete flooding results. This is defined as the exit of one of the entering streams through the effluent line of the other phase. Frequently, but not always, an unstable but operable region of operation occurs just below the flooding threshold. This is usually characterized by periodic coalescence of the dispersed phase in a zone of high dispersed phase holdup and subsequent channeling of these large drops through the column. Generally the extraction efficiency is impaired in the unstable region and often fluctuates widely.

The performance characteristics of a pulsed packed column are similar to those above except that the pulse is not required to produce countercurrent flow and mixer-settler type operation does not properly describe the low pulse energy region.

The effects of increasing frequency and throughput rate on pulse column efficiency are shown in Figure 4. The H.T.U. is relatively high in the mixer-settler zone but decreases rapidly as the frequency passes through the transition zone between mixer-settler and emulsion operation. The H.T.U. is fairly constant throughout the emulsion region but may show a tendency to increase again as the frequency or throughput rate approaches the instability threshold. Optimum over-all H.T.U.'s (raffinate phase controlling) for the Purex system are on the order of one to two feet, with the lower value obtained at higher mass transfer rates. Nearly constant H.T.U.'s over a wide range of throughput rates may be obtained by appropriately adjusting the frequency.

Many of the design and operating variables that affect other types of extraction columns have similar effects on pulse columns. For example, the maximum capacity of the pulse column can be increased by increasing the density difference between the phases, increasing the interfacial tension and decreasing the continuous phase viscosity. Both the capacity and efficiency can be increased by increasing the temperature, presumably because of the favorable effect on viscosity and diffusivity. Neither the height nor the diameter of the column has a significant effect on the capacity of the column; however there may be a channeling tendency that can greatly increase the apparent H.T.U. with systems in which the density of the continuous phase is appreciably higher at the top. Channeling can be reduced and its effect minimized by adding redistributor devices, such as the louver plate which is described later, at three to four feet intervals.

B. <u>Types of Cartridges</u>

Several different types of perforated plates and packings have been investigated for Purex service. Those include sieve-plate cartridges made of stainless steel, plastic, and stainless steel with one surface coated with plastic; nozzle plate cartridges made of stainless steel with the perforations punched to form shallow nozzles; mixed plate cartridges containing both plastic and stainless steel plates; and Raschig rings made of plastic and stainless steel. Each cartridge type has its advantages, and all but the dual-faced plate cartridge are currently represented in the Purex Plant. Typical sieve plates used in pilot plant studies are shown in Figure 5.

1. Sieve Plate Cartridges

Simple sieve plate cartridges were perhaps the first type of pulse column cartridges tested and are still preferred by many investigators because of their simplicity and availability. The operating characteristics of such cartridges have been reported by Sege and Woodfield,(3) Cohen and Beyer(7), Wiegandt and vonBerg(8), and others.

Early in the studies at Hanford, a cartridge containing 2-inch-spaced stainless steel sieve plates with 1/8-inch holes and 23 percent free area was found to be a near optimum compromise between the desiderata of high capacity and efficiency. Such a cartridge has been called the standard cartridge. In the neighborhood of standard cartridge geometry, up to two-fold variations in capacity and H.T.U. are encountered as hole size, plate spacing, or free area is varied three to four fold.

Swift(9) has reported that the effect of the above variables on flooding can be correlated by the following equation:

$$\log \frac{(U_c + U_d)\, F^{0.34}}{d^{0.64}} = C_1 - C_2 \quad \frac{fad^{0.28}}{l^{0.32}\, F^{0.82}}$$

where a = pulse amplitude, in.

d = perforation hole diameter, in.

F = fraction free area

f = pulse frequency, cycles/min.

l = plate spacing, in.

U = superficial volume flow rate, gal./hr.-sq.ft.

C_1 and C_2 = constants dependent on the physical properties of the system.

The above equation is not dimensionless. It was derived empirically from studies made in an aqueous-continuous pulse column using stainless steel sieve plates. The liquid system contained no transferring component and hence was not typical of the Purex system in which gross mass transfer takes place in most of the columns.

It is now known that mass transfer across an interface can greatly affect the physical properties of the interface, particularly those properties which affect the ease of dispersion and coalescence. This effect is perhaps most pronounced in the extraction section of the 1A Column. At the top of this section where a high rate of uranium transfer exists and at the bottom where nitric acid transfer takes place, the pulse energy required to produce a good dispersion of the organic phase is appreciably greater than in the middle of the column where little mass transfer takes place. As a result, with a uniform cartridge this column is particularly susceptible to local flooding in the midsection at amplitude-frequency products of 80 to 90 percent of the complete flooding threshold.

The tendency for local flooding can be greatly reduced by grading the cartridge; that is, by providing greater spacing, hole size, and/or free area in the section where dispersion is most readily accomplished. As might be expected; this grading is still a trial-and-error situation; however it usually takes no more than three or four modifications of a pilot-cartridge to zero-in on one that produces a nearly uniform dispersion. Graded stainless-steel sieve plate cartridges were installed in the original aqueous-continuous Purex 1A and 2D Columns. These operated satisfactorily with respect to throughput rate and uranium and plutonium extraction for several years.

Wetting characteristics of the plates play an extremely important part in the design and operation of conventional sieve plate cartridges. For example, stainless steel plates are preferentially wet by the aqueous phase, and satisfactory operation can be achieved only with the aqueous-phase continuous. When the plates are wet by the dispersed phase, the droplets coalesce on the plates as a film which at low pulse energies leaves the plate reluctantly in streamers or as "balloons" enclosing continuous phase droplets. At higher pulse energies the column may "invert" with the nominally continuous phase becoming the dispersed phase. This, of course, is undesirable because of the existence of a second, uncontrolled interface.

The wetting characteristics of stainless steel plates do not necessarily remain constant. It has been observed, particularly in 1A Column scrub section and 1BX Column studies, that films can form on the plates which greatly increase the coalescing tendency of the dispersed organic phase. This effect is seldom, if ever, observed in columns that have high rates of mass transfer.

Plastic plates made of polyethylene or fluorothene are preferentially wet by the organic phase in the Purex system. Column behavior with plastic plates is just the reverse of that with stainless-steel sieve plates; the most stable, desirable type of operation being obtained with the organic phase continuous. Performance data for typical sieve plate cartridges are illustrated in Figure 6.

These data were obtained in 9-feet-tall, 3 and 8-inch-diameter columns using a 1C Column flowsheet, pulse amplitudes ranging from 0.5 to 1.0 inches, and interface positions as noted. All plates had 1/8-inch-diameter holes and were spaced two to four inches apart. (Optimum H.T.U. values for all cartridges illustrated in this and subsequent figures were in the range of 0.8 to 1.0 feet).

The dual faced plate cartridge, also represented in Figure 6, contains stainless steel plates with the side facing the organic phase inlet to the column coated with polyethylene or fluorothene. This cartridge can be used with either phase continuous. It provides higher capacity than either all-stainless or all-plastic cartridges with equivalent geometries at no sacrifice in efficiency. The plate side first "seen" by the dispersed phase serves to coalesce it, thus preventing buildup of tight emulsions; the other (exit) surface, which is not wet by the dispersed phase, prevents the coalesced disperse phase from clinging to the plate and allows it to be readily redispersed by the pulse. Unfortunately, the integrity of the plastic coating in radiation service cannot be guaranteed, and no plant applications using such plates have been made.

2. Nozzle Plate Cartridges

Stainless steel nozzle plates, a recent Hanford development for pulse columns, are a special class of sieve plates. They are characterized by the indentation of the holes to form many tiny jetting nozzles. A section of a typical nozzle plate cartridge is shown in Figure 7. Nozzle plates are usually oriented with the tips of the nozzles pointing toward the dispersed phase exit of the column; however satisfactory operation can often be obtained with the nozzles pointing either way. Well-designed nozzle plate cartridges have provided capacities 50 to 100 percent higher and H.T.U.'s up to 30 percent lower than equivalent geometry flat sieve plates. The performance of two typical nozzle-plate cartridges in the previously-described 1C Column application is shown in Figure 8. The 10 percent free area plates contained 1/8-inch holes; the 23 percent free area plates had 3/16-inch holes. Plates were spaced 2 to 4 inches apart.

A unique feature of nozzle plates is that they can be used in many applications with either phase continuous. This feature is especially important in high-intensity radiation service where plastic cannot be used. For example, the fission product decontamination factors in the Purex Plant 1A and 2D Columns were improved 10 to 100 fold by changing from aqueous to organic-continuous operation. This change, which diverts highly contaminated interfacial films and solids from the product to the waste end of the column, would have been impractical or even impossible without the development of nozzle plates.

Explanations of the unusual properties of nozzle plates are only conjectural at present. The ability to produce good dispersions even though wet by the dispersed phase is believed due to the inability of the coaleseced disperse-phase film to bridge the nozzle tip as it can a perforation in a flat plate. This theory is supported by the observation that the nozzle tip must extend at least 0.035 inches beyond the base before significant difference between nozzle and sieve plate cartridge operation is seen. Improved capacity over sieve plates is probably related to improved separation of the dispersed and continuous phases by the jet action of the pulse and to smoother flow characteristics of nozzles as contrasted to sharp-edged orifices.

3. Mixed-Plate Cartridges

The most recent cartridge development at Hanford has been the use of mixed cartridges containing alternate groupings of plastic sieve plates and stainless steel sieve plates or nozzle plates. The advantages of this type of cartridge are three-fold: first, the capacity of the column with an appropriate mixed plate cartridge is usually considerably higher than that obtained with either an all-plastic or an all-stainless cartridge; second, the mixed cartridge can be designed to operate with either phase continuous; and third, the efficiency is believed to equal or exceed that obtained with the older, more conventional cartridges.

The benefical effects of inserting plastic plates in a stainless steel cartridge can best be understood by considering again the behavior of the dispersed phase in an all stainless steel cartridge as the pulse frequency or throughput rate is increased. If the organic phase is dispersed, increasing either of these variables progressively increases the holdup of organic phase droplets until the stability limit is exceeded at about 50 to 70 percent holdup. There is usually very little tendency for the dispersed phase to coalesce on the plates during this operation; and the entire column, or at least that portion most subject to flooding, becomes filled with closely packed organic droplets. Now, if we insert plastic sieve plates (which are readily wetted by the organic phase) at about one foot intervals throughout the tightly packed section, we find that they act something like safety valves by coalescing the tight emulsion into large drops. These drops readily pass upward into the next stainless-steel section without greatly impeding the aqueous flow downward. This behavior not only reduces the organic holdup but also permits the column to operate stably at dispersed phase holdups (and hence frequencies or flowrates) that would normally be unstable. It provides in addition complete renewal of the interfacial surfaces at each plastic plate, a factor known to be important for attainment of high extraction efficiency.

The argument for operation with the organic phase continuous is somewhat similar. In this case, operating at high frequencies or flow rates without plastic plates causes phase inversion as mentioned previously. The organic phase actually becomes dispersed as a tightly packed emulsion in an aqueous matrix. Again, addition of plastic plates coalesces this organic emulsion and, in this case, redisperses the aqueous phase.

For application in organic continuous columns, the plastic plates are usually added in pairs alternating with two to four stainless-steel plates, all at one-inch spacing. This is done deliberately to induce development of alternate zones of aqueous and organic continuous operation. Because of the striped appearance of the cartridge and of the emulsion zones therein, this type of cartridge and operation are referred to as the zebra cartridge and zebra-type operation. A short section of a zebra cartridge is shown in Figure 9. The H.T.U.'s for chloride ion transfer in 1A Column scrub section studies using this cartridge were typically 10 to 30 percent lower than those obtained under similar operating conditions with an all-stainless-steel cartridge (aqueous-continuous).

One additional mixed-plate cartridge deserves special mention. This is the "sandwich" cartridge, so-named because the appearance of the inserts of four closely spaced plastic plates (1/16-inch apart) somewhat resembles sandwich filling. A typical sandwich cartridge used in 1C Column studies is shown in Figure 10, and its performance compared with that of standard and 23 percent free area nozzle plate cartridges in Figure 11. This type of cartridge is specifically designed to provide high capacity by completely coalescing the dispersed organic phase. This allows complete settle-out and separation of the phases in the vicinity of the plastic plates even at high pulse energy input. The efficiency of the sandwich cartridge also compares favorably with the other preferred types of cartridges, with typical uranium H.T.U.'s of 1.0

to 1.2 feet. Alternatively the closely-spaced plastic plates can be replaced by one solid sieve plate about one-inch thick.

Despite their superior operating characteristics, mixed-plate cartridges have been installed in only three columns in the Purex Plant - the 1S Column and the 1A and 2D Column scrub sections - because of the limited life of plastic under irradiation and because of the generally satisfactory operation of the other existing cartridges. These three columns use the zebra cartridge which permits high-efficiency operation with the organic phase continuous. Although nozzle plates can and may eventually be used in this application, it was felt that the efficiency of such cartridges would be insufficient to provide the fission product decontamination required.

Linear (Ziegler-process) polyethylene was chosen as the material for the 1A scrub and 1S cartridges because of its high resistance to radiation (stable to an integrated exposure greater than 2×10^9 R). Its life in the 1A Column, however, has only been about 8 months. This failure experience has not been quantitatively related to total exposure and probably occurs as a result of the synergistic effects of flexing and bearing stresses in the high radiation field.

4. Raschig Rings

Subsitution of Raschig rings or other typical tower packing for sieve plates is perfectly feasible and can result in a column with some of the advantages of both packed and pulsed columns. As would be expected, supplying pulse energy to the packed column increases the efficiency. In Purex service, H.T.U.'s have been decreased 1.5 to 3 fold by pulsing.

The capacity of the packed column may first increase slightly as the pulse is applied; however beyond the throughput rate corresponding to the pulsed volume velocity, the capacity falls off fairly rapidly. A typical flooding curve is shown in Figure 12. The data were obtained in an organic-continuous 3-inch-diameter column packed with 1-inch fluorothene Raschig rings. A dilute solution of uranium in nitric acid provided the diffusing component in these tests. For comparison, data obtained with 2-inch-spaced, 23 percent free area nozzle plates are included in Figure 12. At this time, the Purex 2A Column is the only application of a pulsed packed column at Hanford.

5. Louver Plates

It was mentioned previously that channeling may occur when the continuous phase is more dense at the top of the column than at the bottom. This was first observed in A-type extraction columns, aqueous continuous, when the diameter was increased from 16 to 20 inches. Presumably at or above this diameter range, the natural eddy currents that occur between sieve plates are insufficient to provide adequate mixing across the column cross section. Louver plates were developed at Hanford to correct this channeling tendency. These proved to be very effective; they are currently installed in the 1A, 1S, 1BX, 1BS, and 2D Columns. A typical Purex Plant plate is shown in Figure 13; a plate used in the 3-inch pilot plant column was included in Figure 5.

6. "Super" Pulse Column Cartridges

Several years ago a short program was pursued to see how efficient a pulse column could be made. Ultimately H.T.U.'s as low as one-inch were obtained in a 3-inch-diameter column containing stainless steel sieve plates with 0.026-inch holes, 15 percent free area, the plates spaced 0.25 inches apart. The pulse was applied at 0.2 inch amplitude at frequencies up to 300 cycles/minute. The capacity, of course, was quite low, but a 6-inch section of this column had nearly the same extraction efficiency as a 6-foot-tall 1A Column containing the standard cartridge.

At about the same time these studies were carried out, Koski at Hanford developed a miniature laboratory pulse column containing a similar cartridge.

Several such tiny columns (typically 1/4-inch-diameter and sixteen inches tall) have been used for laboratory studies of both the Uranium Recovery (TBP) and Purex processes, particularly fission product decontamination studies using actual plant feed streams. The extraction performance of these tiny columns came amazingly close to that of the plant columns, with H.T.U.'s of about one-inch obtained. The beneficial effects of zebra-type operation were first observed in one of these columns in which fine plastic turnings were inserted between alternate pairs of sieve plates.(10) A typical Koski miniature column is shown in Figure 14.

IV. PULSE COLUMN AUXILIARIES

A. Pulse Generator Design

Piston-type pulse generators chosen for the Purex Plant have proved to be highly reliable. The typical generator consists of a graphite-ringed piston reciprocated by a variable-speed electric motor acting through reduction gears and a conventional crank arrangement. All but one of the generators have a 3/32-inch diameter hole drilled through the piston to prevent formation of air pockets and to vent the pulse leg; the hole was not necessary in the remaining generator.

The presence of an air cushion in the pulse line can cause the pulse wave to deviate considerably from a near-sine wave shape and to either increase or decrease the pulse amplitude, the amount depending on the air volume and pulse frequency. The base of the piston is located 10 to 18 feet above the bottom of the column to reduce the volume of process solution leaking past and through the piston and to permit this leakage to flow by gravity to the proper receiver. During shutdowns, air pressure is supplied beneath the pistons to prevent drainage of the column through the piston bleed holes.

The pulse transmission systems were designed to avoid cavitation, which occurs in a liquid system when the pressure acting on it falls below the vapor pressure of the liquid. In a pulsed system, the total pressure is essentially the summation of three pressure effects: static pressure, pressure drop through the perforated plates, and acceleration pressure (the pressure resulting from the continuous change of momentum in the pulsed system).

The acceleration pressure is calculated from Newton's law: force equals mass times acceleration. In a sinusoidally pulsed system the acceleration pressure was given by

$$P_a = (-7.83 \times 10^{-6}) \frac{k \, L \rho f^2 \sin \theta}{D^2}$$

P_a = acceleration pressure, lb./sq. in.;

D = pipe diameter, in.;

k = pulse volume, cu.in.;

L = pipe length, ft.;

ρ = liquid specific gravity;

f = pulse frequency, cycles/min.;

θ = pulse-cycle phase angle.

To determine the acceleration pressure at any point in a pulsed system, the values of P_a for all sections of the system between the point and the top of the column are added. The greatest acceleration pressure occurs at the face of the pulse-generator piston.

The pressure drop through the column is essentially a function of the pulsed flow rate. For perforated-plate columns, this is given by

$$P_c = (\pm 1.285 \times 10^{-7}) \frac{N k^2 f^2 \rho (1-v^2) \cos^2 \theta}{C^2 A_h^2}$$

where P_c = pressure drop through the column, lb/sq. in.;

N = number of plates;

v = fraction of plate free area;

C = orifice coefficient;

A_h = plate free area, sq. in.

The pulse wave derived from an eccentric - crank arm combination deviates slightly from sinusoidal. For precise calculations of acceleration pressure and velocity pressure drop, it is necessary to use the exact equation of motion of the pulse piston. For the sine wave shape, the maximum acceleration pressure and the maximum velocity pressure occur 90 degrees out of phase; thus usually only that pressure effect which has the higher maximum need be considered. With the Purex-type pulsers, however, it is possible for the sum of the two pressures at some phase angle other than 0 or 90° to be greater than the individual maximum of either.

The maximum power load on the pulse generator is equal to the product of pressure and flow rate:

Power = $(k\pi f \cos \theta)(P_a + P_c + P_s)$, where P_s = static pressure.

The point of maximum power is always at a higher value of θ than the point of maximum pressure.

B. Pulse Column Instrumentation

The physical performance of the Purex pulse columns is indicated by instruments which measure the position of the interface, the specific gravity of the overflowing organic streams, and the differential pressure across a section of the column or across the entire column. In effect, all of these instuments provide a measure of the average specific gravity either at a point in the column or over a section of the column. In addition to detecting the position of the interface, the interface recorder also controls this position by sending an air signal to a diaphragm-operated valve located on the aqueous effluent line. A high interface reading, for example, will increase the valve opening until the interface has dropped to the desired level.

Three types of interface detectors have been utilized in large scale columns: air-purged dip tubes, capacitance probes, and floats. The purge-type instrument uses a flowing stream of air through open ended dip tubes to counterbalance the hydrostatic head of liquid. The back pressure in the dip tubes is measured by a manometer and/or a differential-pressure transmitter. Two dip tubes a measured distance apart are used for interface detection by indicating the average specific gravity across this height. The instrument is set to control the interface by maintaining a mean specific gravity midway between the specific gravities of the aqueous and organic phases at that point in the column. The specific gravity recorders use a similar detection system. Because the columns are pulsed a pressure variation is developed in the dip tube lines with changes in pulse frequency which affects the pressure reading and prevents satisfactory detection of the interface at the bottom of the organic-continuous columns.

Capacitance probes were used for several years to detect the interface in several of the columns - particularly those with bottom interfaces.(12) These detect the interface by the difference in the dielectric constants of the aqueous and organic phases. The probe consists of two electrodes; one a vertical metal cylinder, the other an insulated rod located at the cylinder axis with the liquid filling the annular space between the electrodes. The electrodes are connected by a low capacitance cable to a capacitance bridge which is unbalanced by an amount proportional to the position of the interface relative to the probe. Capacitance probes have been very satisfactory in pilot plant operation but were subject to rather frequent failures in plant service. They also "see" a tight emulsion as aqueous phase and may lose the true interface in trying to control a pseudo emulsion interface.

A float-type sensing mechanism, developed at Hanford in 1957(13), is now used in place of the capacitance probe. It consists of a float made of stainless steel, sized and calibrated to float at the interface. The float rides in a cage in a remote jumper head immersed in the region where the liquid interface is to be controlled. An armature is attached to the top of the float, and its relative position is detected by sealed transmitter induction coils, also located in the jumper. A change in position of the interface causes a change in impedance in a modified Wheatstone bridge circuit. Floats are installed in the IA, IS, 2D, 2A, 2B, IO, and 20 Columns. They have performed well to date and appear to be quite reliable when properly built and installed.

The differential pressure across the column, in effect the static head, is a measure both of the amount of uranium and of the relative amounts of the aqueous and organic phases in the column. At a constant uranium loading, variations in the static pressure are an excellent indication of instability or flooding. Differential pressures are measured individually across the extraction and scrub sections of the IA and 2D Columns because of the physical dissimilarity of the two sections. Air purged dip tubes are used to determine the differential pressure.

V. CONCLUSION

From the preceding discussions, it is evident that adaptation of pulse columns to a given solvent extraction application is still pretty much of a trial-and-error process. The Purex development studies, however, have indicated several short cuts that may be taken to provide a suitable column design with a minimum of pilot plant development. These may be briefly summarized as follows:

1. One of the following stainless steel nozzle plate cartridges should prove a suitable choice in a majority of applications:

   (a) 23 percent free area, 3/16-inch-holes, 2 to 4-inch plate spacing;

   (b) 10 percent free area, 1/8-inch-holes, 2 to 4-inch plate spacing.

   The 23 percent free area cartridge is most suitable for high capacity operation using an easily dispersed liquid-liquid system; the 10 percent free area cartridge is more suitable for high-efficiency operation with less easily dispersed systems. Both can be used for either top or bottom interface operation in most applications, though operation is generally more satisfactory with top interface operation.

2. For those few cases in which the nozzle plate cartridges are unsuitable, addition of one or more plastic sieve plates per foot of cartidge may prove beneficial. These plastic plates improve capacity and, by permitting operation at higher pulse energies may improve the efficiency as well.

3. Efficiency and capacity data obtained at a fixed amplitude are generally comparable to the values obtained at amplitudes within ±50 percent of this amplitude and may be correlated reasonably well by the use of the amplitude x frequency product.

4. Scale-up in column diameter is straighforward. Essentially, the volume velocity and efficiency are independent of the diameter, providing channeling tendencies are recognized and overcome with internal phase redistributors. Scale-up in column height requires some knowledge of how the H.T.U. varies with height or diffusing component concentration.

Perhaps the most severe test of the above rules occurred quite recently in the development of a solvent extraction process for the recovery of strontium-90 from fission product waste. This process uses a new solvent(di-2-ethylhexyl-phosphoric acid diluted with a kerosene-type diluent)and an aqueous feed differing considerably from the relatively simple Purex feed. Emulsification problems were expected with this sytem, but the first cartridge tried (the 23 percent free area nozzle plate cartridge) proved to be highly satisfactory, providing low losses and capacity nearly equivalent to that in some Purex process applications.

It is clear that the inherent flexibility, reliability, and efficiency of the pulse column will lead to its use for many other tasks; but maximum exploitation of pulse column without extensive pilot plant demonstration will require much greater knowledge of the effects of the physical and chemical properties of the liquids employed plus a knowledge of the dynamic effects that evidence themselves under mass transfer conditions. Such information should be the product of the extensive engineering research currently being carried out both in the United States and other countries.

VI. ACKNOWLEDGEMENT

The authors wish to acknowledge the work of the personnel of the Chemical Development Operation who engineered and operated the pilot plant described in this report and the contributions of members of the Chemical Processing Department who operate the Purex Plant. The work was performed under Atomic Energy Commission Contract AT-(45-1)-1350 with the General Electric Company.

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TABLE I

PUREX SOLVENT EXTRACTION COLUMN DATA

	Cartridge		Cartridge Details				Pulser		
Column	Height, Ft.	I.D., In.	Plate Type	Hole Diam., In.	Free Area, %	Plate Spacing, In.	Ampl., In.	Frequency-Range, Cycles/Min.	Continuous Phase
IA Extr.	13.9	24	Nozzle	3/16	23	2	1.1	35-110	Org.
Scrub	19.2	32	Zebra	(a)	23	1	0.6		
2D Extr.	14.0	24	Nozzle	3/16	23	2	1.1	35-110	Org.
Scrub	13.2	32	Zebra	(b)	23	1	0.6		
IS	18.6	34	Zebra	(a)	23	1	0.7	35-110	Org.
IC, 2E	18.0	34	Nozzle	3/16	23	2 (top) 4 (bottom)	1.06	35-110	Aq.
IBX	28.0	27	SS Sieve	3/16	33	4	0.84	35-110	Aq.
IBS	13.3	8	SS Sieve	1/8	23	2	0.84	25-110	Aq.
2A Extr.	20.9	7	1 In. Fluorothene Raschig Rings				1.1	25-110	Org.
Scrub	9.8								
2B	21.0	7	SS Sieve	1/8	23	2	1.1	25-110	Aq.
IO, 2O	26.3	34	Fluor. Sieve	3/16	23	4	0.53	35-110	Org.

(a) Cartridge composed of alternate groups of four stainless steel plates and two linear polyethylene plates. Hole diameters in stainless steel plates = 0.085 in.; polyethylene plates = 0.1875 in.

(b) Cartridge composed of alternate pairs of stainless steel plates and fluorothene plates. Hole diameters in stainless steel plates = 0.085 in.; fluorothene plates = 0.1875 in.

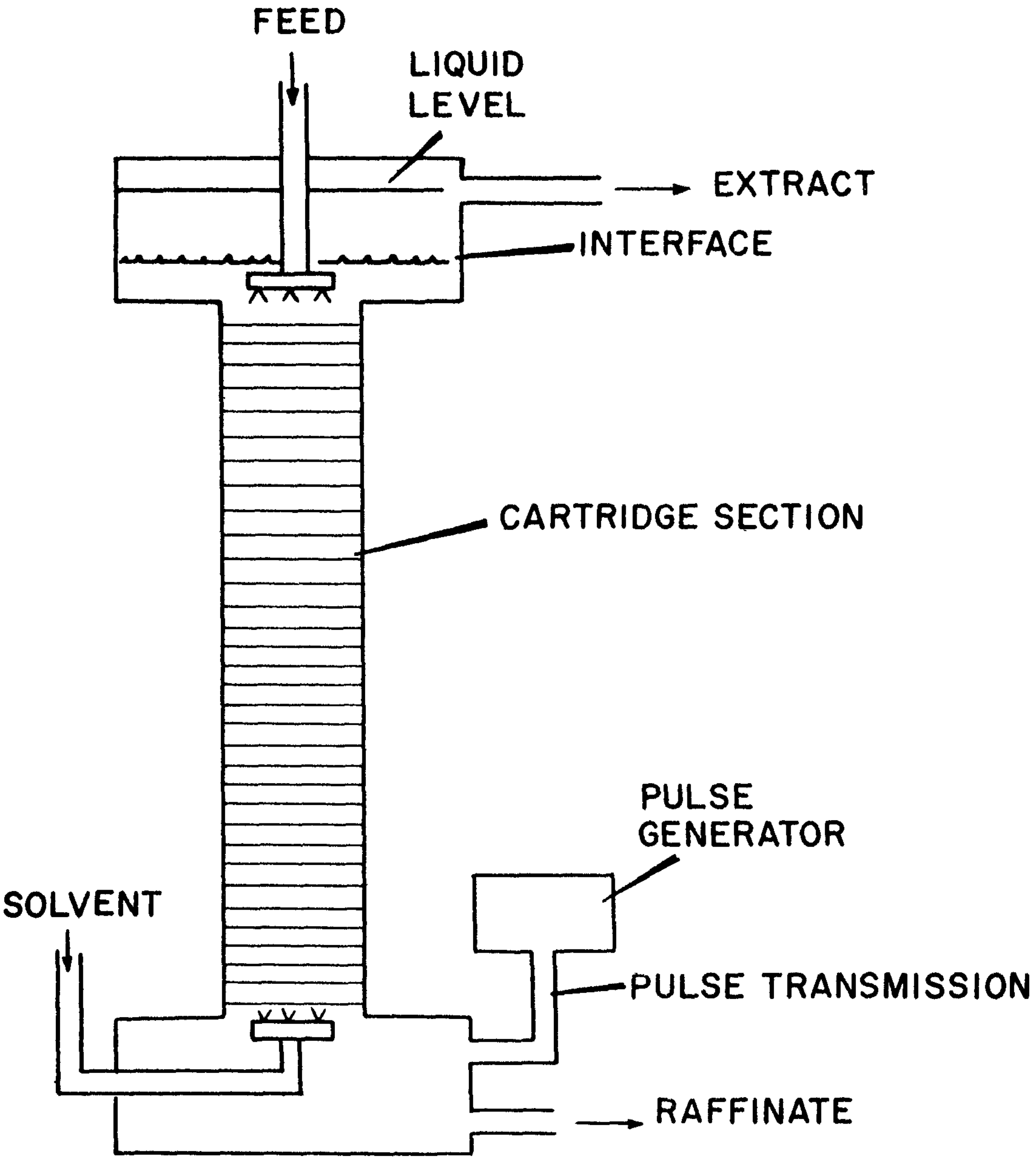


FIGURE 1
Schematic Sketch of a Pulse Column

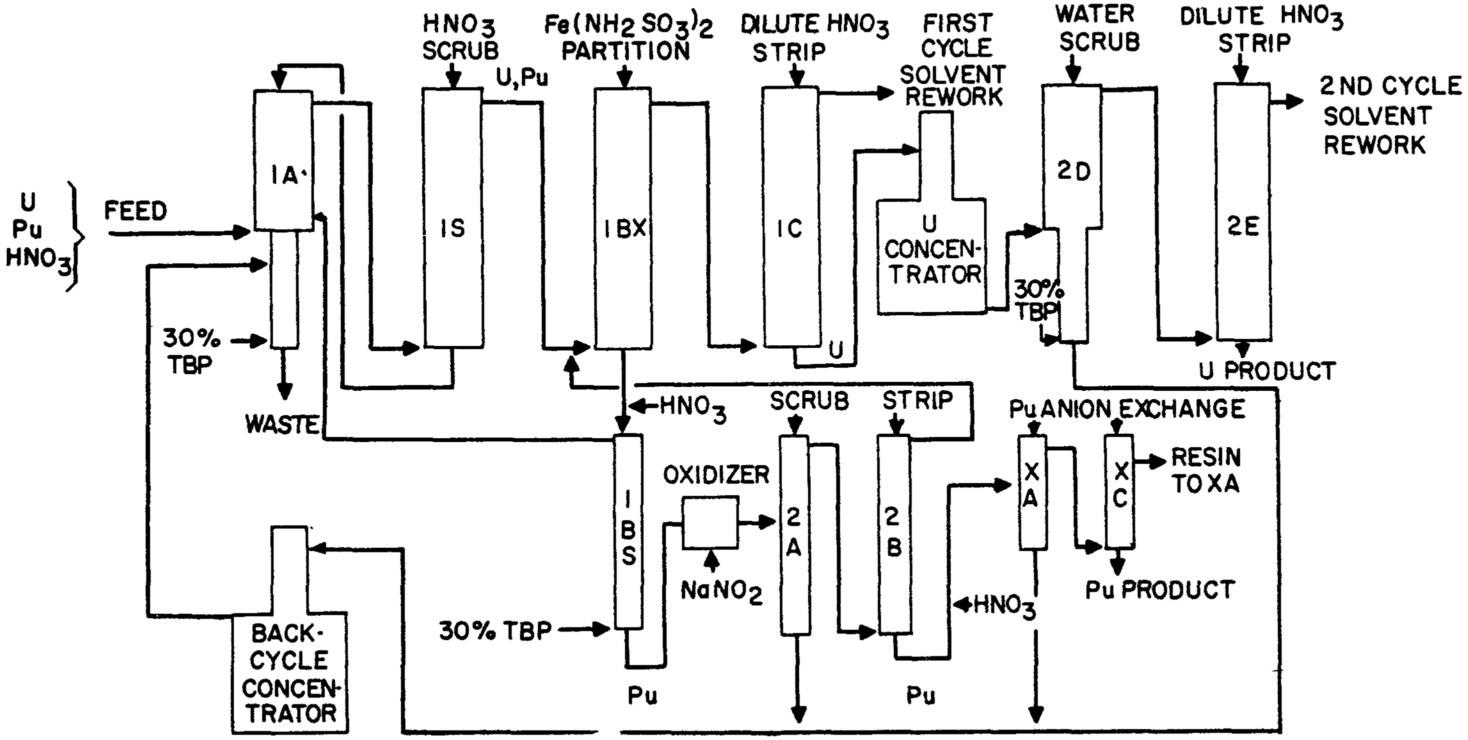


FIGURE 2
Simplified Purex Chemical Flowsheet

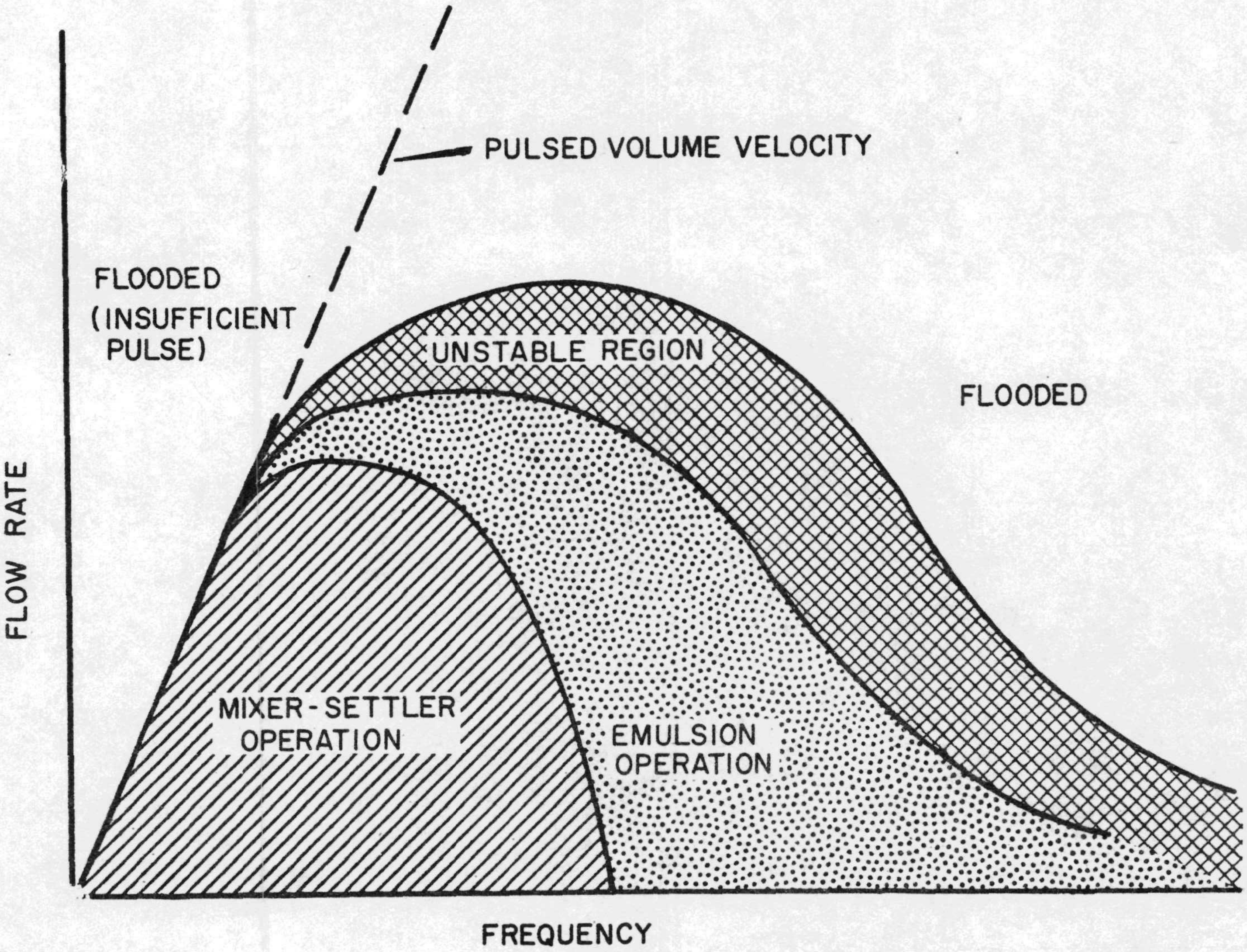


FIGURE 3
Five Types of Dispersion Behavior

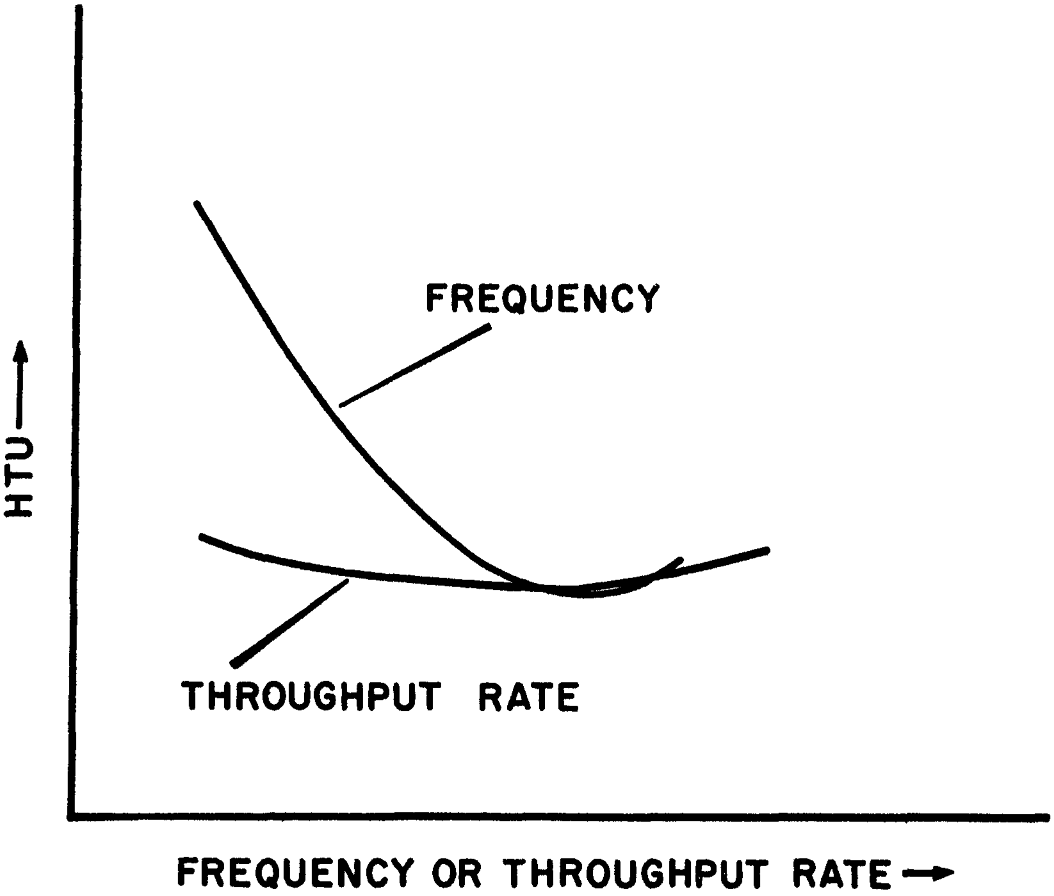
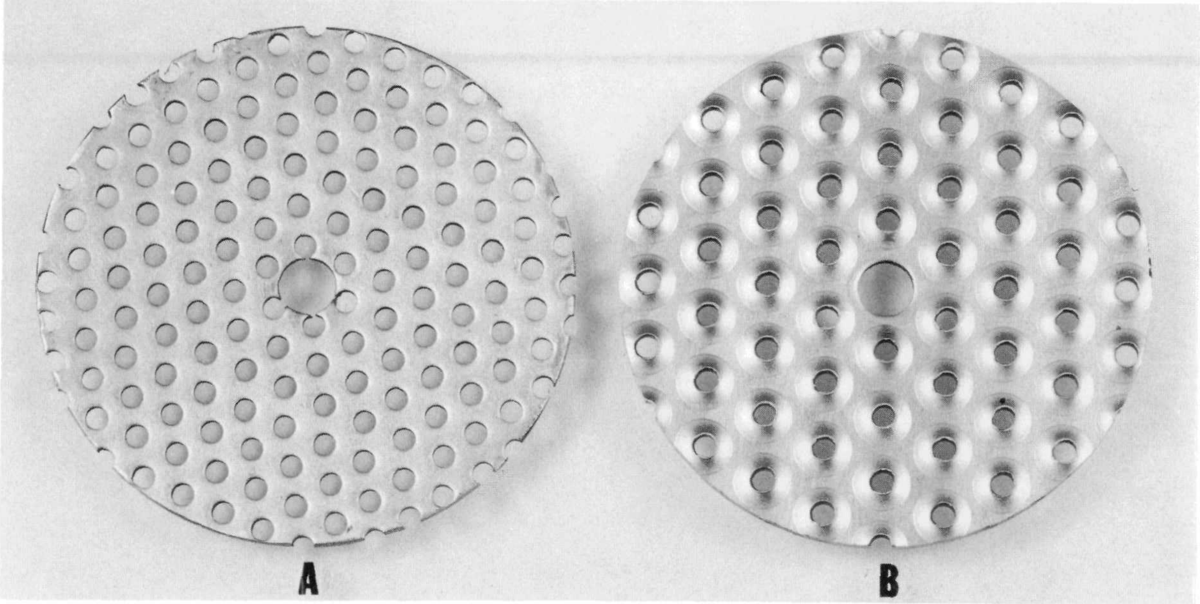


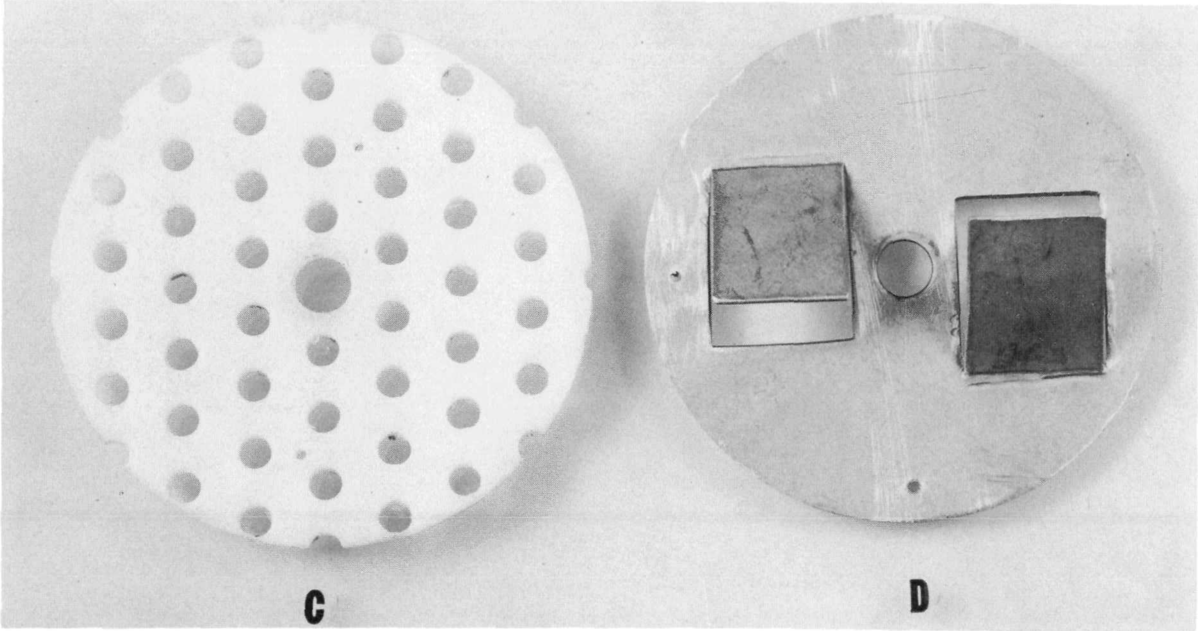
FIGURE 4

Effect of Pulse Frequency and Throughput Rate on Pulse Column Efficiency



"Standard" stainless steel sieve plate, 1/8-in. holes, 23 per cent free area.

"Stainless steel nozzle plate, 1/8-in. holes, 10 per cent free area 0.04-in. nozzle depth.



Linear polyethylene sieve plate, 3/16-in. holes, 23 per cent free area.

Stainless steel louver-type redistributor plate, 25 per cent free area.

FIGURE 5

Typical Sieve Plates Used in Purex Pulse Column Studies:

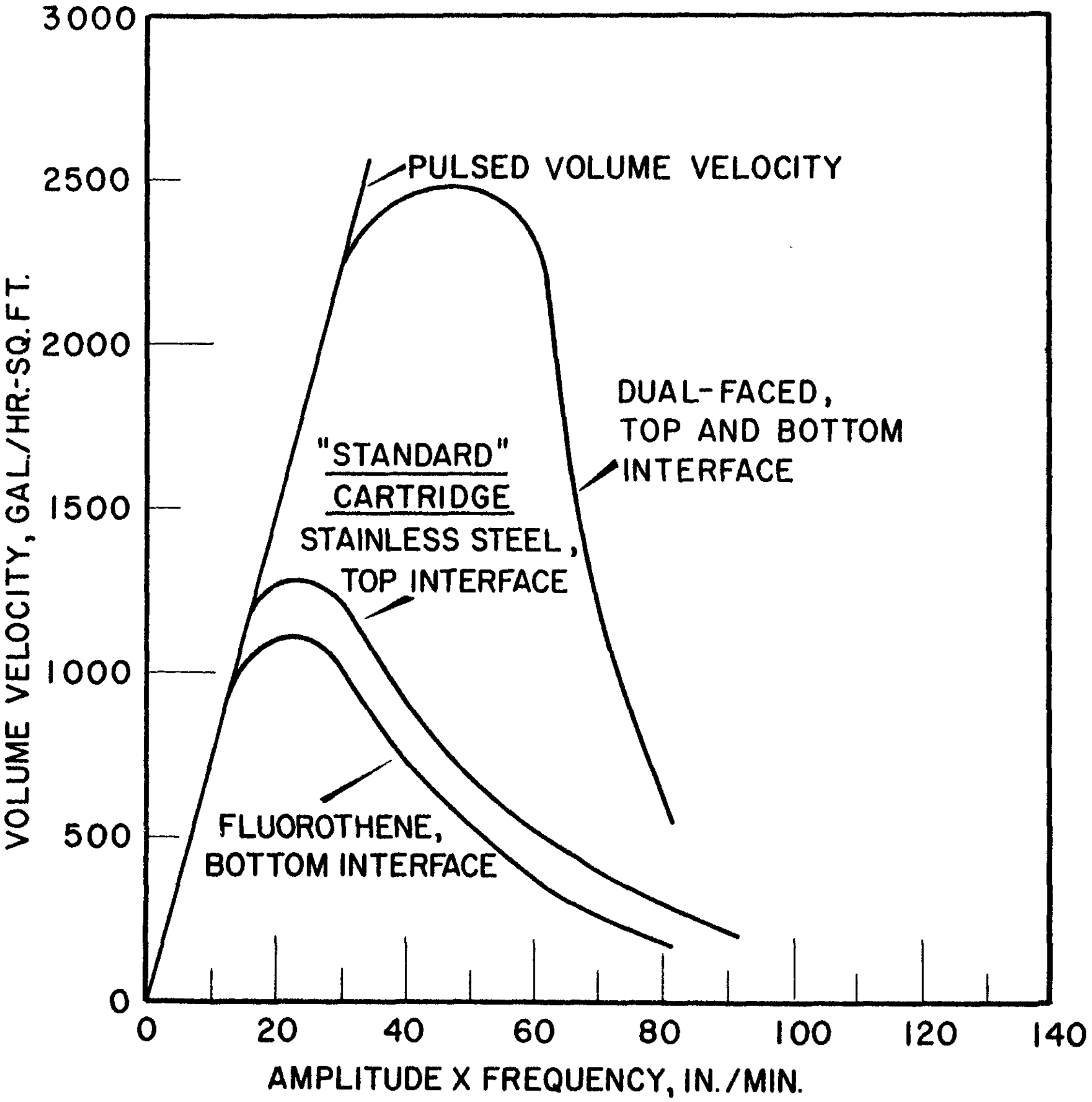


FIGURE 6

Performance Characteristics of Sieve Plate Cartridges
Using a 1C Column Flowsheet

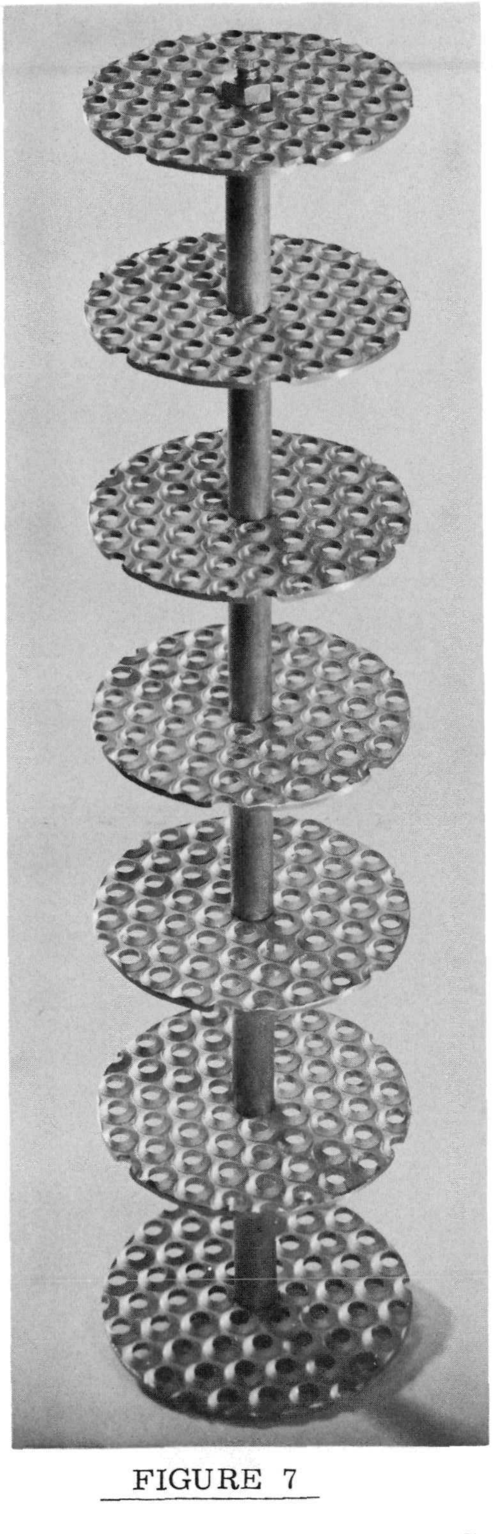


FIGURE 7

Nozzle-Plate Cartridge Used in the HA and 2D Column Extraction Sections (3/16-in. Holes, 23 Per Cent Free Area, 2-in spacing)

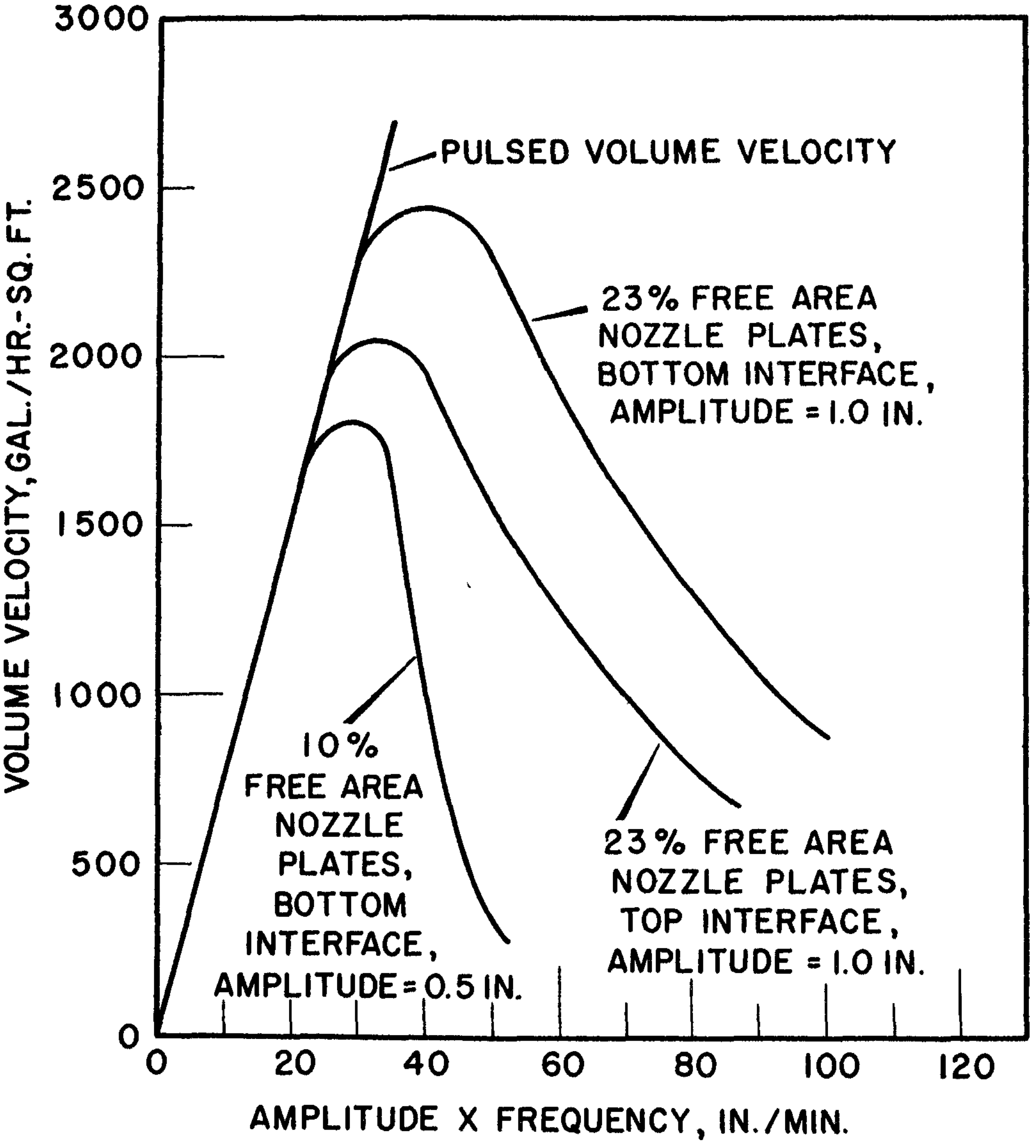


FIGURE 8

Performance Characteristics of Nozzle Plate Cartridges Using a 1C Column Flowsheet

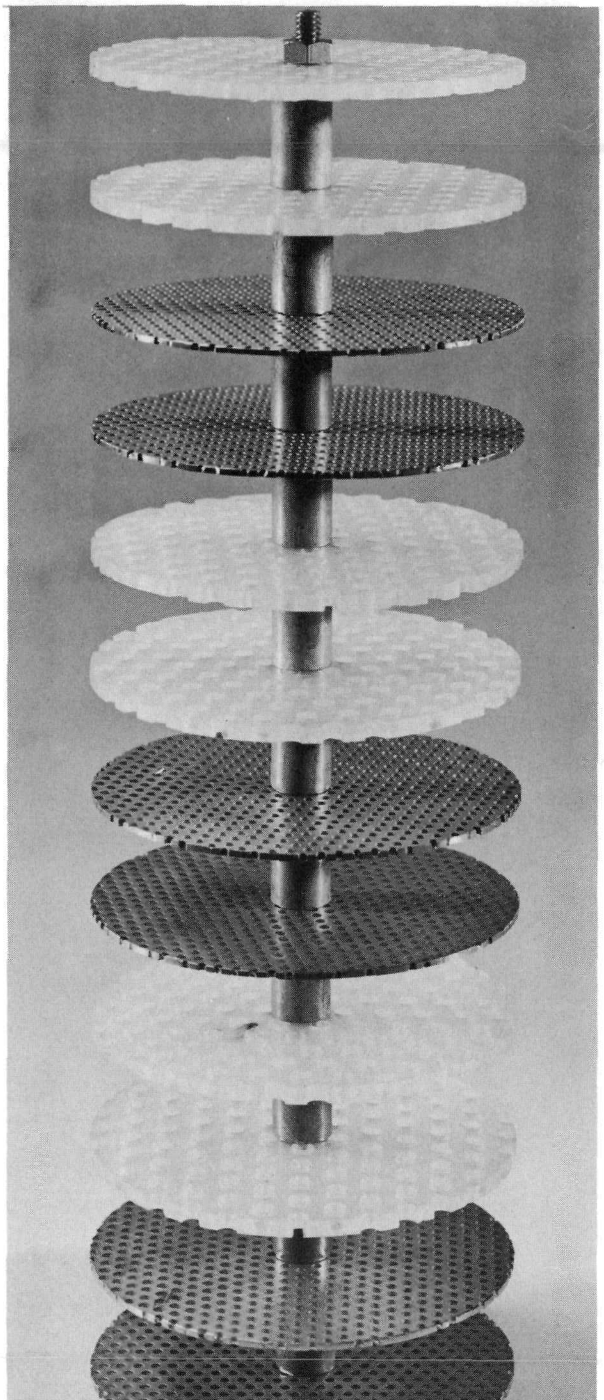


FIGURE 9

Zebra-Type Cartridge in the 2D Column Scrub Section (Stainless Steel Plates Contain 0.08-in. Holes, 21 percent free Area, the Fluorothene Plates Contain 3/16-in. Holes, 23 percent Free Area ; All Plates on One-Inch Spacing)

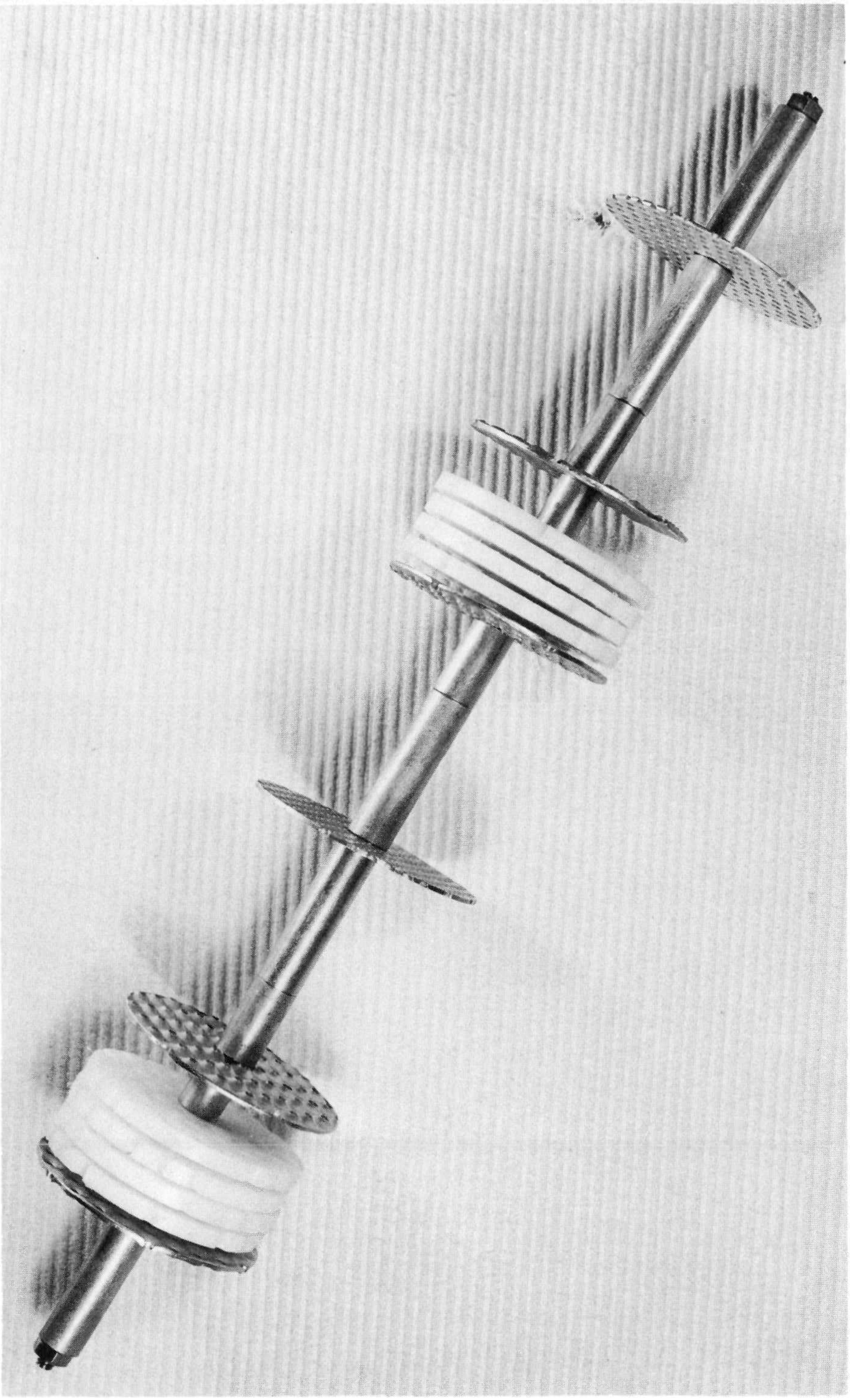


FIGURE 10

Sandwich Cartridge Used in 1C Column Studies

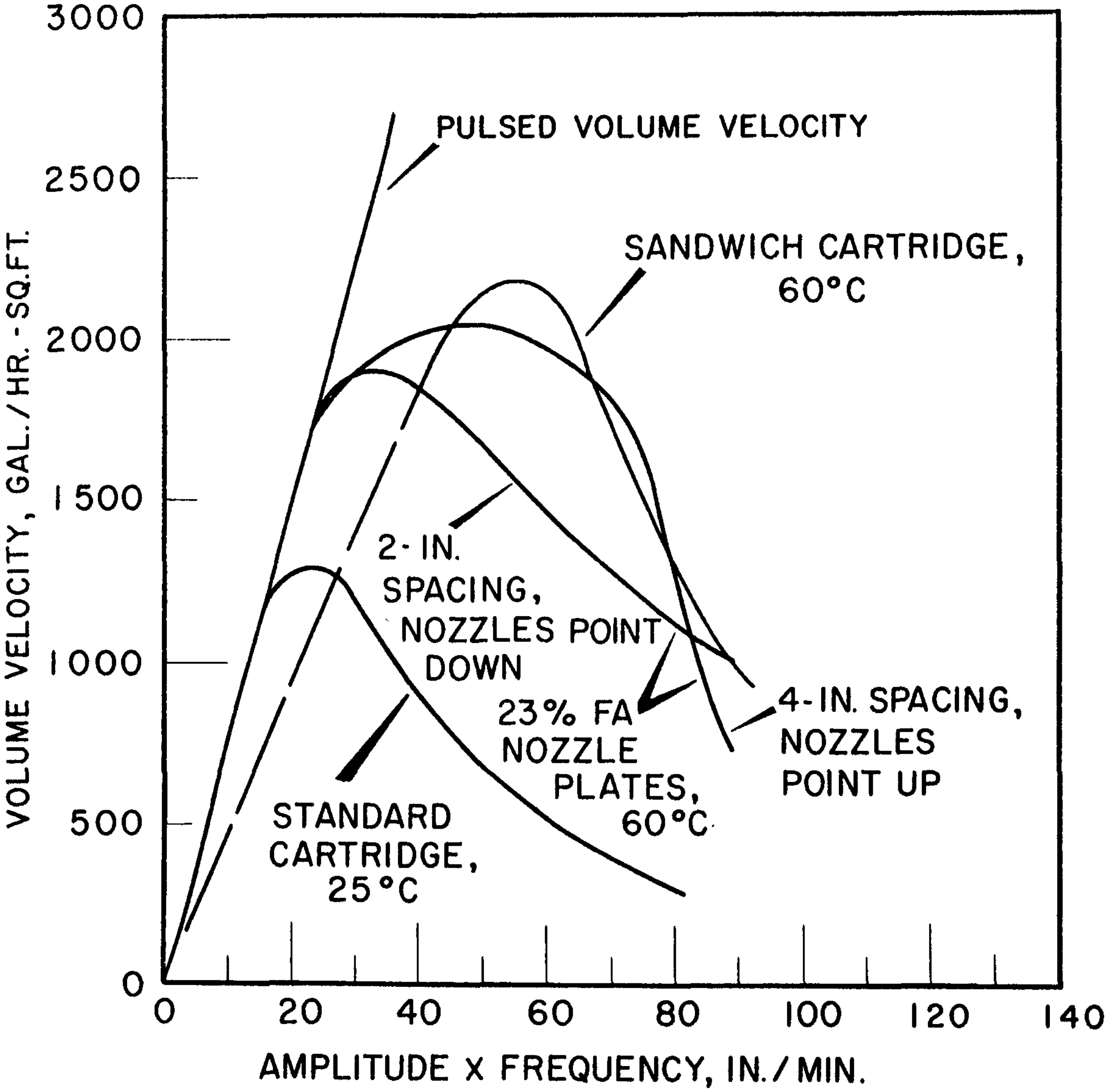


FIGURE 11

Comparison of the Sandwich Cartridge with Nozzle and Sieve Plate Cartridges - 1C Column Flowsheet, Aqueous Continuous

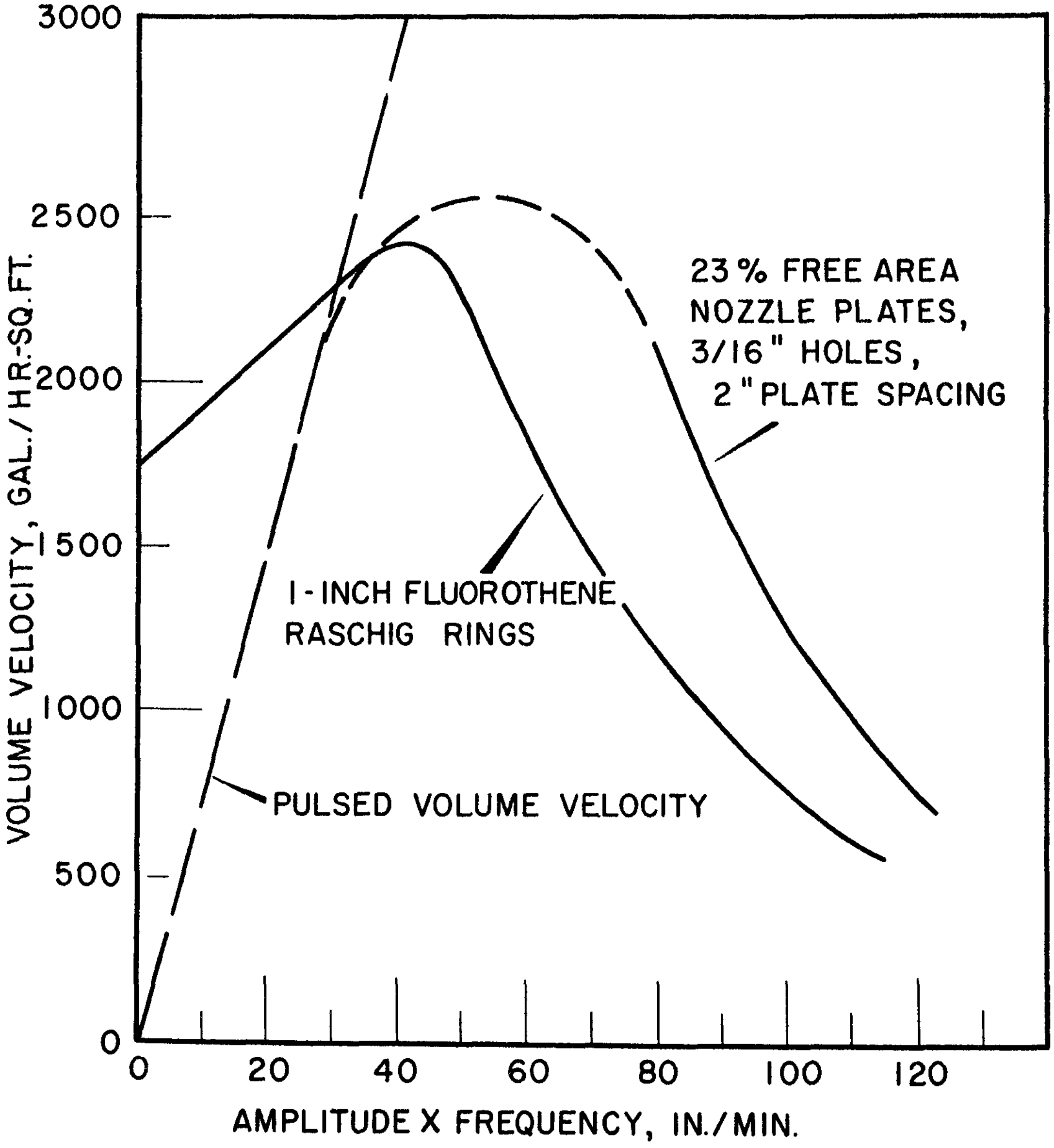


FIGURE 12

Comparison of Raschig-ring Packing with a Nozzle Plate Cartridge - 2A Column Flowsheet, Organic Continuous

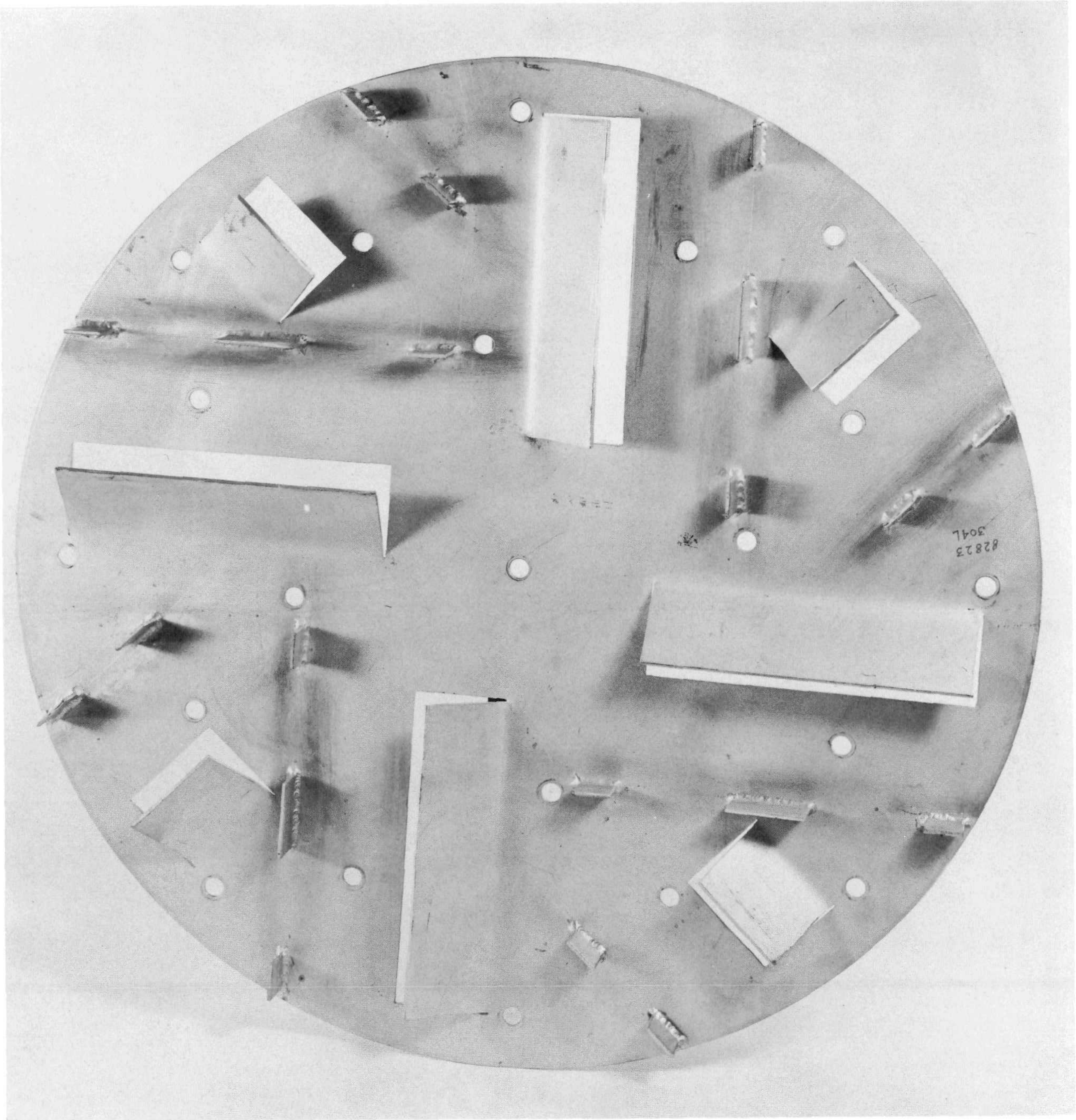


FIGURE 13
Typical Louver Plate Redistributor

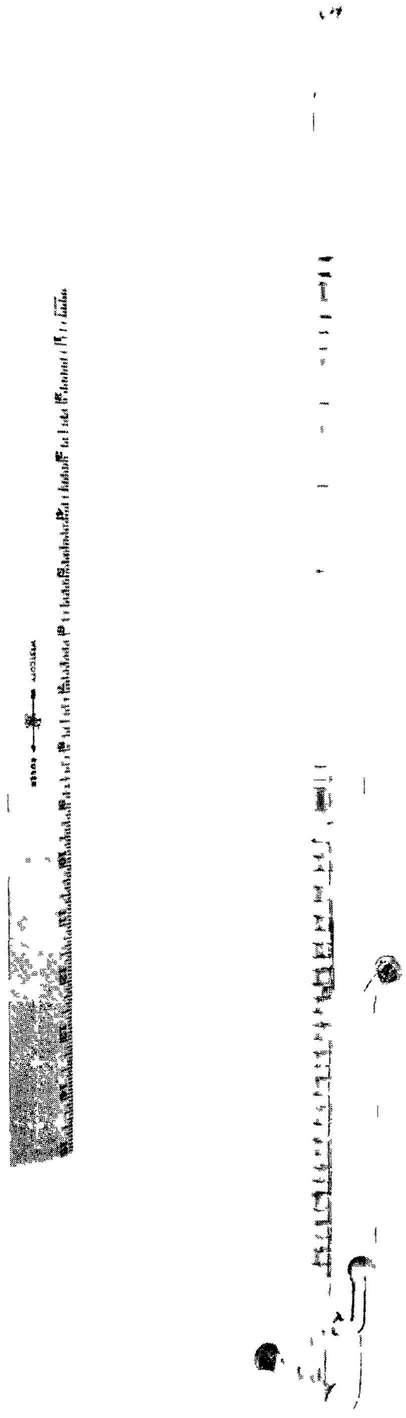


FIGURE 14
Koski Miniature Column