CHAPTER 4

PULSED PACKED COLUMN

4.1 General Considerations

Pulsing can be used with advantage to increase the mass transfer efficiency in packed columns because of the finer dispersions produced by pulsations.

Spaay et. al.(15) have recently published a complete set of data on pulsed packed columns. Their correlations for the characteristic velocity were applied at the flooding point to determine the flooding velocity. Design velocities equal to 65% of flooding, as recommended by Spaay, were utilized to calculate the column diameter. See table below; Design study references.

Data	Pulsed Packed Column
V_k, $\emptyset$	Spaay et al.(15), Widmer(16)
V_{cf} , V_{df}	-
d	Spaay , Widmer
k , HTU	Spaay
E_c	Spaay , Hennico et al.(17)
E_d	Spaay , Hennico

4.2 Flooding Velocities

In the operation of countercurrent extraction columns it is recognized that at a given flow rate of one of the phases, continuous increase of the other phase may finally lead to a condition at which the flow pattern of the lighter liquid becomes erratic. Eventually a portion of the lighter liquid may be swept out of the tower by the heavy liquid: the column is then said to be flooded. These critical flow rates which are independent are called the "limiting flow rates" or "flooding velocities". The material of the packing has been found to affect the flooding rates.

Most extraction column processes operate with a solvent of lower density than the aqueous phase and discrete solvent drops are usually allowed to ascend in thecontinuous aqueous medium, with an interface near the top of the column. Aqueous phase enters just below the interface and solvent phase is fed via the first distributor at the base of the columns.(12)

Sherwood and Pigford(18) concluded that for the above system flooding of the column takes place at certain critical flow rates. This phenomenon arises when the flow rate of the dispersed phase is slowly increased with a constant flow of continuous phase. The additional column hold-up of dispersed phase thus leaves less space for continuous phase and therefore the linear velocity of the of the continuous phase is increased. A tendency to drag the dispersed phase droplets in the direction of the continuous phase thus arises. When the flooding point is reached, the dispersed phase is discharged along with the continuous phase and countercurrent flow ceases. For a given diameter of column, flooding can take place as a result of excessive flow rate by either phase, and particularly by both phases simultaneously. The flooding rate is dependent upon the chemical nature of the system and the difference of density between the twophases, in addition to the mechanical characteristics of voidage and nature of packing. It is usual to operate at about 70% of the flooding rate to allow a reasonable margin for variations of conditions within the system. Temperature, composition of phases, and the presence of finely divided solids in either phase, affect the flooding rate.

Golovko, and Zadorskii, (19)studied flooding rates using a 15x15x2 mm. raschig ring packed, pulsed extraction column (100 x 1650 mm.) for the system carbon tetrachloride - water as a function of pulsation rate, amplitude, and dispersed phase flow rate. Correlations and interactions can be expressed in a regression equation. The flooding curves are characterized by 3 regions.

1. In the boundary - layer - controlling region, the capacity increase with increasing pulsation.

2. In the region characterized by decreasing drop diameter, the capacity decreases slowly with increasing pulsation.

3. In the third region, capacity drops sharply

as an emulsion of small droplets is formed.

Jackson, et al.(3) used a 1/2-in. polyethylene raschig ring packing, a 1/2-in. ceramic raschig ring packing and a 3/8-in. ceramic raschig ring packing in a pulsed packed column for the system water-toluene with varying pulsation rates, amplitudes, and dispersed phase flow rates. Approximate flooding limits were observed and are presented in Fig 1. Flooding limits were observed to decrease linearly with increasing pulse velocity (af). The dotted lines in Fig.1 correspond to ceramic rings of two sizes. The values at zero pulsing velocity were taken from Ballard and Piret(20) and appear to be consistent with the flooding characteristics observed for pulsed packed columns. The use of polyethylene packings leads to lower flooding limits than for the ceramic types. Columns packed with rings so that the ratio of diameter to packing size was 4:1 gave higher flooding limits and better performance than a ratio of 8:1.

In order to understand this surface effect for better designing and enhance the knowledge in this field, systems with different packing materials should be tested. The present work was carried out to partly fill this need. The packing materials used in this present work are 1/2-in. stainless steel raschig rings and 1/2-in. plastic raschig rings.

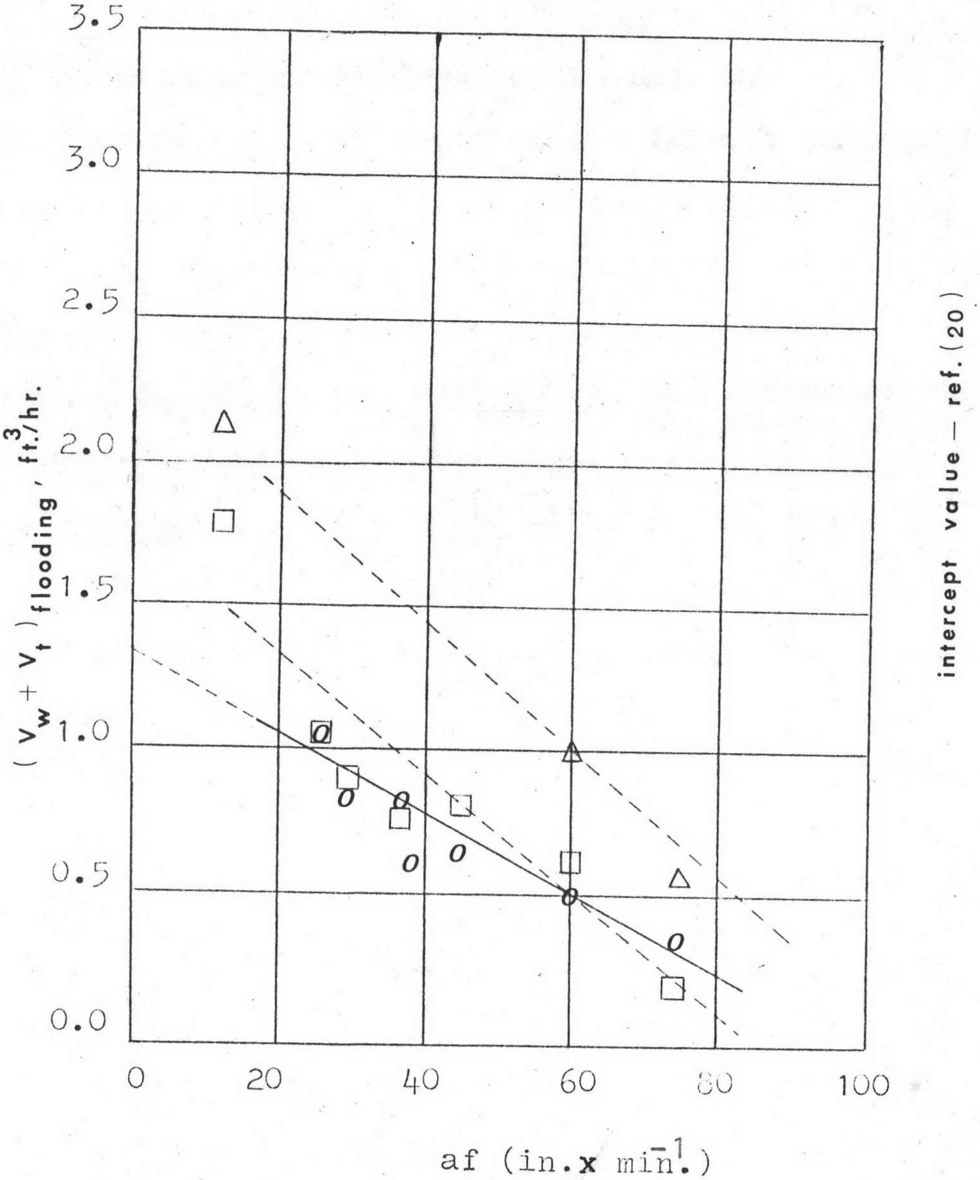

Figure 1 Flooding rates of pulsed packed columns.(5)

o 1/2-in. Polyethylene rings

△ 1/2-in. Ceramic rings

□ 3/8-in. Ceramic rings

4.3 Efficiency of Extraction

Knowledge of mass transfer rates and flooding limits are basic for the use of packed column for liquid-liquid extraction. The input of pulsing energy increases the mass transfer efficiency because of the finer dispersions produced by pulsation and promotes counter flow of the two phases.

The comparison of the overall efficiency of the influence of the wetting characteristics in pulsed packed column has been shown graphically in the Jackson work(3) using the difference of the overall height of transfer unit.

Increasing the mass transfer rate permits an increase in the overall efficiency and a reduction in the overall height of the column required and in capital investment required.

The important factors affecting the overall efficiency are as follows:

1. Dispersed phase hold-up : Wiegant and Von Berg (21) were among the early investigators to observe the hold-up characteristics under pulsing conditions. At low dispersed phase rates pulsations improve the efficiency 10 to 15 fold and the hold-up rises from 5 to 50% or more depending on the pulsing characteristics.

2. Mass transfer rates : Feick and Anderson(9) have reported that the overall mass transfer coefficient was observed to be enhanced by pulsing. Mass transfer data in terms of overall height of transfer unit (HTU) based on dispersed solvent phase have been reported by Potnis et al.(22) for the extraction of acetic acid from an aqueous solution. The extraction efficiencies first showed an increase with pulse velocity, and then decreased after reaching a maximum. Similar results were reported by Wiegant and Von Berg.(21) For each pulse frequency there would be an optimal amplitude giving maximum efficiency. Wiegant and Von Berg noticed that the performance of their pulsed column was practically independent of the continuous phase flow rate but was affected to some extent by the flow rate of dispersed phase. Chantry et al.(4) have indicated that the use of a higher frequency and lower amplitude would result in lower HTU and less consumption of power. They also indicated that higher frequencies tended to create finer dispersions and lower amplitudes tended to lower axial dispersions.

3. Longitudinal mixing : The available mass transfer data reported in terms of overall HTU are based on the assumption of plug flow for both phases. Moon and coworkers(23,6) have reported the axial mixing (longitudinal mixing) in the continuous and disperse phases. It is observed that the extent of axial mixing in the continuous phase remains the same as for unpulsed packed column at

low pulse intensities. But as the pulse intensity is increased (especially with increase in amplitude), greater axial mixing could be observed. It is also shown that the dispersed phase Peclet number, $V_d d_p/E_d$, to account for axial mixing, may be assumed constant having a value of unity. Vermeulen et al.(6) have indicated graphical correlations for the dispersed phase and continuous phase mixing coefficients. It is observed that the wetting characteristic of the packing alters the dispersed phase Peclet number or axial mixing.

4. Surface effects : Several experimental studies have been performed on the above factors by using different systems. Only the studies of surface property effects will be discussed in this investigation with the system carbon tetrachloride-iodine-water, with stainless steel raschig-type rings and plastic raschig-type rings.

4.4 Surface Phenomena

If the continuous phase wets the packing, the dispersed phase move through the column as small droplets only; if the dispersed phase wets the packing, it will also move down the column as a film on the packing surface as well as droplets. In the latter case hold-up of dispersed phase will increase, and flooding will probably tend to occur at low flow rates. However interfacial surface may increase in the packed section, and higher transfer rate may result. Consequently, the overall efficiency of the column may increase and permit a reduction in the overall height of a transfer unit.

The transfer mechanism in the dispersed phase may also be modified by the nature of the solid surface. Circulation in droplets is an important factor in the rate observed which are discussed by Skelland and Garner.(24) If the dispersed phase wets the packing, circulation may be increased through tendency of the droplets to rise or fall which ever the case while in contact with the solid on one side.

Danckwerts(25) proposed the surface renewal theory, an extension of the penetration theory of Higbie, as being a betterdescription of the mechanism of mass transfer for the flow of liquids over packing. The mechanism for this case was postulated as being one of unsteady state diffusion to elements of liquid constantly being renewed at the interface. It is proposed specially for the case of gas absorption but should have equal significance for liquid-liquid systems in packed columns. However no in depth studies have ever been performed for columns using this theory.

Garner(26) cites a quantitative method for the determination of the wetting characteristics of various solid surfaces. Contact angles of drop of either toluene or water, in the presence of the other phase, were measured. These angles corresponded to cases where the water was receding and advancing. The difference in the cosines of the two angles is the measure of the hydrophilic or hydrophobic nature of the surface and some valuesof interest are summarized in the table below :

Table 4.1 Hydrophobic Charactersof Packing and Plate Materials

Materials	Relative Values
Glass	-1.66
Porcelain, glazed	-0.88
Mild steel, rusted	-0.77
Porcelain, unglazed	0.85
Mild steel, clean	1.31
Polyethylene	1.85
Teflon	1.92

The negative and positive values have relative significance, and it is seen that polyethylene is more hydrophobic than unglazed porcelain.

These data were used by Garner et al.(27) to show that the performance of a plate extraction column (not pulsed) was influenced significantly by the wetting characteristics of the plate materials. Buchanan(28) found that the rates of formation and sizes of drops were influenced by the wetting characteristics of orifice plates.

Sobotik and Himmelblau(29) studied the effect of plate wetting characteristics in a pulsed sieve-plate column. The use of stainless steel or polyethylene plates gave about the same rates when the acetic acid was transferred from the water phase to the continuous ketone phase. However substantially higher rates were observed for acid transferred from the continuous ketone phase to water for polyethylene plates. An alternative plate arrangement gave results intermediate between the performance of all polyethylene or all stainless steel plate columns.

Ballard and Piret(20) observed the flow mechanism in a liquid-liquid packed column (not pulsed) and found the flow to depend upon whether the dispersed or the continuous phase preferentially wets the packings. Also flooding limits were correlated for unglazed porcelain ring packing.

Jackson et al.(3) reported the observation of the effect of surface wetting characteristics on performance and compared a packed column with a plate column for the same system and operating conditions. The water-benzoic acid-toluene system was used. The column consisted of a 2-ft. section of 2-in. standard glass pipe. Pulsing was provided by a unique arrangement with a bellows (a 2-in. expansion joint) and a reciprocating air motor. Plates for the column were designed for optimum performance as suggested by Sege and Woodfield.(10) The plates were arranged with a 2-in. spacing with 1/8-in. holes and 23% free area (fifty-two holes on equilateral triangles). The plates were 1/32-in. thick , and the plate cartridge for the 2-ft. section of column contained twelve plates.

Packing material for the 2-ft. transfer section consisted of raschig rings in 1/4- to 1/2-in. sizes. Two material types were selected which had different wetting characteristics as given in Table 4.1 on page 19; these were unglazed ceramic rings and polyethylene tubing cut with length equal to the diameter.

Results for a packed column with 1/2-in. ceramic rings are given in Fig.2. As the pulsing energy increased the overall height of a transfer unit decreased with an obvious abrupt change in behavior. This was apparently caused by approaching emulsification and some change in hold-up within the packing. Although good transfer was obtained at the higher energy levels of pulsing, flooding occured at lower flow rates than for the sieve-plate column.

Figure 3 shows that low values of $(HTU)_{OW}$ were obtained for 1/2-in. polyethylene packing at high pulsing energies, but that flooding occured so early as to render the column relatively ineffective. However, as indicated in Fig.4, good performance was obtained without flooding at lower values of the frequency -amplitude product. The curve lines obtained in some cases are possibly the result of variable circulation rates in the dispersed phase droplets.

Figure 5 shows that the polyethylene packing gave heights of the transfer unit which were about 25% lower than those for the sieve-plate column, with ceramic packing the least effective. It is quite significant that the pulse energy input with polyethylene packing was one-fourth that of the sieve-plate column. The transfer rates were also somewhat better for plastic rings.

Approximate flooding limits were observed and are given in Fig.1 on page 14.

Results for 1/4-in. packing were obtained preliminary to the work with 1/2-in. packing by Grove.(30) The surface area presented by the smaller packing is greater.

Figure 6 gives the results with the small ceramic rings. Contrary to the results cited above the height of a transfer unit increased with increasing pulse energy. This effect possibly was the result of low hold-up of the dispersed phase. Figure 7 indicates the performance observed with polyethylene packing where $(HTU)_{OW}$ decreases with increasing pulse energy. Transfer rates are seen to be definitely much better for the plastic rings.

Figure 8 shows the results obtained with 1/4-in. bands. Contrary to the results with plate columns the flooding limit was drastically reduced and performance was intermediate between that and a column packed with either type of material. Mixed packing, distributed in a random fashion rather than in an alternative bands, might show better flooding limits while retaining good transfer characteristics. The use of larger packing might also lead to better performance of mixed beds.

Jackson concluded that a pulsed packed column showed a performance equal to or exceeding that of a pulsed sieve plate column for water-benzoic acid-toluene system. The pulsed column limited by lower flooding limits may exhibit higher efficiencies because of better transfer performance. The wetting characteristics of the packing surface can have a pronounced effect on performance. An unglazed ceramic packing wet by water gave higher flooding limit than a polyethylene packing wet by toluene. However transfer rates were substantially higher for polyethylene packings. A ratio of column diameter to packing size of 4:1 was found more satisfactory than a ratio of 8:1 because of higher flooding limits. A significantly lower pulse energy input was possible for the plastic packing wet by the organic phase. The energy requirement was only one-fourth that for a sieve-plate column which used the same system, The simplicity of the packed column for large diameters, and the advantage of low energy input and higher transfer rates in some cases, indicates that pulsed packed columnshould be considered for processing applications with liquid-liquid systems. An economic evaluation would dictate a final selection.

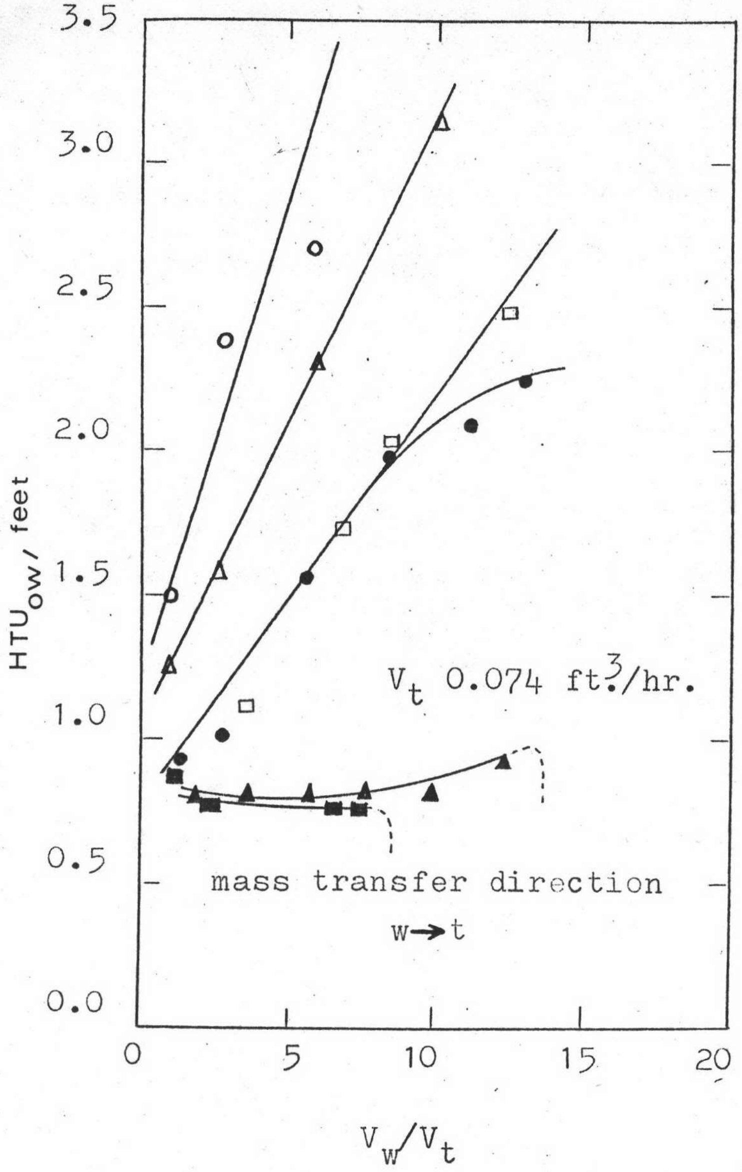

Figure 2 Performance of a pulsed packed column with 1/2-in. ceramic rings.(3)

	f, cpm.	a, in.
○	90	1/4
△	120	1/4
□	150	1/4
●	90	1/2
▲	120	1/2
■	150	1/2
-----	Flood	

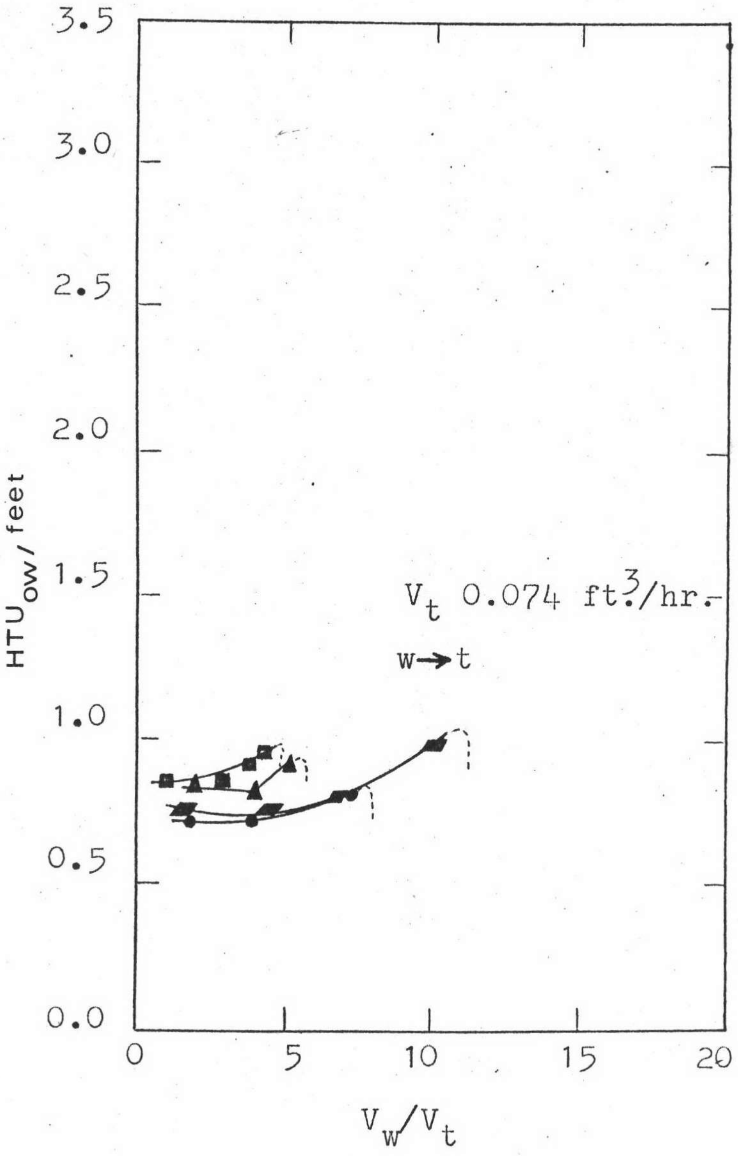

Figure 3 Performance of a pulsed packed column with 1/2-in. Polyethylene rings.(3)

	f, cpm.	a, in.
▰	75	1/2
●	90	1/2
▲	120	1/2
■	150	1/2
-----	Flood	

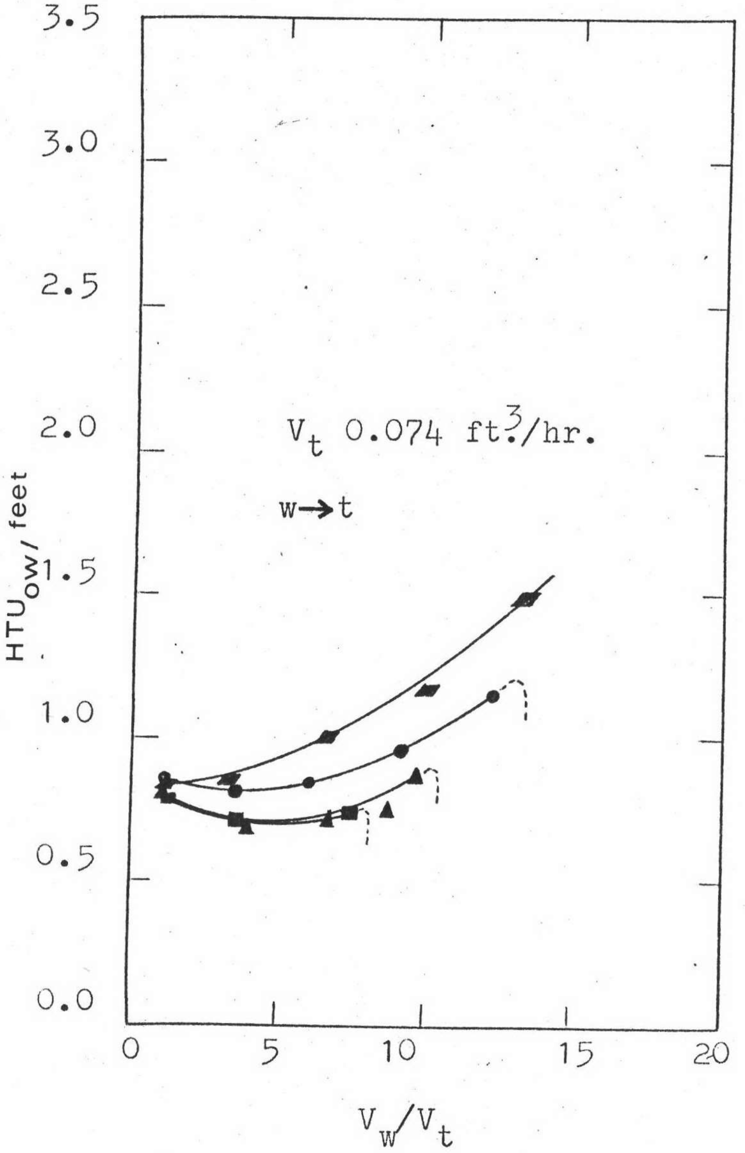

Figure 4 Performance of a pulsed packed column with 1/2-in. Polyethylene rings.(3)

	f, cpm.	a, in.
▰	75	1/4
●	90	1/4
▲	120	1/4
■	150	1/4
------	Flood	

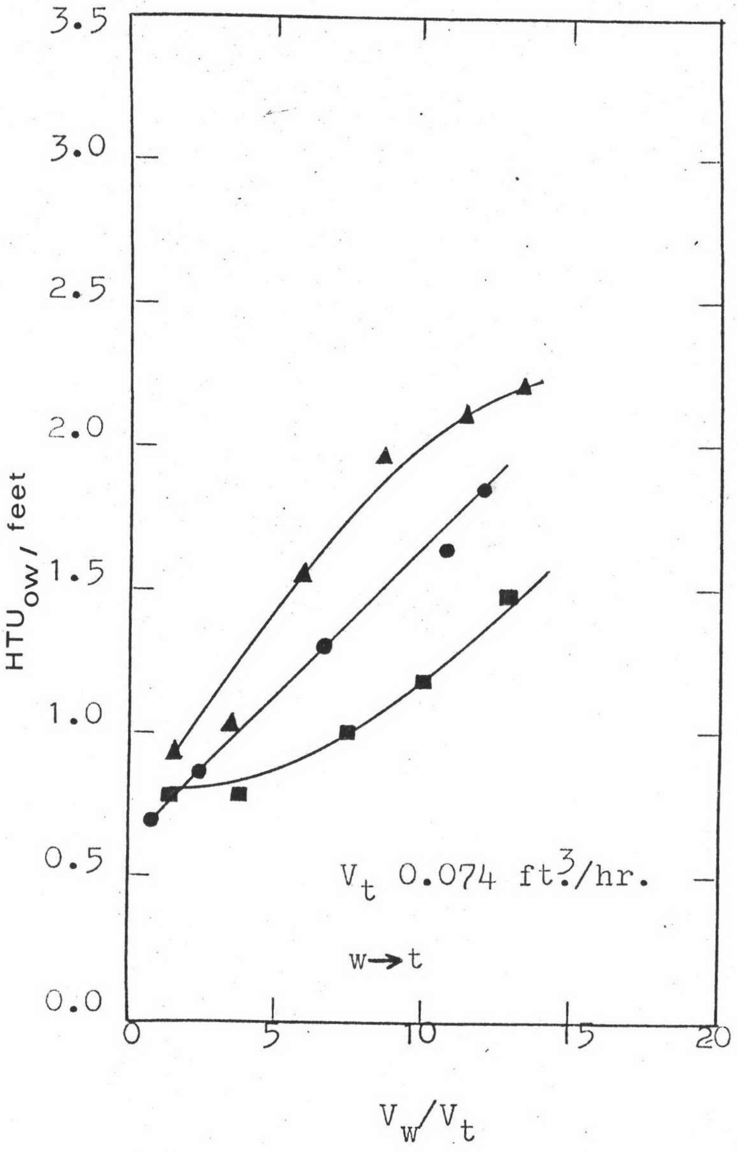

Figure 5 Performance of a pulsed sieve-plate and packed columns.(3)

Type	f, cpm.	a,in.
● sieve-plate	150	1/2
▲ 1/2-in. ceramic rings	90	1/2
■ 1/2-in. polyethylene rings	75	1/4

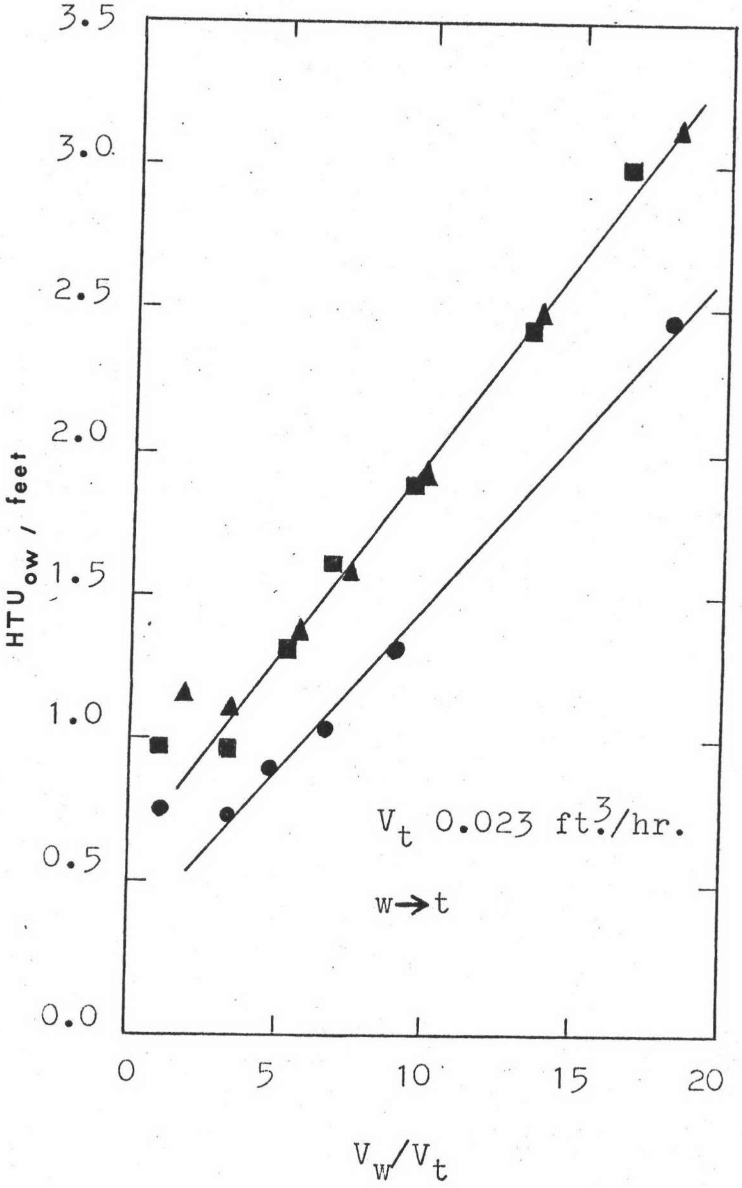

Figure 6 Performance of pulsed packed column with 1/4-in. ceramic rings.(3)

	f, cpm.	a, in.
●	60	1/4
▲	60	5/16
■	60	13/32

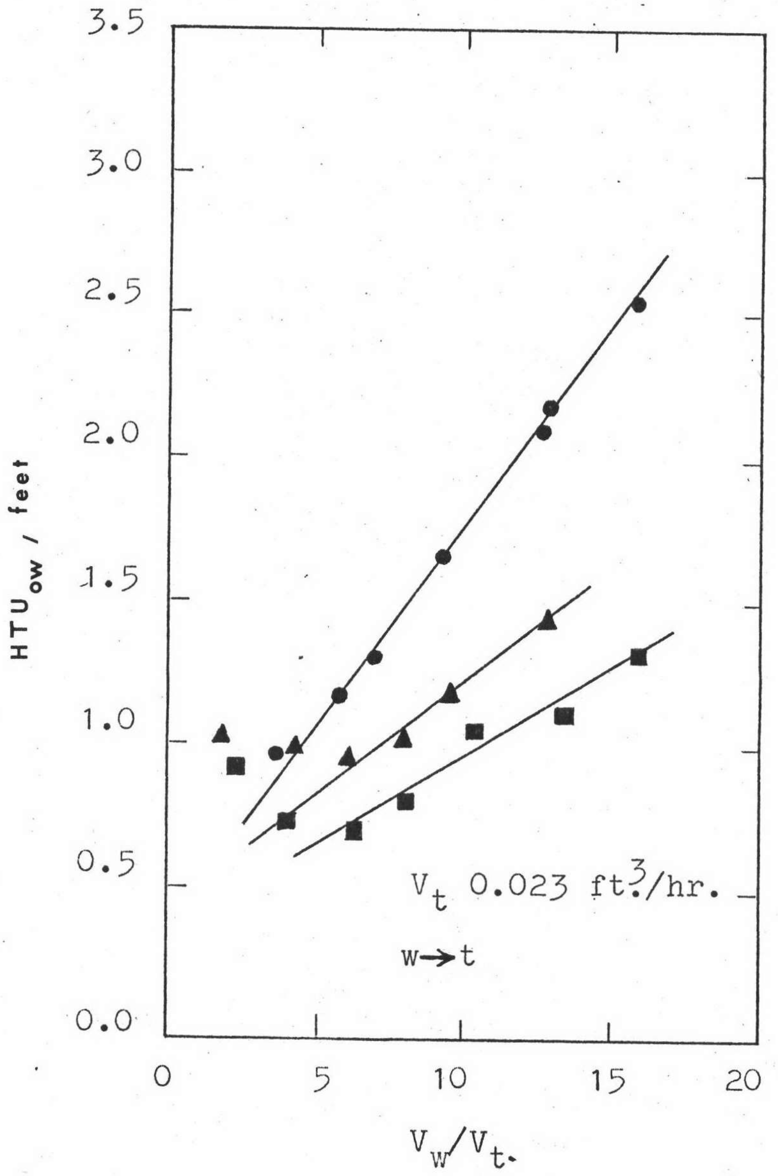

Figure 7 Performance of a pulsed packed column with 1/4-in. polyethylene rings.(3)

	f, cpm.	a, in.
●	60	1/4
▲	60	5/16
■	60	13/32

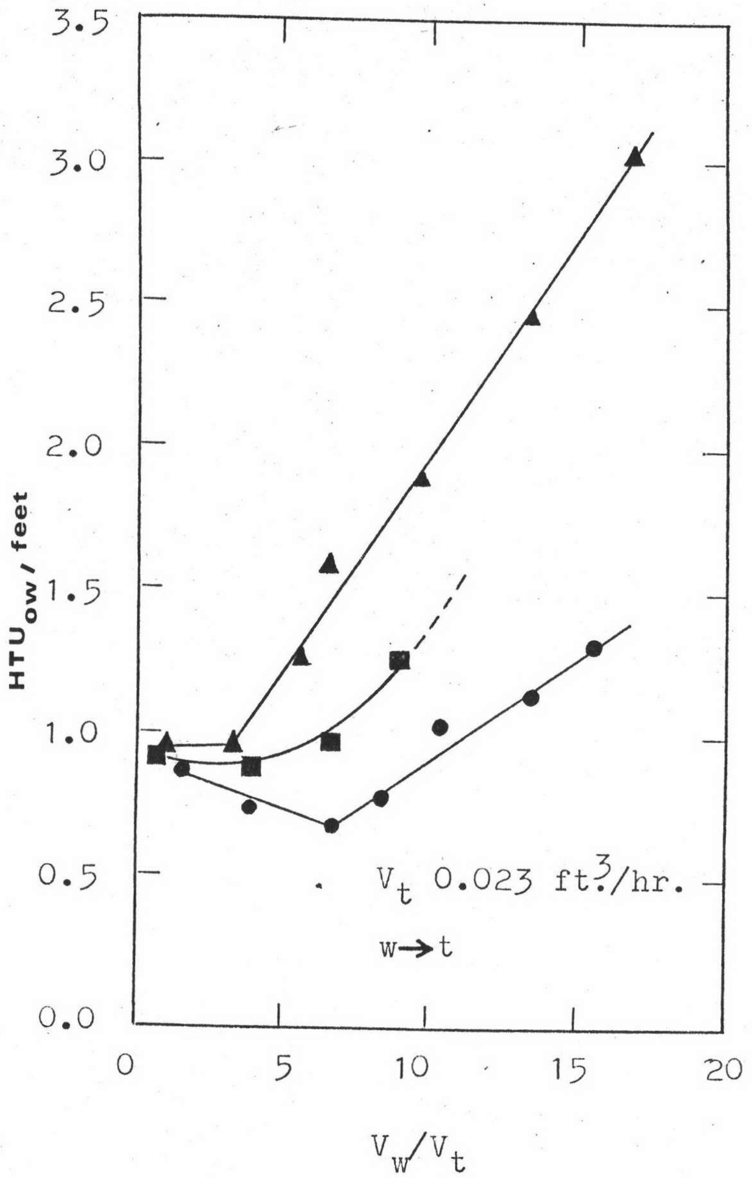

Figure 8 Performance of pulsed packed and mixed beds columns.(3)

Type	f, cpm.	a, in.
● 1/4-in. polyethylene	60	13/32
▲ 1/4-in. ceramic	60	13/32
■ 1/4-in. mixed bed	60	13/32