Radiological Study of Liquid Holdup and Flow Distribution in Packed Gas-Absorption Columns

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A radiological method has been developed to study the local flow characteristics in two-phase contacting equipment. The method presented permits the evaluation of both the vertical and horizontal components of the liquid holdup and flow distribution and can also be used to study the performance of a wide variety of two-phase contacting apparatus. An external source of radiation is used, and hence neither phase is contaminated by the radioactive material.

The application of this method is illustrated in the specific case of air-water contact in a countercurrent packed column.

Knowledge of the local flow phenomena in two-phase contacting equipment will yield important information for a better understanding of its performance. The radiological method with a radioactive isotope used as an external energy source has been found to be capable of measuring the local holdup of liquid inside any two-phase contacting device. From such holdup data, the residence time and the flow distribution can be directly calculated.

For the purpose of verification and demonstration, the water-air system flowing countercurrently in packed columns has been investigated. Packed columns were chosen, since they represent a major class of the two-phase contacting device.

THEORETICAL BACKGROUND

The method of using an electromagnetic wave to detect or measure certain physical properties of matter on the basis of its nature of absorbing such electromagnetic energy is called the *radiological method.*

A high-energy electromagnetic wave, because of its short wave length, can penetrate substances to a great extent. If such a wave consists of only one wave length, then the absorption, through a homogeneous media, can be calculated by an equation similar to that of Lambert's law for the absorption of parallel monochromatic light:

$$I = I_o\, e^{-k_\lambda t} \qquad (1)$$

where I_o is the source intensity of the parallel monenergetic radiation, I is the intensity after it has penetrated a homogeneous material of thickness t, and k_λ is the absorption coefficient which is a function of the wave length and the physical properties of the absorber.

An X-ray generator and similar equipment can be used to produce the high-energy electromagnetic waves required by the radiological study. However the use of a radioisotope is much more convenient and economical.

In the selection of an appropriate radiation source the energy level and the half life of the isotope are two primary factors to be considered. The energy level of an isotope, according to Planck's law, is inversely proportional to the wave length, which in turn is related to the absorption coefficient in Equation (1). The half life is a measure of the decay rate of the radioactive isotope.

$$I_\theta = I_o \left(\frac{1}{2}\right)^{\theta/\gamma} \qquad (2)$$

Gamma radiation is electromagnetic in nature and is highly penetrating; therefore gamma emitters are most suitable for use as the energy source in a radiological study. The intensity of the gamma radiation can be easily measured. The high-frequency electromagnetic pulses emitted by the gamma source, when inpinging on a phosphor crystal, cause molecular excitation and ionization, and part of the energy dissipated in this form is reemitted as visible or ultraviolet photons. These light scintillations, when converted to electrical pulses, can be magnified, scaled, and recorded. The resultant rate of impulse expressed in terms of the number of counts per minute is a direct measure of the radiation intensity.

Theoretically Equation (1) holds true for only the parallel monoenergetic radiation. For a point source, therefore, the objects which are closer to the source will absorb or intercept more radiation than the objects which are farther away; if one assumes that the electromagnetic wave has the property of reflection, then the same is true for objects close to the viewing port of the sensing device. Figure 1 illustrates this relationship, the numbers at the bottom indicating the relative position.

This complication, however, can be eliminated if the cross-sectional area of the absorbing object is greater than the projected area of the cone formed by the radiation wave. In the case of a packed column this requirement can be fulfilled by properly shielding the radiation source so that the maximum

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projected area of the radiation cone is smaller than the diameter of the packed bed.

It is assumed that in a packed bed the column wall, the randomly filled packing ring, and the water and air streams can be approximated by hypothetical layers. T, P, and A denote the thicknesses of the steel, ceramics, water, and air layers respectively; also the consecutive layers of the same material are identified by numeral subscripts. This schematic representation is shown in Figure 2. Furthermore the absorption coefficients for steel, ceramics, water, and air are k_t, k_p, k_l, and k_a respectively. Then by applying Equation (1) to each layer in Figure 2 consecutively one can calculate the radiation intensity behind each layer as follows:

$$\begin{aligned} I_1 &= I_0 e^{-k_t t_1} \\ I_2 &= I_1 e^{-k_p p_1} \\ I_3 &= I_2 e^{-k_l l_1} \\ I_4 &= I_3 e^{-k_a a_1} \\ &\cdots \\ I_{n-2} &= I_{n-3} e^{-k_l l_m} \\ I_{n-1} &= I_{n-2} e^{-k_a a_m} \\ I_n &= I_{n-1} e^{-k_t t_2} \end{aligned} \tag{3}$$

Eliminating I_{n-1} from the last equation and so on, one obtains

$$I = I_n = I_0 e^{-k_t t_1 - k_p p_1 - k_l l_1 - k_a a_1 - \cdots - k_t t_2}$$

or

$$I = I_0 e^{-k_t \Sigma t - k_p \Sigma p - k_l \Sigma l - k_a \Sigma a} \tag{4}$$

Assuming that air absorbs gamma radiation much less than does steel, water, and ceramics and also assuming that the column wall thickness and the cumulative thickness of the ceramic packing rings across a specific segment of the column are constant, one can reduce the above equation to

$$I = (I_0 e^{-k_t T - k_p P}) e^{-k_l \Sigma l}$$

or

$$I = I'_0 e^{-k_l \Sigma l} \tag{5}$$

Equation (5) reduces to

$$I_F = I'_0 e^{-k_l L} \tag{6}$$

when the packed column is filled with water; L is the over-all "thickness" of the water layers, equal to the void space within the column. Similarly, when the column without packing, is filled with water,

$$I^\circ_F = I''_0 e^{-k_l D} \tag{7}$$

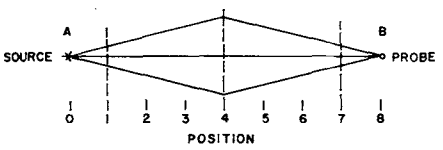

Fig. 1. Geometric configuration of nonparallel radiation source.

If I in Equation (5) is measured while the column is under operating condition, then Σl is the thickness of the running-water layers.

In Equations (5) and (6) I'_0 is the radiation intensity of the packed column when $\Sigma l = 0$, or when the column is completely dry, if one lets

$$I_E = I'_0 \tag{8}$$

for the packed column, and

$$I^\circ_E = I''_0 \tag{9}$$

for the unpacked column, and notes that the ratio L/D is actually the void fraction, and $\Sigma lm/L$ is the local liquid holdup. Then

$$\frac{\Sigma l}{L} = \text{holdup} = \frac{\log (I/I_e)}{\log (I_f/I^\circ_e)} \tag{10}$$

and

$$\frac{L}{D} = \text{void fraction} = \frac{\log (I_e/I_e)}{\log (I^\circ_f/I^\circ_e)} \tag{11}$$

Consequently if the radiation intensities (I, I°_e, I_e, I°_f, and I_f) across a specific segment of the packed column are known, then the liquid holdup and the void fraction of that segment can be calculated. The holdup and the void fraction measured by this method refer only to a specific section within the column, and therefore they are called the local holdup and the local void fraction.

Three types of liquid holdups actually exist, which are defined as follows

1. *Total Holdup*: The total amount of liquid held within the apparatus under operating conditions is defined as the total holdup. It is determined by measuring the radiation intensities while the column is in operation.

2. *Static, or Permanent, Holdup*: The amount of the stagnant liquid which does not flow freely through the apparatus is defined as the static holdup. This part of the liquid either adheres to the surface or is trapped within the interstitial pockets of the packing rings. It can be determined by measuring the radiation intensities across the column after the supply of the influent liquid is discontinued.

3. *Operating Holdup*: The amount of liquid which flows freely through the apparatus under the operating conditions is defined as the operating holdup. It is the algebraic difference of the total holdup and the static holdup. Through this part of the liquid, which represents the constantly moving fresh fluid masses within the column, the mass transfer takes place.

These definitions of holdups are similar to those in the literature (*4, 5, 8*) except that they are expressed as the percentage of the dry void space which is occupied by the liquid. They are so defined because the fractional volume of the void space which is taken up by the liquid is of actual interest. To convert the holdups from percentage void space to percentage over-all volume of the column, the local holdups should be multiplied by the corresponding local void fraction.

In a randomly packed column the packing rings form a certain configuration; consequently the liquid inside the column follows certain channels and the local holdups within the various segments of the column differ considerably from one place to another. To determine the actual flow distribution, the local holdups within the various segments must be measured separately. In Figure 3 the packed column is divided into three concentric rings. The liquid holdups within each of the rings are determined by placing the measuring device at the appropriate lateral and transverse positions. Measurements at several positions are necessary for a representative average.

From similar measurements made at various vertical levels the liquid distribution along the column height can also be studied. The following equation is used to give the average liquid holdup inside a concentric ring and at a particular vertical level:

$$\begin{aligned} &(\text{average holdup})_i \\ &= \frac{1}{4n} \sum_{j=1}^{n} (H_{n,i,j} + H_{e,i,j} + H_{w,i,j} \\ &\qquad + H_{s,i,j}) \end{aligned} \tag{12}$$

where i denotes the specific concentric ring, j denotes each of the n vertical levels in the region of interest, and N, E, W, and S denote the circumferential positions.

REVIEW OF PREVIOUS WORK

Liquid holdup in packed gas-liquid contacting columns has been studied extensively. Jeber and Elgin (*4*) made a comprehensive study of the operating holdup for various packing materials; Otake and Okada (*6*) correlated most of the available literature data on liquid holdup and recommended an empirical formula for calculating the liquid holdup

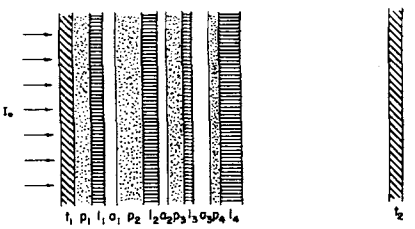

Fig. 2. Hypothetical layers of the packed column.

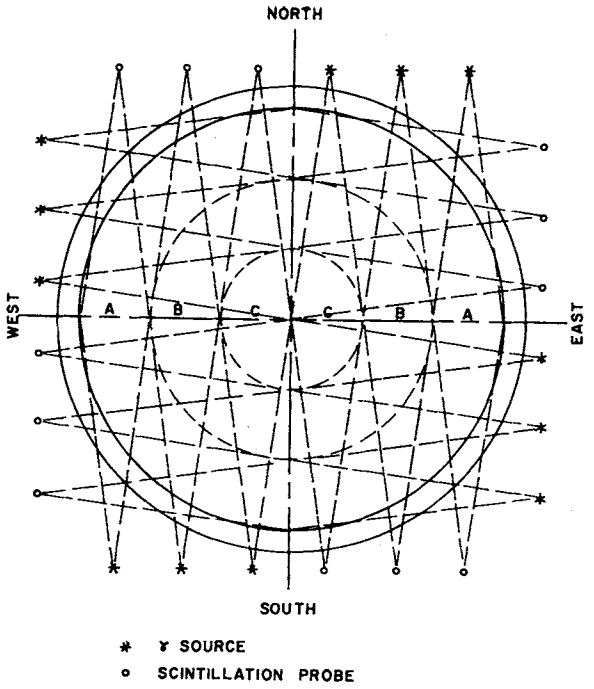

Fig. 3. Three concentric rings and corresponding lateral and traverse positions for scanning.

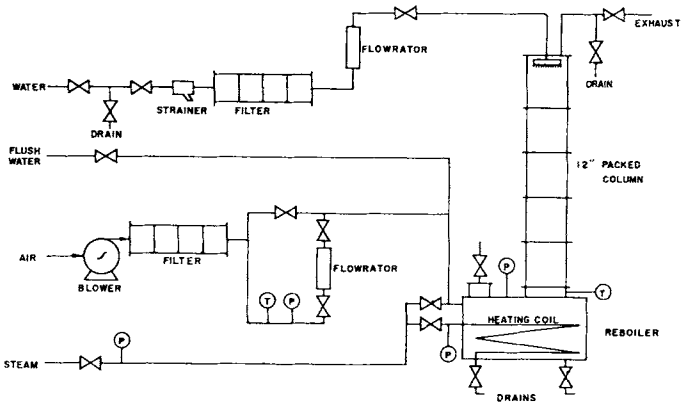

Fig. 4. Schematic flow diagram for the 12-in. packed-column system.

from the flow rates and the fluid properties. Most recently Schulman *et al.* (*7, 8*) measured the total static and operating holdups in a packed column using both aqueous and nonaqueous systems.

The principle of using a radiation-absorption method has been applied successfully to the study of other two-phase problems by earlier investigators. Isbin, Sher, and Eddy (*3*) recently used this technique to study the two-phase steam-water flow in a circular pipe, with selenium-75 as the energy source. These authors cited that Zmola and Bailey (*9*) have used iridium-192 to measure the densities of a boiling liquid; Anson, Belin, and Horlor (*1*) used a beta-ray absorption technique to measure the density of a steam-water mixture at the throat of a circular flow nozzle.

EXPERIMENTATION

Two columns were used in this investigation; one was 6 and the other 12 in. in diameter. The smaller column was used to gain preliminary knowledge of this new technique and to study the effect of the packing size and the flooding condition on the liquid holdup. The larger column was mainly used to investigate the flow distribution, both radially and vertically.

The schematic flow diagram is shown in Figure 4. The accessory equipment used for each of the columns was of the conventional type. Only the liquid distributors were specially made. They were designed to assure initial uniform distribution of the liquid at the top of the tower packing (Figure 5). The detailed description of the equipment is available in reference 2.

The 6-in. column was made from a section of Pyrex-glass pipe, 4 ft. long. Three sizes of unglazed ceramic Raschig packing rings were used (½, ¾, and 1 in.), corresponding to the diameter ratios of 12:1, 8:1, and 6:1 respectively. These ratios represent above, at, and below the critical of 8:1 (*5*). In Addition, the unpacked-column operation was also studied. The water rate was varied from 1,000 to 10,000 lb./(hr.)(sq. ft.), and the air, flowing countercurrently to the water, was varied from 0 to 400 lb./(hr.)(sq. ft.). Thirteen millicuries of mercury-203 were used as the energy source. Mercury-203 has a single-energy level of 0.279 mev. and a half life of 45.8 days.

The experimental results obtained from this column show that the total liquid holdup increases with the liquid rate (Figure 6), whereas the gas rate has negligible effect below the loading point

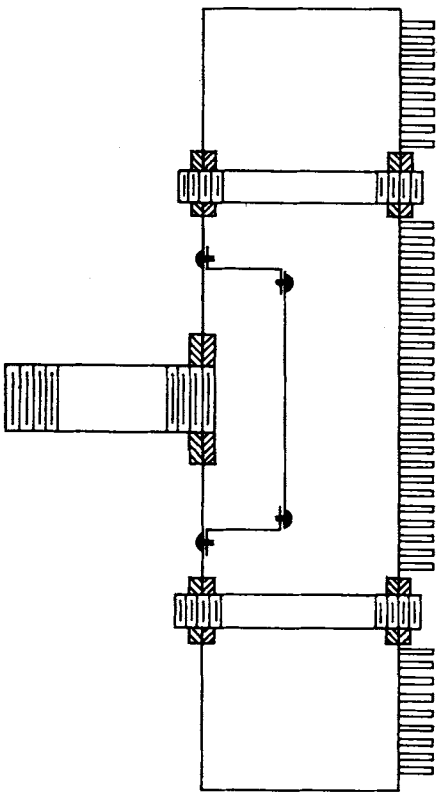

Fig. 5. Liquid distributor for the 12-in. column.

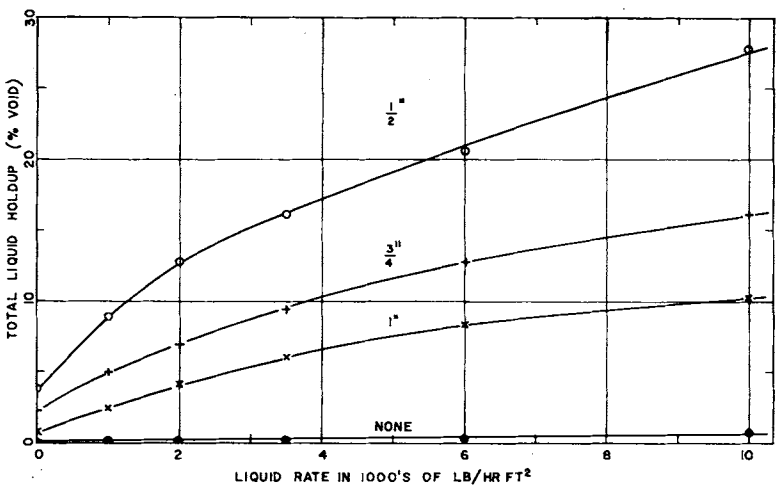

Fig. 6. Effect of liquid rate on total liquid holdup for the unpacked and Raschig-ring packed 6-in. column.

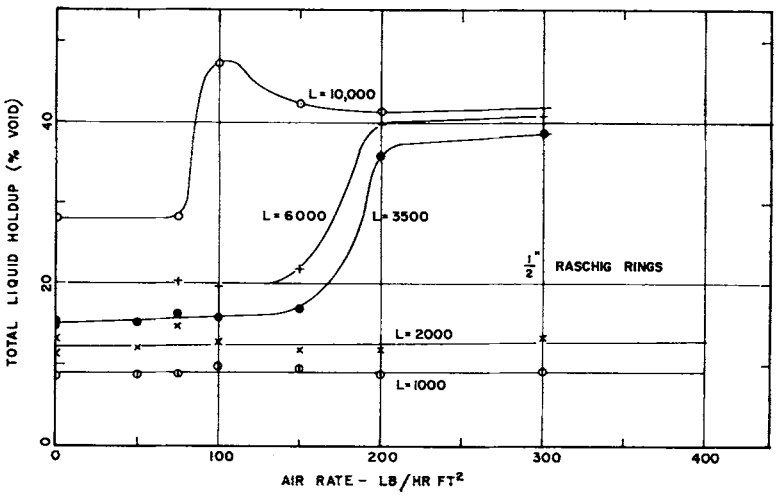

Fig. 7. Effect of gas rate on total holdup in the 6-in. column packed with ½-in. Raschig rings.

(Figure 7). Each of the experimental points is the average value of two separate runs. In each run measurements were made at four circumferential positions and three vertical levels, symmetrically located around the center of the column.

Although the gas rate has negligible effect on the liquid holdup below the loading point, the liquid holdup shows a rapid increase beyond this point (Figure 7). It is believed that at the loading point the water droplets start to stay afloat, since the upward drag force caused by the high air rate balances the gravitational force at this point. This ascending trend continues until the flooding point is reached. Further increase of the air rate, instead of keeping the liquid droplets afloat, carries the droplets away, hence reducing the liquid holdup. This explains the maximum dome appearing in the top curve of Figure 7.

The effect of the packing size and the void fraction was found to be as follows; under the same operating conditions the larger packings always give a lower liquid holdup, and for various sections of the column a lower holdup is always observed at regions where the local void fraction is higher. Figures 8 and 9 show these relationships and the entrance effect when the column is under the normal operating conditions, as well as when it is operated at flooding.

For the study of flow distribution a 20-ft. 12-in. steel column packed with 1-in. Raschig rings was used. This larger column allowed a much more complete scanning procedure for the study of various localized holdup phenomena. Two liquid rates, 1,300 and 13,000 lb./(hr.)(sq. ft.), and two gas rates, 0 and 530 lb./(hr.)(sq. ft.), were investigated. Twenty-three millicuries of thulium-170 were used as the energy source. Thulium-170 has a lower energy level (0.0842 mev.) than mercury-203; hence it yields a more sensitive measuring system. The half life of thulium-170 is 127 days.

The local liquid holdups measured in the 12-in. column, summarized in Table 1. correspond to the average values calculated by Equation (12) with $n = 4$. These results indicate that for the packed column with a column to packing diameter ratio of 12:1 the distribution of liquid remains uniform across the cross section of the column to the entire depth of its 18 ft. of packing. The strong variation of local holdup indicates that the channeling effect exists. Moderate changes of liquid and gas rates do not alter the general fluid-distribution pattern.

The radiation-absorption measuring device used for the 12-in. column consisted of a gamma source and a scintillation-detecting probe. They were shielded and mounted on a set of parallel tracks; the geometric layout is shown in Figure 10, and A and B are such that the intersection of the two cones is located at the center of the column. The tracks were rotated on a steel ring which had been properly grooved and mounted on a platform, and the platform moved vertically along the column, thus allowing the measuring device to scan at any part of the column. The detecting device used for the 6-in. column was similar in nature but somewhat less elaborate.

COMPARISON OF RESULTS WITH PREVIOUS INVESTIGATORS

Shulman, Ullrich, and Wells (8) made an extensive study of liquid holdup on a 10-in. packed column. They correlated their results with the data of several previous investigators and presented the following equations:

$$h_t = \frac{\alpha l^\beta}{D_p^2} \qquad (13)$$

$$h_s = \delta D_p^{-x} \qquad (14)$$

$$h_o = h_t - h_s \qquad (15)$$

$$\beta = \gamma D_p^{\nu} \qquad (16)$$

The values of the constants are given in Table 2. To convert these equations from percentage of the column volume to percentage of the void volume, one should divide by the void fraction:

$$H_t = h_t/\epsilon = (\alpha L^\beta)(\epsilon D_p^2) \qquad (17)$$

$$H_s = h_s/\epsilon = (\delta/\epsilon) D_p^{-x} \qquad (18)$$

$$H_o = H_t - H_s \qquad (19)$$

These equations are applicable below the loading point only.

Table 2. For Porcelain Rashig Rings

	½	1 in.
D · ft.	0.0582	0.1167
α	2.25×10^{-5}	2.25×10^{-5}
γ	0.965	0.965
δ	0.00104	0.00104
x	1.21	1.21
v	0.376	0.376

Figure 11 shows the results of the present study in comparison with the experimental data of Shulman *et al.* and the results calculated from their equations.

The data obtained for the ½-in. rings are shown at the top of the graph. The experimental results of this study are in fair agreement with Shulman's, but they deviate from the calculated results beyond 2,000 lb./(hr.)(sq.ft.). For the 1-in. raschig rings, results are available on both 6 and 12-in. columns from this study. The data on the 6-in.

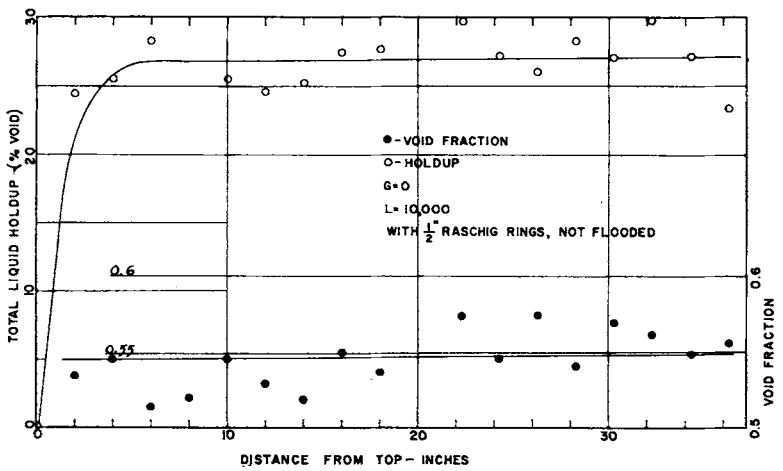

Fig. 8. Longitudinal variation of holdup and void fraction from the top of the packing under normal operating conditions in the 6-in. column.

column are not in good agreement with Shulman's because of the low column-to-packing diameter ratio (6:1), which may have caused excessive channeling. The data obtained on the 12-in. column indicate very good agreement with Shulman's experimental data. Since the 1-in. packing has a much higher loading point than the ½-in. packing, the experimental data follow the calculated results of the correlating equation to a higher liquid rate.

It should be noted in the consideration of this comparison, particularly with regard to the 12-in. column, that the data of the current investigation represent an average of the lateral and vertical scannings which in themselves show considerable variation throughout the column; yet the summation shows good agreement with the cumulative values obtained by other methods.

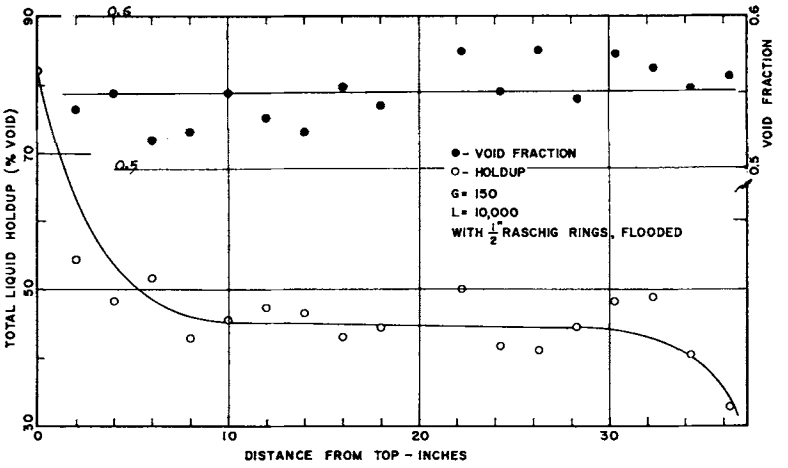

Fig. 9. Longitudinal variation of holdup and void fraction for the top of the packing under flooded conditions in the 6-in. column.

CONCLUSIONS

1. The radiological method described in this paper presents a new simple method of studying the local hydrodynamic characteristics of the packed column or other similar two-phase contacting equipment.

2. This method does not contaminate or disturb the system in any fashion.

3. The liquid-holdup data and the void fraction determined by the radiological method can be calculated by the following equations, which are derived by applying Lambert's law for the absorption of parallel monochromatic light to the radiological system:

$$H_t = \frac{\log I/I_e}{\log I_f/I_e}$$

$$\text{void} = \frac{\log I_f/I_e}{\log I_f^\circ/I_e^\circ}$$

4. By scanning the packed column in a systematic manner, one can obtain the local liquid holdups and the flow distribution.

5. The good agreement of the experimental results from this study and the corresponding data obtained by other methods prove the validity of this new technique.

ACKNOWLEDGMENT

The authors wish to express their gratitude for the Texas Company Fellowship which made this study possible.

Table 1. Local Operating Liquid Holdup in the 12-in. Column

H_o, percentage of void volume operating conditions	From wall to 2 in. from the wall	From 2 to 4 in. from the wall	From 4 in. from the wall of the column
L = 13000, G = 0 lb./(hr.)(sq. ft.)			
0-5	15.04	14.05	14.95
5-9	9.98	12.61	15.18
9-13	12.11	12.42	12.02
13-18	15.46	14.21	16.05
L = 13000, G = 530 lb./(hr.)(sq. ft.)			
0-5	18.60	15.90	14.36
5-9	13.76	13.38	15.03
9-13	12.56	11.89	13.65
13-18	17.58	14.45	17.31
Average void fraction			
0-5	0.719	0.732	0.717
5-9	0.750	0.724	0.680
9-13	0.748	0.749	0.689
13-18	0.701	0.724	0.716

NOTATION

a = thickness of air layer
D = inside diameter of the column
D_p = equivalent diameter of the packing
e = 2.7183
h = liquid holdup, percentage column volume
h_t = total holdup
h_s = static holdup
h_o = operating holdup
H = liquid holdup percentage column void
H_t = total holdup
H_s = static holdup
H_o = operating holdup
I = intensity of the parallel monoenergetic radiation
I_e = intensity when the column is dry
I_f = intensity when the column is filled with water
I_n = intensity after penetrating the R_{th} layer
I_0 = source intensity, or at time O
I_θ = intensity at time θ
I° = intensity without packing
I' = $I_o e^{-k_t t - k_p p}$
I'' = same as $'$ except $p = 0$
k^λ = absorption coefficient for Equation (1)
l = thickness of water layer
L = liquid rate, lb./(hr.)(sq.ft.); cumulative length of the void space
m = number of hypothetical layers
n = number of vertical levels
p = thickness of ceramic layer
t = thickness of the homogeneous absorbing media, the column wall
x = constant
v = constant

Greek Letters

α = constant
γ = constant
δ = constant

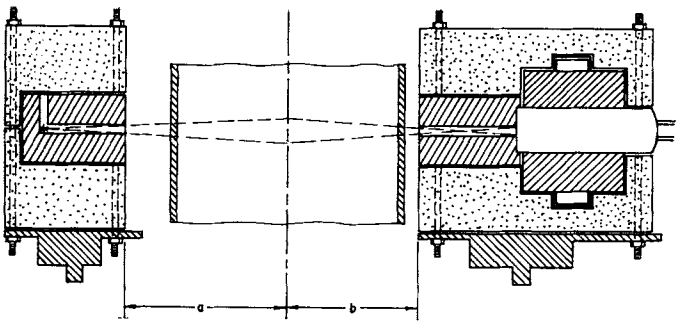

Fig. 10. Scintillation probe and gamma source assembly with the lead shieldings.

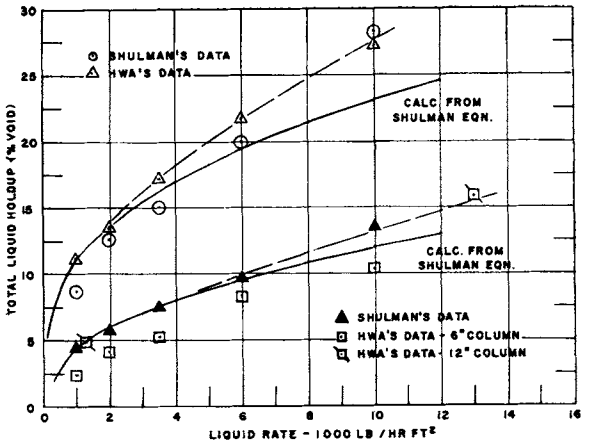

Fig. 11. Comparison of results at $G = 0$ (top two curves for ½-in. packing, lower two curves for 1-in. packing, solid lines for calculated results).

Γ = half life
ϵ = void fraction
λ = wave length
θ = time
Σ = summation

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Manuscript received October 27, 1958; revision received September 2, 1959; paper accepted September 14, 1959. Paper presented at A.I.Ch.E. Atlantic City meeting.

Ion Exchange Kinetics for Systems of Linear Equilibrium Relationships

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The mathematical equations describing the kinetic behavior of ion exchange processes have been solved analytically for systems having linear equilibrium relationships of the type $q^* = K_1 + K_2C^*$. The concentration ratios of the effluent to the influent solution for these cases are found to depend on parameters involving time, position, and relative resistances of the liquid and resin phases. Numerical results have been obtained and are presented in tabular and graphical forms. Furthermore expressions for the constant pattern breakthrough curve for two special cases have also been worked out.

The mathematical equations describing the kinetic behavior of an ion exchange process with the rate-controlling mechanism represented by a combination of resistances for both the liquid and resin phases have been presented elsewhere (*5, 6*). These expressions consist of the equation of continuity, the rate of transfer, the equations describing the concentrations of the resin phase, and the relationship of surface concentrations:

$$\frac{\partial C}{\partial \xi} + \frac{\partial q}{\partial \tau} = 0 \qquad (1)$$

$$\frac{\partial q}{\partial \tau} = k_l (C - C_s) \qquad (2)$$

$$q(\xi, \tau) = \frac{3}{b^3}\int_0^b q_l(r, \xi, \tau) r^2 dr \qquad (3a)$$

$$q_s(\xi, \tau) = q_l(b, \xi, \tau) \qquad (3b)$$

$$q_l(r, \xi, \tau) = \int_0^\tau q_s(\xi, \tau) \frac{\partial}{\partial \tau} H(r, \tau - \lambda) d\lambda \qquad (3c)$$

and

$$q_s = f(C_s) \qquad (4)$$

where

$$\xi = \frac{\rho}{u} z, \quad \tau = t - \frac{\phi}{u} z$$

$$H(r, \tau) = 1 - 2\sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{\left(\frac{n\pi}{b}\right) r} e^{-D\left(\frac{n\pi}{b}\right)^2 \tau} \cdot \sin \frac{n\pi}{b} r$$

and the following initial and boundary conditions exist:

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