Performance of Gas Bubble Columns: Volumetric Liquid-Phase Mass Transfer Coefficient and Gas Holdup

FUMITAKE YOSHIDA and KIYOMI AKITA

Kyoto University, Kyoto, Japan

The gas bubble column in which a gas is bubbled through a deep liquid without mechanical agitation is useful for carrying out various gas-liquid chemical reactions. In the present work the volumetric coefficient for liquid-phase mass transfer k_La in four gas bubble columns of different diameters was studied by means of the sulfite oxidation technique, that is oxidation of an aqueous sodium sulfite solution by air or oxygen with cupric ions as the catalyst. The data were compared with those for desorption of oxygen from pure water and from an aqueous sodium sulfate solution. The gas holdup in this type of column was also determined. One of the objectives of the experiments was to study the effect of the size of the equipment to obtain a basis for the scaling up.

EXPERIMENTAL

Figure 1 shows a schematic diagram of the experimental apparatus. The columns were constructed of transparent vinyl chloride resin, and their dimensions are given in Table 1. Single gas inlet nozzles of various diameters were installed at the bottom center of each column. There was overlapping of nozzle diameters between columns of adjacent sizes. The clear liquid or froth height was varied as given in Table 1 by varying the amount of solution charged at the beginning of an operation.

The sodium sulfite solution was of initial concentration of approximately 0.3 N and contained roughly 10^{-4} g.-moles of cupric sulfate/liter. Air, or in some runs pure oxygen, was fed to the column bottom after being metered with an orifice. Operation was batch-by-batch with respect to solutions. The decrease in the sulfite concentration with time was determined by analyzing liquid samples taken at constant intervals, usually of 2 min. Figure 2 is an example* of the plot of the sulfite concentration against time for two series of runs at two different gas rates with the column 15.2 cm. in diameter and a froth height of 182 cm. at a temperature of 20°C. The data points for liquid samples taken at five different levels fall on single straight lines, indicating that the liquid concentration at a given instant was uniform throughout the vessel within accuracy of analysis. Furthermore, the average concentration of the whole solution discharged from the vessel and well mixed after series of runs was equal to those of liquid samples taken at the end of the last run. This was also true with the other three sizes of columns, indicating a very high degree of liquid mixing in the axial as well as in the radial directions.

Values of the overall coefficient K_La, with respect to the unit volume of aerated liquid, were calculated by

$$-(1-\epsilon_G)\frac{dC}{d\theta} = 2K_La\,H\,p \tag{1}$$

where $dC/d\theta$ is the slope of the concentration-time plot. The Henry's law constant H was estimated from the solubility of oxygen in water with correction for the presence of ions by the method of Van Krevelen (*4*) on the assumption that the solubility of oxygen in the sodium sulfite solution was equal to that in a sodium sulfate solution of the same normality. The partial pressure of oxygen in the bulk of liquid was assumed zero. Even with this assumption, the true driving potential to be used in the calculation should depend on the extent of mixing in the gas phase. However, since the extent of mixing was not known in the present work, the arithmetic mean of the partial pressures of oxygen at the top and bottom of the column, taking into account of the effect of static liquid head, was taken as the mean value of p, which was equal to the mean driving potential in this case. The error involved in this procedure is perhaps less than few per cent, since the mole fraction of oxygen in the air leaving the column obtained by the material balance was always over 0.18. The fact that the lines in Figure 2 are straight indicates that the absorption rate is independent of the concentration of sodium sulfite.

Runs were also made with the 15.2- and 30.1-cm. columns for the desorption of oxygen from the aqueous solution of sodium sulfate and from the aqueous solution of glycerol for the purpose of comparing the oxidation data with those on

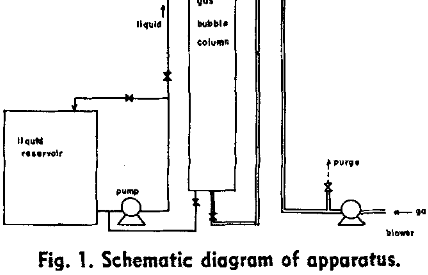

Fig. 1. Schematic diagram of apparatus.

TABLE 1. DIMENSIONS OF COLUMNS

Columns	Inside diameter D, cm.	Clear liquid height Z_L, or froth height Z_F, cm.	Nozzle diameter d, cm.
No. 1	7.7	95 (Z_F) 135 (Z_F) 176 (Z_F)	0.225 0.495 0.778
No. 2	15.2	90 (Z_F, Z_L) 145 (Z_F, Z_L) 182 (Z_F)	0.404 0.833 1.71
No. 3	30.1	120 (Z_F) 189 (Z_F)	1.48 3.00
No. 4	60.0	126 (Z_L) 206 (Z_L) 350 (Z_L)	1.00 2.00 4.00

---

* Tabular material has been deposited as document 8088 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $5.00 for photoprints or $2.25 for 35-mm. microfilm.

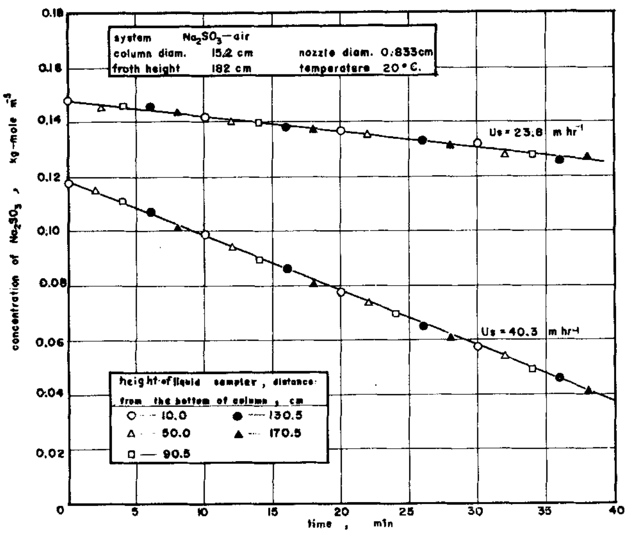

Fig. 2. Concentration vs. time plots for sulfite oxidation.

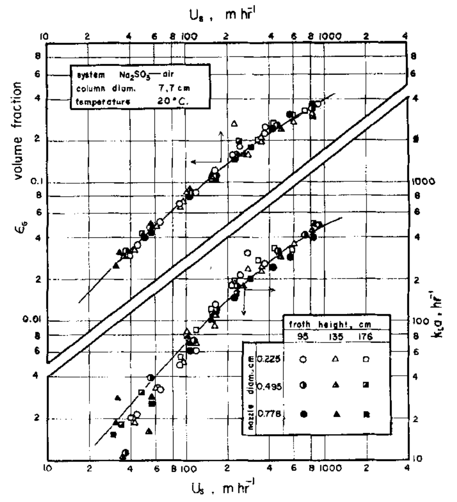

Fig. 4. Gas holdup and k_La in 7.7-cm. column.

pure physical desorption. For analytical procedures the reader is referred to a previous paper (5).

The gas holdup was determined by directly measuring the height of aerated liquid and that of clear liquid without aeration. Thus, the average fractional gas holdup is given as

$$\epsilon_G = \frac{Z_F - Z_L}{Z_F} \tag{2}$$

Since it was rather difficult to read distinctly the level of aerated liquid, the values of ϵ_G thus obtained probably involve a maximum error of about 10%. This error may be reflected in the K_La values calculated by Equation (1), although to a lesser degree depending on the value of ϵ_G.

GAS-PHASE RESISTANCE IS NEGLIGIBLE

Runs for sulfite oxidation were made at 20°C. in the 7.7-cm. column with use of air and pure oxygen. K_La values were plotted against the superficial gas velocity with respect to the cross-sectional area of the column. Within accuracy of experiments difference in K_La values between runs with air and those with oxygen was negligible. This indicates that the mass transfer resistance in the gas phase is negligible compared with that in the liquid phase. The same conclusion was reached by Yoshida and others (5) on the basis of their data on sulfite oxidation in gas-liquid contactors with mechanical agitation. Thus, the overall coefficient K_La for this particular system and apparatus can be regarded as the liquid film coefficient k_La. Furthermore, the fractional gas holdup in the sodium sulfite solution with air was equal to that with oxygen at a given gas rate.

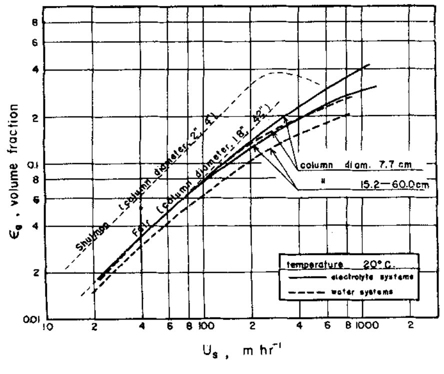

Fig. 3. Effects of system and column diameter on gas holdup.

EFFECT OF ELECTROLYTES ON GAS HOLDUP

Figure 3 compares the values of the fractional gas holdup for the systems of air-water, air-sodium sulfite solution, and air-sodium sulfate solution. The fractional gas holdup in water and electrolyte solutions in the 7.7-cm. column was slightly larger than in the other larger columns, probably due to the wall effect. This observation agrees with that of Fair and others (2) for air-water system. It was visually observed through the column walls that gas bubbles in water were mostly less than 10 mm. in size, whereas bubbles in electrolyte solutions consisted mostly of very fine bubbles and were dotted with larger ones of several millimeter diameter. The occurrence of smaller bubbles in electrolyte solutions can be explained by the electrostatic potential at the gas-liquid interface.

Figure 3 also shows comparative air-water data of Fair for the columns 18 and 42 in. in diameter and the air-water data of Shulman (3) for the 2-in. and 4-in. columns. The disagreement could be explained by the difference in the design of dispersers. Shulman used the porous plate as the disperser, whereas Fair used multiple gas inlet nozzles.

EFFECT OF CATALYST CONCENTRATION

Values of k_La and those of the fractional gas holdup were independent of catalyst concentrations for the range studied, that is from 10^{-5} to 10^{-3} g.-moles/litre of solution.

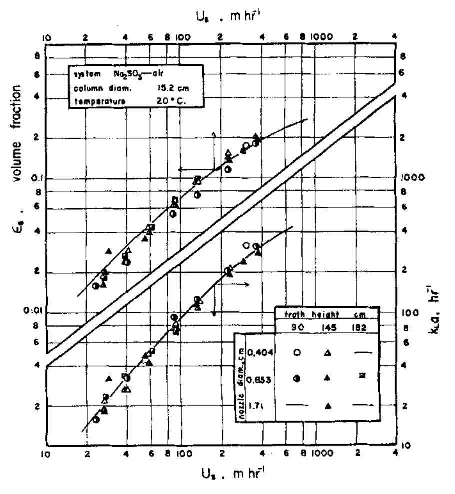

Fig. 5. Gas holdup and k_La in 15.2-cm. column.

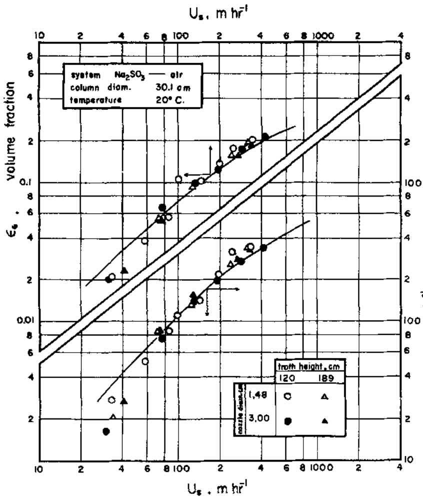

Fig. 6. Gas holdup and k_La in 30.1-cm. column.

EFFECT OF GAS RATE

Figure 4 shows how the fractional gas holdup and k_La values for sulfite oxidation at 20°C. in the 7.7-cm. column vary with the superficial gas velocity. Figures 5, 6, and 7 are similar plots for the data with the 15.2-, 30.1-, and 60.0-cm. columns, respectively. The average rising velocity of bubbles relative to the column wall U_B can be obtained by dividing the superficial gas velocity U_S by the fractional gas holdup ϵ_G.

Figure 8 shows the calculated values of U_B in sulfite solutions plotted against the values of U_S, indicating that the relationship between U_B and U_S is almost independent of the column diameter, except for the 7.7-cm. column, for which U_B was considerably smaller at higher superficial gas velocities. The broken line in the figure represents the maximum value of the rising velocity of single bubbles, which depends on the bubble size, reported by Datta et al. (*1*).

EFFECT OF NOZZLE DIAMETER

In Figures 4, 5, 6, and 7 data with gas inlet nozzles of various diameters in each column are plotted. Neither k_La nor the fractional gas holdup is affected by the nozzle diameter. This can be explained by the fact that, in the range of gas rates studied in the present work, gas flows out of the nozzle as a continuous jet stream and then is split into bubbles by the turbulent motion of liquid in a zone several inches above the nozzle.

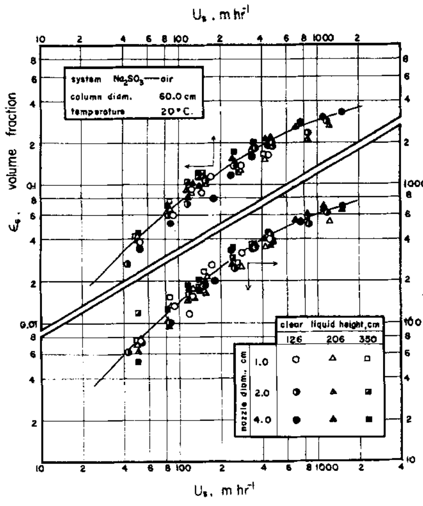

Fig. 7. Gas holdup and k_La in 60-cm. column.

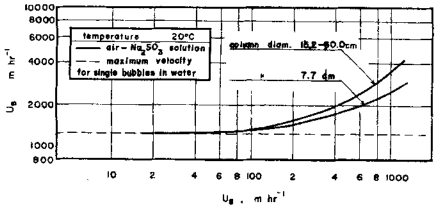

Fig. 8. Average velocity of ascending bubbles.

EFFECT OF LIQUID HEIGHT

Figures 4, 5, 6, and 7 also show that neither k_La nor the gas holdup depends on the liquid height. Since conditions in the vicinity of the gas inlet nozzle are different from other parts of the column, some end effects are expected in shorter columns. However, in such taller columns as studied in the present work, over about 1 m. in height, the end effects are masked by experimental errors, and the fractional gas holdup and k_La are practically independent of the liquid height.

EFFECT OF COLUMN DIAMETER

The effect of the column diameter on the gas holdup was already mentioned in reference to Figure 3. Figure 9 shows that the values of k_La increase with increasing column diameter. The trend does not level off within the range of diameter used. The same trend was observed with the data on desorption of oxygen from water, as will be shown later.

In the 7.7-cm. column, plug flow was observed even at relatively low gas velocities. In the larger columns, however, no plug flow was observed, and more violent motion and more downward movement of fluids along the wall existed. Bubbles near the wall showed very complicated motions. They moved up, around, and down along the wall, and more downward movement was observed at higher gas velocities. At the present stage of knowledge, however, it seems appropriate not to state definitely why values of k_La increase with the column diameter.

EFFECT OF TEMPERATURE

Figure 10 shows the data for the gas holdup and k_La for sulfite oxidation in the 7.7-cm. column at three temperatures, 10°, 20°, and 30°C. Similar correlations were obtained for other columns of different sizes. While the

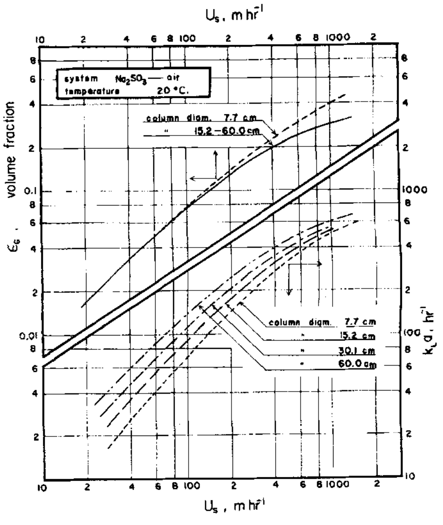

Fig. 9. Effect of column diameter on k_La and gas holdup in sodium sulfite solution.

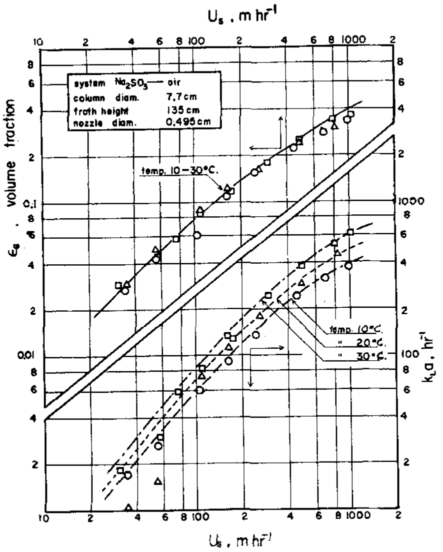

Fig. 10. Effect of temperature on k_La and gas holdup in 7.7-cm. column.

fractional gas holdup is not affected by temperature, k_La increases with increasing temperature, possibly owing to the variation in liquid diffusivity with temperature.

COMPARISON WITH DESORPTION DATA

That the rate of oxidation of sodium sulfite aqueous solutions by air or oxygen with cupric ions as catalyst is mainly controlled by diffusion in the liquid phase under certain conditions was demonstrated by Yoshida and others (5). In Figure 11 comparisons are made among the data on the fractional gas holdup and k_La for oxidation of sodium sulfite, desorption of oxygen from water, and desorption of oxygen from sodium sulfate solutions at 20°C. Values of k_La for sulfite oxidation are larger than those for desorption of oxygen from water but agree with those for desorption of oxygen from sodium sulfate solutions. This fact seems to indicate that the difference in the values of k_La between water system and electrolyte systems is mainly due to the difference in the interfacial area a, in other words due to the difference in bubble size. This is also seen from the difference in the gas holdup between water and electrolyte systems. Although gas diffusivity in an electrolyte solution is lower than in pure water, this effect seems negligible in the present case because of the low salt concentrations.

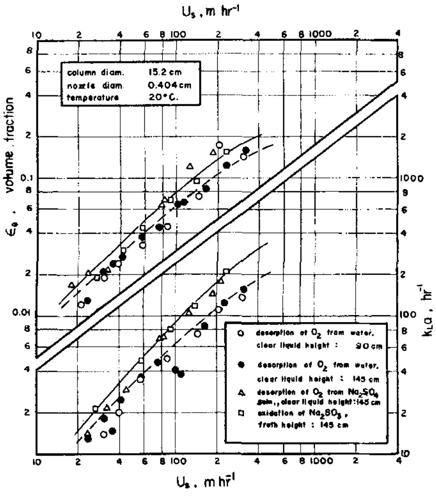

Fig. 11. Gas holdup and k_La for desorption of oxygen from water and sodium sulfate solution compared with those for sulfite oxidation.

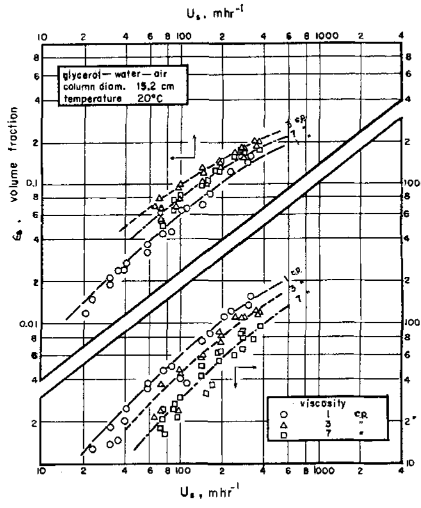

Fig. 12. Effect of liquid viscosity.

Shulman and Molstad (3) report the data on carbon dioxide absorption and desorption and hydrogen desorption into and from water supplied continuously to gas bubble columns. Because of the difference in the disperser design and the mode of operation, it is difficult to compare their data with the present data. However, their data for the 4-in. column at a liquid rate of 3,000 lb./hr./sq. ft. seem to agree approximately with the present data for the 15.2-cm. column, except at higher gas velocities. In his investigation of mass transfer characteristics Shulman observed two different regions depending on the gas rate. This was not revealed in the present work.

EFFECT OF LIQUID VISCOSITY

The effect of liquid viscosity was studied by performing experiments on the desorption of oxygen from aqueous solutions of glycerol at 20°C. in the 15.2-cm. column. As seen from Figure 12, the values of k_La decreases with increasing viscosity, probably owing to the decrease in liquid diffusivity with increasing viscosity. The fractional gas holdup varies with liquid viscosity in an irregular manner as shown in Figure 12.

CONCLUSIONS

Further work needs to be done with gas bubble columns on the extent of mixing in both gas and liquid phases, the magnitude of the interfacial area, the effects of liquid physical properties such as viscosity and surface tension, and continuous operation with respect to liquid.

ACKNOWLEDGMENT

Assistance of A. Okutsu, T. Sato, M. Hattori, Y. Nakai, and J. Watanabe in the experimental work is appreciated. This work was supported in part by a research grant from the Ministry of Education, Japan. Fujinagata Engineering Company donated the 60-cm. column and its accessories.

NOTATION

a = gas-liquid interfacial area, sq. m./cu. m. of aerated liquid
C = concentration of sodium sulfite, kg.-moles/cu. m.
H = Henry's law constant for oxygen, kg.-moles/cu. m./atm.
K_L = overall coefficient of mass transfer based on liquid composition, m./hr.

k_L = liquid-phase mass transfer coefficient, m./hr.
p = partial pressure of oxygen in the gas phase, atm.
U_B = average rising velocity of gas bubbles, m./hr.
U_S = superficial gas velocity, m./hr.
Z_F = height of aerated liquid, cm.
Z_L = height of clear liquid, cm.
ϵ_G = average fractional gas holdup, dimensionless
θ = time, hr.

LITERATURE CITED

1. Datta, R. L., D. H. Napier, and D. M. Newitt, *Trans. Inst. Chem. Engrs.*, 28, 14 (1950).
2. Fair, J. R., A. J. Lambright, and J. W. Anderson, *Ind. Eng. Chem. Process Design and Development*, 1, 33 (1962).
3. Shulman, H. R., and M. C. Molstad, *Ind. Eng. Chem.*, 42, 1058 (1950).
4. Van Krevelen, D. W., and P. J. Hoftijzer, *Chim. Industr. XXI Cogr. Int. Chim. Industr.* p. 168 (1948).
5. Yoshida, F., A. Ikeda, S. Imakawa, and Y. Miura, *Ind. Eng. Chem.*, 52, 435 (1960).

Manuscript received April 13, 1964; revision received July 13, 1964; paper accepted July 17, 1964. Paper presented at A.I.Ch.E. Buffalo meeting.

Heat Transfer Characteristics of Concurrent Gas-Liquid Flow in Packed Beds

VERN W. WEEKMAN, JR., and JOHN E. MYERS
Purdue University, Lafayette, Indiana

The past decade has seen a proliferation of processes in which gas and liquid react in two-phase flow through vessels packed with catalyst particles. In order to ensure adequate throughput, most of these reactors operate with concurrent flow as opposed to countercurrent flow, for which throughput is severely restricted by flooding. Since these two-phase reaction processes are very often exothermic (that is hydrogenation and oxidation), it is necessary to transfer heat from the process. At the high temperatures, high pressures, and corrosive conditions commonly encountered in such two-phase catalytic reactions, the necessary heat-exchange surface may represent a substantial portion of the reactor section cost. When large heats of reaction must be transferred, the catalyst bed will commonly be split into separate vessels with interstage heat exchangers for cooling. Because of the high cost of such heat-exchanger surface, it might be economically more desirable to exchange heat directly in the reactor, provided the necessary design data were available.

It would appear that careful design is required in gas-liquid reactors to ensure proper temperature control and to reduce the capital cost of the equipment. Few, if any, serious attempts have been made to study heat transfer from packed beds operating with gas-liquid flow. In fact only one paper (*1*) has been published dealing with pressure drop and liquid holdup over a wide range of concurrent, two-phase, packed-bed flow conditions. In the present study, experimental data were obtained for heat transfer to beds, packed with various sizes of spheres, through which the two-phase air-water system was flowing concurrently.

APPARATUS

The flow equipment used in the experimental program was described in a previous paper (*10*) and also in detail elsewhere (*2*). The heat transfer section was attached as an integral part of the packed column and consisted of machined pure copper tube surrounded by a steam chest. Thermocouples were emplaced in this copper wall at various axial and radial positions to enable the average wall temperature to be calculated. All thermocouples were calibrated against an N.B.S. standard thermometer. Steam could be introduced at different positions in the chest to help flatten any temperature profile in the copper wall.

An exit mixing device was designed (*2*) to provide an accurate mixing cup temperature of the outlet gas and liquid stream. It consisted of alternating donut types of baffles followed by a short packed section. Thermocouples were located at various radii inside the small packed section.

V. W. Weekman, Jr., is with the Socony Mobil Oil Company, Paulsboro, New Jersey.