Effect of Bed Diameter on Mass Transfer Factors in a Liquid Fluidized Bed

DERAN HANESIAN and VINCENT J. OPALEWSKI[1]
Newark College of Engineering, Newark, N. J.

The effect of bed diameter on mass transfer factors in liquid fluidized beds was investigated. Studies were made in beds ranging in diameter from 2 to 10 inches. A liquid-solid system involving the dissolution of calcium sulfate dihydrate by water was used. Mass transfer factors were correlated with modified Reynolds number. The data indicate that the mass transfer factor decreased as the bed diameter increased at a given Reynolds number. The effect is less pronounced at the higher Reynolds numbers.

THE SCALE-UP of laboratory and pilot plant data for fluidized systems has been difficult and many times the large scale units never met the performance demonstrated in smaller models. In this study, the mass transfer factor, J_D, was investigated for a fluidized system as a function of the modified Reynolds number and column diameter to aid in future scale-up problems.

The concept of the mass transfer factor, J_D, was first introduced by Colburn (*3*). Using the concept of the mass transfer factor, J_D, as a function of modified Reynolds number, N'_{Re}, Chu, Kalil, and Wetteroth (*2*) were successful in correlating the published data of previous investigations for both fixed and fluidized beds. Also pertinent to this study, was the work of McCune and Wilhelm (*8*) involving mass transfer between liquid and solid particles for both fixed and fluidized beds.

Few published data, however, exist pertaining to the effect of bed diameter in a fluidized system. Most of the early studies were in bed diameters ranging from 1.75 to 4.0 inches. Parent, Yagol, and Steiner (*10*) studied fluidization of fine mesh graphite and coke (100-mesh) in 2- and 4-inch columns. They found that at low Reynolds numbers, materials which were fluidized well in a 4-inch column, slugged in a 2-inch unit.

In other areas, the effect of diameter on mass transfer was noted. Barker and Treybal (*1*) measured mass transfer coefficients for the dissolution of boric acid in stirred vessels and reported an effect of column diameter. Liebson and Beckman (*6*) studied the effect of packing size and column diameter in liquid-liquid extraction and found both factors affected the height of a transfer unit.

In summary, therefore, apparently diameter effects in mass transfer have been observed for a variety of systems. Published data relating the effect of column diameter on mass transfer in a fluidized system, however, have not been available.

EQUIPMENT

Fluidizing Column. Three assemblies were used which covered the bed diameter ranges of 2 inches, 4 to 6 inches, and 8 to 10 inches. Each was a similar construction. Two plates were drilled and tapped at the centers with appropriate pipe sizes. A set of threaded tie rods completed the assembly. The lengths of glass pipe were inserted between the plates which were compressed by the tie rods. With this equipment, change-over to a different pipe size necessitated only the loosening of the tie rods and displacement of the existing column with the new size.

The 2-inch column was made of heavy-walled glass tubing. The other columns were of standard Corning flange end glass pipe. The 2- through 6-inch units were of a total height of 2 feet. The 8- and 10-inch beds were 3 feet high.

Calming Section. The calming section was made of a mixture of copper and aluminum shot ranging in size from $\frac{1}{32}$ inch to $\frac{1}{8}$ inch. A 40-mesh Tyler Standard screen headed the calming section and segregated it from the fluidizing area.

Figure 1 shows the calming section assembly. In assembly, an 18-gage bare copper wire was inserted through the bottom flange, associated piping, and out the $\frac{1}{4}$-inch tubing to $\frac{1}{4}$-inch NPT fitting. A small circle of 40-mesh screening to hold the calming section in place was strung over the wire. The rubber gasket and glass pipe were next placed in position with the wire extending through them. The copper and aluminum shot, which actually made up the calming section, were poured into the glass pipe. The 40-mesh screen, which was carefully cut to conform to the inside of the glass pipe, was tied to the loose end of the wire and drawn half way down into the glass pipe. The balance of assembly was completed and water was

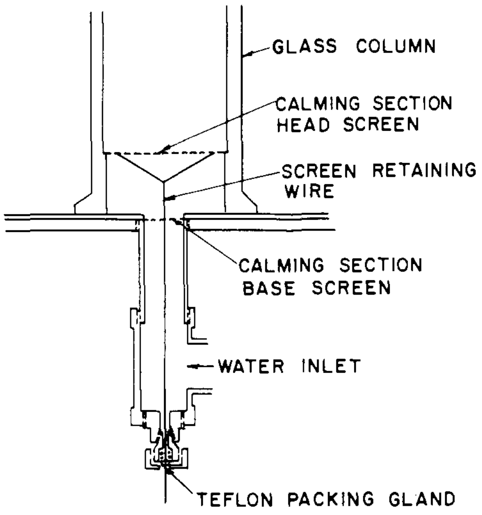

Figure 1. Calming section assembly

---

[1] Present address: Allied Chemical Corp., Whippany, N. J.

introduced at a velocity to fluidize the calming section. The screen was then pulled tightly into place as the water flow was reduced. The calming section in each case included the constricted area of the pipe and was of a height equal to the bed diameter.

Water Supply and Metering. The water supply for the system was a 6-inch water main which was not otherwise used during experimentation. Variations of static pressure were checked and none were seen within the sensitivity of the gage being used. The temperature of the water also was constant during the experiments.

Three Fischer-Porter rotameters were used at various stages of experimentation. These units had 100% flow ratings of 2.7, 8.8, and 14.6 gallons per minute at specific gravity of 1.0 and were calibrated using a minimum flow time of 3 minutes. These units had a high degree of response and were linear.

Temperature. Temperature of operation was measured at the bed outlet using a mercury capillary thermometer with a range of 0° to 50°C. at 1° per division. All samples were withdrawn at the overflow to the sewer.

MATERIALS AND PHYSICAL PROPERTIES

Selection. A liquid-solid system was chosen for experimental purposes. This system provided a minimum of bubbling, channeling, or particle agglomeration in comparison with a gas-solid system. The solution of a material into water appeared to be the least complex from a standpoint of operation, analysis, and interpretation.

The bed material had to have a low but measurable solubility in water, a specific gravity greater than one, and be reasonably tough so that material would not be lost through attrition and elutriation during experimentation. Two major practical considerations were toxicity, since dilute solutions were to be rejected to the sewer, and availability. Approximately 1 cubic foot of closely sized particles would be required for experimentation.

Calcium sulfate dihydrate fulfilled experimental and nontoxic requirements. It was easily and inexpensively available in an anhydrous form as plaster of Paris and, hence, was selected as the fluid bed material.

Preparation. Considerable difficulty is encountered in hydrating a material like calcium sulfate with any degree of reproducibility. Correlation of experimental data requires that the material display uniform characteristics throughout experimental work. In an effort to obtain a uniform material on a macro basis, the total raw material required was prepared in one step.

A charge of 130 pounds of plaster of Paris (unlimed or otherwise modified), 54 pounds of water, and 0.1 pound of concentrated sulfuric acid was mixed in a sigma blade dough mixer for 4 minutes. The slight excess of sulfate ion ensures an extremely hard hydrate form upon curing. The mixture was then dumped on a Plexiglas sheet and spread out to a thickness of approximately 2½ inches and partially cured for ½ hour. The material was then broken up into smaller sheets and allowed to cure at laboratory temperature and humidity conditions for 3 days.

The sheets were broken up and passed through a Wylie mill for particle reduction. A vibrating screen consisting of 16- and 18-mesh U.S. Standard series was fed with the mill discharge. Particles greater than 16 mesh were reground and below 18 mesh were discarded. In this manner 86 pounds of material were produced which had a bulk density of 59.8 pounds per cubic foot.

A statistical 1-pound sample was taken using a sampling weir and the balance was charged into a 12-inch diameter steel fluidizing column. The material was fluidized with water until the overflow contained no indication of fines. The material was dropped from the bed and stored in polyethylene-lined, 5-gallon cans in the presence of water. Storage in water ensured that the somewhat porous material was in equilibrium with respect to moisture, eliminating inconsistent apparent densities.

Characteristics. A statistical sample of the washed particles, which were to be used for experiments, was taken. This material was used to determine the following characteristics: voidage per unit bulk volume, particle diameter, surface area per unit bulk volume, diffusion coefficient, and chemical purity. The results are listed in Table I.

Voidage. A special technique had to be used to determine the voidage characteristics of the material without the apparent error caused by porosity. A sample of the material was allowed to drain for about 5 minutes on a screen. It was then poured into a 2-inch diameter glass tube 2 feet long which was fitted with a stopper and a screen at the bottom. The particles were fluidized slightly with an air stream, and the wet-bulb and dry-bulb temperatures of the exhaust gas were monitored. Initially the humidity began to drop, the air stream was turned off, and the bed was allowed to settle. The air hose was removed and replaced with a length of tubing connected to a buret. Water was added to the system through the buret until it just touched the bottom of the particle bed. The process was then continued with measured quantities of water until the bed was just immersed. The static bed height, volume of required water, and bore diameter of the glass tube were used to determine the voidage characteristics of the material. A check, using mercury displacement with a fixed bed, gave comparable values.

Surface Area and Particle Diameter. A dry sample of the material which had been conserved earlier was passed through a statistical sampling box a number of times until a representative sample of 185 particles was left. The thickness of each of these was measured with a micrometer. The arithmetic average of these values was 0.0306 inch. A small sample of wet particles was partially dried by the method described earlier. Three groups of 500 of these particles were weighed, and their volume was measured by water displacement.

The particles were assumed to be square prisms (*8*). Knowing the average thickness, volume, and mass per unit particle, the average length and width were calculated. These values were used to calculate the approximate surface area. The arithmetic mean of the length, width, and thickness values was used as the particle diameter for calculating the Reynolds number.

Diffusivity of Calcium Sulfate in a 0.01248*M* Aqueous Solution. The calculation of the Schmidt number requires knowledge of the diffusivity of the material being investigated. In the fluidization experimental analysis, the authors assumed that the molecular layer immediately adjacent to the particle was saturated with respect to calcium sulfate. All experimental runs were made at 10° ± 1°C. Therefore,

Table I. Characteristics of $CaSO_4 \cdot xH_2O$ Particles

Physical	
Apparent particles specific gravity	2.278
Particle mass, average 1,500 part., g./part.	2.913×10^{-3}
Particle thickness, average 185 part., inches	0.0307
Particle diameter, average, inches	0.0439
Particle surface area sq. in.	0.0113
Volume fraction part. cu. ft./bulk cu. ft.	0.420
Surface area/bulk volume sq. ft./cu. ft.	732
Chemical	
Degree of hydration, moles H_2O/mole $CaSO_4$	1.97
Emission spectrophotometry analysis	
Ca as $CaSO_4$	99.8%
Si	1 p.p.m.
Mg	Trace

Table II. Diffusivity Cell Standardization

Fixed Data

Cell volume	57.78 sq. cm.
Standardization HCl	0.5000*N*
Volume water in beaker	57.78 cc.
Time period for gradient formation	2 hours
Diffusivity of HCl at 10° C. and 0.5000*N*	7.56×10^{-2}/cm. sq./hr.
Operating temperature	10° C.

Experiment	1	2	3
C_i, initial concn. HCl, g. moles/liter	0.5000	0.5000	0.5000
C_B, concn. in beaker at 2 hours	0.0061	0.0055	0.0062
C_O, $C - C_B$	0.4939	0.4945	0.4938
C_F, final concn. in beaker	0.0521	0.0575	0.0637
t_E, exposure time, hours	16	18	20
K, cell constant	0.0849	0.0845	0.0857

this narrow range was examined with respect to calcium sulfate diffusivity.

The technique of McBain and Lui (7) was used for this determination. A diffusion cell was made from a Corning Glass Works 60-ml. fritted glass filter funnel. The glass membrane was 1.5 inches in diameter and was coded medium porosity by the manufacturer.

The cell was completely cleaned by drawing chrome-sulfuric acid through it with a vacuum pump into a trap, then thoroughly rinsed with distilled water, and dried in a vacuum oven. The cell was weighed, filled completely with degasified distilled water and reweighed to determine its actual volume.

A small unit was fabricated which would hold the diffusion cell rigid and level in the proper position over a constant temperature water bath. The cell was mounted on the unit and leveled with a mercury meniscus. Hydrochloric acid, 0.500*N*, was drawn into the cell, the stopcock was closed, excess was cleaned from the membrane, and the cell was put in contact with a beaker of degasified water equivalent to its own volume. The beaker of water was in a constant temperature bath, and the balance of the unit was enclosed in the chamber which housed the bath. Plastic film was used to seal the gap between the cell and the beaker.

A 2-hour period was allowed for a diffusion gradient to form, and a fresh beaker of water was exposed to the membrane. After 16 hours, the two were separated, and the liquids in each were titrated with 0.500*N* NaOH. This process was repeated three times at three different exposure times, and the relationship proposed by McBain and Lui (7) was used to standardize the cell. The experimental cell constants were 0.0849, 0.0845, and 0.0857 with D in square centimeters per hour (Table II).

The series was repeated using calcium sulfate solutions which were 95% saturated at the temperature being investigated. Since the diffusivity of the $CaSO_4$ was estimated to be lower than that of HCl, the procedure was modified to a 5-hour initial exposure for diffusion gradient formation, and a 24-hour exposure before separation and analysis.

The resultant solutions were analyzed by the disodium EDTA titrimetric technique described in the next section. Utilizing the cell constant determined with hydrochloric acid and the relationship proposed by McBain and Lui (7), the diffusivity of a concentrated solution of calcium sulfate was 0.490, 0.496, and 0.485 sq. cm. per day at 9°, 10°, and 11° C., respectively. The range of values is well within experimental error. Therefore, the average 0.490 sq. cm. per day was used to describe the temperature range (Table III).

Analytical. "Scott's Standard Methods of Chemical Analysis" (4) describes a technique for determining total magnesium and calcium content of water by titration with disodium ethylenediaminetetraacetic acid using Eriochrome Black T as an indicator. The calcium dissolved in the fluidized bed could be determined as the difference in total magnesium and calcium content in the exit and entrance streams.

The recommended procedure had to be modified slightly to sharpen the end point which was wine red to pale blue. Buffer and indicator were increased slightly from 0.5 to 0.6 ml. and from 6 to 7 drops, respectively. The published procedure called for a 50-ml. sample. It was necessary

Table III. Determination of Diffusivity of $CaSO_4$

Fixed Data

Cell volume	57.78 cc.
Volume water in beaker	57.78 cc.
Time period for gradient formation	5 hours
K, cell constant with D in sq. cm./hr. and t_E in hours	0.0850

Experiment

C_i, initial concn. $CaSO_4$ g. moles/liter	0.0125	0.0125	0.0125
C_B concn. in beaker at 5 hours	2.1×10^{-5}	2.3×10^{-5}	1.8×10^{-5}
C_O, $C_i - C_B$	0.01248	0.01248	0.01248
C_F, final concn. in beaker	0.00050	0.00052	0.00051
t_E, exposure time, hours	21	21	21
T, operating temperature, ° C.	9	9	9
D, diffusivity sq. cm./hr.	0.0204	0.0206	0.0202
D, diffusivity sq. cm./day	0.490	0.496	0.485

to dilute a calcium chloride standard, 0.4 mg. of Ca per ml., by 5 ml. of standard per 45 ml. of distilled water. This also was done in order to sharpen end point characteristics.

Purity and Solubility. The source of raw material, commercial plaster of Paris, made a purity analysis mandatory. The technique used was emission spectroscopy. A portion of <100 mesh material, conserved from the grinding operation, was kilned at 500° C. A 1.5-mg. sample was fixed in a 1-gram zinc oxide matrix and examined via emission spectroscopy.

The results of the analysis indicated the material was of high quality. There was 99.8% calcium as sulfate, 1 p.p.m. silica, and a trace of magnesium. A second test checked these values.

Four samples of material (50 grams), which had been cured at laboratory conditions, were dried to constant weight at 200° C. to determine the degree of hydration. The results indicated $CaSO_4 \cdot 1.97H_2O$. The authors assumed that there would be no significant error in using solubility values published in the "International Critical Tables" (5) for $CaSO_4 \cdot 2H_2O$.

Density and Viscosity of Saturated Calcium Sulfate Solution at 10° C. The density and viscosity of a 0.01418*M* aqueous solution of calcium sulfate, which is saturated at 10° C., were determined at 10° C. using a hygrometer and an Ostwald viscometer.

The density of the solution was 62.67 pounds per cubic foot in comparison with 62.42 pounds per cubic foot for water at the same temperature. Using distilled water as a standard, the viscosity of the calcium sulfate solution was 3.177 pounds per foot-hour in comparison with 3.17 pounds per foot-hour.

These values for the calcium sulfate solution represented the condition of the stagnant film about the particle and were, therefore, used in calculating the Schmidt number.

EXPERIMENTAL

Number and Type of Experiments. The experiment was designed to investigate the effect of bed diameter on mass transfer factors over a range of modified Reynolds numbers. The bed diameters used were 2, 4, 6, 8, and 10 inches.

With each column diameter, three sets of runs were made in which the mass rate of water was varied through seven to eight evenly distributed levels. These runs were duplicates of each other, since the same range of modified Reynolds numbers was covered.

The lowest level in each case was the minimum rate required to cause bed fluidization. The maximum velocity was limited by column height in the 2- through 6-inch beds and by sewer capacity in the 8- and 10-inch beds.

A special set of runs was made on the 2-inch column where static bed height was varied through five levels. The static bed height in the experiments had to be kept in a range of 1.5 to 2 inches. When heights exceeding that were used at low Reynolds numbers, the effluent from the bed approached 75% saturation with the material being detected.

Start Up. The bed, with calming section, was assembled as described in the experiment section. The flow of water was started in the equipment at least one hour prior to experimentation in order to ensure that the system was at complete equilibrium with respect to temperatures.

The wet particles were dumped into a graduated cylinder, allowed to settle, and then tapped until they settled to a minimum volume. In each case, the volume added was the quantity required to give a static bed of the desired height.

The materials were charged, with the water off and the column full, through a fitting provided in the top plate. The system was closed, and the bed was fluidized for 1 minute and allowed to settle to check static bed height.

Each run was started at minimum fluidization velocities. At low velocities, 7 minutes were allowed for the system to reach equilibrium. As velocities were increased, equilibrating times were decreased to 2 minutes. These times represent five to seven column turnovers.

Sampling. At each water rate, three 150-ml. samples were collected at the outlet to the sewer. Standard 24/40 glass stopcocks packed with borosilicate glass wool were used to filter each sample immediately after it was taken. In this manner, any elutriated particles would not be in the sample.

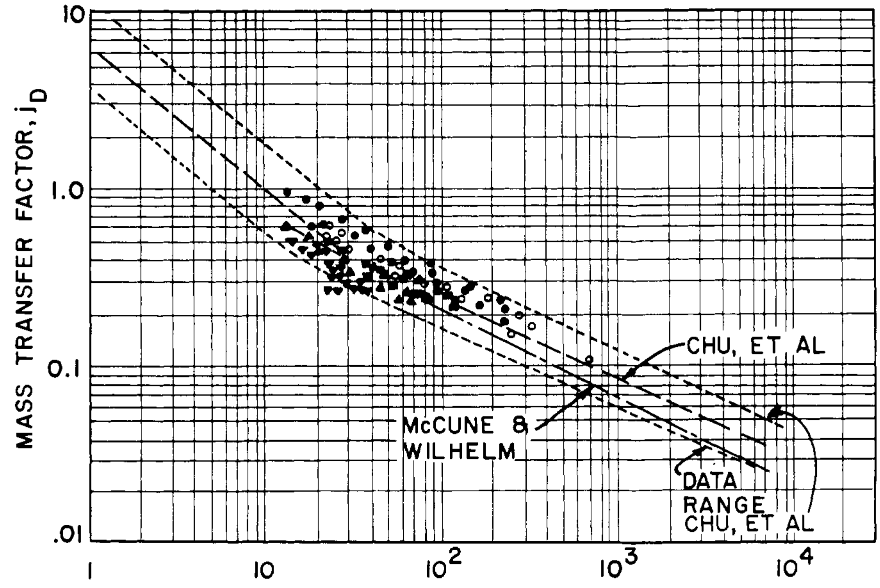

Figure 2. Mass transfer factor vs. modified Reynolds number

● 2-inch diameter bed
○ 4-inch diameter bed
■ 6-inch diameter bed
▲ 8-inch diameter bed
▼ 10-inch diameter bed

Operation. The expanded bed height was noted at the time of sample collection. Immediately after the last sampling at each water rate, the flow of water was turned off, and the static bed height was rechecked. When the static bed height had reduced sufficiently to equal a loss in bed mass of 3 to 5%, the run was terminated, and the bed was emptied by elutriation. A fresh and equivalent charge of solids was added, the start-up procedure was repeated, and experimentation was resumed at the last water rate. At least seven points were taken evenly distributed between minimum and maximum bed expansion for each particular pipe size.

Temperature and flow rate were constantly monitored during the run and no fluctuations were observed. In the experiments with the 8- and 10-inch beds, the modified Reynolds numbers were limited to 70 and 35, respectively. The sewer was undersized at the higher water rates.

Observations. The quality of fluidization achieved was surprisingly good considering the uniformity of particle size. At minimum fluidization velocities, channeling effects were observed to a slight degree. These effects ceased with a slight increase in the velocity of the liquid stream.

The top surface of the fluidized bed was very uniform. No bubbling or inner channeling disrupted its surface. This facilitated measurement of the expanded bed height.

At high velocities, the calming section particle bed tended to expand, slightly warping the retaining screen approximately 2° from horizontal. This did not detract from the experiment or introduce a significant error. Complete experimental details and experimental data are available (*9*).

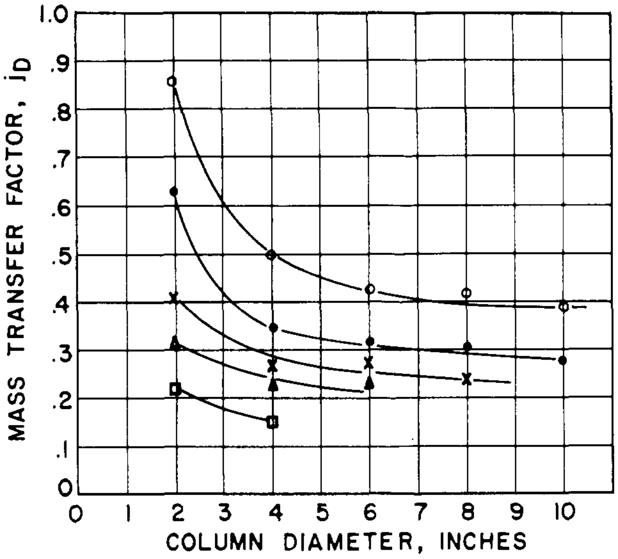

Figure 4. Effect of column diameter on mass transfer factor in a fluidized bed

Modified Reynolds number

○ 19.6–24.7
● 34.9–36.5
× 60.5–36.5
△ 81.0–97.0
□ 223–241

RESULTS

The data for this study, J_D *vs.* modified Reynolds number, are shown in Figure 2. Also shown are the data reported by Chu, Kalil, and Wetteroth (*2*) and McCune and Wilhelm (*8*). The data from the latter study, which was also based on a liquid-solid fluidized system, were recomputed in accordance with the method proposed by Chu, Kalil, and Wetteroth (*2*).

In general, the data from this study agree well with measurements of previous investigators. To show the effect of column diameter, the data for the two extreme diameters, 2- and 10-inch, are shown in Figure 3. At comparable modified Reynolds number, the 2-inch column shows higher mass transfer factors. Higher modified Reynolds numbers for the 10-inch column could not be obtained because of physical limitations in the laboratory.

Figure 4 shows the mass transfer factor correlated with column diameter for various Reynolds numbers. A marked decrease in mass transfer factor occurs with rise in diameter for the smaller units (below 6 inches). The effect of column diameter is also more marked at lower modified Reynolds numbers.

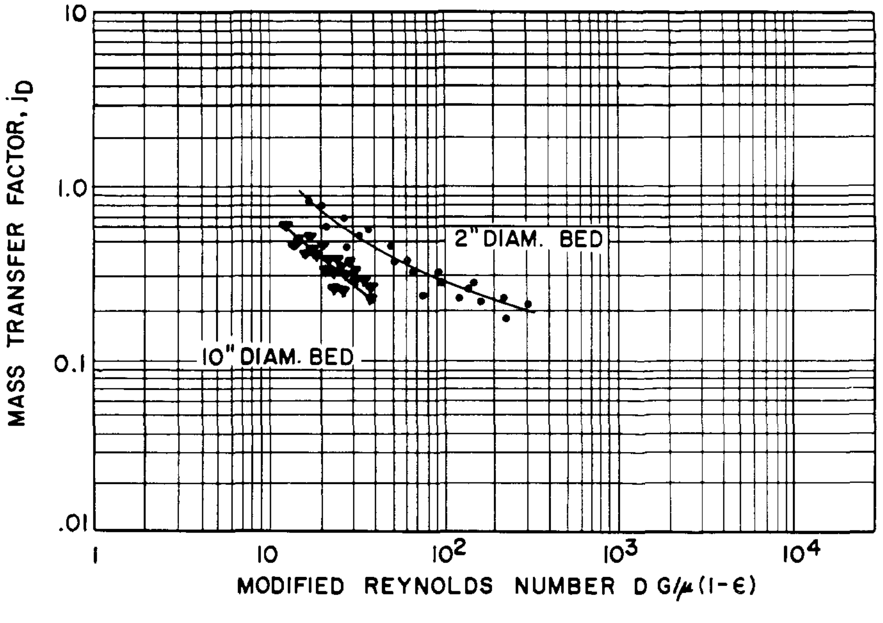

Figure 3. Mass transfer factor *vs.* modified Reynolds number

Table IV. Effect of Bed Height on Mass Transfer Coefficients

(2-inch column)

Static Bed Height, Inches	Static L/D Ratio (Nominal)	Expanded Bed Height, Inches	Expanded L/D Ratio (Nominal)	N'_{Re}	Mass Transfer Coefficient, Lb. $CaSO_4$/Hr. Sq. Ft., Lb. $CaSO_4$/Lb. H_2O
1.00	0.5	1.42	0.71	46	33.7
1.25	0.625	1.87	0.94	54	31.4
1.50	0.75	2.20	1.10	50	35.0
2.00	1.00	2.88	1.44	42	33.8
2.25	1.125	2.97	1.49	43	34.7

The effect of column diameter on the mass transfer factor is believed to be a wall effect on fluidization. Zenz and Othmer (*11*) indicate that the mass transfer factors for fluidized beds generally lie below those for fixed beds (an upper limit). As the velocity increases, the data from fluidized beds approach a lower limit. This lower limit is the mass transfer factor for single particles. For small diameter columns, a greater fraction of the solid is influenced by the wall than in larger diameter columns. Hence this wall effect may give results more related to fixed beds.

Similarly, fixed bed effects would be more nearly approached at lower Reynolds numbers and the mass transfer factors would again be closer to those for a fixed bed, particularly in the smaller diameter columns. Both of these tendencies are shown in Figure 4.

In this study, although the column diameter increased, the static bed height was almost constant for all runs to avoid saturating the water with the solute. Zenz and Othmer (*11*) indicate that this fact is a problem in mass transfer studies in fluidized systems. Since the static height remains constant and bed diameter increased, the L/D ratio from column to column changed.

To determine the effect of L/D ratio on the mass transfer factor, runs were made on the 2-inch column at various static bed heights. The data are shown in Table IV. The data obtained in the 2-inch column show that the static bed height did not influence the mass transfer coefficient. These runs indicated that the variation of the mass transfer factor at a given Reynolds number was caused by a change in the diameter and not a change in the L/D ratio.

Based upon the results of this study, the authors concluded that the mass transfer factor is markedly reduced as the column diameter increases in this liquid, particulate, fluidized system. Beyond 4- to 6-inch diameter columns the effect is less pronounced. The effect is also less pronounced at higher Reynolds numbers. The change in the mass transfer factor with column diameter is believed to be a wall effect.

NOMENCLATURE

J_D = mass transfer factor, $k/G[\mu_F/\rho_F D_F]^{2/3}$
k = liquid film mass transfer coefficient, lb. $CaSO_4$/hr., sq. ft., lb. $CaSO_4$/lb. H_2O
A_c = column cross section, sq. ft.
G'_F = mass rate of liquid stream, lb./hr.
M_F = mean molecular weight of stream
μ_F = viscosity of liquid film about particle, lb./ft.-hr.
ρ_F = density of liquid film about particle, lb./cu. ft.
D_F = diffusion coefficient, sq. ft./hr.
N'_{Re} = modified Reynolds number, $D_pG/\mu\ (1 - \epsilon)$
D_p = particle diameter, ft.
G = mass velocity of liquid, lb./hr. sq. ft.
μ = viscosity of liquid stream, lb./ft.-hr.
e = expanded bed void fraction

LITERATURE CITED

(1) Barker, J.J., Treybal, R., *A. I. Ch. E. J.* **6**, 289 (1960).
(2) Chu, J.C., Kalil, K., Wetterroth, W.A., *Chem. Eng. Progr.* **49**, 141 (1953).
(3) Colburn, A.P., *Trans. Am. Inst. Chem. Engrs.* **29**, 174 (1933).
(4) Furman, N.H., "Scott's Standard Methods of Chemical Analysis," 6th ed., Vol. **I**, p. 270, Van Nostrand, Princeton, N. J., 1962.
(5) "International Critical Tables," McGraw-Hill, New York, 1929.
(6) Liebson, I., Beckman, R.B., *Chem. Eng. Progr.* **49**, 405 (1953).
(7) McBain, J.W., Lui, H.L., *J. Am. Chem. Soc.* **53**, 59 (1931).
(8) McCune, L.K., Wilhelm, R.H., *Ind. Eng. Chem.* **41**, 1124 (1949).
(9) Opalewski, V.J., M.S. thesis, Newark College of Engineering, Newark, N. J., 1965.
(10) Parent, J.D., Yagol, N., Steiner, C.S., *Chem. Eng. Progr.* **43**, 429 (1947).
(11) Zenz, F.A., Othmer, D.F., "Fluidization and Fluid-Particle Systems," p. 471, Reinhold, New York, 1960

RECEIVED for review October 7, 1966. Accepted March 29, 1967.