Lateral Thermal Dispersion in Gas-Liquid Cocurrent Downflow Through Packed Beds

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The characterization of heat transfer in packed beds is relevant in many practical applications, especially those related to packed-bed reactors, since it has a strong bearing on conversion, stability, and catalyst deactivation. A large number of heat-transfer studies in packed beds under one-phase flow conditions are reported in the literature. A review of such works has been presented by Lemcoff et al. (1990). In contrast, relatively few studies have been reported concerning heat transfer in packed beds under two-phase flow conditions, despite its importance in applications in which exothermic reactions are involved, such as hydrotreatment of petroleum fractions in trickle-bed reactors.

One of the first studies on heat transfer in gas-liquid flow through packed beds was carried out by Weekman and Myers (1965). They heated the wall of a cylindrical packed bed to a constant temperature, and then measured radial temperature profiles. The data were analyzed by means of a one-equation model, which assumed implicitly that local thermal equilibrium existed among the phases so that a single temperature could be used as dependent variable. This model allows them to evaluate the radial effective thermal conductivity, which they correlated as the sum of three contributions: a static effective thermal conductivity, and two thermal dispersion coefficients arising from the contribution of gas and liquid flows. Hashimoto et al. (1976) performed an experimental study similar to the work by Weekman and Myers but using wider ranges of gas-and liquid-flow rates. Specchia and Baldi (1979) employed once again the same experimental technique to find correlations for the radial effective thermal conductivity and the wall heat-transfer coefficient.

Crine (1982) criticized the method of determining effective thermal conductivities by heating the walls of the packed bed. It was argued that this procedure gives rise to a strong wall region influence because the local liquid velocity in the vicinity of the wall depends on radial position due to the increase in local porosity that packed beds exhibit close to the wall. This change in local porosity must affect the local values of the effective thermal conductivity and, therefore, the values obtained by one-equation models, which are taken to be uniform. As an alternate experimental procedure, Crine injected hot water into a central region of a cylindrical packed bed while injecting cold water into the remaining annular region. The radial temperature profiles obtained in this manner were then analyzed by means of a one-equation model.

More recently, Lamine et al. (1992) measured radial effective thermal conductivities and wall heat-transfer coefficients in gas-liquid cocurrent upflow through packed beds. They used the wall heating technique. Their results show a strong dependence of the effective thermal conductivity on the flow regime.

Most of the previous works in the literature have used water or aqueous solutions as liquid phase in the experiments to characterize effective thermal conductivities. Since the experiments involve temperature gradients, local variations in the viscosity of the liquid phase occur. These viscosity changes might alter the hydrodynamics of the process by inducing variations in local velocity or liquid holdups, as was shown by Sáez and Ramírez (1993). The presence of liquid holdup or velocity profiles might have a sizable influence on the effective thermal conductivity, which is not accounted for in conventional models.

In this work we present an experimental study of lateral thermal dispersion in gas-liquid downflow through a packed bed for the cases of liquid flowing in the presence of a stagnant gas phase, and two-phase cocurrent flow. In the experiments, temperature profiles are generated by inducing a lateral step change in temperature at the entrance of the bed, which avoids wall heating. The evolution of the temperature profiles is determined by measuring radial temperature variations at various axial positions. The effect of variations of liquid viscosity with temperature are minimized by adjusting the viscosity of the liquid at the entrance of the bed so that it is approximately uniform despite temperature variations, thus avoiding the effect of viscosity changes on velocity or holdup

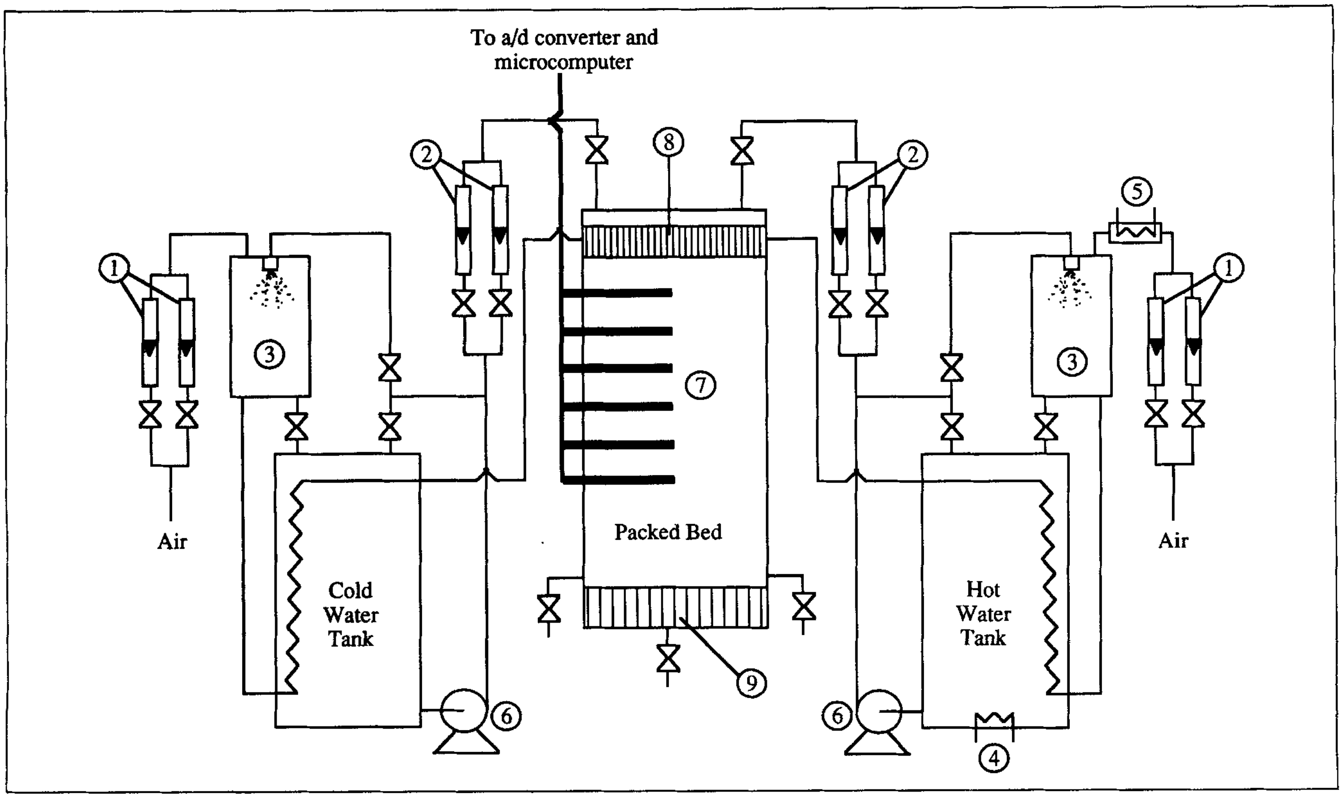


Figure 1. Experimental apparatus

profiles. The temperature profiles are employed to determine lateral effective thermal conductivities, and the effect of various independent variables on this parameter is investigated.

Experimental Studies

The experimental setup used in this work is shown in Figure 1, and the corresponding legend is presented in Table 1. The packed bed was made of Plexiglas. It has a square cross section with internal dimensions 21 cm × 21 cm and a packing length of 106 cm. The bed consisted of disordered packings of monodisperse-size spheres. Different particle sizes and materials were used. Their diameters (d), specific heat (c_{ps}), density (ρ_s) and thermal conductivity (k_s) are presented in Table 2. The bed porosities reported (ϵ) were measured by determining the weight of the packed bed full of water, and subtracting this weight from the weight of the particles.

Table 1. Legend for Figure 1

Item	Description
1	Air rotameters
2	Liquid rotameters
3	Air humidifiers
4	Liquid heater
5	Air heater
6	Pumps
7	Thermocouple probes
8	Inlet distributor
9	Outlet distributor

Lateral temperature profiles were generated by introducing liquid and fully-saturated gas into two separate chambers with different temperatures in the distributor at the top of the column. The distributor consisted of two parallel horizontal copper plates perforated with 798 holes with 0.79 cm pitch. Stainless steel capillaries, 0.8 mm in ID and 20 mm in length were inserted into the holes so that the capillaries were perpendicular to the copper plates. The tubes were soldered to the top plate, and a small tolerance was left between the outside of the capillaries and the bottom plate to allow for gas flow. The distributor was divided in two halves by a vertical Plexiglas wall. Liquid and air at about 45°C were introduced into one side of the distributor whereas liquid and air at about 20°C were fed into the other side. Two type E thermocouple probes were located above the packing section in the hot and cold sides, respectively. They confirmed that, under the conditions explored in our experiments, no appreciable heat transfer occurred between hot and cold fluids until the packing section was reached. This induced a lateral step change in temperature at the inlet of the packing section. The lateral

Table 2. Characteristics of the Packings

Material	d (cm)	ϵ	c_{ps} (J/kg·K)	ρ_s(kg/m^3)	k_s (W/m·K)
Urea-Formaldehyde	0.55	0.40	1.675	1,463	1.05
Urea-Formaldehyde	0.45	0.40	1,675	1,463	1.05
Urea-Formaldehyde	0.20	0.38	1,675	1,463	1.05
Glass	1.11	0.41	779	2,230	1.08
Glass	1.50	0.41	779	2,230	1.08
Glass	2.54	0.42	779	2,230	1.08

temperature profile was measured in each experiment at a fixed distance of 0.254 m downstream of the distributor after steady state was achieved. From this profile, the lateral effective thermal conductivity K^*_{yy}, was evaluated according to the procedure presented below. The analysis leading to this calculation neglects flow nonuniformities due to entrance effects. We verified this assumption in representative experiments by moving the axial position of the measurements to a distance of 0.320 m below the distributor. We found that the K^*_{yy} values calculated at the new axial location coincided, within experimental error, with those calculated at the first axial location.

The temperature probes used for measuring lateral temperature profiles had 14 thermocouples made of Teflon-covered fine-gauge thermocouple wire (type E). The thermocouples were inserted, with a spacing between any two of them equal to 0.95 cm, into holes drilled in a Plexiglas bar with a rectangular cross section of 0.79 cm × 0.32 cm. The thermocouples were cemented to the bar in such a way that the bead lay on the plane of the upper surface of the bar, which is perpendicular to the flow direction in the packed bed. The entire bar was inserted into an external rectangular phenolic-resin tube to maintain rigidity. The low conductivity of the phenolic resin avoids distortion of temperature profiles due to conduction along the probe. The external tube was soldered to a cylindrical brass tube which had two stop positions in the lateral direction. This arrangement made possible to move the entire probe laterally a distance equal to half the spacing between thermocouples so that 28 temperature values could be recorded in each experiment. The thermocouple readings were monitored in a microcomputer after the signal passed through an analog-to-digital converter.

Atmospheric air presaturated with water vapor was used as gas phase in all the experiments. The liquid phase was water, and aqueous carboxymethyl cellulose (CMC) solutions. The CMC used was a high molecular weight sodium salt from Sigma Corporation (CMC C-5013), and it was used to vary the viscosity of the liquid phase. The viscosity of the CMC solutions was measured in an Ostwald capillary viscometer.

Two types of experiments were performed: stagnant gas-phase experiments and two-phase flow experiments. In a stagnant gas-phase experiments liquid flows downward through the bed, but there is no net gas flow through the system. In the two-phase flow experiments cocurrent gas-liquid downward flow was established. Before each experiment, the column was fully wetted in order to ensure an initial uniform saturation of the packing. With this purpose, in the two-phase flow experiments, the system was taken to the pulsing flow regime before the experiment, whereas in the stagnant gas-phase experiments, the bed was flooded and then allowed to drain for 20 min.

Hysteresis in the liquid holdup with respect to changes in the phases flow rates has been reported for two-phase flow and stagnant gas-phase experiments (Kan and Greenfield, 1978; Levec et al., 1986, 1988); tests were done in order to ascertain whether the possible presence of hysteresis produced a measurable effect on the lateral effective thermal conductivity. These tests were performed with the 2-mm urea-formaldehyde particles. For a given set of operating conditions, the gas-flow rate was established and then the liquid-flow rate was increased to the desired value. Temperature profiles were recorded at steady state. Without interrupting the experiments, the liquid-flow rate was increased until pulsing flow was reached, and then it was decreased to the desired value, a point at which a new experiment to measure lateral temperature profile was performed. The values obtained for the lateral effective thermal conductivity in both cases showed no specific trend, and the differences were within the reproducibility limits. This indicates that the possible hysteresis under the conditions explored has no relevance in the measure of effective thermal conductivities.

Theoretical Aspects

Owing to the complexity of the local geometry of a multiphase system, the heat-transfer process is characterized by the spatial average temperature, defined by

$$\langle T \rangle = \frac{1}{V} \int_V T\, dV \tag{1}$$

where the averaging volume V should be large enough so that the average temperature field can be considered a point function in the continuum of the multiphase system. Inside the averaging volume, gas, liquid, and solid phases coexist. Under steady-state conditions, if the thermal conductivities of the phases do not differ by many orders of magnitude and the flow rates are not large enough to cause steep temperature gradients at the phase interfaces, the condition of local thermal equilibrium holds. In this case the heat-transfer process can be reduced to the description of the spatial average temperature. Details have been given by Whitaker (1986, 1991) on the specific constraints that must be satisfied so that the hypothesis of local thermal equilibrium holds for two-phase systems.

For steady-state and local thermal equilibrium conditions, the macroscopic thermal energy equation can be expressed as follows

$$(\rho_l c_{pl} \langle v_l \rangle + \rho_g c_{pg} \langle v_g \rangle) \cdot \nabla \langle T \rangle = \boldsymbol{K}^* : \nabla\nabla \langle T \rangle \tag{2}$$

where $\langle v_\alpha \rangle$ is the superficial velocity vector of phase α, $\boldsymbol{K}^*$ is the effective thermal conductivity tensor, and ρ and c_p are density and specific heat. The indices g and l denote gas and liquid phases, respectively.

Under the conditions explored in this work, the contribution of the gas to the heat capacity of the mixture can be neglected in Eq. 2. Considering that the velocity profile is one-dimensional leads to

$$\rho_l c_{pl} \langle v_l \rangle_z \frac{\partial \langle T \rangle}{\partial z} = K^*_{yy} \frac{\partial^2 \langle T \rangle}{\partial y^2} \tag{3}$$

where z is the longitudinal direction and y is the lateral direction over which temperature gradients are induced. We have neglected the axial effective dispersive term in Eq. 3; it was verified that its magnitude was always much lower than the lateral dispersive contribution. This differential equation was solved with the following boundary conditions

$$\langle T\rangle = T_C \qquad y\to\infty \qquad \forall z \tag{4}$$

$$\langle T\rangle = T_H \qquad y\to-\infty \qquad \forall z \tag{5}$$

$$\langle T\rangle = T_C \qquad y>0 \qquad z=0 \tag{6}$$

$$\langle T\rangle = T_H \qquad y<0 \qquad z=0 \tag{7}$$

where T_C and T_H are the temperatures of cold and hot fluids at the entrance of the packing section, respectively.

The application of boundary conditions (Eqs. 4 and 5) results from the observation that for all the temperature profiles measured, the temperatures close to the wall on each side of the bed were always equal to those at the entrance.

The solution of Eq. 3 subject to boundary conditions (Eqs. 4 to 7) is

$$\theta = \frac{1}{2}\,\mathrm{erfc}\left(\frac{y}{\xi}\right) \tag{8}$$

where

$$\theta = \frac{\langle T\rangle - T_C}{T_H - T_C} \tag{9}$$

$$\xi = \left(\frac{4K_{yy}^{*} z}{\rho_l c_{pl}\langle v_l\rangle_z}\right)^{0.5} \tag{10}$$

Given a lateral temperature profile at a specified axial position K_{yy}^{*} can be calculated by finding the best fit of the profile to Eq. 8. The best fit was found in this work by means of a golden section search method to minimize the mean-squared error.

Results and Discussion

Stagnant gas phase

In the experiments conducted under stagnant gas-phase conditions gravity was the driving force for the downflow of liquid since no pressure drop existed through the bed. In these experiments, the particle diameter was varied from 2 to 25.4 mm, whereas the viscosity of the liquid (water and aqueous CMC solutions) ranged between 0.9 and 5 mPa·s. For each particle diameter and liquid viscosity, the liquid flow rate was varied.

When the first experiments were performed with pure water, it was noticed that the lateral temperature profiles obtained were not centered in the packed bed, i.e., the inflection point of the profiles (see Eq. 8) did not coincide with $y=0$, but was shifted to the hot side of the bed. This shift indicates that the heat-transfer process is not uniform along the cross section (between hot and cold regions). This lack of uniformity results from effects induced by the change in water viscosity with temperature. The lateral temperature variation ranged from 20 to 45°C. In this range, the viscosity of water changes from 1 to 0.6 mPa·s. Under these conditions, calculations performed by Sáez and Ramírez (1993) have shown that a sizable change in liquid holdup with lateral position can be obtained which induces an appreciable change in lateral heat transfer. With the purpose of avoiding the presence of large viscosity profiles, in all subsequent experiments, the viscosity of the hot liquid at the entrance of the bed was raised to that of the cold liquid by the addition of small amounts of CMC. This procedure does not completely eliminate viscosity gradients since lateral temperature profiles, and, therefore, local viscosities, change with axial position. Besides, the lateral gradient in carboxymethyl cellulose concentration leads to lateral mass dispersion of the polymer. However, the effect of viscosity gradients was eliminated for all practical purpose since the lateral temperature profiles were perfectly centered in all experiments. It is worth mentioning that, as reported in the introduction, heat-transfer experiments in trickle beds have usually been performed using water as liquid phase, with temperature changes close to, and even larger than, those used in this work. However, in the majority of the experiments reported in the literature, temperature profiles in the bed are induced by heating the wall. In this case, there is no simple observation (such as a shift in an inflection point) that indicates the existence of a sizable viscosity gradient, so it is more difficult to assess the possible errors that this fact can have on calculated effective thermal conductivities and heat-transfer coefficients.

We studied the effect of liquid viscosity on lateral effective thermal conductivity. The results are presented in Figure 2. The Prandtl and Peclet number are defined as follows,

$$Pr = \frac{\mu_l c_{pl}}{k_l} \tag{11}$$

$$Pe_l = \frac{\rho_l c_{pl}\langle v_1\rangle d}{k_l(1-\epsilon)} \tag{12}$$

The expected increasing trend of lateral effective conductivity with Peclet number is also observed in Figure 2. The continuous increase indicates that the Peclet numbers used are high enough so that dispersion dominates over conduction in the range of Peclet numbers analyzed. The $Pr=6$ curve corresponds to water with equalized hot and cold side viscosities, whereas the other curves correspond to carboxymethyl cellulose solutions. The results show that at the same Peclet (i.e., the same superficial velocity), an increase in

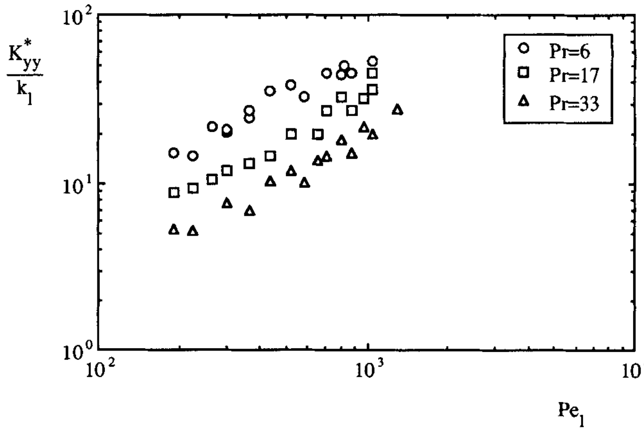


Figure 2. Effect of liquid viscosity on lateral effective thermal conductivity: stagnant gas phase, $d=4.5$ mm.

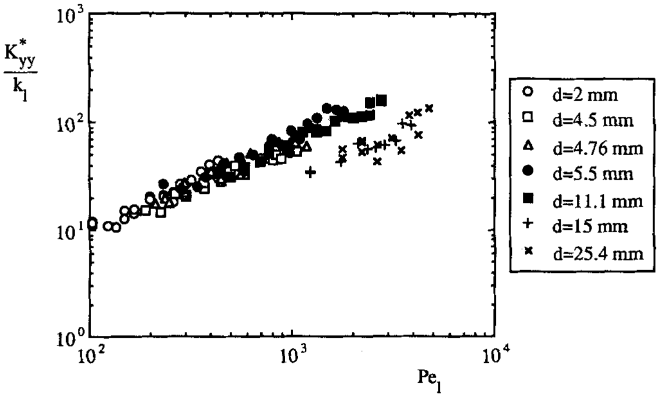


Figure 3. Effect of particle size on lateral effective thermal conductivity: stagnant gas phase, $Pr = 6$.

liquid-phase viscosity results in a decrease in lateral effective thermal conductivity. The liquid-phase viscosity affects thermal dispersion in two ways: first, changes in liquid viscosities affect the local velocity profiles in the liquid phase when inertial effects are present and, secondly, the viscosity alters the liquid holdup. The effect of the liquid viscosity on liquid holdup has been studied in the literature. For example, Sáez and Carbonell (1985) developed a correlation for predicting liquid holdup under stagnant gas-flow conditions in trickle beds, which took into account variations in liquid-phase properties. This correlation was developed under the same experimental conditions used in this work. The correlation shows that the liquid holdup increases with an increase in liquid viscosity unless inertia dominates. Therefore, for the same superficial velocity, an increase in viscosity leads to an increase in liquid holdup and, consequently, to a lower interstitial liquid velocity. The reduction in the level of the local velocity of the liquid phase implies a lower degree of lateral dispersion, observed in Figure 2.

We also studied the effect of particle size on lateral effective thermal conductivity. As the particle size is increased in these experiments, a point is reached for which the ratio of bed to particle size is small enough so that wall effects become noticeable. Figure 3 shows the results obtained. For small particles ($d \leq 11.1$ mm), the relation between lateral effective thermal conductivity and Peclet number is independent of particle size. The bed to particle size ratio for the 11.1 mm particles is $L_B/d = 19$, where L_B is the lateral dimension of the bed: $L_B = 21$ cm. For $d = 15$ mm ($L_B/d = 14$) and larger bed to particle sizes, the effective conductivity decreases. This change can be attributed to wall effects: it is well known that local porosities at distances within several particles of a flat wall are higher than in the bulk of the packing. With small bed to particle size ratios, the region of higher porosity occupies a sizable region of the packed bed. The larger porosity might result in larger liquid holdups than those expected for the same particle size in a large bed and this reduces the interstitial velocity of the liquid, resulting in lower lateral effective conductivities.

If we consider only the data obtained for large bed to particle size ratio, our results indicate that the lateral effective thermal conductivity is a function of liquid viscosity and Peclet number. We can express the dependence on viscosity by employing the Prandtl number, which arises when the point equations are made dimensionless. A fit of the experimental data yields

$$\frac{K^*_{yy}}{k_l} = 0.17\,Pr^{-0.5}\,Pe_l \tag{13}$$

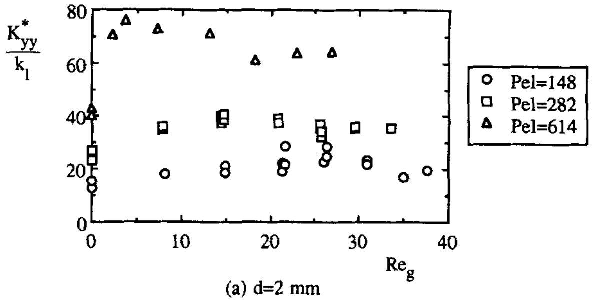
valid for

$$L_B/d > 20$$
$$100 < Pe_l < 5{,}000$$
$$6 < Pr < 33$$

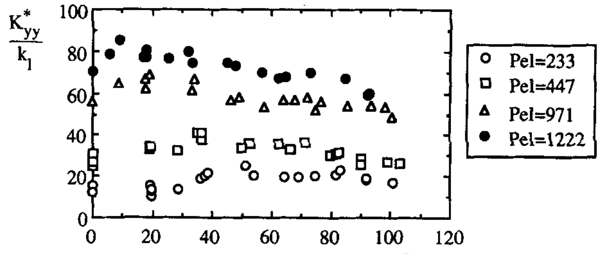
The mean relative deviation between the correlation and experimental values is 18%.

Gas-liquid flow

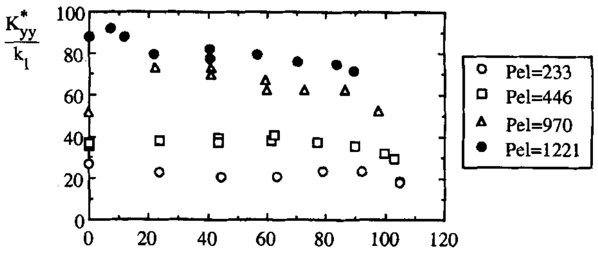
The two-phase flow experiments were conducted with particle diameters lower than or equal to 5.5 mm, so that the wall effects observed during stagnant gas-flow conditions are absent. The effect of gas Reynolds number on the lateral effective thermal conductivity is presented in Figure 4, for three



(a) d=2 mm

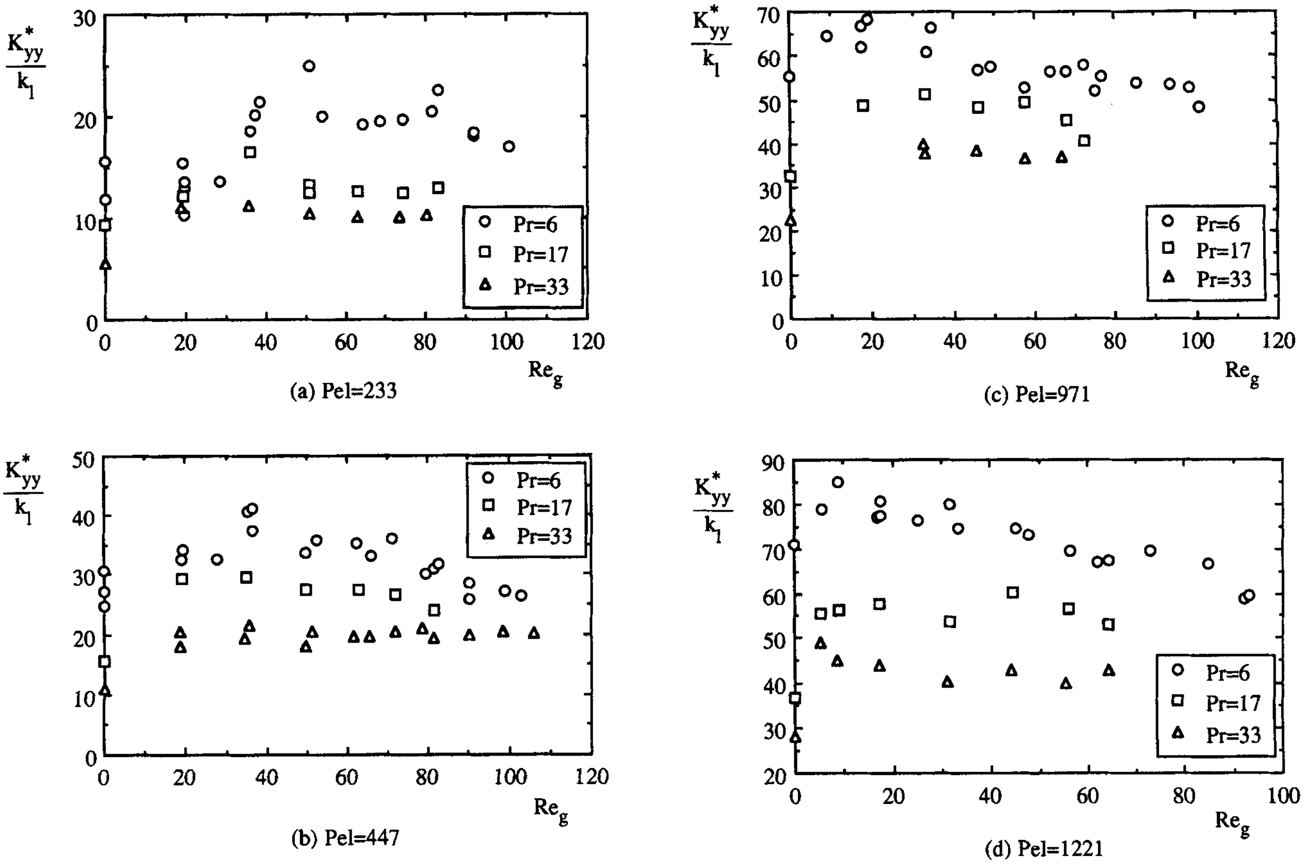


(b) d=4.5 mm



(c) d=5.5 mm

Figure 4. Effect of gas phase Reynolds number on lateral effective thermal conductivity: two-phase flow, $Pr = 6$, urea-formaldehyde spheres.



(a) Pel=233

(c) Pel=971

(b) Pel=447

(d) Pel=1221

Figure 5. Effect of liquid-phase Prandtl number on lateral effective thermal conductivity: two-phase flow, $d = 4.5$ mm.

different particles sizes and a fixed Prandtl number. The gas-phase Reynolds number is defined by

$$Re_g = \frac{\rho_g \langle v_g \rangle d}{\mu_g (1-\epsilon)} \tag{14}$$

The trends observed in Figure 4 are the same for every particle diameter used. First of all, at low liquid Peclet numbers, the effective conductivity is not very sensitive to gas Reynolds number in the range employed. As the liquid Peclet number is increased, the behavior changes: at low gas Reynolds number the effective conductivity increases until it reaches a maximum, and then decreases slowly as the gas Reynolds number is further increased. At the highest liquid Peclet numbers used, this trend is very pronounced since, in some cases, small changes in gas Reynolds number produce a noticeable increase in the effective conductivity.

The trends discussed above are related to those observed in the study of radial distribution of liquid phase by Specchia et al. (1974). They found that the radial distribution of liquid in gas-liquid flows through packed beds was sensitive to the values of gas-and liquid-flow rates. Local liquid velocities were measured as a function of radial distance. It is interesting to point out that the range of liquid-phase Reynolds number in their work was between 50 and 200, which is approximately the same range used in this work. Specchia et al. found that, at low liquid Reynolds numbers, the liquid phase was uniformly distributed throughout the cross section of the bed. As the liquid Reynolds number was increased, the uniformity of the distribution depended on gas Reynolds number: at low gas Reynolds numbers the liquid distribution was uniform; as the gas Reynolds number was increased, the gas flowed preferentially close to the wall of the bed, thus causing an accumulation of the liquid at the center of the bed. This led to a velocity profile of the liquid phase with a maximum at the center. As the gas Reynolds number was further increased, the liquid-phase distribution became more uniform.

The variations of the lateral effective thermal conductivity with gas and liquid Reynolds numbers observed in this work seem to follow closely the trends of lateral distribution of liquid phase. Since the gas phase has a low thermal conductivity, one might argue that the effect of the gas flow on the heat-transfer process leading to the evaluation of the effective conductivity results only from the effect that it has on the hydrodynamics of the liquid phase. At low liquid phase Reynolds numbers, the gas flow affects neither the liquid-phase distribution nor the liquid holdup appreciably and, therefore, its effect on the effective conductivity is small. At high liquid-phase Reynolds number, as the gas Reynolds number is increased the lateral distribution of the liquid becomes nonuniform. In this case the effective conductivity measured reflects not only the thermal dispersion arising from local (pore level) velocity fluctuations but also the macroscopic fluctuations induced by the lateral liquid velocity gradients. This implies a higher degree of lateral mixing and, as observed in Figure 4, a larger effective conductivity. As the gas Reynolds number is further increased, the lateral liquid distribution becomes more uniform and the effective conductivity drops accordingly.

The effect of liquid-phase viscosity in gas-liquid flow on the lateral effective thermal conductivities was also studied. The results are shown in Figure 5. A liquid viscosity increase has the same effect observed under stagnant gas conditions, since it results in lower values of the effective conductivity. Once again, the increase in liquid viscosity increases the liquid holdup and reduces the interstitial velocity of the liquid phase, resulting in a lower degree of lateral mixing.

An interesting observation gathered from Figure 5 regards the decrease of effective conductivity with gas-phase Reynolds number at large values of this parameter. This decrease is always more noticeable for low Prandtl numbers. This is consistent with the explanation given before relating the decrease to a more uniform liquid distribution: at larger Prandtl numbers, the liquid holdup is larger and one would expect a better lateral distribution of liquid phase in all the range of gas flows.

It is possible to correlate the two-phase flow results in the range of gas-phase Reynolds number for which the lateral flow distribution becomes more uniform as that parameter increased, i.e., for values of Re_g beyond the maxima in the effective conductivity vs. Re_g curves.

The resulting correlation is

$$\frac{K_{yy}^*}{k_l} = 2.1\, Pr^{-0.5}\, Pe_l^{0.7}\, Re_g^{-0.13} \tag{15}$$

valid for

$$L_B/d > 20$$
$$100 < Pe_l < 1{,}500$$
$$6 < Pr < 33$$
$$10 < Re_g < 100$$

The mean relative deviation between the correlation and experimental values is 27%. The exponent of the gas-phase Reynolds number in Eq. 15 reflects the slow decrease of the effective conductivity after the maximum in the curves (Figures 4 and 5). Notice that the Prandtl number exponent is the same as that obtained in the correlation for stagnant gas phase (Eq. 13), whereas the dependence on liquid-phase Peclet number is appreciably different. This difference results from the effect that the flow of gas phase has on the hydrodynamics, which in turns affects the dispersion process.

Conclusions

An experimental study for determining lateral effective thermal conductivities in trickle beds has been performed under stagnant gas phase and cocurrent downward gas-liquid flow conditions. At a given liquid-phase Peclet number, the lateral effective thermal conductivity decreases with an increase in the viscosity of the liquid phase due to an increase in liquid holdup and the consequent decrease in interstitial liquid velocity. This sensitivity of the lateral thermal dispersion mechanism to liquid viscosity should be taken into account when performing heat-transfer experiments with liquids that experience a sizable viscosity change with temperature.

The effect of the ratio between column and particle size has also been established. It has been determined that a ratio larger than 20 is necessary to eliminate wall effects from the measurement. Under gas-liquid flow conditions, the lateral effective thermal conductivity follows a complex dependence on gas-phase Reynolds number which closely resembles the trend followed by the lateral distribution of liquid phase reported in previous works. At low gas Reynolds number, the effective conductivity increases with Reynolds number reaching a maximum and then slowly decreases as the Reynolds number is further increased. The initial increase is related to a loss of uniformity of the lateral distribution of liquid phase, whereas the subsequent decrease follows a trend of the liquid distribution to become more uniform at high gas Reynolds numbers.

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