AIChE

Experimental Measurements and Multiphase Flow Models in Solid SiC Foam Beds

David Edouard and Maxime Lacroix
LMSPC (UMR 7515 du CNRS), ECPM, Université Louis Pasteur, 25, rue Becquerel, 67087 Strasbourg Cedex 02, France

Charlotte Pham
Sicat SA, Technical Center, 1, rue du Broetch, 67000 Otterswiller, France

Mamadou Mbodji
Total, Centre Scientifique et Technique Jean Feger, Avenue Larribau, 64000 PAU, France

Cuong Pham-Huu
LMSPC (UMR 7515 du CNRS), ECPM, Université Louis Pasteur, 25, rue Becquerel, 67087 Strasbourg Cedex 02, France

DOI 10.1002/aic.11594
Published online September 29, 2008 in Wiley InterScience (www.interscience.wiley.com).

Solid foam matrices have been recently introduced and present a highly permeable porous structure with a high porosity (0.60–0.95), which enables a considerably reduction of the pressure drops along the catalyst bed even with a high specific surface area. It appears today that despite the hydrodynamics of trickle-bed reactors operating with conventional packings (spheres, extrudates, monoliths, etc.) has become largely documented in the recent literature, trickle-bed hydrodynamic behaviour with solid foam packings, on the contrary, largely remains terra incognita. In this context, two phases flows (air–water) in solid foams under cocurrent trickle flow conditions were analyzed in a fixed-bed at ambient conditions. From Residence Time Distribution (RTD) curves, a new set of experimental data based on the hydrodynamics parameters (pressure drop and liquid holdup) of solid foams are presented. A model based on a cubic lattice approach developed in previous work is used to calculate the equivalent diameter for solid foam packings. Finally, through the modified Eötvös number and the relative permeability concept, the two-phase pressure drop and liquid holdup are well estimated. These news results are important parameters for the design of new process in trickle flow conditions with solid foam packing. *AIChE J*, 54: 2823–2832, 2008
Keywords: porous media, solid foam, two-phase pressure drop, liquid holdup, trickle-bed, permeability concept

Introduction

The use of porous structures represents an important breakthrough in industrial applications. For reactions in multiphasic systems, the reactor packing aims to increase the

Correspondence concerning this article should be addressed to D. Edouard at edouardd@ecpm.u-strasbg.fr.

rate of mass transfer by increasing the gas - liquid contact surface and by increasing the turbulences within the fluid phase. Industrial applications of multiphase reactors included Fisher–Tropsh synthesis and hydrotreating processes of petroleum refining. Many gas–liquid–solid catalytic reactions belonging to the petrochemical industry[1–4] use a gas-liquid trickle-bed reactor (TBR) in which the liquid phase flows downward through the reactor. The gas phase can flow either in a counter - or cocurrently direction. Because flooding may occur in a countercurrent column, cocurrent flow is usually recommended for high loading.[5,6] In this context, the main advantages of using these porous structures are the high contact surface (exchange specific surface area) between the fluid and solid phase and also the low pressure drop along the catalytic bed. One relevant example is the packed bed, which is frequently utilized in catalytic converters and thermal energy storage device. However, due to the low porosity (in the range of 0.3–0.6), packed beds induce important pressure drops at high flow rates which is detrimental for the global process, especially when high space velocity is required in order to maintain acceptable productivity or selectivity. The idea of moving from the traditional packed-bed, i.e., spheres, pellets, to the structured bed, i.e., monolith, wire or foam either made of stainless steel or ceramic, has become more and more popular during the last decade. Foam matrices have been recently introduced to overcome some of the above shortcomings dealing with the use of the packed bed. This new medium has as a highly permeable porous structure with a high porosity (0.60–0.95), which enables a considerably reduction of the pressure drops along the catalyst bed even with a high specific surface area. Permeability is also an important parameter for the characterization of foams employed in industrial applications. Knowledge of pressure drop induced by these foam matrices is essential for successful design and operation of high-performance industrial systems. During the last decade, numerous experiments and models on single-phase flow in foams have been developed and reviewed by Edouard.[7] However, it appears today that despite the hydrodynamics of trickle-bed reactors operating with conventional packings (spheres, extrudates, monoliths, etc.) has become largely documented in the recent literature (see the review of Gianetto et al.[8]), trickle-bed hydrodynamic behaviour with solid foam packings, on the contrary, largely remains terra incognita. The specific two-phase pressure drop $\Delta P/L$ depends on operating conditions and can vary from very small values in trickle flow, to very high values, several bars/m in high interaction regimes. Concerning the TBR operating in low interaction regimes (subject of this work), few articles were found in the literature concerning the biphasic flow moving cocurrent through packed foam structures. Stemmet et al.[9] presented the gas–liquid mass transfer and axial dispersion in solid foam packings. In this work, the authors used air–water system moving in a cocurrent upflow configuration through metallic foam with high porosity. In Topin et al.,[10] the experimental work was based on the stationary pressure profile measurement in a channel filled with metallic foam for several controlled flow rates. Two phases flows (air–water) in cocurrent conditions were analyzed in a plexiglass tube horizontally disposal. The aim of this present work is to quantify the hydrodynamics parameters (pressure drop and liquid holdup) in solid foams under cocurrent trickle flow conditions.

Experimental Apparatus and Procedure

Characteristics and morphological parameters of solid foams

Today, solid foam packing may be produced in a large variety of materials (Al_2O_3, cordierite, metals-based (aluminum, copper etc.) and used in a large range of applications, especially in the field of thermal applications (Lu et al.[11]). Recently, Richardson et al.,[12] Groppi et al.[13] have investigated solid foam packing as potential catalytic support for gas phase reaction. Stemmet et al.[9] described the use of these solid foam materials in multiphase reactors as catalyst support material. However, these foam structures have a relatively low specific surface area ($m^2\ g^{-1}$) due to the high temperature methods of preparation[14,15] for performing good anchorage and dispersion of the active phase. β-SiC foams (studied in this work) with a medium specific surface area and a natural wash-coat layer, i.e., SiO_2 and SiO_xC_y topmost passivation layer (2–4 nm) formed by air oxidation of the ceramic at room temperature, have been synthesized and widely employed in several catalytic reactions. The passivation layer allows the good anchorage of the deposited active phase which enhances the number of active sites per linear surface while its low thickness allows the complete conservation of the intrinsic properties of the underlying support.

In this work, the main morphology of solid SiC foam sample was investigated by means of a scanning electron microscopy (SEM) and carried out on a Jeol JSM-6700F working at 3 kV accelerated voltage, equipped with a CCD camera. The samples was previously coated with carbon and then deposited on a standard holder for observation. The optical images presented in Figure 1 have been taken with an optical microscope Digital Blue equipped with a numerical camera. From this figure, the main morphological characteristics of foams were examined (i.e., the diameters of the cell, pores diameter (a) and the thickness of the struts (d_s)).

Another important parameter of these structures is the foam porosity (ε) which is the volume available for the fluids flow through the open-cell structure. Generally in the literature, ε can be calculated on the basis of mass and volume measurements using the expression $\varepsilon = 1 - \rho_g/\rho_s$, where, ρ_g is the foam apparent density and ρ_s is the materials density of the struts. The material strut bulk density ρ_s and internal porosity can be obtained by different porous distribution techniques (mercury intrusion,[13] water picnometer,[17] He multipycnometer[17]). In this work, the strut bulk density (ρ_s) was determined by mercury intrusion. Finally, external specific surface area (a_c, m^{-1}(i.e., $m^2\ m^{-3}$)) is an important parameter which is responsible for the high-performance and successful design of reactors and which can be calculated from the solid foam geometry as described by Lacroix et al.[18]

In this work, for each sample of SiC foam and/or spherical particles studies, the main morphological characteristics of each bed are summarized in Table 1.

Reactor system

The main elements of the experimental setup are schematically represented in Figure 2. The reactor (I.D. = 37 mm)

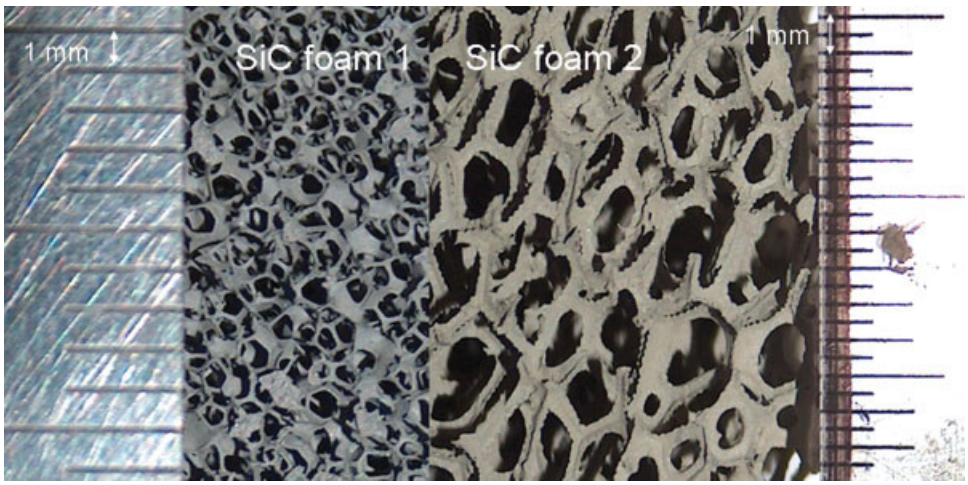

Figure 1. Optical pictures of solid SiC foams showing the detail morphology and connection structure of the material.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

was packed with 3-mm glass spherical particles or solid SiC foam to have a total bed height of 90 cm. The whole facility was maintained in a hermetic casing to prevent vapor leakages.

In the biphasic mode, the liquid water was introduced through the top of the packed column. Gas (air) and liquid (water) were injected through a spray shower to ensure even distribution of the liquid phase on the top of the bed. Different gas flow rates, measured by a rotameter, were explored for different superficial liquid velocity. The maximum superficial gas and liquid velocities determined using the empty reactor cross section were 0.217 and 0.0078 m s^{-1}, respectively.

Two-phase pressure drop and liquid holdup measurements

Liquid holdup is defined as the fractional volume of fixed bed occupied by the liquid phase. For porous media, liquid holdup consists of intra-particle and inter-particle contributions. Inter-particle holdup can be split into dynamic and static contributions:

$$\varepsilon_{Lt} = \varepsilon_L + \varepsilon_{Lint} = \underbrace{\varepsilon_{Ld} + \varepsilon_{Ls}}_{\varepsilon_L} + \varepsilon_{Lint} \quad (1)$$

In Eq. 1, ε_{Lint} (internal liquid holdup) is the ratio of the volume of the liquid held by capillary forces in the pores of porous media to the reactor volume while the ε_L (external liquid holdup) is the ratio of the volume of the liquid located outside the pores, which representing the void volume of the bed occupied by the liquid, to the reactor volume. The external liquid holdup can further be divided into dynamic (or free–draining) (ε_{Ld}) and static (ε_{Ls}) liquid holdup. The dynamic liquid holdup is the free flowing fraction of the liquid and depends upon the hydrodynamics of the flow whereas the static liquid holdup is the ratio of the volume of stagnant liquid and the liquid which is retained between and around the contact points of porous media by capillary forces after draining of the reactor volume. This parameter depends on the operating conditions and support shape and size.

The total liquid holdup (ε_{Lt}) in the column was evaluated using residence time distribution (RTD) analysis measures. A well-known tracer technique (conductance measurements) was performed to determine the RTD curves (for a full description, see Larachi et al.[21]). KCl solution in deionized water was used as tracer. A short tracer pulse was syringe-injected in the liquid inlet pipe. A conductivity cell was

Table 1. Support Characteristics

Support Characteristics	Glass Spherical Particles This Work	SiC Foam1 This Work	SiC Foam2 This Work	Aluminum Foam Stemmet[20,21]		
Cell diameter	–	1650 μm	2650 μm	–	–	–
Pores diameter, a	–	717 μm	1150 μm	2450 μm	612 μm	314 μm
Apparent mass, ρ_{app}	–	187 g l^{-1}	177 g l^{-1}	–	–	–
Open porosity, ε	0.35	0.87	0.88	0.93	0.935	0.94
Strut diameter (d_s) or equivalent strut diameter (calculated*) or particles diameter	3 mm	220 μm	336 μm	505 μm	126 μm	62 μm
Calculated* geometrical specific surface area, ac_g	1300 m^{-1}	2360 m^{-1}	1430 m^{-1}	540 m^{-1}	2146 m^{-1}	4100 m^{-1}
Internal porosity of the support	0 cm^3 g^{-1}	0.03 cm^3 g^{-1}	0.02 cm^3 g^{-1}	–	–	–

*Lacroix.[19]

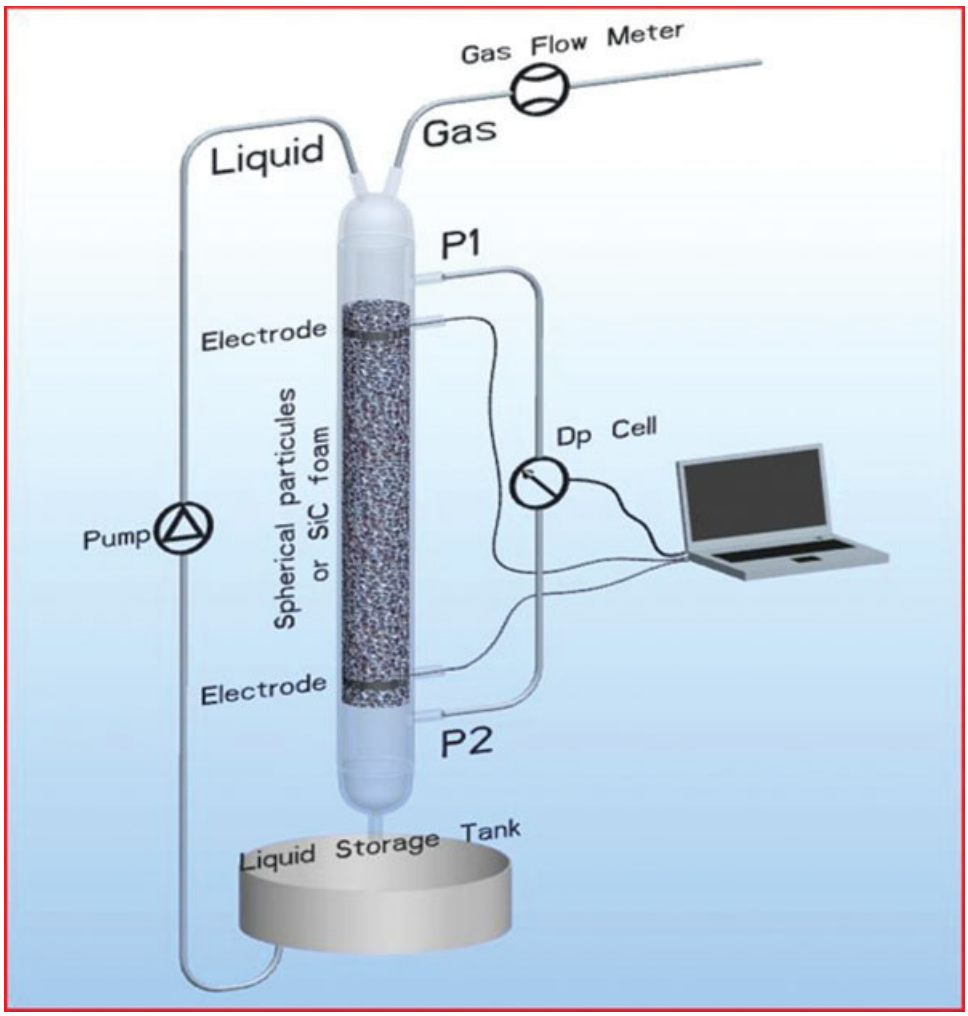

Figure 2. Experimental setup used.

[Color figure can be viewed in the online issue, which is available at www. interscience.wiley.com.]

placed in the top of the reactor to check the pulse injection and the other cell was placed 3 cm above the bottom of the foam bed. The output signals from the probes were received by a conductivity controller (Omega model CDCN-91) and transmitted to a computer by a data acquisition system. From Figure 3, we can see that a *δ-dirac* pulse could be obtained approximated by this technique. To ensure reproducibility and complete wetting of the packing structure, the following procedure was applied: the column was first completely filled with liquid and then, the liquid was withdrawn at once through the entire column. Then the required gas and liquid flow rate was set, and the column regime was stabilized for about 20 min under dynamic flow before starting the different measurements.

From RTD curves and for a pulse injection of tracer, the total liquid holdup (ε_{Lt}) can be calculated as follows:

$$\varepsilon_{Lt} = \frac{\tau Q_L}{V_R} \quad \text{with } \tau = \int_0^\infty tE(t)dt \tag{2}$$

In our case, the tracer concentration is linearly proportional to the output signal of the analytical equipment used, thus $E(t)$ can be evaluated directly from the measured signal as:

$$E(t) = \frac{I(t)}{\int_0^\infty I(t)dt}.$$

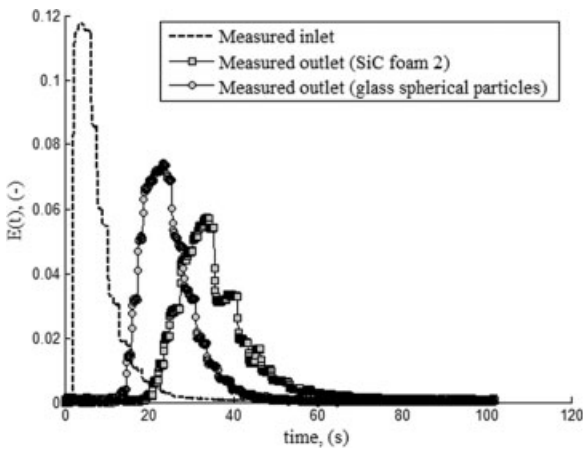

Figure 3. RTD curve measurement.

To estimate the dynamic liquid holdup (ε_{Ld}), the simple drainage method is used (see for instance Yang et al.[22]). In this method, gas and liquid inlets flows are stopped simultaneously; the liquid level in the liquid storage tank is then measured in order to determine the dynamic liquid holdup in the column.

From Eq. 1 the residual liquid holdup ($\varepsilon_{Ls} + \varepsilon_{Lint}$) can be obtained by the difference between the total liquid holdup and the dynamic liquid holdup. In first approximation, it is generally assumed that the pores of catalyst support are completely filled with the liquid (due to capillary forces, particularly in our case where the bed was completely filled with liquid before operation) making the internal liquid holdup (ε_{Lint}) and total pore volume equal. This total pore volume is obtained with BET measurement and given Table 1.

Consequently, by subtracting the total catalyst pore volume from residual liquid holdup, the static holdup (ε_{Ls}) can be determined.

Finally, the two-phase pressure drop was measured with differential pressure sensor (Keller Druckmesstechnik PD-41 (0–200 mbar)) connected to the bed top and bottom sections.

Two-phase flow models

State of the Art Depending on the liquid and gas mass flow rates, several flow regimes may exist in TBR and have given descriptive names such as trickle, pulsing, and spray flow (Stegeman et al.[24]). In the trickle flow condition the gas is the continuous phase while liquid dribbles downward over the packing in the form of rivulets and films. The hydrodynamic parameters necessary for the design and operation of trickle bed reactors are the pressure gradient and liquid saturation. The pressure gradient and liquid saturation are related to the mechanical energy dissipated due to the two-phase flow through the packed bed and the void volume of the packed bed. Therefore, the properties of the packing mode (tortuosity, porosity, geometrical surface area) are important parameters to predict the pressure drop, liquid saturation, transport coefficient, and identify the prevailing flow regime as the function of the operating conditions. A large number of studies have been reported in the literature on various hydrodynamics aspects of the TBR. Several correlations and models of pressure gradient and liquid holdup have been developed from these studies. The main hydrodynamic models can be broadly classified in two different categories. The first one uses an empirical approach based on dimensional analysis to produce explicit correlations for pressure drop and holdup (Ellman et al.,[24,25] and Larachi et al.[21]). These correlations have several parameters which are not universal constants for fitting the experimental data and the estimated values of pressure drop and liquid holdup obtained by these correlations vary considerably. The second category involves the development of models resulting from equation of motion (continuity and momentum) and considers determination of drag forces of gas and liquid phases at various operating regimes (see for instance the excellent review of Carbonell[26]). In this approach, closure relations for the drag forces need to be derived either from first principles or by semiempirical relations to relate them to the flow velocities and volume fractions of each phase, and to the physical properties of the gas, liquid, and solid phases. Saez et al.[27] discussed in detail the macroscopic equations of motion governing two-phase flow in packed beds under steady state and isothermal nonreaction conditions, the volume-averaged momentum equation for the α phase (where α can be either liquid or gas) take the following form:

$$\varepsilon_\alpha \rho_\alpha u_\alpha \frac{du_\alpha}{dz} = -\varepsilon_\alpha \frac{dP_\alpha}{dz} + \varepsilon_\alpha \rho_\alpha g - F_\alpha \tag{3}$$

where, F_α is the drag force per unit volume of bed acting on the α phase and ε_α is the volume fraction of each phase (liquid and gas). Under assumption of one-dimensional flow conditions, the liquid holdup and the average flow velocities are independent of position along the bed, and thus the inertial term on the left-hand side of Eq. 3 is negligible. In this case, the pressure gradients in liquid and gas phases are equal since gradients in liquid saturation are negligible. Knowing that in these conditions, the continuity equations for the gas and liquid phase are reduced to the following form,

$$\begin{cases} \varepsilon_l \rho_l u_l = L \\ \varepsilon_g \rho_g u_g = G \end{cases} \tag{4}$$

where L and G are the total liquid and gas mass fluxes and that the volume fraction of both phases must be added to the total fraction of voids in the medium, ε(i.e., $\varepsilon = \varepsilon_g + \varepsilon_l$); only the knowledge of the expressions for the drag forces F_g and F_l are necessary to calculate both the pressure drop and the liquid holdup in the bed.

The most common approaches admitted in the literature and used in this work to obtain these drag expressions are presented in the following section.

The Relative Permeability Model. In the relative permeability concept, the Ergun equation (Ergun et al.[28]) which is basically for single-phase flow and arises from a momentum balance in assumed bed geometry has been modified to take into account the existence of a second flowing phase. The resulting expression for the drag force per unit volume of each fluid is obtained by Saez et al.[27]:

$$\frac{F_\alpha}{\varepsilon_\alpha} = \frac{1}{k_\alpha(S_\alpha)} \left(A \frac{Re_\alpha}{Ga_\alpha} + B \frac{Re_\alpha^2}{Ga_\alpha} \right) \rho_\alpha g \tag{5}$$

where k_α is the relative permeability, A and B are the Ergun coefficients for single phase flow, Re_α and Ga_α are respectively the Reynolds and Galileo numbers.

Saez proposed to correlate the relative permeability (k_α) with the saturation ($S_\alpha = \frac{\varepsilon_\alpha}{\varepsilon}$) of each phase with the following relationships:

$$\begin{cases} k_l(S_l) = \left(\dfrac{S_l - S_l^0}{1 - S_l^0} \right)^{n_1} \\ k_g(S_g) = (S_g)^{n_2} \end{cases} \tag{6}$$

Once, the expressions for the drag forces of each fluid are known (i.e., for n_1, n_2, A, and B given) and that the pressure gradients in liquid and gas phases are equal, the pressure drop in the bed and the total liquid holdup can be calcu-

lated by coupling Eqs. 3–6. The resulting system is then given by:

$$\begin{cases} \Psi_l = \dfrac{F_l}{\varepsilon_l \rho_l g} = 1 - \left(\dfrac{\Delta P}{Z}\right)_l \dfrac{1}{\rho_l g} \\ \Psi_g = \dfrac{F_g}{\varepsilon_g \rho_g g} = 1 - \left(\dfrac{\Delta P}{Z}\right)_g \dfrac{1}{\rho_g g} \\ \Psi_l = \dfrac{\rho_g}{\rho_l}(\Psi_g - 1) + 1 \end{cases} \quad (7)$$

The Slit Model. The phenomenological slit model developed by Holub et al.,[3] Al-Dahhan et al.[29] and Iliuta et al.[30] involves equations of flow for each phase in the pore scale, and use volume averaging to establish a bed scale model. Initially, the slit model proposed by Holub[3] represented the local flow of liquid and gas around the particles in a TBR by the much simpler flow inside a rectangular slit. In their model, the width of the slit is a function of bed porosity and the angle of inclination of the slit to the vertical axis is related to a tortuosity factor for the packed bed. They introduced the concept of slit of the velocity and stress fields at the gas-liquid interface by introducing two slip parameters (f_v and f_s):

$$\begin{cases} u_g|_i = f_v u_l|_i \\ \tau_g|_i = f_s \tau_l|_i \end{cases} \quad (8)$$

From this system, we can see that if the values of slip parameters are different from 1, there is a discontinuity in the velocity profile and in the shear stress of the gas and liquid at the interface. The resulting pressure drop and liquid holdup obtained by Holub[3] seems to indicate zero for slip parameters at the gas-liquid interface to correlate the experimental data from literature with this approach. Although in reality, a weak interfacial gas-liquid interaction exist (in particular case of very low superficial gas and liquid velocities), this result can be normally assumed to be valid in continuus flows in TBR literature. As result, it leads to a similar coupling between liquid holdup and pressure drop as was found in the relative permeability model. In these conditions, the slip model gives an identical form to system (7) but with a following different relative permeability functions for the gas and liquid phases:

$$\begin{cases} k_l(S_l) = (S_l)^3 \\ k_g(S_g) = (S_g)^3 \end{cases} \quad (9)$$

Later, Al-Dahhan[31] obtained the correlations for the slip parameters as a function of gas and liquid phase Reynolds numbers in order to reconcile their experimental data on the pressure drop and liquid holdup (in the high-interaction and high-pressure regimes) with this slit model (i.e., f_v, $f_s \neq 0$). Since, Iliuta et al.[30] developed the double slit model, which considers the distribution of totally dry slits in addition to the wet slits, as in Holub.[3] They used the same slit factors that Al-Dahhan[31] used.

Recently, Carbonell[26] used the data from the literature (in the high- and low-interaction regimes) to compare the results of the relative permeability model with the slit model. The author concludes that because the models are based on a fundamental approach and contain mechanistic details of the system, they have a wider range of applicability unlike the correlative models, which are system specific. Furthermore, there is no much difference between the relative permeability theory and the slit model. The reason for this is that it is well-known that two-phase flow pressure drop and holdup measurements are subject to significant experimental variations from laboratory to laboratory and indeed, from packing to packing.

In the next section, ,the present work attempts these approaches (relative permeability model and the slit model) in order to model pressure drop and liquid holdup at low-interaction regimes of the trickle bed reactor filled with a solid β-SiC foam. The present study focuses on the original forms of these models (no new fitting parameters are presented in order to reconcile data and theory) to compare these models with the experimental data from the literature and this work.

Results and Discussions

Pressure drop: single-phase flow

Before the analysis of the two-phase flow problem, the single flow was first considered. The single-phase (air flow) pressure drop was measured across the whole packed bed section with a differential pressure transmitter (see experimental section). According to the main characteristics of spherical particles and solid SiC foam, Figure 4 shows a good agreement of simulated pressure drops with experimental data. Lacroix et al.[19] used a cubic cell model with Ergun's equation and used the standard constant Ergun parameters ($E_1 = 150$ and $E_2 = 1.75$). This work has evaluated the applicability of analogy between the traditional spherical particles bed and the foam in order to predict gas phase pressure drops in foams through standard Ergun's equation. A direct analogy between foams and beds filled of spherical particles with the same specific surface area and same porosity than solid foam was developed trough a simple equation ($d_p = \frac{3}{2} d_s$). The estimated pressure drop results were compared with those obtained experimentally under air flow on solid SiC foams with pores diameter (a) ranged between 475

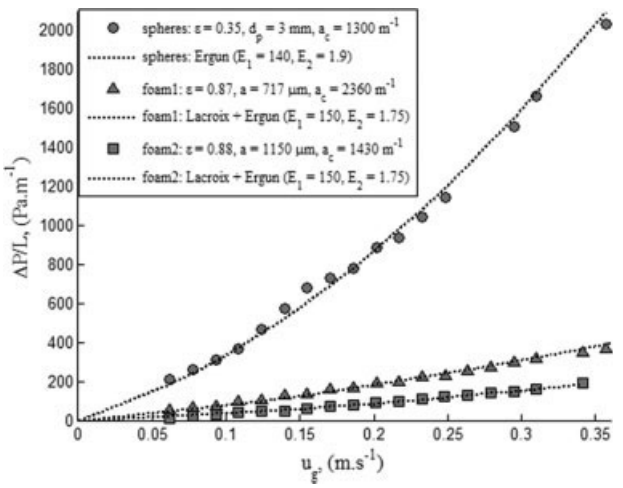

Figure 4. Pressure drop measurement versus gas velocity for different porous media and estimated results.

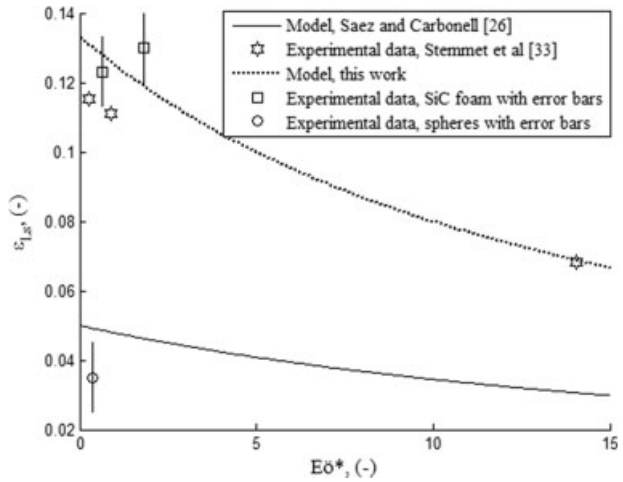

Figure 5. Static holdup correlations.

and 1650 μm, porosity between 0.76 and 0.92 and from fluid velocities as high as 6 m s^{-1}. Recently, the review of Edouard et al.[7] and the work of Giani et al.[32] seem to show that the strut diameter is a good characteristic length and can be used for simulating the pressure drop with standard Ergun's equation on foam matrixes. Figure 4 shows clearly that even with a high specific surface area, the pressure drops through foams were much lower than those measured with the "conventional" packed bed. Foams seem to be an appropriate support for catalytic reactions with short contact times (i.e., high-reactant flows) by preventing excessive pressure drop across the bed.

Static holdup

The measured static liquid holdup for the different packings (glass spherical particles and solid foam) is given in Figure 5 as a function of the modified Eötvos number ($E\ddot{o}^*$) introduced by Saez and Carbonell.[27] The solid and dotted lines correspond to a proposed fit, given by the equation:

$$\varepsilon_{Ls} = \frac{1}{c + dE\ddot{o}^*} \text{ with } E\ddot{o}^* = \frac{\rho_l g d_p^2 \varepsilon^2}{\sigma(1-\varepsilon)^2} \tag{10}$$

The values of c and d found by the authors (Saez and Carbonell[28]) for the packings studied (spheres, extrudates, etc.) are respectively, 20 and 0.9. From Figure 5, it can be seen that in spite of foam packings (SiC foam and Aluminum foam) present a higher porosities, the static liquid holdup measurement is higher and can not be described by Eq. 10 with the originals values of c and d. In this work, the equivalent particle diameter ($d_p = \frac{3}{2} d_s$) given by Lacroix[19] is used in the modified Eötvos number and the static liquid holdup for the foams can be described by Eq. 10 with values of 7.5 and 0.5 for c and d respectively.

Recently, Stemmet et al.[20,21] proposed to correlate the liquid holdup for the Aluminum foams with values of 9 and 0.025 for c and d respectively. Because the authors used the effective length of the strands instead of the equivalent diameter in the modified Eötvos number, they obtained value of d very far from the original values proposed by Saez.[28] There is no question that the data in Figure 5 are widely scattered and that the values of c and d proposed in this work are only a good representation of the experimental data. Effectively, the experimental measurements of the static holdup involve a fair amount of error since the volume of liquid is not measured directly but can be obtained by the difference between the measured total liquid holdup and the dynamic liquid holdup (see experimental section). Another important point is that Saez correlation doesn't take into account the wettability of the solid foam. If in this work the effect of the wettability are minimized because the column was completely filled with liquid before that the required gas and liquid flow rate was set; this is not obvious in study of Stemmet et al.[20] because the experimental trickle flow regime is starting with the void space in the foam filled with only gas phase. When the solid is not perfectly wetted by the liquid, the correlation tends to overestimate the experimental data on the average. The conclusion is that neither of two representations (length of the strands or the equivalent particle diameter) can be used as a way of estimating static liquid holdup from a practical standpoint. However, the Lacroix correlation avoids some possible pitfalls in the characteristic length chosen by previous investigators and gives values of c and d near of the original values. In any case, the static liquid holdup for the foam packing is higher than for classical packings. For solid foam, increasing the porosities and/or the equivalent particle diameter increases the modified Eötvos number and thus decreases the static liquid holdup.

On Figure 6, the apparent thickness of static liquid film (e) remaining on the surface of the solid foam packing versus of strut diameter (d_s) is presented. From the assumption (proposed by Stemmet[21]) that the static liquid holdup spreads evenly over the entire specific surface area, the apparent thickness of liquid film is obtained by the following equation:

$$e = \frac{\varepsilon_{Ls}}{ac_g} \text{ with } ac_g = \frac{4(1-\varepsilon)}{d_s}, \tag{11}$$

given by Lacroix.[19]

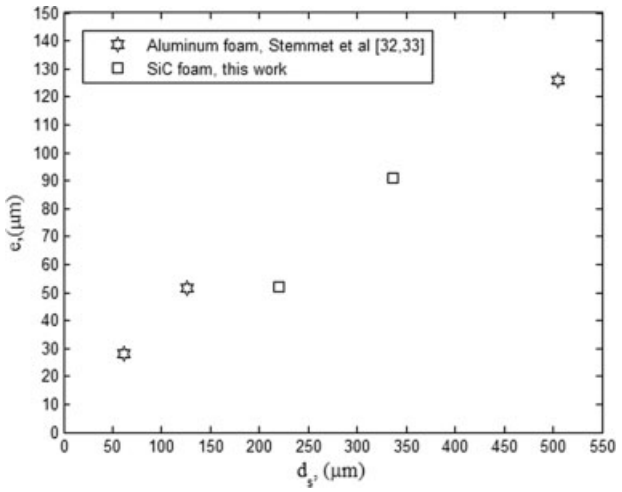

Figure 6. Thickness of static liquid film versus of solid foam strut diameter.

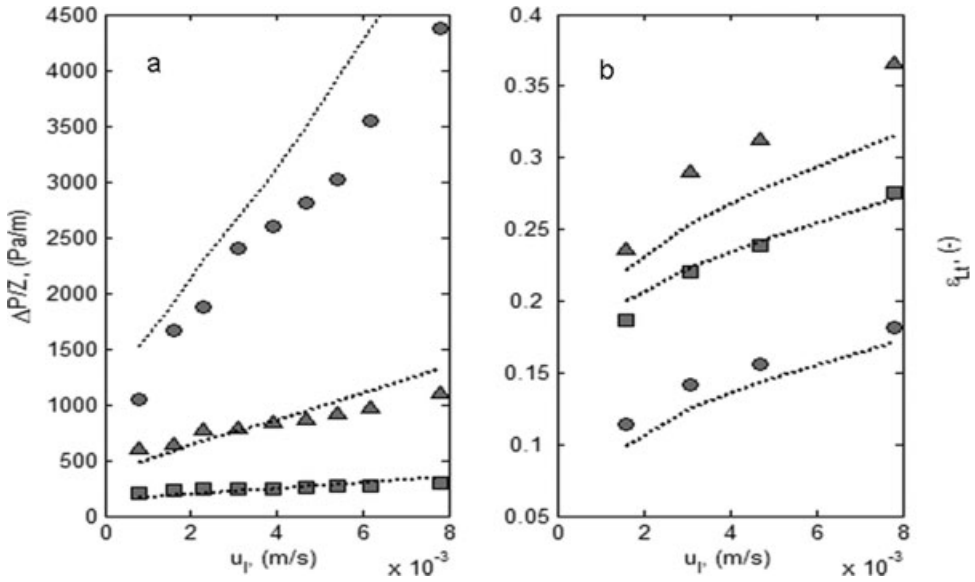

Figure 7. Two-phase pressure drop (a) and total liquid holdup (b) measurements and estimated results (Saez and Carbonell[28]) (---model, ● spherical particles, ▲ SiC foam1, ■ SiC foam2), $u_g = 0.155$ m s^{-1}.

For the range of solid foam samples studied, the apparent thickness of the static liquid film increases with the strut diameter and not only with the porosity (i.e., the apparent density of solid foam). In fact, one would expect an opposite trend, since larger diameters would imply a decrease in capillary forces. Although, it seems that in the case of the solid foams, when the strut diameter decreases the specific surface area increases more rapidly than the capillary forces. Moreover, at this level of observation, it is not obvious that only the forces of capillarity forces due to pore diameter (a) or strut diameter (d_s) play a role, it is also possible that the size, the form and the number of nodes in solid foam are involved in these results. From these results and since the morphology of the foam is controlled, it is clear that the solid foam has larger operating capabilities and there is a need for more research in this area.

Pressure drop and liquid holdup: Two-phase flow

The experimental pressure drop for different liquid flow rates with constant air flow rate ($u_g = 0.155$ m s^{-1}) is illustrated in Figure 7a. This figure compares the experimental pressure drop between spherical fixed bed and two packed bed of solid SiC foam. As shown on this figure, the pressure drop difference is quite important and becomes larger as the liquid velocity increases. According to the single flow model, it appears that an increase in the pore size for similar porosity of foam implies a decrease of the pressure drop.

Liquid holdup plays an important role in several trickle bed processes such as the control of the catalyst wetting efficiency, the radial heat transfer, the gas-liquid mass transfer of gaseous reactant and the solvent evaporation. According to these facts, knowledge of the liquid holdup is another key factor in reactor design. Thus, Figure 7b compares the measured total liquid holdup for a "classical" packed bed (spherical particles) and for a fixed bed filled with solid SiC foams. The total liquid holdup results are shown for different velocities of liquid. It appears that the total liquid holdup increases with increasing liquid velocity (classical result) and is more important with solid SiC foam supports that in the case of spherical particles for the same flow conditions. This could be expected since the foam has a higher static liquid holdup. In the same flow condition and for the same static liquid holdup (in the first approximation), it appears the dynamic liquid holdup is higher for the SiC foam1 compared to the SiC foam2. The same trend is observed by Stemmet et al.[20] and can be explained by a higher restriction to the liquid flow through the foam (i.e., tortuous path and specific surface area increase).

To estimate the pressure drop and liquid holdup in the solid foam, two different approaches as described previously were experimented:

(a) The original model developed by Saez and Carbonell.[28] (b) Holub et al.[3] correlation. In two case, these approaches do not involve any fitting of data and use only a physical characteristic length ($d_p = \frac{3}{2} d_s$), with d_s given by Lacroix.[19]

It appears in Figure 7 that in the case of spherical particles, the estimated data are close to the experimental data (pressure drop and liquid holdup) with Saez correlation.

To check the validity of these models with experimental data for the solid SiC foam1 and the solid SiC foam2, the experimental data and model results for different liquid flow rates and three constant air flow rates ($u_g = 0.078$, 0.155, 0.217 m s^{-1}) are presented in the parity plot given in Figure 8a,b. This graph is obtained from 48 different experimental data. It is easy to see that from the Lacroix analogy (notion of equivalent diameter for foam packing), the estimated pressure drop and liquid holdup are better using the relative permeability model than with the slit model. Because of their high porosities, the bed tortuosity in solid foam may be different from the "classical packings" and that in Holub et al.[3]

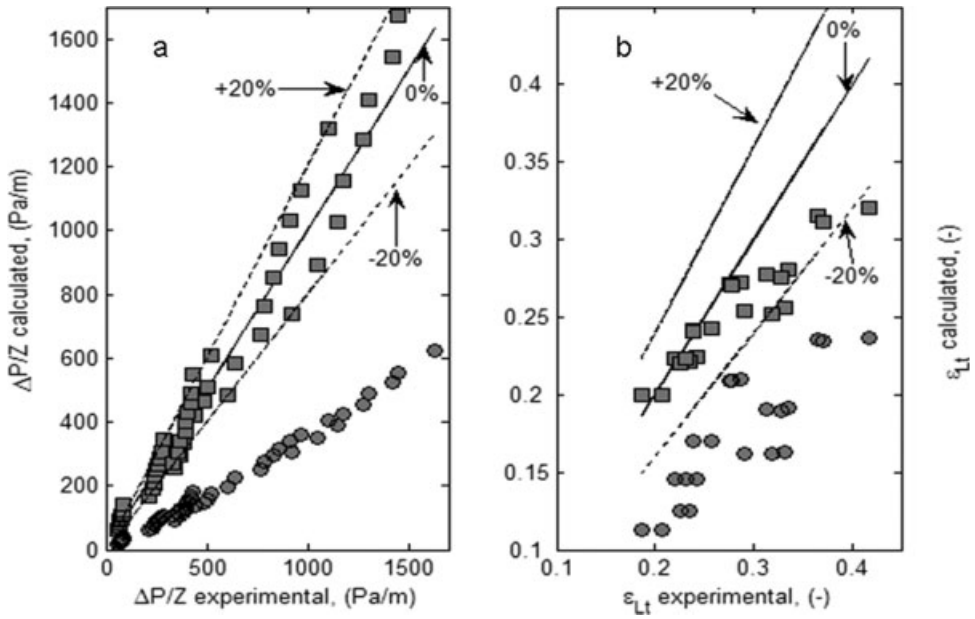

Figure 8. Parity diagrams of theoretical value versus experimental data.

(a) Pressure drop, (b) Liquid holdup. (● Slit-model (Holub et al.[3]), ■ Relative permeability model (Saez and Carbonell[27]).

model, the width of the slit is related to the angle of inclination (which depends on the bed tortuosity); the slit model does not seem to be appropriate for estimating the liquid holdup and thus the pressure drop and in solid foams.

For the relative permeability model, the values of n_1, n_2, A and B given by Saez and Carbonell[27] for all types of "classical packings" are respectively 4.8, 2.43, 180, and 1.8. Because the analogy developed by Lacroix[19] used the standard Ergun parameters ($E_1 = 150$ and $E_2 = 1.75$ proposed by Ergun[29]) instead of values A and B, it is necessary to calculate the corresponding values of n_1 and n_2 for these standard values. From the data used by Saez and Carbonell,[27] we have obtained 5.15 and 2.6 for respectively n_1 and n_2 Note that this new set of parameters (n_1, n_2, E_1, and E_2) is strictly equivalent to the one introduced by Saez and Carbonell[27] to correlate the two-phase pressure drop and total liquid holdup. Thus, the results of simulation presented in Figure 8 do not involve fitted parameters in order to reconcile experimental data for the solid foam with the relative permeability model. Figures 8a, b show how well the correlation compares with experimental data. Effectively, the majority of the pressure drop data lies in the $\pm 20\%$ region, whereas the liquid holdup tends to be underestimated.

These results agree with the measurements made by Stemmet et al.[21] Because in their work the authors have not a good model for the estimation of the pressure drop in single - phase flow (i.e., A and B very higher and not constants), they cannot obtain the values of n_1 and n_2 constants and comparable to the original values found by Saez.[27]

Conclusions

Two phases flows (air–water) in solid foams under cocurrent trickle flow conditions were analyzed in a fixed-bed at ambient conditions. From Residence Time Distribution (RTD) curves, a new set of experimental data based on the hydrodynamics parameters (pressure drop and liquid holdup) of solid foams are presented. The results show a low pressure drop induced by the two phase flows and the higher liquid holdup with foams compared to classical packed bed. A model based on a cubic lattice approach developed in previous work is used to calculate the equivalent diameter for solid foam packings. Finally, through the modified Eötvös number and the relative permeability concept, the two-phase pressure drop and liquid holdup are well estimated. These news results are important parameters for the design of new process in trickle flow conditions with solid foam packing.

Since the morphology of the foam is controlled, it is clear that the solid foam have larger operating capabilities and there is a need for more research in this area (heat and mass transfer under trickle flow conditions for example), and this will be the subject of a future article.

Acknowledgments

The authors thank the Total for financial support. Dr. Daniel Schweich, Dr Jérome Anfray and Dr Régis Philippe are gratefully acknowledged for help and advices.

Notation

Definitions

a = pore diameter, μm
ac_g = external specific surface, m^{-1}
d_s = strut diameter, μm
d_p = particle diameter or equivalent diameter, μm
$E_{1,2}$ or A, B = Ergun constants
$Ga = \left(\frac{\rho_l^2 g d_p^3 \varepsilon^3}{\mu_l^2 (1-\varepsilon)^3}\right)$ = Galileo number
g = gravitational acceleration constant, m s^{-2}
L = length of foam, m

$Re = \left(\frac{\rho_l u_l d_p}{\mu_l(1-\varepsilon)}\right)$ = Reynolds number
u = superficial fluid velocity, (m s^{-1})

Greek letters

ρ_s = strut bulk density, g l^{-1}
ρ_g = apparent density, g l^{-1}
ΔP = pressure drop, Pa
ε = porosity
ε_L = liquid holdup
$E\ddot{o}^*$ = modified Eötvos number

Subscripts

g = gas
l = liquid
i = interface
s = static
d = dynamic
t = total
int = internal

Literature Cited

1. Bakos M, Charpentier J. Taux de rétention pour les écoulements gaz-liquide à co-courant vers le bas dans les colonnes à garnissage arrosé et noyé. *Chem Eng Sci*. 1970;25:1822–1825.
2. Shah YT. *Gas-liquid-solid reactor design*. New York: McGraw-Hill, 1979.
3. Holub RA, Dudukovic MP, Ramachandran PA. Pressure drop, liquid holdup, and flow regime transition in trickle flow. *AIChE J*. 1993; 39:302–321.
4. Aydin B, Larachi F. Trickle bed hydrodynamics and flow regime transition at elevated temperature for a Newtonian and a non-Newtonian liquid. *Chem Eng Sci*. 2005;60:6687–6701.
5. Wammes WJA, Middelkamp J, Huisman WJ, deBaas CM, Westerterp KR. Hydrodynamics in a cocurrent gas-liquid trickle bed at elevated pressure, Part 1: Gas-liquid interfacial areas. *AIChE J*. 1991; 37:1849–1853.
6. Wammes WJA, Middelkamp J, Huisman WJ, deBaas CM, Westerterp KR. Hydrodynamics in a cocurrent gas-liquid trickle bed at elevated pressure, Part 2: Liquid holdup, pressure drop, flow regimes. *AIChE J*. 1991;37:1854–1863.
7. Edouard D, Lacroix M, Pham Huu C, Luck F. Pressure drop modelling on foam: state of the art correlation. *Chem Eng J*. DOI: 10.1016/j.cej.2008.06.007.
8. Gianetto A, Specchia V. Trickle-bed reactor: state of art and perspectives. *Chem Eng Sci*. 1992;47:3197–3213.
9. Stemmet CP, Meeuwse M, van der Schaaf J, Kuster BFM, Schouten JC. Gas-Liquid mass transfer and axial dispersion in solid foam packing. *Chem Eng Sci*. 2007;62:5444–5450.
10. Topin F, Bonnet JP, Madani B, Tadrist L. Experimental analysis of multiphase flow in metallic foam: flow laws, heat transfer and convective boiling. *Adv Eng Mater*. 2006;8:890–899.
11. Lu TJ, Stone HA. Heat transfer in open-cell metal foams. *Acta Materialia*. 1998;46:3619–3635.
12. Richardson JT, Peng Y, Remue D. Properties of ceramic foam catalyst supports: pressure drop. *Appl Catal A: Gen*. 2000;204:19–32.
13. Groppi G, Tronconi E. Design of novel monolith catalyst supports for gas/solid reactions with heat exchange. *Chem Eng Sci*. 2000; 55:2161–2171.
14. Gibson LJ, Ashby MF. *Cellular solids, Structure and properties*, 2nd ed. Cambridge Solid State Science Series, Cambridge: Cambridge Univeraity Press, 2001.
15. Omatete OO, Janney MA, Nunn SD. J Eur Ceram Soc. 1997;17:407.
16. Fourie JG, Du Plessis JP. Pressure drop modelling in cellular metallic foams. *Chem Eng Sci*. 2002;5:2781–2789.
17. Moreira EA, Coury JR. The influence of structural parameters on the permeability of ceramic foams. *Braz J Chem Eng*. 2004;21:23–33.
18. Lacroix M, Nguyen P, Schweich D, Pham Huu C, Savin-Poncet S, Edouard D. Pressure drop measurements and modelling on SiC foams. *Chem Eng Sci*. 2007;62:3259–3267.
19. Stemmet CP, Jongmans JN, Van Der Schaaf J, Kuster BFM, Schouten JC. Hydrodynamics of gas-liquid counter-current flow in solid foam packings. *Chem Eng Sci*. 2005;60:6422–6429.
20. Stemmet CP, Van Der Schaaf J, Kuster BFM, Schouten JC. Solid foam packings for multiphase reactors. Modelling of liquid holdup and mass transfer. *Chem Eng Res Des*. 2006;84:1134–1141.
21. Larachi F, Laurent A, Wild G, Midoux N. Some experimental liquid saturation results in fixed-bed reactor operated under elevated pressure in cocurrent upflow and downflow of the gas and liquid. *Ind Eng Chem Res*. 1991;30:2404–2410.
22. Yang XL, Wild G, Euzen JP. Study of liquid retention in fixed bed reactors with upward flow of gas and liquid. *Int Chem Eng*. 1993; 33:72–84.
23. Stegeman DF, van Rooijen E, Kamperman AA, Weijer S, Westerterp KR. Residence time distribution in the liquid phase in a cocurrent gas-liquid trickle bed reactor. *Ind Eng Chem Res*. 1996;35:378–385.
24. Ellman MJ, Midoux N, Laurent A, Charpentier JC. A new improved pressure drop correlation for trickle-bed reactor. *Chem Eng Sci*. 1988; 43:2201.
25. Ellman MJ, Midoux N, Wild G, Laurent A, Charpentier JC. A new improved liquid holdup drop correlation for trickle-bed reactor. *Chem Eng Sci*. 1990;45:1667.
26. Carbonell RG. Multiphase flow models in packed beds. *Oil Gas Sci Technol-Revue de l'IFP*. 2000;55:417–425.
27. Saez AE, Carbonell RG. Hydrodynamic parameters for gas-liquid cocurrent flow in packed beds. *AIChE J*. 1985;31:52–62.
28. Ergun S. Fluid flow through packed columns. *Chem Eng Prog*. 1952;48:89–94.
29. Al-Dahhan MH, Dudukovic MP. Pressure drop and liquid holdup in high-pressure trickle bed reactors. *Chem Eng Sci*. 1994;49:5681–5692.
30. Iliuta I, Larachi F, Al-Dahhan MH. Double-Slit model for partially wetted trickle flow hydrodynamics. *AIChE J*. 2000;46:597.
31. Al-Dahhan MH, Khadilkar MR, Wu Y, Dudukovic MP. Prediction of pressure drop and liquid holdup in high-pressure trickle bed reactors. *Ind Eng Chem Res*. 1998;37:793.
32. Giani L, Groppi G. Mass-transfer characterization of metallic foams as supports for structured catalysts. *Ind Eng Chem Res*. 2005;44: 4993–5002.

Manuscript received Mar. 13, 2008, and revision received Jun. 13, 2008.