Hydrodynamics and mass transfer in trickle-bed reactors: an overview

Ion Iliuta, Arturo Ortiz-Arroyo, Faïcal Larachi, Bernard P.A. Grandjean, Gabriel Wild
Department of Chemical Engineering & CERPIC, Laval University, Québec, Canada G1K 7P4
Laboratoire des Sciences du Génie Chimique, 1 Rue Grandville, BP 451, 54001 Nancy, France

Abstract

The fluid dynamic and the gas–liquid mass transfer characteristics of trickle-bed reactors were revisited and their quantification methods reevaluated based on extensive experimental historic flow databases (22,000 experiments) set up from the open literature published over the last 50 years. The state-of-the-art of trickle-bed fluid dynamics was summarized and a set of unified and updated estimation methods relying on neural network, dimensional analysis and phenomenological hybrid approaches were discussed.

Keywords: Trickle-bed reactor; Hydrodynamics; Gas–liquid mass transfer

1. Introduction

The use of trickle-bed reactors (TBR) is pervasive in industrial multiphase catalytic processes and spans a broad range of applications from the manufacture of value-added products to the conversion of undesired chemicals into harmless and bio-compatible species (Al-Dahhan, Larachi, Dudukovic & Laurent, 1997). Despite nearly 50 years of worldwide research efforts, a satisfactory approach to trickle-bed reactors is still out of grasp. Even though fluid dynamics continue to be among its most intensely studied areas, TBR state-of-the-art is far from being complete and yet no universal approach has emerged as a panacea to predict conclusively TBR key fluid dynamic parameters. This is partly ascribable to the diverse entangling gas–liquid patterns met in a TBR which make such parameters depend in a complex manner on the fluid properties and throughputs, interfacial interactions and bed geometry.

The aim of this work is to present a cursory glance of 40 years research on TBR major fluid dynamic parameters (flow regime transition, liquid holdup, pressure drop, interfacial area, volumetric liquid- and gas-side mass transfer coefficients), and an attempt to provide the needed tools for better reactor design. Based on massive flow databases gathered from the 1954–1998 packed bed literature, this study pursues a threefold objective: (i) examine thoroughly the trends of hydrodynamic and gas–liquid mass transfer parameters as affected by changes in TBR operation conditions, (ii) assess the value of the numerous available general correlations/models, (iii) provide researchers and plant engineers with a set of unified and updated estimation methods using neural network, dimensional analysis and phenomenological combined approaches whose reliance is judged based on huge historic flow data bases.

2. Trickle-bed flow database

Table 1 describes the hydrodynamic and gas–liquid mass transfer databases in terms of ranges of fluid properties, operating conditions, particle and bed characteristics and gas–liquid systems:

- **Trickle-to-pulse flow transition database** (40 sources) contains 700 measurements over 30 gas–liquid systems, 18 column diameters, 38 packings.
- **Static liquid holdup database** (27 sources) includes 226 static holdup data on 45 liquids, 20 packings, 19 column sizes, 52 packing heights.
- **Liquid holdup–pressure drop database for low (LIR) and high interaction (HIR) regimes** (34 sources)
Table 1

Description of trickle bed flow database

<table>
<thead>
<tr>
<th>Fluid physical properties</th>
<th>Operating conditions</th>
<th>Limits of parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>650 ≤ ρL ≤ 13545</td>
<td>h = 9 × 10⁻⁶ ≤ e, S = 6.45 × 10⁻¹</td>
<td>1.5 × 10⁻² ≤ ReL ≤ 2.7 × 10³</td>
</tr>
<tr>
<td>3 × 10⁻⁴ ≤ ρL ≤ 0.472</td>
<td>u = 2 × 10⁻⁴ ≤ e, S ≤ 4.5</td>
<td>1.94 × 10⁻¹ ≤ ReL ≤ 1.0 × 10⁴</td>
</tr>
<tr>
<td>1 × 10⁻² ≤ e ≤ 0.485</td>
<td>0.1 ≤ P ≤ 10 MPa</td>
<td>1.0 × 10⁻⁷ ≤ WeL ≤ 3.65</td>
</tr>
<tr>
<td>0.16 ≤ νL ≤ 116.4</td>
<td>13 ≤ T ≤ 350°C</td>
<td>6.85 × 10⁻³ ≤ Xu ≤ 5.01 × 10²</td>
</tr>
<tr>
<td>1.5 × 10⁻³ ≤ μL ≤ 2.4 × 10⁻³</td>
<td>0.046 ≤ eL ≤ 0.62</td>
<td>2.843 × 10⁶ ≤ P ≤ 6.223 × 10⁶</td>
</tr>
<tr>
<td>4.76 × 10⁻¹ ≤ Dl ≤ 0.40 × 10⁻⁹</td>
<td>2.13 × 10⁻³ ≤ kLa ≤ 7.04</td>
<td>2.36 × 10⁻³ ≤ kLa ≤ 6.94</td>
</tr>
<tr>
<td>6.6 × 10⁻⁶ ≤ Dl ≤ 1.6 × 10⁻³</td>
<td>23.4 ≤ α ≤ 9070</td>
<td>23.4 ≤ α ≤ 9070</td>
</tr>
</tbody>
</table>

Geometrical properties of packings and columns

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle diameter, dp (m)</td>
<td>10⁻⁵ ≤ dp ≤ 5.64 × 10⁻²</td>
</tr>
<tr>
<td>Column diameter, dc (m)</td>
<td>1.4 × 10⁻² ≤ dc ≤ 5.1 × 10⁻¹</td>
</tr>
<tr>
<td>Bed porosity, e</td>
<td>0.26 ≤ e ≤ 0.94</td>
</tr>
<tr>
<td>Shape factor, φ</td>
<td>0.133 ≤ φ ≤ 1.0</td>
</tr>
<tr>
<td>Static liquid holdup</td>
<td>0.00436 ≤ eL, eL ≤ 0.116</td>
</tr>
<tr>
<td>Particle material</td>
<td>Glass, ceramics, porous alumina, polyethylene, polypropylene, carbon, steel, CuO/ZnO, SiO₂/Al₂O₃/Cr₂O₃ catalysts, quartz</td>
</tr>
<tr>
<td>Particle shape</td>
<td>Spherical, cylindrical, extrudates, Raschig &amp; Pall rings, Berl saddle, three-lobed</td>
</tr>
</tbody>
</table>

**Liquids**

Hydrodynamics: H₂O, H₂O + EtOH [0.1-94%], H₂O + teepol, H₂O + 40% sucrose, H₂O + NaOH [0.2-2.4 N], H₂O + DIPA [1.8 M], H₂O + Na₂SO₄ [0.8 M], H₂O + 2%Na₂SO₄, H₂O + Na₂SO₄, H₂O + 2% (CH₃)₂CO, H₂O + 0.038%H₂SO₄Na, H₂O + [1.5-48%] i-ProOH, H₂O + [10-20 ppm] C₂H₅OH, H₂O + [0.1-2%] tritton X100, H₂O + 0.1% tylose, H₂O + 0.5 ppm lutensol LF711, H₂O + [1-2.5 ppm] tergipol NPP, H₂O + [9-90%] C₂H₅OH, H₂O + [0.1-2%] DEA [1.5 M], H₂O + ETG 40%, H₂O + MEA [0.25-1.26 M], H₂O + [40-80%] MeOH, H₂O + 0.5 NaOH + K₂Fe(CN)₆, K₂Fe(CN)₆, MeOH, C₂H₅OH, PC, EtOH, EtOH + DEA [0.645 M], EtOH + MEA [0.183 M], ETG, ETG + MEA [1.43 M], ETG + DEA [0.061-1.88 M], C₂H₅ + CH₃ + 10% IPA, C₂H₅ + 10% IPA + DIPA, DMA, n-C₃H₇, C₂H₅OH, soybean oil, petroleum ether, gasoline, non-desulfurized gas oil, desulfurized gas oil, kerosene, H₂O + CMC [0.1-1%], PET, H₂O + PET 40%, Dupont petrowet aqueous solutions [10-50%], H₂O + C₆H₄(OH) [1-15%], H₂O + CaCl₂ [10-50%], Cerrobend (Pb-Bi-Cd-Sn alloy), Hg.

Gas mass transfer: H₂O, H₂O + C₆H₅OH, H₂O + NaOH [0.1-2 N], H₂O + Na₂SO₄ [0.8 M], H₂O + Na₂SO₄ [0.25 M] + CO²⁻, H₂O + Na₂SO₄ [0.8 M] + CO₃²⁻, H₂O + NaOH [0.5-2 N] + Na₂SO₄, H₂O + K₂CO₃ [0.8-1.3 M] + KHCO₃ [0.65-1.2 M], H₂O + Na₂SO₄, H₂O + MEA [0.25-0.33 M], DMA, H₂O + DEA [1.5-2 M], H₂O + DIPA [2.4 M], H₂O + DEA [1.5 M] + ETG [20-40%], H₂O + 40% CaCl₂, H₂O + C₆H₅OH, H₂O + CH₃ [0.227-1.77 M] + toluene + 10% IPA, H₂O + EtOH + MEA [0.392 M], H₂O + EtOH + DEA [0.0435-0.875 M], H₂O + EtOH + DEAE [0.0435-1.88 M], ETG + MEA, EtOH + MEA [0.2-0.7 M], EtOH + DEA [0.6-0.8 M], IPA + C₆H₅ + CH₃ [0.1-0.7 M], C₆H₅ + 10% IPA + DIPA, p-C₆H₄(OH) + 10% i-ProOH + CH₃, n-C₆H₅OH + MEA, H₂O + CMC [0.1-1.0%].

**Gases**

Hydrodynamics: air, air + CO₂, air + SO₂, air + O₂, H₂, Ar, CO₂, He, N₂.

Gas-mass transfer: He, air, air + CO₂, air + SO₂, air + O₂, N₂, CO₂, N₂, O₂, CO₂.

CHA = cyclohexylamine, CMC = carboxymethylcellulose, DEA = diethanolamine, DIPA = diisopropanolamine, DMA = dimethylamine, ETG = ethylene glycol, EtOH = ethanol, IPA = isopropanolamine, MEA = mono-ethanolamine, i-ProOH = isopropyl alcohol, C₆H₅O₂ = glycerol, C₆H₅H₂ = cyclohexane, C₄H₅OH = heptanol, PET = polyethylene-glycol, PC = propylene carbonate, C₆H₅ = toluene, p-C₆H₄(OH) = p-xylene, C₆H₅OH = naphthalene, (CH₂)₄CO = acetone, C₆H₄(OH) = sorbitol.

Table 1 encompasses over 17,800 measurements on 74 gas-liquid systems, 29 column diameters, 47 packing materials. Holdup and pressure drop data were recast into total external liquid holdup and frictional pressure gradients. The data were categorized in LIR and HIR classes using a state-of-the-art transition regime correlation developed from the above trickle-to-pulse flow regime transition data.

- Gas-liquid mass transfer data base (70 references) consists of more than 3200 experiments for 52 gas-liquid systems, beyond 60 packing sizes and geometries, 17 column diameters.

Property and operation conditions swept broad ranges up to high pressure/temperature (10 MPa/350°C), Newtonian, non-Newtonian, coalescing, non-coalescing, aqueous, organic, pure and mixed liquids. The data were typical of moderate-porosity trickle beds and high-porosity packed absorption/desorption towers. Bed inventories included few cm size packings (Raschig and Pall
rings, Intalox, Berl saddles) for a bed porosity up to 94%, and few mm size packings (porous and non-porous spheres, extrudates and pellets, three-lobed) for a moderate bed porosity (45% and down). Four order of magnitudes were spanned by the superficial fluid velocity ratio for the mass transfer parameters and the interfacial areas evolved from partial to complete wetting conditions.

3. Artificial neural network regression strategy

From considerations stated elsewhere (Bensetiti, Larachi, Grandjean & Wild, 1997; Iliuta, Larachi & Grandjean, 1998), it is proposed to analyze the impact of fluid velocities, fluid viscosity and density, liquid surface tension, solute diffusivity, gravitational acceleration, particle size and shape, bed diameter and porosity via their combinations into the various dimensionless groups that may affect the hydrodynamic and gas–liquid interfacial mass transfer parameters. Many hundreds of sets of dimensionless groups were thus tested by trial and error with the ultimate objective of identifying the optimal sets of groups best describing each of the hydrodynamic and mass transfer parameters via artificial neural network correlations. The choice of the best sets rests on the fulfillment of the following criteria:

- the optimal set must contain a minimum number of selected dimensionless groups;
- each group (input variable) must be highly cross-correlated with the corresponding hydrodynamic or mass transfer parameter group (output to be predicted);
- the optimal set must lead to the best output prediction, i.e. minimal absolute average relative error and variance;
- the neural network architecture must be of minimal complexity, i.e. the least number of hidden neurons giving the smallest errors both on the learning (70% of the database) and the generalization files (on the remaining population).

Three-layer feed-forward neural network models were designed, using NNFIT software (Cloutier, Tibirna, Grandjean & Thibault, 1996), to derive the needed hydrodynamic and mass transfer correlations. The neural architectures are described by the generic equations (1) and (2) (Table 2) that correlate the network outputs, $S_p$, to sets of normalized input variables, $U_{ik}$. In these equations, $U$ and $H$ define the input and hidden layer vectors, $H_{J+1,k}$ and $U_{J+1,k}$ are the bias constants set equal to 1, $\omega_{ijk}$ and $\phi_{ijk}$ are the weights or the fitting parameters of the neural network models and $J$ is the number of nodes in the hidden layer. The network fitting parameters are a priori unknown, and they have to be determined using a training algorithm by performing a nonlinear least-squares regression over known pseudo-random sets of inputs/outputs (70% of the database). The weights are set as to minimize the training error on the training set using a quadratic objective function which is minimized by the quasi-Newton–Broyden–Fletcher–Goldfarb–Shanno algorithm. A good measure for the extrapolation performance of a well-trained neural network is given by the generalization error which should be comparable to the training error in the case of inputs/outputs not presented during the learning step to the neural network (i.e., 30% of the remaining data). For each of hydrodynamic or mass transfer parameter neural network, the number of hidden neurons, $J$, was varied from 3 to 15. Hidden layers with 3 neurons (for $\phi_{L}$, c-HIR and $\Psi_{L}$-LIR), 4 neurons (for $\Psi_{L}$-HIR), 6 neurons (trickle-to-pulse flow regime transition), 8 neurons (for $Sh_{ad}$), 11 neurons (for $ad_{s}/(1 - \varepsilon)$) and 13 neurons (for $Sh_{L}$) were found to be the optimal neural architectures leading to the smallest average absolute relative errors and standard deviations on the training and the generalization sets.

4. Flow regimes

TBRs operate in a variety of flow regimes ranging from gas-continuous to liquid-continuous patterns. They usually fall into two broad categories referred to as low (LIR) and high (HIR) interaction regimes. While the LIR (trickle flow) manifests at low gas and liquid throughputs, the HIR (i.e., pulse, spray, and bubble/dispersed bubble flow regimes) takes place for moderate-to-high fluid flow rates leading to significant gas–liquid interfacial shears. Because the trickle and pulse flow regimes are of particular industrial interest, the majority of the literature work focused on the trickle-to-pulse flow transition.

The following current trends emerge from the analysis of the trickle-to-pulse flow regime transition database: (i) trickle-to-pulse flow transition is barely affected by intermediate-range bed porosity (0.34 < $\varepsilon$ < 0.40) and particle sizes up to 5 mm. For high porosity beds or large size particles a shift of this transition towards higher liquid throughputs is observed; (ii) a significant drop of the liquid transitional velocity occurs with increasing liquid viscosity; (iii) low surface tension liquids exhibit a peculiar trend as higher liquid transitional velocities occur at higher rather than at lower superficial gas velocities; (iv) an increase in liquid density narrows down the trickle flow region; and (v) the trickle flow domain widens at elevated pressures.

Many flow regime charts and attempts at modeling trickle-to-pulse flow regime changeover have been proposed, a summary of which is available in Saroha and Nigam (1996) and Al-Dahhan et al. (1997). Because of the large number of variables affecting the trickle-to-pulse flow transition, compression into two or three flow chart coordinates seems illusive. Furthermore, since the choice of the compressing variables is not unanimous, the least
Table 2

Set of equations for the neural network correlations

\[ S_k = \frac{1}{1 + \exp \left( -\sum_{l=1}^{L} \omega_{kl} H_l \right)} \]  

(1)

\[ H_k = \frac{1}{1 + \exp \left( -\sum_{l=1}^{L} \omega_{kl} U_l \right)} \]  

(2)

Trickle-to-pulse flow regime transition \((I = 7, J = 6, k = 1)\)

\[ S_1 = \frac{\log v_{st,u} + 3.0804}{3.0} \]  

(3)

\[ U_{11} = \frac{\rho_L - 650}{496}, \quad U_{21} = \frac{\log \mu_L + 3.5086}{2.331}, \quad U_{31} = \frac{\sigma_L - 1.31 \times 10^{-2}}{5.89 \times 10^{-2}}, \quad U_{41} = \frac{\log \varepsilon_{st} + 3.2449}{3.5504} \]  

(4)

\[ U_{51} = \frac{\log \rho_L + 0.7959}{2.8619}, \quad U_{61} = \frac{\log d_p + 2.9355}{1.121}, \quad U_{71} = \frac{\varepsilon - 0.335}{0.405}, \quad U_{81} = 1 \]  

(5)

\( \varepsilon_{st,u} \) - HIR \((I = 5, J = 3, k = 2)\)

\[ S_2 = \frac{\log \varepsilon_{st} + 1.334}{1.1286} \]  

(6)

\[ U_{12} = \frac{\log \text{Re}_L + 0.8105}{4.2485}, \quad U_{22} = \frac{\log \text{Re}_L + 0.7102}{4.7122}, \quad U_{32} = \frac{\log \text{We}_L + 4.148}{4.7104} \]  

(7)

\[ U_{42} = \frac{\log X_L + 2.7}{4.063}, \quad U_{52} = \frac{\log S_k - 0.2068}{1.2942}, \quad U_{62} = 1 \]  

(8)

\( \Psi_L - \) LIR \((I = 6, J = 3, k = 3)\)

\[ S_3 = \frac{\log \Psi_L - 0.0004341}{1.4225} \]  

(9)

\[ U_{13} = \frac{\log \text{Re}_L + 3.301}{5.332}, \quad U_{23} = \frac{\log \text{Re}_L + 0.7113}{3.6513}, \quad U_{33} = \frac{\log \text{We}_L + 8.199}{6.5} \]  

(10)

\[ U_{43} = \frac{\log \text{Ga}_{L} - 0.5932}{4.9638}, \quad U_{53} = \frac{\log X_L + 3.688}{6.174}, \quad U_{63} = \frac{\log S_k - 0.2127}{0.4777}, \quad U_{73} = 1 \]  

(11)

\( (\Psi_L - 1) - \) HIR \((I = 6, J = 4, k = 4)\)

\[ S_4 = \frac{\log (\Psi_L - 1) - 0.00173}{1.9273} \]  

(12)

\[ U_{14} = \frac{\log \text{Re}_L + 0.8105}{4.2025}, \quad U_{24} = \frac{\log \text{Re}_L + 0.7102}{4.7122}, \quad U_{34} = \frac{\log \text{We}_L + 4.148}{4.7102} \]  

(13)

\[ U_{44} = \frac{\log \text{Ga}_{L} - 1.025}{5.229}, \quad U_{54} = \frac{\log X_L + 2.689}{4.052}, \quad U_{64} = \frac{\log S_k - 0.2068}{0.6869}, \quad U_{74} = 1 \]  

(14)

\( \text{Sh}_{L} \) \((I = 6, J = 13, k = 5)\)

\[ S_5 = \frac{\log \text{Sh}_{L} + 0.4738}{5.2428} \]  

(15)

\[ U_{15} = \frac{\log \text{Re}_L + 1.7002}{4.898}, \quad U_{25} = \frac{\log \text{We}_L + 5.7326}{6.298}, \quad U_{35} = \frac{\log X_L + 2.5790}{4.918} \]  

(16)
to be said is that a universal flow chart/correlation is non-existing and that each flow chart/correlation can only be employed interpolatively for conditions falling within those that served to its establishment. To circumvent these setbacks, some conceptual flow regime changeover models were proposed (Sicardi & Hofmann, 1980; Blok, Varkevisser & Drinkenburg, 1983; Ng, 1986; Grosser, Carbonell & Sundaresan, 1988; Holub, Dudukovic & Ramachandran, 1992). Unfortunately, all recent experimental studies reach the same conclusion that none of them is yet entirely successful and no single approach can be recommended.

Using the flow regime transition database, Larachi, Iliuta, Chen and Grandjean (1999) developed an explicit correlation for trickle-to-pulse flow changeover (Table 2) based on neural network modeling. The overall result was an improved prediction of this transition correlation (Fig. 1). Systematically for this correlation (as well as the forthcoming ones) 70% of the database served to train the network while the remaining 30% were used to test its robustness capability. The figure shows a very good agreement for the whole database measurements. For comparison, the parity plot shows also predictions by Larachi, Laurent, Wild and Midoux (1993) correlation, which, among the literature correlations, provided the less-scattered predictions.

![Fig. 1. Predicted versus experimental liquid superficial velocity at the trickle-to-pulse flow changeover.](image)

### Table 2 (continued)

\[
\begin{align*}
U_{a8} &= \log M_{ol} + 14.5943 \quad 10.114, & U_{a8} &= \log S_{cL} - 1.6467 \quad 4.042, & U_{a8} &= \log S_{L} - 0.4069 \quad 1.9011, & U_{a8} &= 1 \\
S_{b6} &= \log Sh_{G} + 3.8824 \quad 6.752 \\
U_{s6} &= \log Re_{L} + 0.6800 \quad 2.6751, & U_{s6} &= \log S_{cL} + 6.3642 \quad 3.615, & U_{s6} &= \log W_{GG} + 6.7667 \quad 6.8823 \\
U_{e6} &= \log X_{G} + 0.0815 \quad 0.55765, & U_{e6} &= \log S_{cG} + 2.6330 \quad 4.393, & U_{e6} &= \log S_{G} - 0.2405 \quad 2.0205, & U_{e6} &= 1 \\
S_{7} &= \log [ad_{a}/(1 - \sigma)] + 2.0021 \quad 4.741 \\
U_{a7} &= \log Re_{L} + 0.9969 \quad 4.077, & U_{a7} &= \log Re_{G} - 0.2327 \quad 3.4213, & U_{a7} &= \log W_{GL} + 4.8945 \quad 5.2032 \\
U_{a7} &= \log Fr_{L} + 4.9633 \quad 5.0172, & U_{a7} &= \log X_{G} + 1.9674 \quad 3.651, & U_{a7} &= \log E_{om} + 1.5728 \quad 4.906 \\
U_{a7} &= \log S_{b} - 0.2188 \quad 2.1742, & U_{a7} &= 1 \\
\end{align*}
\]

5. Total external liquid holdup and frictional two-phase pressure drop

The following current trends emanate from the analysis of the LIR and HIR databases: (i) liquid holdup is an
increasing function of liquid velocity and viscosity, particle diameter; it is a decreasing function of gas superficial velocity and liquid surface tension; (ii) liquid holdup decreases as gas density increases, except for very low gas velocities where it is insensitive to gas density; (iii) non-coalescing liquids exhibit much smaller holdups (and much higher pressure drops) than coalescing liquids; (iv) gas viscosity appears to have marginal effect on liquid holdup and pressure drop; and (v) mixing with fines improves liquid holdup at the expense of increased pressure drop. Increasing gas and liquid superficial velocities or mass fluxes, liquid viscosity, gas density, or decreasing particle diameters increases pressure drops.

Correlations for total external or dynamic liquid holdups and two-phase pressure drops were summarized by Al-Dahhan et al. (1994), Saroha and Nigam (1996) and Al-Dahhan et al. (1997), the majority of them being mostly empirical and restricted to their specific narrow ranges of process conditions. Comparison of the prediction with measurements from the constructed flow databases shows that no method emerges as clearly superior even though those relying on phenomenological considerations seem more reliable.

For the LIR, Holub et al. (1992) pore-scale two-fluid separated-flow phenomenological model and its two extensions (Al-Dahhan, Khadilkar, Wu & Dudukovic, 1998; Iliuta et al., 1998) are recommended for liquid holdup predictions. With an AARE = 15.6%, Holub et al. (1992) shear/slip free model performed better than Ellman, Midoux, Wild, Laurent and Charpentier (1990) (AARE = 25%) and Larachi, Laurent, Midoux and Wild (1991) (AARE = 19.5%) correlations. Implementation of Al-Dahhan et al. (1998) and Iliuta et al. (1998) corrective correlations for non-zero gas–liquid interfacial shear stress and slip velocity into the extended Holub et al. (1992) model led to improved LIR liquid holdup predictions. This is illustrated by Fig. 2 parity plot using the neural network–dimensionless group approach along with the LIR/HIR pressure drop (both for LIR and HIR) and two-phase pressure drop (both for LIR and HIR) are therefore derived using a neural network–dimensionless group approach along with the LIR/HIR flow databases. The liquid holdup and the pressure drop neural-based correlations are given using the same generic equations Eqs. (1) and (2) (Table 2).

For the HIR liquid holdup, the normalized input and output variables of the neural network model are listed in Table 2, the neural network weights are listed in Table 3 and a parity plot is shown in Fig. 2. The present correlation performs 2–3 times better (AARE = 13.6%) than Ellman et al. (1990) and Larachi et al. (1991) correlations.

For the LIR and HIR pressure drop, the normalized input and output variables of the neural network model are shown in Table 2. Table 4 lists the neural network weights for both correlations and Fig. 3 shows the corresponding parity plots. These two correlations outperform all existing correlating tools of pressure drops in both the low (AARE = 16.1%) and the high interaction (AARE = 23.5%) regimes.
Table 4
Fitting parameters of the neural network correlations for pressure drop: LIR \((I = 6, J = 3, K = 3)\) HIR \((I = 6, J = 4, K = 4)\)

<table>
<thead>
<tr>
<th>(\omega_{ijk})</th>
<th>(1)</th>
<th>(2)</th>
<th>(3)</th>
<th>(1)</th>
<th>(2)</th>
<th>(3)</th>
<th>(4)</th>
<th>(5)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>10.992</td>
<td>2.4807</td>
<td>6.1939</td>
<td>7.0682</td>
<td>1.4293</td>
<td>25.119</td>
<td>0.5418</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1.1129</td>
<td>1.1107</td>
<td>0.2757</td>
<td>3.3369</td>
<td>6.1989</td>
<td>4.8916</td>
<td>2.005</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>4.1217</td>
<td>1.3762</td>
<td>1.8968</td>
<td>6.8822</td>
<td>12.295</td>
<td>12.385</td>
<td>1.1747</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>1.4837</td>
<td>6.3772</td>
<td>3.7145</td>
<td>1.3469</td>
<td>24.891</td>
<td>1.8018</td>
<td>1.0354</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>9.9196</td>
<td>2.3686</td>
<td>1.8795</td>
<td>12.295</td>
<td>12.385</td>
<td>1.1747</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>4.6801</td>
<td>4.9743</td>
<td>1.3469</td>
<td>24.891</td>
<td>1.8018</td>
<td>2.6449</td>
<td>1.0354</td>
<td></td>
</tr>
</tbody>
</table>

6. Gas–liquid interfacial areas and interphase mass transfer coefficients

Analysis of the gas–liquid mass transfer database reveals that: (i) gas–liquid interfacial area, \(a\), increases with liquid and gas flow rates and liquid viscosity; (ii) \(a\) for non-coalescing liquids exceeds that for coalescing ones (Midoux, Morsi, Purwasasmita, Laurent & Charpentier, 1984); (iii) \(a\) for organic liquids does not differ significantly from that for ionic liquids (Mahajani & Sharma, 1979); and (iv) \(a\) is higher for non-spherical packings (Larachi, Cassanello & Laurent, 1998). Liquid-side volumetric mass transfer coefficients, \(k_{L,a}\), are higher for viscous liquids (Midoux et al., 1984; Iliuta & Thyron, 1997). At elevated pressure both \(k_{G,a}\) and \(a\) improve. The scarcity of gas-side volumetric mass transfer coefficient is noteworthy; and to the best of our knowledge no experimental data on \(k_{G,a}\) are available for high pressure conditions. Since \(k_{G}\) is expected to diminish with the increase in gas density, disregarding this fact can lead to overestimated overall gas–liquid mass transfer coefficients at high pressure, especially for a mixture of gases involving a highly soluble reactant.

Correlations/models for gas–liquid interfacial area and interphase volumetric gas–liquid mass transfer coefficients in TBRs were summarized by Saroha and Nigam (1996), Al-Dahhan et al. (1997) and Larachi et al. (1998). Systematic testing against the 3200 data of the gas–liquid mass transfer historic database revealed the inconclusiveness of available methods. For the purpose of illustration, predictions by Wild, Larachi and Charpentier (1992) and Yaïci, Laurent, Midoux and Charpentier (1985) correlations, considered among the best correlations, are shown in Figs. 4–6 parity plots along with their corresponding AARE. Their discrepancies are well contrasted when the...
predictions by these correlations are compared to neural network–dimensionless group correlations developed by Iliuta, Larachi, Grandjean and Wild (1999) (see also Figs. 4-6). Despite gas–liquid mass transfer parameters are strongly dependent on flow regimes, these correlations are capable to predict the gas–liquid mass transfer parameters regardless of the prevailing flow regime.

7. Conclusion

New correlations for fluid dynamic parameters were developed for TBR using an extensive historic fluid dynamic database and combined approaches relying on feed-forward neural networks, dimensional analysis and phenomenological models. The database, built from the numerous accessible sources published in the period 1954–1998, consisted of more than 22,000 measurements on (dynamic, static, total, external) liquid holdup, frictional pressure drop, trickle-to-pulse flow regime transition, gas–liquid interfacial areas, volumetric gas- and liquid-side mass transfer coefficients visiting a vast operation condition space including high pressure/temperature: up to 10 MPa, 350°C, >100 liquids: Newtonian, non-Newtonian, coalescing, non-coalescing, pure, mixed, aqueous, organic, >10 gases: from H₂ to CO₂, and ca. 100 packings: of different shapes and sizes. As more experimental data will be released in the literature for new unexplored operating domains, it is expected that the proposed data base will be constantly updated and the correlations re-calibrated (if needed) in order to provide the designers with the most accurate tools to estimate trickle-bed fluid dynamic parameters. Updates and re-tuned correlations are available at the web address: http://www.gch.ulaval.ca/~flarachi.

Notation

\( a \) gas–liquid interfacial area, \( \text{m}^2/\text{m}^3 \)
\( a_s \) external area of particles and wall per unit reactor volume, \( (=6(1-\varepsilon)/\rho_d + 4/\rho_d)^{1/3}/\text{m}^3 \)
\( \text{AARE} \) average absolute relative error, \( (=1/N) \sum_{i=1}^{N} (y_{\text{calc},i} - y_{\text{exp},i})/y_{\text{exp},i} \)
\( d_c \) column diameter, \( \text{m} \)
\( d_h \) Krischer and Kast hydraulic diameter \( (=d_p \sqrt[3]{16\varepsilon/9\pi(1-\varepsilon)^2}) \)
\( d_p \) grain volume mean diameter, \( \text{m} \)
\( D_a \) diffusivity in \( \alpha \)-phase, \( \text{m}^2/\text{s} \)
\( E_o_m \) modified Eötvös number \( (=\rho_d g d_p^2 \varepsilon^2/\sigma_L (1-\varepsilon)^2) \)
\( F_{r_L} \) liquid Froude number \( (=v_{SL}/g d_p) \)
\( g \) acceleration due to gravity, \( \text{m}/\text{s}^2 \)
\( G_a \) Galileo number \( (=d_p^3 \rho_d^2 g \varepsilon^3/[\mu_L^2(1-\varepsilon)^3]) \)
\( k_{\alpha} \) \( \alpha \)-phase side volumetric mass transfer coefficient, \( \text{s}^{-1} \)
\( M_{o_L} \) liquid Morton number \( (=g \mu_L^3/\rho_L \sigma_L^4) \)
\( \Delta P/H \) two-phase frictional pressure drop, \( \text{Pa}/\text{m} \)
\( R_e_s \) Reynolds number \( (=v_{SGp}\rho_d^2/\mu_d(1-\varepsilon)) \)
\( R_e_s' \) Reynolds number \( (=v_{SGp}\rho_d^2/\mu_s) \)
\( S \) network normalized output
\( S_b \) bed correction function \( (=a_d b/(1-\varepsilon)) \)
\( S_{c_\alpha} \) \( \alpha \)-phase Schmidt number \( (=\mu_s/D_p \rho_s) \)
\( S_{h_\alpha} \) \( \alpha \)-phase Sherwood number \( (=k_{\alpha} a_d d_p^2/D_p) \)
\( S_{t_L} \) liquid Stokes number \( (=\mu_L v_{SL}/\rho_L g d_p^2) \)
\( U_i \) normalized input variables
\( v_{SGp} \) superficial velocity of \( \alpha \) phase, \( \text{m}/\text{s} \)
\( X_{Gr}, X_L \) Lockhart–Martinelli numbers, \( (=v_{SGp}^{1/2}/\rho_{SGp}^{1/4})^{1/2}, X_L = 1/X_G \)
\( y \) hydrodynamic/gas–liquid mass transfer parameter
\( W_{e_\alpha} \) \( \alpha \)-phase Weber number \( (=v_{SGp}^2 \rho_d^2/\rho_s \sigma_L) \)

Fig. 5. Whole database parity plot of predicted versus experimental gas Sherwood number (neural network–dimensionless group correlation).

Fig. 6. Whole database parity plot of predicted versus experimental interfacial area (neural network–dimensionless group correlation).

I. Iliuta et al. / Chemical Engineering Science 54 (1999) 5329–5337
Greek letters

$\chi$ subscript meaning gas (G) or liquid (L)
$\varepsilon$ bed void fraction
$\delta_{L,T}$ total external liquid holdup
$\mu_s$ viscosity of $x$ phase, kg/m s
$\rho_s$ density of $x$ phase (kg/m$^3$)
$\sigma$ standard deviation ($= 1/(N-1)$)
$\omega$ network weights
$\Psi_L$ dimensionless body force on liquid phase

$=(\Delta P/H)/\rho_L g + 1$ 

Acknowledgements

Financial support from the Natural Sciences and Engineering Research Council of Canada (NSERC), the Fonds pour la Formation de Chercheurs et d’Aide à la Recherche (Québec) is gratefully acknowledged. Part of this work was done in the frame of the “Coopération scientifique franco-québécoise” (Programme Conception assistée par ordinateur – Génie des Procédés). The financial support of the Ministère des Affaires Étrangères (France), the Ministère des Relations Internationales (Québec, Canada) is gratefully acknowledged. We also express our appreciation to Dr. Z. Benetsetti and Ms. A. André for the collection of the databases. We are also indebted to many contributors who provided us with theses and reports, among them we are thankful to Prof. M. Al-Dahhan (Washington University), Dr. J. P. Euzen (IFP, France), Profs. A. Lakota and J. Levec (University of Ljubljana, Slovenia), Prof. N. Papayannakos (NTU, Athens, Greece), Prof. Y. B. G. Varma (IIT Madras, India), Prof. W. P. M. van Swaaij from Twente University (The Netherlands) and Drs. K. Tahraoui and W. J. A. Wammes.

References