Improving the prediction of liquid back-mixing in trickle-bed reactors using a neural network approach

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Abstract: Current correlations aimed at estimating the extent of liquid back-mixing, via an axial dispersion coefficient, in trickle-bed reactors continue to draw doubts on their ability to conveniently represent this important macroscopic parameter. A comprehensive database containing 973 liquid axial dispersion coefficient measurements (\(D_{AX}\)) for trickle-bed operation reported in 22 publications between 1958 and 2001 was thus used to assess the convenience of the few available correlations. It was shown that none of the literature correlations was efficient at providing satisfactory predictions of the liquid axial dispersion coefficients. In response, artificial neural network modeling is proposed to improve the broadness and accuracy in predicting the \(D_{AX}\) whether the Piston–Dispersion (PD), Piston–Dispersion–Exchange (PDE) or PDE with intra-particle diffusion model is employed to extract the \(D_{AX}\). A combination of six dimensionless groups and a discrimination code input representing the residence-time distribution models are used to predict the Bodenstein number. The inputs are the liquid Reynolds, Galileo and Eötvös numbers, the gas Galileo number, a wall factor and a mixed Reynolds number involving the gas flow rate effect. The correlation yields an absolute average error (AARE) of 22% for the whole database with a standard deviation on the AARE of 24% and remains in accordance with parametric influences reported in the literature.

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Keywords: trickle-bed; gas–liquid downflow; liquid back-mixing database; neural network

NOTATION

\[ \text{AARE} = \frac{1}{N_D} \sum_{i=1}^{N_D} \left| \frac{\text{exp}(i) - \text{pred}(i)}{\text{exp}(i)} \right| \]

ANN

\( \alpha \)
\( \beta \)
\( \gamma \)
\( \omega \)
\( \nu \)
\( \xi \)
\( \eta \)
\( \zeta \)
\( \theta \)
\( \varphi \)
\( \chi \)
\( \psi \)
\( \phi \)
\( \psi \)
\( \omega \)

\( f_z \)
\( g \)
\( G_a \)
\( h_z \)
\( K \)
\( N \)

\( B_0 \)
\( d \)
\( d_h \)
\( d_v \)
\( D_{AX} \)
\( D_C \)
\( D_{eff} \)
\( E_0 \)

\( P_L \)
\( \rho_L \)

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\[ R_{e2} = (\mu, U_2)/(a_2 \mu_2) \]

**RTD**

Residence-time distribution

**S**

Normalized output variable

\[ S_a = \frac{\rho_g}{U_g} \]

\[ U_g = U_i \]

\[ U_i \]

**t**

Time (s)

**T**

Temperature (K)

\[ z \]

Axial coordinate (m)

**Z**

Active bed height (m)

\[ \varepsilon \]

Bed porosity (dimensionless)

\[ \mu_z \]

\[ \mu \]

\[ \rho_z \]

\[ \rho \]

\[ \sigma \]

Standard deviation

\[ \sigma_L \]

Liquid phase surface tension (N m\(^{-1}\))

\[ \phi \]

Particle sphericity (dimensionless)

\[ \omega \]

Neural net fitting parameters

**Subscripts**

D

Dynamic liquid zone

G

Gas

I

Intraparticle liquid zone

L

Liquid

S

Stagnant liquid zone

T

Total liquid, all zones combined

1 INTRODUCTION

Trickle bed reactors (TBR), which are catalytic fixed beds contacted by concurrent downward gas-liquid flows, host a variety of gas-liquid-solid catalytic reactions especially in the petroleum, petrochemical and biotechnology industries. The economic impact of how well these reactors operate is considerable since in the petroleum sector alone the annual processing capacity for various hydrocracking treatments (eg desulfurization, hydrocracking, metals removal, denitrification, etc) exceeds the billion metric tons.\(^1\) With the permanent market shift towards increasing demand for light oil products (naphtha, middle distillates, gas oil), and the decreasing needs for heavy cuts, it is forecast that the refiners will keep improving their ‘bottom-of-the-barrel’ processing units for upgrading heavy oil and residual feedstocks.\(^1\) Any advance in TBR technology, even tiny, will represent substantial cost savings to justify continued research aimed at the improvement of TBR operation and performance.

Reliable design of trickle-bed reactors largely depends on the accuracy of the design models calibrated using laboratory-scale data. Among the most important issues related to TBR macroscopic material transport is the liquid back-mixing which characterizes how much the liquid phase deviates from ideal plug flow through the catalytic bed due to hydrodynamic non-idealities. One simple tool for assessing whether the role of back-mixing is neutral or detrimental to TBR catalytic performance consists of evaluating the liquid axial dispersion coefficient.\(^3\) Then, to maximize reactor productivity, operating flow conditions are tuned in such a way to promote liquid plug flow. Due to the complex nature of the gas–liquid–solid pattern developing in trickle beds, such a choice of operating conditions is not trivial. Therefore, procurement of an accurate tool for predicting the impact of TBR operation on liquid back-mixing is crucial for reactor design.

Several residence-time distribution (RTD) models, summarized in Table 1, have been proposed from which a liquid axial dispersion coefficient is identified. Usually, RTD curves are fitted using multiple-parameter species balance differential equations (RTD model), allowing the extraction of a liquid axial dispersion coefficient (\(D_{AX}\)) or its corresponding dimensionless Péclet number.\(^3\) A wide arsenal of parameter estimation techniques from tracer response measurements has been used, such as the weighted or non-weighted moments method, the transfer function fitting in the Laplace domain, the Fourier analysis, and the curve fitting in the time domain. A comprehensive review on these techniques is provided in the monograph of Wakao and Kagei.\(^6\)

The one-zone two-parameter axial dispersion model, also referred to as the Piston–Dispersion (PD) model, has been the most ubiquitous in the literature for the description of back-mixing. The other differential models can be viewed as its modifications constructed for approximation of more significant flow deviations from the ideal case of plug flow.

The two-zone Piston–Dispersion–Exchange (PDE) model proposed by Van Swaaij et al\(^{12}\) is an improvement over the PD model, which takes into account the mass exchange between the moving (dynamic) liquid phase and the stagnant liquid zone caught in the bed interstices. This pure diffusional mechanism was modeled in an attempt to characterize the tailing symptom often observed in RTD curves in TBRs, which cannot be represented satisfactorily by the PD model. The PDE model (eqns (2a), (2b), Table 1) divides the liquid stream into a dynamic zone in which the liquid flows through the bed as a piston flow with axial dispersion, and a stagnant zone in direct contact with the dynamic zone. Mass transfer retardation is lumped in a mass transfer coefficient or a number of transfer units (\(N\)). The PDE model involves four parameters: the total liquid holdup, the dynamic or static liquid fraction, the liquid axial dispersion coefficient (Péclet number) and the number of transfer units.

For cases involving porous particles, both inter- and
PD model
\[ \frac{\partial c_1}{\partial t} + \frac{U_h}{h_0} \frac{\partial c_1}{\partial z} = D_{\text{ax}} \frac{\partial^2 c_1}{\partial z^2} \] (1)

PDE model
Dynamic liquid zone
\[ \frac{\partial c_D}{\partial t} + \frac{U_L}{h_0} \frac{\partial c_D}{\partial z} + N \frac{U_L}{h_0 z} (c_D - c_S) = D_{\text{ax}} \frac{\partial^2 c_D}{\partial z^2} \] (2a)

Stagnant liquid zone
\[ \frac{\partial c_S}{\partial t} - N \frac{U_L}{2h_0} (c_D - c_S) = 0 \] (2b)

PDE–TD model
Dynamic liquid zone with partial wetting
\[ \frac{\partial c_D}{\partial t} + \frac{U_L}{h_0} \frac{\partial c_D}{\partial z} + N \frac{U_L}{h_0 z} (c_D - c_S) + D'^{\text{eff}} \frac{\partial^2 c_D}{\partial z^2} \bigg|_{z=-R} = D_{\text{ax}} \frac{\partial^2 c_D}{\partial z^2} \] (3a)

Stagnant liquid zone with partial wetting
\[ \frac{\partial c_S}{\partial t} - N \frac{U_L}{2h_0} (c_D - c_S) + D'^{\text{eff}} \frac{\partial^2 c_D}{\partial z^2} \bigg|_{z=-R} = 0 \] (3b)

Intraparticle liquid zone
\[ \frac{\partial c_1}{\partial t} + \frac{D'^{\text{eff}}}{h_1^2} \frac{\partial}{\partial z} \left( \frac{\partial c_1}{\partial z} \right) = 0 \] (3c)

Table 1. Summary of three important RTD models
\begin{tabular}{|c|c|}
\hline
Model & Description \\
\hline
PD & \text{Liquid back-mixing in trickle-bed reactors} \\
\hline
PDE & \text{Dynamic liquid zone} \\
\hline
PDE–TD & \text{Dynamic liquid zone with partial wetting} \\
\hline
\end{tabular}

\* In most of the studies, the RTD curve fitting was performed using time-domain analysis.

\textit{intra-particle} phenomena affect the residence-time distribution, thus invalidating the suitability of the PD and PDE models. Intra-particle tracer diffusion was considered as an additional mechanism contributing to the RTD tails. To interpret the resulting RTD curves, Iiiuta et al.\textsuperscript{7,8} combined therefore the PDE model with a transient diffusion model (PDE–TD) describing the dynamic evolution of the tracer in the porous particles. Deep in trickle-flow regime, the model incorporates contributions of partial wetting from dynamic and stagnant liquid zones in addition to the external and intra-particle mass transfer resistances.\textsuperscript{9} Tracer mass balance equations, respectively, in an axially dispersed dynamic zone, an external liquid stagnant zone and an intra-particle liquid zone are presented in Table 1 (eqns (3a), (3b), (3c)). This three-zone model comprises several parameters: the axial Péclet number, the number of transfer units (characterizing the transfer between the dynamic and static external liquid holdups), the dynamic and static external liquid holdups, the effective diffusion coefficient of the tracer in the pores of the particle, the liquid–solid mass transfer coefficient, and the static and dynamic wetting efficiencies.

The recent contribution of more appropriate RTD models including several tracer diffusion characterizing functions have rendered liquid back-mixing modeling increasingly complex. The problem arises when different RTD models are used to extract liquid axial dispersion coefficients. The resulting coefficients, representing different lumps, must be categorized with respect to the RTD model and interpreted separately. This situation partly explains the lack of correlations predicting \( D_{\text{ax}} \) obtained from the PDE or PDE–TD models since the bulk of the measurements was obtained via the PD model. As a matter of fact, most correlations were developed to predict liquid back-mixing in specific operating conditions suitable to the PD model.

The present work aims at providing one single liquid axial dispersion correlation regrouping the three RTD model cases in order to increase the breadth and accuracy in predicting the liquid axial dispersion coefficient in trickle-bed reactors. Thus, an extensive database containing liquid axial dispersion coefficient measurements was built to first assess the efficiency of important literature correlations on the subject. Following this task, perceptron-like artificial neural network (ANN) modeling was employed to identify which forces, packing properties and external factors are involved in describing liquid back-mixing in terms of the most appropriate set of Buckingham (\( \pi \)) and other characteristic dimensionless groups. The methodology leading to the neural network correlation will
not be detailed in this work since it has been discussed previously.\textsuperscript{10,11} As arguments, a statistical appreciation of literature and ANN correlations will be presented followed by a discussion on the sensitivity of individual physical properties on the calculation of liquid axial dispersion coefficients.

2 BRIEF OVERVIEW

2.1 Database

In total, 973 liquid axial dispersion coefficient measurements have been collected from 22 published independent studies covering the period 1958–2001.\textsuperscript{4,5,8,12–29} The database exhibits results on 14 packing material and shape combinations. From that amount, 195 measurements (20\%) correspond to porous particles for which the PDE–TD model was used to extract the liquid axial dispersion coefficient. The remaining data relate to beds containing nonporous particles: PD model (562 data – 58\%) or PDE model (216 data – 22\%). The database is also associated with 15 liquid compositions (pure, mixtures or solutions) and four gas compositions (Table 2). It should also be emphasized that more than half of the experiments compiled in the database were carried out in the gas-stagnant and liquid gravity-driven trickle flow conditions ($U_L=0$). Inclusion of such data is motivated by the fact that the gas flow rate is considered to have only marginal effects on liquid back-mixing in trickle-bed reactors.\textsuperscript{23} Table 2 summarizes relevant information and offers a detailed description of the range of operating variables that are suspected to influence liquid back-mixing.

2.2 Literature correlations

Since the mid-1950s, investigators have attempted to derive suitable correlations predicting the liquid axial dispersion coefficient or its corresponding Buckingham ($\pi$) group (ie Péclet, Bodenstein or Schmidt numbers) in terms of appropriate bed, liquid, gas, and particle physical properties (Table 3). Actually, all the correlations were developed in dimensionless terms except those of Fu and Tan,\textsuperscript{4} Liles and Geankoplis,\textsuperscript{13} and Tsamatsoulis and Papayannakos.\textsuperscript{17} Frequently used $\pi$ groups describing liquid back-mixing are the liquid Reynolds, Galileo and Froude numbers. Various observations describing the literature correlations presented in Table 3 can be outlined.

- The chief operating variables influencing liquid back-mixing are the liquid superficial velocity and the bed properties, ie packing nominal diameter, grain specific surface area, bed porosity. There is a general consensus that the liquid axial dispersion coefficient is an increasing function of $U_L$, packing nominal diameter, and bed porosity.\textsuperscript{4,12,13,17,30,31}
- Increasing the liquid dynamic viscosity causes a decrease in the liquid axial dispersion coefficient.\textsuperscript{15}
- Despite inclusion of a liquid holdup term in numerous correlations, the gas flow rate factor has been found to affect only marginally the liquid axial dispersion coefficient.\textsuperscript{4,7,8,14,22}
- The effects of the following variables remain

<table>
<thead>
<tr>
<th>Categories</th>
<th>Properties</th>
<th>Min</th>
<th>Median</th>
<th>Max</th>
</tr>
</thead>
<tbody>
<tr>
<td>Operating conditions</td>
<td>Pressure ($P$) [atm]</td>
<td>1.00</td>
<td>1.00</td>
<td>49.35</td>
</tr>
<tr>
<td></td>
<td>Temperature ($T$) [K]</td>
<td>293</td>
<td>293</td>
<td>623</td>
</tr>
<tr>
<td></td>
<td>Superficial liquid velocity ($U_L$) [mm s$^{-1}$]</td>
<td>$3.5 \times 10^{-3}$</td>
<td>$5.0 \times 10^{0}$</td>
<td>$1.1 \times 10^{3}$</td>
</tr>
<tr>
<td></td>
<td>Superficial gas velocity ($U_g$) [m s$^{-1}$]$^a$</td>
<td>0.001</td>
<td>0.085</td>
<td>1.910</td>
</tr>
<tr>
<td>Packing and bed, physical properties</td>
<td>Nominal diameter ($d$) [mm]</td>
<td>0.05</td>
<td>3.00</td>
<td>12.80</td>
</tr>
<tr>
<td></td>
<td>Bed porosity ($\varepsilon$) [%]</td>
<td>33</td>
<td>39</td>
<td>69</td>
</tr>
<tr>
<td></td>
<td>Grain specific surface area ($a_{bz}$) [m$^{-1}$]</td>
<td>468</td>
<td>2000</td>
<td>17143</td>
</tr>
<tr>
<td></td>
<td>Tower diameter ($D_t$) [cm]</td>
<td>1.3</td>
<td>5.1</td>
<td>15.2</td>
</tr>
<tr>
<td></td>
<td>Bed height ($Z$) [m]</td>
<td>0.06</td>
<td>0.92</td>
<td>3.94</td>
</tr>
<tr>
<td>Liquid, physical properties</td>
<td>Density ($\rho_L$) [kg m$^{-3}$]</td>
<td>645</td>
<td>1000</td>
<td>1150</td>
</tr>
<tr>
<td></td>
<td>Viscosity ($\mu_L$) [Pas]</td>
<td>$2.2 \times 10^{-4}$</td>
<td>$1.0 \times 10^{-3}$</td>
<td>$2.7 \times 10^{-2}$</td>
</tr>
<tr>
<td></td>
<td>Surface tension ($\sigma$) [N m$^{-1}$]</td>
<td>$1.0 \times 10^{-2}$</td>
<td>$7.3 \times 10^{-2}$</td>
<td>$7.3 \times 10^{-2}$</td>
</tr>
<tr>
<td>Gas, physical properties</td>
<td>Density ($\rho_g$) [kg m$^{-3}$]</td>
<td>0.164</td>
<td>1.180</td>
<td>2.620</td>
</tr>
<tr>
<td></td>
<td>Viscosity ($\mu_g$) [Pas]</td>
<td>$8.9 \times 10^{-6}$</td>
<td>$1.8 \times 10^{-5}$</td>
<td>$1.8 \times 10^{-5}$</td>
</tr>
<tr>
<td>Liquid axial dispersion coefficient ($D_{Lx}$) [m$^2$ s$^{-1}$]</td>
<td>$9.9 \times 10^{-9}$</td>
<td>$2.1 \times 10^{-4}$</td>
<td>$1.9 \times 10^{-2}$</td>
<td></td>
</tr>
<tr>
<td>Liquid superficial Péclet number ($Pe_L$) [dimensionless]</td>
<td>$6.4 \times 10^{-1}$</td>
<td>$2.0 \times 10^{1}$</td>
<td>$6.2 \times 10^{3}$</td>
<td></td>
</tr>
</tbody>
</table>

Solid phase: glass beads, ceramic sphere, alumina sphere, SiO$_2$Al$_2$O$_3$Cr$_2$O$_3$ sphere, ceramic Raschig ring, glass Raschig ring, ceramic Berl saddle, ceramic Intalox saddle, ceramic extrudate, γ-alumina extrudate, alumina tablets, polyethylene particles, alumina particles, silica powder.

Liquid phase: 0.5\% NaOH solution, methanol, water, n-hexane, cyclohexane, 1-pentanol, 40 vol\% methanol solution, 50\%w glycerol solution, 25\%w glycerol solution, 0.1, 0.5 and 1.0wt\% CMC mixtures, water + 1,2-propylene glycol, heavy vacuum gasoil (HVGO), 15mol\% ethylene glycol solution.

Gas phase: air, nitrogen, hydrogen, helium.

Residence-time distribution model: PD model (562 data), PDE model (216 data), PDE–TD model (195 data).

$^a$ Based on 567 experimental results, those including a gas stream in their study.
uncertain however as no directed studies are available in the literature: column diameter, particle sphericity, surface tension, liquid density and gas density.

3 ARTIFICIAL NEURAL NETWORK METHODOLOGY

The general strategy surrounding the application of neural network computing and the data mining procedure for extraction of the best set of dimensionless groups is akin to the approaches already advocated by the present authors in several other contributions.11,12,13 Indeed, the clustering of physical properties and operating parameters into dimensionless Buckingham (π) groups renders empirical modeling methods such as neural network computing very convenient. The formation of over 15 dimensionless groups as depicted in a previous work14 covers all possible force ratios and external factors encountered in three-phase fixed-bed reactors. In the liquid phase, the Reynolds, Froude, Weber, Morton, Eötvös, Galileo, Stokes, Capillary and Onneshege π groups sweep the complete range of force ratios (ie inertia-to-viscous, gravitational-to-capillary) that might possibly influence liquid back-mixing. As the output, the liquid axial dispersion coefficient can be either merged into a Péclet number, which takes the active bed height as its characteristic length, or a Bodenstein number, which uses a particle characteristic length such as the nominal particle diameter or grain specific surface area.

As explained in Section 2.1, several experiments were performed without a gas stream. For ANN modeling, this fact considerably reduces the efficiency of π groups involving gas inertia. As a result, a mixed Reynolds number which bases itself on gas physical properties and the summation of gas and liquid superficial velocities was added in the study, enabling a possible inclusion of the gas flow rate effect in the correlation. In order to identify the best representation, each π group involving a characteristic length was defined in three versions using the particle nominal diameter (dp), the bed specific surface area (as) and the grain specific surface area (aq).

The three-layer ANN development was carried out using the NNfit software.34 Several aspects were considered to decide whether a set of dimensionless groups presents acceptable results. The choice of the
**Table 4.** Set of equations, normalized inputs, output, schematic diagram and weights for the application of the ANN correlation

<table>
<thead>
<tr>
<th>Set of equations</th>
<th>Normalized inputs</th>
<th>Normalized output</th>
<th>ANN weights</th>
<th>Domain of validity</th>
</tr>
</thead>
<tbody>
<tr>
<td>[ S = \frac{1}{1 + \exp\left(-\sum_{j=1}^{10} \omega_j H_j\right)} ]</td>
<td>[ U_1 = \frac{\log\left(\frac{Ra_h}{2.17 \times 10^{-4}}\right)}{6.025} ]</td>
<td>[ S = \frac{\left(\frac{B_0}{5.91 \times 10^{-4}}\right)}{2.495} ]</td>
<td>( \omega_j )</td>
<td>( 5.91 \times 10^{-4} \leq B_0 \leq 1.85 \times 10^{-1} ) ( 9.71 \times 10^{-6} \leq E_o_2 \leq 6.12 \times 10^{-1} ) ( 2.65 \times 10^{-5} \leq \Delta \lambda \leq 4.92 \times 10^2 ) ( 2.30 \times 10^2 ) ( 6.02 \times 10^{-3} \leq \Delta \lambda \leq 9.52 \times 10^4 ) ( 1.44 \times 10^{-5} \leq \Delta \lambda \leq 3.02 \times 10^2 ) ( 1 \leq \text{RTD} \leq 3 ) (integer only)</td>
</tr>
<tr>
<td>[ H_j = \frac{1}{1 + \exp\left(-\sum_{i=1}^{3} \omega_i U_i\right)} ]</td>
<td>[ U_2 = \frac{\log\left(9.71 \times 10^{-6}\right)}{4.799} ]</td>
<td>( RTD = 1 ) (PD model); ( RTD = 2 ) (PDE model) ( RTD = 3 ) (PDE-TD model)</td>
<td>( K )</td>
<td></td>
</tr>
<tr>
<td>1 \leq J \leq 9 ( H_{10} = 1, U_6 = 1 )</td>
<td>[ U_3 = \frac{\log\left(6.02 \times 10^{-3}\right)}{7.199} ]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>[ U_4 = \frac{\log\left(2.65 \times 10^{-5}\right)}{7.272} ]</td>
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<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>[ U_5 = \frac{\log\left(1.44 \times 10^{-5}\right)}{7.320} ]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>[ U_6 = K - \frac{0.823}{0.173} ]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>[ U_7 = RTD - \frac{1}{2} ] ( RTD = 1 ) (PD model); ( RTD = 2 ) (PDE model) ( RTD = 3 ) (PDE-TD model)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>( B_0 = \frac{U_L}{\alpha D \omega x} )</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>( U_7 = \frac{U_L}{\alpha D \omega x} )</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ANN weights</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \omega_i )</td>
<td>( \begin{array}{cccccccccc} 1 &amp; 2 &amp; 3 &amp; 4 &amp; 5 &amp; 6 &amp; 7 &amp; 8 &amp; 9 &amp; 10 \ 2.28366 &amp; 9.04076 &amp; 1.11151 &amp; 1.67908 &amp; -9.99938 &amp; -5.02841 &amp; 4.45536 &amp; -7.64148 &amp; -6.36805 \end{array} )</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
| Classifications of the neural correlation model are accessible at: http://www.gch.ulaval.ca/~flrachidi or http://www.gch.ulaval.ca/~randjel/
best set rests on the fulfilment of the following criteria:

- The optimal set must lead to the best output prediction with a minimum average absolute relative error and standard deviation.
- The neural network architecture must be of minimal complexity, with the least number of dimensionless numbers and hidden neurons without deterioration in the prediction.
- The trained network must have a generalization ability. Each ANN was systematically trained using 780 learning instances (ca 80% of the database) selected randomly from the database. Once an ANN has been trained, a test file of 973 – 780 = 193 vectors was used to test the correlation robustness.
- The optimal set of dimensionless groups should be closely associated with the phenomenology of liquid back-mixing.

After thorough analysis, six relevant dimensionless numbers and one discrimination code input representing the RTD models have been identified to satisfactorily describe liquid back-mixing, defined as a Bodenstein number ($B_{01}$). The characteristic inputs are the liquid Reynolds ($Re_L$), Galileo ($Ga_L$) and Eötvös ($E_{0L}$) numbers, the gas Galileo ($Ga_G$) number, a wall characterizing group (K) and the mixed Reynolds number ($Sr$). This latter group is introduced in order to provide a correlation which remains asymptotically robust when $U_C \to 0$ as the gas-stagnant trickle flow is approached. The set of equations for the neural correlation as well as the fitting parameters (or weights) is presented in Table 4 along with the appropriate definition of the $\pi$ groups. The applicability of neural networks depends on the domain of validity established for each ANN input and output; they are also depicted in Table 4. Note that the Bodenstein and Reynolds groups have been defined based on the superficial liquid velocity and not on the interstitial liquid velocity as this latter usually implies knowledge of the liquid holdup.

4 PERFORMANCE OF LIQUID BACK-MIXING CORRELATIONS

4.1 Statistical analysis

The statistical performance of the liquid $D_{AX}$ correlations summarized in Table 3, including the ANN correlation, is presented in Table 5. It shows how inadequate the literature correlations are in terms of accuracy and breadth. Among the literature correlations, the Ebach and White correlation yields the best statistical numbers with an average absolute relative error (AARE) of 69% and a standard deviation ($\sigma$) on AARE of 56% for the whole database (first and second rows in Table 5) while others present unacceptable prediction errors on $D_{AX}$. The correlations developed by Hochman and Efron, Buffalo and Rathor, Fu and Tan, and Tsamatsoulis and Papayannakos are found to over-predict the liquid axial dispersion coefficient, whereas those of Liles and Geankoplis and Cassanello et al yield important under-predictions. Narrowing the testing data file to the data corresponding to the domain of validity of the correlations ($N_D < 973$) does not appear to bring much improvement to the predictions (see rows 3–5 in Table 5). The statistical numbers are more or less equivalent whether or not the validity domain is specified. Yet, the correlations present good to average predictive ability on the authors’ own $D_{AX}$ data (last three rows in Table 5). The modest performance achieved by some of the correlations, ie Ebach and White correlation, is

| Table 5. Statistical evaluation of literature and ANN correlations based on the dimensional $D_{AX}$

<table>
<thead>
<tr>
<th>Literature correlations</th>
<th>Whole database without specifications (973 data)</th>
<th>Whole database with specifications$^a$</th>
<th>Author’s own data</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>AARE (%)</td>
<td>$\sigma$ (%)</td>
<td>$N_D$</td>
</tr>
<tr>
<td>Ebach and White$^{12}$</td>
<td>69</td>
<td>56</td>
<td>562</td>
</tr>
<tr>
<td>Liles and Geankoplis$^{13b}$</td>
<td>130</td>
<td>&gt;500</td>
<td>488</td>
</tr>
<tr>
<td>Hochman and Efron$^{14c}$</td>
<td>&gt;500</td>
<td>&gt;1000</td>
<td>288</td>
</tr>
<tr>
<td>Buffalo and Rathor$^{15c}$</td>
<td>340</td>
<td>&gt;500</td>
<td>185</td>
</tr>
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<td>Kobayashi et al$^{16}$</td>
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<td>Fu and Tan$^{4}$</td>
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</tr>
<tr>
<td>Tsamatsoulis and Papayannakos$^{17c}$</td>
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<tr>
<td>Cassanello et al$^{20b}$</td>
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<td>20</td>
<td>227</td>
</tr>
<tr>
<td>Michell and Furzer$^{31}$</td>
<td>157</td>
<td>400</td>
<td>231</td>
</tr>
</tbody>
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$^a$ See Table 3 for the correlations’ specifications.

$^b$ Correlation generally under-predicts liquid axial dispersion coefficient.

$^c$ Correlation generally over-predicts liquid axial dispersion coefficient.

$N_D$ = number of data.
is partly attributed to the fact that liquid holdup is estimated from state-of-the-art correlations and procedures described elsewhere. Liquid holdups are indeed seldom available in the original sources simultaneously with the reported $D_{AX}$. It appears also that most correlations achieve better results for $D_{AX}$ extracted from the PDE and PDE–TD models instead of the $D_{AX}$ obtained from the PD model. This is shown in Table 6, especially for the Michell and Furzer and Hochman and Effron correlations.

With the broadest database, the ANN correlation shows drastic improvement in predicting the liquid axial dispersion coefficient (Table 5). Most conveniently, the improvement factor has at least tripled with respect to the best (Ebach and White) literature correlation. Both correlations are shown in Fig 1 for visual inspection of the quality of fit. As shown in Table 6, the AARE for measurements taken from the PD model is 27% while the axial dispersion data yielded from the PDE and PDE–TD give, respectively, AAREs of 17% and 15%. This performance reinforces the generalization ability of the present ANN correlation which remains suitable regardless of the RTD model used to extract $D_{AX}$.

Further modeling improvement of trickle-bed back-mixing achieved by the new ANN correlation includes its inherent potential to predict quite accurately the $D_{AX}$ in different flow regimes. From the extended range of gas and liquid velocities (Table 2), several experiments gathered in the database were performed in the high interaction flow domain where pulsing, spray and bubble flow are spanned. These different flow regimes are known to induce larger fluid-to-fluid interactions, inflecting thus distinctive liquid dispersion conditions as compared with the low interaction flow domain for which trickle flow predominates. Yet these gas-to-liquid flow distinctions do not curtail the quality of fit characterizing the new ANN correlation as it fits equivalently well the data for both the low interaction and high interactions regimes (Table 7).

### 4.2 Parametric analysis

Providing the means to confirm the robustness of the

<table>
<thead>
<tr>
<th>PD model (562 data)</th>
<th>ANN correlation 27 23</th>
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<tbody>
<tr>
<td></td>
<td>Ebach and White 12 70 38</td>
</tr>
<tr>
<td></td>
<td>Michell and Furzer 11 41 11 700</td>
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<td></td>
<td>Hochman and Effron 14 &gt;1000 &gt;1000</td>
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</table>

<table>
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<tr>
<th>PDE model (216 data)</th>
<th>ANN correlation 17 19</th>
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<tbody>
<tr>
<td></td>
<td>Ebach and White 12 45 45</td>
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<tr>
<td></td>
<td>Michell and Furzer 11 77 76</td>
</tr>
<tr>
<td></td>
<td>Hochman and Effron 14 428 466</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>PDE–TD model (195 data)</th>
<th>ANN correlation 15 12</th>
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<tbody>
<tr>
<td></td>
<td>Ebach and White 12 95 86</td>
</tr>
<tr>
<td></td>
<td>Hochman and Effron 14 149 163</td>
</tr>
<tr>
<td></td>
<td>Michell and Furzer 11 71 73</td>
</tr>
</tbody>
</table>

Table 6. Performance of the ANN correlation and relevant literature correlations divided into RTD model categories.

![Figure 1](image)

Figure 1. Predicted versus measured Bodenstein number of (a) ANN correlation (learning and generalization files) and (b) Ebach and White correlation (measurements within the specified range (see Table 3) and measurements outside the specified range).
ANN correlation, several simulations were performed by attributing different values for one studied variable while holding the others constant. The parametric analysis indeed provides more insights on the influence of important operating variables on liquid back-mixing. A total of eight physical parameters regruped into four diagrams were assessed (Fig 2). The corresponding physical properties of 3 mm glass beads and an air–water system were used for the simulations unless otherwise stated.

As mentioned in Section 2.2, the most influential parameters are the liquid superficial velocity and the packing properties. This is well illustrated in Fig 2(a).

Based on the PD or the PDE model within the simulated interval, the liquid axial dispersion coefficient increases, as suggested in the literature, almost proportionally with the liquid superficial velocity. Likewise, several correlations perceive larger particles, respectively high nominal diameter (d), to increase the D_{AX}. This trend is clearly shown in Fig 2(a) in terms of grain specific surface area. The effect of bed porosity on D_{AX} is also shown. At constant a_{G}, the higher the bed porosity (Intalox saddles empty circles: ε=65%, glass beads empty triangles: ε=35%), the higher the axial dispersion coefficient. This trend is also corroborated from the correlations of Fu and Tan,^{14} and Tsamatsoulis and Papayannakos.^{17} Although it is difficult to highlight explicitly the effect of sphericity, particles exhibiting complex geometry, such as the Intalox saddles, are handled in the ANN correlation through the dimensionless number K. Figure 2(b) represents the actual effect of gas superficial velocity as simulated by the ANN correlation. As widely reported in the literature, the gas superficial velocity has negligible effect on the D_{AX}. In agreement with the

<table>
<thead>
<tr>
<th>Flow regime</th>
<th>Number of data</th>
<th>AARE (%)</th>
<th>σ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single-phase liquid (G=0)</td>
<td>406</td>
<td>25</td>
<td>20</td>
</tr>
<tr>
<td>Low interaction (trickle flow)</td>
<td>359</td>
<td>19</td>
<td>15</td>
</tr>
<tr>
<td>High interaction (pulsing/bubble/spray flow)</td>
<td>208</td>
<td>22</td>
<td>17</td>
</tr>
</tbody>
</table>

Table 7. Performance of the ANN correlation in relation to the operating flow regime

Figure 2. ANN simulations describing the effect of operating variables on liquid axial dispersion coefficient (a – effect of packing physical properties and superficial liquid velocity; b – effect of gas superficial velocity; c – effect of liquid viscosity; d – effect of liquid density and surface tension). Where not specified, default values are: D_{B}=50 mm, a_{G}=2000 m^{-1}, ε=40%, φ=1, U_{L}=5 mm s^{-1}, U_{G}=0.1 m s^{-1}, air–water physical properties and PD model.

work of Buffham and Rathor\(^5\) and as depicted in Fig 2(c), the liquid viscosity decreases the \(D_{AX}\) although the variation may be considered to be relatively small. Finally, Fig 2(d) shows that liquid density and surface tension play a rather limited role in defining liquid axial dispersion.

5 CONCLUSION

A liquid axial dispersion coefficient correlation was developed by combining artificial neural network computing and dimensional analysis. As a result, the prediction of liquid back-mixing in trickle-bed reactors was significantly improved. Based on a 973 measurements database, the neural network correlation yielded an \(AARE\) of 22% with a standard deviation on the \(AARE\) of 24%. The identification of two dominant operating parameters, the liquid superficial velocity and the particle properties, was clearly illustrated. The liquid viscosity affects only slightly the liquid axial dispersion coefficient.

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REFERENCES