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Flowdynamic Modeling of Bale-Type Catalytic Distillation Packings

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ABSTRACT

A model was developed to predict the flowdynamic behavior of bale-type catalytic packings, which are widely used in the industry for several applications. The new model takes into account the effect of the catalyst properties and packing characteristics as well as the properties of the fluids, flow rates, and column diameter to predict liquid hold-up, pressure drop, and the loading and flooding points. The particle model concepts were used as the basis for the generation of the model, introducing considerations that account for a proper representation of a wide variety of data obtained in tests performed in this work or reported previously in the literature. It was found that the nature of the catalyst contained in the “bales” strongly affects the liquid hold-up due to solid–liquid interactions. A factor was also introduced in the model to account for the strong effect of the wall on small-diameter columns. The new model

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significantly improved the prediction of the flowdynamic parameters such as hold-up and pressure drop as well as loading and flooding points, which showed deviations smaller than 10–30%.

Key Words: Bales; Catalytic distillation; Flow dynamics; Column.

INTRODUCTION

The catalytic distillation concept has been widely used over the last decade for a large variety of applications, including ether preparations such as methyl-tert-butyl-ether (MTBE), tert-amyl-methyl-ether (TAME), and ethyl-tert-butyl-ether (ETBE); aromatics alkylation; gasoline desulfurization; dimerization; hydrogenations; and various other reactions.^[1] Most of these applications use the so-called “bale-type” packing, filled with the appropriate catalyst, as the key element of the reactive column. The design of a catalytic distillation column has to take into account not only flowdynamic and separation considerations, as in regular distillation columns, but also conditions that allow significant reaction rates and selectivities. All three factors (mass transfer, reaction rates, and flowdynamics) influence and affect each other very strongly, so it is necessary to have tools that help in the design and evaluation of the operation of this type of columns. In this work, the focus is on development of a tool for predicting the flowdynamics of a catalytic distillation column loaded with bales.

Several studies have been published recently with flowdynamic models for columns loaded with bale packing. A comprehensive flowdynamic and mass transfer model was developed by Subawalla et al.^[2] based on the model for packed distillation columns of Stichlmair et al.^[3] The Subawalla model introduced geometric considerations to account for the irregular shape of the bales as well as principles of the well-known channel model to improve the results of the particle model of Stichlmair. Xu et al.^[4] produced data in a cold simulator column loaded with bale packing to generate flowdynamic correlations to represent that data. Those correlations show that pressure drop and liquid hold-up are only a function of liquid and gas velocities and do not take into account fluids, packing, and catalyst properties, which limit the application to the specific conditions for which they were developed. Akbarnejad et al.^[5] modified the well-known Eckert diagrams for prediction of pressure drop in catalytic distillation columns loaded with bales and validated their model with data obtained in a cold simulator. This model does not predict liquid hold-up, which is an important parameter for columns with chemical reaction, and it is actually believed that today there are more accurate ways of predicting pressure drop than methods based on the Eckert diagram.



Several studies,^[6–8] performed at ambient conditions, found strong effects of the characteristics of the bales (amount of catalyst loaded per volume unit, surface area, and catalyst type) on the flowdynamic performance of small columns. They also showed that none of the available flowdynamic models for bales are capable of properly representing data obtained in columns at different scales.^[6] The Subawalla model is the most accurate model but still shows strong deviations in the prediction of liquid hold-up, flooding, and loading points. All of these are very important parameters in the design and operation of catalytic distillation columns.

The objective of this work is to generate a reliable flowdynamic model for catalytic distillation columns filled with bales of different scales and for applications with a wide variety of fluid and catalytic systems. For this purpose, the various models for catalytic and noncatalytic columns available in the literature were evaluated to identify the more appropriate one to be taken as the basis for the new model. Several modifications were proposed to take care of the observed deviations of the more reliable available model. All the data available in the open literature was used to perform nonlinear regression of the parameters of the new model.

DESCRIPTION OF THE BALE PACKING

Figure 1 is a sketch of the bale packing. This packing contains the catalyst inside pockets of fiberglass cloth (3–5 cm wide), which is wrapped in spiral

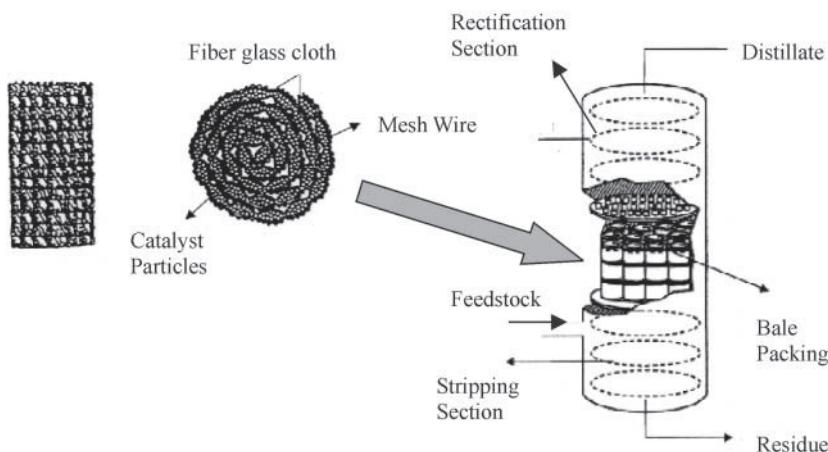


Figure 1. Scheme of the bale-type packing and catalytic column.



with a stainless steel mesh wire. The mesh wire provides both structural strength and open space for the gas and liquid flow, while the cloth provides the surface for mass transfer. Bales are packed inside the appropriate section of the column to favor reaction. The catalytic distillation column may have regular stripping and rectification sections, as in the case of an MTBE column, or different configuration depending on the specific application.

The main flowdynamic properties of the bale packing can be adjusted depending on the amount of fiberglass cloth used per unit of volume, which sets the surface area of the packing, and the amount of catalyst used to fill the holes of the cloth, which largely determines the void fraction.

FLOWDYNAMIC INFORMATION AVAILABLE FOR BALES

Table 1 shows a summary of the available flowdynamic data in the literature for bales. It can be seen that most of the data were obtained with ion-exchange resin as the catalyst (Amberlyst 15 from Rohm and Hass, Philadelphia, PA); however, there is a data set for activated carbon as catalyst filling the bales. On the other hand, even though the largest amount of information is for the air–water system, there are some data sets obtained with organic fluids at actual distillation conditions (cyclohexane/*n*-heptane and acetone/MEK) and at ambient conditions (CO₂–vegetable oil).

The bale-packing flowdynamic properties include a wide variety of data, with surface areas between 100 and 400 m²/m³ and void fractions between 0.66 and 0.83 m³/m³, which assures a good source of information for the model development.

It can also be seen in Table 1 that most of the data were obtained in small columns (4 in. and less). Unfortunately, it was not possible to obtain key properties of the packing for the only set of data published for a large-diameter column. This circumstance impedes the use of this large-scale data for the purpose of evaluating existing or new models.

EVALUATION OF AVAILABLE FLOWDYNAMIC MODELS

Four existing models were evaluated using the available data bank to determine if any could be used to satisfactorily represent the flowdynamics of the column filled with catalytic bales. One of them was specifically developed for bale-type packings (Subawalla et al.^[2]), while the other three models were developed for random packings of regular distillation columns. Those three



Table 1. Available flowdynamic data published for bale-type catalytic packings.

Gas-liquid system	Catalyst inside the bales	Column diameter (m)	Packing surface area (m^2/m^3)	Packing void fraction (m^3/m^3)	Pressure drop data available	Liquid hold-up data available	Reference
Air-water	Amberlyst 15	0.0254	100	0.83	✓	✓	Caetano ^[7]
	Amberlyst 15	0.0254	200	0.80	✓	✓	Caetano ^[7]
	Amberlyst 15	0.0254	200	0.66	✓	✓	Caetano ^[7]
	Amberlyst 15	0.0762	100	0.85	✓	✓	Madrid ^[8]
	Amberlyst 15	0.0762	200	0.83	✓	✓	Madrid ^[8]
	Amberlyst 15	0.0762	200	0.73	✓	✓	Madrid ^[8]
	Amberlyst 15	0.0508	169	0.76	✓	✓	González ^[9]
Cyclohexane- <i>n</i> -heptane	Amberlyst 15	0.0508	169	0.76	✓	✓	González ^[9]
Acetone-MEK	Amberlyst 15	0.0254	100	0.80	✓	✓	Manduca ^[10]
Air-water/MEA	Amberlyst 15	0.0254	200	0.71	✓	✓	Manduca ^[10]
Air-water/MEA	Amberlyst 15	0.0254	200	0.63	✓	✓	Manduca ^[10]
Air-water/MEA	Activated carbon	0.0254	100	0.81	✓	✓	Caetano ^[6]
CO ₂ -vegetable oil	Amberlyst 15	0.0254	100	0.83	✓	✓	Caetano ^[6]
Air-water	Amberlyst 15	0.102	398	0.82	✓	✓	Akbarnejad ^[5]
Air-water	Amberlyst 15	0.600	Not available	0.70	✓	✓	Xu et al. ^[4]



models are the Stichlmair et al.^[3] model, which is based on the assumption of flow around particles inside the column (particle model); the Billet^[11] model, which is based on the so-called channel model; and the Engel et al.^[12] model, which is a further improvement of the original Stichlmair et al. model. The bale-packing models of Xu et al.^[4] and of Akbarnejad et al.^[5] were not included in this comparison since they were developed for very limited conditions and previous work^[6-8] shows that their predictions do not properly represent real data obtained under a wide variety of conditions, such as that indicated in Table 1. Detailed descriptions of the models evaluated in this work can be found in the original publications.

The statistical evaluation of the four models with the data bank of Table 1 was done using the Ansari et al.^[13] method, which calculates six statistical indicators of the deviation of the predicted values in relation to the real data. Those indicators are:

- E1: Percentage average error.
- E2: Percentage average absolute error.
- E3: Percentage standard deviation.
- E4: Average error.
- E5: Average absolute error.
- E6: Standard deviation.

These six indicators are used to calculate a factor of relative performance (FRP), which provides an overall statistical evaluation of the model. The lowest absolute values of the FRP indicate a better representation of the data by the model. An FRP value of zero indicates a perfect prediction. Further details about this statistical method can be found in the original paper of Ansari et al.^[13].

The comparison of models was done on the prediction of the following variables:

- Pressure drop.
- Liquid hold-up.
- Pressure drop at the loading point.
- Liquid hold-up at the loading point.

Table 2 shows a summary of the FRP values for each of the four models. It can be seen that in all cases the lowest values of the FRP, and consequently the better performance, were obtained with the Engel et al. model. The FRP values for the Stichlmair et al. model were also very low and significantly better than the values corresponding to the Subawalla et al. and Billet models.

Table 2. Summary of the evaluation of the existing models using the FRP statistical parameter as indicator.

Parameter	FRP values for the model of Stichlmair et al. ^[3]	FRP values for the model of Engel et al. ^[12]	FRP values for the model of Subawalla et al. ^[2]	FRP values for the model of Billet ^[11]
Pressure drop	0.27	0.14	0.33	0.75
Liquid hold-up	0.47	0.12	4.04	1.57
Pressure drop at the loading point	0.38	0.24	0.34	1.00
Liquid hold-up at the loading point	0.25	0.24	4.80	4.81

Figures 2–7 show parity plots (predicted values vs. experimental data) for the six variables previously listed using the Engel et al.^[12] model. Dotted lines indicate the percent limits in which lie 90% of the points. It can be seen in these figures that these 90% limits are still large values, ranging between $\pm 40\%$ and $\pm 80\%$. Specifically, large data scatter is seen in the prediction of the loading and flooding points. These results show that even the best available model in the literature is not suitable for prediction of the flowdynamic behavior of columns loaded with catalytic bale packing, making the development of better models for these applications necessary.

DEVELOPMENT OF A NEW MODEL

Since the Stichlmair et al.^[3] procedures based on the particle model have been shown to be very reliable for both random packings and catalytic packings, this approach was also followed in this work, introducing corrections to take into account the particularities of the bale-type packing.

Dry Pressure Drop Prediction

The three parameters equation proposed by Stichlmair et al. provides a very accurate representation of the data in both regular and catalytic applications^[6–9] and was used in this work without further modifications. Dry pressure drop is calculated using a relation obtained after performing a force



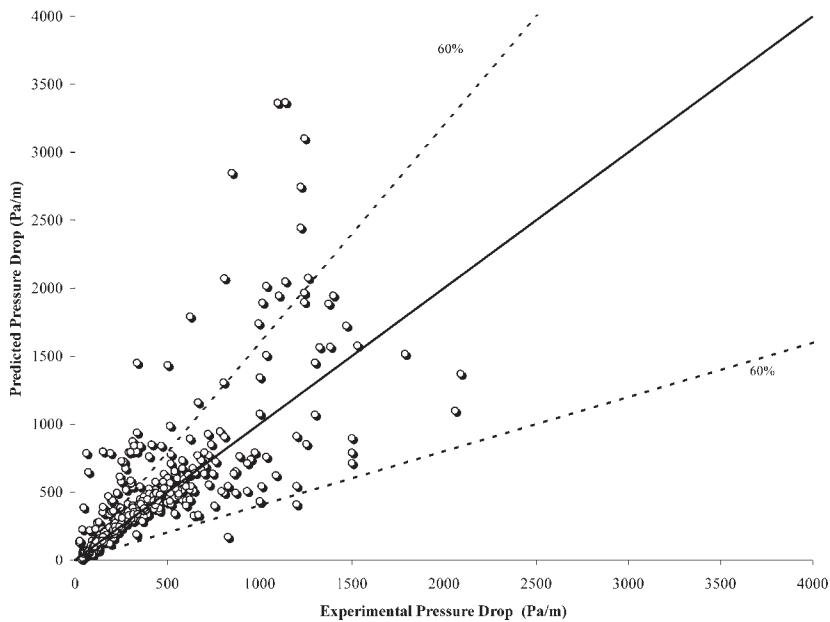


Figure 2. Comparison of the predicted and experimental values of pressure drop using the Engel et al. model.

balance around a single particle:

$$\frac{\Delta P_d}{z} = \frac{3}{4} f_o \left[\frac{(1 - \varepsilon)}{\varepsilon^{4.65}} \right] \frac{\rho_G u_G^2}{d_p} \quad (1)$$

where the f_o parameter is calculated using the following correlation:

$$f_o = \frac{C_1}{Re_G} + \frac{C_2}{Re_G^{1/2}} + C_3 \quad (2)$$

The three parameters, C_1 , C_2 , and C_3 , are regressed from dry pressure drop experimental data. This implies that any bale packing to be modeled has to be previously characterized by data of dry pressure drop at different gas velocities. These are the only experimental parameters of the packing required to run the model, aside from its properties (void fraction and surface area).



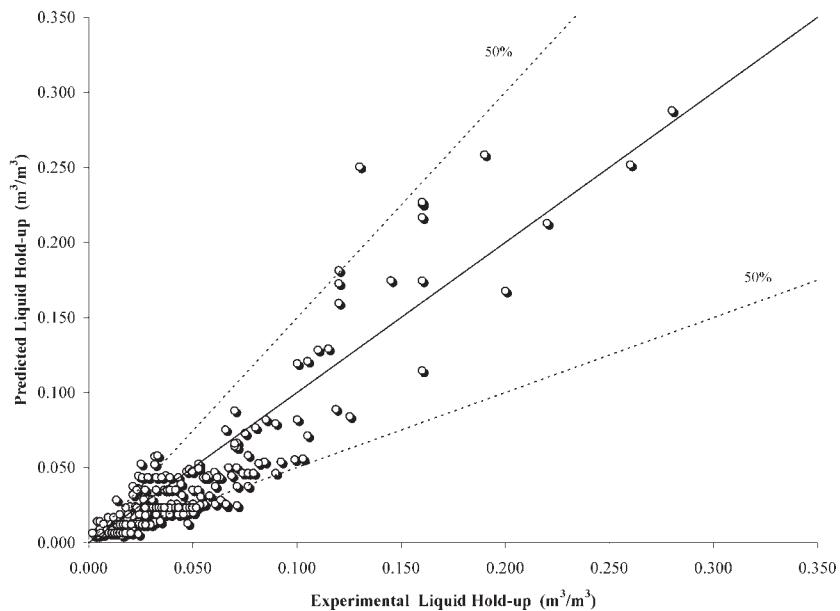


Figure 3. Comparison of the predicted and experimental values of liquid hold-up using the Engel et al. model.

Liquid Hold-Up Below the Loading Point

A review of the literature showed that the liquid hold-up below the loading point could be expressed as a function of three main dimensionless numbers; Froude, Galileo, and Eotvos numbers, which are calculated as follows:

Froude number:

$$Fr = \frac{u_L^2 a_p}{g} \quad (3)$$

Galileo number:

$$Ga = \frac{g \rho_L^2}{a_p^3 \mu_L^2} \quad (4)$$

Eotvos number:

$$Eot = \frac{\rho_L g}{\sigma_L a_p^2} \quad (5)$$



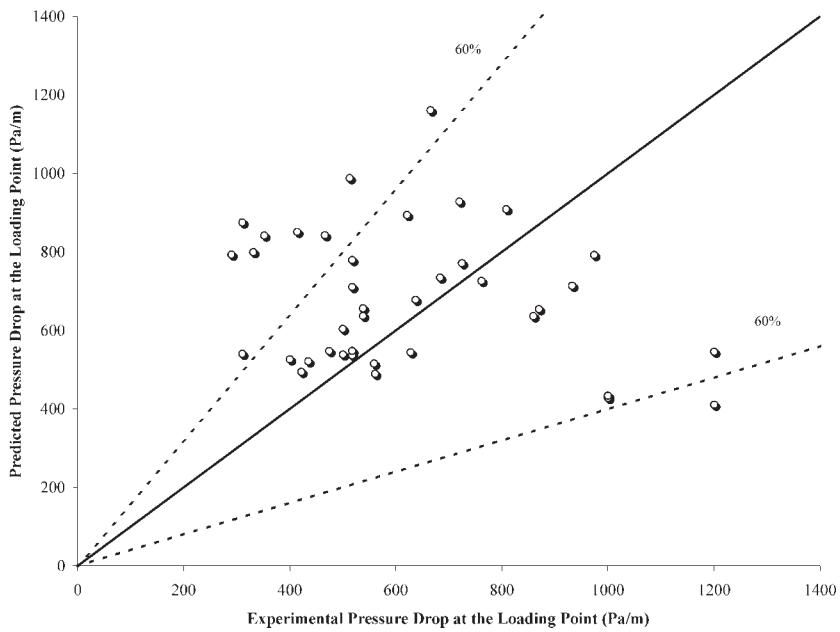


Figure 4. Comparison of the predicted and experimental values of pressure drop at the loading point using the Engel et al. model.

It has also been found that some authors introduce corrections in the hold-up expression to account for the height of the column (liquid maldistribution), diameter of the column, and bed void fraction. Based on this information, five different combinations of dimensionless dependent variables were proposed to generate the expression of liquid hold-up below the loading point.

Combination 1:

$$h_{L,bc} = c_1 (Fr)^{c2} (Ga)^{c3} (Eot)^{c4} \varepsilon^{c5} \quad (7)$$

Combination 2:

$$h_{L,bc} = c_1 (Fr)^{c2} (Ga)^{c3} (Eot)^{c4} \quad (8)$$

Combination 3:

$$h_{L,bc} = c_1 (Fr)^{c2} (Ga)^{c3} \varepsilon^{c4} \quad (9)$$



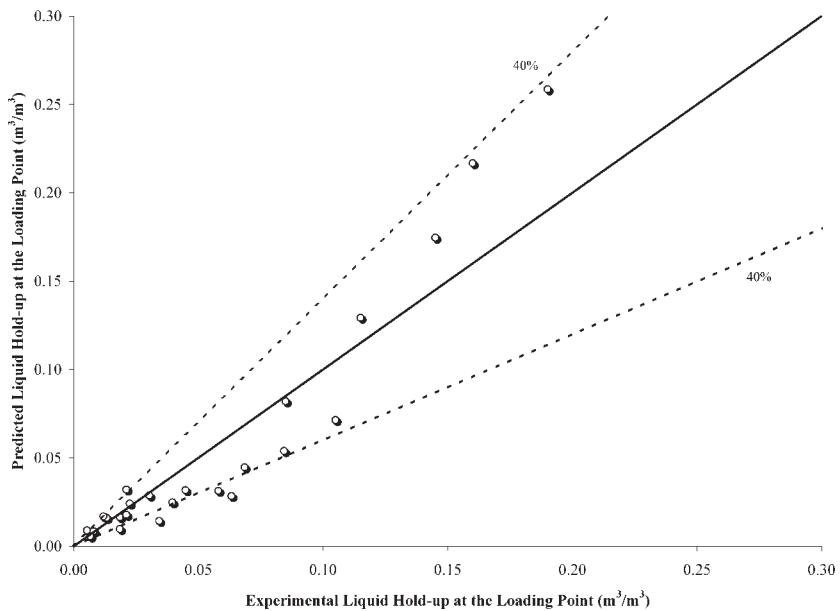


Figure 5. Comparison of the predicted and experimental values of liquid hold-up at the loading point using the Engel et al. model.

Combination 4:

$$h_{L,bc} = c_1(Fr)^{c2}(Ga)^{c3}(Eot)^{c4}\epsilon^{c5}(ad_c)^{c6} \quad (10)$$

Combination 5:

$$h_{L,bc} = c_1(Fr)^{c2}(Ga)^{c3}(Eot)^{c4}\epsilon^{c5}(az)^{c6} \quad (11)$$

The parameters of each of these expressions were calculated by linear regression using the corresponding data from the data bank described in Table 1. The prediction accuracy of each of the previous expressions was evaluated using the Ansari et al. method described earlier. The lowest FRP value was obtained for the first combination [Eq. (7)]. This FRP value was between five and eight times lower than the FRP of the other combinations, which leaves no doubt about the superior performance of the first combination.

The parity plot, comparing the hold-up predicted in Eq. (7) with the experimental results are shown in Fig. 8. The dotted lines indicate that 90% of



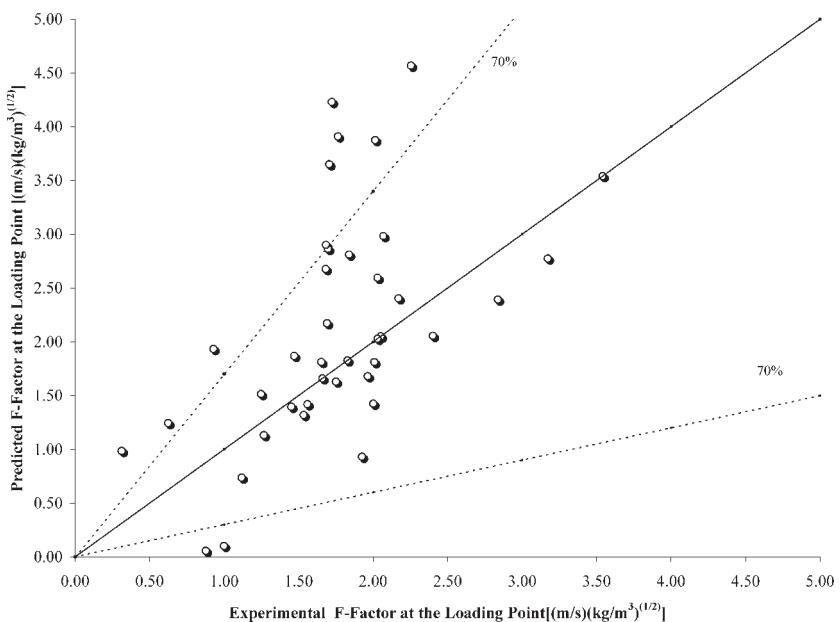


Figure 6. Comparison of the predicted and experimental values of F-factor at the loading point using the Engel et al. model.

the points lie within $\pm 30\%$ of error, which is marginally better than the error obtained with the Engel et al. method ($\pm 40\%$) and still does not meet the expected accuracy of a useful model.

Further evaluation of the data showed significant changes in the experimental values of liquid hold-up when the liquid system or the catalytic solid changed, which was not being taken into account by the proposed expressions. It can be noted that none of the expressions take into account the nature of the catalytic solid inside the bale cloth and the possible liquid–catalyst interactions that may affect the liquid hold-up. In fact, when the catalyst is the hydrophilic resin Amberlyst 15, it is expected that more water is retained inside the column due to the high solid–liquid affinity. On the other hand, less liquid should be retained in the column when the Amberlyst 15 is contacted with organics. This observation led to the definition of an affinity factor that affects the hold-up in the Eq. (7). Taken as the basis the Amberlyst 15–water, system, average values of the affinity factor were



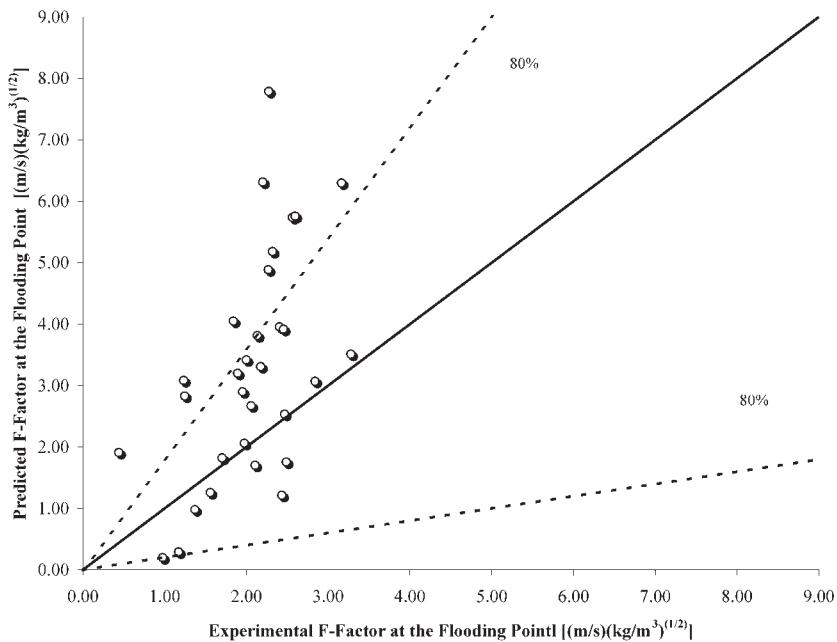


Figure 7. Comparison of the predicted and experimental values of F-factor at the flooding point using the Engel et al. model.

calculated using a few points from the data bank. The following values were obtained:

Liquid	Solid	Affinity factor value (Fa)
Water (hydrophilic)	Amberlyst 15 (hydrophilic)	1.0 (reference value)
Water (hydrophilic)	Activated carbon (hydrophobic)	0.6
Organics (hydrophobic)	Amberlyst 15 (hydrophilic)	0.6
Organics (hydrophobic)	Activated carbon (hydrophobic)	1.0

The affinity factor was incorporated into Eq. (7), and the values previously listed were used in the model to make predictions of the experimental data. Figure 9 shows the parity plot of the prediction and experimental values using Eq. (7) modified with the affinity factor. Dotted



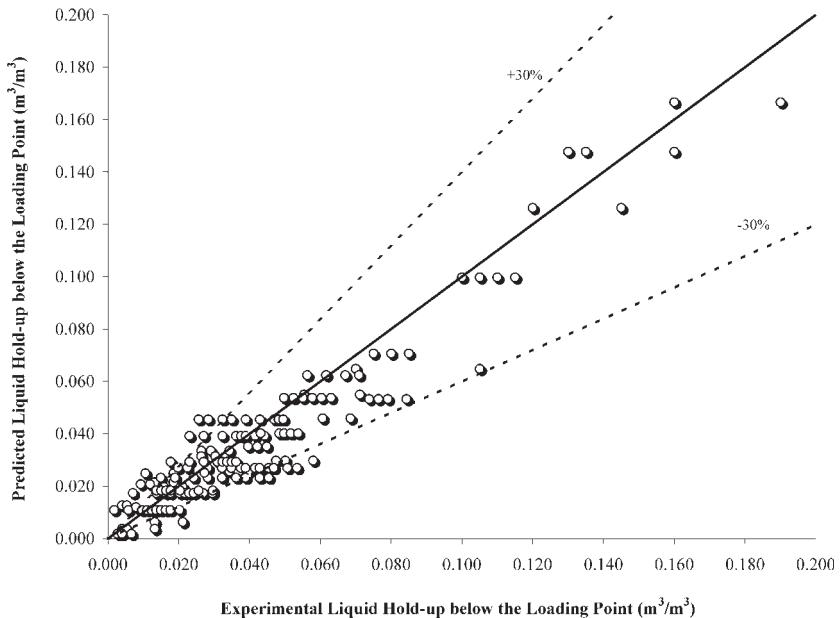


Figure 8. Comparison of the predicted and experimental values of the liquid hold-up at the loading point using Eq. (7).

lines show that in this case 90% of the points were predicted with an error lower than $\pm 15\%$. This is a significant improvement in comparison with previous expressions for liquid hold-up below the loading point. The final expression of the model for liquid hold-up is:

$$h_{L,bc} = 0.017(Fr)^{0.23}(Ga)^{-0.11}(Eot)^{-0.28}\epsilon^{-2.46}F_a \quad (12)$$

Liquid hold-up is a critical parameter that strongly affects the pressure drop, due to the significant reduction of area associated with the liquid accumulation in the column. If good values of dry pressure drop and liquid hold-up are obtained, it is expected that the prediction of pressure drop under irrigated conditions will be automatically improved.

Liquid Hold-Up Above the Loading Point

The following relation, employed previously by several authors,^[2,3,12,14] was also used in this work to model the liquid hold-up value above the loading point:



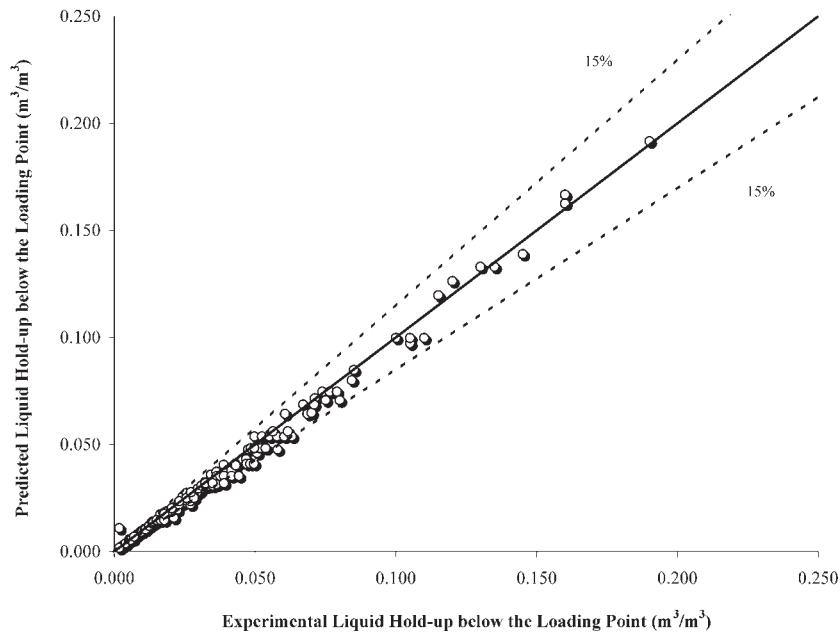


Figure 9. Comparison of predicted and experimental values of the liquid hold-up at the loading point using Eq. (12) (final expression).

$$h_L = h_{L,bc} \left[1 + C_1 \left(\frac{\Delta P_{\text{irr}}}{(z \rho_L g)} \right)^{C_2} \right] \quad (13)$$

As can be seen, this expression has two parameters and relates the liquid hold-up above the loading point with the value of the hold-up at the loading point and the pressure drop value. Once the hold-up at the loading point has been calculated, it is necessary to solve simultaneously with an iterative procedure the equations for liquid hold-up above the loading point and the pressure drop (discussed in the following section). The data bank of Table 1 was used to regress the C_1 and C_2 constants of Eq. (13) for bale-type packing, whose values were:

$$\begin{aligned} C_1 &= 17.8 \\ C_2 &= 1.62 \end{aligned}$$

Figure 10 shows the parity plot comparing the predictions of the liquid hold-up with the experimental data. It can be seen that 90% of the values have absolute



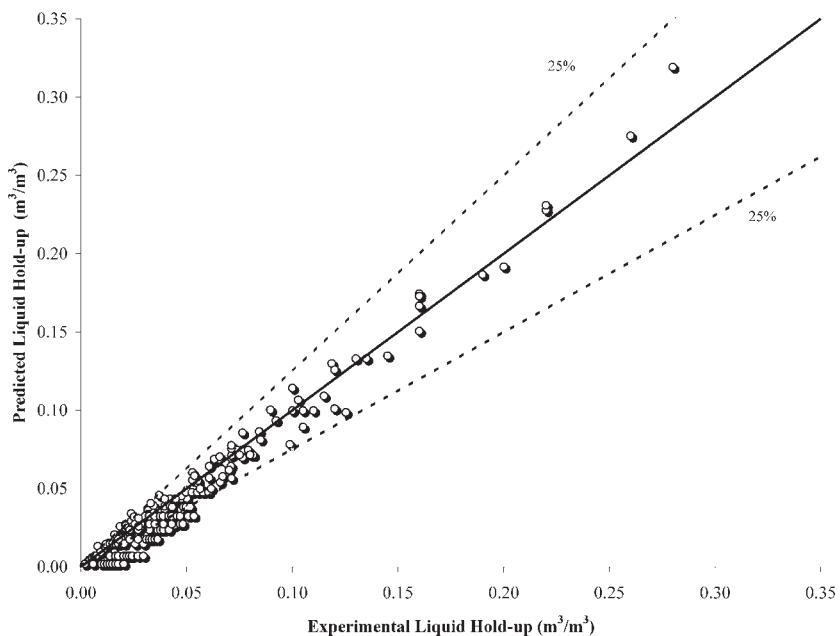


Figure 10. Comparison of the predicted and experimental values of the liquid hold-up using the model developed in this work.

deviations smaller than 25%, which is a significant improvement in relation to the application of the model of Engel et al.^[12] to bale packings, which was 50% (Fig. 3). It can be seen in Fig. 10 that the largest deviations occur at very low values of liquid hold-up.

Pressure Drop Under Irrigated Conditions

The expression for pressure drop under irrigated conditions proposed by Stichlmair et al. was modified by Engel et al.^[12] considering that friction is not only provided by the surface area associated to the packing but also by the area provided by the drops and rivulets of liquid present in the column. In this work, the expression of Engel et al.^[12] was also modified to take into account the fact that most of the data used in this work has been obtained in small columns. It has been shown that in a 7.5-cm column filled with a packing with $200\text{ m}^2/\text{m}^3$ of surface area, the wall provides almost 25% of the total area available in the column, which contrasts with large-diameter columns (larger



than 1 m), where the wall contribution is negligible.^[15] Based on this, the following expression was used for the calculation of pressure drop:

$$\frac{\Delta P_{\text{irr}}}{z} = \frac{1}{8} f_o (a_p + a_L + a_w) \frac{\rho_G u_G^2}{(\varepsilon - h_L)^{4.65}} \quad (14)$$

Figure 11 shows the comparison of the experimental data with the values predicted with the previous equation. In this case, 90% of the points show absolute errors lower than 30%, which is a significant improvement in relation to the application of the Engel et al.^[12] model (Fig. 2), which showed 90% of the data with errors lower than 60%.

Loading and Flooding Points

Loading point was defined here as the gas velocity (or F-factor) at which liquid hold-up in the column starts to increase. This can be easily calculated for each case by combining Eqs. (13) and (14) and recognizing that at the

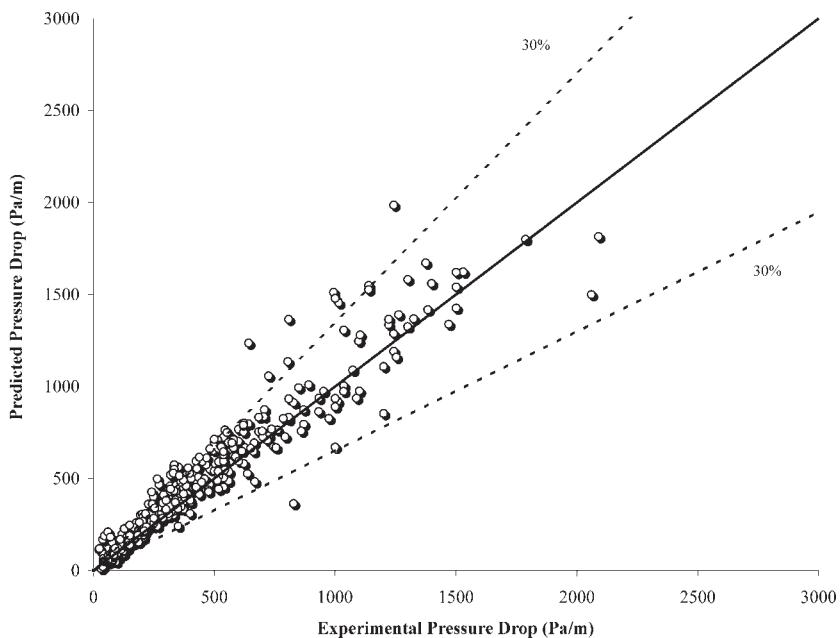


Figure 11. Comparison of the predicted and experimental values of the pressure drop using the model developed in this work.



loading point $h_L = h_{L,bc}$. Figure 12 shows the comparison of the predicted and experimental values of loading point. It can be seen that the agreement of the predicted values with the real data is very good since 90% of the points show absolute deviations smaller than $\pm 10\%$. This is a significant improvement in comparison with the predictions of the Engel et al. model in which 90% of the data were predicted within an error of $\pm 70\%$.

Flooding point can be calculated from Eqs. (13) and (14), recognizing that at this point the change of the pressure drop with the gas velocity is infinite, which can also be expressed as:

$$\begin{aligned} \frac{\partial \Delta P_{\text{irr}}}{\partial \Delta P_d} &= \infty \\ \frac{\partial \Delta P_d}{\partial \Delta P_{\text{irr}}} &= 0 \end{aligned} \quad (15)$$

Combining Eqs. (1) and (14), and with the use of the relation in Eq. (15), an expression for the calculation of the gas velocity at the flooding point can be

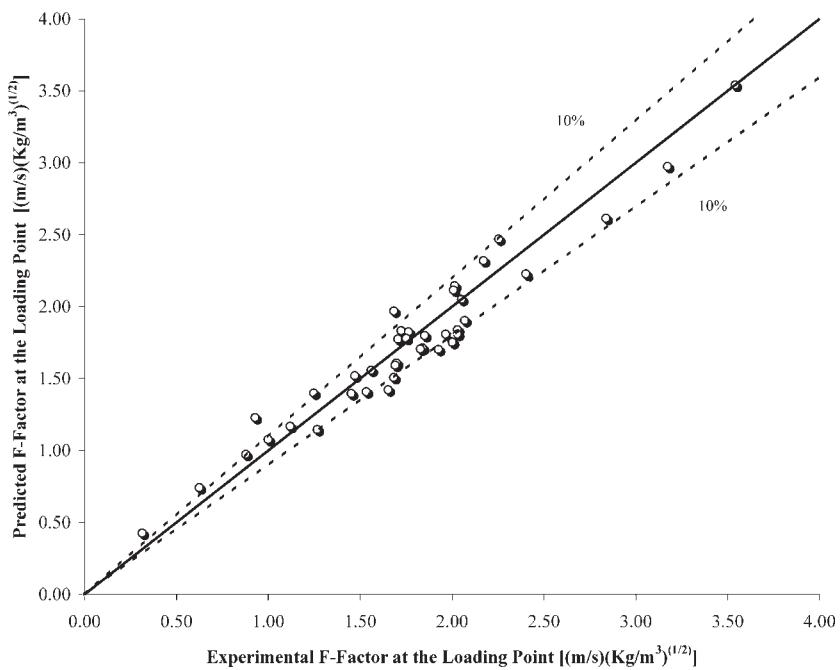


Figure 12. Comparison of the predicted and experimental values of the F-factor at the loading point using the model developed in this work.



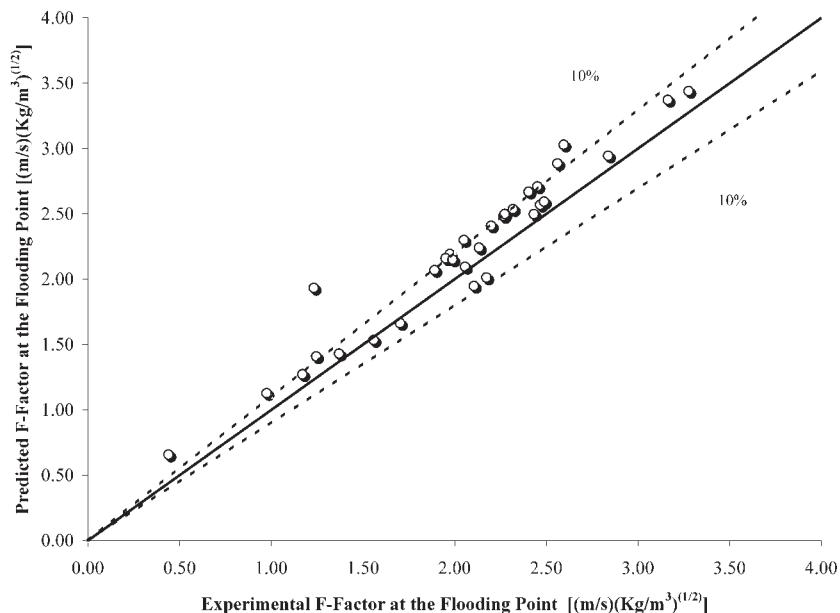


Figure 13. Comparison of the predicted and experimental values of the F-factor at the flooding point using the model developed in this work.

obtained. Figure 13 shows a very good prediction of the flooding point using the model, since 90% of the points show deviations lower than $\pm 10\%$ in relation to the experimental data.

Performance of the Model

Figures 14 and 15 show typical plots of the pressure drop and liquid hold-up as a function of the F-factor at several liquid velocities. It can be seen that the model (continuous lines) represents with reasonable accuracy both the trends and the absolute values of the experimental points.

CONCLUSIONS

A new model has been developed for the flowdynamic representation of catalytic distillation columns loaded with bale-type packing. This model is a modification and improvement of the original Stichlmair et al.^[3] model for



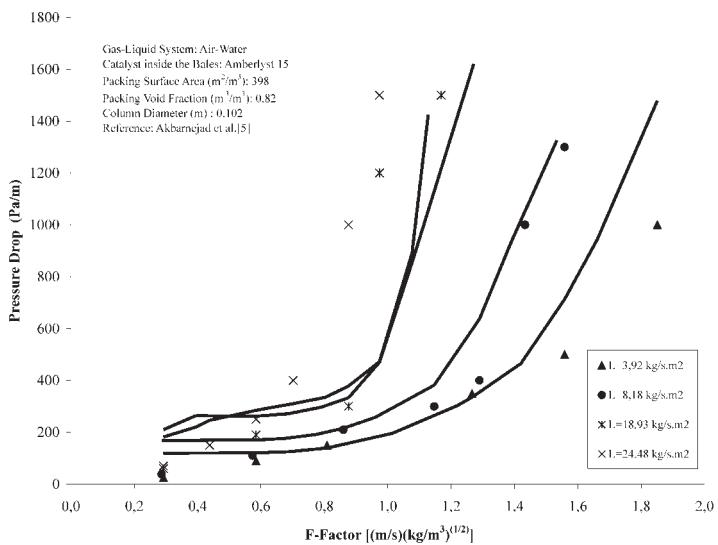


Figure 14. Evaluation of the model performance in the pressure drop prediction at different gas and liquid velocities.

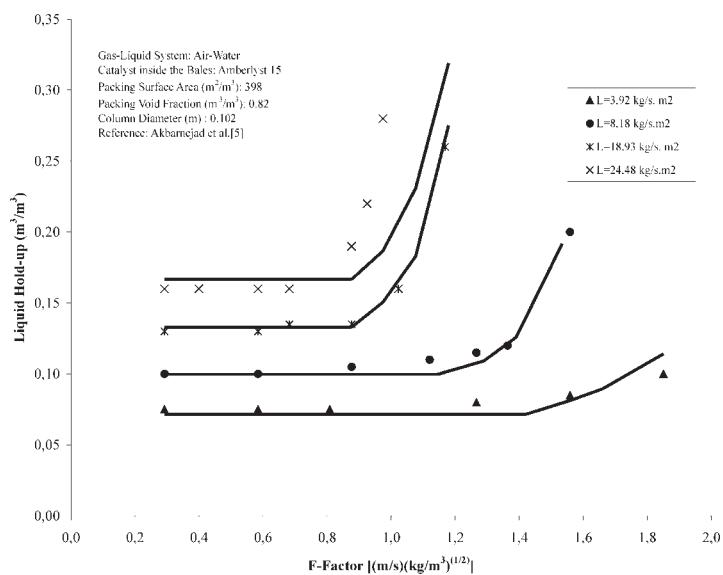


Figure 15. Evaluation of the model performance in the liquid hold-up prediction at different gas and liquid velocities.



random packings and represents a significant improvement in comparison to the application of previous models to this type of packings. Predictions of flooding and loading points match experimental data within an error of $\pm 10\%$, while liquid hold-up is predicted within $\pm 15\%$, of error. The model introduces specific considerations to take into account the nature and amount of catalyst loaded in the pockets of the bales and it is especially useful for low-scale columns typically used in experimentation. Further evaluations are needed to verify the performance of the new model with data obtained on large-scale columns, as well as better understanding of the effect of the nature of the catalyst on the liquid hold-up.

SYMBOLS

a_L	Surface area of the liquid in the column calculated as $a_L = 6h_L/d_L$ (m^2/m^3).
a_p	Specific surface area of the packing (m^2/m^3).
a_w	Surface area of the column per volume unit (m^2/m^3).
C_i	Models constants (dimensionless).
c_i	Models constants (dimensionless).
d_c	Column diameter (m).
d_L	Average diameter of liquid drops in the column calculated as $d_L = C_L \sqrt{6\sigma_L/\Delta\rho \cdot g}$ (m).
d_p	Packing particle diameter calculated as $d_p = 6(1 - \varepsilon)/a_p$ (m).
Eot	Eotvos number defined according to Eq. (5) (dimensionless).
F	Gas capacity factor defined as $F = u_G \sqrt{\rho_G}$ [(m/sec) (kg/m^3) $^{1/2}$].
F_a	Affinity factor between the liquid and the catalyst for Eq. (12) (dimensionless).
f_o	Stichlmair's friction factor calculated according to Eq. (2) (dimensionless).
Fr	Froude number defined according Eq. (3) (dimensionless).
g	Gravity acceleration (m/sec^2).
Ga	Galileo number defined according to Eq. (4) (dimensionless).
h_L	Liquid hold-up (m^2/m^3).
$h_{L,bc}$	Liquid hold-up below the loading point (m^2/m^3).
Re_G	Gas Reynolds number (dimensionless).
u_G	Superficial gas velocity (m/sec).
u_L	Superficial liquid velocity (m/sec).
z	Total height of the packing section of the column (m).



GREEK SYMBOLS

ΔP_d	Dry pressure drop (Pa/m).
ΔP_{irr}	Irrigated pressure drop (Pa/m).
ε	Column void fraction of the packing section (m ³ /m ³).
μ_G	Gas dynamic viscosity (kg/m sec).
μ_L	Liquid dynamic viscosity (kg/m sec).
ρ_G	Gas density (kg/m ³).
ρ_L	Liquid density (kg/m ³).
σ	Surface tension (dynes/cm ²).

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