MODELING THE HYDRODYNAMICS OF LAST-GENERATION CATALYTIC STRUCTURED PACKINGS

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Abstract

Modern catalytic structured packings offer a uniform catalyst support where catalyst particles can be embedded thus combining its features with the well-known advantages of a structured packing (high capacity and high mass-transfer efficiency). In this work, the hydrodynamic properties of a novel family of catalytic structured packings (KATAPAK®-SP) have been modeled using a modified version of a hydraulic model originally proposed by Hoffmann et al.[1] for MULTIPAK®. Modifications to the model included: (1) the adaptation of a more suitable liquid holdup correlation for the open channels, and (2) the use of a different split factor for the liquid flowing through the catalyst bags and the open channels.

1. Hoffmann et al., Chem. Eng. Proc., 43, 383 (2004).

Introduction

The advantages of combining chemical reaction with thermal separation into a single unit have been widely recognized. For example, chemical equilibrium limitations can be overcome by continuously removing reaction products from the reaction mixture; higher conversions can be thus obtained. Further, side reactions are minimized and recycle costs and energy needs are greatly reduced. In this context, reactive distillation emerges as one of the most attractive and popular reaction-separation units nowadays used in the chemical process industry. Reactive distillation continues evolving thanks to the development of last-generation contacting devices such as catalytic structured packings (CSP). A relatively new family of CSP has been developed by Sulzer Chemtech bearing the name of KATAPAK-SP (SP stands for Separation Performance). Unlike conventional structured packings, KATAPAK-SP offers an alternating scheme of corrugated sheets and bags containing the catalyst particles. The corrugated sheet serves as a separation layer and provides the only flow passage for the gas phase through channels of triangular cross section and inclined with an angle of θ to the horizontal. As stated by Gotze et al.[1], the advantage of this CSP relies on its modular design that allows to vary the catalyst fraction and the separation efficiency. This feature in combination with the well-known advantages of a structured packing (high capacity and high mass-transfer efficiency) make KATAPAK-SP a highly competitive internal device for reactive distillation columns.

Comprehensive experimental studies have been recently conducted to investigate the hydrodynamic behavior of various types of KATAPAK-SP using the air-water system at ambient conditions. For example, Gotze et al.[1] reported on experimental measurements in terms of separation efficiency, pressure drop, dynamic liquid holdup and residence time distribution (RTD) for KATAPAK-SP type 12 (2 separation layers per 1 catalytic layer) using a 250-mm I.D. column. Later, Ratheesh and Kannan[2] also experimentally measured the hydrodynamic behavior of KATAPAK-SP 12 but using a rather smaller column diameter (100 mm). More recently, Brunazzi et al.[3] conducted an experimental work in a 100-mm I.D. column filled with KATAPAK-SP 11 (1 separation layer per 1 catalytic layer) and obtained pressure drop, dynamic liquid holdup and RTD measurements. Various hydrodynamic models have been reported in the literature[4,5,6] in an effort to interpret and represent the experimental results so far obtained for several types of CSP. Among these models, the proposal of Hoffmann et al.[6] appears to be most suitable one in correlating and/or predicting the hydrodynamic behavior of the KATAPAK-SP packing family. Accordingly, the main purpose of this work was to properly represent the experimental pressure drop and volumetric liquid holdup data of two last-generation CSP (KATAPAK-SP 11 and 12) via the use of a modified version of the hydraulic model originally proposed by Hoffmann et al.[6]

Original Hoffmann’s Approach for CSP

One of the few pressure drop models for CSP available from literature was that proposed by Hoffmann et al.[6] The authors originally developed their model in an attempt to represent the experimental pressure drop and liquid holdup data they also obtained for two different versions of MULTIPAK, another CSP similar to KATAPAK-SP 11. Unlike the latter, the separation layers of MULTIPAK consist of corrugated wire gauze sheets with arrangements having two different inclination angles: 60° for MULTIPAK-I and 45° for MULTIPAK-II. Hoffmann’s “hybrid” approach comprises different modeling contributions to calculate the pressure drop (of the gas through the open channels) and liquid holdup at given gas and liquid loads:

$$\text{Hoffmann's approach} \begin{cases} \text{Dry Pressure Drop : the channel model of Billet}^{7} \\ \text{Liquid Holdup Below Gas Loading : Mackowiak correlation}^{8} \\ \text{Wet Pressure Drop : Buchanan model}^{9} \\ \text{Liquid Holdup Above Gas Loading : Stichlmair correlation}^{10} \end{cases}$$

Hoffmann et al.[6] also give a set of equations for calculating the volumetric liquid holdup inside the catalyst bags by assuming a reasonable liquid split. According to the authors, total liquid load (L_T) is divided in two main portions:

$$L_T = L_{CB} + L_{OC} \tag{1}$$

where L_{CB} and L_{OC} stand for the amount of liquid flowing through the catalyst bags (CB) and the open channels (OC), respectively. In terms of superficial velocities, the above equation becomes:

$$U_L = \varphi \cdot U_{CB} + \varepsilon \cdot U_{OC} \tag{2}$$

where φ is the catalyst volume fraction and ε is the void fraction of the open channels. In their paper, Hoffmann et al.[6] set the distribution of liquid load by assuming equal superficial velocities through CB and OC:

$$U_{CB} = U_{OC} \tag{3}$$

In terms of known variables:

$$U_{CB} = \frac{U_L}{\varphi + \varepsilon} \tag{4}$$

$$U_{OC} = \frac{(U_L - U_{CB} \cdot \varphi)}{\varepsilon} \tag{5}$$

Eqs. (4) and (5) apply up to the load point (LP) of the catalyst bags. At this point there is a maximum liquid load at which all voids inside the catalyst bags get completely filled of liquid. Hoffmann et al.[6] adapted the Moritz-Hasse[4] approach to determine the load point for the case of MULTIPAK I and II. The application of the Hoffmann's approach for a particular CSP requires the use of six characteristic parameters as follows: 2 constants for the Billet model (D and E), 2 constants for the Mackowiak correlation (A and B), one constant for the Buchanan equation (F) and one constant for the Stichlmair correlation (C). For MULTIPAK I and II, Hoffmann et al.[6] treated the two Billet's parameters as packing-specific constants whereas the others were set as universal constants.

Preliminary Pressure Drop Calculations

In an effort to adequately represent the experimental hydrodynamic behavior (namely, pressure drop and liquid holdup) of KATAPAK-SP 11 and 12, the Hoffmann's approach was chosen in this work as the modeling framework. Firstly, the original Hoffmann's equations and associated model parameters were tested with some exceptions: (1) Billet's constants (D and E) were adjusted to fit KATAPAK-SP 11 and 12 experimental dry pressure drop data. The table below gives these two values obtained for the two CSP under study:

CSP	*D*	*E*	*Experimental Data*
KATAPAK-SP 11	0.01123	14.8638	Brunazzi et al.[3]
KATAPAK-SP 12	0.02402	12.6057	Ratheesh and Kannan[2]

(2) Although presumably used by Hoffmann et al.[6], a calculation procedure devised by Brunazzi et al.[11] was used in this work to correctly compute maximum capacity conditions, namely the flooding point for a given liquid load, and finally (3) liquid holdup through the wire gauze sheets ($H_{L,wg}$) was calculated in a somewhat different manner; unlike the CSP considered by Hoffmann et al., KATAPAK-SP separation layers are of the sheet-metal type and only the bags containing the catalytic beads are made out of wire-gauze material:

$$H_{L,wg} = \varepsilon_{wg} \cdot \varphi_{wg} \text{ (assuming complete wetting)}$$

where ε_{wg} and φ_{wg} correspond to the void fraction and the volume fraction of the wire gauze, respectively. All the geometric characteristics of two KATAPAK-SP packings needed in the Hoffmann's approach were previously estimated by Brunazzi et al.[3]

Other relevant input data for the Hoffmann's approach included suitable values for the various model parameters used in the Mackowiak, Buchanan and Stichlmair equations. As a matter of fact, Hoffmann et al.[6] treated these parameters as universal constants for the two types of MULTIPAK packing they studied. In order to first verify the generality of the aforementioned parameters, the same values proposed by Hoffmann et al. were used in this study for the two KATAPAK-SP packings; that is, A = 0.2, B = 0.25 for the Mackoviak correlation, C = 150 for the Stichlmair equation, and F = 2 for the Buchanan model.

Figures 1 and 2 graphically depict the ability of the original Hoffmann's approach in both correlating dry pressure drop data and predicting wet pressure drop data for KATAPAK-SP 11 and 12 at different liquid loads. Figure 1 shows a parity plot between experimental pressure drop data for KATAPAK-SP 11 and that predicted using the Hoffmann's approach, including dry pressure drop conditions (green circles). As shown by this figure, the agreement between experimental and calculated data is quite acceptable for those points in blue representing pressure drop data below the loading point (about 15% away from the flooding gas velocity). Above the loading point, Hoffmann's model largely under-predicts pressure drop data (red circles). Model performance for KATAPAK-SP 12, however, is not as good as for KATAPAK-SP 11. As evidenced by Figure 2, the Hoffmann model largely over-predicts pressure drop data even below the gas loading point for all liquid loads.

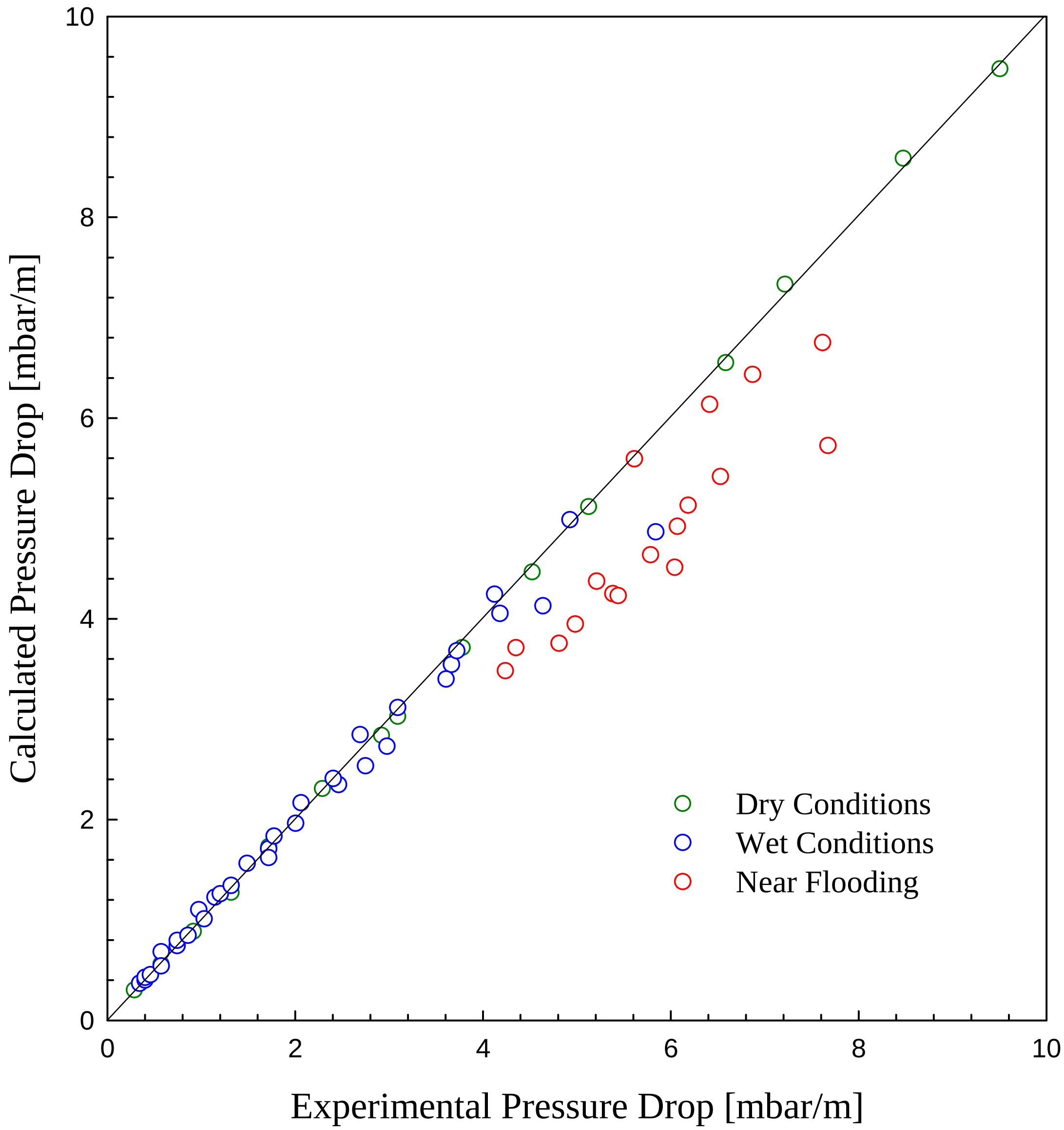

Figure 1. Calculated pressure drop for KATAPAK-SP 11 vs. experimental values. Calculations using original model parameters. Experimental data from Brunazzi et al.[3]

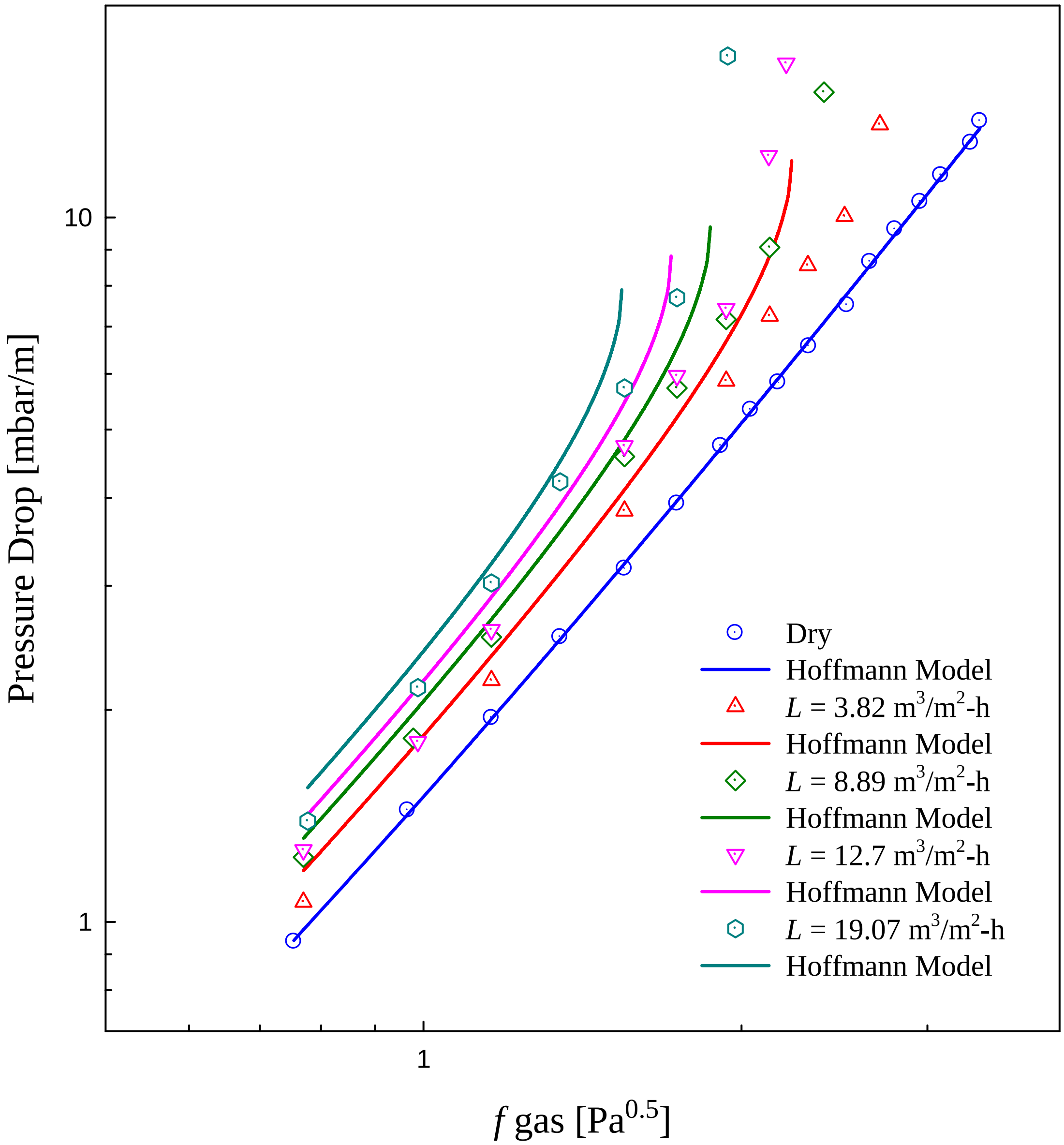

Figure 2. Pressure drop behavior for KATAPAK-SP 12. Calculations using original model parameters. Experimental points from Ratheesh and Kannan[2]

Modified Hoffmann Model

A Different Liquid Holdup Correlation for the Open Channels. As a part of possible refinements and modifications to the Hoffmann's approach, an idea was aimed to incorporate a more suitable liquid holdup model for the open channels. In KATAPAK-SP, the available area for gas flow is formed mainly by combining layers of Mellapak (a sheet-metal packing) with wire-gauze layers of catalytic bags. Accordingly, we recommend the use of the liquid holdup correlation developed by Suess and Spiegel[12], a dedicated equation for the calculation of liquid holdup for the Mellapak packing family: 250.X, 250.Y and 500.Y. The correlation reported by Suess and Spiegel calculates liquid holdup below the loading point as follows:

$$H_L^{Mellapak} = c \cdot a_p^{0.83} \cdot U_L^x \cdot (\eta_L / \eta_{L,w})^{0.25} \tag{6}$$

with

$$c = 0.0169, \quad x = 0.37 \quad \text{for} \quad U_L < 40\ \mathrm{m^3/m^2h}$$

$$c = 0.0075, \quad x = 0.59 \quad \text{for} \quad U_L > 40\ \mathrm{m^3/m^2h}$$

where a_p is the surface area of the packing in m^2/m^3, U_L is the liquid load in m^3/m^2-h, η_L is the dynamic viscosity of the liquid and $\eta_{L,w}$ is the dynamic viscosity of water at 20 ºC. To make use of the Suess-Spiegel correlation, some considerations should be made. First of all, since ambient water was used as liquid, the term $\eta_L / \eta_{L,w}$ becomes almost unity. Secondly, if one uses the Suess-Spiegel correlation directly, the calculated liquid holdup will correspond to a Mellapak-type packing having a surface area equal to that of Mellapak 500.Y ($a_p = 500$ m^2/m^3) and occupying a total cross-sectional area equivalent to that of KATAPAK's open channels. Within the KATAPAK-SP's structure, however, the Mellapak 500.Y layers occupy only a portion of the total cross-sectional area. The calculated liquid holdup should be therefore corrected according to the real geometry of KATAPAK-SP as follows:

$$H_L^{Katapak} = H_L^{Mellapak} \cdot \varepsilon \quad (7)$$

Use of a Variable Liquid Split Factor. The assumption made by Hoffmann et al.[6] about the way liquid distributes through the open channels and the catalyst bags may not be valid for the case of KATAPAK-SP. As mentioned earlier, Hoffmann's main assumption was that $U_{CB} = U_{OC}$ below the load point of the catalyst bag. In this work, the following liquid split factor (α) is introduced in order to better represent numerically the distribution of liquid between CB and OC:

$$\alpha = \frac{L_{CB}}{L_{OC}} = \frac{A_{CB} \cdot U_{CB}}{A_{OC} \cdot U_{OC}} = \frac{\varphi \cdot U_{CB}}{\varepsilon \cdot U_{OC}} \quad (8)$$

From $U_L = \varphi \cdot U_{CB} + \varepsilon \cdot U_{OC}$ it follows that:

$$U_{CB} = \frac{\alpha \cdot U_L}{(\alpha + 1) \cdot \varphi} \quad (9)$$

$$U_{OC} = \frac{U_L - \varphi \cdot U_{CB}}{\varepsilon} \quad (10)$$

Total liquid velocity U_L can be also expressed in terms of U_{CB} and α as follows:

$$U_L = \varphi \cdot U_{CB} \cdot \left(\frac{\alpha + 1}{\alpha} \right) \quad (11)$$

Accordingly, the load point velocity through the catalyst bags is:

$$U_{LP} = \varphi \cdot U_{CB,max} \cdot \left(\frac{\alpha + 1}{\alpha} \right) \quad (12)$$

$U_{CB,\max}$ is again estimated using the Moritz-Hasse[4] approach. The limiting value of α based on Hoffmann's assumption is:

$$\alpha = \frac{\varphi}{\varepsilon} \tag{13}$$

For example, for KATAPAK-SP 11: $\alpha = 0.992$. However, based on experimental evidence found by Brunazzi et al.[3] a more realistic value of α should be considered for both KATAPAK-SP packings, particularly at low liquid loads. For practical reasons, we therefore propose in this study the use of a liquid split factor dependent on liquid load for both KATAPAK-SP packings.

Application of the Modified Hoffmann Model

A final regression effort was undertaken treating α as an adjustable parameter at each liquid load and using the Suess-Spiegel holdup correlation within the Hoffmann's modeling framework. It is important to note that the Buchanan and Stichlmair parameters (*F* and *C*) were also adjusted to better represent the experimental pressure drop at liquid loads above the load point of the catalyst bags. The following table shows the values of the fitted parameters *F* and *C* for both KATAPAK-SP packings:

CSP	*F*	*C*
KATAPAK-SP 11	2.221	172.6
KATAPAK-SP 12	1.322	192.3

As a matter of fact, the above values do not greatly differ from those used by Hoffmann et al.[6] (*F* = 2, *C* = 150). Figures 3 and 4 depict the performance of the modified model in representing the experimental pressure drop data for KATAPAK-SP 11 and 12, respectively. Unlike previous pressure drop calculations using the original Hoffmann model (see Figures 1 and 2), there is a significant improvement in model predictions thanks to the modifications proposed in this work, in particular within the loading-to-flooding region as in the case for KATAPAK-SP 11 (as evidenced by Figures 1 and 3). Generally speaking, the experimental pressure drop data for both KATAPAK-SP packings was very well represented below the gas loading point. As the flooding point is approached, however, model performance gradually deteriorates at all liquid loads, particularly for KATAPAK-SP 12.

Finally, Figure 5 shows the variation of calculated liquid split factors in terms of L_{CB} / L_T ratios with liquid load over a range of 2.5-30 m^3/m^2-h for the two CSP considered in this study. This figure reveals that KATAPAK-SP 12 reaches the load point of the catalyst bags more rapidly than KATAPAK-SP 11. As a matter of fact, KATAPAK-SP 11 reaches the load point at about 18 m^3/m^2-h while KATAPAK-SP 12 does it near 14 m^3/m^2-h. Further, below L = 12 m^3/m^2-h more liquid flows through the catalyst bags for KATAPAK-SP 12 as compared with KATAPAK-SP 11. Above this point, it takes place the opposite. Evidently, all these findings require further verification by either conducting suitable hydraulic experiments or performing CFD simulations of the liquid flow.

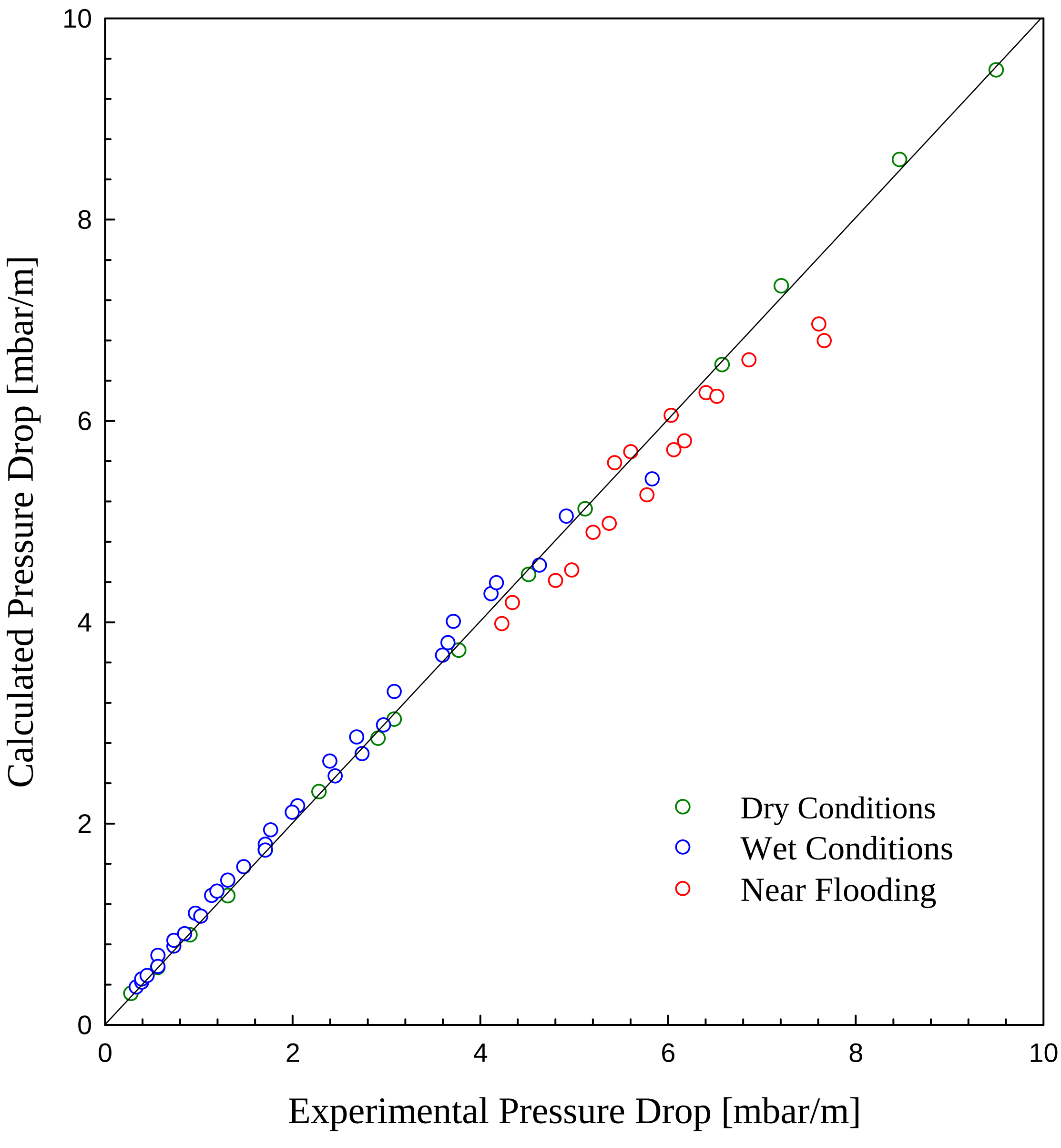

Figure 3. Calculated pressure drop for KATAPAK-SP 11 vs. experimental values. Calculations using modified Hoffmann model. Experimental data from Brunazzi et al.[3]

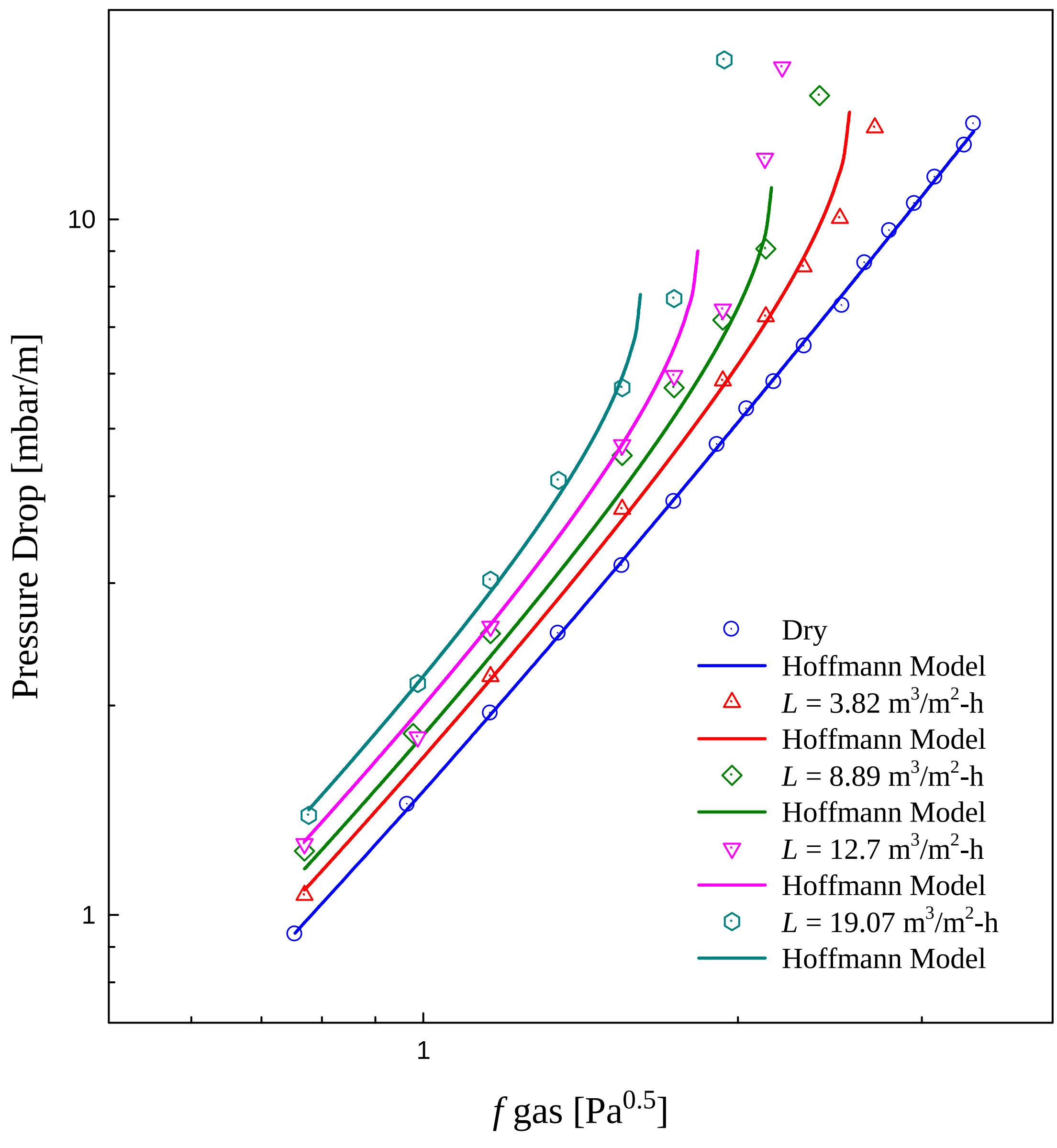

Figure 4. Pressure drop behavior for KATAPAK-SP 12. Calculations using modified Hoffmann model. Experimental points from Ratheesh and Kannan[2]

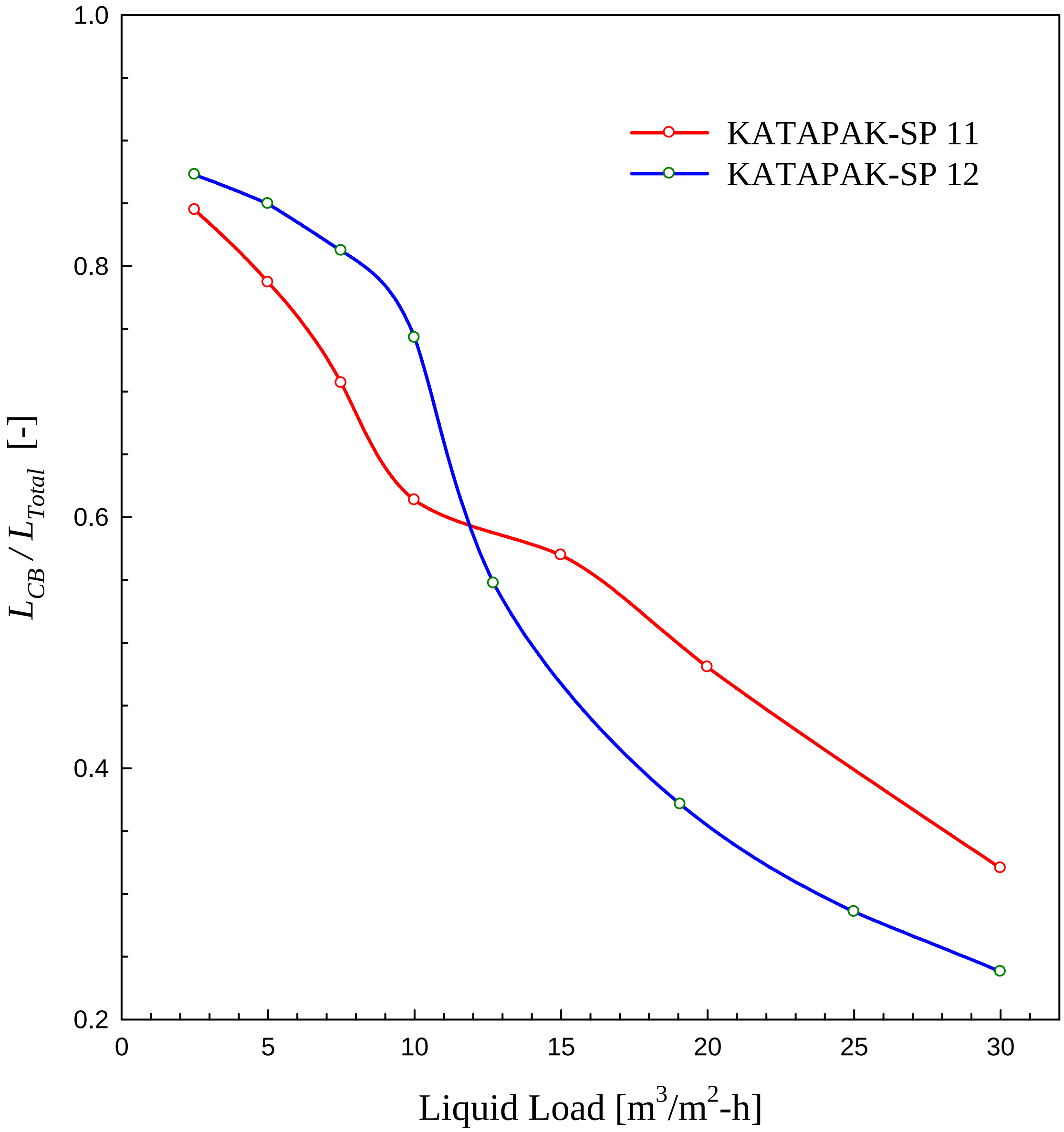

Figure 5. Variation of L_{CB}/L_{total} with liquid load for the two CSP under study

Conclusions

The hydrodynamic model originally proposed by Hoffmann et al.[6] for MULTIPAK was modified in this work in order to adequately represent the experimental pressure drop behavior of two last-generation CSP: KATAPAK-SP 11 and 12. The following conclusions can be drawn from this study:

- The use of a more suitable liquid holdup correlation for the open channels in combination with a liquid split factor dependent on liquid load gave improved pressure-drop representations below the load point of the catalyst bags.
- Above the load point of the catalyst bags, the experimental pressure drop data was equally well represented thanks to the use of adjusted values for the Buchanan and Stichlmair parameters.
- For all liquid loads, the modified model did a pretty good job in representing the observed pressure drop data for both KATAPAK-SP packings below their gas loading points. Although the modified model outperformed the original one within the loading-to-flooding region, calculated pressure drops were still underestimated, particularly for KATAPAK-SP 12.

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