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Holdup and pressure drop studies in structured packings with catalysts

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Abstract

Reactive distillation combines reaction with separation in a single column thereby increasing conversion for equilibrium limited reactions, minimizing side reactions and saving on recycle and capital costs. KATAPAK[®]-SP provides the flexibility in varying the ratio of the reaction zone relative to the separation zone depending on process requirements. The current study provides pressure drop and holdup data for KATAPAK[®]-SP 12 packing contained in a 100 mm diameter column. Correlations are provided for both the dry gas and irrigated pressure drop. The gas loading point was correlated to the liquid flow rate. Flow pattern studies were conducted to estimate the saturation capacity of the packed bed as well as to determine the apportioning of the total liquid flow between the reaction and separation layers. A correlation for the dispersed phase liquid holdup is developed based on the operating variables, equipment parameters and a flow distribution parameter estimated from the flow pattern studies. The holdup and pressure drop correlations were extended to the post-loading region. © 2004 Elsevier B.V. All rights reserved.

Keywords: Reactive distillation; Structured packing; KATAPAK®-SP 12; Pressure drop; Dispersed phase liquid holdup; Flow pattern studies

1. Introduction

In chemical process industries, enhancement of conventional processes is highly desired to minimize capital and operating costs. Combining reaction and separation in a single column enables higher conversion of equilibrium-limited reactions, minimizes side reactions, lowers recycle costs and energy requirements. Several commercial processes have successfully utilized reactive distillation and these include the manufacture of methyl acetate [1] and oxygenated ethers [2]. For the actual realization of this concept, the hardware and design aspects have to be given priority. Data from such studies also ensure reliable simulation of reactive distillation processes, for instance through the rate based models, since the pressure drop along the length of the column affects the separation and permissible flow rates while the holdup influences the reaction rates. The reactive zone in the column can affect the capacity due to the presence of special types of arrangements employed for incorporating the catalysts within

the separation layers. The development of structured packings has enabled the dual goal of reducing pressure drop while retaining the high mass transfer efficiency. The countercurrent mode of contact is usually chosen to increase separation, which however comes at the cost of restricted column capacity. In this study, the hydrodynamic aspects of KATAPAK[®]-SP 12 structured packing incorporated with catalysts are investigated.

2. Literature review on hydrodynamic studies in catalytic packings

Xu et al. [3] conducted hydrodynamic tests in a 600 mm diameter column using the air-water system. Reactive distillation packings comprising of catalyst bundles were used. These authors correlated the irrigated pressure drop and dispersed phase holdup in terms of power law models involving gas and liquid flow rates. Moritz and Hasse [4] obtained pressure drop and holdup data using KATAPAK[®]-S reactive distillation packings in a 70 mm column. They identified two regimes — one above and the other below the liquid load points. The liquid load point corresponded to the sit-

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Nomenclature

a	specific packing surface (m^2/m^3)			
AARD	average arithmetic relative deviation (%)			
A. B. C.	D. E fitting parameters			
$d_{\rm p}$	catalyst particle diameter (m)			
$d_{\rm h}^{\rm P}$	hydraulic diameter (m)			
d_{eq}	$4\varepsilon_{\rm OC}/a$ equivalent diameter (m)			
f	Fanning friction factor			
Fr	Froude number			
g	acceleration due to gravity (m/s^2)			
ĥ	liquid holdup			
L	liquid flow rate (m^3/s)			
Р	pressure (Pa)			
Re	Reynolds Number			
U	superficial gas velocity (m/s)			
w	superficial gas velocity (m/s)			
Z	height (m)			
Greek letters				
α	channel angle relative to vertical axis			
27 17	liquid distribution parameter			
γ Λ	difference			
<u>م</u>	void fraction			
и и	viscosity (kg m/s)			
v	kinematic viscosity (m^2/s)			
0	density (kg/m ³)			
φ	catalyst volume fraction			
~ • •				
Subscripts				
CB	catalyst bed			
OC	open Channel			
LP	loading point			
g	gas			
L ·	liquid			
1rr	irrigated			
dry	dry gas			

uation where the catalyst layers were saturated with liquid causing it to overflow into the adjoining channels. The liquid holdup data was empirically correlated as a ratio of the liquid Froude number to the Reynolds number. Irrigated pressure drop was expressed in terms of an enhancement to dry gas pressure drop, the enhancement being modeled as a function of the dispersed phase liquid holdup. The dry gas pressure drop in turn was modeled using the friction factor approach. This approach is based on the methodology proposed by Stichlmair et al. [5]. Ellenberger and Krishna [6] conducted hydrodynamic studies on KATAPAK[®]-S structured packings of 100 mm and 240 mm diameters. They observed that as the flow of liquid through the packed channel exceeded a certain limit, the excess liquid flowed through the open channels leading to a sharp increase in the gas press-

sure drop. The irrigated pressure drop was given in terms of an enhancement to the dry gas pressure drop with the enhancement factor expressed in terms of the liquid phase Reynolds and Froude numbers. The liquid phase holdup was also correlated in terms of these parameters. Gotze et al. [7] conducted hydrodynamic studies on KATAPAK®-SP structured packing in a 250 mm diameter column. Experimental values of pressure drop and hold up were reported for the air-water system. The values were compared to the results of KATAPAK®-S type structured packings. Gorak and Hoffman [8] investigated pressure drop, loading range and separation efficiency in reactive distillation column using MULTIPAK[®]. The hydraulic model of Rocha et al. [9] were used by these authors to predict pressure drop as a function of the *f*-factor. Hoffman et al. [10] investigated the performance of MULTIPAK® catalytic structured packing of diameters 50 mm and 100 mm. Experimental results on pressure drop and liquid hold-up were presented and models were fitted to the data. Flow visualization experiments were also carried out to identify the different flow regimes. Irrigated pressure drop was modeled in terms of the dispersed phase liquid holdup and dry gas pressure drop. The correlations developed by these investigators are summarized in Table 1.

The scope of the present work is to develop correlations for hydrodynamic factors such as holdup and pressure drop for the relatively less investigated KATAPAK[®]-SP 12 structured packing. The gas loading point is an important factor that delineates the safe operation regime from the loading and subsequent unstable flooding regime. This has to be accurately identified, correlated and incorporated into the models for regime identification. The distribution of liquid between the catalyst bed and the open channels is also to be estimated as a function of liquid flow rate. The post-loading regime, despite its instability, also offers scope for investigation since the column can transcend the loading limit in case of process variable upsets.

3. Experimental details

3.1. Column and packing

Photographs of the KATAPAK[®]-SP 12 packing element from Sulzer Chemtech Ltd. is shown in Fig. 1. SP denotes separation performance [6] while the suffix 12 denotes an alternating arrangement of two corrugated sheets and one catalyst pouch. The corrugated metal sheets serve as separation layers while the catalyst pouch serves as the reaction zone. The ratio of separation to reaction layers can be varied to impart flexibility between separation and reaction. The corrugated sheets are typical structured packing elements of Mellapak PLUS type with channels of triangular cross section. The channels are inclined to the horizontal at 45°. The catalyst pouch is made of wire gauze housing AMBERLYST-15 catalyst particles.

The details of column and packing are given in Tables 2 and 3. The schematic diagram of experimental arrangement is shown in Fig. 2. The column made from Perspex sections has provisions for feeding the gas at the bottom and liquid at the top. Structured packing elements (1) are stacked inside the column above a wedge shaped support located at the column base. Pressure tappings (2) were provided to measure the pressure drop using a 0-100 mbar differential pressure transmitter. The current signal (4–20 mA) from a differential pressure transmitter (Fuji, Japan) was monitored on line with a suitable A-D card and data acquisition software. A cone shaped arrangement at the bottom of the column

Table 2

Details of the experimental column

No.	Column detail	Value
1	Column diameter	100 mm
2	Length of column	2.5 m
3	Length of packed section	2 m
4	i.d. of Perspex section	100 mm
5	o.d. of the Perspex section	110 mm
6	Thickness of Perspex	5 mm

Table	3
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Details	on	catalytic	distillation	packing

No.	Detail	Value
1	Packing element height	20 cm
2	Diameter	10 cm
3	Ratio of number of reaction section to number of separation section	1:2
4	Packing void fraction (ε)	0.82
5	Catalyst diameter (d_p)	0.6-1.2 mm
6	Catalyst volume fraction (%) in the packing element (φ)	24.2
7	Specific surface area (a_{SP})	$236 m^2/m^3$
8	Catalyst bag void fraction (ε_{CB})	0.35
9	Open channel void fraction (ε_{OC})	0.738

(3), and a perforated multi-pipe (4) at the column top were provided to distribute the gas and liquid, respectively. The diagram of the liquid distributor is shown in Fig. 3. The liquid enters in a main line of standard 1/2 in. pipe. The distribution tubes are of 6 mm i.d. with 2 mm holes. A centrifugal pump (5) was used to transport the liquid from a storage tank (10) to the top of the column. The liquid flow rates were measured with the help of pre calibrated rotameters (6). Air from the blower (11) was fed to the bottom of the column and its flow rate is measured with the help of a turbine flow meter (Rockwin Flow meter, India Ltd.) (7). Level measurement provision (8) was provided at the bottom of the column to estimate the liquid hold up. Three rapid closing valves were provided at the air inlet, liquid inlet and liquid outlet (9) for the dispersed phase liquid holdup measurements.

3.2. Experimental procedure

The experiments mainly consisted of pressure drop data acquisition during the course of the steady state column operation and finally the holdup measurements. Initially, the bed was operated at high liquid flow rates for about 10 min to ensure the complete wetness of the bed. The air and liquid flow rates were maintained at constant values during the course of the run. The flow rates of air and water were in the range of 5.6×10^{-3} m³/s to 2.5×10^{-2} m³/s and 8.3×10^{-6} m³/s to 6.7×10^{-5} m³/s, respectively. The dynamic liquid hold up was measured using the volume displacement technique (Xu et al. [3], Ellenberger and Krishna [6]). At the time of actual experimental run, after the column attained a stable operating condition, the air inlet, water inlet and water outlet were stopped simultaneously and instantaneously. The liquid was subsequently allowed to drain and collect at the bottom section of the column. The volume of the liquid drained was measured by checking the level difference in the level meter, attached to the bottom portion of the column.

Table 1 Summary of holdup and pressure drop studies on reactive distillation packings		
Author and packings used	Correlations	
Xien et al. [3] (catalyst bundles,	h = 0.0336U	

Author and packings used	Correlations for holdup and pressure drop
Xien et al. [3] (catalyst bundles, 600 mm)	$h = 0.0336U^{0.0109}L^{0.429}; \ln\left(\frac{\Delta P}{Z}\right) = 5.539U^{0.33}L^{0.048}$
Peter Moritz and H. Hasse [4] (KATAPAK [®] -S, 70 mm)	$\Delta P_{\rm irr} = f_2 \Delta P_{\rm dry}, \text{where} f_2 = K h_{\rm A}^{\rm L} \text{and} \Delta P_{\rm dry} = f_1 \frac{\rho_{\rm g} \Delta H}{2d_{\rm h,K}} \left(\frac{w_{\rm g}}{\varepsilon_{\rm K} \cos \alpha}\right)^2$
Ellenberger and Krishna [6] (KATAPAK [®] -S, 100 mm and 240 mm)	$\varepsilon_{\rm L,OC} = 2.8 \left(\frac{Fr_{\rm L,OC}^2}{Re_{\rm L,OC}}\right)^{0.3}; \frac{\Delta P}{L} = \frac{\Delta P_{\rm dry}}{L} \exp\left[1.3\varepsilon_{\rm L,OC} \left(\frac{Re_{\rm L,OC}}{Fr_{\rm L,OC}}\right)^{0.3}\right]$
Hoffman et al. [10] (MULTIPAK [®] 50 mm and 100 mm)	$h_{\rm L} = \varphi \varepsilon_{\rm CB} \left[1 - 0.5 \left(1 - \frac{U_{\rm L}}{U_{\rm LP}} \right)^2 \right] + (1 - \varepsilon - \varphi) \varepsilon_{\rm wG} + A \left[1 + \left(\frac{\Delta P}{\rho L_{\rm g} \Delta z} \right)^2 \right] Fr_{\rm L,OC}^B (A); h_{\rm L} = \varphi \varepsilon_{\rm CB} + (1 - \varepsilon - \varphi) \varepsilon_{\rm wG} + A \left[1 + \left(\frac{\Delta P}{\rho L_{\rm g} \Delta z} \right)^2 \right] Fr_{\rm L,OC}^B (B); \frac{\Delta P}{\Delta P_0} = \left(\frac{1}{1 - Fh_{\rm L,OC}} \right)^5 \text{ (C); (A) and}$ (B) apply for holdups above and below load points, respectively
	(B) apply for holdups above and below load points, respectively

3.3. Flow pattern studies

A catalyst pouch was held in a rigid Perspex support and water was fed from top through a perforated pipe distributor. At low flow rates the water completely percolate through the catalyst bed and no water was visible on the surface of the gauze layer. At a certain superficial liquid velocity through the catalyst bed ($U_{L,CB,max}$) around 0.008 m/s, the bed's voids

were completely filled with the liquid causing the latter to flow on the gauze surface as rivulets.

3.4. Flow distribution parameter estimation

A separate experimental arrangement (Fig. 4) was created to estimate a measure of the distribution of liquid between the open channels and packed bed as a function of liquid flow rate



(a) Side view



(b) Top view

Fig. 1. Photographs of KATAPAK®-SP 12 packing (a) side view and (b) top view (with permission from Sulzer Chemtech Ltd.).



Fig. 2. Schematic diagram of the experimental setup. 1, Packed section; 2, pressure tappings; 3, air distributor; 4, liquid distributor; 5, pump; 6, rotameters; 7, turbine flow meter; 8, liquid level meter; 9, valves; 10, liquid tank; 11, blower.

for subsequent use in the holdup model development. The arrangement had provisions for separately measuring the liquid rate flowing out of the open channels and the catalyst bed. After several iterations involving various numbers of packing, a packed section involving three elements was finally chosen since the flow distribution represented a mean value over a wide range of liquid flow rates. Care was exercised to use the same distributor (Fig. 3) and Perspex sections as used in the arrangement of Fig. 2. The liquid trickling down the gauze layer of the saturated bed is construed as belonging to the open channel since a separation layer is in contact with it. However due to column capacity limitations the liquid flow rates corresponding to $U_{L,CB,max}$ was not reached.

4. Results and discussion

4.1. Dry gas pressure drop

The dry gas pressure drop (ΔP_0) was measured across the packed section when only air flowed upwards through the apparatus. These results are required to model the irrigated pressure drop as evident from the correlations in Table 1. The results were expressed in terms of a friction factor defined as follows.

$$\Delta P_0 = \frac{f\rho_{\rm g} U_{\rm ge}^2}{d_{\rm eq}} \tag{1}$$

The friction factor estimated from (1) was correlated using POLYMATH in terms of the gas phase Reynolds number as follows

For *Re* < 1500

$$f = \frac{2.293}{Re_{\rm g}^{0.308}} \tag{2}$$

while for Re > 1500, the following equation is obtained

$$f = \frac{0.628}{Re_{\alpha}^{0.131}} \tag{3}$$

where

$$Re_{\rm g} = \frac{d_{\rm eq} U_{\rm g} \rho_{\rm g}}{\mu_{\rm g}}.$$
(4)

The variation of friction factor with gas phase Reynolds number is given in Fig. 5. From these results it may be inferred that there are two regimes for gas flow in the KATAPAK[®]-SP under non-wetted conditions.

The average absolute relative deviations (AARD), defined in Appendix A, are 1.3% and 3.8%, respectively for model predictions (2) and (3), respectively. The experimental errors for calculating the abscissa and ordinate values were estimated to be $\pm 3.7\%$ and $\pm 9.3\%$, respectively.

4.2. Irrigated pressure drop

The irrigated pressure drop values are almost parallel to that of the dry pressure drop for lower gas flow rates (Fig. 6).



Fig. 3. Top view of the distributor for initial liquid distribution. The circle corresponds to the column circumference.

The errors calculated in abscissa and ordinate values were estimated to be $\pm 3.7\%$ and $\pm 2\%$, respectively.

The gas phase is assumed to flow only in the open channels under both dry and irrigated conditions. Significant gas flow through the catalyst bed will be hindered by its low void fraction (0.35) and gauze screen covering the catalyst pouch. The gas flows through inclined narrow channels with high velocities. There are two separation layers on either side of the catalyst pouch and hence the open channels of the separation layers are more exposed to gas flow. The dry gas and irrigated



Fig. 4. Arrangement for estimating flow distribution between packed and open channels. 1, Liquid from open channel; 2, liquid from packed channel; 3, pump; 4, storage tank; 5, rotameter; 6, distributor; 7, open channel; 8, packed catalyst bed; 9, packed section.



Fig. 5. Variation of friction factor with Reynolds number. (\bigcirc) Experimental; (---) Re < 1500; (—) Re > 1500.

pressure drop relations in the pre-loading region would not have been parallel over a wide range of liquid and gas flow rates if significant flow of gas occurred in the catalyst bed that gradually reduced at higher liquid flow rates. The gas flow assumption has also been made by Ellenberger and Krishna [6] and Moritz and Hasse [4] for KATAPAK[®]-S. Hoffman et al. [10] have also used this assumption for MULTIPAK[®] packings where for every catalyst zone, there is only one separation zone. Further, applying Stichlmair et al. [5] and Ergun equation [11] for dry gas pressure drop in packed bed with the catalyst particles leads to an inordinately high pressure drop in the packed channels that were not realized in actual experiments.

After a particular gas superficial velocity whose value decreases with increasing liquid load, the gas loading point is reached where the pressure drop begins to increase signifi-



Fig. 6. Variation of pressure drop with *f*-factor at various liquid flow rates. Liquid flow rate $\times 10^{-6} \text{ m}^3$ /s: (\bigcirc) 0; (\blacksquare) 0.833; (\bullet) 1.11; (\checkmark) 1.94; (\Box) 2.77; (\triangle) 3.33; (\bullet) 4.16.



Fig. 7. Variation of irrigated pressure drop with liquid load. Gas superficial velocity: (\Box) 1.02 m/s; (\triangle) 1.27 m/s.

cantly. The irrigated pressure drop increases sharply after the loading region and a steep increase occurs as shown in Fig. 6 at the proximity to the flooding region.

Unlike Ellen Berger and Krishna's observation [6] with KATAPK[®]-S packings, where the irrigated pressure drop did not immediately increase from the dry gas pressure drop values with increasing liquid flow rates until a certain critical value was reached, it was observed in the current work that the irrigated pressure drop increased with liquid flow rate even at very low values. This is indicated in Fig. 7. The data obtained indicates that even at low liquid flow rates there was some liquid flow in the open channel contributing to increase in the irrigated pressure drop.

Adopting the approach of Stilchmair et al. [5], a correlation for irrigated pressure drop was developed in terms of the enhancement over dry gas pressure drop for the preloading region. This approach has been used as the basis for the models for irrigated pressure drop developed by Ellenberger and Krishna [6] and Moritz and Hasse [4] for KATAPAK[®]-S packings. The irrigated pressure drop is correlated in terms of the liquid holdup and dry gas pressure drop-using POLY-MATH as

$$\left(\frac{\Delta P}{\Delta z}\right)_{\rm irr} = 1.941(h_{\rm L}^{0.446}) \left(\frac{\Delta P}{\Delta z}\right)_{\rm dry}^{1.12} \tag{5}$$

The strong influence of dispersed phase holdup is evident from this correlation. The model prediction compares well to experimental data within $\pm 5\%$ in Fig. 8. AARD is calculated as 2.2% for this fit as well and the maximum expected experimental error is $\pm 3\%$.

4.3. Gas loading

The variation of gas loading velocity ($U_{G,LP}$) with the liquid flow rate was also determined. This is defined in terms of the gas flow rate that causes the dispersed phase holdup to increase from a constant value at a given liquid flow rate. From the experimental results it was observed that the gas load point to decrease with the liquid load as shown in Fig. 9. The correlation obtained for the gas loading point as a function



Fig. 8. Comparison of experimental irrigated pressure drop with model (Eq. (5)) predictions.

of liquid flow rate is given as

$$U_{\rm g,LP} = 0.276 U_{\rm L}^{-0.285} \tag{6}$$

where U_g and U_L are in m/s. This correlation enables the accurate delineation of the preloading region from the postloading region. The AARD based on the model fit is given as 5%. The errors for calculating abscissa and ordinate values were estimated at $\pm 1\%$ and $\pm 3.7\%$, respectively.

4.4. Model for liquid holdup

The liquid holdup is an important parameter determining the packed tower pressure drop, capacity, as well as efficiency. The approach of Hoffman et al. [10] is modified and extended upon in the current work to develop the holdup model that is applicable to the post-loading region as well. The hold up in the packed column arises from contributions of the liquid presence in the open channels as well as in the packed bed. Below the gas loading point ($U_{g,LP}$), the holdup is independent of the gas velocity and hence the pressure drop. The



Fig. 9. Variation of gas load point velocity with liquid velocity. (\Box) Experimental; (-) predicted.

liquid holdup in this regime is expressed as

$$h_{\rm LP} = h_{\rm L,OC} + h_{\rm L,CB} \tag{7}$$

The hold up in the open channels is modeled in terms of Froude number (Fr_{OC}) as follows:

$$h_{\rm L,OC} = A \, Fr_{\rm OC}^{B} = A \left(\frac{U_{\rm L,OC}^{2} a}{g \sin \theta}\right)^{B} \tag{8}$$

where $U_{L,OC}$ is the superficial velocity of the liquid in the open channel. The holdup in catalyst bag is expressed as

$$h_{\rm L,CB} = \varphi \varepsilon_{\rm CB} \left(\frac{U_{\rm L,CB}}{U_{\rm L,CB,max}} \right)^m, \quad \text{if } U_{\rm L,CB} \le U_{\rm L,CB,max}$$
(9)

$$h_{\rm L,CB} = \varphi \varepsilon_{\rm CB}, \quad \text{if } U_{\rm L,CB} > U_{\rm L,CB,max}$$
(10)

Below the liquid saturation point $U_{L,CB,max}$ the holdup in the catalyst bed is modeled in terms of the fractional approach to saturation. It is expressed here as a function of ratio of actual liquid flow rates to that corresponding to saturation. Above the saturation liquid velocity the void spaces are completely filled with liquid. The corresponding $h_{L,CB}$ will then simply be the void spacing of the catalyst bed viz. $\varphi \varepsilon_{CB}$. The parameter '*m*' is an empirical constant that has to be estimated. From Eqs. (7)–(10), the holdup (h_{LP}) in the preloading region is expressed as

$$h_{\rm LP} = A \, Fr_{\rm OC}^{B} + \varphi \varepsilon_{\rm CB} \left(\frac{U_{\rm L,CB}}{U_{\rm L,CB,max}} \right)^{m} \tag{11}$$

$$h_{\rm LP} = A \left(\frac{U_{\rm L,OC}^2 a}{g \sin \theta}\right)^B + \varphi \varepsilon_{\rm CB} \left(\frac{U_{\rm L,CB}}{U_{\rm L,CB,max}}\right)^m \tag{12}$$

Since the superficial liquid velocity (U_L) can be related to the superficial velocities in the open channel and catalyst bag as follows [10]

$$U_{\rm L} = \varepsilon_{\rm OC} U_{\rm L,OC} + \varphi U_{\rm L,CB} \tag{13}$$

Defining the flow distribution parameter γ as

$$\gamma = \frac{A_{\rm OC}U_{\rm L,OC}}{A_{\rm CB}U_{\rm L,CB}} \tag{14}$$

From Eqs. (13) and (14) the liquid flow rate in the catalyst bag and open channel is given as

$$U_{\rm L,CB} = \frac{U_{\rm L}}{\varphi(\gamma+1)} \tag{15}$$

$$U_{\rm L,OC} = \frac{\gamma U_{\rm L}}{\varepsilon_{\rm OC}(\gamma+1)} \tag{16}$$

The expression for holdup in the preloading region (Eq. (12)) becomes

$$h_{\rm LP} = A \left(\frac{\gamma^2 U_{\rm L}^2 a}{\varepsilon_{\rm OC}^2 (\gamma + 1)^2 g \sin \theta} \right)^B + \varphi \varepsilon_{\rm CB} \left(\frac{U_{\rm L}}{\varphi (\gamma + 1) U_{\rm L, CB, max}} \right)^m$$
(17)



Fig. 10. Variation of flow distribution parameter (γ) with liquid velocity.

when $U_{L,CB} \leq U_{L,CB,max}$; and

$$h_{\rm LP} = A \left(\frac{\gamma^2 U_{\rm L}^2 a}{\varepsilon_{\rm OC}^2 (\gamma + 1)^2 g \sin \theta} \right)^B + \varphi \varepsilon_{\rm CB}$$
(18)

when $U_{L,CB} \ge U_{L,CB,max}$.

Above the gas loading point, the liquid holdup becomes a function of gas velocity in addition to that of the liquid. Since the gas velocity is directly related to pressure drop, the holdup expression was found to be suitably fitted as

$$h_{\rm L} = h_{\rm LP} + C \left(\frac{\Delta P / \Delta Z}{\rho_{\rm L} g}\right)^D \tag{19}$$

This model, similar to that proposed earlier by Stichlmair et al. [5] expresses the holdup as an increment to that obtained in the preloading region due to the influence of the gas velocity and consequently the pressure drop.

In a related earlier work by Hoffman [10], the flow distribution parameter (γ) was assumed to be equal to one. However in KATAPAK[®]-SP 12, two separation layers are sandwiched between the catalytic beds (Fig. 1). Hence it is not possible to take γ as unity as it can neither be assumed that an equal distribution of liquid occurs through the catalytic and separation layers nor such a distribution will be independent of liquid flow rate. It was found that γ was indeed a function of the liquid flow rate (Fig. 10) and fitted as in Eq. (20). The AARD was estimated at 22.48%, respectively. The % error in abscissa values is calculated as $\pm 3.1\%$ while for ordinate, the maximum experimental error was estimated at $\pm 20\%$.

$$\gamma = 156.65U_{\rm L} - 0.1512 \tag{20}$$

Substituting Eq. (20) in Eq. (17), the fractional holdup data was correlated.

Below the gas loading point the holdup equation becomes $(h = h_{LP})$

$$h_{\rm LP} = A \left(\frac{(1237U_{\rm L} - 1.19)U_{\rm L}}{(156.65U_{\rm L} + 0.85)} \right)^B + 8.5 \left(\frac{516U_{\rm L}}{156.65U_{\rm L} + 0.85} \right)^m$$
(21)



Fig. 11. Comparison of experimental and predicted holdup values.

•

0

where the estimated parameters were A = 2; B = 0.048; m = 0.587.

In post loading region, the correlation obtained was

$$h_{\rm L} = h_{\rm LP} + C \left(\frac{\Delta P / \Delta z}{\rho_{\rm L} g}\right)^D \tag{22}$$

where C = 1.108 and D = 1.89.

2 27

3.32

The comparison between predicted and experimental holdup is shown in Fig. 11 for a few typical runs. For intermediate liquid flow rates, a similar fit with Eqs. (21) and (22) was obtained. The AARD is calculated as 8.8% with the experimental error in estimating the abscissa and ordinate values being estimated at $\pm 3.7\%$ and $\pm 1.5\%$, respectively.

After the gas loading point $(U_{g,LP})$ the liquid starts to load the column and pressure drop increase is more rapid than in the preloading region. Further increase in gas velocity will cause steep increase in pressure drop and column will flood. In the post-loading region the pressure drop is correlated in terms of liquid superficial velocity and *f*-factor as

$$\left(\frac{\Delta P/\Delta z}{\rho_{\rm L}g}\right) = a U_{\rm L}^{\rm b} (U_{\rm g}\sqrt{\rho_{\rm g}})^c \tag{23}$$

where a = 0.6208; b = 0.815; and c = 4.183.

The comparison of experimental and predicted data is shown in Fig. 12. The variation is within $\pm 20\%$. The experimental error in estimating the pressure drops in the relatively unstable post-loading region was estimated at $\pm 17.9\%$. The flooding data indicates that depending upon the liquid flow rate, the pressure drop ranges between 1200 and 1800 Pa/m (1.48–2.22 in. water/foot of packing).



Fig. 12. Comparison of experimental pressure drop with predictions (Eq. (23)) in post loading region. (\triangle) Loading; (\Box) near flooding.

5. Comparison with other packings

Pressure drop and holdup data of a few typical packings with catalysts reported in the literature for air–water system are shown in Figs. 13 and 14. Also shown are the holdup and pressure drop from a typical structured packing Mellapak 250Y [12] which is closest in comparison with the separation elements of KATAPAK[®]-SP 12. Details are given in Table 4. Due to the absence of the catalysts, the pressure drop and



Fig. 13. Comparison of Pressure drops for various Structured packings. (\blacksquare) Mellapak 250Y [12]; (\Box) KATAPAK[®]-SP [7]; (\Diamond) KATAPAK[®]-SP (current study); (\blacktriangle) KATAPAK[®]-SP [4]; \triangle Catalyst Bundle [3].

Table 4

Details of current and typical studies in literature referred to in the comparison of holdup and pressure drop results (Figs. 12 and 13)

Packing	Liquid load (m ³ /m ² h)	Column diameter (mm)
Mellapak 250Y [12]	16	100
Mellapak 250Y [13]	16	1000
KATAPAK [®] -SP [7]	20	250
Current study	20	100
KATAPAK [®] -S [4]	16.4	70
Catalyst bundle [3]	20	600



Fig. 14. Comparison of holdups for various Structured packings. (**■**) Mellapak 250Y [12]; (**□**) KATAPAK[®]-SP [7]; (\Diamond) KATAPAK[®]-SP (current study); \triangle Catalyst Bundle [3].

holdup in Mellapak 250Y are the least. The values for the KATAPAK family of packings are in the same range. The maximum values are observed for the catalyst bundles of Xu et al. [3].

6. Conclusions

The KATAPAK[®]-SP family of packings offers a flexible option for varying the extent of separation zone relative to the reaction zone. The present study focused on the hydrodynamic parameters holdup and pressure drop on KATAPAK[®]-SP 12 packings using a 2 m tall packed tower of 100 mm diameter. The correlations developed will be useful in reliable operation of the reactive distillation column and its accurate process simulation. The pressure drop in the KATAPAK®-SP 12 packing was in the range of $100-1700 \text{ Pa. m}^{-1}$ over the range of gas flows 5.6×10^{-3} m³/s to 2.5×10^{-2} m³/s and liquid flow rate ranges of 8.3×10^{-6} m³/s to 6.7×10^{-5} m³/s, respectively. Correspondingly the liquid phase dispersed holdup ranged between 8 and 16%. The liquid flow did not distribute uniformly between the catalytic and non-catalytic zones and the ratio of distribution varied with liquid flow rate. Even very low liquid flow rates caused an enhancement of irrigated pressure drop over dry pressure drop values. A correlation for identifying the gas load point is developed. Flow visualization studies indicated that above a threshold limit of liquid flow rate, the catalyst bag was completely saturated and liquid overflowed into the open channels. Further work is recommended on the analysis of different types of structured packings with catalysts including novel configurations for accurate reactive distillation process simulation and equipment design.

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Appendix A

Average absolute relative deviation (AARD) is defined as

$$AARD = \frac{\sum_{i=0}^{N_{p}} (|y_{model} - y_{exptl}| / y_{exptl})}{N_{p}} \times 100$$

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