AIDIC *Conference Series*, Vol. 10, 2011
Editor Sauro Pierucci
DOI: 10.3303/ACOS1110041

ISBN 978-88-95608-58-7 ISSN 2036-5969

THE EFFECT OF LIQUID VISCOSITY ON FLUID PHASE DISTRIBUTION IN MODULAR CATALYTIC PACKINGS

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In the frame of the present study, the influence of liquid viscosity on liquid holdup in a packed column equipped with the modular catalytic packing Katapak-SP 11 is measured. Water and an aqueous solution of glycerine, the viscosity of which equals 10 cP, are used as feed liquids. The estimation of liquid holdup is of great interest due to its strong influence on pressure drop, on solid wetting and on heat and mass transfer coefficients. Besides classical methods (e.g. drainage experiments), high energy X-ray tomography is used. The latter is a unique technique to visualize the local liquid distribution inside the complex structure of packings and to quantify the liquid holdup at different scales. Finally, the applicability of a recently proposed hydrodynamic model to the viscous system is tested.

1. INTRODUCTION

Modular catalytic packings are used in an increasing number of industrial applications where reaction and separation (i.e. distillation, absorption, extraction…) can be efficiently integrated in single equipment. Katapak-SP is the last generation modular catalytic packing manufactured by Sulzer Chemtech. The solid catalyst particles are maintained in baskets made of metallic gauze envelopes which are separated by corrugated sheets of MellapakPlus type.
Among the few experimental studies reported in the literature dedicated to the analysis of hydrodynamics in these modular catalytic packings, most have been carried out with water as working liquid (Behrens et al., 2008, Goetze et al., 2001, Viva and Brunazzi, 2009). But it is well known that industrial liquid fluids have usually a different viscosity than water and there is thus a lack of experimental data available to fully characterize hydrodynamics in modular catalytic packings.
X-ray tomography has been shown to be an efficient non-intrusive tool to see inside and to adequately image the liquid and gas flow distribution in columns filled with metal packings (Green et al., 2007, Aferka et al., 2007, Aferka et al., 2010, Viva et al., 2011a).
The present work aims at studying the influence of liquid viscosity on liquid holdup measured in a packed column equipped with Katapak-SP 11 packing elements by means of X-ray tomography. An experimental campaign has been therefore carried out by using a glycerine solution with a viscosity of 10 cP as working liquid. Results have been compared to those obtained with water in previous studies by the same technique (Viva et al., 2011b) and to those evaluated with a more classical draining method (Viva and Brunazzi, 2007, Viva and Brunazzi, 2009, Viva, 2008). Finally, the hydrodynamic model recently proposed by Viva et al. (2011b) is tested for the viscous case.

Please cite this article as: Viva A., Aferka S., Toye D., Marchot P., Crine M. and Brunazzi E., (2011), The effect of liquid viscosity on fluid phase distribution in modular catalytic packings, AIDIC Conference Series, 10, 373-382 DOI: 10.3303/ACOS1110041

2. EXPERIMENTAL SETUP

The column used for the X-ray tomographic measurements is 4 m high and has an inner diameter equal to 0.1 m. It is made of transparent PVC. The packed bed (1.6 m high) is constituted by the superposition of, from the bottom to the top, one MellapakPlus 752.Y element, four Katapak-SP 11 elements and three MellapakPlus 752.Y elements used to get an initial uniform liquid distribution. These packings are manufactured by Sulzer Chemtech, CH. They are made of stainless steel (Figure 1). Baskets in the Katapak-SP 11 elements are filled with 1 mm glass spheres. Both types of packings are 0.10 m diameter and 0.2 m high. Void fraction and nominal specific area are given in Table 1. Two zones can be distinguished in Katapak-SP 11 packing, the open channels (OC), which correspond to the separation zone where the corrugated sheets are placed, and the catalytic baskets (CB), which correspond to the reaction zone. CB and OC in the packing element occupy a surface fraction (i.e. ratio of the element section surface on the column section surface) which differs from the volumetric fraction (i.e. ratio between the element volume and the packing volume). The superficial and volumetric fractions occupied by OC and CB are summarized in Table 2.

Water and an aqueous solution of glycerine (60% wt and 10 cP) are used as working fluids. The liquid superficial velocity ranges between 5 and 25.5 $m^3/m^2/h$ for water and 4 and 22.8 $m^3/m^2/h$ for glycerine solution. A multiple point source distributor (approx. 4000 drip points/m^2) is used to feed the liquid at the top of the column.

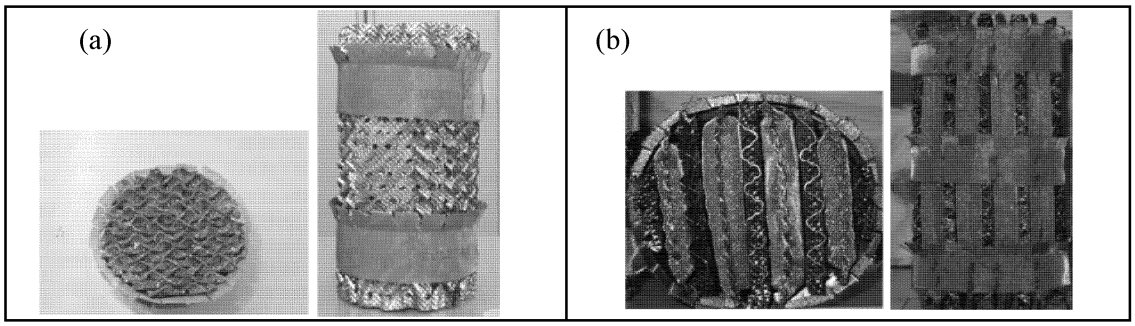

Fig. 1: Top and side view of MellapakPlus 752.Y (a) and Katapak-SP 11 (b)

Table 1: Packings geometrical parameters

	MellapakPlus 752.Y	Katapak-SP 11
Height of packing element (mm)	200	200
Diameter of packing element (mm)	100	100
Void fraction (%)	97.5	76.7
Nominal specific area (m^2/m^3)	510	203

Table 2: Superficial and volumetric fractions of OC and CB in Katapak-SP 11

	symbol	value
OC superficial fraction	ψ_{OC_S}	0.342
OC volumetric fraction	ψ_{OC_V}	0.392
CB superficial fraction	ψ_{CB_S}	0.465
CB volumetric fraction	ψ_{CB_V}	0.418

The X-ray CT facility is a high energy (420 kV) X-ray tomograph equipped with a fan beam X-ray source and with a 1280 photodiode linear detector which are both fixed on an arm able to translate vertically along the scanned object height (Figure 2). Scanned objects are put on a rotating plate which may perform a complete revolution around a vertical axis. Objects with diameters up to 0.45 m diameter and with height up to 3.8 m may be analyzed with a spatial resolution equal to 0.37 mm. More details are provided in Toye et al. (2005). This X-ray CT may work in radiographic mode as well as in tomographic mode. When used in radiographic mode, the rotating plate of the scanner is deactivated and the scanned object remains fixed. 2D radiographic images correspond to X-ray attenuation horizontal profiles measured at different heights by vertically translating the arm. In tomographic mode, the vertical arm remains fixed while the scanned object is rotated in order to get attenuation profiles for all angular positions. From these attenuation data, one may obtain the image of the column cross-section corresponding to the vertical position of the arm. Images of cross sections situated at different heights may be obtained by repeating the measurement procedure for different positions of the arm supporting the X-ray source and the detector. X-ray tomography is thus a time consuming measurement technique as, for each operating condition, a large number of cross section images (70 in the present work) have to be reconstructed to get information on phase distributions relative to the whole bed.
Figure 3 shows a radiographic image (radiogram) of the whole packed bed obtained with the X-ray CT used in radiographic mode. On this radiogram, one may see four MellapakPlus 752.Y (M1, M2, M3, M4) and four Katapak-SP 11 (K1, K2, K3, K4). Above, the liquid distributor is visible.

The specifications concerning the holdup measurements with the glycerine solution using conventional methods and the related experimental setup are described in detail in Viva and Brunazzi (2007) and Viva (2008). It is worth to point out that the same packing and prewetting procedures where used in the two facilities.

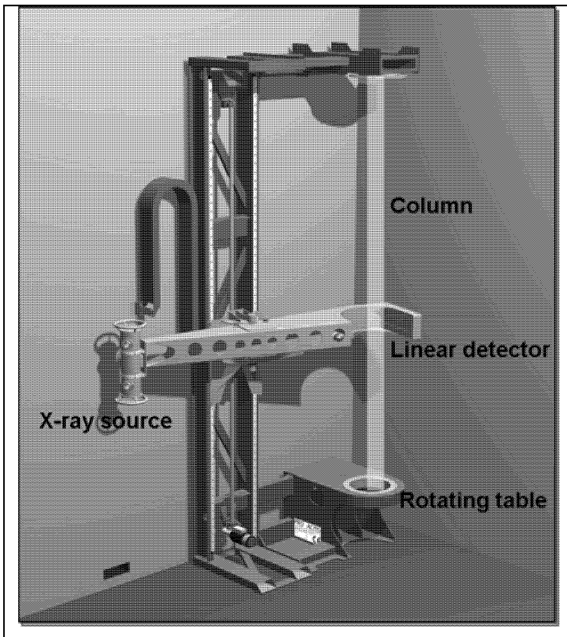

Fig. 2: X-ray tomograph.

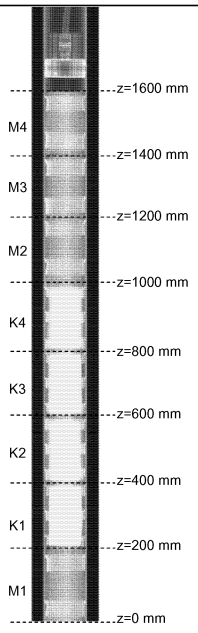

Fig. 3: Radiogram of the packed column.

3. RESULTS

3.1 Qualitative information

Figure 4 presents images of irrigated packing corresponding to cross sections situated at the same height in the column irrigated by the glycerine solution and by water, respectively. On both images, the liquid superficial velocity, UL, equals 10.2 $m^3/m^2/h$ and there is no gas flow. To obtain these images, we proceed as follows. First, dry packing images are recorded, reconstructed and thresholded, leading to the gray part of the images. Then, liquid distributions in the same cross sections are reconstructed and thresholded, leading to the black part of the images. Finally, gray and black are superimposed. Qualitatively, tomographic images show that, even at low liquid loads, catalytic baskets are completely filled by the more viscous solution, while they are partially filled by water in the same operating conditions.

3.2 Quantitative information

From liquid distribution images similar to those presented on Figure 4, liquid holdup values may be computed (Aferka et al., 2010, Viva et al., 2011b). Figure 5 plots the total liquid holdup in different cross sections located at different distances from the bottom of the packed column, as a function of liquid velocity, without gas for the glycerine solution and for water. Liquid is not uniformly distributed along the packed bed height. As expected, the total liquid holdup increases with liquid velocity. Axial profile of liquid holdup is significantly improved (flattened) if liquid viscosity is increased.

For the determination of static liquid hold up, the column is first totally filled up with liquid, and then drained for 24 hours. Figure 6 plots the static holdup obtained by X-ray tomography at various heights for the four Katapak-SP 11 elements with the glycerine solution and with water. One may observe that, at the bottom of each packing element, the static liquid hold up is higher, which corresponds to the liquid retained in baskets by the capillary forces (Aferka et al., 2007). As expected, axial profiles of static liquid holdup also show that static liquid holdup is increased if a more viscous liquid is used.

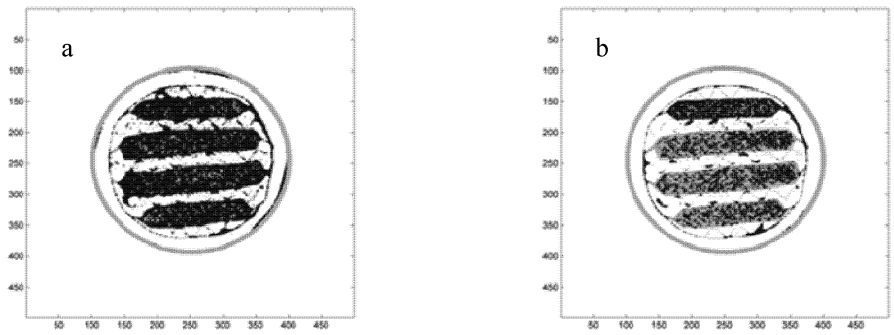

Fig. 4: Cross section of Katapak-SP11 situated at a height of 420 mm from the bottom irrigated with the glycerine solution (a) and with water (b), for UL = 10.2 $m^3/m^2/h$.

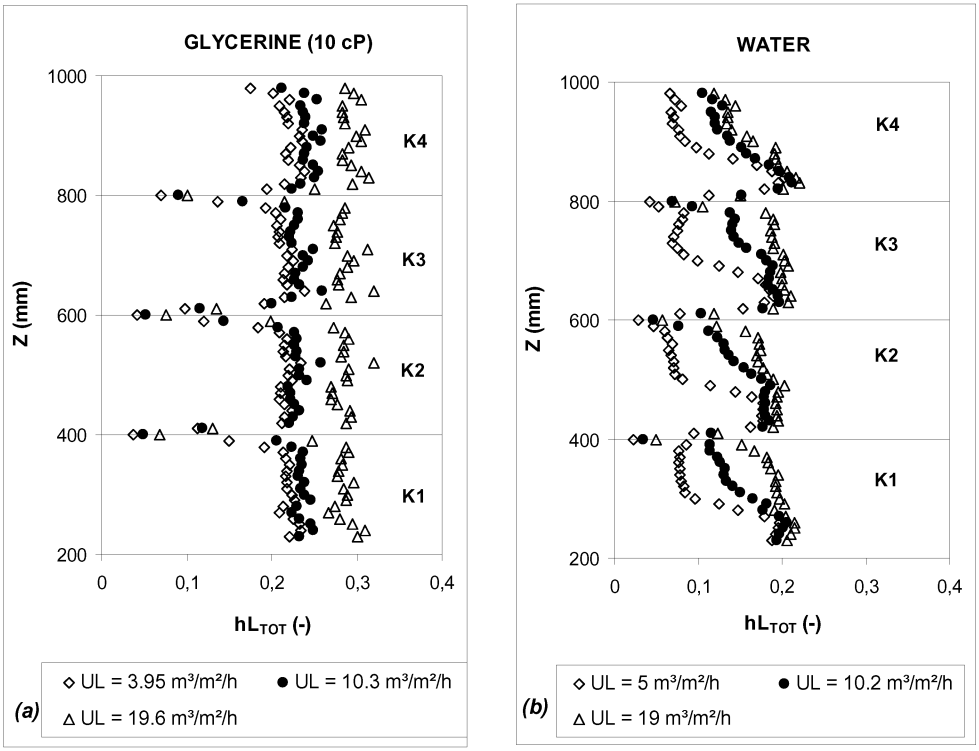

Fig. 5: Axial profile of total liquid holdup with glycerine (a) and water (b).

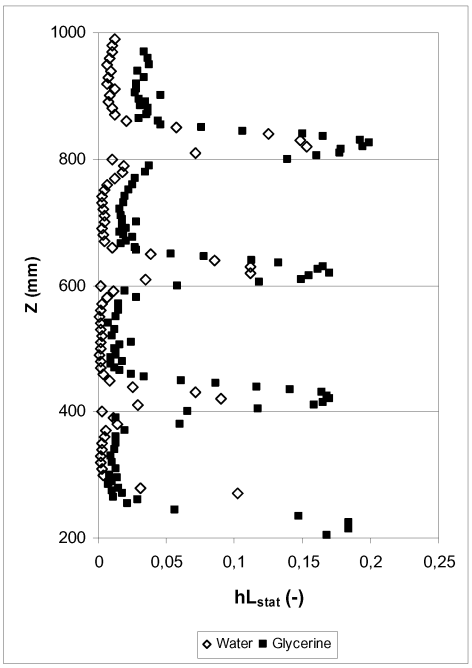

Fig. 6:Axial profile of static holdup with water and glycerine.

3.3 Comparison with other experimental data

By averaging holdup values obtained in different cross sections over the corresponding volume, one can determine global, bed scale, holdup values which can then be compared to those obtained using a more classical draining method. As shown in Figure 7, a very good agreement is observed between global liquid holdup values measured by tomography and values obtained with the draining technique both for water (Viva et al., 2011b) and for glycerine solution.

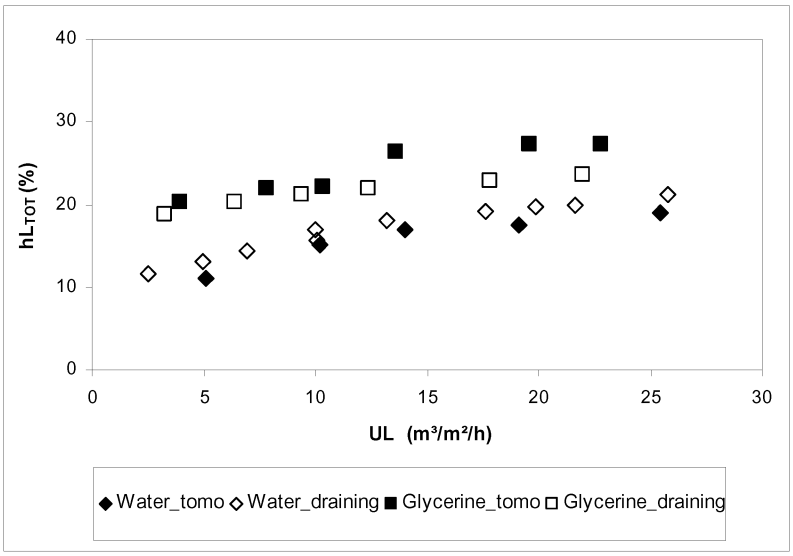

Fig. 7: Comparison between experimental values of liquid holdup obtained by X-ray tomography and by the draining technique.

4. MODELLING OF TOTAL LIQUID HOLDUP

Recently, from experiments with water as working liquid the authors have shown that the liquid holdup can be calculated by summing the holdup in the reaction zone with the holdup in the separation zone (Viva et al., 2011b).

The holdup contribution of the reaction zone can be estimated by means of correlations proposed in the literature for single-phase trickle beds. At the liquid load point, the catalytic baskets get completely filled with liquid (Aferka et al., 2010, Viva et al. 2011b), hence the liquid holdup inside the catalytic baskets is maximum and equal to 0.399 which is the catalytic baskets porosity, ε_{CB}. The maximum liquid superficial velocity inside the catalytic basket, u_{CB_max}, can be determined on the basis of a balance between gravity and the resistance for liquid flow through the bed of spheres (Moritz and Hasse, 1999):

$$\rho_L \cdot g = f \cdot \frac{1-\varepsilon_{CB}}{\varepsilon_{CB}^3} \cdot \rho_L \cdot u_{CB\_max}^2 \cdot \frac{1}{dp} \tag{1}$$

where f is the friction factor, expressed as:

$$f = \frac{160}{Re_{CB\_max}} + \frac{3.1}{Re_{CB\_max}^{0.1}} \tag{2}$$

and the effective Reynolds number in the bed of spheres is:

$$Re_{CB\_max} = \frac{u_{CB\_max} \cdot \rho_L \cdot dp}{(1-\varepsilon_{CB}) \cdot \mu_L} \tag{3}$$

Once u_{CB_max} is known, the total liquid flowrate at the loading point can be estimated by using the splitting factor of 0.9 suggested by Viva et al. (2011b):

$$Q_{L,LP} = u_{CB\_max} \cdot \frac{\psi_{CB\_S}}{0.9} \tag{4}$$

The liquid flowrate flowing on the corrugated sheets at and above the loading point can be determined by means of the following material balance:

$$Q_{L,OC} = u_{OC} \cdot A_{OC} = Q_L - Q_{L,CB\_max} = u_{TOT} \cdot A_C - u_{CB\_max} \cdot A_{CB} \tag{5}$$

Where $Q_{L,OC}$ and $Q_{L,CB}$ are the amount of liquid flowing outside (OC) and inside the catalytic baskets (CB), respectively, and Q_L is the total flowrate. In terms of superficial fractions occupied by the modular elements on the column cross section the above equation becomes:

$$u_{OC} \cdot \psi_{OC\_S} = u_{TOT} - u_{CB\_max} \cdot \psi_{CB\_S} \tag{6}$$

In the present study, use of equations (1)-(3) for the viscous liquid leads to a Q_{L,CB_max} value equal to 1.54 $m^3/m^2/h$. The corresponding liquid load point, $Q_{L,LP}$, equals 1.71 $m^3/m^2/h$. Consequently, the liquid loads used in our experiments are all above the liquid load point. Hence, equation 5 is used to estimate the liquid flowrate in the separation section.

The liquid holdup in the separation zone is estimated by a well established correlation for corrugated sheets. In fact, as reported by Olujic et al. (2007) on the basis of experiments on 450 mm diameter packings and by Aferka et al. (2011) on the basis of tomographic experiments on 100 mm diameter packings, the liquid holdup on the corrugated sheets of MellapakPlus 752.Y packings can be well predicted as a function of the liquid load by using the correlation developed by Suess and Spiegel (1992) for conventional Mellapak packings.

To make use of the Suess-Spiegel correlation in the present case, it has to be adapted to the Katapak-SP geometry according to the following relation:

$$hL_{OC} = c \cdot a_{MP}^{0.83} \cdot u_{OC}^{x} \cdot \left(\frac{\mu_L}{\mu_{water,20°C}} \right)^{0.25} \cdot \psi_{OC\_V} \tag{7}$$

where Ψ_{OC_V} is the volumetric fraction occupied by the separation zone. Empirical constants in Equation 7 depend on the liquid velocity. According to Suess and Spiegel (1992), c = 0.0169 and x = 0.37 for $u_{OC} < 40$ $m^3/m^2/h$ while c = 0.0075 and x = 0.59 for $u_{OC} \geq 40$ $m^3/m^2/h$.

Finally, in the present case the total liquid holdup can be calculated by summing the two contributions (OC and CB) as follows:

$$hL_{TOT} = hL_{OC} + \varepsilon_{CB} \cdot \psi_{CB\_V} \tag{8}$$

where the contribution of the catalytic baskets (CB) to the total liquid holdup is obtained by multiplying the porosity of the catalytic baskets by their volumetric fraction, Ψ_{CB_V}.
Figure 8 compares predicted values and experimental results, as a function of liquid load in the tested flowrate range. The agreement is very good and assesses the suitability of the procedure proposed by Viva et al. (2011b) from experiments carried out with water as working liquid, for the prediction of liquid holdup in a catalytic structured packing irrigated by a viscous solution.

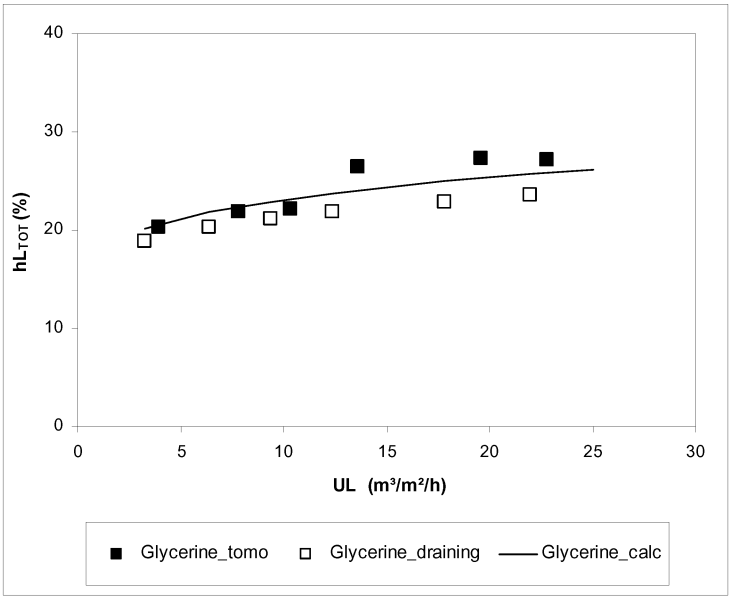

Fig. 8: Comparison between experimental values of liquid holdup, obtained by X-ray tomography and by the draining technique, and model prediction.

5. CONCLUSION

Local results obtained by X-ray tomography enable to get a better understanding of the fluid dynamic behavior in modular catalytic packings when used with more viscous liquids. The analysis of liquid distribution images and of axial profiles of liquid holdup allows quantifying the positive influence of an increase of liquid viscosity on packing wetting. This information is crucial to support on-going efforts in the development of fundamental hydrodynamic models.

6. NOTATION

a	specific geometric area (m^2/m^3)
A_C	area of the column section (m^2)
A_{CB}	area of the section occupied by CB (m^2)
A_{OC}	area of the section occupied by OC (m^2)
c	constant in Equation 7
dp	glass particle diameter (m)
f	friction factor (-)
g	gravitational acceleration (m/s^2)
hL	liquid holdup (-)
Q	volumetric flowrate (m^3/s)

Re	Reynolds number (-)
u	liquid velocity (m/s)
UL	specific liquid load ($m^3/m^2/h$)
x	constant in Equation 7
Z	column axial coordinate (m)

Greek letters

ρ	density (kg/m^3)
ε_{CB}	catalytic basket porosity (-)
ψ_{OC_S}	superficial fraction occupied by the OC (-)
ψ_{OC_V}	volumetric fraction occupied by the OC (-)
ψ_{CB_S}	superficial fraction occupied by the CB (-)
ψ_{CB_V}	volumetric fraction occupied by the CB (-)
μ	viscosity (Pa•s)

Subscripts

CB	catalytic basket section
L	liquid phase
LP	load point
max	maximum
MP	MellapakPlus sheets
OC	open channel section
TOT	total

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