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Influence of the viscosity on the liquid hold-up in trickle-bed reactors with structured packings

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Abstract

The influence of liquid viscosity on liquid hold-up in structured packings under co-current gas–liquid downward flow operation has been investigated for liquid viscosity from 1 to 20 cP. The liquid hold-up has been determined on a 400 mm internal diameter column by gamma tomographic cross-sectional measurements. An important influence of the viscosity on the liquid hold-up is observed. It is shown that, the widely used model supported by Bravo et al. [J.L. Bravo, J.A. Rocha, J.R. Fair, Hydrocarbon Process. January (1985) 91] assuming 1D fully established vertical liquid film flow does not agree with the experimental data. From experiments, the different assumptions used in the 1D model are discussed. On the basis of these results, a new correlation is proposed, which enables to calculate the hold-up from the viscosity, the liquid flow rate and the geometry of the packing. A comparison with data of literature is done.

Keywords: Structured packing; Trickle bed reactors; Liquid holdup; Gas treatment; Distillation

1. Introduction

Structured packing columns are widely used for industrial applications, mainly in distillation and gas treatment processes. For costs reduction and environmental protection purposes, the optimisation of reactor design is of great importance. Thus, the development of reliable models for pressure drop and mass transfer characteristics determination is necessary.

The liquid hold-up is an important hydrodynamic parameter for gas/liquid flow in structured packings columns. As discussed by Iliuta and Larachi [1], the liquid hold-up enables the determination of the pressure drop. It also allows for the calculation of the fluid effective velocity within the packing, which is further used for the determination of the liquid-side mass transfer parameters via the Higbie theory [2]. Most of experiments described in the literature have been carried out in laboratory conditions with water or hydrocarbons as liquid, besides counter-current flow mode is mostly used [3–5]. However, in the case of reactive absorption with non-equilibrated reaction, co-current flow is of high interest since it allows for higher gas and liquid flow rates than in counter-current mode. Recent measurements have been carried out under co-current down-flow mode [6,7]; however, the latter were done with air and water. As underlined by Murrieta et al. [8], there is a lack of experimental data with viscous fluids more representative of industrial solvents used in gas treatment processes.

The present paper aims at studying the influence of liquid viscosity on liquid hold-up in structured packings under co-current downward flow operation. The liquid viscosity has been varied from 1 to 20 cP. The experimental results are compared to those calculated with a 1D model and a new correlation is proposed.

2. Experimental

2.1. Experimental setup

Experiments have been carried out in a 2 m high, 400 mm internal diameter column as shown in Fig. 1 with air and aqueous solutions operating at ambient conditions in the

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Nomenclature

a	slope of the linear curves h_L versus Q_L (Eq. (5))
a_e	effective area ($m^2\ m^{-3}$)
a_g	geometric area ($m^2\ m^{-3}$)
B^P	channel base of packing P (mm)
e	film thickness (m)
h^P	crimp height of packing P (mm)
h_L^P	liquid hold-up in packing P
h_{Lw0}^P	static liquid hold-up in packing P for water
K^P	proportionality coefficient (Eq. (10))
L^P	distance between two contact points in Fig. 6 (m)
Q_L	liquid flow rate ($m^3\ m^{-2}\ h^{-1}$)
$R_{0_{P'/P}}$	ratio between h_{w0}^Y and h_{w0}^X
$R_V^{P'/P}$	ratio of the number of contact points per unit volume of packing P′ by that of packing P
S^P	channel side of packing P (mm)
$u_{eff,L}$	effective liquid velocity ($m\ s^{-1}$)
M250-P	Mellapak250-X or -Y
M500-Y	Mellapak250-Y
V_U^P	packing P unit volume

Greek letters

ε	packing void fraction
Γ	linear liquid flow, based on perimeter ($kg\ s^{-1}\ m^{-1}$)
ν_L	kinematic viscosity of liquid ($m^2\ s^{-1}$)
μ_L	dynamic viscosity of liquid (cP)
ρ_L	density of liquid ($kg\ m^{-3}$)
θ	channel flow angle from horizontal (°)

Subscripts

w	water

Superscripts

X	packing X
Y	packing Y

downward co-current mode. The fluids are distributed with a distributor equipped with 736 gas nozzles and 355 liquid nozzles, which permits a perfect distribution at inlet. This allows for studying the packing effect on distribution without any possible influence of the distributor.

Two packings were considered in this study: P-X and P-Y. These packings are almost identical to Sulzer Mellapak 500-X and 500-Y, respectively, but they are not perforated and have smooth walls. P-X and P-Y packings are made of stainless steel sheets with a channel flow angle from horizontal, respectively, of 60° and 45°. Elements are turned by 90° relative to each other inside the column. The geometric characteristics of the packing are given in Table 1.

In this work, water and three solutions of polyacrylamide with concentration up to 0.45 wt.% were used in order to vary the liquid viscosity, μ_L, from 1 to 20 cP. It was checked that the surface tension of these solutions were identical, equal to that of water. Values have been measured from 70 to 73 mN m^{-1}. Thus, the surface tension has no effect on the present results, the effect of viscosity only being looked at.

The liquid load or superficial liquid flow rate, Q_L, varies from 20 to 205 $m^3\ m^{-2}\ h^{-1}$. Gas superficial velocity, V_{SG}, is maintained constant at 0.1 m s^{-1}. It has been checked experimentally that the variation of the gas flow has no influence on the liquid hold-up distribution up to 0.5 m s^{-1}. This enables direct comparison with data obtained in counter-current mode for very low gas velocities published in the literature.

2.2. Tomographic measurements procedure

Measurements consist in mapping the liquid hold-up, or liquid volume fraction, across the packing. This is done via the use of a unique high-resolution gamma-ray system developed at IFP. This system is described in details by Boyer and Fanget [9]. From tomographic measurements, a local density map is obtained which is further converted into local liquid hold-up values. A similar system was used by Marchot et al. [10] and by Raynal et al. [6] for the study of liquid distribution in structured packings.

All measurements discussed in this paper were done at position (−1) (see Fig. 1) where it has been checked that the two-phase flow is fully established. Fig. 2 shows cross-sectional maps of liquid hold-up obtained with water flow rate of 103 $m^3\ m^{-2}\ h^{-1}$ for both P-X and P-Y packings. Liquid volume fraction ranges from 0 (blue) to 40% (red). One observes that the local liquid hold-up distribution is, in these cases, globally homogeneous, at large scales, with local non-homogeneities, at small scales. Similar observation is made for most of the liquid flow rates and viscosity conditions. To avoid wall effects where liquid accumulates and column's center errors where uncertainty is very large, the liquid hold-up used in this article results from local liquid volume fraction values averaged over 80% of the cross-section.

2.3. Experimental results and comparison with the 1D model

2.3.1. 1D model

The experimental results obtained in this study are compared to those calculated with a standard 1D model [2,5,11]. This model gives the liquid film thickness, e, assuming 1D vertical fully developed laminar flow via the following expression:

$$e = \left(\frac{3\Gamma\nu_L}{\rho_L g}\right)^{1/3} \quad (1)$$

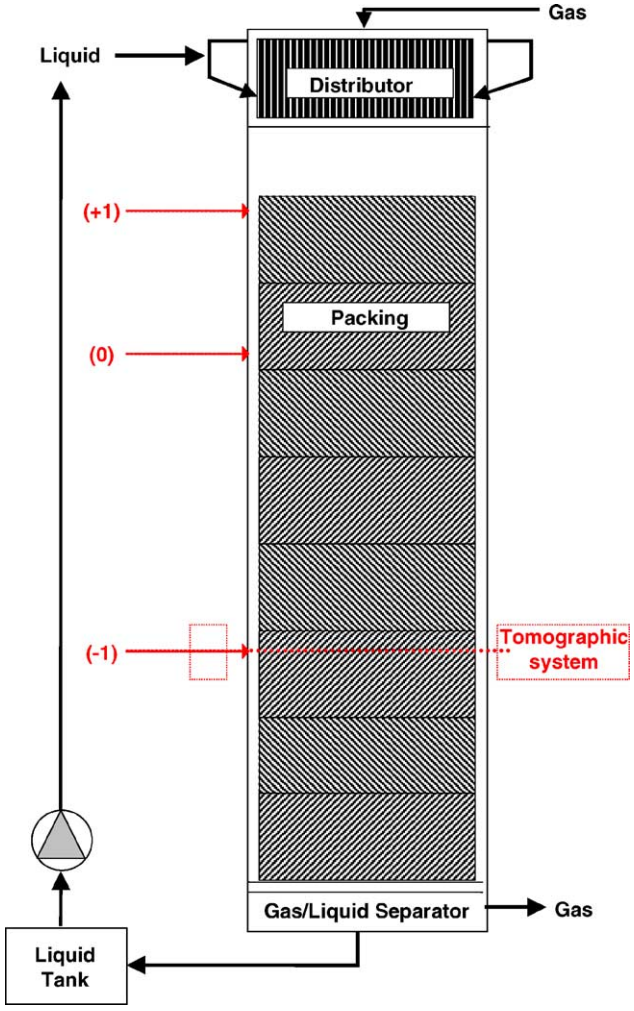


Fig. 1. Experimental setup.

where ν_L is the liquid kinematic viscosity, ρ_L, the liquid density, g being the acceleration of gravity and Γ, the liquid mass flow rate per unit length of wetted perimeter, expressed in kg s^{-1} m^{-1}. It is obtained from the superficial liquid flow rate expressed in m^3 m^{-2} s^{-1} by:

$$\Gamma = \rho_L Q_L \times \frac{B^P h^P}{4S^P} \quad (2)$$

with the geometric characteristics B^P, h^P and S^P of packing P being given in Table 1. From the liquid film thickness and the geometric area of the packing, a_g, one can further deduce the liquid hold-up within packing P, h_L^P, assuming full wetting of the packing:

$$h_L^P = e \times a_g = e \times \frac{4S^P}{B^P h^P} \quad (3)$$

It is also possible to deduce the effective velocity $u_{L,eff}$, the velocity at gas–liquid interface:

$$u_{L,eff} = \frac{3\Gamma}{2\rho_L} \times \left(\frac{\rho_L^2 g}{3\Gamma\mu_L}\right)^{1/3} \quad (4)$$

This effective velocity is commonly used for the determination of the liquid side mass transfer coefficient, k_L, via the Higbie theory (see, e.g. Bravo et. al. [2]). Since this 1D model is widely used, it is important to test its validity versus our present experiments.

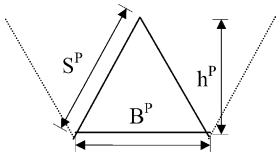
2.3.2. Experimental results

Measurements carried out for P-X and P-Y packings with liquid viscosity varying up to 20 cP are plotted versus the liquid flow rate in Fig. 3. One can first observe that, for a given viscosity, the variation of the liquid hold-up for P-X packing, h_L^X, with the liquid flow rate, Q_L, is linear. It can thus be written:

$$h_L^X = aQ_L + h_{L0}^X \quad (5)$$

where h_{L0}^X corresponds to the static liquid hold-up, that is for $Q_L = 0$. Second, from this figure it is seen that liquid hold-up increases significantly with liquid viscosity. Last, one notes that, for similar operating conditions, liquid hold-up values obtained for P-Y packing (open symbols) are higher than those obtained for P-X packing (closed symbols).

Table 1
Packing geometry information

		P-X[a]	P-Y[a]	M250-X	M250-Y	M500-Y
Crimp height, h^P (mm)		6.3	6.3			
Channel base, B^P (mm)[b]		12.1	14.0			
Channel side, S^P (mm)		8.50	9.82			
Void fraction, ε		0.93	0.93	0.98	0.975	0.975
Metal sheet thickness, t (mm)		0.3	0.3			
Geometric area, a_g (m^2 m^{-3})		446	445	250	250	500
Channel flow angle from horizontal, θ (°)		60	45	60	45	45
Element height, H (mm)		222	222			

[a] Present study.
[b] Horizontal projection.

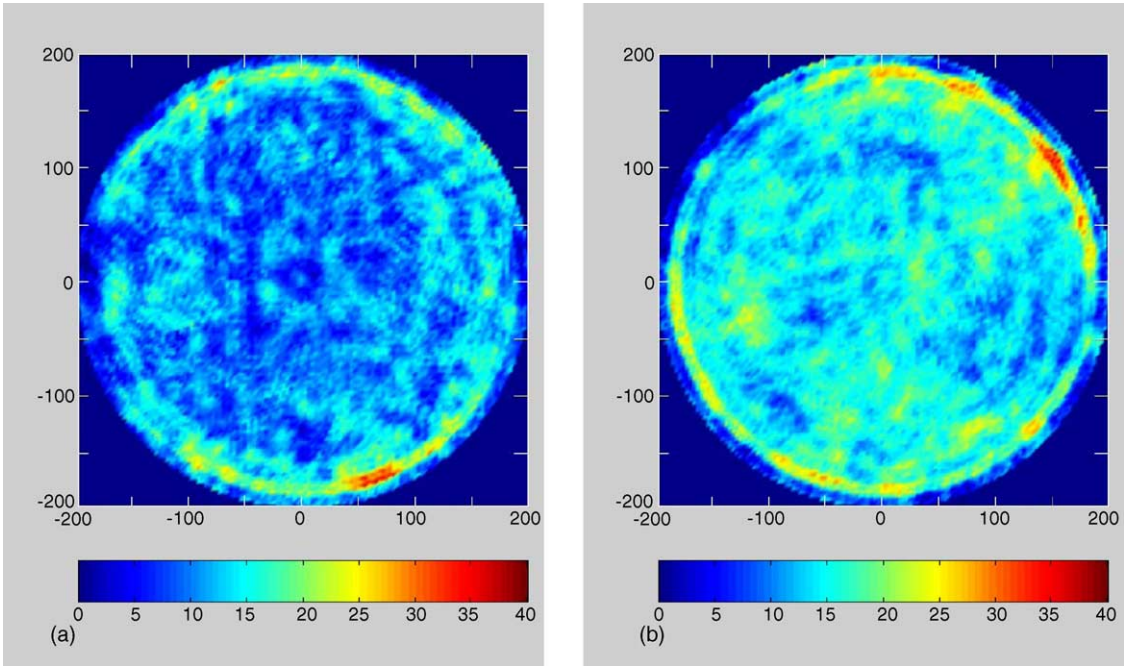


Fig. 2. Tomographic cross-sectional maps of the local liquid hold-up in, (a) P-X and (b) P-Y packings with μ_L = 5 cP, Q_L = 103 m^3 m^{-2} h^{-1}, V_{SG} = 0.1 m s^{-1}.

Coefficients of the best linear fits, as shown in Fig. 3, are reported in Table 2.

In Fig. 4, the calculated liquid hold-up obtained with the 1D model (Eqs. (1) and (3)) are plotted and compared to the experimental values for P-X and P-Y packings at 5 and 20 cP. One first observes an important overestimation by the 1D model for low and intermediate values of liquid hold-up. For experimental values from 5 to 20%, which corresponds to the common range of industrial applications, values obtained with the 1D model range from 10 to more than 30% which means that 1D model may give up to 100% overestimation. Second, since packing geometric characteristics, B, h, S used in Eqs. (2) and (3) are close between the two packings (see Table 1), 1D model gives almost similar results for the two packings which is clearly not the case as can be seen in Fig. 3.

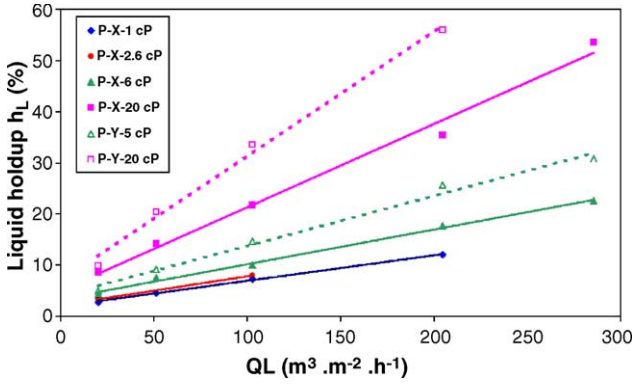


Fig. 3. Liquid hold-up experimental values vs. liquid flow rate for different values of viscosity for P-X (closed symbols) and P-Y packings (open symbols). The lines and the relative equations correspond to the best linear fit of experimental data (straight lines and dashed lines correspond, respectively, to P-X and P-Y packings).

2.3.3. New approach

As the 1D model does not predict properly the experimental data, an original approach for calculating the liquid hold-up in these packings is proposed.

First of all and since from Fig. 3 one observes that static hold-up values are not negligible, it seems interesting to carefully look at the dependence of this parameter on viscosity and on packing geometry. Fig. 5 shows the influence of viscosity on static hold-up for both packings. One actually observes that h_{L0}^{P} follows a power law dependence towards the liquid viscosity, expressed in the

Table 2
Parameters of Eqs. (5) and (10)

Viscosity (cP)		P-X[a]	P-Y[a]	M250-X	M250-Y	M500-Y
1	K	0.043	0.032	0.082	0.044	0.017
1	a	0.050	–	0.05	0.07	0.12
	h_{w0}	1.92	–	4.87	3.58	6.88
2.6	a	0.057	–	–	–	–
	h_{L0}	2.14	–	–	–	–
5	a	0.068	0.098	–	–	–
	h_{L0}	3.42	4.02	–	–	–
20	a	0.16	0.245	–	–	–
	h_{L0}	5.06	6.70	–	–	–

[a] Present study.

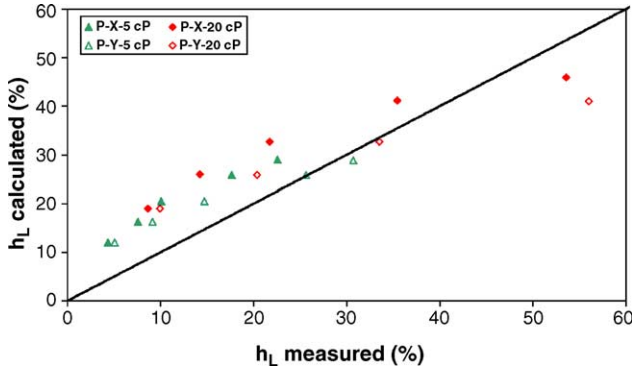


Fig. 4. Calculated liquid hold-up obtained with the 1D model vs. experimental liquid hold-up for P–X (closed symbols) and P-Y packings (open symbols).

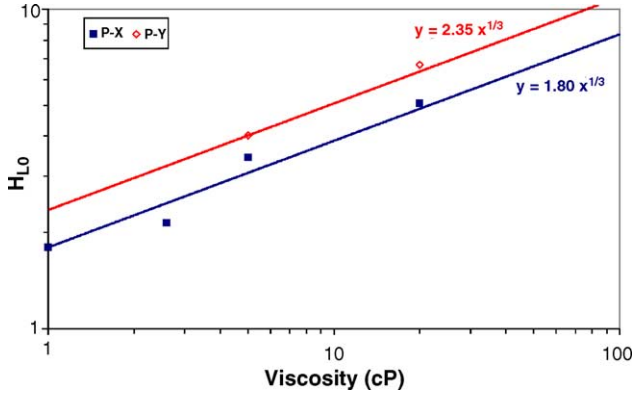


Fig. 5. Static liquid hold-up for P-X (closed symbols) and P-Y packings (open symbols) vs. viscosity. The lines and the relative equations correspond to the best power law fit of data.

following equation:

$$h_{L0}^{P} = h_{w0}^{P} \times \left(\frac{\mu_L}{\mu_w}\right)^{1/3} \tag{6}$$

where the subscript w corresponds to water. In the case of smooth walls, it is believed that static liquid is essentially hold-up at contact points between two adjacent corrugated metal sheets. Those contact points are illustrated in Fig. 6. Four contact points delimit each unit volume, V_U^P, the latter being the smallest periodic volume within packing P. It seems consequently interesting to compare static hold-up between the two P-X and P-Y packings and their respective number of static points per unit volume.

On the first hand, and from Fig. 6, one deduces that the ratio R_0, between h, h_{w0}^Y and h_{w0}^X is:

$$R_0 = \frac{h_{w0}^Y}{h_{w0}^X} = \frac{2.35}{1.80} = 1.31 \tag{7}$$

on the other hand, the ratio of the number of contact points per unit volume of each packings is given by

$$R_V^{Y/X} = \frac{1/V_U^Y}{1/V_U^X} = \frac{B^X L^X h^X}{B^Y L^Y h^Y} = \frac{B^{X^2}\tan(\theta^X)h^X}{B^{Y^2}\tan(\theta^Y)h^Y}$$
$$= \frac{12.1^2\tan(60°)6.3}{14.0^2\tan(45°)6.3} = 1.29 \tag{8}$$

One can consider that these two ratio given by Eqs. (7) and (8) are identical and that the difference in static hold-up values between the two packings directly results from their respective number of contact points per unit volume. One can thus conclude that in order to extrapolate the h_{Lw}^P value for a packing P to another packing P′, one just has to deduce from their respective geometry the ratio $R_V^{P'/P}$ and further deduces:

$$h_{w0}^{P'} = R_V^{P'/P} h_{w0}^P \tag{9}$$

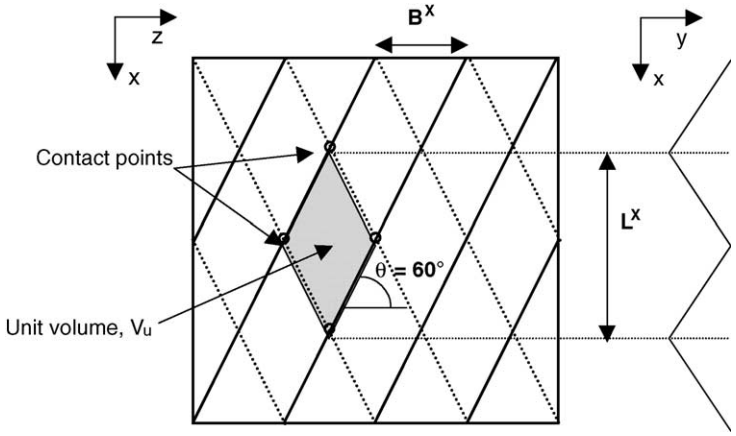


Fig. 6. Sketch of P-X packing geometry.

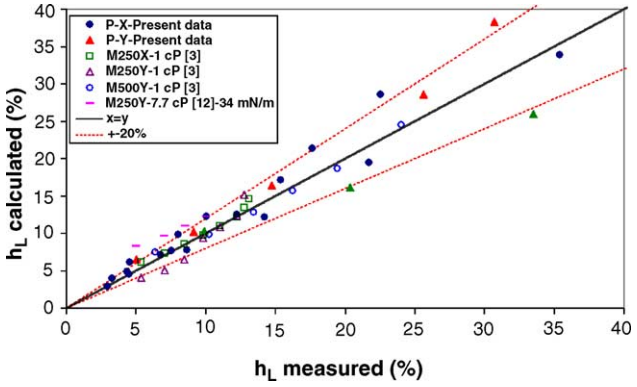


Fig. 7. Calculated liquid hold-up obtained with Eq. (10) vs. experimental liquid hold-up for P-X and P-Y packings and literature data.

Taking into account the preceding static hold-up evolution with viscosity, and assuming that the effect of viscosity on dynamic hold-up should follow the same 1/3 power law as in laminar flow (see Eqs. (1) and (3)), the total hold-up should be given by the following expression:

$$h_{\mathrm{L}}^{\mathrm{P}} = h_{\mathrm{w0}}^{\mathrm{P}} \times \left(\frac{\mu_{\mathrm{L}}}{\mu_{\mathrm{w}}}\right)^{1/3} + K^{\mathrm{P}} \times \frac{\Gamma}{\rho_{\mathrm{L}}} \times \left(\frac{\mu_{\mathrm{L}}}{\mu_{\mathrm{w}}}\right)^{1/3} \quad (10)$$

where K^{P} is a parameter only function of the geometry of the packing. Liquid hold-up values calculated with Eq. (10) are compared with the experimental values obtained for P-X packing and shown in Fig. 7. One observes a good agreement between the proposed correlation and experimental data, meaning that the effect of viscosity is well predicted. Good agreement is also obtained for P-Y packing, the average standard deviations are 11 and 15% for P-X and P-Y packings, respectively.

2.3.4. Comparison with literature data

Most of the experimental data available in the literature deals with counter-current mode and direct comparison with present data is not possible. However, when the superficial gas velocity is much less than the value at flooding, almost no gas influence is observed [5]. Results obtained at zero gas velocities, or at very low gas velocities, in the case of counter-current measurements can thus be compared to present data. This is done with the experimental data of Suess and Spiegel and Spiegel and Meier [3,4]. These data were obtained in a 1 m inner diameter column using Mellapak 250-X and 250-Y packings with air/water system. One of the only reference, which concerns co-current flow is the paper of Brunazzi and Paglianti [12]. The latter experiments were done in a 100 mm inner diameter column using Mellapak 250-Y packings with air/Genosorb 300 systems. The Genosorb 300 has a viscosity of 7.7 cP and a surface tension significantly lower than that of water, 34 mN m^{-1} compared to 73 mN m^{-1}, respectively. The correlation given by Eq. (10) has been adapted to these packings. In this case, the static liquid hold-up $h_{\mathrm{w0}}^{\mathrm{P}}$ and the proportionality coefficients K^{P} of M250-X, M250-Y and M500-Y packings were determined. All data are plotted in Fig. 7. It can be observed that the data of the reference [3] at 1 cP have a global good agreement with average standard deviation of 4.6, 15, 6.0% for Mellapak 250-X, 250-Y and 500-Y, respectively. Data of Brunazzi and Paglianti [12] are not well represented (the corresponding standard deviation is of 39%), but since they are obtained with a low surface tension liquid, the observed discrepancy is attributed to wetting phenomena. This point is further discussed in the following section.

Thus, it seems possible to determine the liquid hold-up for different viscosity values of any structured packing by only making measurements with water. Indeed, measurements with water will provide the water static hold-up, $h_{\mathrm{w0}}^{\mathrm{P}}$, and the proportionality coefficients, K^{P}, the effect on viscosity on both static and dynamic hold-up being the same and is well represented by a 1/3 power law the effect of liquid load being taken into account via the linear mass flow rate at a power of 1.

3. Discussion

The preceding section proposes a new approach giving importance to the static hold-up; however, it does not fully explain why such a discrepancy is observed between the 1D model and experimental data obtained on a real 3D geometry. The two main assumptions of the 1D model concern fully established flow and full wetting of the packing.

Concerning the assumption of fully established flow, some CFD calculations have been carried out by Raynal et al. [7] with a more realistic geometry than an infinite vertical wall as used in the 1D model. Those calculations have shown that, due to the complex geometry of structured packings, characterized by steep changes within short distances, the liquid film thickness is not constant at all along a corrugation. This evolution in shape along a corrugation results in liquid hold-up values higher than in 1D fully established vertical film flow. The complex nature of packing geometry does not explain the observed over-estimation obtained with the 1D model compared to experiments. Note that the study by Raynal et al. [7] was done with physical properties corresponding to air and water and more importantly it was restricted to very large liquid flow rates; further similar work would be useful for lower liquid flow rates corresponding to the present range of experiments and different values of liquid viscosities.

The second main assumption used in the 1D model is that full wetting of the packing is achieved which most probably is not the case. It is indeed well known from literature that the phase contact area or effective area, a_{e}, for gas–liquid mass transfer is less than the geometric area, a_{g} [13]. The ratio $a_{\mathrm{e}}/a_{\mathrm{g}}$ commonly increases from 0 to reach a plateau value around 1 as the liquid flow rate increases. This

phenomenon, observed for both random and structured packings [5], means that part of the packing is not wetted by the liquid and consequently that models assuming fully wetted packing cannot be directly applied to predict liquid hold-up. This observation is reinforced by pictures of the liquid hold-up obtained by tomography such as those shown in Fig. 2. In this figure, one clearly observes local spots with local high non-homogeneities. Since the spatial resolution of the tomograph is about 3 mm, this means that an average is made on small volumes about 3 mm in size. The tomograph resolution acts like a filter towards high frequency changes, which minimizes the small sizes inhomogeneities and allows only for ''large scale'' differences in local hold-up values to be visible. There is consequently a high probability to have local zones, which are actually not wetted. To further support this wettability point, it is seen from Fig. 7 that the only data for which a bad agreement is obtained with the present approach corresponds to data obtained with a different surface tension than water. One can thus deduces that there is a strong effect of surface tension and contact angle, which should deserve thorough studies in order to develop reliable models for liquid hold-up prediction. Last, and as discussed in the previous section, if all packing was fully wetted, there should be almost no effect of surface tension and the relative weight of static hold-up compared to dynamic hold-up would probably be not that large.

4. Conclusion

Determination of liquid hold-up has been made for two structured packings via tomography measurements with different aqueous solutions with liquid viscosities ranging from 1 to 20 cP. An important overestimation of the liquid hold-up by a standard 1D model is observed for low and intermediate values of liquid flow rate (up to about $100\ m^3\ m^{-2}\ h^{-1}$). It is believed that the most important assumption of the 1D model, that is fully wetted walls, is much too restrictive. This point is further supported by the observed differences in static hold-up values when comparing data of the literature. Further studies dealing with wettability effects should thus be carried out for such packings. This would complement the present approach, which enables the determination of liquid hold-up in structured packings with taking into account liquid viscosity effects only on static and dynamic liquid hold-up.

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