

EFFECT OF NON-HOMOGENEOUS LIQUID DISTRIBUTION ON EFFICIENCY OF STRUCTURED PACKING MATERIALS

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Abstract— Uneven liquid distribution (maldistribution) is a recurrent problem in the design and operation of packed towers, regardless of the type of packing. Using a simple but rigorous analysis it can be shown that the effect of liquid maldistribution on holdup is highly nonlinear and has cascading effects on liquid residence times and mass transfer rates. A statistical liquid film thickness description is used to characterize liquid distribution on corrugated structural packing. The distribution has a single adjustable parameter, the volume average variance of the distribution.

Keywords— Packing, Distillation, Holdup

I. INTRODUCTION

Separation columns fitted with ordered packing materials show lower pressure drop and smaller height of a transfer unit (HTU) than columns designed with gas-liquid contacting trays. Thus, structured packings are the preferred choice in vacuum distillation and residual absorption units as well as in revamping of existing units to add more capacity and improve energy efficiency. The main drawbacks of ordered packings are poor performance at low design superficial liquid flow rates, Q ($\text{m}^3/\text{m}^2\cdot\text{s}$) and the apparent uncertainty to predict efficiency in some separation systems. Both issues can be traced to the same cause, bad liquid distribution or maldistribution. If the liquid is perfectly distributed over the entire surface of packing, random or structured, the thickness of the liquid film should be uniform and only dependent of the inclination angle of the solid surface with respect to gravity (Shetty and Cerro, 1995). Any departure from a uniform distribution results in column sections where the film thickness is larger and other where the film thickness is smaller than the uniform distribution. Because of the nonlinear dependence of liquid velocity and flow rates on film thickness, the resulting non-uniform flow always has smaller liquid holdup. In turn, liquid holdup is the variable that leads to smaller average residence time of liquid inside the column and smaller mass transfer rates (Shetty and Cerro, 1997; Billet and Schultes, 1999; Wang *et al.*, 2005).

Given a vapor-liquid separation system, maldistribution can be affected by; a) wetting, b) inlet liquid distribution over the packing and re-distribution after packing sections, and c) specific packing flow characteristics. The most obvious and important parameter in liquid distribution are the wetting properties of the liquid/solid system.

A non-wetting liquid/solid system, i.e. a liquid that rests on the solid surface with a static contact angle close to or larger than 90° will be extremely difficult to distribute with any degree of uniformity. Unfortunately, wetting is a subject that has been misunderstood and improperly treated by many researchers, namely trying to characterize wetting using interfacial vapor-liquid tension. Wetting is associated to a solid/liquid property: static contact angles. The same liquid can have totally different contact angles on different solids. In partially wetting or non-wetting systems, i.e. static contact angles $\beta > 50^\circ$ the formation of liquid rivulets with surrounding dry sections drastically alters the amount of active packing area (Shi and Mersmann, 1985). During the last 30 years industry increased understanding of what was originally labeled as unpredictable behavior of certain separation systems, focusing on materials and surface treatments that deliver improved wetting properties.

Because of the importance and widely use of structured packings, industry spends large amount of resources on experiments to improve maldistribution by more efficient ways to introduce the liquid on top of the packing and on redistribution devices between packing sections (Sulzer Chemtech, 2006). Regardless of packing and distributor configurations, dispersion of a non-uniform film over a solid surface depends on the interaction of three main forces (Shetty and Cerro, 1995):

$$\left\{ \begin{array}{c} \text{hydrostatic} \\ \text{pressure} \end{array} \right\} + \left\{ \begin{array}{c} \text{capillary} \\ \text{pressure} \end{array} \right\} + \left\{ \begin{array}{c} \text{viscous} \\ \text{stress} \end{array} \right\} = 0 \quad (1)$$

Viscous stresses always act to prevent dispersion and capillary pressure always help to decrease non-uniform film thickness. However, the action of hydrostatic pressure depends on the side of the corrugation. On sections of the solid surface looking up, hydrostatic pressure favors leveling while on surfaces looking down, it works against it.

We use a simple flow model plus a statistical distribution of liquid film thickness to characterize maldistribution based on a single parameter, the variance of the distribution. The theoretical relationship of film thickness with packing and liquid parameters is used to derive a relationship of liquid holdup in the column. No general theory could be used to predict the amount of maldistribution because its dependence with wetting, flow distribution and packing flow characteristics. However, an experimental determination of dispersion variance can be used to predict holdup and subsequently evaluate packing design parameters using existing correlations.

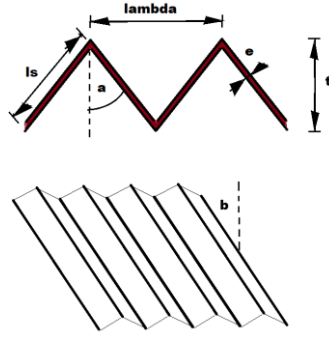


Figure 1: Definition of model packing geometric parameters

II. PACKING GEOMETRY AND LAMINAR FLOW DESCRIPTION

To highlight the fundamentals of this theory a simple model packing geometry will be used, a sheet with straight corrugations as shown in Fig. 1. Outside the angle of inclination of the corrugations with respect to the vertical, β , and the thickness of the material sheet, e , there are only four parameters needed to completely describe the corrugated sheets geometry: the amplitude of the corrugation, λ , the half-angle of the corrugation, α , the length of the side of one corrugation, l_s , and the thickness of the corrugated sheet, t .

Only two out of the four parameters are needed to completely characterize the packing geometry. There are two additional parameters, that can be obtained from simple mathematical identities: the specific area of packing, a_{spec} and the combined length of corrugated sheets on a given column cross section, L_{Flow} .

$$a_{spec} \left(\frac{m^2}{m^3} \right) = \frac{2}{t \sin \alpha}, \quad (2a)$$

$$L_{Flow}(m) \cong \frac{4D}{\sin \alpha} \sum_{i=1}^{\left(\frac{D}{2(t+e)} \right)^{-1}} \left[1 - \left(\frac{2(t+e)i}{D} \right)^2 \right]^{1/2}, \quad (2b)$$

where D is the column diameter and $a_{Cross} = \pi D^2/4$ is the column cross sectional area. The combined length of the corrugated sheets, L_{Flow} , is the length that we will measure by adding the lengths of single corrugated sheets on any horizontal cut of the column. The number of corrugated sheets in a tower of diameter D is given by the ratio $n_i = D/(t+e)$.

Neglecting capillary contributions, gravity film flow, even for flow over complex solid surfaces, shows a parabolic velocity profile (Shetty and Cerro, 1993). For a solid surface described by a function $z_o = \psi(x, y)$, the velocity component in the direction of gravity (x) can be written generically

$$v_x(x, y, z) = \frac{\rho g h^2(x, z) \cos \theta(x, z)}{2\mu} \left[1 - \frac{y^2}{h^2(x, z)} \right], \quad (3)$$

where y is the direction normal to the solid surface and $h(x, z)$ is the liquid film thickness at position on the corrugated sheet where the angle of inclination with respect to the vertical is θ and describes the location of the liquid/vapor interface. Eq. (3) describes a generalized Nusselt velocity profile (Shetty and Cerro, 1993). To compute the flow rate per unit packing length, Eq. (3) can

be integrated over a local position the cross section of the packing, i.e. for constants values of x and z .

$$q(x, z) \left(\frac{m^3}{ms} \right) = \left(\frac{\rho g \langle \cos \theta \rangle}{3\mu} \right) h^3(x, z) = \frac{Q a_{Cross}}{L_{Flow}} \quad (4)$$

In flow over corrugated structured packing the flow rate per unit packing length measured using the combined length of corrugated sheets on a column cross section, $q(x, z)$ is more appropriate to along describe the flow characteristics than the superficial flow rate, Q . We assume that the cosine of the angle of inclination of the corrugated sheets at any point, $\cos \theta = \text{constant}$. The average angle can be computed taking into account the angle of inclination of the corrugations wrt the vertical, β , and the half-angle of corrugations, α , and is given by $\cos \theta = \cos \alpha / \cos \beta$ (Shetty and Cerro, 1997). This relationship is only valid for $\cos \alpha \leq \cos \beta \leq 1$.

For a liquid film with homogeneous distribution, the constant liquid film thickness can be computed using Eq. (4):

$$\langle h \rangle = \left[\frac{3\mu q}{\rho g \cos \theta} \right]^{1/3} = \left[\frac{3\mu Q a_{Cross}}{\rho g \cos \theta L_{Flow}} \right]^{1/3} \quad (5)$$

For any typical column design at low superficial flow rates, $\langle h \rangle$ is a very small number on the order of a fraction of a millimeter. Such a small film thickness on a partially wetting liquid-solid system would very difficult to achieve by hydrodynamic spreading.

III. STATISTICAL DESCRIPTION OF LIQUID FILM DISTRIBUTION

In a typical liquid holdup direct experiment (Olujic *et al.*, 2006) containers are used to trap liquid flowing down at a section of a column and a two-dimensional map of flow rates and average velocities can be made. These are difficult experiments and many times must be made without gas flow. Other indirect experimental procedures with lower resolution include Gama scan and tomography (Alix and Raynal, 2008; Pless, 2017) and electrical impedance (Hewitt, 1978). In all cases, at any given column cross-section, there is a distribution of column zones with different levels of liquid in them. Thus, liquid distribution varies at any given cross section and may be different at different radial and longitudinal positions inside a column. Since the map of liquid holdup includes many points at any column horizontal cross section, we will assume that there is a continuous functional distribution along an independent variable $0 \leq X \leq L_{Flow}$ that is measured along the combined length of corrugated packing. The variable X should not be interpreted as an actual direction along the corrugated sheets but as a particular position where the liquid film thickness has a given value $h(z, X)$. Since L_{Flow} covers all the space points over the corrugated sheets at any value of z , then the mapping (z, X) covers all positions inside our column. Two similar exponential functions are chosen for the statistical distributions of the local liquid film thickness and the local flow rate per unit length of packing. The film thickness distribution has two parameters, one is the maximum liquid film thickness, $h_o(z)$, and the other is the variance of

the liquid film distribution, $m_h(z)$. The statistical distribution of the local flow rate per unit packing length has also two constants, the maximum flow rate per unit packing length, $q_o(z)$, and the variance of the distribution, $m_q(z)$, at any cross section z along the column. At any point in the column values of $h(X, z)$ and $q(X, z)$ are related through Eq. (4).

$$h(z, X) = h_o(z) \exp \left[-\frac{X^2}{2m_h^2(z)} \right] \quad (6a)$$

$$q(z, X) = q_o(z) \exp \left[-\frac{X^2}{2m_q^2(z)} \right] \quad (6b)$$

Integrating Eqs. (6a) and (6b) over the entire length of packing for a given cross section of the column we get the average liquid film thickness for a given cross section and the design superficial flow rate. It is important to point out that at any given cross section of the column, superficial flow rates, $Q = \text{constant}$.

$$\bar{h}(z) = \frac{1}{L_{Flow}} \int_0^{L_{Flow}} h(X, z) dX = \quad (7a)$$

$$= \sqrt{\frac{\pi}{2}} h_o(z) \left\{ \frac{m_h(z)}{L_{Flow}} \operatorname{erf} \left[\frac{L_{Flow}}{\sqrt{2}m_h(z)} \right] \right\}$$

$$Q = \frac{1}{a_{Cross}} \int_0^{L_{Flow}} q(z, X) dX = \quad (7b)$$

$$= \sqrt{\frac{\pi}{2}} \left(\frac{\rho g \cos \theta}{3\mu} \right) \frac{h_o^3(z) L_{Flow}}{a_{Cross}} \left\{ \frac{m_q(z)}{L_{Flow}} \operatorname{erf} \left[\frac{L_{Flow}}{\sqrt{2}m_h(z)} \right] \right\}$$

The statistical distributions between brackets in Eqs. 7 a and b are identical, except for the different values of the variance, $m_h(z)$ or $m_q(z)$, but these variances are related through a simple relationship derived from Eq. (6):

$$m_h(z) \cong \sqrt{3} m_q(z). \quad (8)$$

For large values of the variance, the expressions inside brackets go to one. In the case of the average film thickness, a variance going to infinity is identified with a homogeneous distribution and the maximum film thickness assumes the value $h_o \rightarrow \langle h \rangle$, given by Eq. 5. For small values of $m_h(z)$ the average liquid film thickness is smaller than the ideal value, $\bar{h}(z) < \langle h \rangle$, even if the largest film thickness at a column cross section, $h_o(z)$, is much larger than the ideal film thickness, $h_o(z) \gg \langle h \rangle$. Since the superficial flow rate is constant for any column cross section, it is possible to conclude that the product $h_o^3(z) m_q(z) = \text{constant}$. There is a corollary to derive from this relationship: Since the product $(h_o^3 m_q)$ is a constant for any cross section of the column we can define our local flow rate per unit packing length distribution using a single parameter, either $h_o(z)$ or $m_q(z)$. Since the maximum film thickness, $h_o(z)$, and the maximum flow rate per unit packing length, $q_o(z)$, are related through Eq. (4), and the product $h_o^3(z) m_q(z) = \text{constant}$, any one of the three parameters define the other two.

IV. LIQUID HOLDUP WITH MALDISTRIBUTION

To find an expression for the liquid holdup we integrate the expression for the liquid film thickness, Eq. (7a) over the entire length of the column:

$$H_L = \frac{L_{Flow}}{a_{Cross} L_{Packing}} \int_0^{L_{Packing}} \bar{h}(z) dz = \quad (9)$$

$$= \frac{L_{Flow}}{a_{Cross} L_{Packing}} \int_0^{L_{Packing}} h_o(z) \left\{ \sqrt{\frac{\pi}{2}} \frac{m_h(z)}{L_{Flow}} \operatorname{erf} \left[\frac{L_{Flow}}{\sqrt{2}m_h(z)} \right] \right\} dz$$

Except for the statistical distribution term, the definition of liquid holdup in Eq. (9) is identical to the definitions of holdup used by Spekuljack and Billet (1987) and Shetty and Cerro (1997). After substitution of the expression for $h_o(z)$ from Eq. (7) and writing the terms outside the integral using known dimensionless numbers:

$$H_L = 1.98 N_{Re}^{1/3} N_{Ga}^{1/3} N_{Geom} \times \quad (10)$$

$$\int_0^1 \frac{\left\{ \sqrt{\frac{\pi}{2}} \frac{m_h(z)}{L_{Flow}} \operatorname{erf} \left[\frac{L_{Flow}}{\sqrt{2}m_h(z)} \right] \right\}}{\left\{ \sqrt{\frac{\pi}{2}} \frac{m_q(z)}{L_{Flow}} \operatorname{erf} \left[\frac{L_{Flow}}{\sqrt{2}m_q(z)} \right] \right\}^{1/3}} \left(\frac{dz}{L_{Packing}} \right)$$

where $N_{Re} = 4\rho q/\mu$ is the Reynolds number, $N_{Ga} = \rho^2 g \lambda^3 / \mu^2$, is the Galileo number and $N_{Geom} = (L_{Flow} \lambda) / (a_{Cross} \cos \theta)$ is the packing Geometry number. The cross sectional area, a_{Cross} , contains the column diameter, D , while the flow length, L_{Flow} , the cosine of the angle of flow, $\cos \theta$, and the length of a single corrugation, λ , include all parameters for the definition of the packing geometry as shown in Eq. 2a and b.

On the basis of a single function, the variance of the liquid flow rate per unit length, $m_q(z)$, the integral term in Eq. (10) defines the degree of liquid maldistribution inside a column filled with corrugated structural packing since the variance of the liquid film thickness, $m_h(z)$, can be computed using Eq. (8). When liquid distribution approaches an ideal uniform distribution, the integral in Eq. (10) takes the value of 1 and the ideal liquid holdup correlation becomes just a weighted product of the three basic dimensionless numbers. For a more accurate description of flow distribution, experiments can be used to determine the variance of the flow rates at different positions, z , along the column. If suitable experimental data is available, then the effect of maldistribution can be introduced in the computation of liquid holdup by numerical integration of the integral term in Eq. (10). For a simpler approach we may assume that the variance is not a function of z and replace the variances by average values valid for the entire length of packing:

$$H_L = 1.98 N_{Re}^{1/3} N_{Ga}^{-1/3} N_{Geom} \frac{\left\{ \frac{\bar{m}_q}{L_{Flow}} \operatorname{erf} \left[\frac{L_{Flow}}{\sqrt{2}\bar{m}_q} \right] \right\}}{\left\{ \frac{\bar{m}_h(z)}{L_{Flow}} \operatorname{erf} \left[\frac{L_{Flow}}{\sqrt{2}\bar{m}_h} \right] \right\}^{1/3}} \quad (11)$$

V. LIQUID HOLDUP AND COLUMN PARAMETERS.

The basic assumption of our flow is a laminar falling film flowing under the effect of gravity, Eq. (3). Reynolds numbers should be in the laminar region and below the regime where the column is flooded by an excessive amount of liquid or by the shear stresses acting on the liquid/vapor interface due to the vapor flow. In mass transfer and HTU correlations for structured packing, liquid holdup is a pivoting parameter (Wang *et al.* 2005).

The original holdup correlation based on a parabolic velocity profile was developed by Shetty and Cerro (1997). The packing sheet surface was not corrugated but had a pattern of semispherical parallel “waves” inclined with respect to the vertical. The cosine of the actual angle of inclination was included in the numerical constant.

$$H_L = 6.096 N_{Re}^{1/3} N_{Ga}^{-1/3}. \quad (12)$$

The Reynolds and Galileo numbers in Eq. (12) are identical to the ones defined in Eq. (10). Unlike the relationship developed by Shetty and Cerro (1997), Eq. (13) includes a packing Geometry number that contains all the parameters needed to define and build the corrugated packing. The main difference between Eqs. (11) and (12) is the assumption by Shetty and Cerro (1997) that the packing was perfectly wet and the liquid film had a uniform distribution with no adjustable parameters. Eq. (11) has one adjustable parameter, the variance of the distribution of the liquid flow rate per unit length of packing, $\bar{m}_q$. It would be impossible to predict maldistribution in a general equation unless experimental information is introduced, and in every case it will be a function of wetting properties, type of structured packing and liquid distribution devices. This information could affect the value of the numerical constant as well as the variance of the distribution. To compare Eq. (11) with other holdup correlations, the values of geometric parameters should be the same. This will necessarily require data not always available from research publications. One of the most widely used correlations for liquid holdup was developed experimentally by Billet and Schultes (1999). Using our variables, the B&S correlation (Eq. (11) of Billet and Schultes, 1999):

$$H_L = \left(12 \frac{\mu Q a_{spec}^2}{\rho g} \right)^{1/3} = 4 N_{Re}^{1/3} N_{Ga}^{-1/3} N_{Geo}^{1/3}. \quad (13)$$

Again, the definition of Reynolds and Galileo numbers in Eq. (13) are identical to Eqs. (13) and (14). The Geometry number, $N_{Geo} = \lambda L_{Flow} / a_{Cross}$, is similar to the one in Eq. (13), except that it does not include the cosine of the inclination angle and it is raised to the 1/3 power.

A comprehensive holdup correlation was developed by Rocha *et al.* (1993, 1996) and later corrected for wetting by Gualito *et al.* (1997). This correlation known as the SRP-Model (Fair *et al.*, 2000) has also a similar form as Eqs. (11)-(13) but includes a correction factor for partial wetting. Again, using our definition of variables the corrected SRP-Model correlation contains the same Reynolds and Galileo numbers raised to the 1/3 power:

$$H_L = 0.91 \left(\frac{4 F_t (\sin \alpha)^{1/2}}{\left(\frac{\rho_L - \rho_G}{\rho_L} \right) \varepsilon^{1/2} \phi^{1/2}} \right)^{2/3} N_{Re}^{1/3} N_{Ga}^{-1/3} N_{Geo}^{1/3}. \quad (14)$$

The Reynolds, Galileo and Geometry numbers are identical to the numbers defined by Billet and Schultes (1999). Thus, Eqs. (13) and (14) are very similar except for the correction factor in brackets. Equation (14) is remarkable because it is one of the few instances where the static contact angle of the liquid/solid system is introduced as a variable to determine liquid holdup.

The final example of an empirical correlation is the Delft Model developed by Olujic (1997a; 1997b) and Olujic *et al.* (1999). In our choice of variables, the Delft Model is also surprisingly simple and very similar to all the equations shown before:

$$H_L = 2.39 \left(\frac{(\tan \alpha)^2}{\sin \alpha} \right)^{1/3} N_{Re}^{1/3} N_{Ga}^{-1/3} N_{Geo}^{1/3}. \quad (15)$$

Since Eqs. (11)-(15) all are functions of the Reynolds number to the (1/3) power, these correlations can be shown in a log-log graph as lines with identical slope. Liquid holdup computed using Eqs. (11)-(13) are shown next to each other for an ideal homogeneous liquid distribution in Fig. 2. Eq. (15) is almost identical to the Billet and Schultes (1999) model, except for the coefficient and the tangent term that is a number very close to one, and it is not shown.

When comparing these models, one must take into account two facts: First, all the liquid holdup correlations (Eqs. (11)-(15)) are based on packings with similar but with different flow distributors. Second, the theoretical correlation of Shetty and Cerro (1997) was developed for homogeneous liquid distributions while the experimental correlation of Billet and Schultes (1999) were likely done for large liquid loads approaching a homogeneous distribution. In Fig. 2, the line created with Eq. (11) was computed assuming a very large variance, that is approaching a homogeneous distribution.

If the liquid flow distribution is not homogeneous, actual liquid holdup is always smaller than the ideal computed from a homogeneous liquid film thickness distribution. This behavior is shown in Fig. 3 where maldistribution is computed using Eq. (13) using increasingly large values of the variance, $\bar{m}_q$.

For large values of the variance, $\bar{m}_q \geq 90$, liquid holdup values for a range of liquid load, $10 \leq Q \leq 200 (\text{m}^3/\text{m}^2\text{s})$ cannot be differentiated from the single line of a homogeneously distributed film. For smaller values of the variance liquid holdup takes much smaller values than the ideal distribution. These large deviations from ideality can be better appreciated looking at the values of the maximum film thickness at a cross section of the column.

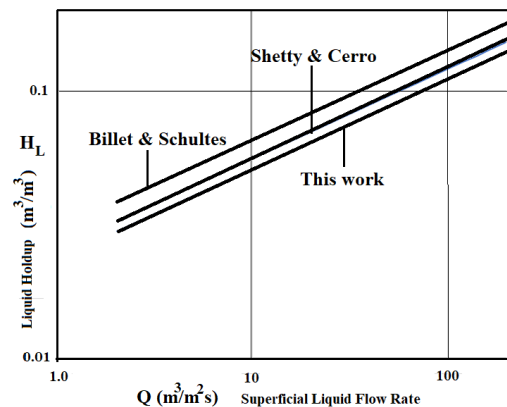


Figure 2. Comparison of liquid holdup correlations. In all cases lines show homogeneous liquid distribution.

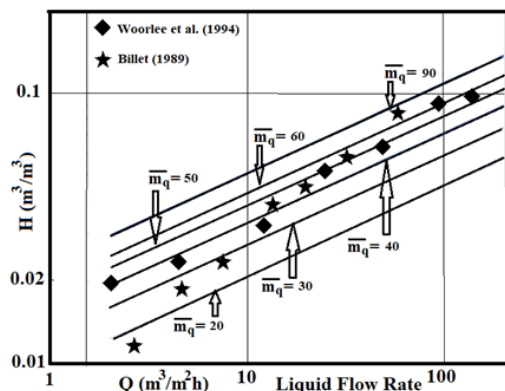


Figure 3: Liquid Holdup for systems with maldistribution. Lines correspond to different variances. Experimental data is from Woorlee *et al.* (1994), $\diamond$, and Billet (1989), $\star$.

VI. CONCLUSIONS.

Not surprisingly, theoretical and experimental correlations of liquid holdup in structured packing show identical behavior as a function of the Reynolds and Galileo numbers. Our theory is based on the sound assumption that on the average there is a laminar flow of a gravity driven falling film. However, as long as the dependence of liquid holdup with flow rate, that is of the Reynolds number to the $1/3$ power, the problem of maldistribution cannot be adequately addressed. Outside of our Eq. (11), the only other correlation addressing maldistribution is the SRP Model.

There is, nevertheless, a big difference between the SRP Model and Eq. (11). SRP deals mostly with wetting through the cosine of the static contact angle but does not address maldistribution due to spreading because this will imply that for such correlation the characteristic of packing and flow distributors apply only to the model used in the experiments. In other words, no two flow distributors are identical and have the same degree of initial maldistribution.

This is the reason why in our model (Eq. (11)) we have one adjustable parameter, the volume average of the distribution variance that must be determined experimentally for every combination of packing and flow distributors and for every vapor/liquid system. Indeed, to perform such experimental determination in a system that is under design is not practical. However, with time accumulation of experimental data will make this design practice more accurate and we are almost certain that such information is already available. More important is the fact that on an existing system, a simple tracer experiment could provide information about the degree of maldistribution and the practical feasibility of improving holdup and mass transfer.

Our equations also highlight the need for using the flow rate per unit packing length as the parameter of choice in structured packing design and to the need for a rational balance between packing specific transfer area and superficial flow rates. It should be obvious that the largest occurrence of maldistribution takes place at the smaller values of the flow rate per unit packing length

because a very small theoretical liquid film thickness, as computed by Eq. 5, has a very reduced spreading power.

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