

REVIEWS

Review of Mass-Transfer Correlations for Packed Columns*

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Mass-transfer correlations developed during the last several decades for packed column used for industrial fractionation, adsorption, etc. are reviewed. Theoretical basis and applicability of the reviewed correlations are discussed and compared concisely. Some dominant correlations and their parameters, as well as some considerations for further improvement, are also summarized and discussed.

Introduction

A packed column is a common, important piece of mass and heat transfer equipment extensively used in chemical process industries, typically in the domain of distillation, absorption, desorption, extraction, etc. Even though, for many cases, the tray column is less expensive and its performance prediction is more accurate, packed columns are increasingly sharing a significant part of the tower equipment market as they demonstrate advantages in their lower pressure drop, higher efficiency, and controlling sensitivity versus the tray column. Especially, the structured packing, having been developed and having gained a booming use in the process industries during last two or three decades, can provide more advanced hydraulic behavior, e.g., higher capacity (throughput) and efficiency, smaller scaling-up effect, and much lower pressure drop, which is precious for the cases of lower pressure operation, typically like vacuum distillation in the refinery crude units. Some heat-sensitive material treatment relies also on this extra low-pressure drop operation.

The extensive utilization of packings, especially structured packing, calls for reliable and accurate models for the prediction of the hydrodynamic and mass-transfer behavior of packed columns for design and analysis purposes. Since the 1930s, many academic organizations and packing suppliers have carried out numerous research studies for the random packing and, since the 1980s, for the structured packings.

A number of mass-transfer models, empirical or semi-theoretical for packed columns, have been published in the literature. These models are built on a variety of experimental bases and theoretical hypothesis, and they have different precisions, limitations, and ranges of suitability for application.

In this paper, we tried to give a comprehensive summary of the models we have found in the open

literatures for predicting the mass-transfer performance of a packed column. Main equations and related parameters of all the cited methods are given, and the theoretical/experimental basis and applicability of these models and methods, especially of some critical parameters (effective interfacial area, for example), are analyzed and compared, with the hope of providing a reference or some guidance for making further improvements for the predictions.

Basic Model for Mass Transfer

The mass-transfer performance of a packed column in distillation or absorption is often expressed by the packed bed height equivalent to a theoretical plate (HETP). Factors affecting HETP have usually been identified as type and size of packing, physical properties of test systems, operation condition, and dimension of columns. According to double-film theory,¹ the relationship between HETP and the height mass-transfer unit for the gas phase (HTU_G) and for the liquid phase (HTU_L) is given by

$$\text{HETP} = \frac{\ln \lambda}{\lambda - 1} (\text{HTU}_G + \lambda \text{HTU}_L) \quad (1)$$

where λ is the stripping factor, which is defined as the slope ratio of the slope of the equilibrium line to that of the operating line. Equation 1 in combination with the definitions of HTU_G and HTU_L, which are based on the concentration driving force across the gas and liquid films, respectively, yields

$$\text{HETP} = \frac{\ln \lambda}{\lambda - 1} \left(\frac{u_{Gs}}{k_G a_e} + \lambda \frac{u_{Ls}}{k_L a_e} \right) \quad (2)$$

where u_{Gs} and u_{Ls} are the gas- and liquid-phase superficial velocity, k_G and k_L are the gas- and liquid-phase mass-transfer coefficients, respectively, and a_e is the effective interfacial area provided by the packing to mass transfer. The accuracy of the model given by eq 2 obviously depends on the accuracy of the correlations used for predicting the basic parameters, i.e., the mass-transfer coefficients for the gas phase and liquid phase,

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respectively, and the effective interfacial area. The overview of the cited works shows that most previous research studies have been focused on estimating and modeling these parameters.

Gas/Liquid-Phase Mass-Transfer Coefficients

Random Packing. Sherwood and co-workers^{2–4} have systematically investigated the hydrodynamic and mass-transfer performance of random packing. On the basis of the data obtained from an oxygen/dioxide–air–water desorption column equipped with random packings, they proposed the earliest correlations, which became the fundamental basis of the later works. It should be pointed out that their correlation, limited to ring and saddle packings, is only applicable to aqueous solution systems or those that have a viscosity close to that of the aqueous systems. Van Krevelen and Hoftijzer^{5–7} introduced gas–liquid chemical reaction into absorption process to investigate the mass-transfer behavior of packings. In combination with the general kinetics theory of gas–liquid reactions, they gave gas/liquid mass-transfer coefficients in packed absorption columns. Morris and Jackson⁸ focused on HTU_G and HTU_L correlation with various factors and have proposed a set of equations to calculate HTU_G and HTU_L , but this model involves too many experimental parameters to be used conveniently.

Another more exhaustive investigative work about the performance of packed column was done by Shulman and co-workers, resulting in a series of papers.^{9–18} Their experimental investigations on the performance of random packed columns have been on aqueous and nonaqueous systems. They determined the gas-phase mass-transfer coefficient by vaporization processes, and then the correlation of k_G was used to determine the wetted areas of packings when irrigated with water and to calculate the effective interfacial areas from data available. These effective areas were then employed to evaluate k_L from available k_1a_e data, and a general correlation was developed for all kinds of random packings. In addition, they distinguished between static and dynamic holdup and between wetted and effective interfacial areas in packed columns as they dwelled on the influence of gas properties, temperature, pressure, solute concentration, and liquid holdup on the gas-phase mass-transfer coefficients.^{13,14,16}

The first and still widely used correlations for mass transfer were developed by Onda and co-workers,^{19–21} who assumed that the wetted surface on packing pieces is identical with effective gas–liquid interface. On the basis of the above assumption, dividing volumetric mass-transfer-coefficient data for gas absorption/desorption and vaporization available in the literature by effective gas–liquid interface, Onda and co-workers proposed the empirical equations of the gas- and liquid-side mass-transfer coefficients. Another frequently adopted model in the distillation process was the Monsanto model, which was presented by Cornell and co-workers^{22,23} Its development was based on a rather limited data set and utilized empirical parameters specific to each packing type and size. Bolles and Fair^{24,25} expanded the database of Cornell and co-workers and fine-tuned the model to new experimental results taken at larger scales of operation. Their adjustments mainly lie in the packing parameters for liquid- and gas-side mass transfer, which are also a function of liquid load and flooding percent, respectively. They

also revised graphically the correction factor for HTU_L at high gas rates. Bravo and Fair²⁶ used the model of Onda and co-workers plus the database of Bolles and Fair for the estimation of mass-transfer coefficients for distillation in random packings, but they provided an alternative correlation for effective interfacial areas. Since the effective interfacial area is used in the liquid-phase Reynolds number, the Bravo and Fair method will predict a different liquid-phase mass-transfer coefficient from that of the Onda and co-workers model. These four approaches to predicting mass transfer were considered to be capable of covering absorption, stripping, and distillation applications.

On the basis of the non-steady diffusion theory and the hydrodynamics of rivulets, Zech²⁷ and Zech and Mersmann^{28,29} have developed a method to calculate the liquid-phase mass-transfer coefficient for packed columns using penetration theory³⁵ and then the wetted specific area. This method in combination with a correlation for gas-phase mass transfer is applied to packed distillation columns. Mangers and Ponter³⁰ investigated the effect of viscosity on liquid-side mass transfer in packed columns. They found transition points at which the flow behavior changes from a partially wetted to a completely wetted packing and presented justification criterion. They also gave the liquid-side mass-transfer coefficient at a wide range of liquid viscosities (0.9–26 cP), below and above this transition point, respectively. Similarly to Zech and Mersmann, Shi and Mersmann³¹ deductively proposed a correlation for the calculation of the liquid-side mass-transfer coefficient from the dimension of rivulets. Mersmann and Deixler³² used the correlation of Shi and Mersmann plus another correlation for gas-phase mass-transfer coefficient and developed a method for the prediction of the mass-transfer performance of packed columns.

Billet and Schultes^{33,34} developed an advanced empirical/theoretical model to calculate the mass-transfer coefficients in gas/liquid countercurrent columns, filled with either random or structured packings, in the entire loading range. The model assumes that the effective free space of the packing is equivalent to a multiplicity of vertical flow channels, wherein the liquid trickles downward along the wall and the gas flows upward countercurrently. Both expressions for the gas and liquid mass-transfer coefficients were developed from the original formulation of Higbie³⁵ on the assumption that mass transfer in both phases follows the law of instantaneous diffusion. These equations require, besides the operating conditions, the theoretical liquid holdup value and two packing-specific constants accounting for shape, material, and size. Unfortunately, no such generalization was done by Billet and Schultes, making the model dependent on the availability of the parameters or experimental data. Wagner et al.³⁶ based on an earlier SRP (Separations Research Program at The University of Texas in Austin) study of liquid holdup and pressure drop in beds of random or structured packing,³⁷ developed a new model for the prediction and correlation of mass-transfer rates in distillation columns containing modern, high-efficiency random packings, which are of high-void-fraction, through-flow type. Building the model uses a large bank of experimental data from the laboratories of FRI (Fractionation Research, Inc. at Stillwater in Oklahoma) and SRP. As with the model of Billet and Schulte, this one is also founded on the penetration theory with the contact time

expressed through the effective liquid velocity and a characteristic length, which is empirically related with the packing characteristic constant and packed bed height.

In addition to the traditional calculation method, some new theories, such as RNN (artificial neural network), are introduced to predict the mass-transfer coefficients of randomly packed columns. Piche and co-workers^{38–40} employed RNN in combination with a reconciliation procedure to propose a set of artificial neural network correlations for the gas–liquid mass-transfer coefficients.

A summary of the various published correlations for the gas- and/or liquid-phase mass-transfer coefficients for random packings is given in Table 1.

Structured Packings. Since the appearance of the structured packing, research studies and industrial utilizations have been very much promoted, with its commercial attraction coming from its superior combination of high mass-transfer efficiency and low-pressure drop. Now experimental and theoretical attempts for deriving general methods for predicting mass transfer of structured packings using double-film theory and dimensionless groups have achieved a varying degree of success. The main published results of these attempts include the works of Bravo and co-workers,^{41–43} Nawrocki et al.,⁴⁴ Henriques de Brito et al.,⁴⁵ Weiland et al.,⁴⁶ Billet and Schultes,³⁴ Hanley et al.,^{47,48} Brunazzi and Paglianti,⁴⁹ Xu et al.,⁵⁰ etc. These models were checked against many sources of pilot plant data. On the other hand, reliable empirical or semiempirical correlations of efficiency specific to a packing supplier's products can be found in their product bulletins (e.g., Saint-Gobain Norpro Corporation⁵¹).

The channels formed by the structured packing elements are presumed to pass liquid and vapor in counterflow contacting. Since the geometry is well-defined and the packing surface area is known precisely, it has been found possible to model the mass-transfer process along lines of wetted wall theory. This idea was adopted by many investigators in the study of structured packed columns.

The first overall investigation about structured packing (gauze) was conducted by Bravo and co-workers,^{41,42} and the result was called SRP (I) model. In this model, the gas-side mass-transfer coefficient is based on earlier investigations of wetted-wall columns⁵² while the liquid-side is based on the penetration theory,³⁵ where the exposure time is taken as the residence time for liquid flow between corrugation changes. Nawrocki et al.⁴⁴ adopted similar relationships to those of Bravo et al.⁴¹ and, in addition, took liquid distribution into consideration to give more reliable liquid mass-transfer coefficients.

Bravo et al.⁵³ and Rocha et al.^{54,55} have improved and updated the SRP (I) model and proposed the SRP (II) model, in which they made modifications on characteristic dimension of packing, effective gas and liquid velocity, and contact time. An impressive characteristic of this model is that mass transfer and hydrodynamic performance of structured packed columns are related by an important parameter—liquid holdup. Billet and Schultes³⁴ and Hanley et al.^{47,48} also mentioned this point and did the same. Brunazzi and Paglianti⁴⁹ closely examined liquid film experimentally and identified a new relationship, incorporating the influence of packing height on liquid-side mass-transfer coefficients. By the

combination of the new relationship with the Rocha et al.⁵⁵ correlation for gas-side mass transfer, but with different effective gas and liquid velocities, the overall mass-transfer performance of a structured packed column can be estimated.

The Billet and Schultes models^{33,34} have been fully discussed in the preceding section. It should be noted, however, that this model is dependent on the liquid holdup calculation. Xu et al.⁵⁰ have used the model of Billet and Schultes and a new correlation for effective interfacial area to predict mass transfer in a packed column containing structured packing. Hanley et al.^{47,48} proposed a unified model for countercurrent vapor/liquid packed columns on the basis of the critical phenomena theory, specifically percolation theory on a conductor/insulator lattice.

Another often used model, the Delft model, was first developed by Olujic,⁵⁶ which has been revised for further refinement^{57,58} and perfection.^{59–61} This model, developed for corrugated packings, considers the gas flow as a continuous zigzag flow through a triangular-shaped corrugation channel and takes into account explicitly all observed macro-geometry-related effects in a fundamentally sound way. It should be noted that, in this model, a common phenomenon, transitional flow, was taken into account, and by analogy with heat transfer, gas-phase mass-transfer coefficients were divided and calculated in laminar and turbulent regimes, respectively. Fair et al.⁶² reported the results of evaluation and comparison of the SRP (II) model and the Delft model based on a comprehensive total reflux distillation study of sheet metal structured packings. On the basis of these results, some possible model-improvement considerations were also suggested. Modifications of the Delft model made by Olujic⁵⁸ were mainly focused on the effective liquid flow angle, calculated according to the theoretically based expression by Spekuljak and Billet,⁶³ and purportedly resulted in only a minor increase of the reliability and accuracy versus the original one.

Shetty and Cerro^{64,65} first proposed the nonempirical model for the design parameters of distillation columns with structured packings. They investigated the geometry of a periodically complex surface as well as the mechanism of liquid flow on the surface and, thus, gave a more fundamental approach to predict the liquid mass-transfer coefficient. According to the authors, the corrugation base length of a packing channel was taken as the characteristic dimension, thus accounting explicitly for the liquid film thickness. The main advantage of this approach is using the knowledge developed on the study of flow patterns and velocity profiles for thin viscous films over complex surfaces, instead of approximate and simplified methods, to calculate contact time and liquid-phase mass-transfer rates. Therefore, it is considered by Fair et al.⁶² that the SRP model and the Delft model could be possibly improved using the Shetty and Cerro correlation for the liquid-phase mass-transfer coefficient.

In addition, Whaley et al.⁶⁶ and Pollock and Eldridge⁶⁷ modeled HETP of structured packings using a neural network. The neural network model was proved to yield a very accurate prediction of experimentally determined HETP values, and it is probably more accurate than a traditional semiempirical model.

Table 1. Correlations for the gas- and/or liquid-side mass-transfer coefficients for random packings

author	correlations		comments ^a		
			PT	CW	OC
Sherwood and Holloway ⁴	$k_L = m \left(\frac{D_L}{a_e} \right) \left(\frac{\rho_L u_L}{\mu_L} \right)^{1-n} Sc_L^{0.5}$	(3)			✓
Van Krevelen and Hoftijzer ^{5,7}	$k_G = 0.2 \frac{D_G}{d_c} \left(\frac{\rho_L u_L}{a_p \mu_L} \right)^{0.8} Sc_G^{1/3}$	(4)		✓	
	$k_L = 0.015 \frac{D_L}{[\mu_L^2 / (\rho_L^2 g)]^{1/3}} \left(\frac{\rho_L u_L}{a_e \mu_L} \right)^{2/3} Sc_L^{1/3}$	(5)		✓	
Shulman and de Grouff ⁹	$k_G = 0.0137 (\rho_G u_G)^{0.65} Sc_G^{-2/3}$	(6)			✓
	k_L is computed by eq 3				✓
Morris and Jackson ⁸	$HTU_G = \frac{99.7 C_0 \epsilon^{0.75} u_G^{0.25}}{R_G C_G a_p} \left(1 - \frac{u_L \epsilon}{u_G} \right)^{-0.75}$	(7)			✓
	$HTU_L = \frac{C_0'}{32.4 R_L C_L} L_w^{0.3}$	(8)			✓
Shulman et al. ¹¹	$k_G = 1.195 u_G \left[\frac{d_p \rho_G u_G}{\mu_G (1 - \epsilon)} \right]^{-0.36} Sc_G^{-2/3}$	(9)		✓	
	$k_L = 25.1 \frac{D_L}{d_p} \left(\frac{d_p \rho_L u_L}{\mu_L} \right)^{0.45} Sc_L^{0.5}$	(10)		✓	
Cornell et al. ^{22,23} , Bolles and Fair ^{24, 25}	$HTU_G = \frac{\psi (d/3.66)^m (Z_p/3.05)^{1/3}}{(3600 \rho_L u_L f_{\mu} f_{\rho} f_o)^n} Sc_G^{0.5}$	(11)			✓
	$HTU_L = \phi C_f \left(\frac{Z_p}{3.05} \right)^{0.15} Sc_L^{0.5}$	(12)			✓
Onda et al. ²⁰ Bravo and Fair ²⁶	$k_G = c \left(\frac{D_G}{a_p d_p^2} \right) \left(\frac{\rho_G u_G}{a_p \mu_G} \right)^{0.7} Sc_G^{1/3}$	(13)		✓	
	$k_L = \frac{0.0051}{(a_p d_p)^{-0.4}} \left(\frac{\mu_L g}{\rho_L} \right)^{1/3} \left(\frac{\rho_L u_L}{a_e \mu_L} \right)^{2/3} Sc_L^{-0.5}$	(14)		✓	
Zech and Mersmann ²⁹	$k_G = K_G \frac{D_G}{d_p} \frac{\epsilon + 0.12}{\epsilon(1 - \epsilon)^{-1}} \left[\frac{\rho_G u_G d_p}{(1 - \epsilon) \mu_G} \right]^{2/3} Sc_G^{1/3}$	(15)		✓	
	$k_L = K_L \sqrt{\frac{6 D_L}{\pi d_{pe}}} \left(\frac{\rho_L g d_{pe}^2}{\sigma_L} \right)^{-0.15} \left(\frac{u_L g d_{pe}}{3} \right)^{1/6}$	(16)	✓		
Mangers and Ponter ³⁰	partial wetting: $k_L = \frac{(3.22 \times 10^3) Sc_L^{0.5} D_L \left(\frac{\rho_L u_L}{\mu_L} \right)^\alpha \left(\frac{\rho_L^2 g h_{pe}^3}{\mu_L^2} \right)^{0.27}}{a_e (1 - \cos \theta)}$	(17a)	✓		
	where $\alpha = 0.49[(1 - \cos \theta)^{0.6} (\rho_L \sigma_L^3 / \mu_L^4 g)^{0.2}]^{0.108}$				
	complete wetting: $k_L = \frac{2.03 D_L Sc_L^{0.5} \left(\frac{\rho_L u_L}{\mu_L} \right)^{1.44} \left(\frac{\rho_L^2 g h_{pe}^3}{\mu_L^2} \right)^{-0.183}}{a_e}$	(17b)	✓		
Shi and Mersmann ³¹ , Mersmann and Deixler ³²	$k_G = K_G \frac{D_G}{d_{pe}} \left(\frac{\rho_G u_G d_{pe}}{\mu_G} \right)^{2/3} Sc_G^{1/3}$	(18)		✓	
	$k_L = 0.86 \sqrt{\frac{6 D_L}{\pi d_{pe}}} \sqrt{\left\{ \frac{u_L^{1.2} g^{1.3} \sigma_L^{0.3} \epsilon^{1.2} (1 - 0.93 \cos \theta)^2}{\nu_L^{1.4} \rho_L^{0.3} a_p^{2.4}} \right\}}$	(19)	✓		
Billet and Schultes ^{33,34}	$k_G = C_G \frac{a_p^{0.5} D_G}{\sqrt{d_h (\epsilon - h_L)}} \left(\frac{\rho_G u_G}{a_p \mu_G} \right)^{3/4} Sc_G^{1/3}$	(20)	✓		
	$k_L = C_L \left(\frac{\rho_L g}{\mu_L} \right)^{1/6} \left(\frac{D_L}{d_h} \right)^{0.5} \left(\frac{u_L}{a_p} \right)^{1/3}$	(21)	✓		
Wagner et al. ³⁶	$k_G = \frac{Z_t^{-0.5}}{C_{pk}} \sqrt{\frac{4 \Phi_G D_G u_G}{\pi (\epsilon - h_t)}}$	(22)	✓		
	$k_L = \frac{Z_t^{-0.5}}{C_{pk}} \sqrt{\frac{4 \Phi_L D_L u_L}{\pi h_t}}$	(23)	✓		
Piche et al. ^{38,39}	$\frac{k_L a_e}{a_p^2 D_L} = f \left[\left(\frac{\rho_L u_L}{a_p \mu_L} \right), \left(\frac{a_p u_L^2}{g} \right), \left(\frac{\rho_L g}{a_p^2 \sigma_L} \right), Sc_L, \left(\frac{Z_t}{Z_t + d_c} \right), \left(\frac{u_G}{u_L} \sqrt{\frac{\rho_G}{\rho_L}} \right) \right]$	(24)			✓
	$\frac{k_G a_e}{a_p^2 D_G} = f \left[\left(\frac{\rho_L u_L}{a_p \mu_L} \right), \left(\frac{a_p u_L^2}{g} \right), \left(\frac{\rho_L g}{a_p^2 \sigma_L} \right), Sc_G, \left(\frac{Z_t}{Z_t + d_c} \right), \left(\frac{u_G}{u_L} \sqrt{\frac{\rho_G}{\rho_L}} \right) \right]$	(25)			✓

^a PT stands for penetration theory, CW stands for correlation based on wetted-wall columns as in eq 74, and OC stands for other correlations.

A summary of the various published correlations for the gas- and/or liquid-phase mass-transfer coefficients for structured packings is given in Table 2.

Other than the above empirical, semiempirical, and theoretical methods, many researchers^{68–75} also have proposed all kinds of shortcut method to estimate the mass-transfer performance of packed column. A sum-up of these methods is shown in Table 3.

Effective Interfacial Area

Although there are a great deal of publications on the effective interfacial area, the problem has not been solved satisfactorily. As far as packed columns are concerned, there exist several definitions of the interfacial area,³¹ but wetted surface area is particularly important because the knowledge of it is essential for basic considerations of two-phase flow in irrigated packing and it also can be taken as a reference surface area when considering experimental mass-transfer results. In addition, wetted surface area is closely linked to effective interfacial area under particular flow conditions because only the wetted area can be effective for mass transfer. But in principle, the difference between the wetted surface and the effective interfacial area lies in that the wetted surface area incorporates liquid surface area in dead zones and the effective interfacial area includes surfaces of drops and jets. Despite these deviations, both the wetted surface area and the effective interfacial area are often applied.

The effective interfacial area can be measured by physical methods, such as electroresistivity, light transmission, and reflection techniques, but mostly it is determined by mass-transfer measurements in the presence of a fast chemical reaction, controlling the absorption process. Danckwerts and Sharma⁷⁶ were among the first to conduct chemical absorption experiments to obtain the effective interfacial area, the results of which were expressed by charts. Danckwerts⁷⁷ also proposed chemical absorption theory and reviewed⁷⁸ two-phase systems employed to measure the interfacial areas of interphase mass transfer. Many workers adopted this method to investigate the effective interfacial areas in packed columns,^{79–85} bubble columns,^{86–88} and agitated reactors.⁸⁹

Random Packings. Many researchers have made a lot of investigations on the effective interfacial area of random packings, which resulted in a number of correlations^{11,20,21,26,28,34,48,83,84,90–93} in the literature. Shulman et al.¹¹ are early investigators to experimentally study the effective interfacial area for absorption, desorption with water or very dilute aqueous solutions, and vaporization. Their experimental results were expressed by an extensive series of graphs for several types of packings and were well-represented by empirical expressions.⁹⁴ It is found from the results obtained by Shulman et al. that the effective interfacial area shows a greater dependence on gas rates and that increased kinetic energy of the gas tends to increase the effective interfacial area. Therefore, the interfacial area available for mass transfer is likely a function of the liquid and gas rates, although many researchers held that only the liquid load will have an impact on the effective interfacial area.

The correlation of Onda and co-workers^{20,21} correlation is recognized as the first powerful, most-accepted predictive equation for randomly packed tower design with regard to the effective interfacial area. The equa-

tion was obtained from detailed investigation on the effects of hydrodynamics and liquid physical properties on the wetted surface area of random packings by physical method and chemical pigmentation. This correlation often overpredicts the effective interfacial area because its prediction includes the area of dead zone, wherein standing liquid remains, wetting the surface area of the packing without any surface renewal, which makes it ineffective for mass transfer. Moreover, as the wettability of the packing increases, for materials of greater critical surface tension, the overprediction grows. It should be noted that, based on the data of Raschig Ring, Berl Saddle, spheres and rods packing, and ceramic Palling Ring, this model is not applicable to new-type random packings.

Puranik and Vogelpohl⁹⁰ also proposed a correlation for estimating effective interfacial area in evaporation and absorption, with and without chemical reaction. Puranik and Vogelpohl also introduced the concepts of static and dynamic areas and measured the maximum area of the packing available for mass transfer, which should be the sum of zones where liquid moves about with surface renewal, i.e., the dynamic area and zones where the liquid is stagnant, termed the static area. Another merit of this correlation is that it is easily capable of identifying operation conditions under which the static component can be regarded as nonexistent and the difference between the wetted surface and the effective interfacial area can be eliminated. In addition, the authors took the maximum area to be identical with the wetted area, valid for evaporation (EV) and absorption with chemical reaction (CR), while the dynamic area was the area available for physical absorption (PA).

Following Kolev,⁹¹ Zech and Mersmann^{28,29} developed a new method to split up the volumetric mass-transfer coefficient to give the liquid-side mass-transfer coefficient and the ratio of effective interfacial area to geometry area, which can be determined theoretically and experimentally, respectively. Theoretical analysis and experimental results²⁷ indicate that the ratio is related with four dimensionless groups i.e., Reynolds, Galileo, Froude, and Weber numbers, and a correlation of this ratio for the distillation process was obtained based on experimental results of mass-transfer coefficients for liquid-phase controlled systems.

Bravo and Fair²⁶ developed an improved correlation of mass-transfer rates for a commercial-scale packed distillation column, in which the Onda et al. correlations were adopted for the mass-transfer coefficients but the method for the effective interfacial area was a new one. The effective interfacial area correlation was obtained by correlating the extensive experimental data bank included in paper by Bolles and Fair,²⁴ which involved a wide range of packings, column size, and systems. The striking characteristic of this data set lies in that the most part of the data are not for absorption but distillation. Compared with previous correlations, this one takes into account the influence of the vapor rate on the effective interfacial area of random packings. In addition, the probable liquid maldistribution due to column height and diameter also was taken into consideration.

Linek et al.⁹² and Costa et al.⁹³ also made investigations on effective interfacial area in absorption or desorption columns and obtained their own correlations. Billet and Schultes^{33,34} proposed a three-equation

Table 2. Correlations for the Gas- and/or Liquid-Side Mass-Transfer Coefficients for Structured Packings

author	correlations		comments ^a		
			PT	CW	OC
Bravo et al. ⁴¹	$k_G = 0.0338 \frac{D_G}{d_{eq}} \left[\frac{\rho_G d_{eq} (u_{Le} + u_{Ge})}{\mu_G} \right]^{0.8} Sc_G^{0.33}$	(26)		✓	
	$k_L = 2 \sqrt{\frac{D_L}{\pi s} \left(\frac{9 \Gamma^2 g}{8 \rho_L \mu_L} \right)^{1/3}}$	(27)	✓		
	where $u_{Ge} = \frac{u_G}{\epsilon \sin \alpha}$, $u_{Le} = \left(\frac{9 \Gamma^2 g}{8 \rho_L \mu_L} \right)^{1/3}$				
Nawrocki et al. ⁴⁴	$k_G = 0.0338 \frac{D_G}{d_{eq}} \left(\frac{\rho_G d_{eq} u_{Ge}}{\mu_G} \right)^{0.8} Sc_G^{0.33}$	(28)		✓	
	$k_L = 2 \sqrt{\frac{D_L}{\pi s} \frac{3 V_L}{2 w \delta_{dyn}}}$	(29)	✓		
Henriques et al. ⁴⁵	$k_L = 2 \sqrt{\frac{D_L}{C_1 \pi l \cos \alpha} u_L^{(1-C_2)/2}}$	(30)	✓		
Hanley et al. ⁴⁸	$k_G = \frac{D_G}{d_{eq}} \left[\frac{d_{eq} K_V (\rho_L / \rho_G)^{0.5}}{\epsilon \mu_G (1 - f f_c)^p} \right]^{0.8} Sc_G^{1/3}$	(31)		✓	
	$k_L = \frac{D_L}{d_{eq}} \left(\frac{d_{eq} u_L \rho_L}{\epsilon \mu_L} \right)^{0.5} Sc_L^{0.5}$	(32)		✓	
Rocha et al. ⁵⁵	$k_G = 0.054 \frac{D_G}{s} \left[\frac{\rho_G s (u_{Le} + u_{Ge})}{\mu_G} \right]^{0.8} Sc_G^{0.33}$	(33)		✓	
	$k_L = 2 \sqrt{\frac{0.9 D_L u_{Le}}{\pi s}}$	(34)	✓		
	where $u_{Ge} = \frac{u_G}{\epsilon (1 - h_L) \sin \alpha}$, $u_{Le} = \frac{u_L}{\epsilon h_L \sin \alpha}$				
Brunazzi and Paglianti ⁴⁹	$k_G = 0.054 \frac{D_G}{d_h} \left[\frac{\rho_G d_h (u_{Le} + u_{Ge})}{\mu_G} \right]^{0.8} Sc_G^{0.33}$	(35)		✓	
	$k_L = A \frac{D_L}{d} \left(\frac{\mu_L g}{\sigma^3 \rho_L} \right)^{B_1} \left(\frac{\delta \sin \alpha_L u_{Le} d}{Z_t D_L} \right)^{B_2}$	(36)		✓	
	where $u_{Ge} = \frac{u_G}{(\epsilon - h_L) \sin \alpha}$, $u_{Le} = \frac{u_L}{h_L \sin \alpha_L}$				
Olujic et al. ^{57,58 b}	$k_G = \sqrt{\left(\frac{Sh_{G,lam} D_G}{d_{hG}} \right)^2 + \left(\frac{Sh_{G,turb} D_G}{d_{hG}} \right)^2}$	(37a)		✓	
	where $Sh_{G,lam} = 0.664 Sc_G^{1/3} \sqrt{Re_{Grv} \frac{d_{hG}}{l_{G,pe}}}$ $Sh_{G,turb} = \frac{(Re_{Grv} Sc_G \xi_{GL} \varphi / 8) [1 + (d_{hG} / l_{G,pe})^{2/3}]}{1 + 12.7 \sqrt{\xi_{GL} \varphi / 8} (Sc_G^{2/3} - 1)}$	(37b)			
	$k_L = 2 \sqrt{\frac{D_L u_{Le}}{0.9 \pi d_{hG}}}$	(38)	✓		
Shetty and Cerro ⁶⁴	k_G is computed by eq 33			✓	
	$k_L = \frac{0.4185 D_L}{b} \sqrt{\frac{\sin \alpha}{l_{ratio}}} \left(\frac{4 \rho_L g}{\mu_L} \right)^{1/3} \left(\frac{\rho_L^2 g b^3}{\mu_L^2} \right)^{1/6} Sc_L^{1/2}$	(39)	✓		
Xu et al. ⁵⁰	$k_G = \sqrt{\frac{4 D_G u_G}{\pi l \left[\epsilon - \left(\frac{4 F_t}{s} \right)^{2/3} \left(\frac{3 \mu_L u_L}{\rho_L \epsilon g_{eff} \sin \alpha} \right)^{1/3} \right]}}$	(40)	✓		
	$k_L = \sqrt{\frac{4 D_L u_L}{\pi l \left(\frac{4 F_t}{s} \right)^{2/3} \left(\frac{3 \mu_L u_L}{\rho_L \epsilon g_{eff} \sin \alpha} \right)^{1/3}}}$	(41)	✓		

^a PT stands for penetration theory, CW stands for correlation based on wetted-wall columns as in eq 74, and OC stands for other correlations. ^b Adjustment is using α_L developed by Spekuljak and Billet⁶³ for calculations of δ and u_{Le} instead of α .

semiempirical procedure to predict mass transfer of either structured packed or randomly packed columns,

which includes a correlation for calculating the effective interfacial area. The correlation was obtained and

Table 3. Sum-Up of Shortcut Methods for Mass Transfer of Packed Columns

authors	shortcut correlations
Whitt ⁶⁸	$HTU_G = 36/\sqrt{\frac{d_N \mu_L}{\rho_L \mu_L}}$ (42)
Murch ⁶⁹	$HETP = c(\rho_G \mu_G)^a d_c^b Z_t^{1/3} \left(\frac{\alpha \mu_L}{\rho_L}\right)$ (43)
Ellis ⁷⁰	$HETP = \left[18d_N + 0.305m_{eq} \left(\frac{\rho_G \mu_G}{\rho_L \mu_L} - 1\right)\right] \left(\frac{Z_p}{3.05}\right)^{0.5}$ (44)
Harrison and France ⁷¹	$HETP = 100/a_p$ (45)
Kister ⁷²	$HETP = 100/a_p + 0.102$ (46)
Strigle ⁷³	atmospheric distillation (0.4–4 atm): $HETP = \exp(m - 0.187 \ln \sigma_L + 0.213 \ln \mu_L)$ restrictions: $\sigma_L = 4\text{--}36 \text{ mN/m}$, $\mu_L = 0.08\text{--}0.83 \text{ cP}$ pressure distillation (>4 atm): $HETP = n - 0.213 \ln M_L$ restrictions: $M_L = 22\text{--}72 \text{ g/mol}$ (47a) (47b)
Lockett ⁷⁴	$HETP = \frac{1.54g^{0.5}(\rho_L - \rho_g)^{0.5}\mu^{-0.06}}{a_p[1 + 0.78 \exp(0.00058a_p)(\rho_g/\rho_L)^{0.25}]^2}$ (48)
Carillo et al. ⁷⁵	sheet packing: $HETP = \frac{1.54g^{0.5}(\rho_L - \rho_g)^{0.5}\mu^{-0.06}}{a_p[1 + 0.78 \exp(0.00058a_p)(\rho_g/\rho_L)^{0.25}]^2}$ (49a)
	gauze packing: $HETP = \frac{P\sqrt{\rho_L}F_V^{0.42}}{(2712 + 82.0P)[1 + 1.505(\rho_g/\rho_L)^{0.25}]^2}$ (49b)

validated using a large data bank available in the literature, including different systems and different types and sizes of packing. This correlation, the general form of which comes from a dimensional analysis of the influencing parameters, reflects well the results of the experiments if the surface tension increases from top to bottom (i.e., positive system). In the case of rectification of negative systems, which are characterized by the fact that the surface tension along the column decreases from the top to the bottom, the Marangoni effect must be taken into account, because this effect will cause a reduction in the effective interfacial area. Therefore, the authors subsequently introduced a correction factor related to the Marangoni number to account for negative systems. Recently, the authors gave an overall summary of the calculation method of Billet and Shultes.⁹⁵

Careful analysis of the above existing relationships indicates that there is some conflicting evidence concerning the influence of some factors, e.g., viscosity and gas flow, on the interfacial area. However, we must bear in mind that the range of viscosities on which these relationships are based was very narrow. In view of this point, Nakajima et al.⁸⁵ measured and correlated interfacial area by the chemical absorption of CO₂ on Raschig rings within a wider range of viscosity (0.95–4.5 cP). Their experimental results confirm the conclusion that the system undergoes a transition during the formation of the effective interfacial area, which was also suggested by Mangers and Ponter.³⁰ Incorporating the effect of gas flow, the authors developed a correlation to predict the effective interfacial area in packed columns.

A summary of the various published correlations for the effective interfacial area for random packings is given in Table 4, with the exponents or expressions of the principal parameters given in Table 6.

Structured Packings. The effective interfacial areas for structured packing are more easily investigated on a theoretical basis because of the packing's orderly arranged structure. Among models and correlations^{31,43,48,50,55–58,80,96–99} available in the literature for

structured packings, the representative model was proposed by Shi and Mersmann,³¹ on the basis of the assumption that the liquid flows in the form of uniformly distributed rivulets on a flat plate inclined to the horizontal at an angle, which partly wet the surface of the packing. A rivulet is a narrow film whose width is not determined by the equipment, but it spreads freely. The dimensions of the rivulet (width and thickness) were calculated by theoretical methods to obtain the wetted surface area and were measured by experiments to validate predictive results, respectively. Considering the difference between wetted area and effective interfacial area, the authors also introduced a correction factor in the model, which is a function of packing element diameter. Because of its mechanistic feature, some subsequent research studies^{44,55,100} were performed on the basis of this model.

Bravo et al.⁴¹ reasonably considered the total surface to be wetted, which was attributed, in part, to the capillary action within the finely woven wire gauze. It is clear that sheet metal structured packings do not have the benefit of capillary action and may not be completely wetted. For structured packings, Fair and Bravo^{43,97} introduced a discount factor, which is a function of flooding percentages, on the geometric specific area for partial wetting.

Henriques et al.⁹⁸ studied three kinds of Mellapak packing in their experimental work and proposed a general correlation to predict interfacial mass-transfer area. The authors found that the packing employed can provide an effective interfacial area higher than the geometric area of the packing defined by the packing surface up to a factor of 2, depending on the liquid and gas load and the geometric area of packing. They considered that the mechanism responsible for this remains to be studied in detail, but probably is the liquid flow instabilities between the sheets. Brunazzi et al.⁹⁹ reported an equation that correlates the interfacial area only with the liquid holdup evaluated experimentally, geometric parameters, and physical properties.

Table 4. Correlations for the Effective Interfacial Area for Random Packings

authors	correlations
Shulman et. al. ¹¹	$a_e = m(808\rho_G^{0.5}u_G)^n L^p \frac{BL^{0.57}\mu_L^r}{\rho_L^{0.84}(AL^q - 1)} \left(\frac{\sigma_L}{0.073}\right)^{s-0.262 \log L} \quad (50)$ <p>where A, B, m, n, p, q, r, and s can be obtained from ref 94, p 205, Table 6.4</p>
Onda et al. ^{20,21}	$\frac{a_e}{a_p} = 1 - \exp\left[-1.45\left(\frac{\sigma_c}{\sigma_L}\right)^{0.75} \text{Re}_L^{0.1} Fr_L^{-0.05} We_L^{0.2}\right] \quad (51)$ <p>where $Re_L = \rho_L u_L / (\mu_p \mu_L)$, $Fr_L = u_L^2 a_p / g$, $We_L = \tilde{n}_L \mu_L / (\sigma_L a_p)$ limitations: $0.04 < Re_L < 500$, $1.2 \times 10^{-8} < We_L < 0.272$; $5 \times 10^{-9} < Fr_L < 1.8 \times 10^{-2}$, $0.3 < \sigma_c / \sigma_L < 2$</p>
Puranik and Vogelpohl ⁹⁰	$\frac{a_w}{a_p} = \left(\frac{a_e}{a_p}\right)_{EV} = \left(\frac{a_e}{a_p}\right)_{CR} = 1.045 \left(\frac{\rho_L u_L}{\mu_L a_p}\right)^{0.041} \left(\frac{\rho_L u_L^2}{\sigma_L a_p}\right)^{0.133} \left(\frac{\sigma_c}{\sigma_L}\right)^{0.182} \quad (52a)$
	$\frac{a_{stat}}{a_p} = 0.229 - 0.091 \ln\left(\frac{\rho_L g}{a_p^2 \sigma_L}\right) \quad (52b)$
	$\frac{a_{dyn}}{a_p} = \left(\frac{a_e}{a_p}\right)_{PA} = \frac{a_w}{a_p} - \frac{a_{stat}}{a_p} \quad (52c)$
Kolev ⁹¹	$\frac{a_e}{a_p} = 0.583 \left(\frac{\rho_L g}{a_p^2 \sigma_L}\right)^{0.49} \left(\frac{u_L^2 a_p}{g}\right)^{0.196} (a_p d_p)^{0.42} \quad (53)$
Zech and Mersmann ^{28,29}	$a_e = K_L \left(\frac{\rho_L u_L}{\mu_L d_{pe}}\right)^{0.5} \left(\frac{\rho_L g d_{pe}^2}{\sigma_L}\right)^{0.45} \quad (54)$
Bravo and Fair ²⁶	$\frac{a_e}{a_p} = 0.498 \left(\frac{u_L \mu_L}{\sigma_L} \frac{6\rho_G u_G}{a_p \mu_G}\right)^{0.392} \sigma_L^{0.5} Z_t^{0.4} \quad (55)$
Rizzuti et al. ⁸³	$a_e = 39\nu_L^{0.7} u_L^{0.326} \quad (56)$
Rizzuti et al. ⁸⁴	$a_e = (2.944 \times 10^4) \nu_L^{0.28} u_L^{0.313} \quad (\nu < 1.54 \times 10^{-6}) \quad (57a)$
	$a_e = 0.165 \nu_L^{-0.625} u_L^{0.313} \quad (\nu > 1.54 \times 10^{-6}) \quad (57b)$
Linek et al. ⁹²	$\frac{a_e}{a_p} = 0.0277 \left(\frac{1 + \cos \theta}{2}\right)^{3.477} (a_p d_p)^{1.585} \left(\frac{\rho_L u_L}{a_p \mu_L}\right)^{0.641-0.407(1+\cos \theta)/2} \quad (58)$
Costa Novella et al. ⁹³	$\frac{a_e}{a_p} = a \left(\frac{\rho_L u_L}{a_p \mu_L}\right)^{0.13} \left(\frac{u_L^2 a_p}{\rho_L \sigma_L}\right)^{0.38} \left(\frac{\sigma_c / \sigma_L}{(a_p Z_t)^b}\right)^{0.18} \quad (59)$
Billet and Schultes ^{33,34,95}	$\left(\frac{a_e}{a_p}\right)_{neu,pos} = 1.5(a_p d_h)^{-0.5} \left(\frac{\rho_L u_L d_h}{\mu_L}\right)^{-0.2} \left(\frac{u_L^2 \rho_L d_h}{\sigma_L}\right)^{0.75} \left(\frac{u_L^2}{g d_h}\right)^{-0.45} \quad (60a)$
	$\left(\frac{a_e}{a_p}\right)_{neg} = \left(\frac{a_e}{a_p}\right)_{neu,pos} (1 - 2.4 \times 10^{-4} Ma_L ^{0.5}) \quad (60b)$
Nakajima et al. ⁸⁵	$\frac{a_e}{a_p} = 0.6119 \left(\frac{u_L \mu_L}{\sigma_L}\right)^{0.4305} \left(\frac{\rho_G u_G}{a_p \mu_G}\right)^{1.0-0.192\mu_L/\mu_L^R} \quad (61)$

Rocha et al.,⁵⁵ on the basis of the Shi and Mersmann correlation, focused on sheet packing and added an alternative correction factor. At the same time, a surface enhancement factor was also introduced to account for the surface treatment of the packing. Gualito et al.¹⁰¹ corrected the Rocha et al.⁵⁵ correlation by a factor to make it more applicable to high-pressure conditions. Olujic⁵⁶ developed a pure empirical correlation from experimental results but took holes on the packing surface into consideration. The correlation underwent a big modification⁵⁸ by introducing the Onda and co-workers correlation, adopting the effective liquid flow angle and considering the effect of the corrugation angle.

The Xu et al.⁵⁰ correlation, on the basis of the Billet and Schultes model,⁹⁵ adopted the relative stabilizing index S_R to account for the Marangoni effect and included the influence of gas rates, which is usually omitted. Siminiceanu et al.⁸⁰ determined the effective mass-transfer area of Mellapak750Y structured packing and correlated by a critical equation.

A summary of the various published correlations for the effective interfacial area for structured packings is

given in Table 5, with the exponents or expressions of the principal parameters summarized in Table 6.

Model Refinement Considerations

Liquid-Phase Mass-Transfer Coefficient. In general, it was considered that the mass-transfer resistance in packed columns mainly lies on the gas-phase side and that $HTU_{OG} = HTU_G$. However, in the processes where mass transfer is liquid-phase controlled, for example, some cases for high-pressure distillation and the most cases for absorption or desorption, the liquid-phase mass-transfer coefficient is an important factor. Liquid-phase mass-transfer coefficients could be obtained by dividing the volumetric mass-transfer coefficient $k_L a_e$ measured experimentally by the effective interfacial area a_e determined by experiments or predicted by correlations. The penetration model³⁵ can also be used to estimate the liquid-side mass-transfer coefficient.¹⁰² According to penetration theory, the liquid mass-transfer coefficient, mainly depending on liquid rate and contact time or exposure time, was independent of the

Table 5. Correlations for the Effective Interfacial Area for Structured Packings

authors	correlations
Shi and Mersmann ³¹	$\frac{a_e}{a_p} = \frac{0.76md_{pe}^{1.1}u_L^{0.4}\nu_L^{0.2}}{1 - 0.93 \cos \theta} \left(\frac{\rho_L}{\sigma_L g} \right)^{0.15} \frac{a_p^{0.2}}{\epsilon^{0.6}} \quad (62)$
Spiegel and Meier ⁹⁶	$a_e = A(\rho_L u_L)^{0.2} \quad (63)$
Bravo and Fair ^{43,97}	$a_e/a_p = 0.5 + 0.0058F_r \quad (F_r < 0.85) \quad (64a)$
	$a_e/a_p = 1 \quad (F_r > 0.85) \quad (64b)$
Henriques de Brito et al. ⁹⁸	$\frac{a_e}{a_p} = 0.465 \left(\frac{\rho_L u_L}{\mu_L a_p} \right)^{0.3} \quad (65)$
Hanley et al. ⁴⁸	$\frac{a_e}{a_p} = (1 - f) \left[1 - \exp \left(-\omega \frac{d_{eq} u_L^2 \rho_L}{\epsilon \sigma_L} \right)^z \right] \quad (66a)$
	$\frac{a_e}{a_p} = \frac{1 - (ff_c)^p}{(1 - f)^{-1}} \left[1 - \exp \left(-\omega \frac{d_{eq} u_L^2 \rho_L}{\epsilon \sigma_L} \right)^z \right] \quad (66b)$
Brunazzi et al. ⁹⁹	$\frac{a_e}{a_p} = \frac{d_{eq}}{4} \left(\frac{h_L}{\epsilon} \right)^{1.5} \left[\frac{\rho_L g \epsilon (\sin \alpha)^2}{3 \mu_L u_L} \right]^{0.5} \quad (67)$
Rocha et al. ⁵⁵	$\frac{a_e}{a_p} = F_{se} F_t = F_{se} \frac{29.12 u_L^{0.4} \nu_L^{0.2} s^{0.159}}{(1 - 0.93 \cos \theta)(\sin \alpha)^{0.3} \epsilon^{0.6}} \left(\frac{\rho_L}{\sigma_L g} \right)^{0.15} \quad (68)$
Olujic et al. ⁵⁶	$\frac{a_e}{a_p} = \frac{1 - \Omega}{1 + A/u_L^B} \quad (69)$
Gualito et al. ¹⁰¹	$\frac{a_e}{a_p} = \left(\frac{a_e}{a_p} \right)_{eq\ 68} \left[\frac{1.2}{1 + 0.2 \exp(15u_L/u_G)} \right] \quad (70)$
Siminiceanu et al. ⁸⁰	$\frac{a_e}{a_p} = 0.1245 \left[\frac{6 \rho_L u_L (1 - \epsilon)^{0.4}}{\mu_L a_p} \right] \quad (71)$
Xu et al. ⁵⁰	$\frac{a_e}{a_p} = \frac{0.00842}{(a_p d_h)^{0.5}} \left(\frac{\rho_L u_L d_h}{\mu_L} \right)^{-0.2} \left(\frac{u_L^2 \rho_L d_h}{\sigma_L} \right)^{0.614} \left(\frac{u_L^2}{g d_h} \right)^{-0.45} \left(\frac{\rho_G d_h u_G}{\mu_G} \right)^{0.647} (1 \pm S_R^{0.358}) \quad (72)$
	where \pm stands for surface tension positive and negative systems, respectively.
Olujic ⁵⁸	$\frac{a_e}{a_p} = (1 - \Omega) \left(\frac{a_e}{a_p} \right)_{eq\ 51} \left(\frac{\sin 45}{\sin \alpha_L} \right)^n \quad (73)$
	where $n = \left(1 - \frac{a_p}{250} \right) \left(1 - \frac{\alpha_L}{45} \right) + \ln \left(\frac{a_{e,Onda}}{250} \right) + (0.49 - \sqrt{760/P}) \left(1.2 - \frac{\alpha_L}{45} \right)$

gas rate, which was indicated by recent investigations made below the loading point by Murrieta et al.¹⁰² Early work by Sherwood and Holloway⁴ has shown that, in the loading region, the gas rate increase can enhance the liquid-phase mass transfer. However, just as pointed out by Murrieta et al.,¹⁰² very little experimental work related to the liquid-phase mass-transfer coefficient has been performed with liquids other than water.

Up to now, almost all correlations in the literature for the liquid-side mass-transfer coefficient are based on the penetration theory,³⁵ with few exceptions, for example, the correlations developed by Shulman et al.¹¹ and Onda et al.²⁰ The difference between these correlations lies in the calculation method of contact time, which was often determined by empirical expressions for the sake of simpleness. In the previous works, the contact time estimation has been demonstrated to be critical for calculating the liquid-phase mass-transfer coefficient via penetration theory.

Shetty and Cerro⁶⁴ studied the flow patterns and other transport parameters over the rugged surfaces of structured packings and through their constricted channels. Their investigations essentially involved modeling the liquid-film thickness over the packing surface and understanding the influence of the gas phase on it. The exposure time was computed using the analytical expressions derived for the velocity of the free surface. The velocity of the free surface was calculated using the flow angle defined by Spekuljak and Billet⁶³ for a corrugated

surface. This approach also takes into consideration the fact that the path length traveled by the liquid between two peaks is larger. Unlike empirical modeling, this approach leads to a better understanding of the role of the design parameters and, more importantly, their interactions and can be termed film model.¹⁰³ Essentially, liquid mass-transfer-coefficient correlations based on the film model seems to be more reasonable. However, research studies on film models for structured packings are very new, and few publications^{104–107} available relate to this subject. Anyhow, improvements in contact time estimation could then be considered as a possible way to improve the liquid-phase mass-transfer-coefficient prediction.

Gas-Phase Mass-Transfer Coefficients. The gas-side mass-transfer coefficient is usually computed using the relationships based on the extensive investigations of wetted wall columns by Sherwood et al.¹⁰⁸ They concluded that the relationship of Johnstone and Pigford⁵² should be used for the gas side mass-transfer coefficients. This relationship is as follows,

$$Sh_G = C Re_G^m Sc_G^n \quad (74)$$

where C is a constant in the range of 0.18–0.04⁹⁷ and m and n are exponents of the Reynolds and Schmidt numbers, respectively. There exist a few exceptions, for example, the models proposed by Billet and Schultes³⁴ and Wagner et al.,³⁶ which were deduced from penetra-

Table 6. Exponents or Expressions Used in Correlations for the Effective Interfacial Area

	eq 50	eq 51 ^a	eq 52a	eq 53	eq 54	eq 56
ρ_L	-1.74 to -0.15 ^b -1.04 to 0.08 ^c	0.3	0.174	0.49	0.95	
u_L	-0.90 to 0.69 ^b -0.20 to 0.92 ^d	0.4	0.307	0.392	0.5	0.392
μ_L	0.13 ($\mu_L < 12$ cP) 0.31 ($\mu_L > 12$ cP)	-0.1	-0.041		-0.5	0.392
σ_L	$\alpha = f(\rho_L, u_L)$	-0.95	-0.315	-0.49	-0.45	0.108
ρ_G	$\beta = g(\rho_L, u_L)$					0.392
u_G	2β					0.392
μ_G						-0.392
σ_c ^d		0.75	0.182			
a_p		0.65	0.826	0.216	0	0.608
ϵ				$(1 - \epsilon)^{0.42}$		
d_{pe}					0.4	
α						
Z						-0.4
g		0.05		0.294	0.45	
	eq 58	eq 61	eq 59	eq 60a	eq 65	eq 67
ρ_L	$0.641 - 0.407w^e$	-0.05	-0.25	0.55	0.3	0.5
u_L	$0.641 - 0.407w$	0.4	0.89	0.4	0.3	$h_L^{1.5}$
μ_L	$0.407w - 0.641$	0.2	-0.13	0.2	-0.3	-1.0
σ_L		-0.15	-0.56	-0.75		
ρ_G						
u_G						
μ_G						
σ_c	$(1 + \cos \theta)^{3.477}$	$(1 - 0.93 \cos \theta)^{-1}$	0.18			1.0
a_p	$0.407w + 0.359$	1.2	$0.6 - 1.25$	0	0.7	-1.0
ϵ	$(1 - \epsilon)^{1.585}$	-0.6		0.5		-1.0
d_{pe}		1.1				1.0
α						$\sin \alpha$
Z			-0.65 to 0			
g		-0.15		0.45		0.5
	eq 68	eq 61	eq 71	eq 72	eq 73 ^a	
ρ_L	-0.05		0.4	0.414	0.3	
u_L	0.4	0.4305	0.4	0.128	0.4	
μ_L	0.2	0.4305	-0.4	0.2	-0.1	
σ_L	-0.15	-0.4305		-0.614	-0.95	
ρ_G		$\zeta = f(\mu_L)^f$		0.647		
u_G		ζ		0.647		
μ_G		$-\zeta$		-0.647		
σ_c	$(1 - 0.93 \cos \theta)^{-1}$				0.75	
a_p	2.2	$1 - \zeta$	0.6	-1.011	0.65	
ϵ	-0.6		$(1 - \epsilon)^{0.4}$	1.511		
d_{pe}	$0.359(s)$					
α	$\sin^{-3} \alpha$				$\sin^{-n} \alpha_L^g$	
Z						
g	-0.15			0.45	0.05	

^a The analysis is based on the power progression of corresponding correlations. ^b From Raschig. ^c From Berl. ^d The critical surface tension σ_c is related with contact angle by $\cos \theta = 1 + m(\sigma_c - \sigma_L)$. ^e $w = (1 + \cos \theta)/2$. ^f $\zeta = 0.1 - 0.8$. ^g $n = f(\alpha_p, P, \alpha_L)$.

tion theory. In general, the flow of two phases in wetted-wall columns was within the laminar region, and eq 74 was based on the assumption of laminar flow. But in fact, liquid flow instabilities due to waves, film detachment, and drop formation often violate this assumption. Olujic⁵⁶ was the first author who directly considered the more common phenomenon, transitional flow, which usually exists in packed columns. On the basis of the assumption that the extent of mass-transfer enhancement, which is due to entrance effects imposed by abrupt changes in flow direction at each transition between packing elements, is equal to that observed with heat transfer in laminar and turbulent regimes under similar conditions; the author gave Sherwood number expressions for both the laminar and turbulent flow regimes. This modeling idea could be considered as a more realistic and favorable approach.

Anyway, it is reasonable and necessary to take transitional flow into consideration for modeling mass-

transfer coefficients. Improvements in predicting the gas-phase mass-transfer coefficient could then be made by incorporating the more-complex flow regimes.

Effective Interfacial Area. Among numerous correlations for the effective interfacial area, the mechanistic model developed by Shi and Mersmann³¹ can be considered as a sound one. It is based on the model that Rocha et al.⁵⁵ developed, which is considered by Xu et al.⁵⁰ as the best model available for predictions in structured packing. On the other hand, it is quite conceivable that the liquid holdup in a column is closely connected with the interfacial area, i.e., the static holdup correlates with the static area and the dynamic holdup correlates with the dynamic area. This model, in comparison with previous ones, is more fundamental and considers more factors involved. It is consistent with the correlations of Onda et al.²⁰ and Puranik and Vogelpohl⁹⁰ in that they correlate the effective interfacial area with Reynolds number, Froude number, Weber

number, and wettability of the packing surface (via critical surface tension or wettability angle). At the same time, it agrees with the dimensional analysis by Gandhidasan.¹⁰⁹ Therefore, this correlation seems to be more reasonable and to have a wider applicability.

But this model does not involve the following factors: surface treatment of packing, liquid distribution, operating pressure, and gas rates. For the packing undergoing surface treatments, corrective terms similar to those of Olujic⁵⁶ and Rocha et al.⁵⁵ to account for the perforated and embossed treatment could be added.

As to liquid distribution, a lot of investigations were made^{44,56,100,110,111} by different researchers. The models obtained from these research studies are either strongly dependent on experiments or cumbersome mathematical calculations. Liquid distribution is a complex problem in that it relates to packing type and size, column dimensions, performance of internals, physical properties of the test system, and operating conditions; thus, it is difficult to express quantitatively. But qualitative analysis about some factors can be made, e.g., Henriques de Brito et al.⁹⁸ have indicated that a large packed aspect ratio enhances maldistribution of liquid and, thus, reduces the effective interfacial areas.

As for the operating pressure, besides the gas and liquid physical and transport properties, it can affect gas and liquid volumetric flow rates and, thus, interfacial contact status. It was found that the overall efficiency of structured packing is worse than expected in high-pressure applications. However, Zuiderweg and co-workers^{112,113} pointed out that a basic HETP is approximately constant in a wide pressure range. Analysis indicates that the deterioration likely comes from vapor backmixing induced by initial liquid maldistribution other than small-scale maldistribution and wall flow. On the other hand, a pressure increase may also give rise to vapor backmixing, and thus, apparent HETP increases. Therefore, the authors, on the basis of basic HETP, added HDU (height of dispersion unit), which is a function of the ratio of the gas-liquid velocity (total reflux) and the characteristic dimension of packing. Gualito et al.¹⁰¹ introduced a factor based on the ratio of the gas-liquid velocity to account for the pressure effect on the mass-transfer area under high-pressure distillation. It should be noted that, with an operating pressure increase, the difference between vapor and liquid density decreases and two-phase countercurrent operation was hindered.

As far as gas loads are concerned, most of the investigators considered that they have no influence on effective interfacial area or are contributing above the flooding point.¹¹⁴ As aforementioned, Shulman et al.¹¹ showed that the gas velocity can affect the interfacial area. Bravo and Fair²⁶ and Nakajima et al.⁸⁵ indicated a strong influence of the gas rates, via 0.392 and 0.8–0.1 powers of gas-phase Reynolds number, respectively, on the interfacial area for random packings. Xu et al.⁵⁰ also introduced the gas-phase Reynolds number in their equation for the effective interfacial area with an exponent of 0.647. These results indicate that the gas rate is an indispensable factor for the evaluation of mass-transfer performance. On one hand, increasing kinetic energy of gas is helpful to the formation of film surface rippling, droplet dispersion, and the occurrence of gas bubbling in puddles, which is favorable to mass transfer. On the other hand, gas velocity shows a great influence on liquid holdup, which is closely connected

with the interfacial area. A lot of studies show that the effect of the gas/vapor rate on interfacial area for structured packings is not as strong as that for random packings.

In addition, the liquid flow status also should be considered. For the liquid flow, on the wetted surface there may exist a stagnation zone, which is not renewed and does not participate in mass transfer. Liquid flow instabilities could bring extra area in addition to the fully wetted packing surface. Bravo and Fair²⁶ pointed out that the effective interfacial area is composed of not only the wetted area over the packing but also the area provided by suspended and falling droplets, gas bubbles within liquid puddles, ripples on the liquid film surface, and any contribution from film falling on the walls of the column. Generally speaking, the latter accounts for the considerable part of total areas available for mass transfer above the loading point in packed columns. Therefore, how to quantify their influence is very important for the accurate estimation of the effective interfacial area and the mass-transfer performance of packed columns.

Others. Almost all existing mass-transfer correlations are validated by the data obtained from two sources. One source is experiments carried out in pilot distillation columns packed with specific packing using recommended test mixtures, ordinarily at total reflux. Because of the equality between gas and liquid flow, the influence of liquid load on efficiency cannot be investigated independently of gas load. One way to address this problem is to make measurements at partial reflux conditions, but such experiments would be more difficult.

The other source is absorption experiments using systems such as $\text{NH}_3/\text{air}/\text{water}$ or $\text{CO}_2/\text{air}/\text{water}$. Here, the liquid load can be varied independently from the gas load. Although the results are valuable for the design of absorption columns, their use for organic distillation systems is limited because of the different wetting properties of organic and water.

Conclusions

Although a lot of studies relating mass-transfer prediction for packed columns have been conducted, the research on modeling still remains open, with a margin for further improvement in both fundamentals and applications. Because of the lack of understanding of the complex transport phenomena occurring in the packed columns, the present state is that the modeling is mainly based on empirical and semiempirical correlations, most of which were obtained from experimental data-fits. This lack of a scientific basis for predicting the performance of new equipment in large diameter columns slows down both the development and the acceptance of new equipment and necessitates trial-and-error techniques to tune their performance.¹⁰³

Therefore, more research works are needed to understand more fundamental phenomena occurring in packed columns and then to improve the precisions of predictive models. The deeper investigations should be directed to the basic fluid mechanics and mass-transfer phenomena. Detailed analysis of liquid film flow over packings can lead to a better understanding of the role of the design parameters and, more importantly, their interactions, as demonstrated by the model of Shetty and Cerro,^{64,65} which was unfortunately the only model we have found based on theoretical liquid-phase fluid

modeling. A hopeful improvement on this model could be reached by investigating spatial liquid distribution inside the packings. This has been considered as one of the weak areas of research, and serious experimental as well as theoretical works are very much expected.

On the other hand, there are opportunities for rigorous film modeling now with the availability of higher computational power and better numerical methods such as finite element techniques and CFD (computational fluid dynamics) methods. The CFD can help give the detailed pictures of the fluid flow, heat and mass transfer, and associated phenomena in chemical process equipment by means of a computer-based simulation.¹¹⁵ A recent work¹¹⁶ on tray columns shows that, with CFD, it is possible to model the concentration fluctuation terms and to give a closure to a combined equation set for both the fluid dynamics and the mass transfer. The solution of this equation set can lead to a rigorous simulation and can give velocity and concentration distributions simultaneously. However, there are only a few publications^{117–120} on the use of this technique in modeling mass transfer in a packed column. The main reason is that, with the CFD tools available, it is still difficult to deal with the complicated boundary conditions associated with vapor–liquid two-phase flow in a packed column, and the simulation turns out to be too expensive in terms of computing resources.

Nevertheless, the accuracy of film modeling and the three-dimensional rigorosity of CFD techniques, as shown by Gu et al.¹²¹ in their elementary work for liquid film simulation on a corrugated surface, open up tremendous possibilities of reliable modeling of hydrodynamics and mass transfer in packed columns.

As for experiments, broadening the range of experimental conditions and improving the equipment to obtain more reliable data is badly needed. In addition, new noninvasive measurement techniques such as gamma- or X-ray scanning, radioactive tracing, and advanced imaging and identifying means such as X-ray tomography and electrical resistance tomography should continue to be tested.

Nomenclature

a = constant in eqs 43 and 59
 a_e = effective specific interfacial area, 1/m
 a_{dyn} = dynamic specific surface area, 1/m
 a_p = packing specific surface area, 1/m
 a_{stat} = static specific surface area, 1/m
 a_w = wetted specific surface area, 1/m
 A = constant in eqs 36, 50, 63, and 69
 b = corrugation base length, m; constant in eq 43 and 59
 B = constant in eq 50 and 69
 B_1, B_2 = exponents in eq 36
 c = packing-specific constant in eqs 13 and 59; constant in eq 43
 C = physical property parameters in eqs 7 and 8; packing-specific constant in eqs 20 and 21
 C_0, C_0' = correction coefficients in eqs 7 and 8
 C_1, C_2 = empirical coefficients in eq 30
 C_f = correction factor for effect of approach to flooding
 C_{pk} = dimensionless packing characteristic in eq 23
 d = liquid film characteristic dimension defined by 4δ , m
 d_c = column internal diameter, m
 d_{eq} = equivalent diameter of flow channel, m
 d_h = hydraulic diameter of packing defined by $4\epsilon/a_p$, m
 d_{hG} = hydraulic diameter for the gas phase defined by $(bh - 2\delta h)/(s + b/2)$, m
 d_N = packing nominal diameter, m

d_p = particle diameter defined by $6(1 - \epsilon)/a_p$, m
 d_{pe} = diameter of a packing element, m
 D = diffusion coefficient, m²/s
 f = fraction of cells occupied
 f_c = fraction of cells occupied at the percolation threshold
 f_μ = correction factor for viscosity defined by $(\mu_l/\mu_{\text{water}})^{0.16}$
 f_ρ = correction factor for density defined by $(\rho_l/\rho_{\text{water}})^{-1.25}$
 f_σ = correction factor for surface tension defined by $(\sigma_l/\sigma_{\text{water}})^{-0.8}$
 F_V = gas load factor defined by $u_G(\rho_G)^{0.5}$, Pa^{0.5}
 F_r = flood percentages
 F_{se} = packing surface enhancement factor
 F_t = correction factor for total liquid holdup
 g = gravitational constant, m/s²
 g_{eff} = effective gravity, m/s²
 h = corrugation height, m
 h_L = liquid holdup, m³/m³
 h_{pe} = height of a packing element, m
 HDU = height of dispersion unit, m
 $HETP$ = height equivalent to a theoretical plate, m
 HTU = height of mass-transfer unit, m
 k_G = gas-side mass-transfer coefficient, m/s
 k_L = liquid-side mass-transfer coefficient, m/s
 K = dimensionless coefficient in eqs 15, 16, and 18
 K_V = density-corrected superficial vapor velocity defined by $u_G(\rho_G/(\rho_L - \rho_G))^{0.5}$, m/s
 l = contact length in eq 30, m; the characteristic length of packings in eqs 40 and 41, either d_{eq} or d_h , m
 $l_{G,pe}$ = length of gas flow channel in a packing element, m
 l_{ratio} = ratio of actual path length to path for $\alpha = 90^\circ$ C in eq 39, defined by $3.7617 - 0.123\alpha + (1.976 \times 10^{-3})\alpha^2 - (1.1167 \times 10^{-5})\alpha^3$ (α in radians)
 L = superficial liquid mass flow rate in eq 50, kg/(m²·s)
 L_W = liquid wetted rate, m³/(m·s)
 m = packing-related constants in eqs 3, 11, 47a, 50, and 62
 m_{eq} = slope of the equilibrium line
 n = packing-related constants in eqs 3, 11, 47b, and 50
 M = molecular weight, g/mol
 Ma_L = Marangoni number defined by $(d\sigma_L/dx)[(x - x^*)/(D_L\mu_L a_p)]/[\lambda/(\lambda + HTU_G/HTU_L)]$
 p = scaling exponent in eqs 31 and 66b; constant in eq 50
 P = operation pressure, mmHg
 q = liquid flow rate per unit packing width, m³/(m·s); constant in eq 50
 r = constant in eq 50
 R = film fill coefficients in eqs 7 and 8
 Re = Reynolds number defined by $\rho u d/\mu$
 Re_{Grv} = relative velocity Reynolds number defined by $\rho_G(u_{Ge} + u_{Le})d_{hG}/\mu_G$
 s = corrugation side length, m; constant in eq 50
 S_R = relative stability index defined by $-(d\sigma/(\sigma dx)) - (x - x^*)$
 Sc = Schmidt number defined by $\mu/\rho D$
 Sh = Sherwood number defined by kd_{eq}/D
 u = superficial velocity, m/s
 V_L = rivulet flow rate, m³/s
 w = width of rivulet, m
 x = mole fraction of more volatile component in liquid phase
 z = stretched exponential power in eqs 66a and 66b
 Z_p = height of each packed bed, m
 Z_t = total height of packed bed, m

Greek Letters

α = corrugation inclination angle, deg; relative volatility in eq 43
 α_L = effective liquid flow angle, deg
 Γ = liquid flow based on perimeter, kg/(m·s)
 δ = liquid film thickness, m
 ϵ = void fraction of packing

θ = contact angle, deg
 λ = stripping factor
 μ = viscosity, kg/(m·s)
 μ_L^R = reference liquid viscosity (pure water at 20.2 °C), kg/(m·s)
 ν = kinematic viscosity, m²/s
 ξ_{GL} = gas-liquid friction factor
 ρ = density, kg/m³
 σ = surface tension, N/m
 σ_c = critical surface tension, N/m
 φ = fraction of the triangular flow channel occupied by liquid defined by $2s/(b + 2s)$
 ψ = packing-related parameter in eq 11, m
 ϕ = packing-related parameter in eq 12, m
 Φ = enhancement factor for turbulent diffusion in eqs 22 and 23
 ω = decay rate of the stretched exponential used in eqs 66a and 66b
 Ω = packing surface void fraction

Subscripts

CR = chemical reaction
 dyn = dynamic
 e = effective
 EV = evaporation
 G = gas or vapor
 lam = laminar flow
 L = liquid phase
 neu = surface tension neutral systems
 neg = surface tension negative systems
 pos = surface tension positive systems
 PA = physical absorption
 stat = static
 turb = turbulent flow

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