

Review

A Review on Gas-Liquid Mass Transfer Coefficients in Packed-Bed Columns

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Abstract: This review provides a thorough analysis of the most famous mass transfer models for random and structured packed-bed columns used in absorption/stripping and distillation processes, providing a detailed description of the equations to calculate the mass transfer parameters, i.e., gas-side coefficient per unit surface k_y [$\text{kmol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$], liquid-side coefficient per unit surface k_x [$\text{kmol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$], interfacial packing area a_e [$\text{m}^2\cdot\text{m}^{-3}$], which constitute the ingredients to assess the mass transfer rate of packed-bed columns. The models have been reported in the original form provided by the authors together with the geometric and model fitting parameters published in several papers to allow their adaptation to packings different from those covered in the original papers. Although the work is focused on a collection of carefully described and ready-to-use equations, we have tried to underline the criticalities behind these models, which mostly rely on the assessment of fluid-dynamics parameters such as liquid film thickness, liquid hold-up and interfacial area, or the real liquid paths or any mal-distributions flow. To this end, the paper reviewed novel experimental and simulation approaches aimed to better describe the gas-liquid multiphase flow dynamics in packed-bed column, e.g., by using optical technologies (tomography) or CFD simulations. While the results of these studies may not be easily extended to full-scale columns, the improved estimation of the main fluid-dynamic parameters will provide a more accurate modelling correlation of liquid-gas mass transfer phenomena in packed columns.

Keywords: separation technologies; unit operations; gas-liquid mass transfer; mass transfer review; mass transfer coefficients for packed columns; structured packing; random packing; absorption column; stripping column; distillation column; cooling tower



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1. Introduction

Gas-liquid contactors are used extensively in chemical process industries, especially in the field of distillation, evaporation, humidification, gas absorption and desorption. These operations are based on the transfer of one or more components between two or more fluid phases to produce high added-value compounds, condition gas streams or purify one of the phases.

Most of the mass-transfer contactors in the chemical industry are either trays or packing towers. The latter is preferred when the process benefit for low-pressure drops and a limited number of feed and side-streams are required. Among column packings, random elements have been used since the beginning of the 20th century, and they consist of discrete structural elements that are randomly dumped in the contact vessel [1,2].

Since the second and half of the 1980s, structured packings entered the chemical industry market. Structured packings are made of corrugated metal or plastic sheets or wire meshes that are placed vertically into the column as blocks of assembled layers. These packings have gained a fast-growing spread in the process industries, as they can provide higher capacity and interfacial area with high separation efficiency, small scale-up limits and much lower pressure drops than random packings [3]. These features make the

structured packings more convenient for those applications requiring lower pressure drops and smaller space requirements.

The proper design and rating of packed towers require accurate models to predict pressure drops and mass-transfer coefficients. Several models have been proposed in the past to estimate the mass transfer coefficients of packings. These models are normally semi-empiric and require assessment of calibration parameters to account for the specific geometry of the packing. Their utilization in process design tends to provide a different estimation of the mass transfer coefficient that will translate into a different degree of accuracy in the estimation of column height and in a different bias for the process design.

The history of mass transfer coefficients correlations for packing towers has more than 60 years. The first models of large diffusion in the unit operation textbooks were the Cornell's method [4] for Raschig rings and Berl saddles, the McCabe et al. method [5] experimentally based approach and the Onda's method [6]. Years later, Bolles and Fair [7] extended the Cornell's method also for Pall ring and Intalox saddles. Instead, Bravo et al. [8] proposed a new model for mass transfer estimation in random packed columns.

For structured packings, one of the first models adopted in several textbooks was provided by Bravo et al. [8] and modified by Shi and Mersmann [9] which revised the estimation of interfacial surface area of the original model. These were based on semi-empirical equations, two-films theory for estimation of gas-side coefficient and the penetration theory for the liquid-side coefficient.

After their pioneering work, Bravo et al. [10] reported a revised version of their model adding liquid hold-up and film thickness data as physical parameters and introducing two correction factors: the first to correct the total liquid hold-up and the second to take into account the surface renewal of the packing. These parameters required a suitable calibration based on experimental data.

One year later, Billet and Schultes [11] proposed a descriptive model using the theory of penetration for the calculation of both gas and liquid coefficients, introducing corrective factors for both k_y and k_x equations. The authors provided a number of calibration parameters based on extensive experimental tests conducted under specific operating conditions (different gas and liquid loads, pressure and temperature and several chemical-physical properties of gas and liquid) and specific packing both random and structured.

Other authors (e.g., Brunazzi and Paglianti [12] and Olujić et al. [13]) provided a second generation of models following either the approach proposed by Bravo et al. [10] or Billet and Schultes [11], always considering suitable calibration parameters.

Lately, Hanley and Chen [14] used a new data fitting procedure derived from Bravo and Billet and Schultes experiments [8,10,11,15], which were adopted as reference mass transfer models for distillation and absorption columns in the ASPEN PLUS software. Hanley and Chen [14] proposed a new set of equations based on dimensionless groups, fitting parameters and functional dependencies found for specific random and structured packing classes.

To the best of our knowledge, only a limited number of review papers are available in the specialized literature for mass transfer in packed towers equipped with random and structured packing. One of the most recent was proposed by Wang et al. [16] who reviewed available correlations to calculate the mass-transfer coefficients in liquid and gas-side and the interfacial area. However, Wang et al. [16] focused the study on the theories adopted to develop the mass-transfer models and provided only a short summary of the main model equations examined. Other works such as Olujić and Seibert [17] or Wang et al. [18] report a brief summary of the mass transfer models.

This review work aims to provide a comprehensive and critical overview of the most recognized models in the last decades for predicting mass-transfer coefficients and the interfacial areas for packed towers with random and structured packing. The work has carefully selected and examined the most successful correlation models and adopted in the open scientific literature [19–42] also reporting the correlations by Hanley and Chen [14], which turns out to be one of the most recent and approved models in the literature.

This review has the merit of providing the reader with both the theoretical approach adopted by the authors and the relative mathematical formulation for the estimation of the gas-side (k_y) and the liquid-side (k_x) mass-transfer coefficients and of the effective interfacial area (a_e) reported in a rigorous and complete form together with the fitting parameters of the models necessary for their use. This study also provides the ranges of models' validity and applicability together with their main pros and cons, to help the reader in selecting the most suitable one for specific packing/application. Finally, the last part of the paper describes the new possibilities to refine the proposed correlations offered by new experimental findings and modeling approaches available in the last 20 years literature.

2. Mass Transfer Models for Packed-Bed Columns

The works reported in this review are based on different theories and experimental evidence on mass transfer for packed columns developed by several authors in the last fifty years. The models are suitable for specific types of random and structured packings and provide the predictive correlations for liquid and gas mass-transfer coefficients (k_x and k_y) per surface unit [$\text{kmol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$] and wet effective surface area to the mass transfer (a_e , [$\text{m}^2\cdot\text{m}^{-3}$]).

Most of these correlations are based on the use of dimensionless groups (e.g., Reynolds gas Re_G and liquid Re_L numbers, Schmidt gas Sc_G and liquid Sc_L numbers, Froude liquid number Fr_L , Weber liquid number We_L , Kapitza liquid number Ka_L , Graetz liquid number Gr_L), characteristic dimensions of packing (e.g., equivalent or hydraulic diameters, specific packing sizes and surface area, corrugation or inclination angle of packing sheet, void and holes fractions of the packing) and physical and fluid-dynamics properties of gas and liquid streams (e.g., molecular weight, density, viscosity, surface tension and diffusivity). The equations also included a series of model fitting parameters to adapt their correlations to specific types and models of packings.

2.1. Onda et al., 1968 (The OTO Model)

The OTO model presented correlations for the mass transfer coefficients in gas absorption and desorption processes, based on the vaporization of water and the gas absorption into organic solvents in columns filled with randomized packings.

In a number of papers dating back to the early sixties [43–45], the same authors supposed that the wet surface area of packing (a_e) is identical with the gas-liquid interface. However, they derived a new empirical equation for wet surface area, taking into account the liquid surface tension (σ_L , [$\text{N}\cdot\text{m}^{-1}$]) and the critical surface tension of packing material (σ_c , [$\text{N}\cdot\text{m}^{-1}$]) as a model parameter:

$$\frac{a_e}{a_n} = 1 - \exp \left[-1.45 \left(\frac{\sigma_c}{\sigma_L} \right)^{0.75} Re_L^{0.1} Fr_L^{-0.05} We_L^{0.2} \right] \quad (1)$$

in which a_n is the nominal surface area [$\text{m}^2\cdot\text{m}^{-3}$], Re_L is the Reynolds liquid number, We_L is the Weber liquid number, and Fr_L is the Froude liquid number. In Table 1 are listed the critical surface tension (σ_c) values for different packing materials [46].

Table 1. Critical surface tension for different packing materials [46].

Packing Material	Ceramic	Steel	Plastic	Carbon
σ_c [$\text{N}\cdot\text{m}^{-1}$]	0.061	0.075	0.033	0.056

The abovementioned dimensionless numbers are given by the following expressions:

$$Re_L = \frac{\rho_L u_{Ls}}{a_n \mu_L} \quad (2)$$

$$We_L = \frac{\rho_L u_{Ls}^2}{a_n \sigma_L} \quad (3)$$

$$Fr_L = \frac{a_n u_{Ls}^2}{g} \quad (4)$$

where u_{Ls} [$\text{m}\cdot\text{s}^{-1}$] is the superficial liquid velocity, ρ_L [$\text{kg}\cdot\text{m}^{-3}$] is the mass liquid density, μ_L [$\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$] is the mass liquid viscosity, and g is the acceleration of gravity ($9.81 \text{ m}\cdot\text{s}^{-2}$).

Onda et al. [6] showed that a_e equation can be applicable within $\pm 20\%$ error to the column filled with Raschig rings, Berl saddles, Spheres and Rods made of ceramic, glass, polyvinylchloride and also coated with paraffine film.

The liquid-phase mass transfer coefficient (k_x) data are referred to gas absorption into water, desorption from water, gas absorption of CO_2 into water adding a non-foaming surfactant (Newpol PE-61) and gas absorption of pure CO_2 into methanol and carbon tetrachloride. The authors obtained the following correlation:

$$k_x = C_L^{OTO} \left(\frac{\mu_L g}{\rho_L} \right)^{-1/3} Re_L^{2/3} Sc_L^{-1/2} (a_n d_p)^{0.4} \rho_x \quad (5)$$

where C_L^{OTO} is a proportionality model factor set to 0.0051, Re_L is given by Equation (2) replacing the nominal surface area (a_n) by the wet surface area (a_e), d_p [m] is the diameter of a sphere possessing the same surface area as a piece of packing (i.e., the packing sizes), ρ_x [$\text{kmol}\cdot\text{m}^{-3}$] is the molar liquid density, and Sc_L is the Schmidt liquid number defined by

$$Sc_L = \frac{\mu_L}{\rho_L D_L} \quad (6)$$

in which D_L [$\text{m}^2\cdot\text{s}^{-1}$] is the gas diffusivity in the liquid phase. The exponent of Re_L in Equation (5) coincides with that derived on the wet surface area basis by Vankrevelen and Hoftijzer [47] and Fujita and Hayakawa [48] and also is nearly equal to 0.61 of that derived by Norman and Sammak [49].

The liquid-side mass transfer coefficients (k_x) for gas absorption and desorption in packed columns have been correlated within an error of $\pm 20\%$ for organic solvents as well as water.

The gas-side mass transfer coefficient (k_y) data are referred to gas absorption processes reported in the literature [45,48,50–52]. The empirical equation that best represents experimental data is reported below.

$$k_y = C_G^{OTO} Re_G^{0.7} Sc_G^{1/3} (a_n d_p)^{-2.0} D_G \rho_y \quad (7)$$

where C_G^{OTO} is a proportionality model factor set to 5.23 for packing sizes above 15 mm and to 2.0 for sizes below 15 mm, D_G [$\text{m}^2\cdot\text{s}^{-1}$] is the gas diffusivity in the gas phase, and ρ_y [$\text{kmol}\cdot\text{m}^{-3}$] is the molar gas density; Re_G and Sc_G are, respectively, the Reynolds and the Schmidt gas numbers, which are expressed as

$$Re_G = \frac{\rho_G u_{Gs}}{a_n \mu_G} \quad (8)$$

$$Sc_G = \frac{\mu_G}{\rho_G D_G} \quad (9)$$

where u_{Gs} [$\text{m}\cdot\text{s}^{-1}$] is the superficial gas velocity, ρ_G [$\text{kg}\cdot\text{m}^{-3}$] is the mass gas density, and μ_G [$\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$] is the mass gas viscosity. The k_y equation for gas absorption is also applicable to the vaporization process with $\pm 30\%$ error. Onda et al. [6] also found that the difference between the mass transfer data for absorption and that for vaporization is quite small and could be neglected.

The authors have also studied the applicability of this model for mass transfer to distillation processes with packed columns, including the systems benzene-toluene, methanol-

water and ethanol-water at total and finite reflux ratios, where both the gas and liquid side resistances should be considered. The agreements between the calculated value for the mass transfer coefficients with the experiments are within $\pm 30\%$ except for columns higher than 1.0 m in which maldistribution of liquid might have occurred.

2.2. Bravo et al., 1985 (The BRF Model)

In 1985 Bravo, Rocha and Fair [8] proposed one of the first models to predict mass transfer coefficients and interfacial area for structured packing. The authors studied distillation processes for which liquid-side mass transfer resistance could be neglected, and consequently, *HETP* (height equivalent to a theoretical plate) could be approximated to the gas-side resistance of the mass transfer.

The model was developed for gauze-type structured packing Sulzer BX, where complete wetting of the packing surface is assumed. Based on the two-film theory [53], it allowed determining gas and liquid mass transfer coefficients.

The gas-side mass transfer coefficient (k_y) was estimated using the relationship proposed by Johnstone and Pigford [54] for counter-current evaporation in a wetted-wall column as

$$k_y = C_G^{BRF} Re_G^{0.8} Sc_G^{1/3} \left(\frac{D_G}{d_{eq}} \right) \rho_y \quad (10)$$

in which C_G^{BRF} is a proportionality factor set to 0.0338, d_{eq} (equivalent diameter, [m]) is the characteristic packing dimension, Sc_G is the Schmidt gas number defined as in Equation (9), Re_G is the Reynolds gas number for the BRF model which is defined in Equation (11):

$$Re_G = \frac{d_{eq} \rho_G (u_{Ge} + u_{Le})}{\mu_G} \quad (11)$$

where u_{Ge} and u_{Le} are the gas and liquid effective velocity through the channel. The equivalent diameter (d_{eq}) can be calculated through the knowledge of the packing dimensions that are, in particular, its base width (B_p), slant height (S_p) and height (H_p) of packing corrugation as

$$d_{eq} = B_p H_p \left(\frac{1}{B_p + 2S_p} + \frac{1}{2S_p} \right) \quad (12)$$

The packing dimension parameters (B_p , S_p and H_p) of structured packings are shown in Table 2.

Table 2. Packing dimension parameters for different packing type [36]. * The data for Mellapak 250.X are available in Flagiello et al. [22].

Structured Packing	B_p mm	S_p mm	H_p mm
Flexipac 1.Y, Metal/Plastic	12.7	9	6.4
Mellapak 2.Y, Metal/Plastic	33	21.5	13.8
Mellapak 125.Y, Metal/Plastic	55	37	24.8
Mellapak 250.Y, Metal/Plastic	24.1	17	11.9
Mellapak 350.Y, Metal/Plastic	15.3	11.9	8.9
Mellapak 500.Y, Metal/Plastic	9.6	8.1	6.53
Mellapak Plus 252.Y, Metal/Plastic	24.1	17	11.9
Mellapak 250.YS, Metal/Plastic	24.1	17	11.9
Mellapak 250.X, Metal/Plastic *	24.1	17	11.9
Sulzer BX, Metal/Plastic	24.1	17	11.9

The effective gas velocity (u_{Ge}) is given by the following expression:

$$u_{Ge} = \frac{u_{Gs}}{\varepsilon_p \sin \theta_c} \quad (13)$$

where ε_p [$\text{m}^3 \cdot \text{m}^{-3}$] is the void volumetric fraction of the packing, and θ_c [$^\circ$] is the inclination or corrugation angle. The liquid effective velocity (u_{Le}), instead, is assessed through the classical falling film equation [55] as

$$u_{Le} = \frac{3\Gamma}{2\rho_L} \left(\frac{\rho_L^2 g}{3\mu_L \Gamma} \right)^{1/3} \quad (14)$$

where Γ is the liquid flow per unit length of perimeter [$\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$], expressed as

$$\Gamma = \frac{\rho_L u_{Ls}}{P_s} \quad (15)$$

The perimeter per unit cross-sectional area (P_s , [$\text{m} \cdot \text{m}^{-2}$]) in Equation (15) is given by the following expression:

$$P_s = \frac{4S_p + B_p}{B_p h_p} \quad (16)$$

The penetration theory of Higbie [56] is used for the liquid-side mass transfer coefficient (k_x), which is expressed as

$$k_x = C_L^{BRF} \sqrt{\frac{D_L}{\pi t_e}} \rho_x \quad (17)$$

where t_e [s] is the exposure time, that is, the time it takes for a fluid element to flow between corrugation channels, and C_L^{BRF} is a model parameter, and its value is set to 2. According to this theory, the liquid-side mass transfer resistance could be neglected in comparison to the gas-side one. The exposure time (t_e) is defined as the ratio between the slant height of the corrugation (S_p) and the effective liquid velocity through the channel (u_{Le}).

$$t_e = \frac{S_p}{u_{Le}} \quad (18)$$

Regarding to the effective mass transfer surface area (a_e), the authors assumed the wet surface equal to the nominal surface (a_n), due to the corrugation and the capillarity of the packing which led to assume a unitary wettability efficiency.

$$\frac{a_e}{a_n} = 1 \quad (19)$$

Rocha, Bravo and Fair provided some modifications to the original model, extending it to the study of Sulzer Mellapak packings [10]. In particular, they proposed a novel expression for the effective surface area (a_e) based on Shi and Mersmann equation [9]:

$$\frac{a_e}{a_n} = 1 - 1.203 \left(\frac{u_{Ls}^2}{S_p g} \right)^{0.111} \quad (20)$$

In recent years, Orlando Jr [57] used several BRF model for the comparison between theoretical and experimental HETP values, the latter obtained with a lab column containing structured packing. Orlando Jr [57] showed that the model deviation from the experimental HETP was reduced from 47% to 8% average if Equation (20) was used instead of the unitary wettability assumption. The author also proposed to replace the slant height of the corrugation (S_p) with the hydraulic equivalent diameter (d_{eq}) in Equation (20) to reduce average deviation [57].

2.3. Bravo et al., 1992 (The SRP Model)

In 1992, Bravo et al. [10] proposed a new version of the previous Equations (10)–(20) called *SRP* (Separations Research Program) model. The authors modified the previous assumption about complete wettability of the packing surface area (a_e). Following the study by Shi and Mersmann [9] that proposed an expression base on fluid hydraulics over an inclined plane, Bravo and Fair [58] developed an equation for randomized packing under distillation and absorption/stripping conditions. For structured packing, Fair and Bravo [59] and Bravo et al. [10] observed a relatively influence by gas flow rate and a much more dependence by liquid rate.

Bravo et al. [10] took into account the problems due to the liquid distribution, such as initial liquid distribution, radial migration of liquid in the packed-bed, surface wettability and surface texturing. The estimation of wet surface area (a_e) in *SRP* model was based on the extensive study by Shi and Mersmann [9] about hydraulic consideration for sheet-metal packing.

$$\frac{a_e}{a_n} = F_{SE}F_t \quad (21)$$

The *SRP* model included two corrective factors to predict the effective surface area, following the first studies reported by Shi and Mersmann [9]. The first parameter is the surface enhancement factor (F_{SE}) which accounts for variations of surface packing (lancing, fluting, etc.), and the second is a correction factor for total liquid hold-up due to effective wetted area (F_t).

The F_{SE} represents a corrective parameter based on observation of liquid flow on packing surfaces relative to distillation experiments by McGlamery [60]. Some values for F_{SE} are reported in the studies of Rocha et al. [33,61] for several packing types. In Table 3 are shown the nominal surface area (a_n), void volumetric fraction (ϵ_p) and surface enhancement factor (F_{SE}) for some common packings.

Table 3. Characteristic of several structured packings [33,61].

Structured Packing	a_n $\text{m}^{-2} \cdot \text{m}^{-3}$	ϵ_p $\text{m}^{-3} \cdot \text{m}^{-3}$	S_p mm	F_{SE} -
Flexipac 2.Y, Metal	233	0.95	18	0.35
Gempak 2A, Metal	233	0.95	18	0.34
Gempak 2AT, Metal	233	0.95	18	0.312
Intalox 2T, Metal	213	0.95	22.1	0.415
Maxpak, Metal	229	0.95	17.5	0.364
Mellapak 250.Y, Metal	250	0.95	17	0.35
Mellapak 350.Y, Metal	350	0.93	11.9	0.35
Mellapak 500.Y, Metal	500	0.91	8.1	0.35
Sulzer BX, Metal	492	0.9	17	0.35

Bravo et al. [10] and Rocha et al. [33,61] proposed an expression for the correction parameter F_t :

$$F_t = \frac{29.12(We_L Fr_L)^{0.15} S_p^{0.359}}{Re_L^{0.2} \epsilon_p^{0.6} (1 - 0.93 \cos \gamma_c) (\sin \theta_c)^{0.3}} \quad (22)$$

where γ_c [°] is the contact angle which accounts for surface material wettability, Re_L is the Reynolds liquid number, We_L is the Weber liquid number, and Fr_L is the Froude liquid number for *SRP* model.

$$Re_L = \frac{u_{Ls} \rho_L S_p}{\mu_L} \quad (23)$$

$$We_L = \frac{u_{Ls}^2 \rho_L S_p}{\sigma_L} \quad (24)$$

$$Fr_L = \frac{u_{Ls}^2}{gS_p} \quad (25)$$

The value of $\cos\gamma_c$ in Equation (22) could be calculated for two different conditions:

$$\cos\gamma_c = \begin{cases} 5.211 \times 10^{-16.835\sigma_L} & \text{for } \sigma_L \geq 0.055\text{N} \cdot \text{m}^{-1} \\ 0.9 & \text{for } \sigma_L < 0.055\text{N} \cdot \text{m}^{-1} \end{cases} \quad (26)$$

based on the surface tension of the liquid.

The gas-side mass transfer equation was similar to previous model developed (BRF):

$$k_y = C_G^{SRP} Re_G^{0.8} Sc_G^{1/3} \left(\frac{D_G}{S_p} \right) \rho_y \quad (27)$$

$$Re_G = \frac{u_{Ge} \rho_G S_p}{\mu_G} \quad (28)$$

where C_G^{SRP} is a proportionality Sherwood gas number factor, and its value is 0.054 by Rocha et al. [33] and Re_G is the Reynolds gas number for SRP model. In this formulation, the side dimension of a corrugation cross section (S_p) was used as characteristic dimension of packing in place of the equivalent diameter (d_{eq}) adopted in BRF model [8]. The calculation of effective gas velocity (u_{Ge}) takes into account of the corrugation packing angle (θ_c), the space occupied by liquid using the void volumetric fraction of packing (ε_p) and the liquid hold-up (h_L).

$$u_{Ge} = \frac{u_{Gs}}{\varepsilon_p(1 - h_L) \sin\theta_c} \quad (29)$$

where h_L , [$\text{m}^3 \cdot \text{m}^{-3}$] is the volumetric fraction of the liquid hold-up which was expressed in terms of the liquid film thickness (δ_f , [m]), the side dimension of a corrugation cross section (S_p) and a correction factor for total hold-up (F_t).

$$h_L = 4F_t \left(\frac{\delta_f}{S_p} \right) \quad (30)$$

The liquid film thickness is obtained by a modification of the classical falling film equation [62,63]:

$$\delta_f = \left[\frac{3\mu_L u_{Ls}}{\rho_L \varepsilon_p h_L g_{eff} \sin\theta_c} \right]^{0.5} \quad (31)$$

in which g_{eff} [$\text{m} \cdot \text{s}^{-2}$] is the effective gravity related to a force balance on the liquid film which flows on the packing surface. The effective gravity can be expressed as a function of the liquid-gas densities, the pressure drops ($\Delta P/Z$, [$\text{Pa} \cdot \text{m}^{-1}$]) and K_1 constant (which depends only on the packing type [61]).

$$g_{eff} = 1 - \frac{\rho_G}{\rho_L} - K_1 \left(\frac{\Delta P}{Z} \right) \left(\frac{1}{g\rho_L} \right) \quad (32)$$

The effective gravity tends to decrease with increasing of pressure drops and for high loadings. When $g_{eff} = 0$, it means that the flooding condition occurs.

The K_1 parameter is constant for a particular shape of packing regardless of size or surface characteristic.

$$K_1 = \frac{g(\rho_L - \rho_G)}{\left(\frac{\Delta P}{Z} \right)_{flood}} \quad (33)$$

The $\Delta P/Z_{flood}$, [$\text{Pa} \cdot \text{m}^{-1}$] is a pressure drops per meter of packing at flooding condition, and it is a specific packing parameter like to K_1 constant. Fair and Bravo [64] observed that

the flooding appears in the 900–1200 Pa·m⁻¹ range for the packing investigated. For the sake of simplicity, the value of $\Delta P/Z_{flood}$ was fixed at 1025 Pa·m⁻¹ in order to calculate K_1 .

Finally, Equations (30) and (31) may be combined to give the final expression of liquid hold-up:

$$h_L = \left(4 \frac{F_t}{S_p}\right)^{2/3} \left(\frac{3\mu_L u_{Ls}}{\rho_L g_{eff} \sin \theta_c}\right)^{1/3} \quad (34)$$

For the liquid phase, the penetration approach [56] can be used with a revisited exposure time (t_e) adopted by BRF model:

$$t_e = \frac{S_p C_E^{SRP}}{u_{Le}} \quad (35)$$

where C_E^{SRP} is the surface renewal factor for SRP model. Murrieta [65] estimated, with experiments on oxygen-air-water system, that the value was slightly less than unity ($C_E^{SRP} \sim 0.9$) for structured packings.

The effective liquid velocity (u_{Le}) is defined as

$$u_{Le} = \frac{u_{Ls}}{\varepsilon_p h_L \sin \theta_c} \quad (36)$$

The liquid-side mass transfer equation is calculated according to the penetration theory [56] adding a correction factor (C_E^{SRP} , so-called surface renewal factor) for exposure time evaluation:

$$k_x = C_L^{SRP} \sqrt{\frac{D_L u_{Le}}{\pi S_p C_E^{SRP} \rho_x}} \quad (37)$$

where C_L^{SRP} is the same model parameter in BRF model ($C_L^{BRF} = C_L^{SRP}$).

The authors observed that the revised model deviation from the HETP experimental values was approximately $\pm 24\%$.

2.4. Billet and Schultes, 1993 (The BS Model)

Billet and Schultes [11] studied the mass transfer process into packed columns for gas absorption and distillation operations using 31 different liquid-gas systems and 67 different types of packings. The experiments were performed in different columns (in terms of height and diameter) working in counter-current flow and using both structured and random packings. The authors applied the penetration theory to both the gas and liquid mass transfer. The liquid mass transfer coefficient is defined as

$$k_x = 2 \sqrt{\frac{D_L}{\pi t_L}} \rho_x \quad (38)$$

where t_L [s] is the time necessary for the renewal of interface area.

$$t_L = h_L d_h \frac{1}{u_{Ls}} \quad (39)$$

in which d_h is the characteristic dimension of the packing (hydraulic diameter, [m]) and h_L is the liquid hold-up (below the loading point [66]) for this model:

$$d_h = 4 \frac{\varepsilon_p}{a_n} \quad (40)$$

$$h_L = \left(12 \frac{\mu_L a_n^2 u_{Ls}}{g \rho_L}\right)^{1/3} \quad (41)$$

Combining Equations (38)–(41) it is possible to obtain the expression for liquid-side mass transfer:

$$k_x = C_L^{BS} \left(\frac{g\rho_L}{\mu_L} \right)^{0.166} \left(\frac{u_{Ls}}{a_n} \right)^{0.333} \sqrt{\frac{D_L}{d_h}} \rho_x \quad (42)$$

where C_L^{BS} is a specific constant parameter for the liquid phase.

The mass transfer model in the gas phase is based on the assumption that the gas flow takes different directions through the packing and a theoretical time (t_G , [s]) required for renewal of the contact area between phases can be defined as

$$t_G = (\varepsilon_p - h_L) d_h \frac{1}{u_{Ls}} \quad (43)$$

The gas contact time (t_G) is comparatively short in the packed-beds and for the gas mass transfer can be assumed the Higbie analogy [56]:

$$k_y = 2 \sqrt{\frac{D_G}{\pi t_G}} \rho_y \quad (44)$$

The gas-side mass transfer equation is obtained by Equations (41), (43) and (44):

$$k_y = C_G^{BS} \left(\frac{1}{\sqrt{\varepsilon_p - h_L}} \right) \sqrt{\frac{a_n}{d_h}} D_G Re_G^{0.75} Sc_G^{0.333} \rho_y \quad (45)$$

where C_G^{BS} is a specific constant parameter for the liquid phase. In this model, Re_G is the Reynolds gas number expressed as

$$Re_G = \frac{u_{Gs} \rho_G}{\mu_G a_n} \quad (46)$$

The authors clearly stated that the packing specific constants, C_L^{BS} and C_G^{BS} depend on the characteristic constructions and material of the packing [11,15] and dedicated experimental tests are needed for their determination. Billet and Schultes estimated the values of the C_L^{BS} and C_G^{BS} for several random and structured packings [11,15]. Table 4 reports some values of the BS model fitting parameters.

Table 4. Characteristic data of several packings and specific constants C_L^{BS} and C_G^{BS} [15].

Random/Structured Packing	Material	Size mm	a_n $m^{-2} \cdot m^{-3}$	ε_p $m^{-3} \cdot m^{-3}$	C_L^{BS} -	C_G^{BS} -
Raschig Super-Rings	Metal	0.3	315	0.96	1.5	0.45
	Metal	0.5	250	0.975	1.45	0.43
	Metal	1	160	0.98	1.29	0.44
	Metal	2	97.6	0.985	1.323	0.4
	Metal	3	80	0.982	0.85	0.3
Raschig rings	Plastic	2	100	0.96	0.377	0.337
	Ceramic	50	95	0.83	1.416	0.21
	Ceramic	38	118	0.68	1.536	0.23
	Ceramic	25	190	0.68	1.361	0.412
	Ceramic	15	312	0.69	1.276	0.401
	Ceramic	13	370	0.64	1.367	0.265
	Ceramic	10	440	0.65	1.303	0.272
Ralu Flow	Carbon	25	202.2	0.72	1.379	0.471
	Plastic	1	165	0.94	1.486	0.36
Pall Rings	Plastic	2	100	0.945	1.27	0.32
	Metal	50	112.6	0.951	1.192	0.41
	Metal	38	139.4	0.965	1.012	0.341
	Metal	25	223.5	0.954	1.44	0.336

Table 4. Cont.

Random/Structured Packing	Material	Size mm	a_n $m^{-2} \cdot m^{-3}$	ϵ_p $m^{-3} \cdot m^{-3}$	C_L^{BS} -	C_G^{BS} -
Ralu rings	Plastic	50	111.1	0.919	1.139	0.368
	Plastic	38	151.1	0.906	0.856	0.38
	Plastic	25	225	0.887	0.905	0.446
	Ceramic	50	155.2	0.754	1.278	0.333
	Metal	50	105	0.975	1.192	0.345
	Metal	38	135	0.965	1.277	0.341
	Metal	25	215	0.96	1.44	0.336
Hiflow rings	Plastic	50	95.2	0.983	1.52	0.303
	Plastic	38	150	0.93	1.32	0.333
	Plastic	25	190	0.94	1.32	0.333
	Metal	50	92.3	0.977	1.168	0.408
	Metal	25	202.9	0.962	1.641	0.402
	Metal	50	117.1	0.925	1.478	0.345
	Plastic	50 hydr.	118.4	0.925	1.553	0.369
	Plastic	50 S	82	0.942	1.219	0.342
	Plastic	25	194.5	0.918	1.577	0.39
	Ceramic	50	89.7	0.809	1.377	0.379
NOR PAC rings	Ceramic	38	111.8	0.788	1.659	0.464
	Ceramic	20	286.2	0.758	1.744	0.465
	Plastic	50	86.6	0.947	1.08	0.322
	Plastic	35	141.8	0.944	0.754	0.425
	Plastic	25 type B	202	0.953	0.883	0.366
	Plastic	25	197.9	0.92	0.976	0.41
Glitsch rings	Metal	30 PMK	180.5	0.975	1.92	0.45
	Metal	30 P	164	0.959	1.577	0.398
VSP rings	Metal	50	104.6	0.98	1.416	0.21
	Metal	25	199.6	0.975	1.361	0.412
Envi Pac rings	Plastic	80	60	0.955	1.603	0.257
	Plastic	60	98.4	0.961	1.522	0.296
	Plastic	32	138.9	0.931	1.517	0.459
Bialecki rings	Metal	50	121	0.966	1.721	0.301
	Metal	35	155	0.967	1.412	0.39
	Metal	35	176.6	0.945	1.405	0.377
	Metal	25	210	0.956	1.462	0.331
Raflux rings	Plastic	15	307.9	0.894	1.913	0.37
TOP-Pac rings	Aluminum	50	105.5	0.956	1.326	0.389
Berl saddles	Ceramic	38	164	0.7	1.568	0.244
	Ceramic	25	260	0.68	1.246	0.387
	Ceramic	13	545	0.65	1.364	0.232
Intalox saddles	Ceramic	13	625	0.78	1.677	0.488
	Plastic	70	110.7	0.938	1.527	0.326
DIN-PAK	Plastic	47	131.2	0.923	1.69	0.354
Ralu Pak	Metal	YC-250	250	0.945	1.334	0.385
	Metal	250	250	0.975	0.983	0.27
Impulse packing	Metal	250	250	0.975	0.983	0.27
	Plastic	100	91.4	0.838	1.317	0.327
Montz packing	Metal	B1-200	200	0.979	0.971	0.39
	Metal	B1-300	300	0.93	1.165	0.422
	Plastic	C1-200	200	0.954	1.006	0.412
Euroform	Plastic	PN-110	110	0.936	0.973	0.167

The model is completed by the value of the effective wet surface area, expressed as

$$\frac{a_e}{a_n} = \frac{1.5}{\sqrt{d_h a_n}} Re_L^{-0.2} We_L^{0.75} Fr_L^{-0.45} \quad (47)$$

In this model, the Reynolds, Weber and Froude liquid numbers are defined as follows:

$$Re_L = \frac{u_{Ls}\rho_L d_h}{\mu_L} \quad (48)$$

$$We_L = \frac{u_{Ls}^2 \rho_L d_h}{\sigma_L} \quad (49)$$

$$Fr_L = \frac{u_{Ls}^2}{g d_h} \quad (50)$$

Billet and Schultes also provided the mean relative deviations of the calculated mass transfer coefficients from the experimentally determined values for absorption and desorption processes which were 8.3% for the liquid-side and 12.4% for the gas-side [11]. The mean relative deviation for the height of transfer units (HTU_{OG}) was also estimated at around 14.1%.

2.5. Brunazzi and Paglianti, 1997 (The BP Model)

Brunazzi and Paglianti [12] developed a mass transfer model by experiments concerned the desorption of CO_2 from water into air and in column working co-currently and the absorption of chlorinated compounds with two commercial high boiling liquids (Genosorb 300 and Genosorb 1843). The tests were performed in a column working counter-currently, using structured packings, such as Mellapak 250.Y and BX.

In this model, the evaluation of the mass transfer coefficient in the gaseous phase (k_y) is equal to that of the SRP model ($C_G^{BP} = C_G^{SRP} = 0.054$).

$$k_y = C_G^{BP} Re_G^{0.8} Sc_G^{0.33} \left(\frac{D_G}{d_h} \right) \rho_y \quad (51)$$

In Equation (51), the Reynolds gas number (Re_G) is defined as in Equation (11), and the characteristic dimension of the channels (d_h) is the same of BS model in Equation (40).

The effective gas velocity for BP model (u_{Ge}) set out in the Reynolds gas number (Re_G) is defined hereafter:

$$u_{Ge} = \frac{u_{Gs}}{(\varepsilon_p - h_L) \sin \theta_c} \quad (52)$$

The authors argued that the liquid mass transfer coefficient (k_x) depended on the packing height (Z , [m]) and the mixing factor; for this reason, they introduced two dimensionless numbers to describe the liquid phase: the Graetz (Gr_L) and the Kapitza (Ka_L) liquid numbers, which were correlated to k_x through the following equation.

$$k_x = a \frac{Gr_L^b}{Ka_L^c} \left(\frac{D_L}{d} \right) \rho_x \quad (53)$$

where a is a coefficient of proportionality for the Sherwood liquid number (Sh_L), b is the functional dependence on Graetz liquid number, and c is the functional dependence on Kapitza liquid number.

The authors obtained the model fitting parameters for metal/plastic Sulzer Mellapak Y-type and plastic BX-type [12] shown in Table 5.

Table 5. Characteristic parameters in Equation (53) for different packings [12].

Type of Packing	Material	a	b	c
Mellapak Y	Metal/Plastic	16.43	0.915	0.09
Sulzer BX	Plastic	63.10	0.676	0.09

The dimensionless numbers in Equation (53) are expressed as

$$Gz_L = Re_L Sc_L \frac{\delta_f}{H} \quad (54)$$

$$Ka_L = \frac{\sigma_L^3 \rho_L}{\mu_L^4 g} \quad (55)$$

where H [m] is the flow distance which represents the path of the liquid-phase from the top to the bottom of the column.

The flow distance (H) depends on the type of liquid mixing at the junctions of the structured packing. In the case of complete mixing, H is a function of the channel dimension, whereas in the case of partial mixing, it must be evaluated as the distance covered by the liquid phase flowing into the column. In the latter case, in addition to the geometric characteristics of the packings, H can also be related to the packing height (Z , [m]). The flow distance (H) is calculated as

$$H = \frac{Z}{\sin \theta_L} \quad (56)$$

in which θ_L [°] is the slope of the steepest descent line with respect to the horizontal axis. This geometric parameter can be computed using Spekulijak and Billet correlation [67], once the corrugation angle (θ_c) and the packing dimension parameters (B_p and H_p) are known:

$$\theta_L = \arctan \left(\frac{\cos(90 - \theta_c)}{\sin(90 - \theta_c) \cos \left[\arctan \left(\frac{B_p}{2H_p} \right) \right]} \right) \quad (57)$$

In this model, the Schmidt liquid number (Sc_L) was expressed as in Equation (9), while the Reynolds liquid number (Re_L) was defined as below:

$$Re_L = \frac{\rho_L u_{Le} d}{\mu_L} \quad (58)$$

The effective velocity of the liquid (u_{Le}) is a function of the superficial liquid velocity (u_{Ls}), of the dynamic liquid hold-up (h_L) and the slope of the steepest descent line with respect to the horizontal axis of the packing (θ_L).

$$u_{Le} = \frac{u_{Ls}}{h_L \sin \theta_L} \quad (59)$$

Furthermore, the characteristic dimension of the liquid film (d , [m]) is four times the liquid film thickness (δ_f):

$$d = 4\delta_f \quad (60)$$

The liquid film thickness (δ_f) is a function of liquid properties, geometric characteristics of the packing, liquid velocity (u_{Ls}) and the dynamic liquid hold-up (h_L). If the liquid-phase is Newtonian liquid and flows in laminar conditions, the liquid film thickness can be calculated as

$$\delta_f = \left(\frac{3\mu_L}{\rho_L \sin \theta_L} \cdot \frac{u_{Ls}}{h_L \sin \theta_L} \right)^{0.5} \quad (61)$$

Finally, for the evaluation of the effective wet surface (a_e), the authors used a model based on experimental measurements of the liquid hold-up [68,69]:

$$\frac{a_e}{a_n} = \frac{d_h \sin \theta_c}{4\epsilon_p} \sqrt{\frac{g\rho_L}{3\mu_L u_{Ls}}} (h_L)^{0.666} \quad (62)$$

However, the authors suggested the Suess and Spiegel approach to calculate the liquid hold-up for Mellapak-type structured packings [70]:

$$h_L = k_{hL} a_n^{0.83} (3600 u_{Ls}^x) \left(\frac{\mu_L}{\mu_{Lo}} \right)^{0.25} \quad (63)$$

in which μ_{Lo} is the dynamic viscosity of water at 20 °C [$\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$]; k_{hL} is a proportionality factor, which is set to 0.0169 for $u_{Ls} < 0.0111 \text{ m}\cdot\text{s}^{-1}$, otherwise to 0.0075; x is a model parameter, which is set to 0.37 for $u_{Ls} < 0.0111 \text{ m}\cdot\text{s}^{-1}$ and to 0.59 otherwise. Instead, for BX packings Torelli [71] proposed the following empirical equation to evaluate the liquid hold-up.

$$h_L = 0.82 u_{Ls}^{0.64} \left(\frac{\mu_L}{\mu_{Lo}} \right)^{0.25} \quad (64)$$

Finally, Brunazzi and Paglianti [12] showed that the values of $k_x a_e$ and $k_y a_e$ within an error of $\pm 15\%$ and $\pm 19\%$, respectively, for desorption of CO_2 from water in air and for absorption of chlorinated solvents with Genosorb 300 and Genosorb 1843 using Mellapak 250.Y.

2.6. Olujić et al., 2004 (The Delft Model)

The Delft model was first developed in 1997 by Olujić [72] and further improved between 1999–2004 [13,73,74]. This model, developed for corrugated structured packings, considers the gas “animated” by a zig-zag flow through the corrugated triangular packing channels, since the packing layers consists of single elements each rotated by 90° with respect to the adjacent elements.

Olujić [73] estimated the gas-side mass transfer coefficient from an analogy to the heat transfer, considering common transition from the laminar to the turbulent gas regime, so that the mass transfer coefficient is represented by the mean of the two individual contributions of the laminar ($k_{y,lam}$, [$\text{kmol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$]) and turbulent ($k_{y,turb}$, [$\text{kmol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$]) regime conditions:

$$k_y = \sqrt{(k_{y,lam})^2 + (k_{y,turb})^2} \quad (65)$$

$$k_{y,lam} = \frac{Sh_{G,lam} D_G}{d_{hG}} \quad (66)$$

$$k_{y,turb} = \frac{Sh_{G,turb} D_G}{d_{hG}} \quad (67)$$

where: $Sh_{G,lam}$ is the Sherwood gas number for laminar flow; $Sh_{G,turb}$ is the Sherwood gas number for turbulent flow, and d_{hG} [m] is the hydraulic diameter for the gas phase. The gas-side mass transfer coefficients referred to laminar and turbulent flow are defined below:

$$k_{y,lam} = C_G^{Delft} \left(\frac{D_G}{d_{hG}} \right) Sc_G^{1/3} \sqrt{Re_{Grv} \frac{d_{hG}}{l_{G,pe}}} \rho_y \quad (68)$$

$$k_{y,turb} = \frac{Re_{Grv} Sc_G \frac{\varphi \xi_{GL}}{8}}{1 + 1.27 \sqrt{\frac{\varphi \xi_{GL}}{8}} (Sc_G^{2/3} - 1)} \left[1 + \left(\frac{d_{hG}}{l_{G,pe}} \right)^{2/3} \right] \left(\frac{D_G}{d_{hG}} \right) \rho_y \quad (69)$$

where C_G^{Delft} is a proportionality coefficient for laminar flow case, and its value is 0.664; $l_{G,pe}$ [m] is the length of gas flow channel in a packing element, h_{pe} [m] is the height of structured packing element unit, φ is the fraction of the triangular flow channel occupied by liquid, Re_{Grv} is the Reynolds gas number based on relative effective velocities between

gas and liquid, and ζ_{GL} is the gas-liquid friction factor by Colebrook and White [75]. The expressions provided for these parameters are:

$$l_{G,pe} = \frac{h_{pe}}{\sin \theta_c} \quad (70)$$

$$\varphi = \frac{2S_p}{B_p + 2S_p} \quad (71)$$

$$Re_{Grv} = \frac{\rho_G(u_{Ge} + u_{Le})d_{hG}}{\mu_G} \quad (72)$$

$$\zeta_{GL} = \left[-2 \text{Log} \left[\frac{\delta_f}{3.7d_{hG}} - \frac{5.02}{Re_{Grv}} \text{Log} \left(\frac{\delta_f}{3.7d_{hG}} + \frac{14.5}{Re_{Grv}} \right) \right] \right]^{-2} \quad (73)$$

in which u_{Ge} is the effective gas velocity (expressed as BP model in Equation (52)), and u_{Le} is the effective liquid velocity.

$$u_{Le} = \frac{u_{Ls}}{\varepsilon_p h_L \sin \theta_L} \quad (74)$$

As in the Brunazzi and Paglianti model [12], the correlation of Spekuljak and Billet [67] was used to calculate the effective liquid flow angle θ_L , shown in Equation (57).

The data relative to geometric characteristic parameters of the packings used by Olujić works are reported in Table 6.

Table 6. Packing geometric parameters by Olujić et al. [74]. * The data for Mellapak 250.X are available in Flagiello et al. [22].

Structured Packing	a_n $\text{m}^{-2} \cdot \text{m}^{-3}$	θ_c °	ε_p $\text{m}^{-3} \cdot \text{m}^{-3}$	H_p mm	B_p mm	S_p mm	h_{pe} m
Montz B1-250, Metal	244	45	0.98	12	22.5	16.5	0.197
Montz B1-250.60, Metal	245	60	0.98	12	22.3	16.4	0.211
Montz B1-400, Metal	394	45	0.96	7.4	14	10.3	0.197
Montz B1-400.60, Metal	390	60	0.96	7.4	14.3	10.3	0.215
Montz BSH-400, Metal	378	45	0.97	7.4	15.1	10.6	0.194
Montz BSH-400.60, Metal	382	60	0.97	7.4	14.8	10.5	0.215
Mellapak 250.X, Metal/Plastic *	250	60	0.98	17	24.1	11.9	0.223

The Equation (73) also requires knowledge of the mean liquid film thickness (δ_f). Olujić et al. [73] proposed, based on experimental evidence, that the liquid film thickness (δ_f) is not significantly affected by the gas flow rate in the pre-loading region. Therefore, δ_f can be determined through correlations developed for liquid films in stagnant gas. If the liquid film flows in laminar regime on the packing surface, it can be estimated as

$$\delta_f = \left(\frac{3\mu_L u_{Ls}}{g\rho_L a_n \sin \theta_L} \right)^{1/3} \quad (75)$$

The *Delft* model defined the liquid hold-up (h_L) as a product of the nominal surface packing area (a_n) and the liquid film thickness (δ_f):

$$h_L = \delta_f a_n \quad (76)$$

The resistance to the mass transfer in the liquid phase is considered negligible by Olujić [72], and the penetration theory was considered valid for its estimation. The authors used the same expression proposed by Bravo et al. [8] to the liquid-side mass transfer coefficient:

$$k_x = C_L^{Delft} \sqrt{\frac{D_L u_{Le}}{\pi d_{hG} C_E^{Delft} \rho_x}} \quad (77)$$

where C_L^{Delft} is a proportionality Sherwood liquid number factor set to 2, while C_E^{Delft} (the surface renewal factor) is fixed at 0.9 as suggested by Murrieta [65].

The hydraulic diameter of triangular gas flow channel d_{hG} was used in place of the S_p [10] and d_{eq} [8]. The expression provided by the authors for this parameter is

$$d_{hG} = \frac{\frac{(B_p H_p - 2\delta_f S_p)^2}{B_p H_p}}{\left[\left(\frac{B_p H_p - 2\delta_f S_p}{2H_p} \right)^2 + \left(\frac{B_p H_p - 2\delta_f S_p}{B_p} \right)^2 \right] + \frac{B_p H_p - 2\delta_f S_p}{2H_p}} \quad (78)$$

The last parameter to be addressed for the mass transfer estimation is the wet surface area, which was subjected to several modifications over the years. In the first version of the *Delft* model, Olujić [72] proposed that the wet surface area (a_e) cannot exceed the nominal surface area (a_n) and that in the case of uniform distribution the liquid in the bed, poor distribution occurs only at low liquid flows. Therefore, the authors obtained an empirical formulation in which the percentage of wettability of the packing is a function of the liquid flow:

$$\frac{a_e}{a_n} = \frac{(1 - \Omega_p)}{\left(1 + \frac{A}{(u_{Ls})^B} \right)} \quad (79)$$

where Ω_p [$\text{m}^3 \cdot \text{m}^{-3}$] is the volumetric fraction of packing surface area occupied by holes, A and B are the constants depending by type and size of packing.

Fair et al. [76] later compared the *Delft* and *SRP* models, concluding that the *Delft* model overestimated the effective wet areas. Following this suggestion, Olujić et al. [13] developed a revisited version of the correlation adopted by Onda et al. [6], to calculate a_e for structured packings:

$$\frac{a_e}{a_n} = (1 - \Omega_p) \left[1 - \exp \left[-1.45 \left(\frac{\sigma_c}{\sigma_L} \right)^{0.75} Re_L^{0.1} Fr_L^{-0.05} We_L^{0.2} \right] \right] \quad (80)$$

in this model, the liquid Reynolds, Weber and Froude numbers are defined as in Equations (2)–(4), while the σ_c can be evaluated from Table 2, for different construction materials.

No specific fitting parameters are required in the revisited Onda equation [6] model for a_e of the *Delft* model. This formulation has been developed both for unperforated and perforated packings and takes into account of the fraction of packing surface area occupied by holes (Ω_p). For unperforated packings, (Montz B1 series) $\Omega_p = 0$, while for the perforated packings with common size of the holes around 4 mm (e.g., Montz BSH, Koch-Glitsch Flexipac and Sulzer Mellapak packings), the perforated fraction is generally in the range of 10–15% ($\Omega_p = 0.1$ – 0.15).

In addition, Olujić et al. [13] improved the Equation (80) by adding a correction term to account for packing inclination angle other than 45°:

$$\frac{a_e}{a_n} = (1 - \Omega_p) \left[1 - \exp \left[-1.45 \left(\frac{\sigma_c}{\sigma_L} \right)^{0.75} Re_L^{0.1} Fr_L^{-0.05} We_L^{0.2} \right] \right] \left(\frac{\sin 45^\circ}{\sin \theta_L} \right)^n \quad (81)$$

in which the exponent n accounts for the observed loss of effective area. The authors provided the following expression for this parameter:

$$n = \left(1 - \frac{a_n}{250} \right) \left(1 - \frac{\theta_L}{45} \right) + \left(0.49 - \sqrt{\frac{1.013}{P_{op}}} \right) \left(1.2 - \frac{\theta_L}{45} \right) + \text{Ln} \left[1 - e^{-1.45 \left(\frac{\sigma_c}{\sigma_L} \right)^{0.75} Re_L^{0.1} Fr_L^{-0.05} We_L^{0.2}} \right] \quad (82)$$

In Equation (82), P_{op} [bar] is the operating pressure; the specific surface area of 250 m²·m⁻³, the corrugation angle of 45° and the atmospheric pressure are taken as reference parameters. The correction term on the right hand-side of Equation (81), with the exponent n described by the above expression, reduces the size of effective area predicted by Equation (80) to the extent corresponding to the observed effects in the experiments by Olujić et al. [13].

The authors considered total reflux distillation data obtained with different Montz packings, i.e., B1-250.60, B1-400 and B1-400.60 model, using cyclohexane/n-heptane system at 1.03 bar. The model showed that calculated *HETPs* correlated experimental ones within errors of ±12%.

2.7. Hanley and Chen, 2012 (The HC Model)

Hanley and Chen [14] reviewed the performance of a few of the more commonly used packed-column mass-transfer correlations when they are used to predict the Height of Equivalent to a Theoretical Plate (*HETP*, [m]) for binary separations (i.e., Ar/O₂, p/o-Xylene, Chlorobenzene/Ethylbenzene, i-Cyclobutane/n-Butane, Cyclohexane/n-Heptane and cis-Decalin/trans-Decalin) using Flexipac and Mellapak type packings. In particular, they focused on several packed-column mass-transfer/interfacial area correlations found in commercially available simulation software like Aspen Technology's Aspen Rate Based Distillation component [77]. The results demonstrated that predicted *HETPs* are more than 20% above or below the experimental results [14]. Consequently, they used a new data fitting procedure related to distillation and acid gas absorption with amines operations in order to develop a set of dependable and dimensionally consistent correlations for the mass transfer related quantities k_x , k_y and a_e available for metal Pall rings, metal IMPT, sheet metal structured packings of Mellapak type and metal gauze structured packings in the X configuration.

Concerning the mass-transfer coefficient for the gas phase (k_y), the authors derived a correlation from the classical hydraulic analogy proposed by Chilton and Colburn [78] which is reasonably accurate for flows in which no form drag is present:

$$k_y = C_G^{HC} Re_G^\beta Sc_G^{1/3} \left(\frac{\rho_y D_G}{d_h} \right) \quad (83)$$

where C_G^{HC} and β are model fitting parameters (listed in Table 7), Sc_G is the Schmidt gas number given by Equation (9), d_h [m] is the hydraulic diameter given by Equation (40), Re_G is the Reynolds gas number, whose expression is reported below:

$$Re_G = \frac{d_h \rho_G u_{Gs}}{\mu_G} \quad (84)$$

where u_{Gs} [m·s⁻¹] is the superficial gas velocity. It is important to underline that in this model, the Reynolds number for the gas phase (Re_G) is linearly dependent on the gas-phase superficial velocity (u_{Gs}), and the gas-phase effective velocity is not required. In particular, in Equation (83) β is the functional parameter for the Reynolds gas number, Re_G .

According to the abovementioned analogy, the random packings are considered to have open structures; therefore, form drag should be small and the friction factor is found to be weakly dependent on the Reynolds number. Consequently, the gas-side mass transfer coefficient (k_y) is a linear function of the Reynolds number ($\beta = 1$).

Table 7. Liquid-side and gas-side characteristic parameters in the HC model for different metal packings [14].

Packing Type	C_G^{HC}	β	γ	C_L^{HC}	α
Pall rings	0.00104	1.0	-	1.0	-
IMTP	0.00473	1.0	-	1.0	-
Mellapak	0.0084	1.0	-3.072	0.33	4.078
Sulzer X	0.3516	0.5	-	12	-

For the liquid-side mass transfer coefficient (k_x), the authors considered that the induced shear at the liquid interface due to the turbulent counter-current flow of the gas interferes with the path of the liquid film in most packed columns. There is much less general consensus for the liquid-film mass transfer coefficient correlation under this case. Two different models are often chosen to describe mass transfer phenomena in a turbulent liquid film, which are the penetration/surface renewal model and the film model [56,79]. According to these theories, the Sherwood liquid number (Sh_L) is a function of two dimensionless numbers, which are the liquid Reynolds (Re_L) and Schmidt (Sc_L) numbers; the former is showed below, whereas the latter is given by Equation (6).

$$Re_L = \frac{u_{Ls}\rho_L d_h}{\mu_L} \quad (85)$$

However, Chilton and Colburn proved that the functional influence of the Schmidt liquid number (Sc_L) can generally approximated by a 1/3 power function [78]. Instead, for the Reynolds liquid number (Re_L), Potnis and Lenz [80] studied liquid desiccant systems for gas drying using random as well as structured packings reporting that the exponent of Re_L ranged from 0.9 to 1.2: for this reason, it was set by authors to 1. The liquid-side mass transfer coefficient (k_x) is given by the following expression:

$$k_x = C_L^{HC} Re_L Sc_L^{1/3} \left(\frac{\rho_x D_L}{d_h} \right) \quad (86)$$

in which C_L^{HC} is a model fitting parameters (listed in Table 7), and d_h [m] is the hydraulic diameter, which is given by Equation (40).

For sheet metal structured packings (i.e., Mellapak type), there is an additional effect of the corrugation angle both in the gas-side (k_y) and liquid-side (k_x) mass transfer coefficients. For the packings abovementioned, the authors considered adjusting factors $F_{\theta,G}$ and $F_{\theta,L}$, respectively, which are dependent on the corrugation angle (θ_c , [°]):

$$F_{\theta,G} = \left(\frac{\cos \theta_c}{\cos 45^\circ} \right)^\gamma \quad (87)$$

$$F_{\theta,L} = \left(\frac{\cos \theta_c}{\cos 45^\circ} \right)^\alpha \quad (88)$$

where γ and α are the exponents for the gas-side ($F_{\theta,G}$) and the liquid-side ($F_{\theta,L}$) mass transfer dependence on crimp inclination angle (θ_c , [°]), which are set to -3.072 and 4.078 for Mellapak type packings, respectively.

Finally, for the evaluation of the effective wet surface (a_e), the authors proposed a dependence upon Reynolds gas and liquid numbers (Re_G and Re_L), Froude liquid number (Fr_L) and Weber liquid number (We_L). The last two dimensionless parameters Fr_L and We_L are given by the following equations, respectively.

$$Fr_L = \frac{u_{Ls}^2}{gd_h} \quad (89)$$

$$We_L = \frac{u_{Ls}^2 \rho_L d_h}{\sigma_L} \quad (90)$$

The effective wet surface area (a_e) is determined using Equation (91), where η , κ , λ , ν , χ , ω and ψ are model fitting parameters listed in Table 8 for metal Pall rings, IMTP random packings, sheet metal and gauze metal packings.

$$\frac{a_e}{a_n} = C_m^{HC} \eta Re_G^\kappa Re_L^\lambda We_L^\nu Fr_L^\chi \left(\frac{\rho_G}{\rho_L}\right)^\omega \left(\frac{\mu_G}{\mu_L}\right)^\psi \quad (91)$$

Table 8. Effective wet surface area characteristic parameter in the *HC* model for different metal packings [14].

Packing Type	η	κ	λ	ν	χ	ω	ψ
Pall rings	0.25	0.134	0.205	0.075	−0.164	−0.154	0.195
IMTP	0.332	0.132	−0.102	0.194	−0.2	−0.154	0.195
Mellapak	0.538	0.1455	−0.1526	0.2	−0.2	−0.033	0.090
Sulzer X	2.308	−0.274	0.246	0.248	−0.161	−0.180	0.233

In particular, η is a proportionality coefficient for the wet surface area (a_e); κ , λ , ν and χ represent the wet surface area functional parameters for Reynolds gas and liquid numbers, Weber liquid number and Froude liquid number, respectively; ω represents the wet surface area dependence parameter on the gas to liquid density ratio; ψ represents the wet surface area dependence parameter on the gas to liquid viscosity ratio; C_m^{HC} is a correction factor related to construction material. The authors specified that this factor (C_m^{HC}) is equal to 1 for metal packings and 0.75 for plastic ones. Further, no adjustment is made to account for the expected mass transfer improvement of Y type over X type ones.

Lastly, they showed that the interfacial area participating in mass transfer can be greatly in excess of the geometrical surface area of the packing itself.

Hanley and Chen [14] showed that Bravo et al. [8], Bravo et al. [10] and Billet and Schultes [11] models gave calculated *HETPs* which are correlated the experimental values within an error of $\pm 22\%$, $\pm 24\%$ and $\pm 25\%$, respectively. Using a new data fitting procedure [14], obtained higher accuracy, with errors of about $\pm 10\%$ for a wide range of chemical systems and column operating conditions, including distillations as well as gas capture with amines.

3. Final Considerations and Models Refinement

3.1. Model Comparison and Field of Application

The prediction models for mass transfer rates in packed towers examined in the previous section have been developed in the last 50 years and are based on different theoretical approaches and experimental conditions. Table 9 shows the field of application of each model in use for different packing/application together with the different operating conditions, the number of fitting model parameters required and the estimated model prediction error with respect to the experimental values of $k_x a_e / k_y a_e$ or *HETPs*.

Table 9. Summary of the validity and applicability ranges of the correlations examined in this Section, based on packing/application, experimental conditions, number of fitting parameters and estimated error adopted by the authors for their formulations.

Models	Application	Column Size		Operating Conditions				Packing Type	Error	Fitting Param
		D, m	Z, m	P, atm	T, K	$F_G, Pa^{0.5}$	$F_L, m/h$			
<i>OTO</i>	Absorption/Desorption	0.06–0.1	0.1–0.3	1.0	293–298	0.75–2.95	up to 295	Raschig rings, Berl saddles, Spheres, Rods	$\pm 30\%$ ¹	2
<i>BRF</i>	Distillation	0.43	3.0	0.33–4.14	334–427	0.6–3.2	9.35	Sulzer BX	47% ² 8.0% ³	2
<i>SRP</i>	Distillation Absorption	0.43	3.0	0.33–20.4	334–427	0.2–3.6	9.35	Sulzer BX, Gempak 2A, Gempak 2AT, Intalox 2T, Flexipac 2Y, Maxpak, Mellapak: 250Y, 350Y, 500Y	$\pm 24\%$ ⁴	4
<i>BS</i>	Distillation Absorption/Desorption	0.06–1.4	0.15–3.95	0.033–1.0	288–407	0.01–2.77	up to 118.20	See Table 4	$\pm 8.3\%$ ⁵ $\pm 12.4\%$ ⁶	2
<i>BP</i>	Absorption/Desorption	0.05–1.0	0.42–1.89	1.00	298	0.5–3.1	1.2–79.2	Sulzer BX, Mellapak: 125Y, 250Y, 500Y	$\pm 15\%$ ⁵ $\pm 19\%$ ⁶	4
<i>Delft</i>	Distillation	0.2–1.4	3.4–6.0	0.33–4.14	334–427	0.5–4.0	9.0–35	Montz: B1-250, B1-400 B1-250.60 B1-400.60 BSH-400 BSH-400.60	$\pm 12\%$ ⁴	3
<i>HC</i>	Distillation Absorption/Desorption	See <i>BRF</i> , <i>SRP</i> and <i>BS</i> model							$\pm 10\%$ ⁴	10 (random) 11/12 (structured)

Note: ¹ refers to the range errors of the two coefficients; ² refers to the average error with respect to the experimental *HETPs* using original a_c equation; ³ refers to the average error with respect to the experimental *HETPs* using revisited a_c equation; ⁴ refers to the range error with respect to the experimental *HETPs*; ⁵ refers to the range error for k_x coefficient; ⁶ refers to the range error for k_y coefficient.

The most dated and used correlations in the textbooks are the models of Onda et al. [6] for random packing and Bravo et al. [8] for structured packing. The Onda model was tested with Raschig rings, Berl saddles, Spheres and Rods in different sizes and materials in absorption and desorption processes, while Bravo model was characterized only with Sulzer BX gauze-type structured packing in distillation applications. The correlations of Onda need two fitting parameters for the gas-side and liquid-side coefficient and provide a very high error from experiments in the range $\pm 30\%$. Moreover, the correlation of Bravo requires only two parameters for the coefficients and even if fluid-dynamics parameters, i.e., the liquid hold-up and film thickness, are not taken into account, the Bravo model records an average error of approximately 8% from experimental *HETP* values using the revised equation for the interfacial area. Despite this, on the other hand, the field of applicability for this model is very limited.

The correlations of Bravo et al. [10] or *SRP* and Billet and Schultes [11] are valid for different types of packing. In particular, the *SRP* model has been tested in distillation applications, also for column pressure higher than 1 atm, for a fair number of structured packing (Koch-Glitsch, Sulzer, Jaeger and Norton) greater than the previous version, while Billet and Schultes characterized a large number of packings both random and structured type in different sizes/materials (see Table 4) for distillation and absorption/desorption processes. The Billet and Schultes model thanks to a large number of experiments and used packings, which makes the strength of this correlation, turns out to be quite reliable with average errors of 8.3% for the liquid-side and 12.4% for the gas-side coefficient, providing fairly easy formulations that require only the use of two fitting parameters for the coefficients (k_y and k_x). It should be noted that when used for structured packings, only the void fraction and nominal surface area of the packing data are needed, unlike the other correlations which require other characteristic dimensions of the packing. On the contrary, the *SRP* model requires the use of four fitting parameters, one of which for the interfacial area. Generally, the model equations proposed for a_e calculation are not calibrated through the use of fitting parameters because the interfacial area is difficult to measure experimentally, and in fact, a calibration procedure on the coefficients $k_x a_e$ and $k_y a_e$ is preferred, introducing fitting parameters in the equations for the gas-side (k_y) and liquid-side (k_x). However, despite the authors' efforts to revise the previous version, this model provides an error of about $\pm 24\%$ considering the experimentally measured *HETP* values. Furthermore, among the other correlations examined, the *SRP* model couples a predictive model for pressure drops in the mass transfer model to calculate the variation in liquid hold-up and film thickness with an iterative algorithm. This complication makes this correlation more complex in use but on the other hand allows to estimate pressure drops and mass transfer coefficients simultaneously.

The new generation correlations proposed by Brunazzi and Paglianti [12] and Olujić et al. [13] or *Delft* model are very accurate and complex formulations compared to the previous models but are valid only for specific types of structured packing and applications. Brunazzi and Paglianti tested Mellapak Y series and BX structured packing by Sulzer in plastic and metal material for absorption and desorption applications while Olujić characterized the Montz metal packing class by Koch-Glitsch in different types (nominal areas and corrugation angles) for distillation applications also for column pressure higher than 1 atm. The use of different theoretical approaches compared to two-films and penetration theories makes these correlations more complex to use because several physical, geometric and fluid-dynamic parameters have to be calculated. The *Delft* model requires the use of three fitting parameters and provides errors of approximately $\pm 12\%$ with respect to the experimentally calculated *HETP* values. While the Brunazzi and Paglianti model requires the use of four fitting parameters, of which three only for the liquid-side coefficient, furthermore the correlation for k_x is based on an iterative algorithm on the height of the packing column which significantly increases the computational efforts required. Despite this, the errors found compared to the experiments are $\pm 15\%$ for the liquid-side and $\pm 19\%$ for the gas-side coefficient.

Among the new generation correlations, the model of Hanley and Chen [14] is probably the most reliable one since it is based on a set of equations containing dimensionless groups, fitting parameters and functional dependencies obtained with data fitting procedure from Bravo and Billet and Schultes experiments. This correlation based on a large number of experimental data has the benefit of being valid for distillation and absorption/desorption applications in different operating conditions and for both random and structured packing types (corrugated and gauze types) with errors of $\pm 10\%$ from experimentally calculated *HETPs* by Billet and Schultes and Bravo. Although this model appears to be the most accurate, on the other hand, a large number of fitting parameters are required (ten for random packing and eleven/twelve for structured packing). It is also worth noting that with respect to the two previous models, Hanley and Chen provide a dedicated calibration formulation for the interfacial area as in the *SRP* model but using seven fitting parameters.

Despite the efforts made up to the last decade by the authors to improve the correlations examined in this work, further refinements are required to achieve a definitive model equation able to overcome the problems related to the liquid distribution in packed towers, which plays a key role in the mass transfer phenomena by modifying the mass transfer coefficients and the interfacial packing area. To this end, a better description of the fluid-dynamics and mass transfer behavior in multiphase flow processes is needed through a correct estimation of some fluid-dynamics parameters (i.e., the liquid film thickness and hold-up and the interfacial surface area) that are considered the most controversial and debated parameters in the authors' vision.

Thanks to the development of new methodologies to support scientific research, novel experimental and modelling approaches have been proposed to overcome the limitations posed by the evaluation of these fluid-dynamics parameters through large-scale experiments and empirical or semi-empirical equations, with the final goal of improving the accuracy of existing predictive models. A short review of the most acknowledged and promising of these methodologies are described in the following paragraph.

3.2. New Insights on the Characterization of Liquid Distribution in Packed Columns

In the last decades, the most promising experimental and modelling approaches that have been proposed to support liquid-gas mass transfer modelling in packed towers are the optical technologies (e.g., tomography) and numerical CFD simulations.

Different tomographic techniques have been adopted in the literature, such as X-ray and Gamma-ray computed tomography (CT), electrical capacitance tomography (ECT) and electrical resistance tomography (ERT). The tomography allows to evaluate the gas-liquid distributions in packed columns as well as the liquid hold-up, the film thickness and the interfacial area.

Doan and Lohi [81] measured the liquid distribution and liquid velocity in the packed bed using electrical resistance tomography (ERT). ERT method was able to capture the effect of liquid channeling, liquid hold-up and the wall flow on the actual liquid flow in a packed bed. The authors observed a radial and axial liquid maldistribution as well as liquid channeling in a trickle flow mode and the calculated liquid residence time was about 1.5–2 times higher than that measured by the ERT method.

Wehrli et al. [82] using X-ray computed tomography (CT), provided liquid distribution, interfacial area and liquid-hold-up in columns packed with Mellapak 250.Y and 500.Y, using water and isopropanol as tracer fluids. The authors agreed with the simulation data in CFD from Olenberg and Kenig [83] for a packing with comparable geometric features that showed a similar distribution of liquid. Some differences may be explained by the absence of fine surface texture in their model.

Wu et al. [84] used electrical capacitance tomography (ECT) to analyze the distribution of a liquid-phase of a counter-current gas-liquid packed column with Mellapak 250.Y and to quantify the liquid hold-up. The authors found that local liquid hold-up values show reasonable agreement with both global liquid hold-up values measured experimentally

and also with empirical correlations from literature. The ECT tomography method could be suitable for packed columns in the industrial field service by providing in situ liquid distribution and local liquid hold-up measurements and using them to set a more accurate real-time liquid distribution [84].

Green et al. [85] adopted X-ray computed tomography (CT) to study liquid distribution, liquid hold-up and interfacial area in packed columns with structured packings and in different operating conditions of the liquid and gas load. This method can be adopted to validate results collected by traditional methods improved the prediction models, or other novel approaches with CFD simulations. Indeed, also the use of numerical simulations can provide a significant gain in time and could limit the number of experiments. Another reason to use CFD is the possibility of accessing information on a local scale which is not measurable with experimental methods. Thus, this method can provide a comprehensive and descriptive insight about governing mechanisms of fluid-flow inside packed systems and reveal qualitative/quantitative effects of the process variables. In recent years, the Volume Of Fluid (VOF) CFD approach [86] has gained more and more attention to investigate on liquid paths and the distribution along the packed column and estimate some fluid-dynamics parameters, i.e., the interfacial area, liquid hold-up and film thickness in order to calculate the mass transfer coefficients.

Recently, Haroun et al. [87] applied a VOF method to investigate the interfacial area and liquid hold-up for the Mellapak 250.X structured packing. In the simulations, the values of liquid-solid contact angles were varied to reduce the geometrical imperfections of the packing surface. The results showed significant variations of the interfacial area depending on the characteristics of the liquid-solid contact angle and the liquid flow rate. The comparison between the CFD interfacial area and the experimental data of Tsai et al. [37] showed that the best agreement with experimental data was obtained with a static angle of 10° , the deviation being about 20%. For the liquid hold-up prediction, the authors obtained good agreement between CFD and literature modelling [88].

Lassauce et al. [89] evaluated the liquid film thickness from CFD simulations and then calculated the liquid hold-up by applying the model of Bravo [8]. Optimal agreement with experimental data was observed for small-scale plants operated under laminar flow conditions.

Haroun and Raynal [88] calculated the liquid-side and gas-side coefficient through CFD simulations applying a VOF method using the Mellapak 250.X packing and compared their results with predictive models and data from the literature. The numerical liquid-side mass transfer coefficients were in very good agreement with the Higbie theory [56], and also matched the correlation of Brunazzi and Paglianti [12] with an accuracy of 75%. The gas-side mass transfer coefficient results described the model and experimental data provided by Wang [38] with an accuracy within 90%.

Macfarlan et al. [90] used the Simcenter STAR-CCM + 2019.3.1 CFD software to simulate the SO₂ scrubbing with concentrated aqueous NaOH in a Mellapak 250.Y column, varying the gas flow rate. The authors showed that the dependence of the gas-side mass transfer coefficients on the gas flow rate agrees very well with the experimental data, with errors in the range of 3%. These predictions are better than the results obtained with different semi-empirical correlations, such as Bravo et al. [10] and Olujic et al. [13].

Finally, it is worth noting that CFD application revealed significant limitations for random packings, caused by the huge scale difference between the numerical liquid flow resolution and large representative computational domains and to the treatment of interfacial mass transfer in multi-phase systems with intensive phase contact [1]. To overcome scale limitations and difficulties regarding species mass transfer, Salten and Kenig [91] proposed a novel 3D Eulerian-Eulerian model based on the concept of hydrodynamic analogies. Under this approach, the complex fluid dynamics inside the real packing is governed by a combination of simplified film, jet and droplet flows, with the gas phase in counter-current flow. The packed bed is represented by a bundle of tubular channels with a diameter equal to the hydraulic diameter of the packing. Mixing points are incorporated in

the model, their positions derived from the actual packing geometry. The mixing points are important elements of the model representing the redirection of the liquid and gas flows as well as formation and merging of jets and droplets. The model is used to simulate the reactive absorption of CO₂ from air into aqueous solutions containing sodium hydroxide under three different operating conditions. The simulation study highlighted the influence of random packing geometry on separation efficiency. It is found that a small hydraulic diameter, an increased frequency of liquid film mixing and an increased liquid hold-up lead to higher absorption rates. In contrast, the frequency of gas or jet/droplet mixing has a negligible influence on the absorption rate. These performed “virtual experiments” bring about useful recommendations for the development of random packings. The method can be also extended to support the design and optimization of structured packings [1,91] to understand the impact that the structured packing geometry, physical parameters of fluids and the shape of flow have on physical and reactive mass transfer phenomena.

4. Conclusions

This work reviewed a number of predictive models for mass transfer coefficients and wet interfacial area for packed columns equipped with both random and structured packing. The paper describes the most recognized models reported in the scientific literature [6,10–14] and includes specific data on the geometric and model fitting parameters found by the authors. The models are also scrutinized according to their range of validity and their accuracy in describing experimental data purposely selected by the authors.

The model equations mostly rely on a mix of theoretical modelling and semiempirical approaches. The two-films and penetration theories have been used extensively for most of the works, while the use of mixing factor for liquid-side coefficient was adopted by Brunazzi and Paglianti and the Chilton and Colburn analogy was used by Olujić in the *Delft* model to estimate gas-side coefficients. More recently, Hanley and Chen performed a dimensional analysis of mass transfer coefficient based on the model equations available in ASPEN PLUS software. From the analysis of the pros and cons of the correlations examined, the Billet and Schultes model seems to be the most reliable one, thanks to its wide range of validity and applicability to a large number of packing applications and the low errors of estimation (equal to 8.3% for the liquid-side and 12.4% for the gas-side coefficient), despite the use of only two fitting parameters.

While unavoidably suffering for the geometric constraints posed by the specific packing considered in their studies and, to a lower extent, to the size of the test plant, the modelling efforts made available in the pertinent literature by a number of authors had the undiscussed merit of having discovered the key physical variables and packing geometry characteristics which influence the mass transfer coefficients on both the gas and the liquid-side. The parameters that are definitely more controversial in the authors' visions are the liquid hold-up, the liquid film thickness and the interfacial surface area, which are strictly related, and the definition of a characteristic dimension of the packing (i.e., of a hydraulic diameter).

To overcome the problems related to the indirect estimation of film thickness, liquid hold-up and the interfacial surface area, several studies have proposed alternative ways to measure these parameters. Among them, probably, the most interesting results derive from the use of optical technologies, e.g., tomography. Similarly, several researchers have performed Computational Fluid Dynamic (CFD) studies to estimate the flow characteristics in packed columns in reference conditions.

These efforts testify how alive is the interest in estimating the mass transfer coefficients for packing towers and its relevance as a fundamental tool both for process designer and for specialist developer of packing and internal equipment. In spite of the current efforts, the availability of a definitive model able to predict the mass transfer coefficients for a generic packing geometry is still far to come.

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List of Symbols

a	Proportionality coefficient for the Sherwood liquid number in the <i>BP</i> model, [-]
A	Constant depending by type and size packing in <i>Delft</i> model, [-]
a_e	Wet effective surface area of packing, [$\text{m}^2 \cdot \text{m}^{-3}$]
a_n	Nominal surface area of packing, [$\text{m}^2 \cdot \text{m}^{-3}$]
b	Functional parameter for Graetz liquid number in the <i>BP</i> model, [-]
B	Functional parameter for superficial liquid velocity in <i>Delft</i> model, [-]
B_p	Base width of a packing corrugation, [m]
c	Functional parameter for Kapitza liquid number in the <i>BP</i> model, [-]
C_E^{SRP}	Surface renewal factor of the packing in the <i>BRF</i> model, [-]
C_E^{Delft}	Surface renewal factor of the packing in the <i>Delft</i> model, [-]
C_G^{BP}	Gas proportionality factor in the <i>BP</i> model, [-]
C_G^{BRF}	Gas proportionality factor in the <i>BRF</i> model, [-]
C_G^{BS}	Gas-side specific constant in the <i>BS</i> model, [-]
C_G^{Delft}	Gas-side proportionality coefficient for laminar flow case in the <i>Delft</i> model, [-]
C_G^{HC}	Gas proportionality factor in the <i>HC</i> model, [-]
C_G^{SRP}	Gas proportionality factor in the <i>SRP</i> model, [-]
C_G^{OTO}	Gas proportionality model factor in the <i>OTO</i> model, [-]
C_L^{BP}	Liquid-side proportionality model factor in the <i>BP</i> model, [-]
C_L^{BRF}	Liquid-side proportionality model factor in the <i>BRF</i> model, [-]
C_L^{BS}	Liquid-side proportionality model factor in the <i>BS</i> model, [-]
C_L^{HC}	Liquid-side proportionality model factor in the <i>HC</i> model, [-]
C_L^{SRP}	Liquid-side proportionality model factor in the <i>SRP</i> model, [-]
C_L^{OTO}	Liquid-side proportionality model factor in the <i>OTO</i> model, [-]
C_m^{HC}	Correction factor related to construction material, [-]
d	Characteristic dimension of the liquid film, [m]
d_{eq}	Equivalent diameter, [m]
D	Column diameter, [m]
D_G	Gas diffusivity in the gas phase, [$\text{m}^2 \cdot \text{s}^{-1}$]
d_h	Hydraulic diameter, [m]
d_{hG}	Hydraulic diameter of triangular gas flow channel, [m]
D_L	Gas diffusivity in the liquid phase, [$\text{m}^2 \cdot \text{s}^{-1}$]
d_p	Diameter of a sphere possessing the same surface are as a piece of packing, [m]
Fr_L	Froude liquid number, [-]
F_{SE}	Surface enhancement factor in the <i>SRP</i> model, [-]
F_t	Correction factor for total hold-up due to effective wetted area in the <i>SRP</i> model, [-]
$F_{\theta,G}$	Gas-side mass transfer coefficient dependence on crimp inclination angle, [-]
$F_{\theta,L}$	Liquid-side mass transfer coefficient dependence on crimp inclination angle, [-]
g	Acceleration of gravity, [$\text{m} \cdot \text{s}^{-2}$]
g_{eff}	Effective acceleration of gravity, [$\text{m} \cdot \text{s}^{-2}$]
Gr_L	Graetz liquid number, [-]
H	Flow distance, [m]
$HETP$	Height equivalent to a theoretical plate, [m]
h_L	Volumetric liquid hold-up, [$\text{m}^{-3} \cdot \text{m}^{-3}$]
H_p	Peak height of a packing corrugation, [m]

h_{pe}	Height of structured packing element unit, [m]
Ka_L	Kapitza liquid number, [-]
K_1	Parameter for packing type dependence, [-]
k_{hL}	Proportionality factor for liquid hold-up in <i>BP</i> model, [-]
k_x	Liquid-side mass transfer coefficient per surface unit, [$\text{kmol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$]
k_y	Gas-side mass transfer coefficient per surface unit, [$\text{kmol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$]
$k_{y,lam}$	Gas-side mass transfer coefficient for laminar regime, [$\text{kmol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$]
$k_{y,turb}$	Gas-side mass transfer coefficient for turbulent regime, [$\text{kmol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$]
$l_{G,pe}$	Length of the triangular gas flow channel in a packing element, [m]
n	Correction exponent for the effective area in <i>Delft model</i> , [-]
P_{op}	Operating pressure, [bar]
P_s	Perimeter per unit cross-sectional area, [m]
Re_G	Reynolds gas number, [-]
Re_{Grv}	Reynolds gas number based on relative effective velocity between gas and liquid, [-]
Re_L	Reynolds liquid number, [-]
Sc_G	Schmidt gas number, [-]
Sc_L	Schmidt liquid number, [-]
Sh_L	Sherwood liquid number, [-]
$Sh_{G,lam}$	Sherwood gas number for laminar flow, [-]
$Sh_{G,turb}$	Sherwood gas number for turbulent flow, [-]
S_p	Slant height of a packing corrugation, [m]
t_e	Exposure time, [s]
t_G	Gas contact time, [s]
t_L	Time necessary for renewal of interface area, [s]
u_{Ge}	Gas effective velocity through the packing channel, [$\text{m}\cdot\text{s}^{-1}$]
u_{Le}	Liquid effective velocity through the packing channel, [$\text{m}\cdot\text{s}^{-1}$]
u_{Gs}	Superficial gas velocity, [$\text{m}\cdot\text{s}^{-1}$]
u_{Ls}	Superficial liquid velocity, [$\text{m}\cdot\text{s}^{-1}$]
We_L	Weber liquid number, [-]
Z	Packing height, [m]

Greek Symbols

α	Liquid-side mass transfer coefficient dependence on crimp inclination angle in the <i>HC</i> model, [-]
β	Functional parameter for Reynolds gas number in the <i>HC</i> model, [-]
γ	Gas-side mass transfer dependence on crimp inclination angle in the <i>HC</i> model, [-]
Γ	Liquid flow per unit length of perimeter, [$\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$]
γ_c	contact angle accounts for surface material wettability, [°]
δ_f	Liquid film thickness, [m]
$\Delta P/Z$	Total pressure drops per meter of packing, [$\text{Pa}\cdot\text{m}^{-1}$]
$\Delta P/Z_{flood}$	Pressure drops per meter of packing at flooding condition, [$\text{Pa}\cdot\text{m}^{-1}$]
ε_p	Void volumetric fraction of the packing, [$\text{m}^{-3}\cdot\text{m}^{-3}$]
ζ_{GL}	Interaction coefficient for gas-liquid friction losses in the <i>Delft</i> model, [-]
η	Proportionality coefficient for the wet surface area in the <i>HC</i> model, [-]
θ_c	Inclination or corrugation angle, [°]
θ_L	Slope of the steepest descent line with respect to the horizontal axis, [°]
κ	Functional parameter for Reynolds gas number in the <i>HC</i> model, [-]
λ	Functional parameter for Reynolds liquid number in the <i>HC</i> model, [-]
μ_G	Mass gas viscosity, [$\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$]
μ_L	Mass liquid viscosity, [$\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$]
μ_{Lo}	Dynamic viscosity of water at 20 °C, [$\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$]
ν	Functional parameter for Weber liquid number in the <i>HC</i> model, [-]
ρ_G	Mass gas density, [$\text{kg}\cdot\text{m}^{-3}$]
ρ_y	Molar gas density, [$\text{kmol}\cdot\text{m}^{-3}$]
ρ_x	Molar liquid density, [$\text{kmol}\cdot\text{m}^{-3}$]
ρ_L	Mass liquid density, [$\text{kg}\cdot\text{m}^{-3}$]
σ_c	Critical surface tension of packing material, [$\text{N}\cdot\text{m}^{-1}$]
σ_L	Liquid surface tension, [$\text{N}\cdot\text{m}^{-1}$]
φ	Fraction of the triangular flow channel occupied by liquid, [-]
χ	Functional parameter for Froude liquid number in the <i>HC</i> model, [-]
ψ	Wet surface area dependence parameter on the gas to liquid viscosity ratio in the <i>HC</i> model, [-]
ω	Wet surface area dependence parameter on the gas to liquid density ratio in the <i>HC</i> model, [-]
Ω_p	Fraction of packing surface area occupied by holes, [$\text{m}^{-3}\cdot\text{m}^{-3}$]

Abbreviations

BP	Referred to the work of Brunazzi and Paglianti (1997)
BRF	Referred to the work of Bravo et al. (1985)
BS	Referred to the work of Billet and Schultes (1993)
Delft	Referred to the work of Olujić et al. (2004)
HC	Referred to the work of Hanley and Chen (2012)
OTO	Referred to the work of Onda et al. (1968)
SRP	Referred to the work of Bravo et al. (1992)

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