

Improved Mass Transfer Correlations for Random and Structured Packings

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Introduction

Rate-based calculations for trayed and packed columns would seem to offer process engineers a more rigorous and reliable basis for assessing column performance than the traditional equilibrium stage approach, especially for multicomponent separations. Unfortunately, rate-based simulations for packed columns suffer from a serious weakness – they are ultimately tied to underlying correlations for the mass transfer coefficients, k_x and k_y , as well as for the specific area participating in mass transfer, a_m . It is well-known that packing correlations available in the public domain are unreliable when they are applied to chemical systems and column operating conditions outside of those used to develop the correlations in the first place. We will develop, in this paper, reliable correlating expressions for metal Pall type rings, IMTP type random packings, and sheet metal structured packings of the MELLAPAK type for both the “Y” and “X” configurations. We will demonstrate the superior performance of these correlations for a wide range of chemical systems and column operating conditions, including standard distillation as well as acid gas capture with amines.

The Performance of Public Domain Mass Transfer Correlations

Perhaps the most straightforward way to demonstrate the limitations of several of the more popular public domain mass transfer correlations is by comparing simulation predictions with experimental measurements. Table 1 is such a comparison for binary distillation experiments with random packings. For each experimental system, we have calculated HETPs using three mass transfer correlations – Billet & Schultes¹, Bravo & Fair², and Onda, et al.³ - within the Aspen Plus *Rate Based Distillation* module. Since most distillations are vapor phase controlled the results in the table can be considered tests of the robustness of each correlation’s $k_y a_m$ predictions (we expect $K_{Oy} \approx k_y$ for such systems). Calculated HETPs more than 20% different from the measured value range are bolded and underlined. Table 2 is a similar comparison for structured packings. For each experimental system, we have calculated HETPs using three mass transfer correlations – Billet & Schultes¹, Bravo, Rocha, and Fair 1985 (BRF85)⁴, and Bravo, Rocha, and Fair 1992 (BRF92)⁵ - within the Aspen Plus *Rate Based Distillation* module. Once again, calculated HETPs more than 20% different than the measured value range are bolded and underlined. In Table 1, 2/3 of the calculated HETPs are more than 20% above or below the experimental values. In Table 2, about 40% of the predicted HETPs are outside the $\pm 20\%$ envelope.

These same correlations fail to adequately describe the mass transfer performance of systems which are controlled on the liquid side because of chemical reaction. Let us consider the experiments of Lawal, et al.⁶ for the absorption of CO₂ into aqueous MEA solutions in a column equipped with IMTP #40 random packing. These researchers describe the results of two such experiments – referred to as “case 32” and “case 47”. Figure 1 compares Lawal’s experimental findings for case 32 with predictions calculated using the Bravo & Fair and Onda mass transfer correlations for random packings within the Aspen *Rate Based Distillation* environment using the ELECTNRTL properties model. Note that the Bravo & Fair correlation comes close to matching the experimentally measured rich amine loading and the CO₂ removal from the inlet gas stream but that its column temperature profile prediction is markedly

Table 1. Comparison of Measured/Calculated HETPs for Several Random Packings*

System	Pressure (torr)	Packing	HETP (Billet)	HETP (BF82)	HETP (Onda)	HETP (Expt)
C ₆ /n-C ₇	760	2" Pall ring	<u>17</u>	<u>10</u>	28	25-28
1,2PG/EG	10	2" Pall ring	<u>25</u>	34	<u>56</u>	38
EB/SM	100	3½" Pall ring	25	25	<u>56</u>	29
EB/SM	100	1½" Pall ring	<u>23</u>	<u>10</u>	17	16-19
EB/SM	100	¾" Pall ring	16	<u>6</u>	<u>7.5</u>	11-15
p/o-xylene	50	⅝" Pall ring	16	<u>8</u>	<u>6.75</u>	14-15
i-C ₄ /n-C ₄	8533	2" Pall ring	<u>6.5</u>	<u>10</u>	21	20
i-octane/tol	760	1" Pall ring	<u>9.7</u>	<u>5.5</u>	<u>8.4</u>	16
i-octane/tol	760	2" Pall ring	<u>13</u>	<u>11</u>	<u>19.5</u>	26-28
D ₂ O/H ₂ O	350	HY-PAK #1	<u>25.5</u>	<u>5.5</u>	<u>12</u>	19

* Calculations performed with Aspen Plus v7.2 *Rate Based Distillation* module. Experimental results from Kister⁷ and Schultes⁸. In all cases the countercurrent flow model was used. All experiments at total reflux. HETPs are reported in inches.

Table 2. Comparison of Measured/Calculated HETPs for Several Structured Packings**

System	Pressure (torr)	Packing	HETP (Billet)	HETP (BRF85)	HETP (BF92)	HETP (Expt)
Ar/O ₂	1550	Flexipac 500Y	6.15	7.5	<u>8.7</u>	6.9
p/o – xylene	16	Mellapak 250Y	15	14	<u>20</u>	13
p/o – xylene	100	Flexipac 700Y	9.3	<u>3.6</u>	<u>5.8</u>	8
CB/EB	76	Mellapak 350Y	11.5	9.75	<u>13</u>	10.5
CB/EB	76	Flexipac 500Y	<u>12</u>	11	11	9
i-C ₄ /n-C ₄	8533	Flexipac 250Y	<u>4.7</u>	12.5	10	8.5-12
C ₆ /C ₇	1241	Flexipac 250Y	<u>8.6</u>	16	14	15-16
cis-/trans-decalin	304	Mellapak 250Y	<u>9.5</u>	16.5	15.5	13-14
TEG/H ₂ O/CH ₄	31030	Flexipac 250Y	<u>186</u>	<u>137</u>	<u>470</u>	66

**Calculations performed with Aspen Plus v7.2 *Rate Based Distillation* module. Experimental results from Kister⁷, Fitz, et al.,⁹ Bennett, et al.,¹⁰ and Kean, et al.¹¹ In all cases the countercurrent flow model was used. All experiments at total reflux. HETPs are reported in inches.

different from the measured profile. On the other hand, the Onda correlation matches the rich amine loading and the observed temperature profile reasonably well but its prediction of the CO₂ removal from the inlet gas stream is quite low.

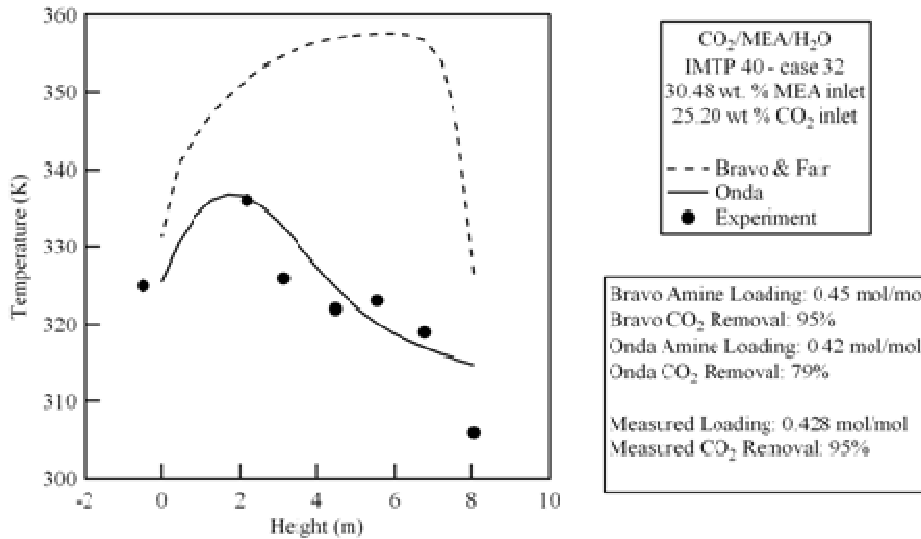


Figure 1. Calculated and measured temperature profiles for the absorption of CO₂ by aqueous MEA. Data from Lawal, et al.⁶ for case 32. Calculations performed with Aspen Plus v7.2 *Rate Based Distillation* module using the VPlug option and the ELECTNRTL properties model.

We next consider data of Shivelor, et al.¹² for the simultaneous removal of CO₂ and H₂S from a high pressure natural gas stream into an aqueous MDEA solution using MELLAPAKPLUS 252Y sheet metal structured packing. Oftentimes in natural gas processing H₂S must be removed almost completely while a certain amount of CO₂ can be allowed to “slip” into the treated gas. MDEA is a selective amine because there is a significant difference in the reaction rates between it and H₂S versus CO₂. Thus these types of experiments are sensitive tests of the suitability of the thermodynamic and kinetic models within the rate based model on the one hand and the packing mass transfer correlations used in the simulation on the other. Figure 2 is a comparison of the measured selectivity (y_{CO_2}/y_{H_2S}) in the outlet gas stream versus the predictions of the Bravo, Rocha, and Fair 1985 (BRF85) correlation for mass transfer in wire-mesh and sheet metal structured packings for three different limiting cases. We note here that the performance of the Billet and Schultes correlation and the modified Bravo, Rocha, and Fair correlation of 1992 (BRF92) are significantly worse and are therefore not included in the figure. Clearly all three mass transfer correlations prove inadequate in this case.

Improved Mass Transfer Correlations for Pall Rings, IMTP, and MELLAPAK/FLEXIPAC

Scaling and Dimensional Consistency

We start with dimensional analysis applied to the vapor phase and liquid phase mass transfer coefficients k_y , k_x , as well as to the specific mass transfer area, a_m . If $k_y = k_y(d_e, v_V, \rho_V, c_V, \mu_V, D_V)$ then dimensional consistency requires that a dimensionless group containing k_y be a function of two other independent dimensionless groups (seven parameters and four independent units of measure). This relationship is usually taken to be $Sh_V = (k_y d_e / c_V D_V) = Sh_V(Re_V, Sc_V)$. By a similar argument if $k_x = k_x(d_e, v_L, \rho_L, c_L, \mu_L, D_L)$ then $Sh_L = (k_x d_e / c_L D_L) = Sh_L(Re_L, Sc_L)$.

We now turn our attention to the specific mass transfer area, a_m . If we postulate that a_m depends upon the flow and physical properties in both phases (think about rain hitting your windshield as you’re driving during a thunderstorm) then it is reasonable to assume that

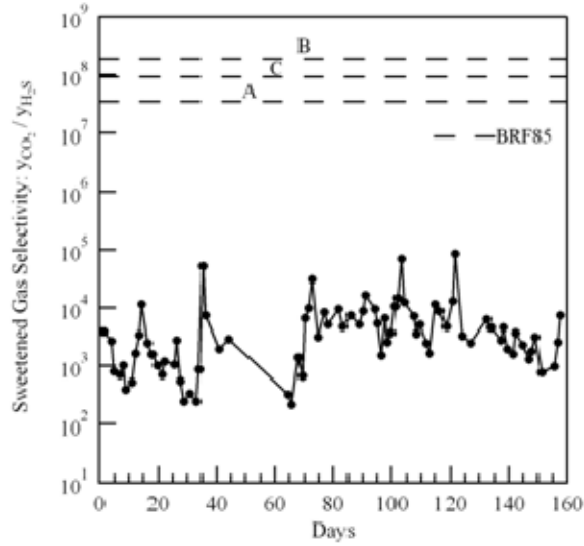


Figure 2. Calculated and measured selectivity (selectivity defined as the ratio y_{CO_2}/y_{H_2S} in the treated gas stream) for the Bravo, Rocha, Fair (BRF85) correlation for mass transfer using wire-mesh or sheet metal structured packings. Data taken from Shivelor, et al. Calculations performed with Aspen Plus v7.2 *Rate Based Distillation* module using the VPlug option and the ELECTNRTL properties model.

$$(a_m / a_d) = A_m (\rho_V, \mu_V, V_V, \rho_L, \mu_L, V_L, \sigma, d_e, g) \quad (1)$$

It is not difficult to show that a_m/a_d can be expressed thusly

$$\frac{a_m}{a_d} = A_m \left(Re_V, Re_L, We_L, Fr_L, \frac{\rho_V}{\rho_L}, \frac{\mu_V}{\mu_L} \right) \quad (2)$$

It is the appearance of the groupings ρ_V/ρ_L and μ_V/μ_L , required for dimensional consistency in the expression for a_m/a_d , that are new to this development.

The Defining Expression for the HETP at Total Reflux

Since most distillation experiments carried out to measure a packing's HETP in a binary system are carried out at total reflux we consider only that case here. If we assume that the relationships for k_y , k_x , and a_m are all power laws then the defining expression for the column average HETP is

$$\langle HETP \rangle = \frac{G d_e}{\left(A_m \left(\frac{\rho_V}{\rho_L} \right)^A \left(\frac{\mu_V}{\mu_L} \right)^B (Re_L^x) (Fr_L^A) (We_L^E) (Re_V^\phi) a_d \right)} \left(\frac{C_y}{A_V Re_V^m Sc_V^n c_V D_V} + \frac{C_x}{A_L Re_L^b Sc_L^c c_L D_L} \right) \quad (3)$$

where Hanley has previously defined the parameters C_x and C_y .¹³

We immediately see that the above expression for the HETP expressed in terms of independent expressions for k_y , k_x , and a_m (opposed to expressions for the combined quantities $k_y a_m$ and $k_x a_m$) is not unique. It is possible to factor out the front factor A_L (for example) and define two new relative front factors for k_y and a_m

$$\langle \text{HETP} \rangle = \frac{G d_e}{\left(A_M' \left(\frac{\rho_V}{\rho_L} \right)^A \left(\frac{\mu_V}{\mu_L} \right)^B (\text{Re}_L^x) (\text{Fr}_L^A) (\text{We}_L^E) (\text{Re}_V^\Phi) a_d \right)} \left(\frac{C_y}{A_V' \text{Re}_V^m \text{Sc}_V^n c_V D_V} + \frac{C_x}{\text{Re}_L^o \text{Sc}_L^c c_L D_L} \right) \quad (4)$$

where $A_M' = (A_M)(A_L)$ and $A_V' = A_V/A_L$. It is thus not possible to develop an unambiguous fit for k_y , k_x , and a_m from data for the HETP alone. In light of this finding, we proceed as follows. We assume that the mass transfer coefficients are adequately described by the expressions

$$k_y = \left(\frac{A_V}{d_e} \right) \text{Re}_V^1 \text{Sc}_V^{1/3} c_V D_V \quad (5)$$

$$k_x = \left(\frac{A_L}{d_e} \right) \text{Re}_L^1 \text{Sc}_L^{1/3} c_L D_L \quad (6)$$

We can now take $A_L = 1$ and determine all fitting parameters including the values for the relative front factors A_V' and A_M' from fits to publicly available binary HETP data.

Next these complete (yet relative) mass transfer coefficient and mass transfer area correlations are used to predict the performance of the packing in some other type of experiment (for example, CO_2 absorption by aqueous amines). If the results do not match experiment then the coefficient for A_L (originally assumed to be unity) can be adjusted, leading to automatic adjustments in the values for A_V and A_M (necessary to keep the HETP fit good). Some combination of parameters A_L , A_V , and A_M will appear optimal across many experiments in the sense that the HETP will be fit well for binary distillation experiments concurrently with the temperature profile, rich amine loading, and acid gas removal for acid gas absorption into amines.

The calculations described above have been carried out for metal Pall rings, metal IMTP, and sheet metal structured packings of the MELLAPAK/FLEXIPAC type in both the "X" and "Y" configurations using publicly available HETP data

Metal Pall Rings

$$k_x = \left(\frac{1}{d_e} \right) \text{Re}_L^1 \text{Sc}_L^{1/3} c_L D_L \quad k_y = \left(\frac{0.00105}{d_e} \right) \text{Re}_V^1 \text{Sc}_V^{1/3} c_V D_V \quad (7)$$

$$\frac{a_m}{a_d} = 0.25 \text{Re}_V^{0.134} \text{Re}_L^{0.2052} \text{We}_L^{0.0752} \text{Fr}_L^{-0.164} \left(\frac{\rho_V}{\rho_L} \right)^{-0.154} \left(\frac{\mu_V}{\mu_L} \right)^{0.195} \quad (8)$$

Metal IMTP

$$k_x = \left(\frac{1}{d_e} \right) \text{Re}_L^1 \text{Sc}_L^{1/3} c_L D_L \quad k_y = \left(\frac{0.00473}{d_e} \right) \text{Re}_V^1 \text{Sc}_V^{1/3} c_V D_V \quad (9)$$

$$\frac{a_m}{a_d} = 0.3325 \text{Re}_V^{0.132} \text{Re}_L^{-0.1018} \text{We}_L^{0.194} \text{Fr}_L^{-0.2} \left(\frac{\rho_V}{\rho_L} \right)^{-0.154} \left(\frac{\mu_V}{\mu_L} \right)^{0.195} \quad (10)$$

MELLAPAK/FLEXIPAC Sheet Metal Structured Packings[†]

$$k_x = \left(\frac{0.33}{d_e} \right) \text{Re}_L^1 \text{Sc}_L^{1/3} c_L D_L \quad k_y = \left(\frac{0.0084}{d_e} \right) \left(\frac{\cos(\Theta)}{\cos(45)} \right)^{-7.152} \text{Re}_V^1 \text{Sc}_V^{1/3} c_V D_V \quad (11)$$

$$\frac{a_m}{a_d} = 0.5386 \left(\frac{\cos(\Theta)}{\cos(45)} \right)^{4.078} \text{Re}_V^{0.1455} \text{Re}_L^{-0.1526} \text{We}_L^{0.2} \text{Fr}_L^{-0.2} \left(\frac{\rho_V}{\rho_L} \right)^{-0.033} \left(\frac{\mu_V}{\mu_L} \right)^{0.09} \quad (12)$$

Results

Tables 3 and 4 are similar to Tables 1 and 2 except that HETP predictions for the newly proposed correlations above have been included. Clearly the predictions for the HETP from the new correlations are, in general, better than those from the other correlations examined.

Figure 3 contains the data of Figure 1 along with the predictions from the proposed correlations above. Note that the new correlation matches the temperature profile, the rich amine loading, and the removal of CO₂ from the gas phase quite well. Figure 4 contains the data of Figure 2 along with the predictions from the correlations proposed above. The three case studies using the new correlations here match the measured outlet gas selectivity well.

Table 3. Comparison of Measured/Calculated HETPs for Several Random Packings

System	Pressure (torr)	Packing	HETP (Billet)	HETP (BF82)	HETP (Onda)	HETP (Aspen)	HETP (Expt)
C ₆ /n-C ₇	760	2" Pall ring	<u>17</u>	<u>10</u>	28	23	25-28
1,2PG/EG	10	2" Pall ring	<u>25</u>	34	<u>56</u>	42	38
EB/SM	100	3½" Pall	25	25	<u>56</u>	<u>22</u>	29
EB/SM	100	1½" Pall	<u>23</u>	<u>10</u>	17	18	16-19
EB/SM	100	⅝" Pall ring	16	<u>6</u>	<u>7.5</u>	13	11-15
p/o-xylene	50	⅝" Pall ring	16	<u>8</u>	<u>6.75</u>	13	14-15
i-C ₄ /n-C ₄	8533	2" Pall ring	<u>6.5</u>	<u>10</u>	21	19	20
i-octane/tol	760	1" Pall ring	<u>9.7</u>	<u>5.5</u>	<u>8.4</u>	15.7	16
i-octane/tol	760	2" Pall ring	<u>13</u>	<u>11</u>	<u>19.5</u>	<u>19.7</u>	26-28
D ₂ O/H ₂ O	350	HY-PAK #1	<u>25.5</u>	<u>5.5</u>	<u>12</u>	<u>13.7</u>	19

[†] where Θ is the corrugation inclination angle (in degrees) from the vertical

Table 4. Comparison of Measured/Calculated HETPs for Several Structured Packings

System	Pressure (torr)	Packing	HETP (Billet)	HETP (BRF85)	HETP (BF92)	HETP (Aspen)	HETP (Expt)
Ar/O ₂	1550	Flexipac 500Y	6.15	7.5	<u>8.7</u>	6.93	6.9
p/o – xylene	16	Mellapak 250Y	15	14	<u>20</u>	<u>17</u>	13
p/o – xylene	100	Flexipac 700Y	9.3	<u>3.6</u>	<u>5.8</u>	6.8	8
CB/EB	76	Mellapak 350Y	11.5	9.75	<u>13</u>	10.8	10.5
CB/EB	76	Mellapak 500Y	<u>12</u>	11	11	8.8	9
i-C4/n-C4	8533	Mellapak 250Y	<u>4.7</u>	12.5	10	9.5	8.5-12
C6/C7	1241	Flexipac 250Y	<u>8.6</u>	16	14	12	15-16
cis-/trans-decalin	304	Mellapak 250Y	<u>9.5</u>	16.5	15.5	15	13-14
TEG/H ₂ O/CH ₄	31030	Flexipac 250Y	<u>186</u>	<u>137</u>	<u>470</u>	55	66

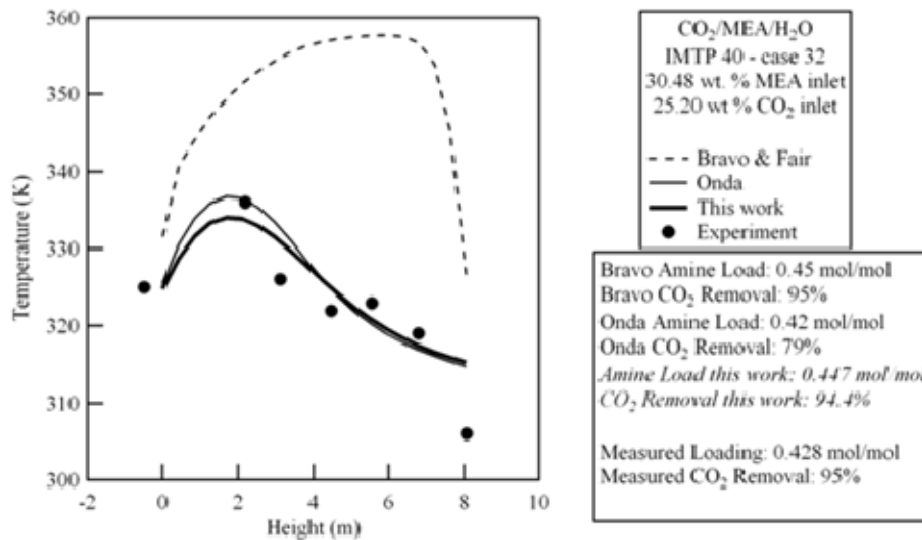


Figure 3. Calculated and measured temperature profiles for the absorption of CO₂ by aqueous MEA. Data from Lawal, et al. Calculations performed with Aspen Plus v7.2 *Rate Based Distillation* module using the VPlug option and the ELECNRTL properties model.

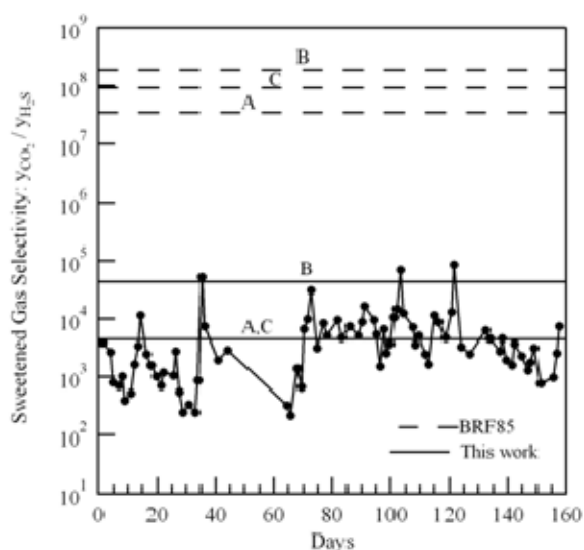


Figure 4. Calculated and measured selectivity (selectivity defined as the ratio $y_{\text{CO}_2}/y_{\text{H}_2\text{S}}$ in the treated gas stream) for the Bravo, Rocha, Fair (BRF85) correlation and the new correlation presented in this paper.

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