Predicting Effective Mass Transfer Area in Randomly-packed Column — Design Considerations

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The paper describes the performance of a randomly packed distillation column depending on the effective vapor-liquid interfacial area ($a_{el}$) and the flood ratio (per cent $Fl$) estimated by the Eckert flooding model. Capacity, pressure gradient, and efficiency data obtained on 6.25 mm size of a Raschig-type ceramic ring are used to establish the basis for the model reliability analysis. The reliability of design models is analyzed against observed performance statistically by using a log-ratio objective function. The proposed $a_{el}$ approach for random packings is expected to be an improvement in data fit, clarifying the simultaneous impact of packing geometry, loading, and system properties on the performance of a moderate scale packed column. Details underlying some aspects of selection of an appropriate packed column design algorithm are discussed. An example of scaling-up a pilot-plant with respect to the resulting statistical factors is elucidated. Finally, an approach to design in terms of the derivatives of the function $a_{el} = f$(per cent $Fl$) is interpreted analytically.

Keywords: Mass transfer, Randomly-packed column, Distillation column

1 Introduction

The scaling-up of a randomly packed distillation column depends on a reasonable choice of proper variables and operating parameters to minimize production costs. Recently, a comprehensive study of designing a packed column confirmed that four major considerations need to be specified with reliability, i.e., capacity or flood point, pressure gradient, effective interfacial area, and height equivalent to a theoretical plate (HETP). Traditionally, determination of the packed column diameter has been attributed to two approaches based on the use of either a gas flooding percentage (per cent $Fl$) estimated by a Sherwood type flooding correlation or a percentage value of pressure drop line with respect to the generalized correlations, and Leva. Many attempts have been made to predict the column capacity using the concept of the maximum efficient capacity (MEC) point. Besides considering these factors the paper discusses the efficiency of a pilot-plant for distillation in terms of two critical variables, effective area, and flood ratio. However, analysis is mainly focused on an analytical solution of conditions optimizing effective interfacial area ($a_{el}$) and HETP as a function of flood ratio estimated by the Eckert flooding model. In the design practice the efficiency behavior of packings is usually interpreted against various vapor velocities or a vapor rate dependent factor such as, flood ratio, loading factor or density-corrected capacity parameter. The flood ratio is defined as a ratio of operating vapor superficial mass velocity to the flooding one for a given constant liquid rate (per cent $Fl = 100G/G_0$). This requires quantitative knowledge of the flood velocity so that the flood ratio can be quantified. Bolles and Fair have proposed an analytical approach to design a packed column using a flood ratio fixed between 70-80 per cent flooding. Rukovena and Koshy have extended the per cent $Fl$ range to a 90 per cent limit related to the MEC point.

It is conceivable that the implementation of a packed column design strategy argues an uncoupling of effective area ($a_e$) from the volumetric mass transfer coefficient ($ka_e$) to establish a sentence structure distinguishing the divergent behaviors of $k$ and $a_e$ as the flood point is approached in a vapor phase controlled system. Hanley et al. have presented evidence of monotonical increase of the vapor phase mass transfer coefficient over the whole vapor loading range, while $a_e$ first increases with increase in vapor flux and then begins to decrease as the flood point is
approached. They have concluded that the divergence of HETP near flooding depends sensitively upon the dominating effect of the rate of loss of mass-transfer area in contrast to the rate of increase in the overall mass-transfer coefficient. As HETP is inversely proportional to both $k$ and $\alpha_e$, it turns out that the trends observed in (HETP)$^4$ at varying loading conditions should apply for effective area. Process considerations, dealing with the competition between $\alpha_e$ and $k$ about the loading point, are still challenging since such systems show extreme behaviors of changing from a stable operating regime to the flooding one by any small perturbation. Examples of successful application of a rate-based model are precise evaluation of optimum flow conditions and interfacial area, as well as effective solution of non-ideal equilibrium equations regarding the validation limits.

In this study, attempts are made to optimize a distillation plant performance by reducing a complex problem requiring simultaneous characterization of the complementary effects of functionally dependent quantities, HETP, $k$, $\lambda$, and $\alpha_e$, and their derivatives. A newly proposed log-basis approach for $\alpha_e$ of moderate-type random packings allows one to obtain criteria for improvement of the column performance. The study aims to develop a general optimization structure for distillation applications that would not require previous validation for the packing type or size. The proposed optimization structure could then be applied to numerous packing sizes and shapes while still being general with regard to the system properties and flow conditions. Additionally, a new conceptual definition for optimum $d_i$ as the locus of the statistical design factors has been proposed

1.1 Eckert Flooding Model. Effective Area

Approaches

Characterization of a packed column efficiency is intimately connected to the definitions of flooding point and effective mass-transfer area. Recently, postulating a continual thickening of the falling liquid film with increasing liquid flow and shear stress in random packings, theoretical interpretations of flooding in terms of the interactive effects of liquid holdup and pressure drop have been carried out\textsuperscript{5,6,7}. Among the proposed versions of the empirical Sherwood flooding model for gas flood velocity ($G_f$) the Eckert flooding model is still widely accepted as a reliable tool of designing packed columns, since it covers the performance of a large scale older random packings and metal Pall rings classified also as a through-flow modern packing\textsuperscript{8}. The model defined as a plot of $y = (G_f^2 F_i)^{\alpha_e}/(\mu_L/\mu_W)^{0.5}}{(gpc_p)}$ vs. $x = F_{1G} = (L/G_0)(\rho_C/\rho_L)^{0.5}$ has been interpolated into a polynomial explicit form available for analysis of a distillation plant

\[ F(G_f) = \ln[(AB^2) + 3.84445 - 0.9182] \\
\ln[B/G_f] + 0.1282[\ln(B/G_f)]^2 = 0, \tag{1} \]

where $A = F_0(M_u)^{0.5}\Psi' / (\rho_D/\rho_L)$, $B = L_1(\rho_C/\rho_L)^{0.5}$, $\Psi = \rho_D / \rho_L$, $\mu_L = 1$ mPa·s, and $F_{1G} = \alpha_e / \lambda^3$ an adjustable parameter called packing factor. $F_{1G}$ is the liquid-gas flow parameter. Eq. (1) coincides with the flooding curve for the $F_{1G}$ ranging from 0.01 to 8.0, showing 0.03 per cent mean deviation. Kessler and Wankat\textsuperscript{9} have suggested a similar approach with slightly different coefficients relating to the points and the $F_{1G}$ range selected.

A summary of four generalized methods, based on two-film model, reliable for predicting $\alpha_e$ of larger-scale packed distillation columns described earlier\textsuperscript{10} is given by Wagner et al.\textsuperscript{11}. In this study, using the transfer unit concepts of film theory ($H_0$, $H_1$, and $H_{OC}$) the mass transfer algorithm which combines the Bravo-Fair effective area model\textsuperscript{11}, as well as the Onda et al.\textsuperscript{12} wetted area ($\alpha_w$) approach with the Onda individual side mass transfer coefficients has been applied. The Bravo-Fair model for effective interfacial area ($\alpha_w$) was performed using an adjusted exponent value $r = 0.88$ of the liquid surface tension ($\sigma$) instead of 0.5 leading to an amount of the estimated effective area larger than the wetted one regarding to the Onda $\alpha_w$ prediction for a small packing ($d_i < 12.7$ mm).

\[ \frac{\alpha_w}{\alpha_p} = 0.498 \left( \frac{\sigma}{Z^{0.4}} \right) \left( \frac{C_{dL} \cdot R_e \cdot G}{\mu_L} \right)^{0.392}, \tag{2} \]

where $Z$ is the height of packed bed, $m$; $\sigma$ is the liquid surface tension, $N/m$; $C_{dL} = \mu_L / (\rho_D \cdot \sigma_p)$ is the liquid capillary number; $R_e = 6G(\alpha_w \cdot \mu_L)$ is the modified gas Reynolds number.

There is some evidence that suggests the existence of several "kinds" of area related to a packed column operation\textsuperscript{13}. It is recognized that hydrodynamic instabilities in the liquid flow due to the waves or ripples on the liquid film surface, gas
bubbling within liquid puddles, droplets, localized flooding\textsuperscript{11,13,17}, as well as the occurrence of the vertical density and viscosity gradients through the packed bed\textsuperscript{14,15} give rise to an additional amount of interfacial area beyond the wetted packing part. In fact the stationary waves with steady finite amplitudes appearing on the surface of vertically falling liquid films throughout a wall, qualified as travelling waves, are attributed to the surface tension that compensates the destabilizing effect of gravity\textsuperscript{16}.

In a preceding study\textsuperscript{8}, an explicitly formulation of dimensionless groups affecting primarily the flooding limit along with a dimensional analysis of experimental data in terms of a log-basis relation are proceeded to realize a generalized estimation method for the ratio of effective area (\(a_e\)) and its upper limit (\(a_l\)).

\[
\frac{a_e}{a_l} = a_o \left( \frac{\sigma_L / \sigma_o}{100 - \% F_i} \right)^{n} (F_i)_{n}^n
\]

\[
(Ca_L, Re_G) \left( \frac{We_L}{Re_L} \right)^{0.5} \left( \frac{\sigma}{\sigma_o} \right)^{0.5}, \quad \ldots (3)
\]

where the liquid film number, i.e. the ratio of Froude (\(F_i = a_o L^2 (\rho_o \rho_i^{\frac{1}{2}})\)) and Reynolds (\(Re_L = L/(a_o \mu_i)\)) numbers, \(F_i = Fr_i/Re_{L}\), represents the behavior attributed to the \(i/e\) group; the \((We_L/Re_L)\) term including the liquid Weber (\(We_o = L^2 (\rho_o \sigma_o)\)) and Reynolds numbers accounts for the kinematic wave character of the falling liquid film\textsuperscript{16}; the Bravais-Fair “loading term”, \(Ca_o \times Re_G\), incorporates the simultaneous effects of vapor and liquid loads. The Onda surface tension term, \(\sigma \sigma_o\), accounts for the liquid/packing surface interaction; \(\sigma_i\) is a critical liquid surface tension for a particular material: 61 N/m (ceramic), 75 N/m (steel), and 3.3 N/m (polyethylene)\textsuperscript{16}. The \(\sigma_i/\sigma_o\) group designates the binary system character. Subscripts “\(L\)” and “\(i\)” denote light and heavy components. The experimental findings reveal that the enhanced magnitude of interfacial area is highly sensitive to the range of changes of both variables \(d\), and \(Z\), thereby causing an interactive effect on \(a_e\)\textsuperscript{11,17}. Thus the influence of these variables has been incorporated into \(c_6\). Coefficients \(c_i\) of Eq. (3), obtained for three systems using multivariable procedures of linpack are given in Table I. The model satisfies two limiting conditions in accordance with the theory, i.e. if per cent \(FI \rightarrow 0\), \((a_o \mu_o)\) approaches 0, and if \(G \rightarrow G_i \) (per cent \(FI \rightarrow 100\)) \(a_o\) is indefinable in nature. An important consequence of Eq. (3) is that the estimated \(a_o\) is less than \(a_o\) for the whole per cent \(FI\) range. The amount of effective area is assumed to be the same for vapor- and liquid-side mass transfer\textsuperscript{2,17}.

2 Experimental Procedure

A pilot-scale column of 9 cm inside diam randomly filled at a depth of 1.90 m with a Raschig-type ceramic ring and equipped with a cylindrical reboiler, a pipe type liquid distributor at the top packed section containing a distribution point density of 470 points/m\(^2\), and a total condenser was used in experiments. To avoid dealing with the effect of variable packing shape and initial voidage, only a single size of packing was studied. Details about the distillation plant and associated control equipment, as well as the geometry of a packing element were reported earlier\textsuperscript{8}. There are seven temperature sensors (Pt100) and six liquid sampling ports. Experimental runs were conducted to determine the capacity and efficiency at total reflux for several constant column pressure drops. The efficiency tests were made using three binary systems of negative I (trichloroethylene/n-heptane), positive II (n-heptane/toluene), and neutral III (benzene/toluene) types due to the ratio of liquid surface tensions of light component to heavy one (\(\sigma_i/\sigma_o\)). Initial liquid mole-fraction of light component \(x_i = 0.33\) for systems II and III, and

<table>
<thead>
<tr>
<th>System</th>
<th>(c_0)</th>
<th>(c_1)</th>
<th>(c_2)</th>
<th>(c_3)</th>
<th>(c_4)</th>
<th>(c_5)</th>
</tr>
</thead>
<tbody>
<tr>
<td>All systems</td>
<td>2.552</td>
<td>0.033</td>
<td>-0.239</td>
<td>0.306</td>
<td>0.290</td>
<td>-0.530</td>
</tr>
<tr>
<td>Positive*</td>
<td>25.462</td>
<td>-0.0065</td>
<td>-0.439</td>
<td>0.027</td>
<td>1.061</td>
<td>-2.50</td>
</tr>
<tr>
<td>Negative*</td>
<td>3.203</td>
<td>0.078</td>
<td>-0.394</td>
<td>0.387</td>
<td>0.136</td>
<td>2.45</td>
</tr>
</tbody>
</table>

* Modelled values give a supreme point
various \( x_i \) compositions in the range 0.25-0.40 for system I were used. The data obtained for system I with \( x_i = 0.33 \) was previously studied. The properties of ring and testing systems are given in Table 2. Solvents (> 99 per cent, G C) were furnished from Fluka. The attainment of constant boiling temperatures for 4 h at the bottom, top, and two middle (0.6 m and 1.20 m high) packed bed sections was the sign of steady-state reached. Liquid samples withdrawn from the column sections were analyzed by a Hewlett-Packard G C, Model 5890A, equipped with FID and HP-I-type capillary column. All runs were performed at normal pressure (\( P = 101.32 \) kPa). Table 3 represents the efficiency behaviors of packings varying with the column pressure gradient (\( P_g \)) attributed to system I. Furthermore, results covering the range of changes of efficiency against the variety of initial liquid composition (\( x_i \)) are shown in Figure 1, that actually may validate a negligible variation of HETP with composition. However,

Table 2 — Summary of packing characteristics and system properties \(^a\)

<table>
<thead>
<tr>
<th>Nominal size, ( d_p ) (mm)</th>
<th>Void fraction, ( \varepsilon ) ((m^3/m^3))</th>
<th>Specific area, ( a_s ) ((m^2/m^3))</th>
<th>System ( b ) ((\sigma/\sigma_0)^v)</th>
<th>Slope, ( n^d )</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.25</td>
<td>0.545</td>
<td>1037.3</td>
<td>I (1.453)</td>
<td>0.880</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>II (0.710)</td>
<td>0.895</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>III (1.007)</td>
<td>0.850</td>
</tr>
</tbody>
</table>

\(^a\) Properties of mixture evaluated through the methods given by Reid \textit{et al.} \(^25\); \(^b\) (negative) trichloroethylene/\( n \)-heptane, II (positive) \( n \)-heptane/toluene, III (neutral) benzene/toluene; \(^c\) Ratio of liquid surface tensions of light component to heavy one; \(^d\) Slope of equilibrium curve

Table 3 — Summary of the packed column efficiency properties obtained with trichloroethylene/\( n \)-heptane system for different initial liquid compositions (\( x_i \)) and various column pressure gradients \(^a\)

<table>
<thead>
<tr>
<th>( \Delta P ) ()kPa</th>
<th>( x_i ) Mole frac.</th>
<th>Column section</th>
<th>Liquid ( x ) Mole frac.</th>
<th>Temperature ( T ) K</th>
<th>Volatility ( \alpha ) (mean)</th>
<th>( N_p^d )</th>
<th>( N_{OL}^e )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.133</td>
<td>0.375</td>
<td>Bottom</td>
<td>0.260</td>
<td>366.8</td>
<td>1.4041</td>
<td>8.23</td>
<td>9.09</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.872</td>
<td>361.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.333</td>
<td>0.312</td>
<td>Bottom</td>
<td>0.207</td>
<td>367.8</td>
<td>1.4031</td>
<td>8.78</td>
<td>9.51</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.858</td>
<td>361.2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.533</td>
<td>0.345</td>
<td>Bottom</td>
<td>0.216</td>
<td>376.6</td>
<td>1.4034</td>
<td>9.02</td>
<td>9.83</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.874</td>
<td>361.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.800</td>
<td>0.300</td>
<td>Bottom</td>
<td>0.185</td>
<td>368.2</td>
<td>1.4027</td>
<td>9.30</td>
<td>10.01</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.862</td>
<td>361.2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.333</td>
<td>0.290</td>
<td>Bottom</td>
<td>0.138</td>
<td>369.6</td>
<td>1.4012</td>
<td>9.74</td>
<td>10.26</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.835</td>
<td>361.5</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.667</td>
<td>0.290</td>
<td>Bottom</td>
<td>0.132</td>
<td>369.7</td>
<td>1.4012</td>
<td>10.11</td>
<td>10.64</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.845</td>
<td>361.4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2.000</td>
<td>0.284</td>
<td>Bottom</td>
<td>0.128</td>
<td>369.7</td>
<td>1.4014</td>
<td>10.57</td>
<td>11.13</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.860</td>
<td>361.2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2.400</td>
<td>0.370</td>
<td>Bottom</td>
<td>0.180</td>
<td>368.3</td>
<td>1.4030</td>
<td>10.99</td>
<td>11.92</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.915</td>
<td>360.8</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3.200</td>
<td>0.335</td>
<td>Bottom</td>
<td>0.152</td>
<td>369.0</td>
<td>1.4023</td>
<td>11.25</td>
<td>12.06</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.905</td>
<td>360.9</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4.000</td>
<td>0.350</td>
<td>Bottom</td>
<td>0.158</td>
<td>368.9</td>
<td>1.4023</td>
<td>10.54 (^e)</td>
<td>11.29</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.887</td>
<td>361.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5.333</td>
<td>0.315</td>
<td>Bottom</td>
<td>0.124</td>
<td>369.7</td>
<td>1.4012</td>
<td>10.35</td>
<td>10.86</td>
</tr>
<tr>
<td></td>
<td></td>
<td>top</td>
<td>0.846</td>
<td>361.4</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^a\) Total reflux condition; \(^b\) Column pressure gradient; \(^c\) Initial composition of distilled binary liquid mixture, mol fraction of light component; \(^d\) Evaluated due to the Fenske equation \(^26\); \(^e\) Evaluated due to the Chilton-Colburn equation \(^26\).
pressure gradient characteristics of packing for air/water system were also evaluated in the model reliability analysis. The efficiency and pressure gradient \((P_k, \text{kPa/m})\) and the results are given in Figure 2 and 3, where HETP is obtained by the Fenske equation. The assumption inherent in this approach, fulfilled for all the systems studied, is attributed to a geometric average of the relative volatility \((\alpha)\) at the top and bottom in terms of the Antoine equation.

3 Results and Discussion

3.1 Design and Analytical Solution of Optimum Conditions

Section 3.1 outlines the steady state two-film model, as well as the concepts of theoretical stage (HETP) and effective interfacial area \((a_e)\) providing the basis for an analytical prediction of the optimum per cent \(Fl\). \(a_e\) was selected as a critical variable by reason of reducing the number of degrees of freedom, as compared to HETP. It is recognized that the behavior of \(a_e\) along the per cent \(Fl\) range presumably tracks the behavior of HETP\(^3\) and the contribution of \(k\) to the overall impact of coupled volumetric coefficient \((ka_e)\) disapprovingly is small enough to reduce a "pocket" type divergence of efficiency near flooding\(^7\). So, the implication for a dependently variation of \(a_e\) and HETP\(^1\) assuming a mean value estimation rule for the range of changes of \(k\) and \(\lambda\) is proceeded.

The opinion of achieving a sentence structure for optimization, based on the independent analysis the divergent behavior of HETP alone, by neglecting the effect of changes of \(k\), \(\lambda\) and \(a_e\) quantities is important but not exactly a sufficient factor for a thorough design process. Unfortunately the solution of extremely complex problem pertained to a simultaneous dealing with the rate of changes of four functionally dependent variables regarding the conditions attributed to the HETP derivatives is highly difficult. However the method to formulate the optimization structure including HETP and \(a_e\) as two
basic variables along with distinguishing the antagonistic changes of \( k \) and \( a_e \) about the loading point was thought to be an acceptable basis for this study. Nevertheless, HETP is chosen as a basic variable establishing the final step of an overall design process and a main comparison criterion for estimates.

Starting from the Fenske equation for the number of theoretical plates, \( \text{Z}_{\text{HETP}} \), and the transfer unit approach preferred as a fundamental basis in the film theory, it has been achieved as analytical structure of optimization related to a “hump” type divergence of both \( a_e \) and HETP, which justifies the derivative conditions of a supreme point.

\[
\frac{\text{d}(a_e)}{\text{d}(\text{per cent } F_l)} = 0; \quad \frac{\text{d}^2 \left( \text{HETP}^\text{'} \right)}{\text{d}(\text{per cent } F_l)^2} = 0. \quad \ldots (4)
\]

For vapor-phase controlled mass transfer process the same remarks hold for the HETP quantity obtained from the height of overall gas-phase transfer unit \( \text{Z}_{\text{HETP}} = \text{Z} + \Delta \text{H}_k = \lambda \), as

\[
\text{HETP} = \frac{\text{ln} \lambda}{\lambda - 1} \left( H_e + \Delta H_k \right), \quad \ldots (5)
\]

where \( \lambda = \frac{mG}{aL} \) is the ratio of slopes of equilibrium (m) to operating lines. \( H_G = G_{\text{eq}}(k,a_e) \) and \( H_L = L_{\text{eq}}(k,a_e) \) are the heights of gas- and liquid-side transfer units, respectively. Incorporating Eq. (5) into Eq. (4) along with supposing a mean estimated value for the overall range of changes of mass-transfer coefficients \( (k, \text{mole-fraction driving force}) \), constant molar flow rates \( (G_{\text{eq}} \text{ and } L_{\text{eq}}) \), and an average \( \lambda \) for all \( Z \), then, the basic optimum conditions attributed to HETP result in

\[
\frac{\text{d}(\text{HETP}^\text{'} \text{per cent } F_l)}{\text{d}(\text{per cent } F_l)} = 0; \quad \frac{\text{d}^2 \left( \text{HETP}^\text{'} \text{per cent } F_l \right)}{\text{d}(\text{per cent } F_l)^2} = 0. \quad \ldots (6)
\]

Similar derivative conditions of HETP have been obtained by Mell and Spekuljak using a heavy component basis along with neglected variation of \( a_e \).

A classical design method mainly utilizes the simple process of setting up the model and then adjusting the design variable in trying to minimize a given objective function. The usual approach to solve the problem focused on Eq. (4 and 6) is to seek an appropriate function between \( a_e \) (or HETP) and flood ratio so that the derivative of the function will be zero at a strongly prescribed value of the independent variable (per cent \( F_l \)). The additional test with the sign of the second derivative separates the location of supreme (optimum) point. To reduce the complexity of optimization problem, an uncoupling and independent dealing only with the derivatives of \( a_e \) is proceeded. From the definition of per cent \( F_l \) the Reynolds number for gas is rearranged as:

\[
\text{Re}_G = \frac{6G}{(a_e \mu_G)} = 0.06G \frac{\% F_l}{(a_e \mu_G)}, \quad \ldots (7)
\]

Incorporating Eq. (7) into Eq. (3) the \( a_e \) derivative is defined.

\[
\frac{\text{d}(a_e)}{\text{d}(\text{per cent } F_l)} = Q \left( \frac{\sigma_e / \sigma_i}{(100 - \% F_l)} - \frac{\sigma_e / \sigma_i}{(100 - \% F_l)} \right) (\text{Re}_G)^{-1} + \frac{1}{(100 - \% F_l)} (\text{Re}_G)^{-1} (\text{Re}_G)^{-1} (0.06G, /a_e \mu_G), \quad \ldots (8)
\]

where \( Q = a_e / (a_e + (C \sigma_i) \cdot (C \sigma_i) - (C \sigma_i) / (C \sigma_i)^{-1} (0.06G, /a_e \mu_G)) \).

Similarly, differentiating Eq. (2) (Bravo-Fair model) in terms of per cent \( F_l \) gives,

\[
\frac{\text{d}(a_e)}{\text{d}(\text{per cent } F_l)} = 0.498a_e \left( \frac{G^{0.98}}{Z^{0.1}} \right) (C \sigma_i)^{0.92} 0.392(\text{Re}_G)^{0.92} - 0.06G, /a_e \mu_G) \quad \ldots (9)
\]

Thus, in searching for the optimum per cent \( F_l \) the derivatives of \( a_e \) to obtain the zero value, as defined by Eq. (8 and 9), need analysis. Study of a large bank of observations by Boiles and Fair has revealed that two-film models do not accurately reflect the HETP behavior in the 80-100 per cent \( F_l \) range. By the analysis of Bravo-Fair and Onda models this tendency can also be verified (Figures 2 - 4). This leads to the modelled derivative values, \( \frac{\text{d}(a_e)}{\text{d}(\text{per cent } F_l)} \) and \( \frac{\text{d}(\text{HETP})}{\text{d}(\text{per cent } F_l)} \), which do not change their sign about the loading point (Figures 5 and 6). The inadequacy of both models for this working range, therefore, does not allow for a sentence structure to obtain the optimum per cent \( F_l \). For practical reasons, in the absence of an interpolation expression for 80-100 per cent \( F_l \) range the evaluation of supreme point based on experiments should be utilized. The optimum per cent \( F_l \) is indicative from the variation of observed \( a_e \) with per cent \( F_l \), as depicted in Figure 4.
Figure 5 presents a quantitative assessment of the \( \alpha \) predictions achieved for both models, defined by Eq. (2 and 3), as well as for their derivative variations with regard to Eq. (9 and 8), respectively. The reliability of Bravo-Fair design approaches Eq. (2 and 9) proved to be disapprovingly less accurate for the supremely ranging zone reproducing the derivative variation with no zero value, as compared to the newly proposed ones Eq. (3 and 8) yielding the optimum conditions, i.e. a zero derivative value at a strongly prescribed value of per cent \( FL \). The same remarks hold for HETP and its derivatives estimated by these models on the basis of the Onda \( k \) coefficients, as depicted in Figure 6. It is expected to apply the obtained optimum per cent \( FL \) to any packed system, so long as the distillate systems are identical with respect to the \((\sigma/\bar{\sigma})\) ratio, i.e. systems are either positive or negative, and a geometrically proportional plant in terms of the \((dL/dG)\) ratio is tested. The observed \( \alpha \) in Figures 4 and 5 was obtained from experimental data of HETP and \( \lambda \), and the Onda liquid- and vapor-side mass transfer coefficients, \( k_L \) and \( k_G \), according to Eq. (10) using the measured system properties.

\[
(\alpha_c)_{\text{obs}} = \frac{\ln \lambda}{\lambda - 1} (\alpha_c H_G + \alpha_c \lambda H_L) \text{HETP}^{-1}, \quad \text{(10)}
\]

where \( H_G = L/(k_G \alpha_P)/H_G = G/[k_G (\alpha_P) M_H] \). The ratio of slopes, \( \lambda = m_G H_G/L_m \), was correlated using the Yoshida and Koyanagi\(^\text{9}\) method for an average slope of equilibrium curve, \( m = \sum \alpha_P L_m/(\sum \alpha_P G_m) \). The number of transfer units, \( N_{OL} = \int(dx/(x - x)) \) and \( N_{OG} = \int(dy/(y - y)) \), were estimated by the Simpson method; \( x \) and \( y \) are the operating liquid and vapor mole fractions; \( y' \) and \( x' \) are the equilibrium values of \( x \) and \( y \), respectively. \( \alpha_c \) was evaluated using a mean \( m \) value given in Table 2 and \( L_m/G_m = 1 \).

Consequently the inadequacy of the estimated derivative variation through Eq. (2 and 9) for 80-100 per cent \( FL \) range would call for assumptions of a
more complex criterion should be adapted to the main structure (CaLiReG) of model to extend the prediction to the flooding zone. A preliminary analysis of formulating a perceptible criterion structure was focused on categorizing and coupling functionally the dimensionless-group variables listed, 

\[ DG = f(d_c, d_p; \sigma/\sigma; \text{h}; \sigma/\sigma; \text{c}) \]

where \( h \) is the liquid holdup, \( m^3/m^3; \sigma \) and \( \sigma \) are the surface tension of liquid and its critical value for a packing material, respectively, N/m. A positive system \( (\sigma_{\text{c}}/\sigma < 1) \) provides for a more interfacial area during distillation. Also, variation in the column diameter \( (d_C > d_p) \) probably has a little further effect for a \( (d_C/d_p) \) ratio larger than 8-10, quoted as a minimum value.

### 3.2 Model Reliability Analysis

The reliability of packed column design models has been analyzed analytically by application of a log-ratio objective function (OF) proposed by Bolles and Fair:

\[ X = \ln(Y_{\text{obs}}/Y_{\text{mod}}) \]  \hspace{1cm} \ldots(11)

where \( X \) is OF of model reliability; \( Y_{\text{obs}} \) is observed value of performance, i.e. flood velocity \((G_f)\), HETP, or \( P_g \); \( Y_{\text{mod}} \) is model predicted (modelled) value. In the design practice of distillation applying a new class rate-based algorithm, success is assumed to be critical at only one end of the confidence band. Using the overall design factor \((F_{\text{od}})\), the model normalization factor \((F_{\text{m}} = \exp(\tilde{X}))\), and the safety factor \((F_{\text{r}} = \exp(S))\) given by Bolles and Fair the following expression for upper \((F_{\text{od}} = F_{\text{m}}F_{\text{r}}; Y = \text{HETP}, P_g)\) and lower \((F_{\text{ol}} = F_{\text{m}}F_{\text{r}}; Y \equiv G_f, a_c)\) bounds critical is obtained.

\[ Y_{\text{desn}} = e^{\tilde{X}} e^{S} Y_{\text{mod}} = (F_{\text{m}}F_{\text{r}})^{1/2} Y_{\text{mod}} = F_{\text{od}} Y_{\text{mod}}, \quad \ldots(12) \]

where \( Y_{\text{desn}} \) = Design value; \( Y_{\text{mod}} \) = Modelled value; \( \tilde{X} = \sum x_{/n} \) is mean (bias) of the objective function; \( \chi_{/n} \) = OF for the \( i \)-th observation; \( n = \) Number of observations; \( s = \sqrt{\sum(x-x_{/n})^2/(n-1)} \) is standard deviation of the OF; \( t = \) Student’s \( t \) for a certain degree of confidence. The overall design factor, \( F_{\text{od}} \) is the overall correction factor that must be applied to the model to achieve a specified probability of success. For a selected 95 per cent probability of success (1 chance in 20 failures) and an infinite number of degrees of freedom related to a 90 per cent confidence band Student’s \( t = 1.645 \) (ref. 21).

The resulting statistical factors of a pilot-scale column, as well as a comparison with the results of Bolles and Fair for the commercial plant are presented in Table 4. These factors indicate the need to overdesign as a result of insufficient model reliability.

<table>
<thead>
<tr>
<th>Model</th>
<th>Present work</th>
<th>Eckert</th>
<th>Bemer-Kalis( ^a )</th>
<th>Onda</th>
<th>Eq. (2)</th>
<th>Eq. (3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( n ) (runs)</td>
<td>14( ^a )</td>
<td>48</td>
<td>103( ^a )</td>
<td>21( ^a )</td>
<td>21( ^a )</td>
<td>21( ^a )</td>
</tr>
<tr>
<td>( \tilde{X} )</td>
<td>-0.3505</td>
<td>-0.204</td>
<td>0.0127</td>
<td>0.0409</td>
<td>0.4268</td>
<td>0.1225</td>
</tr>
<tr>
<td>( s )</td>
<td>0.1273</td>
<td>0.167</td>
<td>0.1212</td>
<td>0.1213</td>
<td>0.0419</td>
<td>0.0216</td>
</tr>
<tr>
<td>( F_{\text{m}} )</td>
<td>0.7043</td>
<td>0.82</td>
<td>1.0128</td>
<td>1.0417</td>
<td>1.5324</td>
<td>1.1303</td>
</tr>
<tr>
<td>( F_{\text{c}} )</td>
<td>1.2337</td>
<td>1.32</td>
<td>1.2214</td>
<td>1.2216</td>
<td>1.0715</td>
<td>1.0363</td>
</tr>
<tr>
<td>( F_{\text{ol}} )</td>
<td>0.5709</td>
<td>0.62</td>
<td>1.2370( ^d )</td>
<td>0.8527</td>
<td>1.4301</td>
<td>1.0907</td>
</tr>
<tr>
<td>Student’s ( t )</td>
<td>1.6500</td>
<td>1.65</td>
<td>1.6500</td>
<td>1.6500</td>
<td>1.6500</td>
<td>1.6500</td>
</tr>
</tbody>
</table>

\( ^a \) \( P = 0.29 (\rho_s^{1/3} G)^{1/3} F_s \left( 1 - \frac{h}{2\tilde{X}} \right) \) \( \cdot \text{C} \), where \( h = 0.34 \mu_s (\rho_s/\rho_s)^{1/3} R_e^{1/3} \) (Bemer-Kalis model\( ^{22} \)); \( ^b \) Estimates due to Bolles and Fair\( ^4 \); \( ^c \) Data for air/water and organic systems I, II and III; \( ^d \) Systems I and II; \( ^e \) Lower bound critical, \( F_{\text{ol}} = F_{\text{m}}F_{\text{r}} \) and 95 per cent confidence; \( ^f \) Upper bound critical, \( F_{\text{ol}} = F_{\text{m}}F_{\text{r}} \).
e.g. a safety factor of 1.234 for the Eckert flood model means that a pilot scale column designed with this model must have, on an average, a cross section 23.4 per cent larger than the modeled value to achieve a reasonable (95 per cent) confidence of success. Table 4 reveals that the standard deviation of the Eckert flood model is remarkably close to 0.15, indicating the consistency of predictions for a wide range of packing type and size and various scaling plants. The reliability tests covering the Berner-Kalis pressure gradient model\textsuperscript{23} (Table 4) were performed concerning air/water system and iteratively-developed packing studied. The convenience of Berner-Kalis is 0.52 and the constriction factor model was also studied by plotting estimates against the experimental data with a mean relative error $\varepsilon(a_e)$, per cent $= 27.3$ and root-mean-square-deviation $\chi(a_e/m^3) = 148.9$, as compared with $\varepsilon$, per cent $= 61.7$ and $\chi = 364.4$ for Bravo-Fair model. The Onda model gives a moderate confidence of $\varepsilon(a_e)$, per cent $= 21.79$ and $\chi(a_e/m^3) = 135.8$, but disapprovingly not convenient for designing a distillation system, since the predicted HETP increases with increase in per cent $Fl$ over the entire working range, i.e. a trend of changes indicative for absorption rather than distillation, as depicted in Figure 3. In fact, besides the accuracy of the model prediction, an important concern is whether the proposed model actually tracks the trend of a pocket type divergence of HETP and its derivative variation from the positive region to the negative one as the flood point is approached. However, it is essential that this phenomenon should have a significant impact on the implementation of a simulation algorithm applying a rate-based method, e.g. a packed column design method of Mori et al.\textsuperscript{24} As evident from Figures 2, 3 and 6 the new model matches the observed HETP reasonably through the whole per cent $Fl$ range except for system III (benzene/toluene).

### 3.3 Design Application Using Statistical Factors

It is observed from Figure 4 that the supremely variation of $a_e$ with per cent $Fl$ indicatively represents a maximum point. In the region above the loading point the effective mass transfer area decreases abruptly along with an unavoidable accumulation of liquid and a large change in vapor pressure in the packed bed.

In this section, an attempt has been made to interpret analytically both optimum per cent $Fl$ and $d_e$. Experimental data of a pilot-plant obtained with trichloroethylene/n-heptane system pertaining to the top column conditions at the beginning of flooding is considered. The significant properties of vapor and liquid estimated by the methods given by Reid et al.\textsuperscript{25} are: Liquid composition, $x = 0.862$ mole-fraction; operating pressure, $P = 101.32$ kPa; liquid load (superficial), $L = 1.22$ kg/m$^2$/s; vapor load (mass), $U = 7.753 \times 10^{-1}$ kg/s; $\rho_l = 1196.4$ kg/m$^3$; $\rho_v = 4.291$ kg/m$^3$; $\mu_l = 0.414$ mPa·s.

Execution of the Eckert flood model with a packing factor ($F_p$) of 6407.9 m$^2$/m$^3$ gives a modelled
gas flood velocity, $G_{\text{mod}} = 1.383 \text{ kg/m}^2\text{s}$. Application of Eq. (12) for $F_1$ with statistical parameters from Table 4 results in a design mass flood velocity, which for a selected 95 per cent probability level should favorably fall to a scope of 90 per cent confidence band ($t = 1.65$) restricted between the following bounds of $G_{\text{des}}$: Upper bound = $1.383F_m \times F_i = 1.202 \text{ kg/m}^2\text{s}$; Mean = $1.383F_m = 0.974 \text{ kg/m}^2\text{s}$; Lower bound = $1.383F_i/F_i = 0.790 \text{ kg/m}^2\text{s}$, with the most probable value of $0.974 \text{ kg/m}^2\text{s}$. Because the maximum hydraulic limit of a packed column is restrained by the flood point, or a condition defined as the 100 per cent MEC point, i.e. a point related to an infinite slope of the pressure drop curve (Figure 2 and 3), the column cross-section with respect to the design flooding load of 0.974 kg/m²/s is:

$$d_c = \left( \frac{U}{G_{\text{des}} \pi} \right)^{0.85} = \left( \frac{7.753\times 10^{-4} \times 4^{0.85} \times 0.974 \pi}{0.974 \pi} \right) \times 100 = 10 \text{ cm} \ldots \text{(13)}$$

Using the concept of Rukovena and Koshy, the design with a proposed load of a 90 per cent MEC can be evaluated, assuming that one of the modeled flood velocity is the 100 per cent MEC: $d_c = \left[ \frac{4\pi(U/F_i) \times G_{\text{mod}}}{4\pi(U/F_i) \times G_{\text{mod}}} \right]^{0.85} = \left[ \frac{4\pi(7.753 \times 10^{-4} \times 0.9 \times 1.383)}{4\pi(7.753 \times 10^{-4} \times 0.9 \times 1.383)} \right]^{0.85} \times 100 = 8.91 \text{ cm}$. Nguyen has proposed an implausible in nature approach to design $d_c$ with a 50 per cent modelled flood velocity value. The implementation of a design method, improved by Bolles and Fair, has been fulfilled by using a flood ratio fixed between 70 and 80 per cent $F_1$. Assuming a flood ratio of 73 per cent, the estimated $d_c$ in terms of the model load (1.383 kg/m²/s) is: $d_c = \left[ \frac{4\pi(U/F_i) \times G_{\text{mod}}}{4\pi(U/F_i) \times G_{\text{mod}}} \right]^{0.85} = \left[ \frac{4\pi(7.753 \times 10^{-4} \times 0.73 \times 1.383)}{4\pi(7.753 \times 10^{-4} \times 0.73 \times 1.383)} \right]^{0.85} \times 100 = 9.89 \text{ cm}$. Correspondingly, a valuable design algorithm of Koch-Glitsch, Inc. (Koch-Glitsch Tower Design Software – KTD 7.0 version) performs perfectly an appropriate result for $d_c$. On the other hand, from Eq. (14) the designed $d_c$ with respect to the upper and lower critical confidence levels of $F_{\text{ad}}$ must be approximately 7 to 32 per cent greater than the model predicted value, with the most probable increase of 19 per cent, according to the mean of objective function, assuming one hopes for a 95 per cent probability of success.

$$d_c \text{ (designed)} = (F_{\text{ad}})^{0.85}d_c \text{ (modelled)} \ldots \text{(14)}$$

Assuming that the designed optimum load does not exceed 90 per cent of the statistically defined lower bound of flood velocity, application of Eq. (12) results in a mass velocity of: $G_{\text{up}} = 0.9\times F_{\text{ol}} \times G_{\text{mod}} = 0.9\times 0.571\times 1.383 = 0.711 \text{ kg/m}^2\text{s}$. With a designed optimum load of 0.711 kg/m²/s the 90 per cent confidence band in terms of flood ratio (per cent $F_1$) is defined as follows:

Lower bound: $(0.711/1.202) \times 100 = 59.6 \text{ per cent}$

Mean: $(0.711/0.974) \times 100 = 73.0 \text{ per cent}$

Upper bound: $(0.711/0.790) \times 100 = 90.0 \text{ per cent}$

The results indicate that the optimum flood ratio is probably between 59 and 90 per cent, with expected value of 73 per cent $F_1$. This is considered a good design loading because it is desirable to avoid the region above 90 per cent $F_1$, where the efficiency decreases beyond control. The observed values are: optimum per cent $F_1 = 80.5$ per cent; $d_c = 9 \text{ cm}$; minimum HETP = 17 cm due to the Fenske equation.

4 Conclusions

Characterization of the mass transfer efficiency with $\alpha_e$, $k$ and per cent $F_1$ variables provides a basis for the description of some aspects of the packed column performance. The evaluated efficiency results for three binary systems give evidence of three critical factors, i.e. the flood ratio, system properties, and packing geometry which affect significantly the magnitude of effective interfacial area. The implication for an elevated amount of effective area in distillation, as compared with the wetted one, is attributed to a tendency toward rippling, wave, and droplet formation in the falling liquid film can be validated by observing various packings. Figure 4 illustrates that the observed and predicted behaviors of $\alpha_e$ are very sensitive to the vapor and liquid loads, as well as to the system properties. Inherent to the uncoupling of $\alpha_e$ from the mass transfer coefficient the effect of changes of $k$ on a “pocket”-like divergence of efficiency near loading are assumed to be negligible, as compared to $\alpha_e$ closely approximating the behavior of HETP. Consequently, a complex challenging problem of treating simultaneously with the derivatives of HETP, $k$, $\alpha_e$, and $\lambda$ quantities has been reduced to the analysis of two dependent changing variables, HETP and $\alpha_e$. The way to formulate the optimization problem, including these design variables, is critically discussed. Some aspects
of designing statistically the \( d_e \) variable in terms of the per cent \( FL \) is expounded.

The proposed \( \alpha_e \) approach, Eq. (3), reflecting the combined effect of loading, packing geometry, and system properties appears to be improvement in data fit for moderate random packings. A modified version of the Kuramoto-Sivashinsky equation for travelling liquid waves including an additional kinetic energy term of gas flow is expected to reflect the influence of the factors mentioned.

Acknowledgments

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References

5 Nomenclature

A = Quantity as defined in Eq. (1)
\( a_e \) = Effective interfacial area \( (m^2/m^3) \)
\( a_p \) = Specific surface area of dry packing \( (m^2/m^3) \)
\( a_w \) = Wetted area of packing \( (m^2/m^3) \)
B = Quantity as defined in Eq. (1)
C = Constant, or coefficient
\( C_{al} \) = Capillary number for the liquid, \( \mu_l L/(\rho_l \sigma_{lg}) \)
DG = Dimensionless group
dc = Column diameter (m)
\( d_p \) = Nominal size (or diameter) of packing (m)
e = Relative mean error, \( e = (100/n) \sum (Y_{obs} - Y_{mod})/Y_{obs} \), (per cent)
F, f = Function symbol
\( F_{lg} \) = Liquid-gas flow parameter
Fp = Packing factor, \( a_f L^3 \) (m\(^3\))
Fl = Film number for the liquid, \( Fr_l/Re_l = a_p^2 \mu_l L/(g \rho_l^2) \)
Per cent Fl = Flood ratio, \( (G/G_i)100 \) (per cent)
FrL = Froude number for the liquid, \( a_p L^2/(g \rho_l^2) \)
G = Gas superficial mass velocity \( (kg/m^2/s) \)
g = Acceleration of gravity \( (m/s^2) \)
gc = Force/mass conversion factor (dimensionless)
\( H_G; H_L \) = Heights of gas- and liquid-side phase transfer units \( (m) \)
\( H_{OG} \) = Height of an overall gas phase transfer unit \( (m) \)
HETP = Height equivalent to a theoretical plate \( (m) \)
h = Liquid holdup \( (m^3/m^3) \)
\( k_G \) = Individual mass transfer coefficient for gas-phase \( (kmol/m^2/s/Pa) \)
\( k_L \) = Individual mass transfer coefficient for liquid-phase \( (m/s) \)
\( k_i; k_l \) = Mass transfer coefficients for the liquid- and gas-side phases, respectively, related to the mole fraction concentration \( (kmol/m^2/s) \)
L = Liquid superficial mass rate \( (kg/m^2/s) \)
M = Molar mass \( (kg/kmol) \)
m = Slope of the equilibrium curve
\( N_{OG}; N_{OL} \) = Number of overall transfer units based on gas- and liquid-side phases
n = Number of observations
P = Total pressure \( (Pa) \)
\( P_e \) = Pressure gradient \( (Pa/m) \)
Q = Quantity as defined in Eq. (8)
\( Re_G \) = Modified Reynolds number for the gas, \( 6G/(a_p \mu_G) \)
\( Re_L \) = Reynolds number for the liquid, \( L/(\alpha_d \mu_L) \)
\( S \) = Standard deviation of objective function
\( U \) = Vapor mass velocity (kg/s)
\( We_L \) = Weber number for the liquid, \( L^2/(\alpha_p \rho_L) \)
\( X \) = Objective function of model reliability
\( \bar{X} \) = Arithmetic mean (bias) of objective function
\( x, x^* \) = Liquid phase mole fractions, operating and equilibrium, respectively
\( \hat{x} \) = Constriction factor
\( Y \) = Independent variable used in objective function, Eq. (11)
\( y, y^* \) = Vapor phase mole fractions, operating and equilibrium, respectively
\( Z \) = Height of packed bed (m)

Greek letters
\( \alpha \) = Relative volatility
\( \varepsilon \) = Fraction void in packing (m³/m³)
\( \lambda \) = Ratio of slopes of equilibrium to operating lines, \( m_G / m_L \)
\( \mu \) = Viscosity (Pa·s)
\( \rho \) = Density (kg/m³)
\( \sigma \) = Surface tension of liquid (N/m)
\( \sigma_c \) = Critical surface tension for a packing material (N/m)
\( \phi \) = Correction factor for packing voidage
\( \chi \) = Root-mean-square-deviation
\( \psi \) = Correction factor for density (\( \rho_w / \rho_L \))

Subscripts
desn = Design
f = Flooding
G (OG) = Gas (overall gas)
h = Heavy (less volatile) component
L (OL) = Liquid (overall liquid)
l = Light (more volatile) component
m = Referred to a molar quantity
mod = Modelled value of performance
obs = Observed value of performance
opt = Optimum
W = Water