Novel Model for Separation Efficiency of Packed Distillation Columns

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We present a novel approach to the determination of the distillation packed bed height utilizing procedure involving new mass-transfer and hydrodynamics models for structured packings. Our approach is based on: a) advanced analyses of the distillation data utilizing composition profiles (instead of HETP); b) measurement and separation of the non-ideal hydrodynamics influence on the packed bed efficiency; c) utilization of the data acquired with proven absorption test methods with non-air (kG) and non-aqueous (kL) systems. The parameters of the here presented model are derived from the experimental data acquired with i.d. 150 mm distillation & absorption column and might be modified and supplemented when industrial scale experimental data and data for more packing types are available.

1. Introduction

The modern rate-based design of the packed-bed mass-transfer exchangers is usually connected with a simple hydrodynamic plug-flow model for both phases, which anticipates no axial mixing. Axial mixing of the phases is an undesirable phenomenon in the multistage and packed mass exchangers as it decreases their separation efficiency through mixing of the matter in the direction of separation and due to the reduction of the interfacial mass-transfer driving force. This phenomenon, known as very important e.g. for extraction towers, is not featured in the contemporary simulation models for distillation columns. Conversely, plug flow is usually assumed, although the axial mixing can be anticipated significant especially for the dispersed (liquid) phase. Match of the simulation results (utilizing plug-flow) to the experimental separation behavior (being affected by axial mixing) has to be achieved by usage of the mass-transfer coefficients with somewhat lower values, or, as is usual, by usage of the mass-transfer coefficients evaluated out of the experimental data with (erroneous) assumption of plug-flow. However, this classical approach brings undesirable consequences. 1) the relative effect of the certain degree of axial mixing increases with overall degree of separation (number of transfer units) and the values of the mass transfer coefficients evaluated with the assumption of the plug flow are therefore dependent also on the height of the packed bed. 2) Modified values of the mass-transfer coefficients become intertwined functions of the quantities affecting not only mass-transfer rate but also the mixing intensity and one can expect their correct correlations to be complex.

On the other hand - introduction of the axial mixing phenomenon into the distillation model makes the description more complex and solution tedious. The situation is not improved by the fact, that the contemporary mass-transfer coefficient models necessary for rate-based modelling application are based on the absorption data or distillation HETP data only and there is complete lack of the mixing data under distillation conditions.

Our workplace has developed a “profile method” enabling determination of the volumetric mass-transfer coefficients kLaPM, kGaPM under the distillation conditions through analysis of the composition profiles along the packed-bed. Its results come out in the form of multiplicative parameters bL, bG correcting first guesses of kL, kG introduced in the model, usually in the form of literature models of kLaLM and kGaLM correlating them with physical properties and phase flows. This procedure is applied mainly due to some changes of kL, kG.
along the packed bed, while the profile method has a potential of determination of just single $k_L$ and $k_G$ for its entire length.

\[
k_L a_{PM} = b_L k_L a_{LM}
\]

\[
k_G a_{PM} = b_G k_G a_{LM}
\]

The method was first applied with an assumption of the plug flow of the phases (Linek et al., 2005), (Rejl et al., 2006). Significant differences between the molar fractions of the component in the liquid and gas phase at one column cross-section under total reflux have been observed (disagreement, spacing of the liquid and vapor phase composition profiles), signaling non-ideal flow behavior. The axial mixing of the phases has been therefore incorporated into the process description through the axial dispersion model utilization (Valenz et al., 2011). Its parameters, Bodenstein numbers $Bo_L$, $Bo_G$, have been first taken from (Macias Salinas and Fair, 1999) rearranged according to the results of the air/water/tracer experiments (Valenz et al., 2010) performed under the identical phase flows on the same packing

\[
Bo_L = 1.022 \times 10^{0.478} \left( \frac{3}{5\sin^2\alpha} \right)^{1.159} (d_{eq} a_g)^{6.337}
\]

\[
Bo_G = 2.807 \times 10^6 Re_G^{-0.00895} 10^{-0.00394} Re_L (d_{eq} a_g)^{-7.792}
\]

Simulation of the distillation process using axial dispersion model substantially improved match of the calculated and experimental composition profiles (Valenz et al., 2011). However, the experimentally found spacing of the vapor and liquid profile was still systematically larger than the simulated one, suggesting that the degree of axial mixing is in fact larger than predicted by correlations Eq\( (3),(4) \). As the axial mixing in the gas phase was of minor effect during air/water experiments, it was assumed that majority of the mixing effect can be attributed to the liquid phase also for the distillation conditions. Under such assumption experimental spacing of the liquid and vapor composition profiles brings a possibility for search of its value through optimization of $\xi$ in (5)

\[
Bo_L = \xi \times 1.022 \times 10^{0.478} \left( \frac{3}{5\sin^2\alpha} \right)^{1.159} (d_{eq} a_g)^{6.337}
\]

Optimized $\xi$ values were significantly lower than unity, moreover exhibited significant dependence on the liquid velocity and therefore suggested insufficiency of (3) for description of the liquid phase mixing in the distillation column.

Study of the mass-transfer under hybrid conditions (absorption experiments with non-air and non-aqueous systems) brought further valuable insight in the mechanism of the mass-transfer. Mass transfer coefficient in the gas phase for Mellapak packings has been found to be well correlated by dimensionless equations

\[
Sh_G = 2.7 + 0.038 Re_G^{0.88} Sc_G^\beta
\]

\[
\beta = 0.33 + 0.9 \exp (-0.6 Sc_G)
\]

derived from the theoretical considerations and experiments with $SO_2$ chemisorption from helium, nitrogen and $SF_6$ into aqueous NaOH solution (Haidl et al., 2017). The gas Schmidt number range of $Sc_G$ 0.49-2.32 includes that of ordinary distillations.

Dimensionless equation for the mass transfer coefficient in the liquid phase for the Mellapak packing

\[
Sh_L = 0.207 Re^{0.798} Fr^{-0.563} We^{0.511} Sc^{0.528}
\]

is derived from the experiments with oxygen desorption from water and from primary alcohols (Rejl et al., 2017). Compilation of the results from the hybrid absorption experiments and distillation experiments evaluated using profile method lead us to the development of a new efficiency calculation approach for structured packings under distillation conditions involving new mass-transfer and hydrodynamics models. The approach is based on: a) advanced analyses of the distillation data utilizing composition profiles (instead of $HETP$); b) measurement and separation of the non-ideal hydrodynamics influence on the packed bed efficiency; c) utilization of the data acquired with proven absorption test methods with non-air ($k_G$) and non-aqueous ($k_L$) systems. While the new model assumes plug flow for the gas phase, plug flow with axial dispersion is introduced for the description of the liquid phase hydrodynamics.
2. Experimental

The distillation experiments have been performed in the stainless steel atmospheric distillation column with an inner diameter of 150 mm under total reflux. The packed bed consisted of ten elements of Mellapak packing with overall height of 2.1 m. The composition profiles were compiled from seven individual composition of vapour and liquid samples taken from the packing along the bed height. The analysis was performed using refractometer Atago RX5000 with accuracy of approximately 0.1 mol. %.

The experiments have been performed with metal Mellapak 250.Y and Mellapak 452.Y packings using binary systems consisting of methanol, ethanol and n-propanol and with cyclohexane/n-heptane system under several power inputs into reboiler up to loading point, thus covering relatively large range of physical and hydrodynamic conditions as shown in the Tab. 1. The detailed set-up of the distillation and absorption facilities has been described elsewhere (Rejl et al., 2009, 2010).

Table 1: Physical properties of the distilled mixtures and hydrodynamic conditions.

<table>
<thead>
<tr>
<th>System</th>
<th>Phase flow rate range</th>
<th>Density</th>
<th>Viscosity</th>
<th>Surface tension</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>B [mh⁻¹]</td>
<td>ρ₇ₕ [kgm⁻³]</td>
<td>η₇ₕ [mPa s]</td>
<td>η₇ₕ x 10⁻² [mPa s]</td>
</tr>
<tr>
<td>MeOH/EtOH</td>
<td>2 - 14</td>
<td>743</td>
<td>0.39</td>
<td>1.05</td>
</tr>
<tr>
<td>EtOH/PrOH</td>
<td>3 - 16</td>
<td>740</td>
<td>0.47</td>
<td>0.98</td>
</tr>
<tr>
<td>MeOH/PrOH</td>
<td>2 - 18</td>
<td>750</td>
<td>0.43</td>
<td>0.97</td>
</tr>
<tr>
<td>C₆/C₇</td>
<td>5 - 23</td>
<td>662</td>
<td>0.27</td>
<td>0.76</td>
</tr>
</tbody>
</table>

Density, viscosity and surface tension for equimolar mixture

3. Results

3.1 Preliminary analysis of kG and kL data evaluated by profile method from distillation experiments

The results of the distillation experiments (kG, kL) recalculated to Sh₅ₕA and Sh₌ₕA have been compared with the data from absorption experiments and with their correlations, assuming identical value of the interfacial mass-transfer area for both processes (Figure 1, 2). The average relative deviation of Sh₅ₕA data (Figure 1) measured under absorption as well as distillation conditions is 17 %, while distillation Sh₃ₕA data differ from the absorption ones significantly (average relative deviation 46 %). Very good agreement of absorption and distillation Sh₅ₕA led us to the belief that the Sh₃ₕ values is the safest candidate for assumption of transferability between the absorption and distillation, while the interfacial area, liquid side mass-transfer and axial mixing is left to be determined from the outcome of distillation experiments. The hybrid absorption correlation for Sh₅ₕ Eq(6), (7) was assumed valid for the distillation process and in the consequence, the corrective parameter B₅ₕ in Eq(1) expresses actually the corrective parameter of the distillation effective interfacial area, determining thus its value.

![Figure 1: Assessment of the gas-phase mass transfer transferability from absorption to distillation conditions. Comparison of the dist. gas phase mass-transfer data with correlation from the hybrid absorption experiments.](image1)

![Figure 2: Assessment of the liquid-phase mass transfer transferability from absorption to distillation conditions. Comparison of the dist. liquid phase mass-transfer data with correlation from hybrid absorption experiments.](image2)
3.2 Deduction of the interfacial area

Application of the modifications mentioned in 3.1 enables the interfacial area under distillation conditions to be deduced by comparison of $k_{G}\alpha_{PM}$ evaluated by the profile method Eq(1) with predictions of the $k_G$ by the model Eq(6), (7). This comparison is depicted in the Fig 3, 4. Figure 4 documents results of ordinary application of the profile method assuming utilization of Eq(2), while Figure 3 demonstrates results for the case, when not only $Sh_G$ Eq(6), (7) but also $Sh_L$ Eq(8) is assumed transferable from (hybrid) absorption to the distillation conditions. The outcome is similar: the effective interfacial area seems to be for all conditions (all type of mixtures, all phase flows) relatively high and constant fraction of the geometrical area of the packing: 80 % for the case when $Sh_L$ is subject of optimization by the profile method, 86 % when Eq(8) is assumed valid.
3.3 Evaluation of the liquid phase axial mixing

The degree of axial mixing of the liquid phase in the form of Bodenstein number \( \text{Bo}_L \) has been evaluated from the distillation experiments by the profile method in the form of the multiplicative parameter \( \xi \) according to \( \text{Eq}(5) \) and are compared with the results of original correlation \( \text{Eq}(3) \) in Figure 5. It can be observed, that the original correlation substantially overestimates the \( \text{Bo}_L \) values (experimentally found values are lower – i.e. the axial mixing is higher) and it also overestimates dependence of the \( \text{Bo}_L \) values on the liquid phase flow. For most experimental conditions the \( \text{Bo}_L \) values are around 10. Suboptimal agreement of the original correlation with experimentally found results lead us to the development of a new correlation for liquid phase Bodenstein numbers under distillation conditions.

3.4 The development of the new liquid phase axial mixing correlation

The new correlation has been developed on the basis of an imagination that the ratio of the convective and dispersion mass transport (Bodenstein number) is a function of inertial, viscous, gravitational and surface forces in the liquid flowing down the packing. These forces stand behind Reynolds', Froude's and Weber's numbers.

\[ \text{Bo}_L = f(Re, Fr, We) \]  

Correlation, with \( \alpha \) according to the packing type, provided

\[ \text{Bo}_L = a \text{Re}^{-0.168}\text{Fr}^{-0.923}\text{We}^{1.097} \]  

signaling cancelation of inertial forces effect and thus reducing after reoptimization into

\[ \text{Bo}_L = a \text{Fr}^{0.178}\text{Eo}^{1.097} \]  

\( \text{Mellapak 250. Y: } \alpha = 0.78; \text{ Mellapak 452. Y: } \alpha = 1.76 \)

Its success in fitting of the experimental data with values is shown in the Figure 6.

3.5 Relative effects of mass-transfer phenomena and axial mixing of the liquid phase on the packing separation efficiency

The results of the distillation experiments can be used for judgment of the relative effect of the individual phenomena (gas-phase mass transfer resistance, liquid-side mass-transfer resistance, axial mixing) on the distillation packed bed efficiency. This effect evaluation has been expressed as a ratio of the simulated packing height necessary for performance of the chosen mixture separation with 1) all three parameters characterizing individual phenomena \( (k_a, k_Ga, \text{Bo}_L) \) taken as their optimization by the profile method 2) the value of the parameter in question set to infinity, while the other two remaining unchanged. (E.g. – setting of \( k_a \) value to infinity assumes no mass-transfer resistance in the liquid phase. Setting of the \( \text{Bo}_L \) to infinity corresponds to the plug flow of the liquid phase). The results identify the gas-phase mass-transfer resistance as a key factor determining the packing distillation efficiency. For high loads the gas phase resistance forms 66 – 90 % of the overall resistance against separation. The liquid phase axial mixing is found to be the next most important phenomenon with about 25 % of the overall resistance against separation. The liquid side mass-transfer resistance is of minor effect, especially under high loads. The results are summarized in the Tab. 2.

<table>
<thead>
<tr>
<th>P [kW]</th>
<th>system</th>
<th>packing</th>
<th>H [m]</th>
<th>Relative effect - parameter [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>k_a</td>
<td>k_Ga</td>
<td>Bo_L</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10 MeOH/EtOH Mellapak 250.Y</td>
<td>2.35</td>
<td>7</td>
<td>61</td>
<td>32</td>
</tr>
<tr>
<td>40 MeOH/EtOH</td>
<td>2.74</td>
<td>0</td>
<td>77</td>
<td>23</td>
</tr>
<tr>
<td>10 MeOH/PrOH</td>
<td>1.07</td>
<td>14</td>
<td>60</td>
<td>26</td>
</tr>
<tr>
<td>40 MeOH/PrOH</td>
<td>1.34</td>
<td>0</td>
<td>82</td>
<td>18</td>
</tr>
<tr>
<td>8 C6/C7</td>
<td>3.10</td>
<td>23</td>
<td>52</td>
<td>25</td>
</tr>
<tr>
<td>24 C6/C7</td>
<td>2.91</td>
<td>0</td>
<td>90</td>
<td>10</td>
</tr>
<tr>
<td>10 MeOH/PrOH Mellapak 452.Y</td>
<td>0.88</td>
<td>19</td>
<td>62</td>
<td>19</td>
</tr>
<tr>
<td>30 MeOH/PrOH</td>
<td>0.99</td>
<td>2</td>
<td>68</td>
<td>30</td>
</tr>
</tbody>
</table>

4. Conclusions

The profile method has been used for an advanced analysis of the distillation data which reveals that the axial mixing of the liquid phase is a significant undesirable phenomenon in the packed distillation columns, forming
about 25% of the overall resistance against separation. There is suggested a new approach for the distillation packed bed height determination, which assumes the plug flow with axial dispersion in the liquid phase. A new correlation for the liquid phase Bodenstein number
\[
B_\text{O}_L = \alpha F_i^{0.178} E_i^{1.097}
\]
Mellapak 250 Y: \( \alpha = 0.78 \); Mellapak 452 Y: \( \alpha = 1.76 \)
has been developed on the basis of the distillation data. The gas phase mass-transfer is modeled using relations
\[
S_h L_g = 2.7 + 0.038 \text{Re}_{L_g}^{0.88} \text{Sc}_L^\beta; \quad \beta = 0.33 + 0.9 \exp \{-0.6 \text{Sc}_L\}
\]
derived from the hybrid absorption experiments, which were found transferable to the distillation conditions. The liquid phase mass transfer correlation
\[
S_h L_L = 0.207 \text{Re}_{L_L}^{0.798} F_r^{-0.563} W \cdot 0.511 \text{Sc}_{0.528}
\]
also derived from the hybrid absorption experiments, has been found poorly transferable to the distillation conditions. However, its predictions are on the safe side from the designer point of view and it was also demonstrated that the liquid side mass-transfer resistance is of minor effect on the packing distillation efficiency. The effective mass-transfer area is found to be well estimated as appx. 86% fraction of the geometrical area. The parameters of the here presented model are derived from the experimental data acquired with i.d. 150 mm distillation and absorption column and might be modified and supplemented when industrial scale column experimental data are available. The magnitude of the axial mixing in the columns of different diameter can differ due to the different wall effect significance and different quality of the initial liquid distribution.

Nomenclature

- **aeff**: effective interfacial area \([m^2 m^{-3}]\)
- **ag**: geometrical area of the packing \([m^2 m^{-3}]\)
- **deq**: packing characteristic dimension \([m]\)
- Sh: Sherwood number
- Sc: Schmidt number
- Fi: Film number \(Fi = u \eta/ (\rho g d^2_{eq})\)
- Eö: Eötvös (Bond) number \(E_0 = \rho g d_{eq}^2/\sigma\)

Reference