Experimental Validation of a Flexible Modeling Approach for Distillation Columns with Packings

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DOI 10.1002/aic.14560

The two main concepts for the modeling of distillation columns are the equilibrium-stage (EQ) and the nonequilibrium-stage (NEQ). A model is presented which combines decisive features of both conventional concepts. Based on the idea of a reduced nonequilibrium-stage (RNEQ), this model can be used for the simulation of distillation columns with packings. In contrast to the conventional NEQ approach, this model neglects the influence of liquid side mass-transfer coefficients, which ultimately allows to come up with only one empirical equation describing the overall mass transfer. Thus, a considerable reduction in model complexity is reached, which allows for an efficient consideration of new experimental distillation results. Fitted to experimental data, the model is able to predict, how different pressures and chemical systems might affect the separation efficiency. By comparing calculation results with experimentally determined separation efficiencies for three different packing types, these valuable RNEQ qualities are illustrated. © 2014 American Institute of Chemical Engineers *AIChE J*, 60: 3833–3847, 2014

Keywords: distillation, mass transfer, structured packing, mathematical modeling, design

Introduction

The focus of this article lies on the mathematical representation of distillation columns. Research in this area started already at the end of the 19th century, when Sorel[1] proposed an equilibrium stage model to simulate the behavior of a distillation column for the purification of ethanol. More than 100 years later, modeling of distillation columns is still a research topic of ongoing interest within the chemical engineering community, which is far from being finished.[2] It is common knowledge among chemical engineers that current distillation column models can be divided into three main categories: equilibrium-stage models (EQ), nonequilibrium-stage models (NEQ) also known as rate-based models, and models applying methods from computational fluid dynamics with the first two categories being the most widely used ones. Interestingly, researchers do not completely agree on the question of which model complexity is needed for the modeling of distillation columns. Although some researchers clearly promote the concept of equilibrium-stages for distillation problems,[3,4] others strongly insist on the application of more complex models (especially in the case of multicomponent distillation).[2,5] For this reason, a detailed comparison between EQ and NEQ approach is presented within this article. The first part of this comparison covers the differences of both types concerning the general model structure, thus giving an overview about how the model complexity differs. The second part of the comparison is related to the question of how experimental data are used to develop empirical equations and parameters describing the separation efficiency of distillation column internals. Even though this difference is of equal importance when it comes to the question of which model type should be used for the solution of a certain problem, it is most often completely overlooked. Therefore, within this article EQ and NEQ models are thoroughly compared regarding both their model structure and the input of experimental data. Based on this comparison, the reduced nonequilibrium-stage (RNEQ) is being presented. This modeling approach will be explained in detail in this article. Major characteristics of this model are qualitatively compared with both conventional concepts in Figure 1. It can be seen that in terms of model complexity the RNEQ approach lies in between the other model types. Another important fact becomes visible when the second category is looked at. Compared to the conventional NEQ approach, this model requires considerably less experimental data to determine empirical parameters describing separation efficiency.

The purpose of this article is to present the RNEQ approach and to discuss its characteristics in comparison with the conventional EQ and NEQ concepts. Therefore, after the general presentation of all three model types, a section is presented in which both the strengths and the limitations of the different models become visible. From an industrial point of view, it is shown how typical engineering problems could be solved with the traditional EQ and NEQ models and with the RNEQ approach. The article's subsequent main part illustrates valuable RNEQ qualities by comparing simulation results with experimental investigations of separation efficiency for three commercial packing structures. It is shown that the RNEQ approach allows to efficiently consider new experimental distillation results. Fitted to experimental data, it gives the possibility to predict

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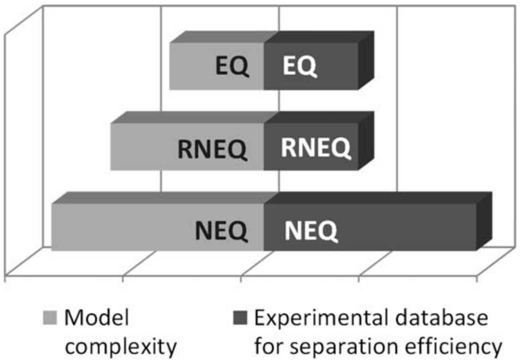

Figure 1. Qualitative comparison of EQ, NEQ, and RNEQ approach.

separation efficiencies for varying chemical systems and process conditions.

Comparison of EQ and NEQ Models

The differences between EQ and NEQ models for the description of distillation columns with structured packings are to be presented in this section of the article. The plural form of the word model is used in this context, because both EQ and NEQ models are published by a lot of different authors. As mentioned earlier, the model types are compared regarding two different categories. First, the main model equations are examined. As this comparison can be found elsewhere,[6,7] it is presented here in a very summarized way. Further books showing NEQ equations are available.[8–10] The second point to be looked at is the huge discrepancy regarding the way experimental data describing the separation efficiency of structured packings is usually considered to determine vital model parameters. As the input of experimental data ultimately determines the accuracy of the model predictions, this second part of the comparison is of major importance.

Model structure

The main equations of EQ models are the well-known MESH equations.[7] The balance equations for material and enthalpy are set up together for the liquid and the vapor phase. The main assumption within EQ distillation models is the fact that liquid and vapor streams leaving a stage are expected to be in thermodynamic equilibrium. Thus, component streams going from one phase to another are accounted for indirectly by incorporating Eq. 1

$$y=K\ x \tag{1}$$

When it comes to separation efficiency of column internals, the one decisive input parameter of EQ models is the height equivalent to a theoretical plate (HETP).[3] For a distillation column with structured packings, this is equivalent to the section height that can be represented by one equilibrium-stage.

The balance equations for NEQ models are set up separately for the vapor and the liquid phase. Both phases are linked by component streams going from one phase to another. In contrast to EQ models, these interfacial streams are calculated directly within NEQ models based on mass-transfer correlations. Equations 2 and 3 show how the calculation of interfacial component fluxes can be done.[6] To calculate the interfacial molar streams on a stage, these fluxes simply have to be multiplied with the stage's volume. It can be seen that for each phase N is assumed to consist of two parts: a diffusional part with the concentration difference between bulk phase and phase interface as driving force and a convective part. It is important to notice that the vapor and liquid concentrations at the interface are assumed to be in phase equilibrium. Hence, both EQ and NEQ models need equilibrium constants which are capable of accurately describing vapor–liquid equilibria

$$N=k_{\mathrm{L}}\ a_{\mathrm{eff}}\ c_{t,\mathrm{L}}\ (x_{\mathrm{int}}-x)+x\ N_t \tag{2}$$

$$N=k_{\mathrm{V}}\ a_{\mathrm{eff}}\ c_{t,\mathrm{V}}\ (y-y_{\mathrm{int}})+y\ N_t \tag{3}$$

Equations 2 and 3 show the three decisive empirical model parameters needed for the NEQ approach to simulate distillation columns equipped with structured packings. These are the mass-transfer coefficients in both phases and the effective interfacial area available for mass transfer between the phases. Empirical correlations are generally used to determine these parameters for different packing types and process conditions.

At this point, it must be stated that the distillation problems looked at in this article are deliberately limited to the binary case. This limitation facilitates the scientific analysis, as diffusional interaction effects which are possible in multicomponent systems, are eliminated. As a further result, the equations in this work do not contain matrices but scalar values only. Therefore, in order to improve the clarity of the equations, subscripts indicating chemical components could be consequently left out in this work. For the same reason, subscripts indicating the stage are left out as well. The reader should simply bear in mind that each individual equation shown in this article is set up individually for each EQ, NEQ, or RNEQ stage.

We are perfectly aware that the possibility to account for diffusional interaction effects in multicomponent systems (e.g., by the use of the Stefan–Maxwell equations) might be regarded as a very decisive advantage of the NEQ approach.[6] However, at the same time, it is obvious that a proper description of binary distillation problems is an absolute prerequisite for that. This, together with the fact that most experimental distillation data are generated with binary test systems,[3] makes the limitation to binary systems a reasonable choice for this work. Nonetheless, the consideration of Stefan–Maxwell diffusion is, of course, possible with the RNEQ approach for multicomponent systems.

Input of experimental data

As mentioned earlier, both conventional types of distillation models require empirical constants or correlations in order to simulate distillation processes. Therefore, the results of scientific distillation experiments are needed to develop and test these empirical correlations. For the case of distillation processes, the most important and most widely used experimental procedure to analyze separation efficiency of different packing types is the distillation at total reflux.[3] For these experiments, several binary test systems are applied, with chlorobenzene/ethylbenzene, ortho-/para-xylene, and cyclohexane/*n*-heptane being the predominantly used ones. Recently, Ottenbacher et al.[3] published detailed practical suggestions about how total reflux experiments should be carried out in order to accurately determine the separation efficiency of distillation column internals. At this point, it should be mentioned that within this article the term separation efficiency is used in accordance with the work of Ottenbacher et al.[3]

EQ Models: HETP. For EQ models, the one empirical parameter describing separation efficiency is the height equivalent to a theoretical plate (HETP). This value can be directly deduced from binary distillation experiments at total reflux with the well-known equation of Fenske.[11] Under the assumption of constant relative volatility, the only other input parameters for its calculation are the purities above and below the packed bed and the packed bed height. HETP can be directly used as input parameter for the EQ approach in order to account for the separation efficiency of a packing. However, this parameter is not constant for all kinds of process conditions. Therefore, it is empirically determined for different test systems and pressures. Its industrial relevance is illustrated by the fact that today, vendors of structured packings offer diagrams showing the dependence of HETP values on *F*-factor for nearly all of their packing types.

NEQ Models: Common Empirical Correlations. For NEQ models, based on numerous experimental results, a huge number of empirical correlations have been developed which claim to predict mass transfer not only for varying process conditions, but also for various packing geometries. Concerning the vast amount of structurally different packing types available on the market, this goal can be called ambitious to say the least. Of course, only a limited number of mass-transfer correlations can be shown in this chapter to give an overview about what type of correlations can be used by chemical engineers to determine the behavior of packed distillation columns with NEQ models. In the following part, three of the most cited correlations are to be compared. The main objective of this comparison is to reveal the way in which the authors use experimental data to determine the mathematical form of their correlations as well as the values of empirical parameters. Additionally, it is shown how the authors tend to include geometric characteristics of different packing types in order to be able to make predictions for various geometries. The correlations to be looked at are the ones from Olujic et al.[12,13] (Delft model), from Rocha et al.[14] (SRP model) and from Billet and Schultes[15] (BS model). As the following comparison is supposed to only give an overview about the main equations and the incorporation of experimental data, the equations will be shown in a very shortened way. Showing all equations, especially all different definitions of dimensionless numbers, characteristic lengths and velocity calculations presented by the authors, would be simply out of the scope of this study. Thus, detailed questions about these mass-transfer correlations should be answered using the original articles.[12–15]

NEQ Models: Calculation of k_L. When comparing the correlations used for the calculation of liquid side mass-transfer coefficients within the empirical models mentioned earlier, one finds that all authors use the penetration model approach to account for liquid side mass transfer (Eq. 4). Higbie[16] derived this equation from Fick's second law for the case of gas absorption in 1935. The penetration model requires estimation of an exposure time describing the time for which each fluid element stays at the phase interface before being replaced.[6] However, even though the same equation form for liquid side mass-transfer coefficient is used within all three models, the exposure time of the penetration approach is described quite differently[12,14,15]

$$k_{\mathrm{L}}=2\left(\frac{D_{\mathrm{L}}}{\pi\ t_{\mathrm{e}}}\right)^{0.5} \tag{4}$$

Billet and Schultes[15] calculate the exposure time with the use of a characteristic length describing the packing type, the liquid velocity, and a packing-specific constant as seen in Eq. 5. The value of C_1 is empirically determined by comparing calculation results with various separation experiments. This value does not change with varying conditions inside the column. However, it is assumed to be dependent on the type of packing structure used. Concerning empirical parameters, the following notation is used within this article. Empirical constants being dependent on packing type are denoted with the letter *C*. Empirical constants which are assumed to be valid for different packing types are named *B*

$$t_{\mathrm{e}}=\frac{C_1\ L}{u_{\mathrm{L}}} \tag{5}$$

Rocha et al.[14] use a slightly different approach to calculate liquid side mass-transfer coefficients with the penetration model (Eq. 6). One important difference to the BS model is the fact that the empirically determined value of B_1 is chosen to be constant for different packing structures and varying conditions inside the column. It is assumed that the differences between packing structure types can be expressed simply with the use of a characteristic length, which, in contrast to the BS model, is set equal to the corrugation side length. The authors justify this assumption by referring to the results of unpublished desorption experiments run at Separations Research Program at The University of Texas at Austin. These experiments were executed for "several well-known structured packings" with an "oxygen-air-water system."[14] However, it is very important to keep in mind that for these kind of stripping experiments, only values for the product of k_{L} and a_{eff} can be determined. Thus, a reliable correlation describing the effective interfacial area has to be applied to get authentic k_{L} values

$$t_{\mathrm{e}}=\frac{B_1\ L}{u_{\mathrm{L}}} \tag{6}$$

Olujic et al.[12,13] also use Eq. 6 in combination with Eq. 4 to calculate liquid side mass-transfer coefficients. They declare to use the same equation as Rocha et al.[14] for liquid side mass transfer. However, in contrast to Rocha et al.,[14] the characteristic length is set equal to a definition of the hydraulic diameter of the flow channels inside the packing structure and not equal to the corrugation side length. Interestingly, the value of constant B_1 in Eq. 6 is not changed accordingly. Thus, depending on the packing type, the model of Olujic et al.,[12,13] should produce different values for k_{L} than the SRP model. However, it has to be kept in mind that these differences are not due to a different experimental dataset. The only reason is the fact that Olujic et al.,[12,13] decide to choose a different characteristic length than Rocha et al.[14] while using the same numerical value for empirically determined constant B_1 in Eq. 6.

Olujic et al.[12] made an attempt to compare the results for k_{L} for all three model types. The authors present a diagram showing calculated k_{L}-values for an exemplary system. The differences discovered between the Delft model and the SRP model are not surprising as explained earlier. But it becomes clear that all three model predictions show remarkable differences. For high *F*-Factors, the deviations shown between each of the three model are bigger than 30% of the calculated mean value.[12] For distillations, however, these differences should not be overrated, because the influence of liquid side mass transfer on distillation is generally considered to be small.[12,14,17–19]

NEQ Models: Calculation of k_V. The mathematically simplest form of k_V-equation is used by Rocha et al.[14] (Eq. 7). In this equation, B_2 represents a constant that is empirically determined by comparing calculation results with various separation experiments. However, it is assumed to be constant for the different types of structured packing.[14] The exponent of the Reynolds number is empirically determined with distillation experiments under total reflux in an earlier publication.[20] The exponent of Sc_V is chosen to be 0.33. These exponents are also assumed to be constant for various packing types and process conditions. The only packing specific information included in Eq. 7 is the characteristic length of Reynolds and Sherwood numbers which is chosen to be the corrugation side length of the packing structure. With this simple approach, it is expected that k_V can be accurately determined for different kinds of corrugated sheet type packings[14]

$$Sh_V = B_2 \; Re_V^{0.8} \; Sc_V^{0.33} \tag{7}$$

As shown in Eq. 8, Billet and Schultes[15] also use an equation where the Sherwood number is described as being proportional to a product of Reynolds and Schmidt numbers with certain exponents. However, in this equation, the prefactor is not a constant value. It is a mathematical term depending on packing type and also on equations describing liquid holdup. As within the BS model, liquid holdup is supposed to be a function of both liquid velocity and viscosity, Γ_1 turns out to be dependent both on the packing type and on process conditions. The value of the packing specific constant being part of Γ_1 is empirically determined by mass-transfer experiments for different packing types[21]

$$Sh_V = \Gamma_1 \; Re_V^{0.75} \; Sc_V^{0.333} \tag{8}$$

The most complex equations for the calculation of vapor side mass-transfer coefficients are given by Olujic et al.[12,13] The equations are divided into a laminar and a turbulent part. In addition to *Re* and *Sc*, they also contain a description for a gas–liquid friction factor and for the liquid film thickness. A very special thing about this model is the fact that k_V is calculated solely with conventional correlations derived for heat and mass transfer in tubes. Neither equation forms nor parameter values are fitted to any kind of experiments with structured packings. Nonetheless, Olujic et al. use their model equations to calculate k_V values for all kinds of packing types.[22,23] This is assumed to be possible simply by incorporating certain geometric characteristics of the gas flow channels like their length per packing segment and their hydraulic diameter.[12,13]

Using the same case study as for the comparison of k_L-values, Olujic et al.[12] also present calculation results for k_V for all three model types (Delft model, BS model, and SRP model). Compared to the calculation of k_L-values, the differences between the models are even bigger in this case. Between each of the models, average differences with a remarkable value of around 60% compared to the mean value of k_V predictions are shown.[12]

NEQ Models: Calculation of a_{eff}. When it comes to the description of the effective interfacial area, the equation forms proposed by Olujic et al.,[13] Rocha et al.,[14] and Billet and Schultes[15] show huge differences. Olujic et al.[13] use an equation which is based on the description for interfacial area presented by Onda et al.[24] The original equation was derived for random packings.[24] To reproduce the results of distillation experiments run at total reflux for various types of structured packings with their overall model, Olujic et al.[13] introduce a complex correction term, which is multiplied with the equation of Onda et al.[24] This correction term contains parameters describing geometric characteristics of the packing structure as well as the operating pressure and the area predicted by the Onda correlation[24] itself (Eqs. 9 and 10). With this information included, Olujic et al.[13] assume the final area equation to be able to reliably predict the effective interfacial area for various corrugated sheet packings

$$a_{\text{eff}} = a_{\text{eff,Onda}} \; (1-\Omega) \; \left(\frac{0.707}{\sin \alpha_L}\right)^n \tag{9}$$

$$n = f\left(a_{\text{geom}}, \alpha_L, a_{\text{eff,Onda}}, p\right) \tag{10}$$

Rocha et al.[14] use a modified version of an equation proposed by Shi and Mersmann.[25] The original equation was also developed to predict the interfacial area for columns filled with random packings (Raschig rings, Berl saddles, Pall rings, and spheres).[25] Rocha et al.[26] add typical geometric parameters of structured packings to this equation and determine empirical constants that allow to reproduce measurements of liquid holdup for different structured packing types. However, in order to match the results of distillation experiments, this fitted equation has to be multiplied with another empirically determined constant. This packing specific constant C_2 is shown in Eq. 11. For some standard packing types, Rocha et al.[14] show this factor to have values in the range of 0.312–0.415

$$a_{\text{eff}} = C_2 \; \frac{29.12 \; (We_L Fr_L)^{0.15} L^{0.359} \; a_{\text{geom}}}{Re_L^{0.2} \varepsilon^{0.6} (1-0.93 \; \cos\theta)(\sin\alpha)^{0.3}} \tag{11}$$

The most complex set of equations used to describe the interfacial area among the three models is proposed by Billet and Schultes.[15] The authors use one equation up to the loading point and another one in the region between loading and flooding point. Main variables of these equations, which are not shown here, are Reynolds, Weber, and Froude numbers for the liquid phase. However, Billet and Schultes[15] do not suggest to use these empirical relations for every distillation task. For distillation systems with decreasing surface tension from column top to bottom, they recommend to further add a correction term Γ_2 which is shown in Eq. 12. Regarding the complex definition of Marangoni number chosen by Billet and Schultes,[15] Eq. 13 only shows a reduced list of input parameters being part of this definition. In this equation, subscript "L,V" means that the respective quantity is incorporated within the equation both for the liquid and the vapor phase

$$\Gamma_2 = \left(1 - 2.4\times10^{-4} \; |Ma|^{0.5}\right) \tag{12}$$

$$Ma = f\left(\sigma_L, \rho_{L,V}, M_{L,V}, \nu_{L,V}, D_{L,V}, u_{L,V}, m_{yx}, \varepsilon\right) \tag{13}$$

Summarizing, it can be stated that the equations of the presented mass-transfer correlations, even though comparable in some parts, show huge differences. Naturally, substantial differences in simulation results may result from that. As mentioned earlier, an attempt of Olujic et al.[12] to exemplary compare the calculation results of all three correlations for the mass-transfer coefficients was published in 1999. There, average differences between the models with a value of 30% for k_L at high column loads and 60% for k_V are presented. Despite these remarkable differences, all authors are able to show good agreement between measurements of separation efficiency determined with binary distillation experiments at total reflux and their simulation results for different structured packing types.[13–15] How is this possible? The answer to this question is given by the third empirical parameter being part of these models: the effective interfacial area. As shown earlier, all three mass-transfer models come up with own developments of area equations. Even though some of them are structurally based on formerly published equations for the effective area, each area equation of the presented mass-transfer models is individually fitted to experimental data. However, this final adjustment is not exclusively done with the use of direct measurement methods for the interfacial area. For each of the presented models, the area equations are rather altered in a way that each overall mass-transfer model is able to show good agreement with experimental measurements of separation efficiency. However, as stated by Ronge,[27] even the best interpolation of experimental results does not imply that each individual contribution to the overall mass transfer is correctly described. Similar criticism can be found in the book by Stichlmair and Fair.[19] They strictly warn against using the area equations of one author with the equations for mass-transfer coefficients originating from another article, as these equations are usually not determined independently.[19] Therefore, the possibility is given that for many cases the calculated effective area does not accurately describe the actual fluid dynamic situation inside the packing, but mainly correct the deviations of calculated mass-transfer coefficients from more realistic values.

Development of RNEQ Model

In this chapter, both the overall model structure of the RNEQ approach is presented as well as the form of the empirical mass-transfer correlations. The model equations are set up and solved with the software Aspen Custom Modeler by Aspen Technology. Thus, the model expands the so-called DWC:toolbox, which is a collection of Aspen Custom Modeler (ACM)-based distillation column models created by our group. In addition to the RNEQ approach, the DWC:toolbox contains distillation models of varying complexity following both the EQ concept[28,29] and the NEQ approach.[30] As indicated by the collection's name, these other distillation models were originally created to simulate the behavior of dividing wall columns.

As NEQ models, the RNEQ model is supposed to directly account for diffusion controlled mass transfer between vapor and liquid phase for distillation processes. This request is based on the reasonable assumption that, using this physically founded approach, the model allows for simulation results with a broader range of validity than possible with simple EQ models. However, compared to the huge number of highly complex mass-transfer correlations published for the simulation of distillation columns equipped with structured packings, the number of empirical equations used for the RNEQ approach is drastically reduced. At the same time, the model equations are supposed to be usable for all kinds of structured packing types. Finally, starting from a point, where the model is fitted to exemplary experimental results, accurate predictions of separation efficiency should be possible for changing process conditions.

Model structure

The balance equations of the RNEQ approach are formulated in a way typical for NEQ models.[6,31] They are set up separately for the vapor and the liquid phase. Shown below are the molar balances, which have to be formulated for each component (Eqs. 14 and 15), and the enthalpy balances (Eqs. 16 and 17). The summation operators in Eqs. 16–18 indicate a summation over all chemical components of the system. Within these equations, an interfacial flux N is defined to be positive when going from the vapor to the liquid phase. The heat flows Q_L and Q_V (Eqs. 16–18) are defined to be positive when entering the vapor or liquid phase

$$0=S_{\mathrm{in,L}}\ z_{\mathrm{in,L}}-S_{\mathrm{out,L}}\ x+N\ V_s \tag{14}$$

$$0=S_{\mathrm{in,V}}\ z_{\mathrm{in,V}}-S_{\mathrm{out,V}}\ y-N\ V_s \tag{15}$$

$$0=S_{\mathrm{in,L}}\ h_{\mathrm{in,L}}-S_{\mathrm{out,L}}\ h_L+V_s\sum\left(N\ h_{\mathrm{C,L}}\right)+Q_L-Q_{\mathrm{loss}} \tag{16}$$

$$0=S_{\mathrm{in,V}}\ h_{\mathrm{in,V}}-S_{\mathrm{out,V}}\ h_V-V_s\sum\left(N\ h_{\mathrm{C,V}}\right)+Q_V. \tag{17}$$

Both phases are linked by component streams going from one phase to another through an infinitesimally thin interfacial layer. This phase interface is crossed both by material streams as well as enthalpy streams. Obviously, the model equations have to make sure that within this infinitesimally thin phase interface no energy can accumulate. This constraint is expressed with Eq. 18, which is the enthalpy balance over the phase interface. It can be seen that the interfacial heat flows Q_L and Q_V are needed to fulfill this enthalpy balance

$$V_s\sum\left(N\ h_{\mathrm{C,V}}\right)-Q_V=V_s\sum\left(N\ h_{\mathrm{C,L}}\right)+\ Q_L \tag{18}$$

In contrast to typical NEQ models, the transfer equation for the liquid phase (Eq. 2) is reduced to Eq. 19 for the RNEQ approach. This formulation implies that the mass-transfer resistance on the liquid side is negligible. As mentioned earlier, it is commonly accepted that for most distillation tasks the mass-transfer resistance is predominantly located on the vapor side.[12,14,17–19] Therefore, this simplification seems to be justified

$$x=x_{\mathrm{int}} \tag{19}$$

Another simplification of the RNEQ approach is the assumption of infinitely high values for heat-transfer coefficients between both phases and the phase interface. Own calculations show this simplification to be justified because of the small effect on separation efficiency, which is in accordance with the findings of Krishnamurthy and Taylor.[32] Thus, for each RNEQ stage both the liquid and the vapor phase are assumed to have the same temperature. It is, however, important to understand that this assumption does not imply the heat flows Q_L and Q_V to be zero. Their values are determined with the enthalpy equations mentioned earlier.

The second mass-transfer equation of NEQ models (Eq. 3) is only slightly changed for the RNEQ case (Eq. 20). To emphasize the fact that the resistance for the overall mass transfer is related to the vapor phase, the subscript "O,V" is

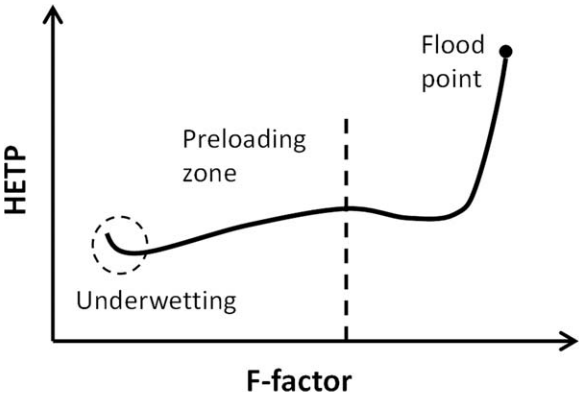

Figure 2. Separation efficiency of structured packings (total reflux distillation).

introduced. Also, for each packing type, the effective interfacial area is characterized by the product of a standard unit area of 1 m^2/m^3 and a dimensionless constant A. Besides that, the mass-transfer equation stays unchanged. Still, it consists of a diffusional part with the concentration difference between bulk phase and phase interface as driving force and a convective part (Eq. 20)

$$N=\left(k_{\mathrm{O,V}}\ A\right)\ a_{\mathrm{unit}}\ c_{t,\mathrm{V}}\ \left(y-y_{\mathrm{int}}\right)+y\ N_t \tag{20}$$

An important issue to mention at this point is that with the simplifications made, the RNEQ model contains only one mathematical term which has to be determined with the use of an empirical correlation: the product of $k_{\mathrm{O,V}}$ and A. This means that the contribution of vapor side mass-transfer coefficients and of wetted surface area to the overall mass transfer is determined altogether. Therefore, the model is not able to make predictions about the wetted fraction of the overall surface. However, it does not assume a certain degree of wetting either. The equations for this calculation are introduced in the following part.

Input of experimental data

Apparently, an equation is needed that describes the product of overall mass-transfer coefficient on the vapor side and the correlated effective interfacial area. To answer the question of how such an equation should look like, one has to define for which situations the RNEQ approach should be used later on. The RNEQ model is supposed to be used for the determination of separation efficiency of structured packings implemented in industrial distillation columns.

Figure 2 gives an impression of how the dependence of separation efficiency on vapor loading usually looks like for structured packings.[33] It can be seen that the HETP value slightly increases with rising F-factor in the preloading range. For even higher vapor loads, close to the flood point, HETP might even decrease again, before ultimately rising at flooding conditions. This effect is due to an increase in liquid holdup,[33] which leads to a rise in interfacial area.[27] However, as stated by Ronge,[27] this local minimum of HETP value is not observed for all packing types.

Most industrial distillation columns are designed to be operated in the preloading zone to prevent the effect of flooding. Therefore, the equation for the product of $k_{\mathrm{O,V}}$ and A is supposed to be valid within this specific region. But how should an equation look like that allows for the combined description of $k_{\mathrm{O,V}}$ and A in the preloading zone?

Obviously, the effective interfacial area is dependent on many physical quantities. Stichlmair and Fair,[19] for example, show an impressive illustration of how surface tension and contact angle can influence the wettability of a flat surface. However, two decisive problems occur, when the interfacial area of structured packings is to be described with the use of an empirical equation using dimensionless numbers. First, in order to correctly describe all influences, one would have to include many different dimensionless groups. Therefore, a lot of experimental data with varying values for these quantities must be available to accurately determine the influence of each group on the interfacial area. However, such an extensive set of experimental data is missing for many different packing types used in industrial distillation columns. Second, even if a lot of different experimental results are available, many decisive physical properties are not known exactly. The final problem is that the interfacial area itself cannot be directly determined with standard distillation experiments.[19]

For these reasons, the equation for the combined calculation of $k_{\mathrm{O,V}}$ and A, which is tried to be kept simple, solely utilizes dimensionless numbers used for the description of vapor side mass-transfer coefficients. Own calculations for the vapor flow show that for most industrial distillation processes, slightly turbulent conditions should be present inside the corrugation channels of structured packings. Therefore, our empirical equation for $k_{\mathrm{O,V}}$ and A is based on a common empirical approach used for turbulent vapor side mass-transfer coefficients in tubes (Eq. 21)

$$Sh_{\mathrm{O,V}}=C_A\ Re_{\mathrm{V}}^{C_B}\ Sc_{\mathrm{V}}^{0.33} \tag{21}$$

As usual for such a correlation, the exponent of the Schmidt number is chosen to be 0.33.[34] Vapor phase Schmidt numbers, as will be shown later, usually do not change a lot for distillation processes. Therefore, a constant exponent not fitted to individual experiments seems to be justified in this respect. As commonly known, for turbulent conditions in tubes, Sherwood numbers are proportional to Reynolds numbers with an exponent of around 0.8.[35] This finding is valid for an interface of constant size. Therefore, with the assumption of only slightly changing interfacial area, Reynolds numbers with an exponent of approximately 0.8 are to be expected for constant C_B. The values of both parameters C_A and C_B of Eq. 21 have to be determined with the use of experimental distillation results.

In this work, the characteristic lengths being part of Sherwood and Reynolds numbers are chosen to be equal to 1 m for all cases. This means that Eq. 21 together with the definitions of dimensionless numbers being part of this equation (Eqs. 22–24) form the total set of equations being used for the description of mass transfer within the RNEQ approach. No further equation has to be solved in this respect

$$Sh_{\mathrm{O,V}}=\frac{\left(k_{\mathrm{O,V}}\ A\right)\ L_{\mathrm{unit}}}{D_{\mathrm{V}}} \tag{22}$$

$$Re_{\mathrm{V}}=\frac{V_{\mathrm{V}}\ \rho_{\mathrm{V}}\ L_{\mathrm{unit}}}{A_{\mathrm{cross}}\ \eta_{\mathrm{V}}} \tag{23}$$

$$Sc_{\mathrm{V}}=\frac{\nu_{\mathrm{V}}}{D_{\mathrm{V}}} \tag{24}$$

In his Ph.D. thesis, Ronge[27] uses a model very similar to the one proposed in this article to successfully describe the separation efficiency of a specific structured packing type. The main differences of his model equations to the ones proposed here are the choices for the interfacial area and for the characteristic lengths. Ronge[27] decides to include both the geometric surface area of the packing and a packing-specific definition for its characteristic length into his model equations. Though entitling this choice to be arbitrarily made,[27] the impression might arise that empirical constants determined with those equations for one packing type could be used for other types as well, if geometrical parameters are changed accordingly. Regarding recent findings about the very complex and locally varying distribution of both liquid holdup and interfacial area for an exemplary structured packing type,[36] it must be questioned whether such a procedure would lead to accurate simulation results. Therefore, in this article, not a single geometric parameter describing the packing geometry is included in the equations of RNEQ model.

Summarizing, it can be stated that the RNEQ approach, in addition to a general negligence of liquid side mass-transfer resistance, has two other decisive features. First, as stated earlier, the model contains not a single geometrical parameter describing the packing structure. Second, typical dimensionless numbers being present in many equations for the effective interfacial area are decided not to be included. Both positive and negative consequences resulting from these simplifications are presented in Sections Application of RNEQ Model and Discussion.

Application of EQ, NEQ, and RNEQ Models

As stated by Marquardt,[37] models are created and used to answer questions about real systems. Mathematical models are neither good nor bad in general. They are either well suited or not so well suited for the solution of a certain problem. Therefore, when analyzing the strengths and limitations of a model, one has to refer to a certain problem to be solved with the model. Without doing that, different model types can be compared but not evaluated. Therefore, in the following part, the determination of separation efficiency for packed distillation columns is discussed for EQ and NEQ models as well as for the RNEQ approach from an engineering perspective. The different model types are evaluated regarding their applicability to determine the separation efficiency for a hypothetical packing type XY. In this regard, two different situations are looked at for each model type.

First, it is shown how separation efficiency can be calculated, if no experimental data for this specific packing is available. As shown earlier, all model types (EQ, NEQ, and RNEQ) use empirically determined parameter values. Therefore, if no experimental distillation results are available, predicting the separation efficiency for specific distillation processes is a very challenging task.

After that, it is illustrated how the determination of separation efficiency is possible if experimental data for packing type XY is available for certain test systems. Even though this problem seems to be considerably easier than the first one, it is by no means trivial. This is due to the fact that engineers ultimately are interested in predictions of separation efficiency for industrially relevant chemical systems. The physical properties of such systems for which distillation columns are to be designed usually differ from the properties of the original test system. Thus, even for the case that experimental distillation results with test systems are both available and in correspondence with simulation results for a certain packing, chemical engineers have to answer one crucial question. It has to be decided whether the applied model also allows for accurate predictions concerning the industrial relevant system for which a distillation column has to be designed.

Application of EQ models

If no experimental data concerning the separation efficiency is available for a packing structure, its prediction is difficult. For EQ models, one possibility to solve this problem is to use HETP values of geometrically similar packings for which experimental distillation results are available. These values could be used as a basis for the estimation of separation efficiency of the untested packing type. Understandably, huge errors might be introduced with such a procedure. Another possibility is to use shortcut methods that claim to directly predict HETP values for various packing geometries.[38,39] However, the low number of citations in scientific journals for articles following this idea illustrates that shortcut models of this type do not seem to be widely used.

If for a test system, experimental results concerning separation efficiency are available for a packing type, HETP values accurately describing the experimental results can be derived from the available distillation data. These values can be directly used with EQ models to simulate the separation behavior of the respective packing type for certain distillation tasks. However, the general limitations of EQ models considering the predictability of processes with different physical properties have to be considered. The EQ model equations do not give a hint about how HETP might be affected by changing physical properties. Therefore, mainly based on personal experience, the engineer being faced with this problem has to guess how HETP value might change.

Application of NEQ models

For NEQ models the situation is a little bit different. As mentioned earlier, frequently cited correlations are available that claim to predict mass transfer in distillation columns for various packing types. Therefore, if experimental data for a certain packing type is missing, one could use empirical parameters determined for geometrically similar packings, when the correlations of Billet and Schultes[15] or Rocha et al.[14] are used. The geometric differences between the tested packing type and untested type XY can then be considered within the model equations. Olujic et al.[13] even claim that their correlations do not need any empirically determined, packing-specific input parameter to predict the separation efficiency of untested corrugated sheet packings. Therefore, this model can be used directly to make predictions of separation efficiency for untested packing types. For each of the three models, however, the accuracy of such predictions is hardly possible to evaluate in advance.

If experimental data about the separation efficiency of packing type XY is available for certain test systems, the engineer's situation is a little better but still far from being easy. Many different empirical mass-transfer models are available for packed distillation columns.[40] Therefore, for engineers, it is hard to decide which correlation should be used. Even if one decides to only choose between the three models presented in this article (BS model, Delft model, and SRP model), this question is hard to answer. As presented earlier, one decisive difference between the three models is the form of their highly complex area equations. A lot of experimental data originating from experiments carried out with all types of packing was used to create them. In the

Table 1. Basic Data for All Experiments Analyzed in this Article

Packing and Organization[a]	Column Diameter (m)	Packed Bed Height (m)	Test Conditions[b]
B1-250MN, FRI	1.22	2.99	PX/OX (10.0 kPa) C6/C7 (31.0 and 162 kPa)
B1-350MN, FRI	1.22	2.60	PX/OX (10.0 and 100 kPa) C6/C7 (31.0 and 162 kPa)
B1-500MN, BTS	0.59	3.49	CB/EB (10.0 and 100 kPa)

[a] Packing type and organization running the experiments.
[b] Chemical system and column top pressures (in brackets).

end, the authors come up with totally different and highly complex mathematical forms whose strength is to successfully interpolate a lot of experimental data when used with the corresponding correlations for mass-transfer coefficients. Additionally, at least two of the presented NEQ correlations contain empirical parameters, which are supposed to be packing-specific. Therefore, by fitting these parameters to the hypothetical data of packing type XY, at least one of the models should be able to reproduce the experimental data for this packing. Ronge,[27] however, strongly emphasizes the fact that the sheer ability of an equation to interpolate experimental data, does not imply a sound physical foundation, which is needed to make reasonable predictions for changing physical properties (i.e., other column pressure and other chemical system). Therefore, when the separation efficiency of one specific packing type is to be determined for process conditions deviating from the ones for which experimental data are available, one decisive question has to be asked: for this specific problem, is it of any help to use a set of equations which in its entirety was created to interpolate experimental data also for a lot of other packing types?

Application of RNEQ model

Is it possible to use the RNEQ model to make predictions for a packing type for which no experimental data concerning the separation efficiency is available? As for the NEQ approach, it is possible to use empirical parameters of geometrically similar packing types. However, we strongly recommend against doing so, as the RNEQ model was created without a single parameter describing packing geometry. It is supposed to only be used for packing types, for which experimental data concerning separation efficiency is available. From the industrial point of view, however, this constraint hardly limits the applicability of the RNEQ approach. This is due to the fact that for industrial production plants, distillation columns are only rarely equipped with packing types for which experimental data for separation efficiency is totally missing.

The second problem to be looked at is of considerably higher industrial relevance. Fortunately, this is where the RNEQ approach has its major impact. For three exemplary packing structures, the following part of the article illustrates how the RNEQ model can successfully be used for the calculation of separation efficiency, if some experimental data are available.

Validation of RNEQ Model

This part of the article starts with the presentation of experimental HETP results which were provided by Julius Montz GmbH, Hilden, Germany. For three of the company's structured packing types, HETP values are presented which were determined at total reflux. The RNEQ model is fitted to some of the data and subsequently used to predict the remaining data points.

Experimental distillation results

The packing types to be looked at in this part belong to Julius Montz' structured packing series. Montz MN packings are of the corrugated metal-sheet type with a corrugation angle which at the bottom of each packing element gradually changes to the vertical direction. These packings were experimentally examined for different chemical systems and pressures at total reflux by Bayer Technology Services GmbH (BTS) and by Fractionation Research, Inc. (FRI). A total of nine test runs conducted at different process conditions is analyzed within this article. Some of these experiments were published already in the work of Olujic et al.[22,23] who compared the experimental findings with the predictions of the Delft model. Table 1 lists test conditions and column dimensions for all of the nine test runs. The experiments were run at pressures ranging from 10 to 162 kPa. The test systems used within the experiments conducted by FRI are para-xylene/ortho-xylene and cyclohexane/*n*-heptane. The drip point density of the liquid distributor, which was used during these experiments, has a value of 143 points/m^2. The system used by BTS is chlorobenzene/ethylbenzene. For these experiments, two different distributors were used. The drip point densities were 198 points/m^2 for the run at 100 kPa and 516 points/m^2 for the measurements at 10 kPa.

Figure 3 shows the experimental results of both the FRI and BTS measurements. It gives the reader an impression of how strongly the measured HETP values can differ for each packing type between the test runs. For each point in this graph, the column was run at total reflux in a steady-state mode. The HETP values shown in Figure 3 are the original ones determined by FRI and BTS. However, for reasons of data reduction, data points with F-factors differing by less than 1% were combined for each of the test runs. The HETP values were calculated with the Fenske equation[11] using the relative volatility and the compositions measured above and below the packed bed. For the FRI tests, measurements of composition taken from a distributor

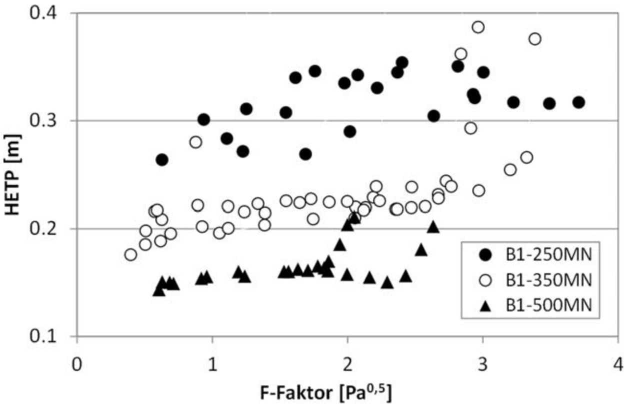

Figure 3. Experimental HETP values of Montz B1-250MN, B1-350MN, and B1-500MN at column pressures ranging from 10 to 162 kPa.

Table 2. Calculated Physical Properties for Pure Components at Process Conditions[a]

Chemical System	Chlorobenzene/Ethylbenzene				Cyclohexane/*n*-Heptane				Para-/Ortho-Xylene			
Top pressure (kPa)	10.0		100		31.0		162		10.0		100	
Component name	CB	EB	CB	EB	C6	C7	C6	C7	PX	OX	PX	OX
Boiling temperature (°C)	63.3	67.0	131.5	135.7	45.9	62.7	97.2	115.2	68.9	74.1	137.9	143.8
Vapor density (kg/m^3)	0.40	0.38	3.35	3.12	0.98	1.11	4.43	5.03	0.37	0.37	3.11	3.06
Vapor viscosity (μPa s)	8.4	7.4	10	8.8	7.6	6.6	8.7	7.6	7.2	7.5	8.7	8.9
Vapor diffusion coefficient[b] (mm^2/s)	30	31	4.4	4.5	9.4	10	2.4	2.7	28	29	4.1	4.2
Liquid density (kg/m^3)	1061	827	984	761	754	649	704	598	819	835	754	770
Liquid viscosity (mPa s)	0.51	0.40	0.29	0.24	0.64	0.27	0.35	0.16	0.38	0.43	0.22	0,25

[a] Calculated with Aspen Properties V7.3 by Aspen Technology, Inc. using pure component databank PURE25.

[b] For respective binary system (Wilke-Lee model).

above the packed bed and from a sample position directly below the packed bed were used to directly compute HETP values. For the BTS tests, composition measurements were available for column top and for the bottom. Thus, for this case, the results of Fenske equation[11] were reduced by one to account for the reboiler stage.

The numerical value being part of the packing names of the MN-series is related to the surface area per overall packing volume. Thus, B1-250MN packing has a surface area of approximately 250 m^2/m^3. As expected, it can be seen in Figure 3 that with rising area installed the HETP values decrease. However, comparing the measured results for the different packing types, one easily notices differences in the variation caused by changing process conditions. For example, both test runs conducted for type B1-500MN basically give the same HETP values in the preloading region, whereas for the other packing types the measured separation efficiency partly varies a lot (e.g., more than 25% difference in HETP for B1-250MN at $F = 1.7$ Pa^5). Now, the question is whether, despite those differences in experimental findings, the RNEQ model is capable of coping well with all packing types presented in this article.

Calculation of physical properties

This section gives simulation results for all decisive physical properties in order to illustrate how test conditions differed between the test runs and to give an impression of the property data used in this work. Table 2 lists the calculation results for the different components and pressures present during total reflux experiments. The presented values for viscosity, density, and diffusion coefficients are calculated for the given values of pressure and boiling temperature. Figure 4 gives an illustrative overview of how the calculated physical properties differ for the different test conditions.

It can be seen that liquid density and the viscosities in both phases only show moderate differences for the different test conditions. Both vapor density and vapor phase diffusion coefficients, however, show considerable differences. To estimate the resulting impact on mass transfer, Figure 5 gives normalized values both for Reynolds and Schmidt numbers. It should be noted at this point that the Reynolds numbers shown in Figure 5 are calculated for a constant F-factor. It can be seen that basically no change is present according to Schmidt numbers. Reynolds numbers, however, do change a lot even though the F-factor is kept constant.

Determination of empirical parameters

As mentioned earlier, the RNEQ model is created to be used in the preloading region (see Figure 2). Therefore, when determining parameters C_A and C_B of the RNEQ approach (Eq. 21), it is crucial to only use data points from this region. Data points being close to the flood point concerning F-factor should be obtained, as well as data points at the far left of HETP-diagrams, if HETP values are rising with decreasing gas load. As indicated in Figure 2, this last effect can be explained with the liquid load falling below a minimum value which causes an underwetting of the packing surface.[19] As commonly known, for a constant F-factor, the liquid load falls with decreasing column pressure. Therefore, for the subsequent regressive determination of C_A and C_B, the test run with the highest pressure was used for each

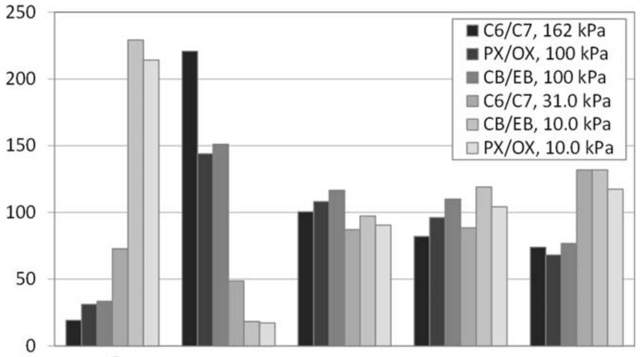

Figure 4. Normalized mean values for physical properties (100 = mean value).

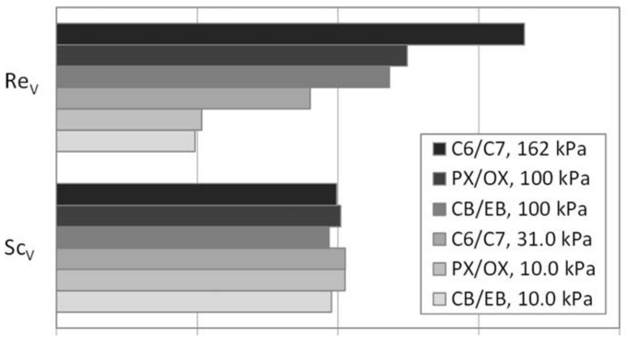

Figure 5. Normalized mean values for dimensionless numbers (100 = mean value).

Table 3. Data Points Used for the Regressive Determination of C_A and C_B

Packing Type	Test Conditions: Pressure (kPa)	Test Conditions: Chemical System	Regression Range: F_{min} ($Pa^{0.5}$)	Regression Range: F_{max} ($Pa^{0.5}$)	Number of Data Points[a]	Regression Results: C_A	Regression Results: C_B
B1-250MN	162	C6/C7	0.63	1.76	5	59.8	0.75
B1-350MN	162	C6/C7	0.51	1.54	5	70.2	0.77
B1-500MN	100	CB/EB	0.96	1.78	5	28.1	0.88

[a] Being used for regression.

packing type in order to assure high liquid loads even for low F-factors. Another important issue is the question of how many experimental data points should be used for the determination of C_A and C_B. To answer this question, it is important to understand that C_B of Eq. 21 basically determines the dependence of $k_{O,V}$ on the gas load. It is, therefore, strongly related to the slope in HETP diagrams. And, in order to determine this slope with a high accuracy, it is important to choose data points with a decent variation concerning the gas loads. Otherwise, errors in these data points would strongly affect the extrapolation to other Reynolds numbers. Following these advices, the regression ranges listed in Table 3 were identified, which ultimately led to regression values for C_A and C_B given in the same table.

Table 4 gives an impression about how the simulation runs carried out in this work were executed. Exemplarily, both the input values of the RNEQ model are shown there for one data point per packing type as well as the measured and calculated compositions at the column top and the resulting HETP values. The HETP values of these data points can also be found in Figure 6. To allow for a sensible comparison between experimental HETP values and HETP values originating from RNEQ model results, the same value for the relative volatility is used within the Fenske equation in both cases. Within this article, for each data point, the relative volatilities originally determined by FRI and BTS were used to calculate HETP values.

It must be stated at this point that the RNEQ model was not fitted to the HETP values determined by FRI and BTS but it was rather fitted directly to measured compositions. If possible, regression runs should be always done like this. This is due to the fact that considerable errors might be introduced during calculation of HETP depending on the value of relative volatility which was used.[3] HETP values are only used within this article to effectively visualize and compare calculation results with the results of the distillation experiments.

In this work, the regression was run in a way that for the calculation of each data point the corresponding input values shown in Table 4 were used with the RNEQ model. Subsequently, for each packing, C_A and C_B were varied in order to minimize the sum of squared errors between calculated and measured compositions above the packed bed. The sets of available measurements slightly differ between FRI and BTS runs. For example, for BTS experiments reboiler and condenser duties were not available, but vapor and reflux streams. Therefore, Table 4 lists slightly different input values for FRI and BTS runs.

It is important to notice that the input parameters shown in Table 4 allow for the consideration of both heat loss and pressure drop with the RNEQ model. Therefore, for each data point calculated in this work, both the overall heat loss and the overall pressure drops are considered. They are equally distributed on a total of 100 RNEQ stages, which were used for all calculations. Concerning vapor–liquid equilibria, it is important to notice that for all calculations ideal vapor and liquid phases were assumed (i.e., activity coefficients = 1).

Table 4. Exemplary Presentation of RNEQ Input and Output Values and Comparison with Experiment

Packing type Process conditions Organization/company		B1-250MN C6/C7, 162 kPa FRI	B1-350MN C6/C7, 162 kPa FRI	B1-500MN CB/EB, 100 kPa BTS
RNEQ input	Lower composition[a] (mol/mol)	0.084[b]	0.074[b]	0.126[c]
	Pressure drop (mbar/m)	0.48	0.72	2.45
	F-factor ($Pa^{0.5}$)	1.25[d]	1.24[d]	1.19[e]
	Inner column diameter (m)	1.22	1.22	0.59
	Packed bed height (m)	2.99	2.60	3.49
	Reflux temperature (°C)	64.7	13.0	114.2
	Condenser duty (kW)	900	775	–
	Reflux stream (kg/s)	–	–	0.559
RNEQ output	Upper composition[a] (mol/mol)	0.873	0.948	0.688
	HETP[f] (m)	0.307	0.214	0.159
Experimental	Upper composition[a] (mol/mol)	0.868[g]	0.945[g]	0.683[h]
	HETP[f] (m)	0.311	0.216	0.160

[a] Molar fraction of light boiling component.
[b] Measured directly below the packed bed.
[c] Measured in the column bottom.
[d] Below packed bed.
[e] In the middle of the packed bed.
[f] HETP calculated from measured lower and respective upper composition (RNEQ output or experimental).
[g] Measured in distributor directly above the packed bed.
[h] Measured at column top.
-Not used.

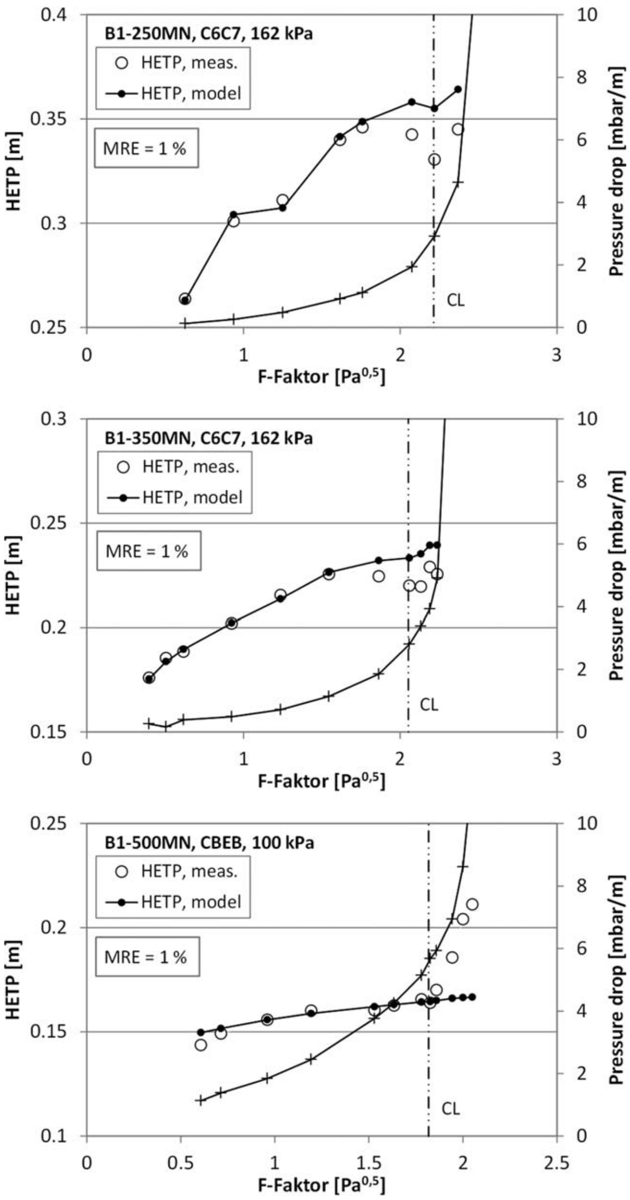

Figure 6. Regression results for B1-250MN, B1-350MN, and B1-500MN.

Figure 6 gives a graphical impression of the very good model accuracy. For each regressive run, these diagrams show a comparison between HETP values calculated with the use of the RNEQ model and measured values of HETP. The mean relative error of this comparison has a value of only 1% for each of the packing types. For the calculation of this average error, each data point up to the capacity limit CL is considered. This line marks the gas load which, for the present test conditions, would usually not be exceeded in industrial applications. In accordance with the work of Spiegel and Meier,[17] CL is defined as 90% of the *F*-factor at flooding conditions, which are supposed to be present at specific pressure drops of 10 mbar/m. Comparing the position of the CL between the different diagrams shown in Figures 6 and 7, one sees that CL decreases with increasing pressure for the specific packing types. These flooding characteristics can also be seen in Figure 3, where different branched groups of experimental points are shown for B1-350MN and B1-500MN at high *F*-factors. It should be noted at this point that the straight lines connecting calculated HETP values are only inserted because of graphical reasons. They do not indicate calculation results for the space between presented data points. The same is valid for Figure 7. Finally, it is important to state that for the calculation of HETP values in Figures 6 and 7, as explained earlier, the Fenske equation was used.[11] The relative volatilities needed for this calculation were in accordance with the original values used by BTS and FRI.

Despite the remarkably good agreement between HETP values originating from the experiments and those calculated from our simulation results, it is important to check the accuracy with which the regressed parameters can be determined. Do the five data points which are used for each packing type allow for an accurate determination of the regression parameters? As explained earlier, the accuracy of parameter C_B strongly affects the extrapolation to other Reynolds numbers. It basically determines the slope in HETP diagrams. Low values of C_B result in high values for the slope. For a good extrapolating behavior of the RNEQ model, it is therefore of crucial importance to be able to accurately determine this value. Changing the parameter C_A, basically results in a vertical movement of the points within HETP diagrams. As can be seen in Eq. 21, this parameter can be understood as the overall vapor-side Sherwood number, for the case that both *Re* and *Sc* equal 1. Vapor phase Schmidt numbers are usually close to one for distillation processes. Reynolds numbers, however, are far from that. Therefore, knowing the accuracy of C_A is of minor importance for the user. One is rather interested in the accuracy with which Sherwood numbers can be determined for more realistic values of Reynolds numbers. To get this information, Eq. 21 can be rearranged to give Eq. 25

$$Sh_{\mathrm{O,V}} = C_A^{'} \left(\frac{Re_{\mathrm{V}}}{Re_{\mathrm{V,m}}}\right)^{C_B} \left(\frac{Sc_{\mathrm{V}}}{Sc_{\mathrm{V,m}}}\right)^{0.33} \qquad (25)$$

In this equation, subscript "m" indicates a mean value. For each of the three regression runs being listed in Table 3, these mean values were determined from the five data points on which the regression was based. Thus, $C_A^{'}$ in Eq. 25 can be understood as the overall vapor-side Sherwood number, for the case that both *Re* and *Sc* equal their mean value present in the respective dataset being used for the regression. The accuracy of this value together with the accuracy of C_B is important to know for the user of the RNEQ approach. Statistical analysis carried out with ACM shows a mean 95% confidence interval of ±1.8% for the determination of $C_A^{'}$ and ±7.1% for parameter C_B for the regression runs. Basically, this means that both the slope of the curves shown in HETP diagrams as well as their vertical position can be determined with a high accuracy from the dataset being used for the regression. It should be clear at this point that the calculation of the confidence intervals is based on the assumption of normally distributed residuals with an expected value of zero.

Calculation of separation efficiency for deviating process conditions

The preceding section shows very good agreement between calculated and measured HETP values. However, it has to be kept in mind that for those test runs C_A and C_B were determined to produce a best fit. The real value of the RNEQ approach can only be determined when model

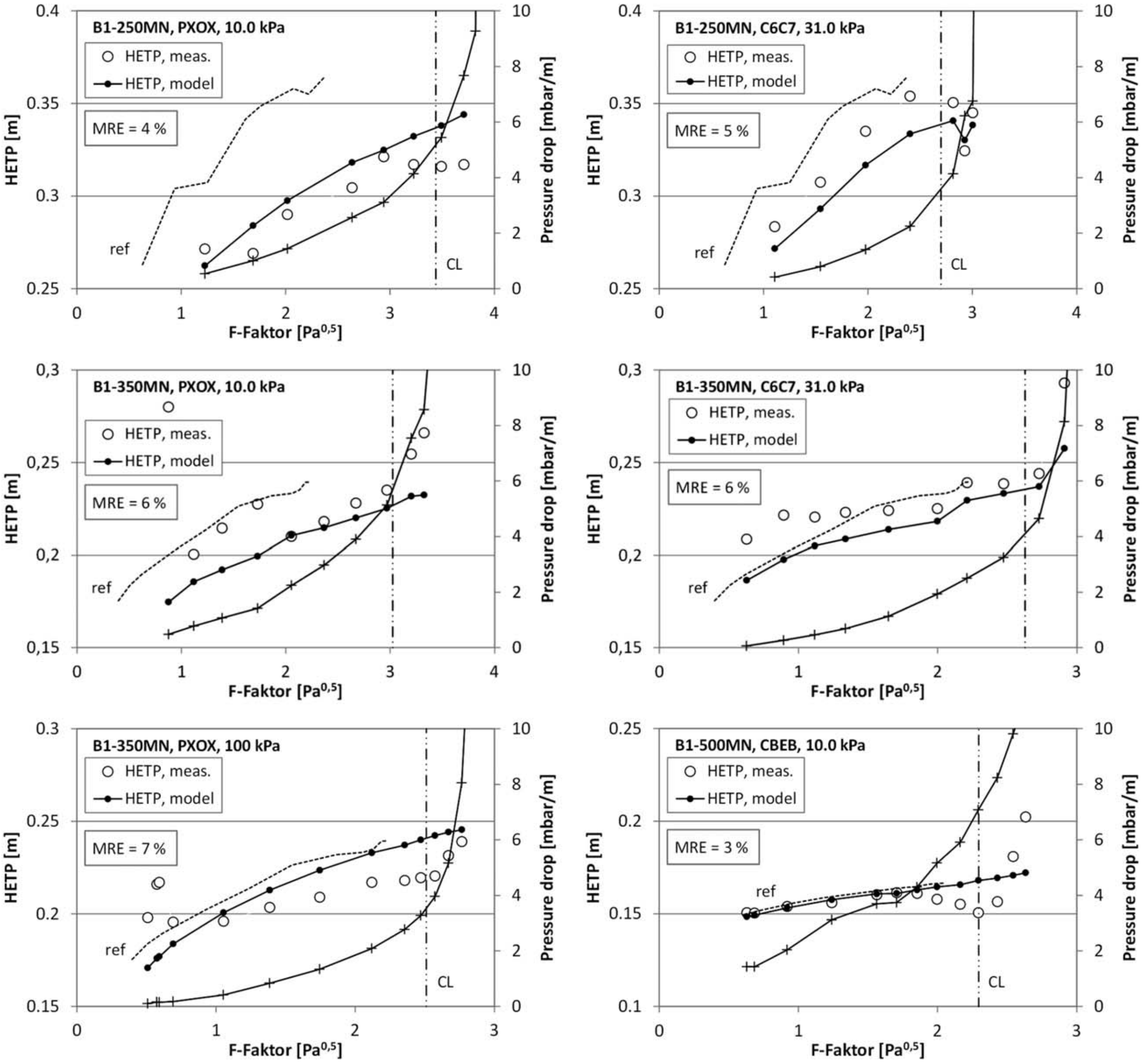

Figure 7. Calculated and measured HETP values and measured pressure drops for test conditions deviating from the regression cases (B1-250MN, B1-350MN, and B1-500MN).

predictions are compared with experimental results which were not used for the fitting. This is done below. For both B1-250MN and B1-350MN packing types, column top pressures are deviating from the regression case by a factor of up to 16. Also, different chemical systems are used. For the case of B1-500MN packing type, the chemical system stays the same and the column pressure differs by a factor of 10. The comparison between calculated and measured HETP values for the six test runs not being part of the regression procedure is shown in Figure 7. All of these diagrams show a dotted reference curve (ref), which is identical with the simulated HETP curves of Figure 6 for the respective case. These curves are included to facilitate the graphical examination of Figures 6 and 7 for the reader. Because of the good agreement between measured and calculated HETP values in Figure 6, the reference curves in Figure 7 also give an impression of how the measured HETP values change for the different operating conditions.

It can be seen that the relative differences between calculated and measured HETP values shown in Figure 7 have an average value of about 5% up to the CL. For the calculation of this average value, only the clearly deviating measurements at low vapor loads for B1-350MN with the PX/OX-system are excluded. The excluded points are seen at the following coordinates (F-factor = 0.88/HETP = 0.28 and F-factor = 0.58/HETP = 0.22).

Discussion

This article gives a methodology of how the RNEQ approach can be used to simulate distillation columns. It is illustrated how experimental results from the preloading region can be used to calculate constants C_A and C_B in Eq. 21 for a specific packing type. Knowing these values, the model can be used to simulate column behavior for that packing type even for process conditions deviating from the original regression case. This is done for three different packing types. It can be seen that the differences between measured results for separation efficiency and calculation results have an average value of 5% up to the CL for test runs not being part of the regression.

Now, the question is how this average relative error is to be evaluated compared to other mass-transfer models. Neither Rocha et al.[14] nor Olujic et al.[12,13] give numerical values about average relative errors for the simulated separation efficiencies. Billet and Schultes,[15] however, mention an average relative deviation of 14% for the calculation of mass-transfer efficiency for the case of distillation with their model equations. At first glance, considering the enormous size of the experimental database used for the BS model, the value of 14% is hard to compare with the relative deviation seen in this work. However, just like the RNEQ approach, the BS model also uses two different empirical constants for each individual packing type in order to calculate separation efficiency of column internals. Keeping this in mind, the relative deviation of 5%, seen in this work for data points not being part of the fitting procedure, seems to be a very good result.

Of course, it should be clear that the model is not suitable for situations in which the basic assumptions are not valid. This means that the approach should not be used for situations in which the mass-transfer resistance is strongly located on the liquid side. Besides that, possible limitations of the RNEQ approach can be classified into two categories: liquid load and wettability of the packing. Liquid load is not directly accounted for within RNEQ equations. Therefore, some effects on separation efficiency, which are explained here with a change in vapor side Reynolds number, can be actually due to changes in liquid load. However, this is considered not to be too problematic, as both vapor and liquid mass flows are strongly related, for distillation processes. Nonetheless, for very low reflux ratios, the effect of reduced liquid flows on separation efficiency should be kept in mind. For these situations, it is advisable to verify the simulation results with additional experiments run at comparable conditions.

The second limitation of the RNEQ approach is related to systems with very high values for liquid viscosity, vapor–liquid surface tension, or liquid–solid contact angle, which can cause strong underwetting effects.[19] As these properties are not used within the RNEQ approach, the reader is encouraged to determine a new set of parameters C_A and C_B for situations in which these properties are assumed to have values strongly deviating from the original regression case. At this point, however, it must be kept in mind that the consideration of these properties within empirical equations should generally be questioned critically. This is due to the fact that their predictive calculation is, for many industrial applications, not at all possible with a high accuracy.[27,41,42]

Also, the following should be kept in mind. It has long been known that packed columns do not show ideal plug flow behavior. A lot of different experimental works give an impression on that. Some of them are referenced here.[36,43–45] As correctly stated by Stichlmair and Fair,[19] this means that parameters describing the separation efficiency of a packing which are derived from experimental column operation always include maldistribution effects. This, of course, is valid for the EQ, NEQ, and RNEQ approach and should be considered for scale-up calculations.

As explained within this article, RNEQ parameters are to be determined for each packing type individually. This might look as a drawback of this modeling approach, because it implies the model's inability to predict the influence of geometrical changes on separation efficiency. However, when it comes to the calculation of separation efficiency for a specific packing type for which some experimental results are available, the flexibility of the RNEQ approach can be regarded as a major advantage over more complex mass-transfer models. Empirical parameters C_A and C_B can easily be determined by the end-user for a specific packing type. Subsequently, these values can be used for the prediction of separation efficiency for deviating process conditions. Depending on the available experimental database, parameters C_A and C_B can be determined for all different situations. Experimental results with test systems can be included as well as data from actual production plants. This is a major advantage over more complex mass-transfer models. As demonstrated by Olujic et al.,[13] the consideration of new experimental results into such models of high complexity might require to drastically alter the form of the empirical equations. This, of course, can hardly be done by the end-user without negatively affecting the model accuracy for the vast amount of experimental results on which these equations are based.

Conclusions

Within this article, the RNEQ approach for the modeling of packed distillation columns is presented. Though based on the direct calculation of mass transfer, this model shows fundamental differences compared to the NEQ approach being used with many conventional mass-transfer correlations. The whole idea of developing a set of empirical parameters and equations simultaneously valid for all types of chemical systems and for all types of structured packings was deliberately put aside. With a clear focus on the reduction of model complexity, the RNEQ model is rather created to allow for the accurate simulation of specific distillation problems. The reduction of model complexity is reached by the neglection of liquid side mass-transfer coefficients. This finally allows to come up with only one empirical equation describing the overall mass transfer instead of three. Within this article, the flexibility of the RNEQ model to account for specific experimental results is demonstrated. After being fitted to experimental results, the model is able to accurately predict separation efficiencies for deviating process conditions.

Summarizing, it can be stated that the presented RNEQ approach definitely widens the range of applicable models in the area of distillation modeling. It is not intended to fully replace either the EQ or the conventional NEQ approach. These models will continue to be used for a lot of interesting tasks. However, especially for the design of industrial distillation columns, the RNEQ approach can play an important complementary role.

Acknowledgments

This work was financially supported by the German Federal Ministry of Economics and Technology as part of the ZIM program (Zentrales Innovationsprogramm Mittelstand, project number: KF 2267706WZ2). The authors want to express their greatest gratitude for this support. The authors also want to thank their project partner Julius Montz GmbH for the possibility to publish experimental data. Finally, the authors thank Prof. Dr.-Ing. Dr. h. c. Dieter Mewes for his support concerning questions about mass transfer modeling.

Notation

a_{eff} = effective interfacial area, $\mathrm{m^2/m^3}$
a_{geom} = geometric packing surface, $\mathrm{m^2/m^3}$
a_{unit} = standard unit area (= 1), $\mathrm{m^2/m^3}$
A = dimensionless prefactor for effective interfacial area
A_{cross} = free cross-sectional area of distillation column, $\mathrm{m^2}$
B_1, B_2 = empirical constants
C_1 C_2, = empirical parameters dependent on packing type
C_A, C'_A, C_B = RNEQ parameters
c_t = molar density, $\mathrm{mol/m^3}$
D = binary diffusion coefficient, $\mathrm{m^2/s}$
F-factor = vapor loading factor, $\mathrm{Pa^{0.5}}$
h_{C} = specific enthalpy of interfacial molar flux of one component, J/mol
h = specific enthalpy of the bulk phase, J/mol
h_{in} = specific enthalpy of stream entering RNEQ stage, J/mol
HETP = height equivalent to a theoretical plate, m
k = mass-transfer coefficient, m/s
K = vapor–liquid equilibrium constant
L = characteristic length, m
L_{unit} = standard unit length (= 1), m
M = molar mass, kg/kmol
m_{yx} = slope of vapor–liquid equilibrium line
n = corrective exponent of Delft model area equations
N = volumetric interfacial molar flux of one component, $\mathrm{mol/(s\ m^3)}$
N_t = sum of volumetric interfacial molar component fluxes, $\mathrm{mol/(s\ m^3)}$
p = pressure, kPa
Q = heat flow rate, J/s
Q_{loss} = heat loss to surroundings, J/s
S_{in} = molar stream entering RNEQ stage, mol/s
S_{out} = molar stream leaving RNEQ stage, mol/s
t_{e} = exposure time, s
u = velocity, m/s
V = volumetric flow, $\mathrm{m^3/s}$
V_{s} = stage volume, $\mathrm{m^3}$
x = liquid molar bulk fraction, mol/mol
x_{int} = liquid molar fraction at phase interface, mol/mol
y = vapor molar bulk fraction, mol/mol
y_{int} = vapor molar fraction at phase interface, mol/mol
z_{in} = molar fraction of stream entering RNEQ stage, mol/mol

Greek letters

α = angle of corrugation channel, deg
α_{L} = effective liquid flow angle, deg
ε = void fraction of packing, $\mathrm{m^3/m^3}$
η = dynamic viscosity, Pa s
θ = contact angle, deg
v = kinematic viscosity, $\mathrm{m^2/s}$
π = circle constant (= 3.14159...)
ρ = mass density, $\mathrm{kg/m^3}$
σ = surface tension, N/m
Γ_1, Γ_2 = empirical terms dependent on packing type and process conditions
Ω = fraction of packing surface area occupied by holes

Subscripts

L = liquid phase
m = mean value
O = overall
Onda = originating from Onda et al.[24]
V = vapor phase

Dimensionless numbers

Fr = Froude number
Ma = Marangoni number
Re = Reynolds number
Sc = Schmidt number
Sh = Sherwood number
We = Weber number

Abbreviations

ACM = Aspen Custom Modeler
BS model = mass-transfer model by Billet and Schultes[15]
BTS = Bayer Technology Services GmbH, Leverkusen, Germany
C6 = cyclohexane
C7 = *n*-heptane
CB = chlorobenzene
CL = capacity limit
Delft model = mass-transfer model by Olujic et al.[12,13]
EB = ethylbenzene
EQ = equilibrium stage
FRI = Fractionation Research, Inc., Stillwater, OK
MRE = mean relative error between measured and simulated HETP value
NEQ = nonequilibrium stage
OX = ortho-xylene
PX = para-xylene
RNEQ = reduced nonequilibrium stage
SRP model = mass-transfer model by Rocha et al.[14]

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Manuscript received May 6, 2014, and revision received July 1, 2014.