

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 (k_G) and non-aqueous (k_L) 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 $k_{L,PM}$, $k_{G,PM}$ under the distillation conditions through analysis of the composition profiles along the packed-bed. Its results come out in the form of multiplicative parameters b_L , b_G correcting first guesses of $k_L a$, $k_G a$ introduced in the model, usually in the form of literature models of $k_{L,LM}$ and $k_{G,LM}$ correlating them with physical properties and phase flows. This procedure is applied mainly due to some changes of $k_L a$ and $k_G a$

along the packed bed, while the profile method has a potential of determination of just single $k_{L,a}$ and $k_{G,a}$ for its entire length.

$$k_L a_{PM} = b_L k_L a_{LM} \quad (1)$$

$$k_G a_{PM} = b_G k_G a_{LM} \quad (2)$$

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

$$Bo_L = 1.022 Fi^{0.478} \left(\frac{3}{\sin^2 \alpha} \right)^{1.159} (d_{eq} a_g)^{6.337} \quad (3)$$

$$Bo_G = 2.807 \cdot 10^6 Re_G^{-0.00895} 10^{-0.00394 Re_L} (d_{eq} a_g)^{-7.792} \quad (4)$$

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 ξ in (5)

$$Bo_L = \xi 1.022 Fi^{0.478} \left(\frac{3}{\sin^2 \alpha} \right)^{1.159} (d_{eq} a_g)^{6.337} \quad (5)$$

Optimized ξ 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

$$Sh_G = 2.7 + 0.038 Re_{LG}^{0.88} Sc_G^\beta \quad (6)$$

$$\beta = 0.33 + 0.9 \exp \{-0.6 Sc_G\} \quad (7)$$

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

$$Sh_L = 0.207 Re^{0.798} Fr^{-0.563} We^{0.511} Sc^{0.528} \quad (8)$$

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.

System	Phase flow rate range		Density		Viscosity		Surface tension
	B [mh ⁻¹]	u _G [ms ⁻¹]	ρ _L [kgm ⁻³]	ρ _G [kgm ⁻³]	η _L [mPa s]	η _G x 10 ⁻² [mPa s]	σ [mNm ⁻¹]
MeOH/EtOH	2 - 14	0.37 – 1.89	743	1.36	0.39	1.05	18.3
EtOH/PrOH	3 - 16	0.34 – 1.74	740	1.77	0.47	0.98	17.8
MeOH/PrOH	2 - 18	0.37 – 1.98	750	1.58	0.43	0.97	18.8
C ₆ /C ₇	5 - 23	0.32 – 1.22	662	3.16	0.27	0.76	14.9

Density, viscosity and surface tension for equimolar mixture

3. Results

3.1 Preliminary analysis of k_{Ga} and k_{La} data evaluated by profile method from distillation experiments

The results of the distillation experiments (k_{Ga}, k_{La}) recalculated to Sh_{La} and Sh_{Ga} 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 Sh_{Ga} data (Figure 1) measured under absorption as well as distillation conditions is 17 %, while distillation Sh_{La} data differ from the absorption ones significantly (average relative deviation 46 %). Very good agreement of absorption and distillation Sh_{Ga} led us to the belief that the Sh_G 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 Sh_G Eq(6), (7) was assumed valid for the distillation process and in the consequence, the corrective parameter b_G in Eq(1) expresses actually the corrective parameter of the distillation effective interfacial area, determining thus its value.

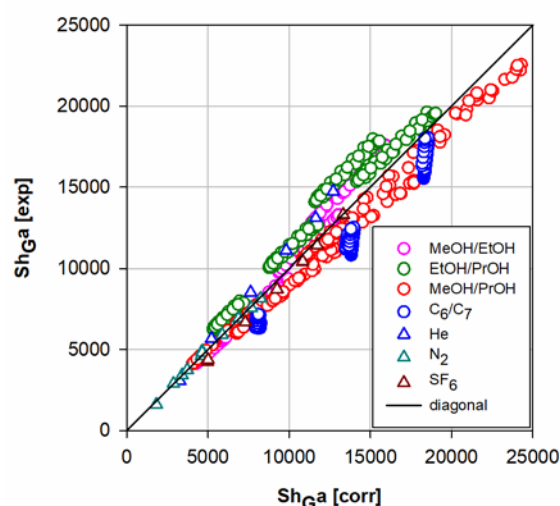


Figure 1: Assessment of the gas-phase mass transfer transferability from absorption to distillation conditions. Comparison of the dist. gas phase mass-transfer data with correlation from the hybrid absorption experiments.

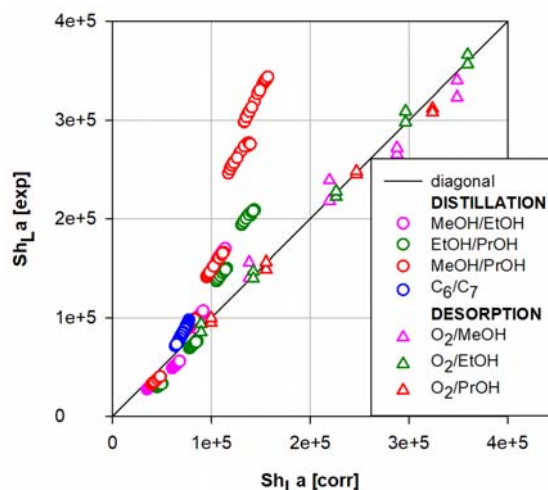


Figure 2: Assessment of the liquid-phase mass transfer transferability from absorption to distillation conditions. Comparison of the dist. liquid phase mass-transfer data with correlation from hybrid absorption experiments.

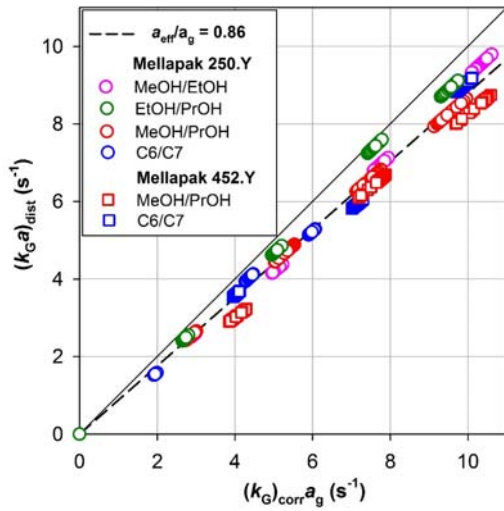


Figure 3: Evaluation of the distillation interfacial mass-transfer area. Comparison of the experimental distillation $k_G a$ values evaluated by the profile method assuming validity of Eq. (6,7 and 8) with $k_G a_G$ values. (Bo_L and a are subject of optimization)

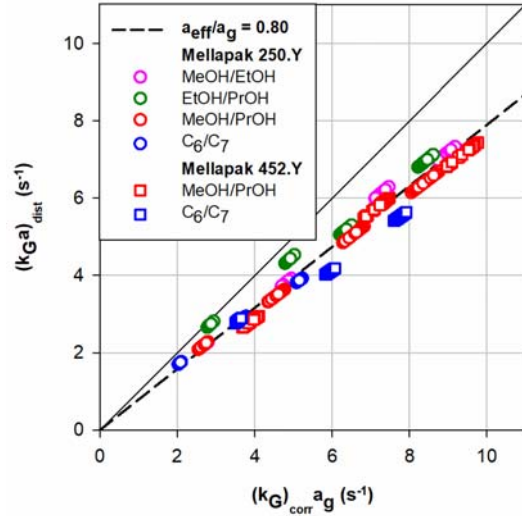


Figure 4: Evaluation of the distillation interfacial mass-transfer area. Comparison of the experimental distillation $k_G a$ values evaluated by the profile method assuming validity of Eq. (6),(7) with $k_G a_G$ values. (Bo_L , k_L and a are subject of optimization)

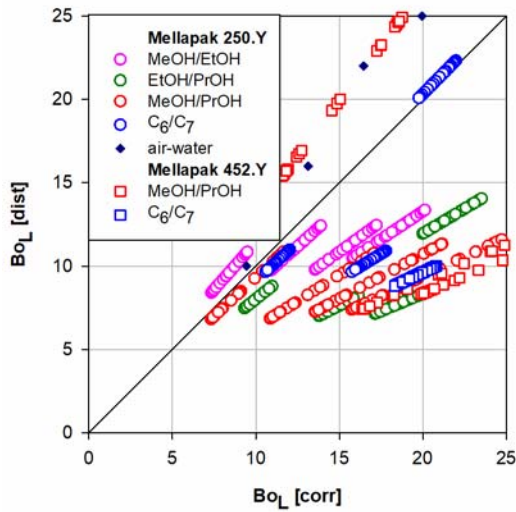


Figure 5: Axial mixing in the liquid phase under distillation conditions. Values of the liquid phase Bodenstein number, Bo_L , evaluated from the distillation experiments compared the results of the original correlation Eq. (3). Data for single packing and system are provided for several reboiler duties.

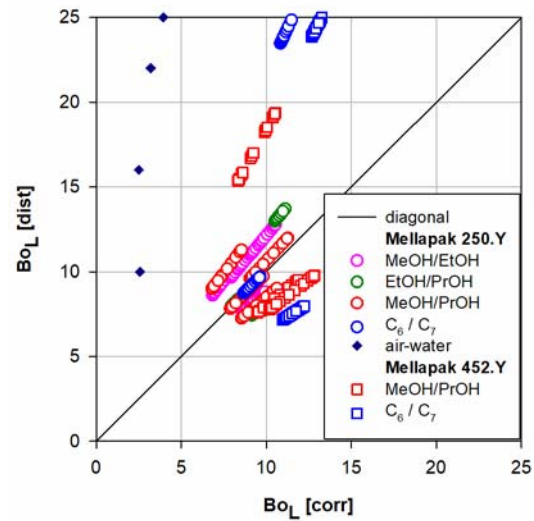


Figure 6: Axial mixing in the liquid phase under distillation conditions. Values of the liquid phase Bodenstein number, Bo_L , evaluated from the distillation experiments compared to the results of the newly developed correlation Eq. (11).

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 $k_{GA_{PM}}$ 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 Bo_L has been evaluated from the distillation experiments by the profile method in the form of the multiplicative parameter ξ according to Eq(5) and are compared with the results of original correlation Eq(3) in Figure 5. It can be observed, that the original correlation substantially overestimates the Bo_L values (experimentally found values are lower – i.e. the axial mixing is higher) and it also overestimates dependence of the Bo_L values on the liquid phase flow. For most experimental conditions the 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.

$$Bo_L = f(Re, Fr, We) \quad (9)$$

Correlation, with α according to the packing type, provided

$$Bo_L = \alpha Re^{-0.168} Fr^{-0.923} We^{1.097} \quad (10)$$

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

$$Bo_L = \alpha Fi^{0.178} E\ddot{o}^{1.097} \quad \text{Mellapak 250.Y: } \alpha = 0.78; \text{ Mellapak 452.Y: } \alpha = 1.76 \quad (11)$$

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 ($k_{L,a}$, $k_{G,a}$, 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 $k_{L,a}$ value to infinity assumes no mass-transfer resistance in the liquid phase. Setting of the 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 2: Relative effects of the parameters $k_{L,a}$, $k_{G,a}$, Bo_L on the packed bed height H , needed to separation of the mixture from 0.9 to 0.1 in mole fractions.

P [kW]	system	packing	H [m]	Relative effect - parameter [%]		
				$k_{L,a}$	$k_{G,a}$	Bo_L
10	MeOH/EtOH	Mellapak 250.Y	2.35	7	61	32
40	MeOH/EtOH		2.74	0	77	23
10	MeOH/PrOH		1.07	14	60	26
40	MeOH/PrOH		1.34	0	82	18
8	C ₆ /C ₇		3.10	23	52	25
24	C ₆ /C ₇		2.91	0	90	10
10	MeOH/PrOH	Mellapak 452.Y	0.88	19	62	19
30	MeOH/PrOH		0.99	2	68	30

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

$$Bo_L = \alpha Fi^{0.178} E\ddot{o}^{1.097} \quad \text{Mellapak 250.Y: } \alpha = 0.78; \text{ Mellapak 452.Y: } \alpha = 1.76$$

has been developed on the basis of the distillation data. The gas phase mass-transfer is modeled using relations

$$Sh_G = 2.7 + 0.038 Re_{LG}^{0.88} Sc_G^\beta; \beta = 0.33 + 0.9 \exp \{-0.6 Sc_G\}$$

derived from the hybrid absorption experiments, which were found transferable to the distillation conditions. The liquid phase mass transfer correlation

$$Sh_L = 0.207 Re^{0.798} Fr^{-0.563} We^{0.511} Sc^{0.528},$$

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

a_{eff}	effective interfacial area [m^2m^{-3}]	C_6/C_7	Cyclohexane – n-Heptane
a_g	geometrical area of the packing [m^2m^{-3}]		
d_{eq}	packing characteristic dimension [m]	Indexes	
Sh	Sherwood number	PM	Profile method
Sc	Schmidt number	LM	literature model
Fi	Film number $Fi = u \eta / (\rho g d_{\text{eq}}^2)$	L	liquid phase
$E\ddot{o}$	Eötvös (Bond) number $E\ddot{o} = \rho g d_{\text{eq}}^2 / \sigma$	G	vapour phase

Reference

- Haidl, J., Rejl, F. J., Valenz, L., Moucha, T., Petříček, R., 2017, General mass-transfer model for gas phase in structured packings, *Chemical Engineering Research and Design*, 126, 45 – 53.
- Linek V., Moucha T., Prokopová E., Rejl F. J., 2005, Simultaneous determination of vapour and liquid-side volumetric mass transfer coefficients in distillation column, *Chemical Engineering Research and Design*, 83(A8), 979 – 986.
- Macias-Salinas R., Fair J. R., 1999, Axial mixing in modern packings, gas and liquid phases: I. Single-phase flow, *AIChE Journal*, 45, 222 – 239.
- Rejl F. J., Linek V., Moucha T., Prokopová E., Valenz L., Hovorka F., 2006, Vapour- and liquid-side volumetric mass transfer coefficients measured in distillation column. Comparison with data calculated from absorption correlations, *Chemical Engineering Science*, 61, 6096 – 6108.
- Rejl F. J., Linek V., Moucha T., Valenz L., 2009, Methods standardization in the measurement of mass-transfer characteristics in packed absorption columns, *Chemical Engineering Research and Design*, 87, 695 – 704.
- Rejl F. J., Valenz L., Linek V., 2010, “Profile Method” for the Measurement of $k_L a$ and $k_V a$ in Distillation Columns. Validation of Rate-Based Distillation Models Using Concentration Profiles Measured along the Column, *Industrial & Engineering Chemistry Research*, 49, 4383 – 4398.
- Rejl, F. J., Haidl, J., Valenz, L., Marchi, A., Moucha, T., Petříček, R., Brunazzi, E., 2017, Liquid-phase mass-transfer coefficients of Mellapak structured packings under desorption of oxygen from primary alcohols, *Chemical Engineering Research and Design*, 127, 1-9.
- Valenz L., Rejl F. J., Linek V., 2010, Gas and Liquid Axial Mixing in the Column Packed with Mellapak 250Y, Pall Rings 25, and Intalox Saddles 25 under Flow Conditions Prevailing in Distillation Columns, *Industrial & Engineering Chemistry Research*, 49, 10016–10025.
- Valenz L., Rejl F. J., Linek V., 2011, Effect of Gas- and Liquid-Phase Axial Mixing on the Rate of Mass Transfer in a Pilot-Scale Distillation Column Packed with Mellapak 250Y, *Industrial & Engineering Chemistry Research*, 50, 2262 – 2271.