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Random Packing Modelling under High Pressure for Gas-Liquid Systems

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This work presents a design principle for the prediction of the separation efficiency of random, non-perforated and lattice-type, packing with different size and type for gas-liquid systems under high pressure up to 90 bar for distillation and absorption systems in the entire operating range up to the flooding point.

In the first part the known correlations from literature were used for calculation of separation efficiency of different random packings for distillation data taken from literature, valid for the systems used under higher pressure. As a conclusion of this analysis, big deviations of calculation results in comparison to experimental data were observed, Hanley & Chen (2012).

The application of the new droplet model presented by Maćkowiak (2015) was validated for more than 24 test systems and 115 packing types in the low pressure range from 13 to 1000 mbar. In this work the model was validated using additionally 10 different test systems in the pressure range up to 90 bar. The presented correlation does not require empirical, individual packing specific constants, which is a very important advantage of this model for practical design of packed columns.

Good consistency was found between experimental values of the separation efficiency and calculated values based on the model for different types of random packings of 1st and 2nd generation such as Berl Saddle, Raschig-Ring, Pall-Ring but also for lattice type packing of 3rd and 4th generation.

1. Introduction

Random packings are widely and successfully used in distillation, absorption and stripping processes but also in environmental protection processes like waste gas and wastewater treatment. Their form has changed significantly during the last decades. The field and range of applications as well as their efficiency compared to the 1st generation of packings have significantly increased. A big progress in the available design methods was observed during that period of time, especially in the modelling of fluid dynamics, which is presented in the literature Maćkowiak (2010).

One of the first mass transfer models for the design of random packing of 1st and 2nd generation were the models of Onda et al. (1968), Zuiderweg (1978) and Zech & Mersmann (1978). The first model for the design of distillation columns not only for classical random packings but also for *lattice type* packings of the 3rd generation was the model developed by Billet & Maćkowiak (1984), which has been modified by Maćkowiak (1990). The model from Maćkowiak (1990) was developed for more than 60 different packings for systems with the main mass transfer resistance located in the vapor phase. Based on the assumption of film formation in the packed bed, Billet & Schultes (1993, 1999) derived correlations for determining the effective interfacial area per unit volume a_e and mass transfer coefficients in the gas and liquid phase for 55 different classical and lattice-type packings. These correlations are mainly applicated to systems at ambient or low pressure.

The main aim of this work is to present the application of droplet model from Maćkowiak (2015) for determining the separation efficiency n_t/H or HTU_{OV} for distillation and absorption systems under higher pressure. The value of the overall vapor height of transfer unit HTU_{OV} is calculated by Eq(1), which can be transformed to theoretical stages per meter n_t/H according to Eq(2).

$$HTU_{OV,cal} = \frac{u_v}{\beta_V \cdot a_e} + \frac{m_{yx}}{\dot{L}/\dot{V}} \cdot \frac{u_L}{\beta_L \cdot a_e} \qquad [\mathrm{m}] \qquad (1)$$

$$\left(\frac{n_t}{H}\right)_i = \left(\frac{1}{HTU_{OV}}\right) \cdot \left(\frac{\lambda + 1}{2}\right) \qquad [\mathrm{n/m}] \qquad (2)$$

In Table 1 exemplary the resulting separation efficiency $(n_t/H)_{exp}$ from experimental distillation data with metallic 50 mm Pall-Rings, published by Schultes (2003, 2013), are compared to the calculation using known correlations from literature by deviation of separation efficiency $\delta_i\left(\frac{n_t}{H}\right)_i$ to experiment (Eq(3)).

$$\delta_\mathrm{i}\left(\frac{n_t}{H}\right) = \frac{\left(\frac{n_t}{H}\right)_{cal} - \left(\frac{n_t}{H}\right)_{exp}}{\left(\frac{n_t}{H}\right)_{exp}} \cdot 100\ \% \qquad [\%] \qquad (3)$$

Table 1: Deviation of separation efficiency of known correlations $\delta(n_t/H)_i$ to published experimental data from Schultes (2003, 2014) $(n_t/H)_{exp}$ below loading point ($F_V/F_{V,Fl} \leq 0.65$) at total reflux for two systems with higher pressure in 50 mm Pall-ring packings.

System	p_T [bar]	Schultes (2003, 2014) $(n_t/H)_\mathrm{exp}$ [1/m]	Onda et al. (1968) $(n_t/H)_\mathrm{cal}$ [1/m]	Zuiderweg (1978) $(n_t/H)_\mathrm{cal}$ [1/m]	Zech&Mersmann (1979) $(n_t/H)_\mathrm{cal}$ [1/m]	Maćkowiak (1990) $(n_t/H)_\mathrm{cal}$ [1/m]	Billet & Schultes (1993, 1999) $(n_t/H)_\mathrm{cal}$ [1/m]
iso-butane/n-butane; $F_V = 0.85\ Pa^{0.5}$							
	11.4	2.44	1.65	2.61	10.12	2.72	6.9
$\delta_i(\mathrm{n_t}/H)_\mathrm{i}$			-32.4%	7.0%	314.6%	11.6%	182.6%

Table 1 showing that the use of various known and common correlations resulting in significant deviations compared to experimental data from literature (Schultes 2003, 2014). The same observation was made by Hanley & Chen (2012). Consequential the droplet model from Maćkowiak (2015), validated at low and ambient pressure, will be reviewed at high pressure systems.

2. Model validation

The existing ENVIMAC data bank (EDB) was used to check the model presented in Maćkowiak (2015) for 23 different test systems for 115 packing types of 1st – 4th generation with size of d= 8 to 100 mm made of metal, ceramic and plastic. The total number of experimental data evaluated within this work was extended to about 200 additionally data sets for high pressure distillation systems and absorption systems. The 9 additional test systems and their physical properties are presented in Table 2.

Table 2: The physical properties of investigated systems for distillation and absorption, high pressure range

System	p_T [bar]	T_S [K]	ρ_V [kg/m³]	$\nu_V \cdot 10^6$ [m²/s]	$D_V \cdot 10^6$ [m²/s]	Sc_V [-]	ρ_L [kg/m³]	$\nu_L \cdot 10^6$ [m²/s]	$\sigma_L \cdot 10^3$ [kg/s²]	$D_L \cdot 10^9$ [m²/s]	Sc_L [-]
cyclohexane/n-heptane	1.6	378	4.97	1.65	2.18	0.756	641	0.35	13.0	6.19	57.2
iso-butane/n-butane	6.9	329	15.9	55.3	0.63	0.878	521	0.22	8.0	12.96	16.9
iso-butane/n-butane	11.4	351	28.7	33.5	0.41	0.816	487	0.18	5.0	17.67	10.3
methanol/N_2-CO_2	10.0	240	14.4	1.1	1.13	0.946	841	1.81	26.8	2.64	685
methanol/N_2-CO_2	20.0	240	29.0	0.54	0.56	0.959	841	1.81	26.2	2.64	685
methanol/N_2-CO_2	30.0	240	43.9	0.36	0.37	0.964	842	1.81	25.6	2.64	684
methanol/N_2-CO_2	50.0	240	74.5	0.22	0.22	0.993	842	1.81	24.4	2.64	684
methanol/N_2-CO_2	70.0	240	105.7	0.16	0.16	1.032	842	1.81	23.2	2.64	684
methanol/N_2-CO_2	90.0	240	137.0	0.13	0.12	1.083	842	1.81	22.0	2.64	684

The EDB contain various data for higher operating pressures up to 90 bar, e.g experimental data of Krehenwinkel (1986), Schultes (2003, 2014) and others. To predict the efficiency of random packing n_t/H or HTU_{OV}, see Eq(1) and Eq(2), the knowledge of the liquid and gas phase volumetric mass transfer coefficients $(\beta_L \cdot a_e)$ and $(\beta_V \cdot a_e)$ are required. For the calculation of both products the model presented earlier by

Maćkowiak (2015) was used, see Eq(4) and Eq(5). The liquid hold-up is an important factor for mass transfer and is calculated according to Eq(6), valid below loading point.

$$(\beta_L \cdot a_e) = \frac{15.1}{C_T \cdot (1-\varphi_P)^{\frac{1}{3}} \cdot d_h^{\frac{1}{4}}} \cdot \left(\frac{D_L \Delta\rho g}{\sigma_L}\right)^{0,5} \cdot \left(\frac{a}{g}\right)^{\frac{1}{6}} \cdot u_L^{\frac{5}{6}} \qquad [1/s] \qquad (4)$$

$$(\beta_V \cdot a_e) = 6 \cdot \frac{h_L}{d_T^2} \cdot D_V \cdot \left(2 + 0.0285 \cdot \frac{u_R \cdot d_T}{\nu_V} \cdot \left(\frac{\nu_V}{D_V}\right)^{\frac{1}{3}}\right) \cdot \left(1 - \frac{h_L}{\varepsilon}\right)^6 \qquad [1/s] \qquad (5)$$

$$h_L = 0.57 \cdot Fr_L^{\frac{1}{3}} = 0.57 \cdot \left(\frac{a u_L^2}{g}\right)^{\frac{1}{3}} \; ; \mathrm{F_V} \leq 0.65\, \mathrm{F}_{\mathrm{V},Fl} \qquad [m^3/m^3] \qquad (6)$$

Furthermore the operating pressure and physical properties have significant influence on the droplet size and their stability. For higher pressure and low surface tension of the system the droplet size increases, what was observed and reported in the literature by Braun (1990) and Billet & Maćkowiak (1984). This fact is taken into account by using the correction factor C_T, shown in Figure 1, to calculate the droplet size d_T at different pressure conditions by Eq(7).

$$d_T = C_T \cdot \sqrt{\frac{\sigma_L}{\Delta\rho \cdot g}} \qquad [m] \qquad (7)$$

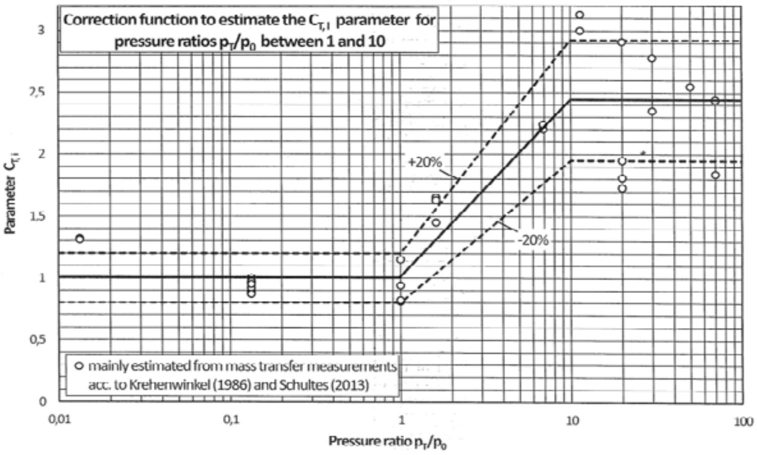
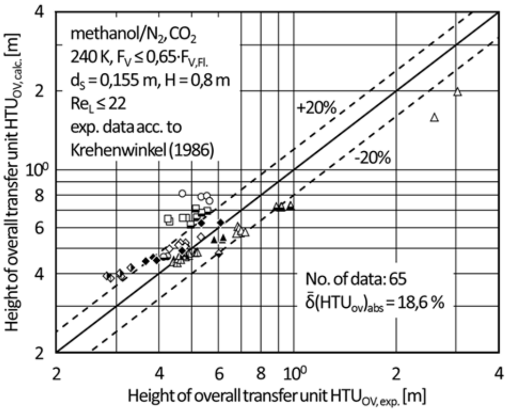


Figure 1: Correction factor C_T for droplets at various pressures from EDB.



<table>
<tr><td>Packing</td><td colspan="6">PR15-PP</td></tr>
<tr><td>N [1/m³]</td><td colspan="6">247600</td></tr>
<tr><td>Symbol</td><td>◭</td><td>△</td><td>▲</td><td>◬</td><td>⟁</td><td>◮</td></tr>
<tr><td>p_T [bar]</td><td>10</td><td>20</td><td>30</td><td>50</td><td>70</td><td>90</td></tr>
<tr><td>Packing</td><td colspan="6">BS15-C</td></tr>
<tr><td>N [1/m³]</td><td colspan="6">228000</td></tr>
<tr><td>Symbol</td><td>⬙</td><td>◇</td><td>◆</td><td>-</td><td>◈</td><td>⬗</td></tr>
<tr><td>p_T [bar]</td><td>10</td><td>20</td><td>30</td><td>50</td><td>70</td><td>90</td></tr>
<tr><td>Packing</td><td colspan="2">BS10-C</td><td colspan="2">PR15-M</td><td>RR15-M</td><td>IP20-M</td></tr>
<tr><td>N [1/m³]</td><td colspan="2">630000</td><td colspan="2">210000</td><td>239800</td><td>202500</td></tr>
<tr><td>Symbol</td><td colspan="2">◒</td><td colspan="2">⬓</td><td>□</td><td>○</td></tr>
<tr><td>p_T [bar]</td><td colspan="2">20</td><td colspan="2">20</td><td>20</td><td>20</td></tr>
</table>

Figure 2: Experimental verification of the model presented in this work for absorption system under high pressure up to 90 bar for different random packing. (PR: Pall-ring; BS: Berl saddle; RR: Raschig-ring; IP: Interpack; PP: plastic; C: ceramic; M: metal)

Figure 2 shows the comparison between the experimentally determined HTU_{OV} to the calculated results, according to the presented model above. The model is valid for the experimental absorption system methanol/N_2-CO_2 of Krehenwinkel (1986) in the operating pressure range up to 90 bar for different random packing, such as 10-20 mm metallic and plastic Pall-rings, ceramic Berl Saddles and metallic Interpack.
The estimated mean deviation between experimental and calculated values for the evaluated packings of 1st and 2nd generation is 18.6 %. This implies a good agreement of the model to high pressure systems.

3. Comparison of the presented model with correlations from literature

3.1 Absorption systems

The results of the evaluation of the absorption data from the EDB-databank for gas-liquid systems with different correlations from literature is shown as an example in *Table 3*.

Table 3: Results of experimental data of absorption in methanol/N_2-CO_2-system Krehenwinkel (1986) under higher pressure up to 90 bar & comparison with correlation from literature and this work.

System: *Methanol/ N_2-CO_2*	p_T [bar]	Experiment $HTU_{OV,exp}$ [m]	Onda et al. (1968) $\delta_i(HTU_{OV})_i$ [%]	Zuiderweg (1978) $\delta_i(HTU_{OV})_i$ [%]	Zech & Mersmann (1979) $\delta_i(HTU_{OV})_i$ [%]	Billet & Schultes (1993, 1999) $\delta_i(HTU_{OV})_i$ [%]	Maćkowiak (this work) $\delta_i(HTU_{OV})_i$ [%]
15 mm Pall ring, metal; $u_L = 31.9\,m/h,\ F_V = 0.7\,Pa^{0.5}$							
	20	0.56	44.4	-27.3	18.4	-44.3	25.1
15 mm Raschig ring, metal; $u_L = 18.0\,m/h,\ F_V = 0.4\,Pa^{0.5}$							
	20	0.48	41.2	-15.6	26.2	-	35.7
20 mm Interpak, metal; $u_L = 22.8\,m/h,\ F_V = 0.5\,Pa^{0.5}$							
	20	0.56	13.8	-19.5	16.7	-	34.5
10 mm Berl saddle, ceramic; $u_L = 18.4\,m/h,\ F_V = 0.4\,Pa^{0.5}$							
	20	0.42	33.3	-32.2	41.4	-35.6	9.9
15 mm Berl saddle, ceramic; $u_L = 17.2\,m/h,\ F_V = 0.4\,Pa^{0.5}$							
	20	0.46	63.5	-15.2	44.9	-31.3	12.3
	30	0.47	55.7	-21.9	41.1	-34.6	3.8
15 mm Pall-ring, plastic, $u_L = 21\,m/h,\ F_V = 0.5\,Pa^{0.5}$							
	20	0.70	24.4	-39.9	-24.9	-56.2	-16.6
	50	0.49	66.3	-17.7	-1.8	-47.0	-4.4
	70	0.46	82.6	-7.6	8.1	-42.9	0.0
	90	0.48	82.1	-10.7	8.0	-44.6	-2.2
Abs. mean deviation [%]			42.8	22.0	72.5	65.7	15.8

The model presented in this work allows the prediction of the separation efficiency HTU_{OV} in the absorption system methanol/N_2-CO_2 below the loading point with best accuracy in comparison to other models taken into account.

3.2 Distillation systems

Furthermore a generally better prediction of the separation efficiency in distillation processes is achieved compared to classical correlations like that of Onda et al. (1968), Zuiderweg (1978), Zech & Mersmann (1978) or Billet & Schultes (1993, 1999). This can be confirmed by the data listed in Table 4, which shows a comparison of calculated separation efficiencies to experimental distillation data of Schultes (2003). It becomes apparent that all correlations are useful for calculation of separation efficiency for classical packings, but the model presented in this work leads to best accuracy and is also valid for modern random packings.

4. Final remarks

This work showed the prediction of separation efficiency columns with random packings according to the equilibrium n_t/H-model or the transfer stage $HTU_{OV} \cdot NTU_{OV}$ –model. Different correlations from literature were compared to the newly developed model by Maćkowiak (2015). It was shown that the droplet flow model allows the prediction of the separation efficiency in packed column filled with random packing, for all investigated types of packings from 1st to 4th generation in the entire operation range from vacuum

(Maćkowiak (2015)) up to 90 bar (this work) below loading point with a satisfactory accuracy for practical applications. Furthermore applicability to absorption and distillation processes where confirmed. The use of individual packing constants for mass transfer prediction is not necessary in the presented model, what distinguishes the presented model from other models in literature and is a big advantage for design of columns. *Table 5* shows the validity range of the presented model for the investigated systems.

Table 4: Comparison of the model prediction with literature correlations with experimental data of Schultes (2003,2013) for 25-50 mm metallic Pall rings and SRR-rings-no.2, d_s=1.2 m;

Experiments $\left(\frac{n_t}{H}\right)_{exp}$ [1/m]	Onda et al. (1968) $\delta_i\left(\frac{n_t}{H}\right)_i$ [%]	Zuiderweg (1978) $\delta_i\left(\frac{n_t}{H}\right)_i$ [%]	Zech & Mersmann (1979) $\delta_i\left(\frac{n_t}{H}\right)_i$ [%]	Maćkowiak (1990) $\delta_i\left(\frac{n_t}{H}\right)_i$ [%]	Billet & Schultes (1993, 1999) $\delta_i\left(\frac{n_t}{H}\right)_i$ [%]	Maćkowiak (this work) $\delta_i\left(\frac{n_t}{H}\right)_i$ [%]
<u>50 mm Pall ring, metal</u>						
cyclohexane/n-heptane; $F_V = 1.49\ Pa^{0.5}, p_T = 1.6\ bar$						
1.73	-5.8	14.2	151.5	25.7	111.7	23.9
iso-butane/n-butane; $F_V = 0.85\ Pa^{0.5}, p_T = 11.4\ bar$						
2.44	-32.4	7.0	314.6	11.6	182.6	12.4
<u>Raschig Super-ring No. 2, metal</u>						
cyclohexane/n-heptane; $F_V = 1.49\ Pa^{0.5}, p_T = 1.6\ bar$						
1.73	-0.7	-3.3	95.0	13.9	106.2	21.6
	iso-butane/n-butane		$F_V = 1.24\ Pa^{0.5}, p_T = 6.9\ bar$			
2.00	-21.1	-2.7	172.0	10.2	128.7	-1.6
	iso-butane/n-butane		$F_V = 1.13\ Pa^{0.5}, p_T = 1.6\ bar$			
2.44	-35.7	-9.3	222.9	8.6	178.9	12.0
<u>25 mm Pall ring, metal</u>						
cyclohexane/n-heptane; $F_V = 1.13\ Pa^{0.5}, p_T = 1.6\ bar$						
3.15	21.1	-18.6	48.9	-5.4	24.3	-12.0
Abs. mean deviation [%]	19.5	12.1	167.5	12.6	122.1	13.9

Table 5: Proven validity range of presented model.

d	0.008 – 0.090 m	ρ_V	0.076 – 105,7kg/m³	$F_V/F_{V,FL}$	0.90
a	54.2 – 550 m²/m³	ρ_L	487 – 1100 kg/m³	p_T	0.013 – 90 bar
ε	0.696 – 0.987 m³/m³	σ_L	5 – 72.7 mN/m	$Re_L = \frac{u_L}{a\,\nu_L} \in \langle 0 - 150\rangle$	
d_S	0.10 – 1.4 m	$Sc_L = \frac{\nu_L}{D_L} \in \langle 10 - 10000\rangle$		$Re_V = \frac{u_V d_P}{(1-\varepsilon)\nu_V} K \in \langle 400 - 17500\rangle$	
d_s/d	6	$Sc_V = \nu_V/D_V \in \langle 0.4 - 1.25\rangle$			
H	0.8 – 4 m				

Symbols

a	[m²/m³]	geometric surface area of packing per unit volume
a_e	[m²/m³]	interfacial area per unit volume
C_T	[–]	correction factor droplet, Eq(2)
d	[m]	packing diameter
d_h	[m]	hydraulic diameter; $d_h = 4 \cdot \varepsilon/a$
d_S	[m]	column diameter
d_P	[m]	Partial size of packing; $d_p = 6 \cdot (1-\varepsilon)/a$
d_T	[m]	mean droplet diameter
D_V, D_L	[m²/s]	diffusion coefficient in the gas or liquid phase
F_V	[Pa$^{0.5}$]	gas load factor in relation to full column cross section, $F_V = u_V \cdot \sqrt{\rho_V}$
g	[m/s²]	gravitational acceleration
h_L	[m³/m³]	liquid hold-up in relation to total packing volume V_S
HTU_{OV}	[m]	height of an overall transfer unit related to vapor phase
H	[m]	height of packed bed

K	[–]	wall factor, acc. to [1]
l	[m]	mean contact path
$\dot{L}$	[kmol/h]	molar flow of liquid
m_{yx}	[-]	slope of equilibrium line
n_t/H	[1/m]	number of theoretical stages per 1 m packing height
NTU_{OV}	[–]	number of overall transfer units, related to vapor phase
p_0	[bar]	ambient pressure
p_T	[mbar, bar]	operating pressure
u_L	[m/s]	specific liquid load in relation to full column cross section
u_R	[m/s]	relative phase velocity $u_R = \frac{u_V}{\varepsilon - h_L} + \frac{u_L}{h_L}$
u_V	[m/s]	linear gas velocity in relation to full column cross section
V_L	[m^3]	liquid volume
V_S	[m^3]	packing volume, $V_S = H \cdot \pi \cdot d_S^2 / 4$
$\dot{V}$	[kmol/h]	molar flow of gas or vapor
Greek symbols		
$\beta \cdot a_e$	[1/s]	volumetric mass transfer coefficient
$\delta_i(\dots), \bar{\delta}(\dots)$	[%]	relative error, middle value of relative error
λ	[-]	stripping factor $\lambda = m_{yx} \cdot \dot{V} / \dot{L}$
φ_P	[-]	packing form factor acc. to (Maćkowiak 2010)
$\rho, \Delta\rho$	[kg/m^3]	density, density difference $\Delta\rho = \rho_L - \rho_V$
σ_L	[N/m]	surface tension of liquid
ν	[m^2/s]	kinematic viscosity
Indices		
L		relating to liquid
S		relating to operating point at loading point; $F_V = 0.65 \cdot F_{V,Fl}$
V		relating to gas or vapor

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