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EXPERIMENTAL INVESTIGATION ON THE EFFECT OF PACKING MATERIAL TEXTURES ON THE LIQUID-SIDE MASS TRANSFER

Martin Kohrt[1], Ilja Ausner[2] and Jens-Uwe Repke[1]
[1] Technical University Berlin, Chair of Process Dynamic and Operation; Strasse des 17. Juni 135; 10623 Berlin; Germany; Martin.Kohrt.1@tu-berlin.de
[2] Sulzer Chemtech Ltd; R&D Laboratory Mass Transfer Technology, Winterthur, Switzerland

Abstract
In this work, the impact of different packing material textures to liquid-side controlled mass transfer for the CO_2 absorption into silicone oil is analysed. Typical industrially applied textures with bidirectional pyramidal and unidirectional wavy topographies are investigated for a wide range of liquid loads and viscosities. It is found that the texture design has a significant influence on the mass transfer efficiency, e.g. mass transfer intensification up to 80% can be performed by a textured surface compared with a flat plate.

Keywords: Structured packings, textured surface, falling film absorption, liquid-side mass transfer, mass transfer intensification

1. Introduction
The efficiency of distillation and absorption processes depends on different factors. One important criterion is the choice of the geometry of column internals like structured packings where the gas and liquid phase are brought in contact and heat and mass transfer take place. Several authors have already investigated e.g. the effect of packing corrugations on mass transfer, but less knowledge concerning the influence of the packing texture or grooving on the mass transfer is available.

In recent and former research activities textured or corrugated surfaces are investigated theoretically[1-4] and experimentally[5-8], but the effect of surface textures on film flow or rivulets is analysed mainly with focus on the fluid dynamics. The influence on mass transfer is only studied for flat plates regarding e.g. to different liquid properties, counter-current flow and wave characteristics[9-14]. It is known that due to the texture the wetting can be clearly improved but its influence on the mass transfer is still unknown. To bridge this scientific gap, absorption measurements on different textured surfaces are carried out in order to investigate the effect of textured surfaces on the liquid-side controlled mass transfer. Preparing these measurements, it is necessary to identify a suitable test mixture, to provide the needed physical properties and to enable reproduction of mass transfer experiments which is done with an inclined steel plate as benchmark[15]. As test system, the absorption of gaseous carbon dioxide into silicone oil is chosen. Silicone oil provides the benefits of good liquid spreading on different solid surfaces and selectable viscosities. Three different oils are used to cover a viscosity range of 1-10 x10^{-6} m²/s, which is the most suitable range for different industrially applied solvents. CO_2 is absorbed physically in the oils without any chemical reaction. The gas phase consists of almost pure carbon dioxide in order to reduce mass transfer resistances only to the liquid phase ($k_G >> k_L$).

In this paper, the experimental procedure, liquid properties and types of textured surfaces for the mass transfer determination will be presented first. Then, a unifying mass transfer correlation for three different silicone oils is developed for a flat plate. Finally, the effect of the used textured surfaces to liquid-side mass transfer is presented for a variety of process parameters and their importance for process design is discussed.

2. Experimental set up
Mass transfer measurements are accomplished for falling film absorption on an inclined plate with an inclination angle of 30° to the vertical, see figure 1. The geometry of a plate was chosen to enable a detailed observation of the liquid flow behaviour for future campaigns as done e.g. in[10, 16-17]. An overflow weir is used to ensure uniform wetting of the plate. The gas absorption is done at constant conditions of head pressure (PIR01=1.053±0.001 bar absolute), outlet temperature (TIR03=28.5±0.2 °C) and base area (0.1 m x0.3 m). Since the gas solubility and the heat of mixing of carbon dioxide in

silicone oil are small, the temperature gradient over the plate length of 0.3 m can be neglected and isothermal absorption is assumed.

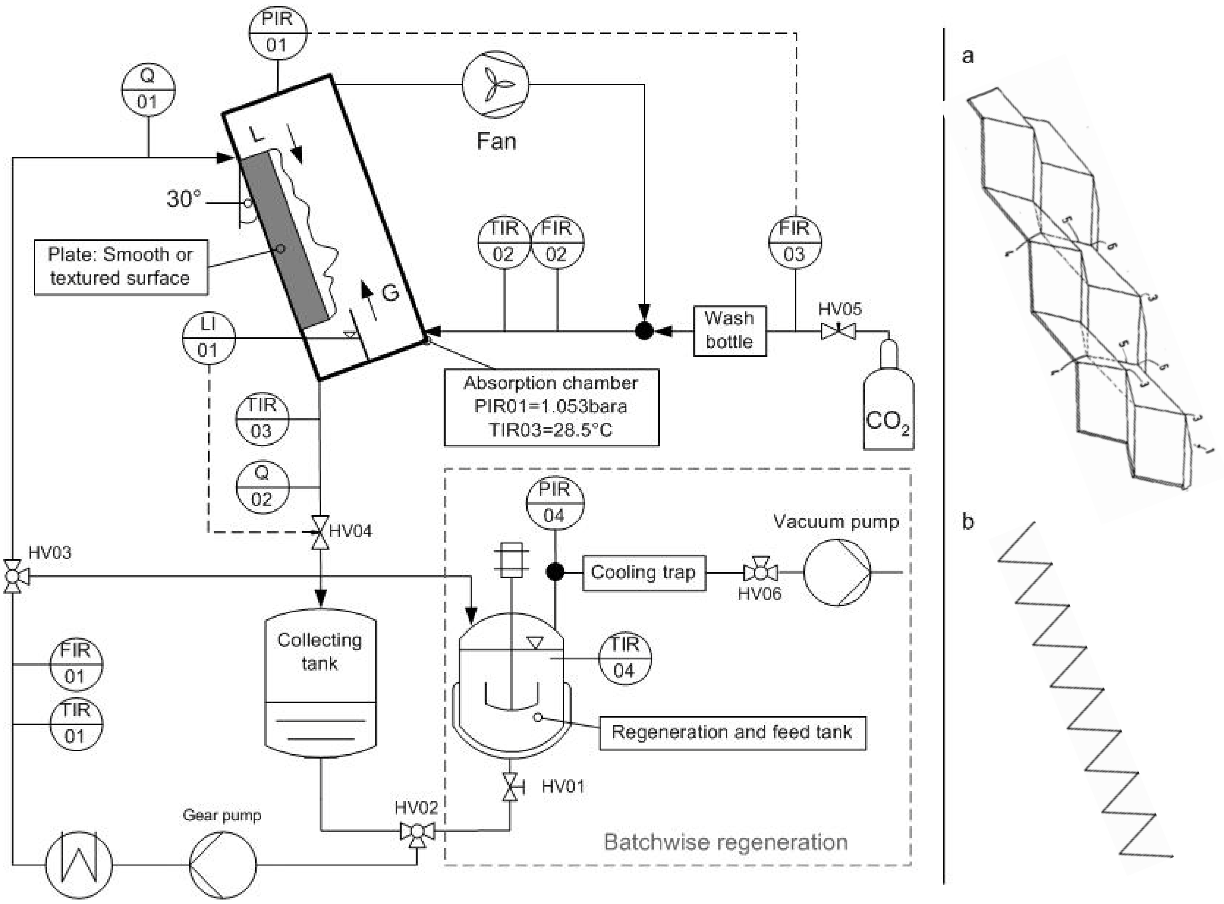

Figure 1. Schematic view of the test rig for falling film absorption and batchwise regeneration. Bidirectional[18] (a) and unidirectional (b) texture are shown schematically on the right hand side.

The type of silicone oil (Dow Corning Fluid 200® with different viscosities), liquid load and counter-current gas flow will be changed to study the influence on the mass transfer efficiency for different textured surfaces. The gas flow is expressed by the F-factor which varies between 0 and 2.5 $Pa^{0.5}$, see tab.1. The liquid flow rate $\dot{V}_L$ is transferred to the liquid load referring to the structured packing Mellapak 250.X® by Sulzer Chemtech Ltd, which has a surface area of approx. 250 m²/m³. Using the plate width of 0.1 m, $\dot{V}_L$ is computed with 2500 times the liquid load. This is leading to film Reynolds numbers and Nusselt film thicknesses[19] depicted in tab.1 for the experimental campaign:

Table 1. Varying process data for absorption operation.

Silicone oil	F-factor [$Pa^{0.5}$]	Liquid load [m³/m²h]	*Re* [-]	Nusselt film thickness δ_N [mm]
DC 10	0; 1.6; 2.5	25-90	2-10	0.55-0.80
DC 5	0; 1.6; 2.5	25-90	5-20	0.45-0.65
DC 1	0; 1.6	30-90	30-100	0.20-0.30

In addition to the falling film absorption, regeneration or degassing of the liquid is necessary for its reuse. The degassing of the loaded silicone oil is done batchwise in a stirred 20 l vessel while using a standardized procedure, where the pressure will be reduced to 40 mbar absolute. With this procedure the CO_2 concentration in the liquid is reduced to only some ppm. Therefore, it can be set to zero in comparison to the concentration at the end of the plate after absorption (‰-range). Within the experimental space the working liquid properties are varied systematically by changing the viscosity, see table 2:

Table 2. Physical properties of pure silicone oil (* measured at 28.5 °C and 1 atm).

Silicone oil	Viscosity* ν [m²/s]	Density* ρ_L [kg/m³]	Surface tension* σ [mN/m]	Diffusivity[15] (25°C) $D_{CO2,oil}$ [m²/s]	*Sc* [-]	*Fi* [-]
DC 10	$9.9 \cdot 10^{-6}$	935	18.9	$1.27 \cdot 10^{-9}$	7800	$8.8 \cdot 10^{4}$
DC 5	$4.9 \cdot 10^{-6}$	915	18.3	$1.81 \cdot 10^{-9}$	2700	$1.4 \cdot 10^{6}$
DC 1	$0.97 \cdot 10^{-6}$ [20]	812	16.0	$4.30 \cdot 10^{-9}$	226	$8.8 \cdot 10^{8}$

In table 2, Schmidt number ($Sc=\nu/D_{CO2,oil}$) and Film number ($Fi=\sigma^3/g\rho_L^3\nu^4$) are introduced. Further dimensionless numbers used in this paper are film Reynolds number ($Re=\dot{V}_L/\nu\, w_{plate}$)[6] and Sherwood number ($Sh=k_L\,\delta/D_{CO2,oil}$). Here, the width of the plate w_{plate}=0.1m and the characteristic length $\delta \approx \delta_N$. The mass transfer rate $\dot{M}_{CO2} = \rho_L \dot{V}_L (X_{CO2,out} - X_{CO2,in})$ can then be obtained by analysing the liquid samples at the inlet (Q01) and outlet (Q02) with Fourier Transform Infrared Spectroscopy. While using the equilibrium data of CO_2 solubilities in silicone oil[15], the following equation for the liquid-side mass transfer coefficient can be applied[11, 13]:

$$k_L = \frac{\dot{V}_L}{A} \cdot \ln\left(\frac{X_{CO2,eq}}{X_{CO2,eq} - X_{CO2,out}} \right) \tag{1}$$

Where it is assumed, that the gas liquid interface is equal to the area *A* of the plate (0.03 m²). This assumption is reasonable for the flat plate due to the fact that the low surface tension and contact angle of silicone oil ensures complete wetting in the operating range. In fact, the occurring waves increase the gas liquid interface only by a few percent[14].

2.1 Textured surfaces

As denoted in fig.1, the plate itself can be changed while using the same absorption chamber. Therefore, different surfaces can be installed and analysed within the working space. At first, a flat stainless steel plate is used as reference. Additionally, conventional packing material textures are used, i.e. bidirectional pyramidal structures with crossing grooves (bd-type) and unidirectional textures with transversal waves (ud-type). These and slightly modified geometrical types can be found as a texture or grooving in almost every common high performance packing on the market, which is of great industrial interest.

Here, the ud-type has longitudinal waves with small amplitudes and perpendicularly orientation to the flow direction. This texture has comparable dimensions as the packing surfaces used by Zhao and Cerro[5] and Ataki et al.[8]. In contradiction to the bd-type, the film flow has to flow over the longitudinal waves[5], i.e. there are no dry spots in the operating range for the ud-type. Since the bd-type consists of crossing grooves, there are channels which provide a good cross mixing and therefore an excellent wetting even for difficult systems like aqueous solutions and stainless steel[18]. The amplitude for the bd-type is in all cases bigger than the Nusselt film thickness in table 1, thus channel flow can be expected as the dominant flow pattern and dry peaks are possible. More details about the geometry of the bd-type are documented in the patent by Pluess[18].

Since the interfacial area between gas and liquid were not predicted for the textures yet, equation (1) has to be written in the form of the volumetric mass transfer coefficient (k_LA) for result discussion. This equation will be used for the flat plate only to develop a suitable Sh-correlation.

3. Results and discussion

Before discussing the differences in mass transfer between flat and textured surfaces, dimensionless mass transfer results for the three oils will be presented for the reference case of the flat plate.

3.1 Mass transfer correlation

In general, it is difficult to predict mass transfer characteristics precisely for new working pairs while using film theory, penetration theory or empirical correlations. It was shown in[15] that well established correlations for aqueous systems in the form $Sh = a \cdot Re^b Sc^{0.5}$ cannot be applied for silicone oil because the surface tension is not taken into account. Furthermore, experimentally determined *Sh* for all used silicone oils are in the same range despite the big differences in *Re*. Following the idea to take the surface tension with *Fi* into account[11], the following unifying mass transfer correlation for the falling film absorption of CO_2 in silicone oil is proposed:

$$Sh = a \cdot Re^b Sc^{0.5} Fi^{-0.25} \tag{2}$$

The parameter *a* and *b* are changing with the flow regime, e.g. laminar, wavy or turbulent flow, which depends on *Re* and *Fi* [21-22]. In fact, plotting all results of the accomplished mass transfer measurements for the wavy flow regime, equation (2) can be fitted with a very high accuracy (96%) for all the three different viscous oils, see figure 2.

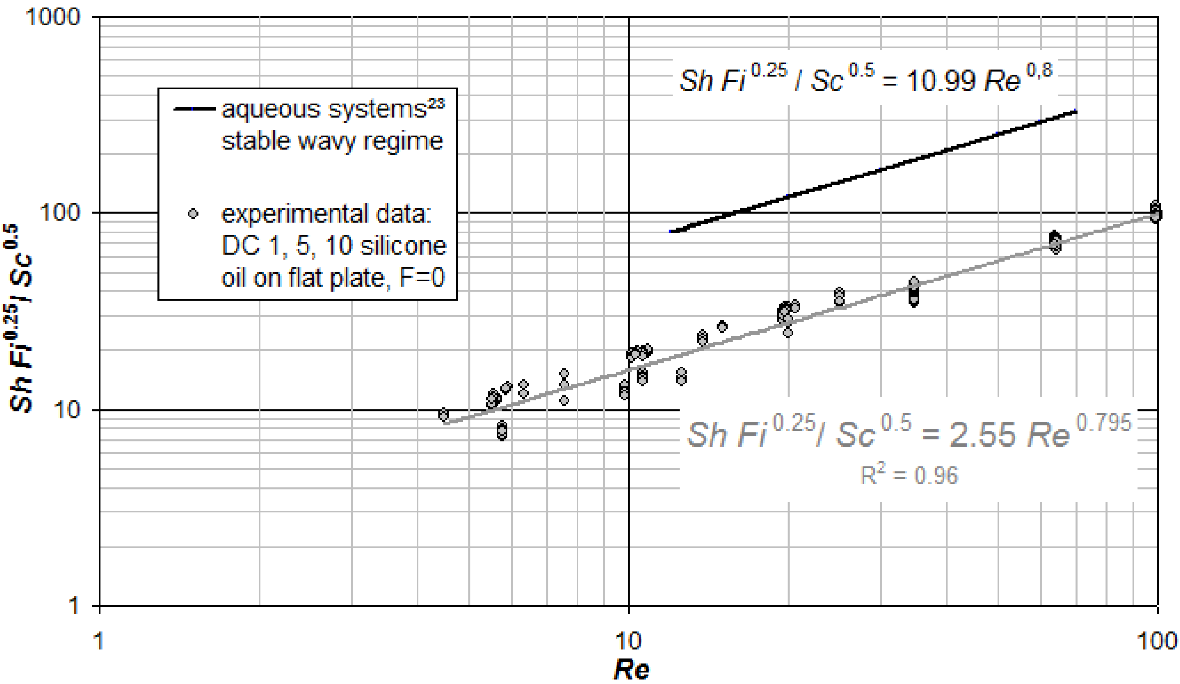

Figure 2. Dimensionless plot of falling film absorption results for flat plate within the stable wavy flow regime referring to Al-Sibai[22]. Additionally, a plot of aqueous systems taken from the literature[23] is shown for comparison reasons (Fi_{water}=5.8 x10^{10}).

Using the parameters *a*=2.55 and *b*=0.795, the maximum deviation is about 30% in fig.2. In comparison to aqueous systems, the Re-exponent is almost the same (b_{water}=0.8) in the stable wavy pattern, but four times higher absolute values in fig.2 occur for water compared to silicone oil at the same *Re* (a_{water}≈11). To sum up, equation (2) is unifying the mass transfer characteristics for different viscous silicone oils with high accuracy, but other systems may not been predicted precisely as can be seen e.g. for water. In future work a more general correlation has to consider the surface tension in a different manner.

3.2 Influence of packing's texture

The characterization of the mass transfer performance for flat and textured surfaces will be done with the volumetric mass transfer coefficient (k_LA) as mentioned above. Although tests with different F-factors were accomplished, the most interesting result is shown in this paper: the impact of the textured surfaces in dependency of the liquid load without counter-current gas as depicted in fig.3. It can be seen in fig.3 that for the low viscosity oil (DC 1) the highest percentage changes appear in the volumetric mass transfer coefficient. For the bd-type, (k_LA) increases up to 80% for low liquid loads and 30% for high loads in comparison to the flat plate. The mass transfer enhancement due to the bd-type is clearly higher than the error bars which represent the variance of several measurement points from different batches. Despite dry peaks for this type with losses in the interfacial area, advantageous phenomena like cross mixing and channel flow yield the observed performance. The ud-type is closer to the flat plate, but especially for low liquid loads an enhancement can be clearly identified (approx. 25%) for DC 1. For the maximum load, the ud-type is only 5% above the flat plate which can be neglected compared with the variance. This behaviour can be explained by the amplitude of the lamella structure which is at small flow rates close to the Nusselt film thickness. With growing flow rates the texture amplitude becomes small in comparison to the film thickness, thus its influence on the gas-liquid mass transfer becomes small as well.

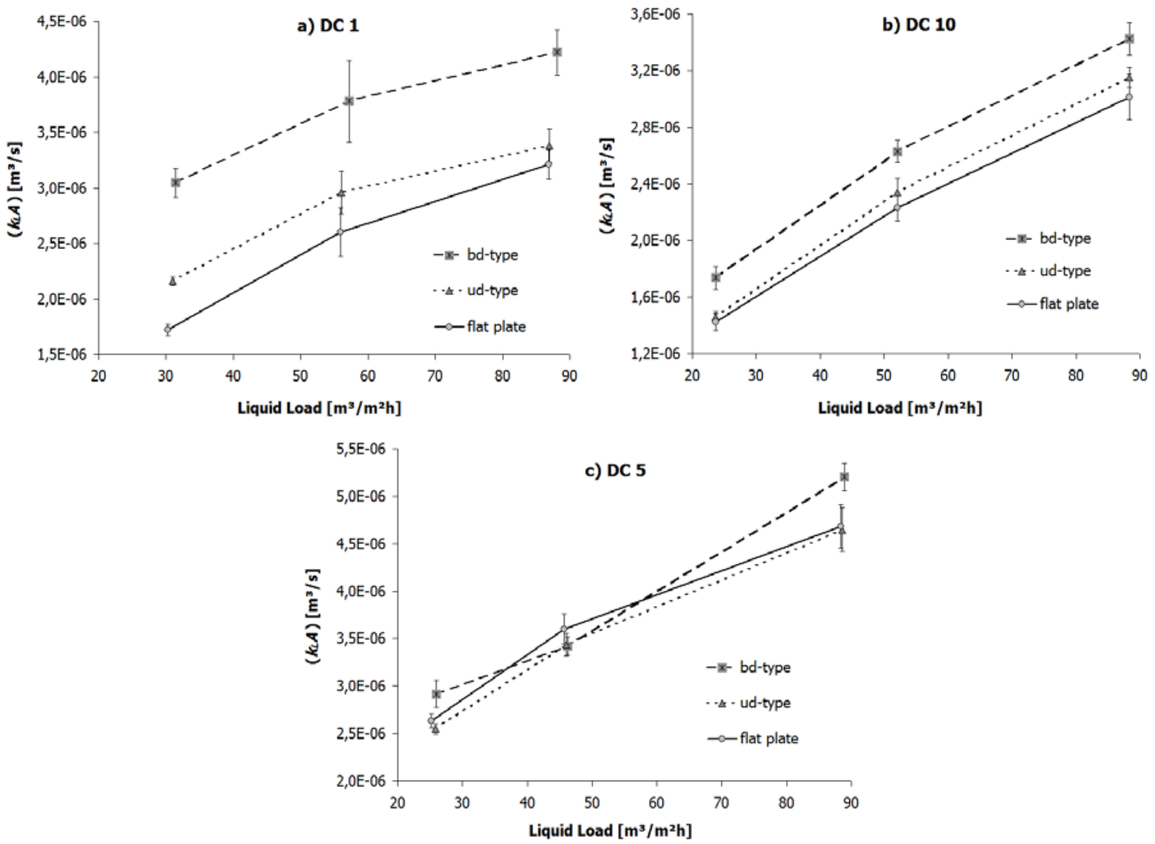

Figure 3. Volumetric mass transfer coefficients for different surfaces without counter-current gas flow for low viscosity a), high viscosity b) and moderate viscosity oil c).

This trend for the ud-type can be seen also for higher viscous silicone oils. For the cases in fig.3b-c), the higher viscosities yield to higher film thicknesses for the same volume flow, see tab.1, so the amplitude of the ud-type is maximum one-fifth of the film thickness. For this reason, there is no significant difference between the flat plate and the ud-type using DC 5 and DC10 oils. Fig.3b) shows for the bd-type that even for relatively thick films a mass transfer enhancement of 15-20% can be achieved compared to the other surfaces. Here, the film thickness is slightly lower than the pyramids amplitude. Therefore, channel flow effects (increased surface velocities) and mass transfer enhancement are smaller compared to the case in fig.3a). The bd-type in fig.3c) shows for low and high liquid loads analogous behaviour as for DC10 oil, i.e. (k_LA)-values are 10% higher in comparison to the flat plate. However, the bd-type has slightly lower performance compared to the flat plate for liquid loads of about 46 m³/m²h. This results in a different slope for the bd-type in comparison to all other cases in fig.3a-c). The reasons for this observed behaviour have to be found in the complex effects of the texture's topography on flow patterns. These flow patterns have to be investigated in future campaigns by analysing the fluid dynamics with non-intrusive flow measurements like film thickness and velocity measurements as described in[16-17].

CONCLUSION

Falling film absorption of gaseous CO_2 in different silicone oils is accomplished in order to investigate the effect of textured surfaces of structured packing material on liquid-side mass transfer in comparison with a flat inclined plate. The used liquids cover a wide range of technical relevant solvents; and liquid loads are chosen as under typical industrial operating conditions. The textures consist of bidirectional and unidirectional types which are the most common textures in practice.
It was known that textured surfaces improve the wetting of liquids in many cases, but a positive impact on the mass transfer, independently from wetting, could at least be assumed. Now it is shown experimentally that textured surfaces can lead to a significant increase of liquid-side controlled mass transfer. This positive effect is most dominant for lower viscosities (approx. 1 $x10^{-6}$m²/s) with approx. 80% enhancement compared to the flat plate and it can be identified even for relatively high viscosities (approx. 10 $x10^{-6}$m²/s).

The conclusion from Zhao and Cerro[5] that the influence of textures is most interesting with ratios of film thickness and amplitude between 0.1 and 1 (inferred from fluid dynamic tests) can be confirmed with the performed mass transfer measurements. Beside clear mass transfer enhancement also deterioration of performance can be observed for special combinations of working liquid and texture type.

LITERATURE

1. S.J. Baxter et.al, *Int. J. Numer. Meth. Fluids* (2009), DOI: 10.1002/fld.2029
2. H. Luo. and C. Pozrikidis, *Acta Mechanica* 188 (2007) 209-225
3. P. Valluri, O.K. Matar, G.H. Hewitt, M.A. Mendes, *Chem. Eng. Sci*. 60 (2005) 1965-1975
4. Y.Y. Trifonov, *J. Applied Mech. & Tech. Phys*. 45 (2004) 389–400
5. L. Zhao and R.L. Cerro, *Int. J. Multiphase Flow* 18 (1992) 495-516
6. K. Helbig et.al, *J. of Heat Transfer* 131 (2009) 011601
7. K. Argyriadi, M. Vlachogiannis, V. Bontozoglou, *Phys. of Fluids* 18 (2006) 012102
8. Ataki, P. Kolb, U. Bühlmann, H.-J. Bart, Proceed. of Distillation and Absorption (2006) 534-543
9. S.V. Alekseenko, S.E. Nakoryakov, B.G. Pokusaev, Wave flow of liquid films, Begell House (1994)
10. S. Paschke, R. Thiele, J.-U. Repke, 8th World congress of Chem. Eng. in Montréal (2009)
11. Y.S. Won and A.F. Mills, *Int. J. Heat and Mass Transfer*. Vol. 25 (1982) 223–229
12. L.M. Yu, A.-W. Zeng, K.T. Yu, *Ind. Eng. Chem. Res*. 45 (2006) 1201-1210
13. Z.F. Xu, B.C. Khoo, N.E. Wijeysundera, *Chem. Eng. Sci.* 63 (2008) 2559-2575
14. S.M Yih and K.Y. Chen, *Chem. Eng. Commun*. 17 (1982) 123-136
15. M. Kohrt, I. Ausner, J.-U. Repke, G. Wozny, 5th IBW on Transport Phen. Moving Bound. (2009)
16. Ausner, A. Hoffmann, J.-U. Repke, G. Wozny, 3th IBW on Transport Phen. Moving Bound. (2005)
17. M. Kohrt, S. Paschke, J.-U. Repke, G. Wozny, 12th Workshop on Transport Phen. in Two-phase flow (2008) 27-34
18. R. Plüss, European Patent 0190435, 1989
19. W. Nusselt, VDI-Zeitschrift 60 (1916) 541-546
20. S.B. Gunst, *Trans. Am. Soc. Mech. Eng*. 72 (1950) 401-407
21. S. Ishigai, S. Nakanisi, T. Koizumi, Z. Oyabu, Bulletin of the JSME, 15 (1973) 594-602
22. F. Al-Sibai, PhD-thesis RWTH-Aachen, Germany (2005)
23. M. Kraume, *Transportphänomene in der Verfahrenstechnik*, Springer (2004)