ON THE TRACK TO UNDERSTANDING THREE PHASES IN ONE TOWER

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Packed towers are widely used for distillation processes due to their higher separation efficiency and capacity in comparison to tray towers. The separation efficiency of the tower strongly depends on the liquid flow behaviour inside the packing, which becomes more complex with the appearance of a second liquid phase. Such flow behaviour can be observed in the three-phase distillation process in packed towers, where two immiscible liquid phases flow down the packing co-currently and a vapour phase flows in counter-current flow. For a better understanding of this process it is necessary to investigate the three-phase flow behaviour inside packed towers in detail. Numerical investigations are carried out with CFD and are validated with own flow measurements.

KEYWORDS: multiphase flow, three phase distillation, tower performance, packings, CFD

INTRODUCTION

Packed towers as they occur in distillation and absorption processes mainly depend on the flow behaviour inside the packing elements. The flow is governed by the liquid and vapour loads as well as system parameters like the contact angle [1]. The complex behaviours of film break-up, rivulet and droplet formation play an important role. The problem gets even more complicated when there is more than one liquid phase apparent, e.g. in the distillation of heterogeneous azeotropic mixtures. Because of the complex fluid dynamics, the separation performance of such towers is hardly predictable; therefore substantial work is needed [2]. Recent investigations of three different packings with four heterogeneous mixtures show no deterioration of the performance for relevant f-factors. A detailed description of these investigations would go beyond the scope of this paper. Further information can be found in [3].

For the performance understanding, a detailed knowledge of the fluid dynamics on the packing surface is urgently required. In general, it can be observed that one liquid phase forms a closed film on the packing while the other one forms rivulets and droplets, which move below and above the film surface. In a distillation or absorption tower a vapour phase is in counter-current flow to the liquid which has an effect on the multiphase film flow. Since the interfacial areas between all phases are significant for transport phenomena, the determination of the interfacial areas is one goal of this contribution. Another objective is the investigation of the flow performance, i.e. mainly the spreading of the liquids on the packing and the velocity of each phase. These investigations are carried out numerically using the commercial CFD-package CFX 5, ANSYS Inc., as well as experimentally with optical measurement methods like particle tracking velocimetry (PTV) and light induced fluorescence (LIF).

For first investigations the packing is simplified and substituted by a flat inclined plate of stainless steel, which can be modelled more easily and allows a free optical access to the flow for measurements. Additionally, in the first stage of the investigation the vapour phase is neglected. The vapour field is initially motionless and dragged by interfacial forces. With the experimental data the numerical models are validated and prepared for the simulation of complex structured packings, which can be investigated experimentally only with great endeavours.

EXPERIMENTAL METHODS

The experimental setup is shown in Figure 1. The two immiscible liquids water and toluene are pumped separately with peristaltic pumps out of a separation tank and are delivered into feed tubes. A feed tube is a horizontal adjusted tube with several holes, which let out the containing liquid (one tube for each liquid) as jets on to the plate. With a variable inclination angle for example of 45° or 60° to the horizontal, the plate of stainless steel leads the liquids back to the separation tank at the end of the plate. The flow rates are measured with two rotameters.

Perpendicular to the plate a CCD-camera is installed, which takes pictures of the flow behavior and the spreading of both liquids. Tracers on the liquid surfaces are double exposed with a strobe light and tracked with the camera. The images are analysed with a PTV method [4], which gives the information about the surface velocity for each phase. Additionally, a LIF method [5] is applied for the measurements of the fluid thicknesses. Fluorescent dyes are solved into the liquids and excited with light of a specific wavelength. The emitted fluorescent light intensity increases proportional to the fluid thickness and is detected with the camera.

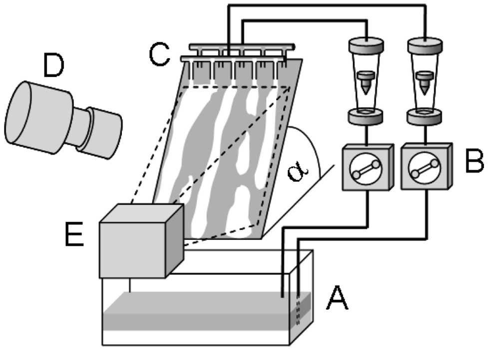

Figure 1. Experimental setup with separation tank (A), peristaltic pumps and rotameters (B), feed tubes (C), camera (D), and light source (E)

More details of the experimental setup and the implemented measurement technique can be found in [6].

NUMERICAL METHODS

Since multiphase film flows deal with closed film flow including film break-up as well as rivulet and droplet flow, the numerical simulation makes high demands on the simulation tool. That requires a three-dimensional and transient model, which is implemented in CFX 5. It uses an Euler algorithm with one set of equations for all phases. Like the VOF method [7] the phases are implemented with averaged phase fractions for each finite volume cell, i.e. one velocity and pressure field for all phases. The interfaces then are defined at specific phase fractions.

Surface tensions are implemented with the continuous surface force model [8] and contact angles for each phase are given as wall boundary condition. The liquid inlets are realized as jet-inlets, which lead to the same boundary condition as the experimental setup. Left and right hand side boundaries are defined as wall boundaries, since in the experimental setup there are the plate supports. Additionally, the mesh near the plate has to be very fine for accurate resolution of the liquid flow. With variations of the grid size near the plate it was found that a resolution of ten cells for the film height is sufficient.

RESULTS AND DISCUSSION

First of all, morphological comparisons between simulation and experiment are carried out, which show qualitatively the ability of CFD to simulate complex free surface effects like film break-up or rivulet and droplet flow. In Figure 2 images taken from the single and multi-liquid flow in simulation and experiment are shown. The comparison of a single liquid phase (Figure 2a) is taken from a water flow at $\mathrm{Re} = 165$ on an inclined steel plate. As can be seen, the morphology of the water film break-up in simulation and experiment is almost identical. The more complex flow behaviour of the water-toluene flow is shown in Figure 2b. While toluene forms a more or less closed film that covers nearly the whole plate, the water flow breaks up and forms rivulets, which move below the toluene phase, and droplets, which mainly flow on the toluene phase. This flow behaviour is observed in the experiments as well as in the simulations. The morphological agreement between simulation and experiment for the case of multiphase flow is not as perfect as it was found in the single liquid phase flow. Nevertheless, the simulation agrees sufficiently well with the experiment that predictions of the flow behaviour for other sets of parameters (e.g. flow in complex geometries) will be successful.

The next step is the quantitative comparison between simulation and experiment. One quantity, which can be determined with this special experimental setup, is the surface velocity. The surface velocity represents the maximum velocity of a liquid phase and can be extracted out of the simulations as well. In the measurements a factorial design of the experiments was used to identify significant parameters, which influence the surface velocity of a liquid phase. With that linear relationships between the varied parameters (water and toluene flow rates) and the objective quantities toluene surface velocity

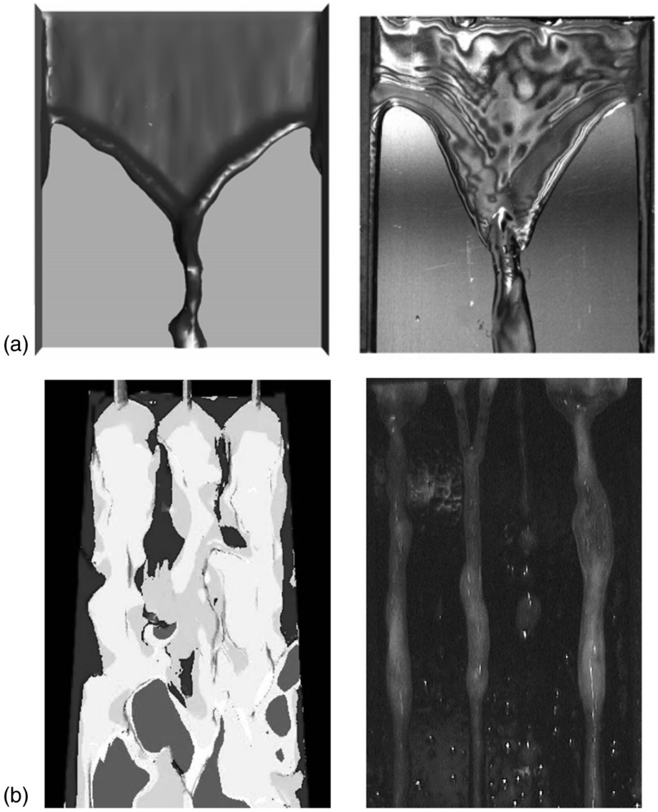

Figure 2. Morphological comparison between simulation (left) and experiment (right) for a) the single-liquid water flow and b) the multi-liquid flow of water (blue) and toluene (white)

and water surface velocity were developed [9]. These relationships now can be used for the validation of the simulations. A comparison between simulated and relationship-wise computed surface velocities for different sets of flow rates is shown in Figure 3. The flow rates are represented by the Reynolds number $\mathrm{Re} = Q/b{\cdot}v$ for each phase, which is defined by flow rate Q, the width of the plate b, and the kinematic viscosity v.

In Figure 3 simulated surface velocities are tagged by the dotted lines while the solid lines indicate the surface velocities calculated with the experimentally determined relationships. While the solid lines represent the linear assumption of the relations, the dotted lines between the simulated points help to visualize the trends.

In general, it can be found that a simulated value is always close to the experimental based calculated value for a set of Re-numbers and the deviation in the considered range of

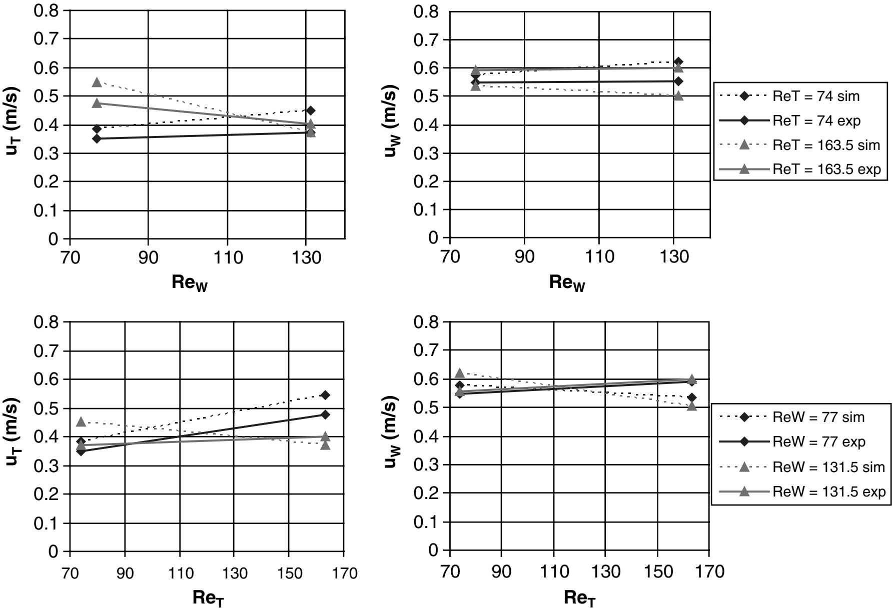

Figure 3. Comparison of surface velocities for different Re-numbers between simulation (sim) and experiment (exp)

Re-numbers is quite small. The simulated trends agree with the measured trends quite well – except for the diagram of water surface velocity u_W plotted against the toluene Re-number Re_T, but the variation is very small and in the precision of measurements range. Very surprising is the observation that some surface velocities are decreased when the flow rate is increased. This can be seen for instance in the diagram of the toluene surface velocity u_T as a function of the water Re-number Re_W: on a low level of Re_T the toluene surface velocity increases slightly with an increase of Re_W. But on a higher level of Re_T the toluene surface velocity is decreased with an increase of Re_W, and this trend is found in the simulations as well as in the experiments.

This result demonstrates that there must be a strong interaction between both liquid phases, which even leads to a deceleration of a liquid phase although the flow rate is increased. Another effect that indicates strong interactions between the phases is the stabilization effect, which can be observed mainly in the experiments: a water flow on a steel plate at low flow rates forms a rivulet flow that meanders transiently down the plate. After the addition of the toluene phase to the flow, the water rivulets spontaneously form straight rivulets with a very fixed and stationary path. Due to the presence of the toluene phase the highly non-stable water flow is stabilized and decelerated in comparison to the surface velocity of the single phase flow.

The stabilization effect of both liquids can be found also in the simulations. Water rivulets are not that straight as in the experiments, but the formation of single and separated rivulets can be observed as well. Additionally, the deceleration of the liquid phases in the multiphase flow compared to the single liquid phase flow is obtained, too: single water or toluene flows move with higher surface velocities than both phases together in the multiphase film flow [10].

Measurements of the liquid thicknesses also show that the toluene phase forms a very large contact area with the water flow and with that high friction forces can affect. For the thickness measurements a two-phase LIF method is developed, which is able to measure both phases separately, as can be seen in Figure 4. Figure 4a shows the bright toluene phase, which covers the whole plate and forms an interfacial boundary along the dark water rivulets. The interface of the water rivulets in Figure 4a is much brighter than the film between the rivulets. This indicates that toluene forms meniscuses on the borders of water rivulets, which leads to larger contact areas between the toluene and the water phase compared to the case of a flat toluene film without meniscuses. In Figure 4b mainly the bright water rivulets with different thicknesses can be seen, while the toluene phase between the water rivulets is very dark. These detected pixel intensities are calibrated for each phase using prismatic cells with defined fluid thicknesses, so that one detected pixel intensity is assigned to one specific fluid thickness. Since the toluene phase more or less flows as a closed film, further investigations are concentrated to the more interesting water rivulet thickness.

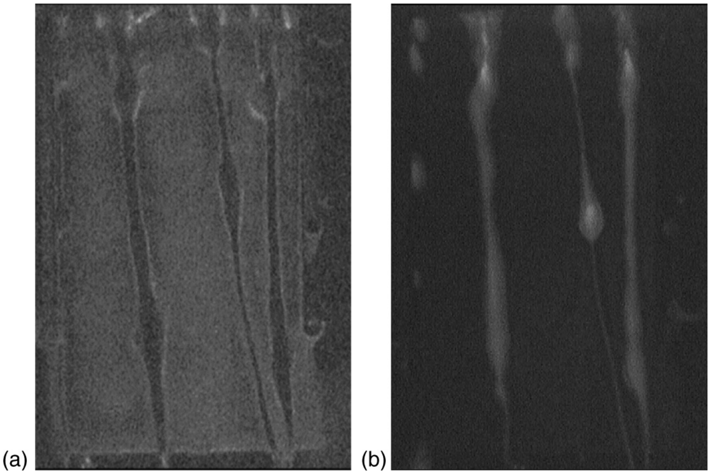

Figure 4. Phase separation and thickness measurement with 2-phase LIF. a) bright toluene phase with dark water rivulets, b) bright water rivulets with dark toluene film

In our measurements it is found that the water rivulet thickness is almost constant with an increase of Re_W, but it is decreased with an increase of Re_T. However, the averaged width of the rivulets increases with higher toluene Re-numbers (Re_T). A comparison of these results to the multiphase CFD simulations and to relations for single-phase water rivulets of other authors is shown in Figure 5.

On the left hand side of Figure 5, the rivulet thicknesses at a low level of Re_T are shown and at a higher level of Re_T on the right hand side, respectively. Measured and simulated thicknesses are taken from a number of time steps at different positions for each set of Re-numbers. Therefore, in Figure 5 the averaged thicknesses and the according standard deviations are plotted. It can be seen that the standard deviation of the measured water rivulet thickness decreases at higher water Re-numbers as well as at higher toluene Re-numbers. Compared to the measurements, the rivulet thicknesses from the simulations show identical trends regarding to the decrease with higher toluene loads and the decreasing standard deviations. All of the averaged thicknesses are found below the measured mean values, but with a constant distance, which is an indicator for a still unknown systematic deviation.

On the other hand, the comparison to thicknesses of other authors shows strong discrepancies. For comparisons with the multiphase rivulet thickness, common relations for the thickness computation of a single liquid phase flow are applied. At first, the well known Nusselt-relation for the film thickness [11] was adapted to the rivulet flow, referred to the measured rivulet widths. As can be seen in Figure 5, the computed thicknesses are underestimated, but show a slight dependence of the Re-numbers. Compared to that, Doniec [12] has developed an equation for the prediction of the critical rivulet thickness. This equation is only a function of fluid properties and doesn't consider the flow rates. For the surface tension the interfacial tension of water-toluene with 0.036 N/m is set and the

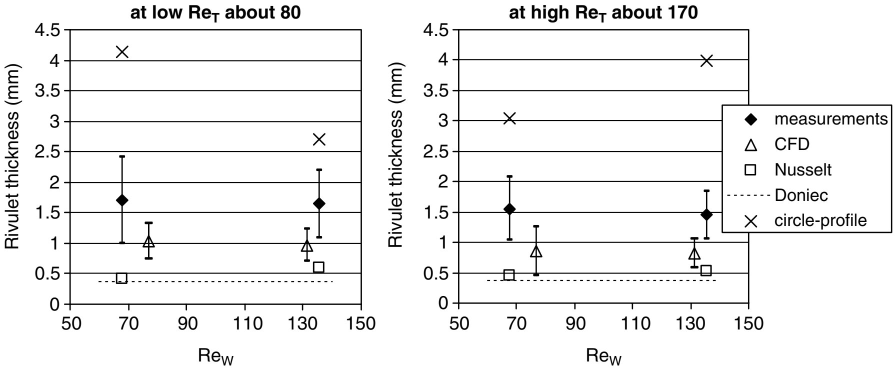

Figure 5. Measured and simulated rivulet thicknesses at different levels of Re in comparison to other relations

contact angle for the water phase is chosen at the water-toluene-steel interface of 124°. It can be seen that the values are close to the Nusselt thickness and are also underestimated. However, if it is assumed that the rivulet profile agrees with a segment of a circle as can be found e.g. in [13], then it leads to an overestimation of the thicknesses and, additionally, shows a very strong dependency of the rivulet width.

The investigation of the film thickness shows that thickness relations of a single liquid flow are not applicable for the case of multiphase film flow. As a reason for that the strong interactions between both phases can be seen again.

The quality of the interaction between both liquids inside of a structured packing has to be investigated next. Due to the strong effort of measurements inside the packing, the previously validated CFD-simulations are applied to more complex geometries. As a first case the macroscopic geometry of a Sulzer Mellapak Y-series is chosen and implemented into CFX. Figure 6 shows the model geometry of the simulated packing segment: two corrugated sheets of stainless steel are arranged in a way that the undulated profiles are crossing and due to that channels for liquid and vapour flow are formed. The liquid inlet on the top is implemented as a closed film with given velocity. For two-liquid phases this film inlet is divided into separate regions with one phase each. On the left and right hand side of the packing, periodical boundaries are applied.

In Figure 7 two qualitative results of the Mellapak simulations are given. The flow behaviour of the water phase on the back sheet (compared to Figure 6) – actually the water-wetted wall area – is shown for the case of single-liquid water flow (Figure 7a)

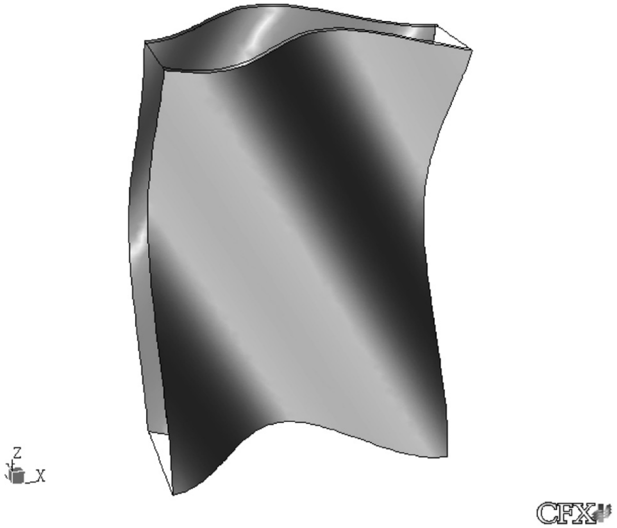

Figure 6. Mellapak Y-series segment

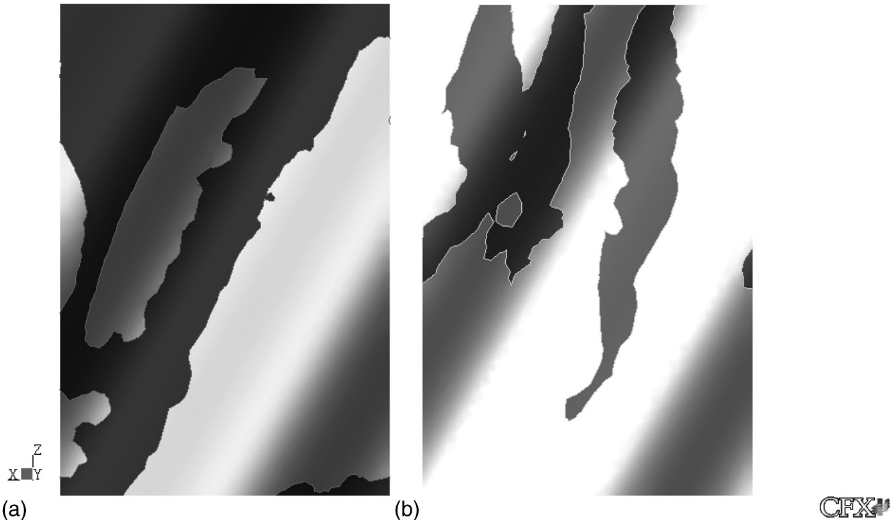

Figure 7. Water-wetted area (dark-grey) on the back sheet. a) two-phase flow (water-air), b) three-phase flow (toluene-water-air)

and for the two-liquid phase flow (Figure 7b). The water-wetted area on the plate gives information about the path of the water flow, which is different between the two-phase and the three-phase flow. In the two-phase flow (Figure 7a), the closed water film at the inlet breaks-up soon and water rivulets are formed. These rivulets mainly follow the macroscopic structure of the sheet and flow along the inclined channel to the left border of the sheet. Transferred to a whole structured packing, that means the liquid phase moves with high probability to the outer layers of the packing. This effect is also known as mal-distribution.

With the appearance of the toluene phase, the water rivulets are forced to the sheet and flow straight downwards to the bottom, as can be seen in Figure 7b, where only the water phase on the sheet is displayed. Even after the formation of capillary liquid hold-up at the crossing point of two corrugations (such a crossing point is located on the top left of Figure 7b), water forms rivulets again and flows down to the bottom of the segment. This flow behaviour due to the interaction of the two liquids reduces mal-distribution of the packing. These effects can be considered to explain the quite good performance of three-phase operated packed towers [3].

CONCLUSION

On the track to understanding three phases (two liquid phase and one vapour phase) in one packed tower a detailed knowledge about the complex fluid dynamics on the packing surface is required. Comprehensive flow studies of the two immiscible liquids toluene and water on inclined steel sheets are carried out numerically as well as experimentally. For simulations a commercial CFD-tool is used, for measurements optical methods are developed and applied. While the investigations of the surface velocities, fluid thicknesses, and liquid morphologies, it was found that strong interactions between both liquid phases exist. Due to these interactions the liquid flow is decelerated and stabilized. This stabilisation effect leads to a better performance of three-phase operated packed towers, which was shown in CFD-simulations as well.

Further efforts will aim at more detailed investigations of the interaction between both liquids. These will be carried out with μPIV-measurements at the liquid-liquid interface. Additionally, the influence of the counter-current vapour flow on the liquid flow behaviour has to be studied experimentally as well as numerically.

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REFERENCES

1. Hoffmann, A., Ausner, I., Repke, J.-U., Wozny, G.; Comp. Chem. Eng. 29 (6), 2005, pp. 1433–1437
2. Repke, J.-U., Wozny, G.; Chem. Eng. Technol. 25 (5), 2002, pp. 513–519
3. Villain, O., Repke, J.-U., Wozny, G.: Evaluation of the separation efficiency of three-phase operated packed towers; in: AIChE Spring Meeting, 10–14 April 2005, Atlanta, GA, USA
4. Adrian, R.J.; Annu. Rev. Fluid Mech. 23, 1991, pp. 261–304
5. Adomeit, P., Renz, U.; Int. J. Multiphase Flows 26, 2000, pp. 1183–1208
6. Ausner, I., Hoffmann, A., Repke, J.-U., Wozny, G.: Multiphase film flow investigations on inclined plates; in: 3rd International Berlin Workshop (IBW3) on Transport Phenomena with Moving Boundaries, 6–7 October 2005, Berlin, Germany
7. Hirt, C.W., Nichols, B.D.; J. Comp. Phys. 39, 1981, pp. 201–225
8. Brackbill, J.U., Kothe, D.B., Zemach, C.; J. Comp. Phys. 100, 1992, pp. 335–354
9. Ausner, I., Hoffmann, A., Repke, J.-U., Auracher, H., Wozny, G.: Two liquid phase film flow over inclined plates; in: 7th World Congress of Chemical Engineering (WCCE7), 10–14 July 2005, Glasgow, Scotland/UK
10. Ausner, I., Hoffmann, A., Repke, J.-U., Wozny, G.; Chem. Ing. Tech. 77 (6), 2005, pp. 13–19
11. Nusselt, W.; VDI-Z. 60 (27), 1916, pp. 541–546
12. Doniec, A.; Can. J. Chem. Eng. 69, 1991, pp. 198–202
13. Ataki, A., Barth, H.-J.; Chem. Eng. Tech. 27 (10), 2004, pp. 1109–1114