

Loading & Flooding in Mellapak Structured Packing: Radiographic & Computer-Tomographic Insights

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Abstract. The combination of radiography and computer tomography was used in this work to overcome the temporal resolution limitations of X-ray computed tomography. Its unique spatial (80 μm) and temporal (100 fps) resolution combined with an image processing routine enable the determination and quantification of highly dynamic flow phenomena in structured packings. Investigations in a Mellapak 500.Y packing show that at low F-factors, dynamic effects are of little significance. Therefore, computed tomography remains valid over a wide operating range, but droplets can be added as another flow phenomenon to complete the picture. In the loading and flooding region, however, a significant change in flow morphology is observed, with highly dynamic effects gaining importance. These cause a uniform distribution of the fluid over the cross-section, but significant differences are observed over the packing height. The improved understanding is not only of academic interest but especially for the further development of structured packings.

Keywords: Radiography, Mellapak, Tomography, Flooding, Droplets

1. Introduction

In the last decades, X-ray computed tomography has shown to be a suitable non-invasive measuring technique, which allows the determination of local parameters in columns under process conditions [1-7]. However, especially for highly dynamic and small-scale flow phenomena, such as droplets, this measurement technique reaches its resolution limits. Object size, spatial and temporal resolution are dependent parameters. Therefore, a compromise must be made in this respect. A detailed description of these relationships can be found in literature [8, 9]. However, to further increase the temporal resolution, various new technical approaches are being pursued. In this place, the ultrafast electron beam X-ray computed tomography should be mentioned, which allows a very high temporal resolution, but also reaches its limits in terms of spatial resolution [6, 10, 11].

In this work, the same measurement technique but different measurement modes have been used. Fig. 1 illustrates the basic procedure and the four measurement modes which have been applied. The left-hand side shows the computed tomography mode, where the X-ray source and the detector rotate around the column. In contrast, in the radiographic mode on the right, the radiation passes through the column from only one angular position. In addition, it is possible for both operating modes to measure for a longer period at one height position (top row: cross-sectional images or stationary measurement) or to move the measuring unit vertically (bottom row: helix or transient measurement). The advantage is that all measurement modes can be carried out subsequently in the same set-up under constant conditions. This means that all modes can be combined, and the advantages of the individual methods can be integrated into the overall result. On the one hand, it is possible to record highly dynamic effects radiographically at 100 fps and, on the other hand, to utilize the high spatial resolution over the cross-section of the computed tomography. This provides the maximum gain in knowledge and enables characterization of all flow phenomena that occur over the entire operating range. Using the combination of the previously mentioned techniques, systematic measurements in MellapakTM 500.Y structured packings were conducted. One major focus was put on the behavior of the structure packing at increasing gas load around the loading point up to flooding. In section 2, the measuring technique used for this purpose, the experimental set-up, and the measurement procedure are explained. The results are presented and discussed in section 3 and afterwards summarized in section 4.

2. Material and Methods

2.1. X-ray Scanner & Experimental Set-up

As described above, the same measurement set-up was used for both the computed-tomographic and radiographic measurements. The experiments were performed using the computer tomograph available at the Institute of Separation Science and technology in Erlangen, Germany. It was developed and manufactured by the Fraunhofer Development Center for X-ray Technology for the determination of fluid dynamics in columns and reactors. The line detector used, type SEZ T3/W1/W2/W3 480 from NTB elektronische Geräte GmbH, has a highest resolution of 12 pixels per

millimeter. Pictures are recorded at 500 fps by averaging five images, the resulting transient resolution of the radiographic images is 100 fps.

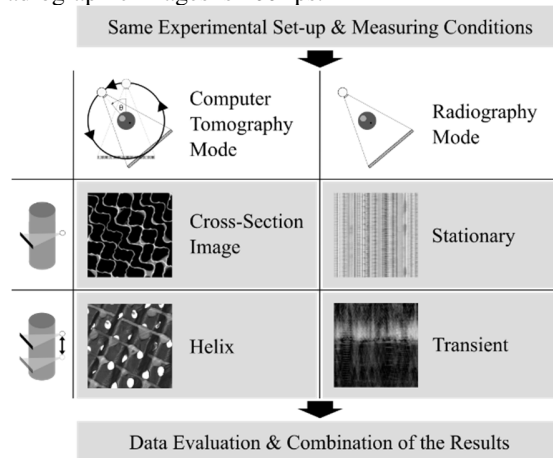


Fig. 1 Schematic illustration of the measurement modes used and the procedure from measurements to results.

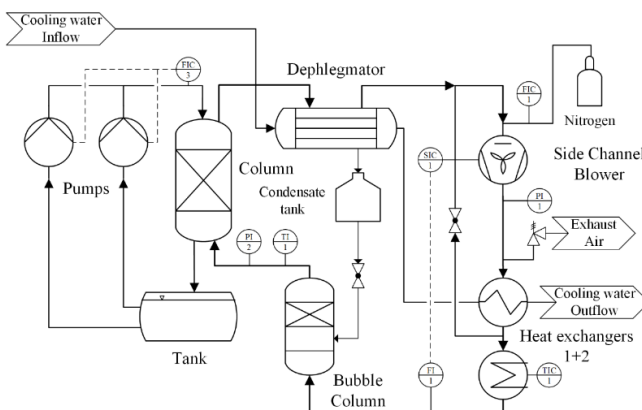


Fig. 2 Experimental set-up used with all relevant components in the liquid and gas cycles.

The column is operated stationary at all measurements. During the computed tomography scans, the X-ray source and the detector rotate around the column, as depicted in Fig. 1. The rotational speed is set to 0.33 rps, resulting in a measurement time of 3 seconds for one cross-sectional image. As described previously, the setup allows direct switching between radiographic and computed-tomographic modes. This ensures the maximum transferability of the results. A detailed description of the measuring system, as well as results in the field of distillation, extraction, and reaction technology, can be found in literature [3-6, 9, 12, 13]. The geometrical and reconstruction parameters for computed tomography correspond to the conditions as they have already been described for the 100 mm column in previous publications [3, 5]. A detailed description of the settings for the radiographic measurements can be found in Eder et al. [9]. The experimental set-up is designed to realize all desired operating points up to flooding with the water-air system. Within this paper, only this for fluid dynamic measurements common reference system is used, but similar computer tomographic and radiographic measurements with the same set-up and organic systems have been published already [3, 6, 9, 12]. Liquid and gas phase flow countercurrently to each other and the liquid circulation is shown on the left side in Fig. 2. The liquid is pumped from the storage tank into the head of the column via gear pumps of the type Ismatec MCP Z Standard from Cole Parmer GmbH. The flow of the liquid phases is controlled by Coriolis mass flow meters of the type M55 RGD 44 O S from Wagner Mess- und Regeltechnik GmbH in combination with LabVIEW software from National Instruments AG. The liquid is fed via a splash plate distributor from Sulzer Chemtech AG. It has a distribution density equivalent to about 400 dripping points per square meter. The gas counter flow is shown in Fig. 2 on the right. The experimental setup allows any variation of the F-factors up to the flooding point. The core of the gas counter-flow system is the side channel compressor SKV CL 14/21 G 2D from Mapro GmbH. This design enables oil- and pulsation-free flow. The gas volume flow is measured by a flow meter type F 106AI RGD 03 V from Wagner Mess- und Regeltechnik GmbH. The task of the other devices is preventing water entry into the compressor or ensuring that the gas entering the column is completely saturated with water at 25 °C to prevent evaporation in the column. To ensure a uniform inlet gas distribution, the distance between the gas inlet and the lower edge of the packing is four column diameters. Three modifications of Sulzer structured packings Mellapak™ 500.Y were investigated. First the standard type with w-grooving and holes. The second packing has got a smooth surface and holes and the third packing a w-grooving but no holes. Four elements were installed, each rotated by 90 degrees and fixed at the top by the distributor and the bottom by a support grid.

2.2. Preliminary Tests & Experimental Procedure

Before the actual experiments were conducted, preliminary tests were carried out to obtain the most meaningful results. At first, the computed-tomographic and radiographic methods and the respective evaluation routines were validated. A detailed description of the procedure and the results are described in literature [3, 9]. Through further preliminary tests it was found that under the present conditions, the liquid needs approx. 30 minutes after start-up or a change of the liquid load to form a largely stationary flow. This does not include the periodic fluctuations typical for water, which occur, for example, due to the periodic rupture and reformation of bridges. By measuring at several height positions, these local fluctuations can be averaged out. These local measurement deviations are in the same order of magnitude as the deviations caused by the repacking of a column. The hold-up measurements are therefore reproducible with a

2 % measurement deviation. After a change in the gas load, approx. 15 minutes are required for the formation of a sufficiently stationary flow. The measuring positions were distributed over the second and third element, since preliminary tests revealed that the liquid does not yet have the characteristic distribution in the first element. The fourth element was not measured to ensure a typical gas distribution and minimize inlet effects.

Taking these preliminary examinations into account, a typical measurement proceeds as follows. At first, dry images are taken for the radiographic evaluation. Then the liquid load is set to $20 \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1}$, the only liquid load used in this work. After reaching a largely steady state, the radiographic and computed-tomographic measurements are carried out one after the other. Both stationary measurements at 3 mm intervals from each other and transient measurements over the second and third element are performed. When setting a new operating point, care must be taken to increase the gas loads only, to exclude hysteresis effects. Otherwise, the increased static hold-up from the previous measurement will falsify the measurement result for the subsequent measurement at a lower load. For this reason, the column is completely dried when a measurement at a lower load is carried out afterwards.

2.3. Data Processing

The large amount of data obtained by the measurement requires efficient and automated data processing with the aim of providing meaningful and reproducible information. In this section, only the basic procedure and previously unpublished passages are described. Most of the evaluation routines used have already been presented in previous publications and are therefore only mentioned here [3, 9]. These papers also describe in detail the validation methods that are necessary to avoid falsification in the evaluation. In the computer tomographic mode, image processing is necessary because every tomographic measurement is accompanied by image artifacts and therefore no direct assignment of the individual phases is possible [8, 14]. The individual projections are first reconstructed into cross-sectional images and then a complete and overlap-free segmentation is performed. Based on the resulting segmented cross-sectional images, the image evaluation takes place, in which the various fluid dynamic parameters are determined. An overview of these parameters provides Hampel et al. [6]. The image processing program used for this is the ImageJ extension Fiji and the software Matlab for the subsequent segmentation and image evaluation. [3, 8] An implementation in Python would also be possible, historically Matlab was used. The helix measurement is not suitable for quantitative comparison, because the missing possibility of time-averaging leads to an insufficient signal-to-noise ratio, and consequently the segmentation is error-prone. Since it is not used for qualitative analysis in this paper, as it has been in the past, it will not be discussed in more detail [5].

In the radiographic mode, the signal is corrected first with the help of light and dark images. After this so-called flat-field correction, the images are averaged with factor five to reduce the data amount and to increase the signal-to-noise ratio, resulting in a temporal resolution of 100 fps. The next main step is calculating the quotient between the wet and dry measurements to calculate the portion of liquid within the individual X-rays. Using the geometrical data of the experimental set-up and a forward projection, a so-called ray length array is calculated which holds the length each X-ray travels inside the column. Using both arrays, the hold-up first for each array and later for the whole cross-section is calculated. The data evaluation routine is implemented in Python. [9, 15] The big advantage of determining the hold-up from the radiographic measurement is, besides the higher temporal resolution of 100 fps, the direct calculation via the attenuation coefficients without reconstruction and segmentation errors. However, the horizontal spatial information is lost. When determining the dynamics, a distinction is made between dynamic and highly dynamic effects. Dynamic effects are obtained from computed-tomographic data. Since a data set is averaged over 3 seconds and the dynamics are derived from the deviation of ten images, only effects that change within seconds are detected here. The data for the highly dynamic effects, on the other hand, are generated from the radiographic measurement and the deviations are evaluated to the hundredth of a second. A breaking liquid bridge thus flows into both dynamics, while droplets are only assigned to the highly dynamic effects.

3. Results and Discussion

Fig. 3 shows the dynamics over increasing F-factor as obtained from computed tomography images over one entire cross-section. Pixels in which the phase changes within 30 seconds are shown brighter, pixels in which the phase does not change remain black. At low gas loads below the loading point, only isolated dynamic areas are visible. These are mainly liquid bridges that break and form again or rivulets that flow through unevenly. Examples are marked in yellow in Fig. 3. Even after the column has been restarted and has been running under different loads, these dynamic areas usually reappear at the same positions. Operating point a) at an F-factor of $1.7 \text{ Pa}^{0.5}$, shows a typical behavior before loading. As depicted in the cross-sectional image a), these dynamic areas are unevenly distributed and in this example are located mainly in the lower part of the image. With an increase to $1.8 \text{ Pa}^{0.5}$, not only the hold-up increases slightly, but also the dynamics, as shown in operating point b) in Fig. 3. This shows the typical behavior when loading begins. The liquid flows through the column in a pulsating manner, because the gas flow repeatedly accumulates it for a short time. With a further increase in the gas counter-flow, the liquid accumulates more evenly over the cross-section. The

individual rivulets merge more into a uniform film over the entire packing. At this point c), the hold-up and the pressure drop increase significantly and the packing shows almost complete wetting. A further increase of the F-factor leads to flooding of the column and the dynamics are maximum at point d). Whereas the temporal changes before flooding take place over several seconds and the phase changes can still be detected well by computed tomography, it reaches its limits in the area of loading and flooding. Highly dynamic effects can no longer be detected by computed tomography. Therefore, the focus in the following is on radiographic measurements. Nevertheless, the deviations between the computed-tomographic measurement and the radiographic measurement below loading are small. However, this only is true for a comparison between cross-sectional images and stationary measurements taken at the same height. For structured packings, the underestimation of computed tomography over the entire packing is less than 2 %. As shown in Fig. 3 and discussed in detail in the following, this can be accounted to the low dynamics below loading.

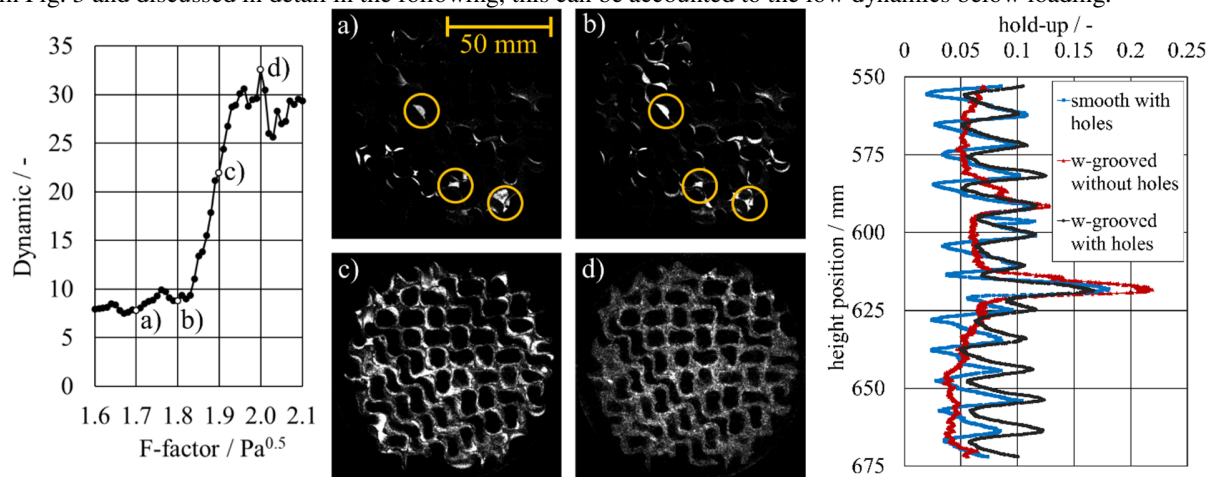


Fig. 3 Dynamics (phase change) at different F-factors; Computer tomographic measurement at one height position in the third element (Mellapak 500.Y (w-grooved with holes); liquid load = $20 \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1}$); Cross-sectional images show the dynamics; dynamic areas (phase change) are shown brighter than non-dynamic areas (no phase change).

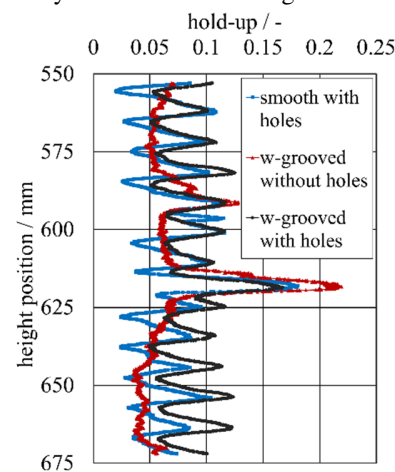


Fig. 4 Hold-up over height for 3 modifications of Mellapak 500.Y (liquid load = $20 \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1}$; F-factor = $2.0 \text{ Pa}^{0.5}$); Transient radiographic measurement over 2nd and 3rd element.

As described before, the helix measurement is not suitable for a quantitative comparison. Therefore, the following analysis over the height is done with transient radiographic measurements. Fig. 4 shows the hold-up over the lower half of the second packing element and the upper half of the third packing element for each of the three packing modifications examined. In the middle, the transition zone with the characteristically increased hold-up is clearly visible [2, 3]. The maximum value varies within the same packing modification and is therefore not representative of the respective modification. The reason for this is the different offset between the sheets and the manufacturing tolerances. However, the course in the rest of the packing is characteristic. In Fig. 5, the changes with increasing F-factors during loading and flooding will be discussed in more detail. In Fig. 4, the F-factor is $2 \text{ Pa}^{0.5}$, so the total hold-up is already increased by the slight loading, but it differs only slightly in the course from that with lower F-factors. While the unperforated packing shows hardly any fluctuations over height, the perforated packings show the fluctuations already known from computed tomography [3, 5]. Due to the transient recording, a very high resolution over the y-axis is possible. The high temporal resolution of 100 fps and the direct evaluation via the Lambert-Beers equation lead to more exact hold-up results compared to earlier computed-tomographic evaluations [3, 5, 9]. A comparison between the unperforated and the perforated w-grooved modification shows that the holes increase the hold-up. The same applies for the w-grooved versus the smooth modification, whereby the w-grooving prevents the strong drop-off of the hold-up below the holes, as can be seen in Fig. 4. The reason for this is a better small-scale liquid distribution, as has already been discussed in the literature [3, 5].

The focus in the following is on the gas counter-flow. Below the loading point, the influence of gas flow on liquid distribution is minor and therefore Fig. 5 shows only F-factors over $1.9 \text{ Pa}^{0.5}$. The reason for the loading and flooding at low F-factors is the small column diameter of 100 mm. The wall wipers block a disproportionately large amount of cross-sectional area for the gas, so the real F-factor in the packing is higher. Furthermore, due to the small column diameter, wall effects which reduce the capacity are more significant. The absolute values are therefore not directly transferable to larger column diameters [16]. Graph b) in Fig. 5 shows the hold-up averaged over the second and third packing element from the transient radiographic measurement for different gas loads. While the hold-up is not yet affected at low gas loads, it increases significantly above an F-factor of $2.1 \text{ Pa}^{0.5}$. Fig. 5 a) shows that this increase is not uniform over the entire packing height.

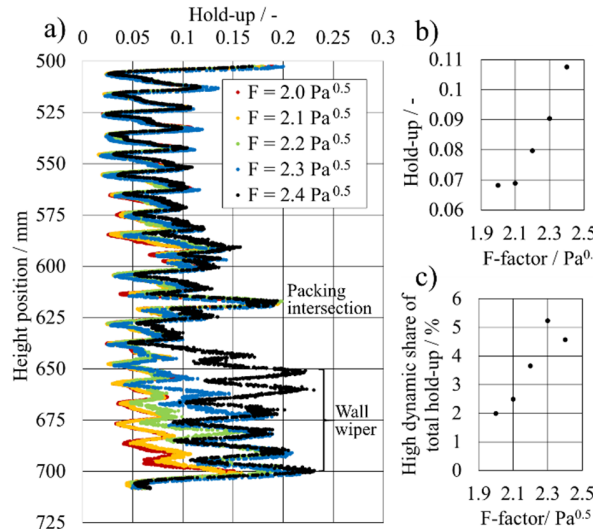


Fig. 5 Radiographic measurement at different F-factors (Mellapak 500.Y; liquid load = $20 \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1}$);
a) Hold-up over height (transient measurement over 2nd and 3rd element);
b) Mean hold-up (transient measurement over 2nd and 3rd element);
c) High dynamic shares (e.g. droplets) of total hold-up (stationary measurement over 2nd and 3rd element).

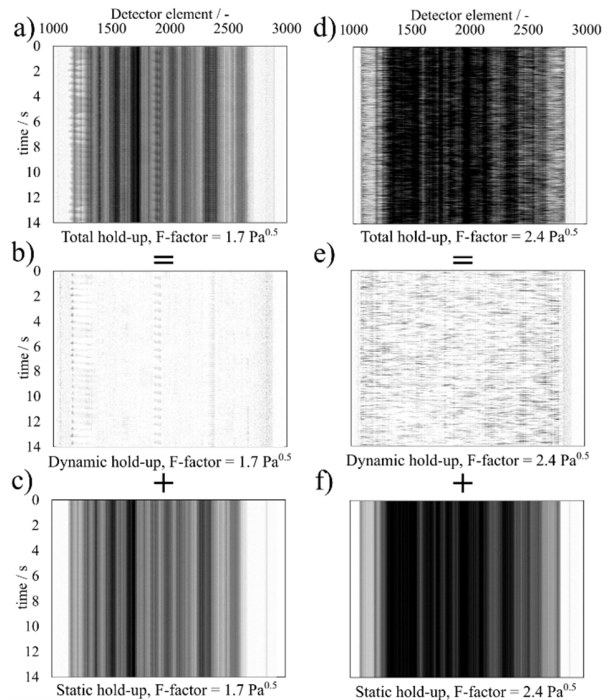


Fig. 6 Stationary radiographic images (Mellapak 500.Y (w-grooved with holes); liquid load of $20 \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1}$).

As before in Fig. 4, Fig. 5 a) shows the lower part of the second and the upper part of the third packing element. It is noticeable that in the second packing element (500 mm to 620 mm) the hold-up changes only slightly. The increase in the hold-up takes place much more in the area of the wall wiper in the third element (650 mm to 700 mm). As can be seen from the hold-up curves for the different F-factors, the hold-up starts first to increase in the lower area of the wall wiper (green) and then continues upwards with rising gas load (blue) until the hold-up is strongly increased in the entire area of the wall wiper (black). With a further increase in the F-factor, this behavior continues upwards. The initial point for loading behavior is often the transition zone between two packings. This is especially true for columns with larger diameters, where the wall wipers are of less importance, but could also be observed in these measurements. The reason is the reduced free cross-section for the gas due to the increased hold-up. Furthermore, with the help of the stationary recordings, it could be shown that the loading in the packing is already noticed beforehand by increased dynamics in the column. This applies both to the dynamic as well as the highly dynamic effects and is caused by the damming up of bridges and rivulets, as discussed earlier. In Fig. 5 c) this effect can already be seen at an F-factor of $2.1 \text{ Pa}^{0.5}$ and at rising F-factor the proportion of highly dynamic effects increases significantly, as this was already the case for the dynamic effects. As shown in Fig. 5, the maximum value for the dynamic share averaged over all height positions is 5.2 %. Here the three packing variations differ only slightly. Locally, this proportion can increase up to 8 % in the packing. At very high F-factors, at which the total hold-up also increases significantly, the highly dynamic share drops again.

The flow phenomena causing these highly dynamic effects can be seen on radiographic images, whereby computer tomographic images are helpful for interpretation. Fig. 6 shows stationary images at a height position just below the packing intersection between the second and the third element. On the left, the images were taken at an F-factor of $1.7 \text{ Pa}^{0.5}$, and on the right the F-factor was $2.4 \text{ Pa}^{0.5}$. At the top, figures a) and d) show the total hold-up, which in turn can be divided into the dynamic part b) and e) and the static part c) and f). In the static areas is no phase change visible within 14 seconds. This time was chosen as an example and has no influence on the results. For the liquid phase these are mainly rivulets, bridges, and films [3, 5]. The highly dynamic hold-up in Fig. 6 b) is mainly liquid dripping from the packing bottom edge, which is only small compared to the static hold-up c). In dynamic hold-up, there is a high degree of regularity of the individual phenomena in relation to location and time. Increasing the gas load to $2.4 \text{ Pa}^{0.5}$ rises not only the static hold-up f) but also the highly dynamic hold-up e). As already shown at the beginning by computed-tomographic measurements for the dynamic share, it can also be seen for the highly dynamic share that the dynamic effects are distributed over the entire cross-section. As already mentioned at the beginning of this chapter, the individual flow phenomena merge into each other at high gas loads and due to the strong superposition on the radiographic images, the individual flow phenomena can no longer be assigned clearly.

4. Summary and Conclusions

To overcome the temporal resolution limitations of X-ray computed tomography, radiography was used as a complementary method. The results show that computed tomography underestimates the hold-up values by less than 2 % over a wide operating range. Thus, many computed-tomographic results from the past can be confirmed and extended by results of radiography measurements. The influence of the w-grooving and the perforation can be determined with a 300 times higher temporal resolution and droplets can now also be detected. The higher temporal resolution of the radiographic measurement is especially important at drip points as well as with high gas loads, where the error using computed tomography is increasing due to the higher dynamics. The combination of both measurement techniques makes it possible to assign and quantify all flow phenomena over a wide operating range until the loading point. Rivulets, bridges, and films are the dominant flow phenomena here, showing hardly any change over time. Droplets play only a minor role and occur almost only at packing lower edges. Shortly before loading begins, the dynamics increases. With rising gas flow, rivulets, and bridges pulsate stronger and these dynamic areas are increasingly evenly distributed over the entire cross-section. Towards flooding, the highly dynamic effects dominate, and the gas and liquid phases alternate several times per second over the entire cross-section. At these high gas loads, the flow phenomena are no longer separated from each other, but rather form a permanently changing film over the entire packing. The transient measurements show that the packings do not start to flood uniformly over the entire height, but that the accumulation begins at height positions with maximum interaction between gas and liquid phase and then spreads over the height. With the developed evaluation routine, the stationary measurements enable a distinction between highly dynamic and stationary flow phenomena. Integrated over the column, these highly dynamic flow phenomena can be up to 5.2 % of the total hold-up, locally their value can rise as high as 8 %. Furthermore, the droplet amount of the total hold-up can be quantified. At high gas loads, however, this method reaches its limits due to the limited resolution and the strong superposition. Therefore, an assessment of droplet count during entrainment is not possible with this set-up. In the future, further developments in tomography and digital image processing will further improve the spatial and temporal resolution. This will contribute to this measurement technology becoming established in process engineering.

References

- [1] A. Janzen, J. Steube, S. Aferka, E. Y. Kenig, M. Crine, P. Marchot, *et al.*, "Investigation of liquid flow morphology inside a structured packing using X-ray tomography," *Chemical Engineering Science*, vol. 102, pp. 451-460, 2013.
- [2] S. Aferka, A. Viva, E. Brunazzi, P. Marchot, M. Crine, and D. Toye, "Tomographic measurement of liquid hold up and effective interfacial area distributions in a column packed with high performance structured packings," *Chemical Engineering Science*, vol. 66, pp. 3413-3422, 7/15/ 2011.
- [3] S. Schug and W. Arlt, "Imaging of Fluid Dynamics in a Structured Packing Using X-ray Computed Tomography," *Chemical Engineering & Technology*, vol. 39, pp. 1561-1569, 2016.
- [4] T. Linder, P. Leicht, M. Sommerschuh, A. Bösmann, P. Preuster, K. Müller, *et al.*, "Fluiddynamische Untersuchungen mittels Computertomographie " *Jahrestreffen der ProcessNet-Fachgruppen Fluidverfahrenstechnik, Adsorption und Extraktion*, 2020.
- [5] M. Wehrli, T. Koegl, T. Linder, and W. Arlt, "An Unobstructed View of Liquid Flow in Structured Packing," *Chemical Engineering Transactions*, vol. 69, pp. 775-780, 2018.
- [6] U. Hampel, M. Schubert, A. Döb, J. Sohr, V. Vishwakarma, J.-U. Repke, *et al.*, "Recent Advances in Experimental Techniques for Flow and Mass Transfer Analyses in Thermal Separation Systems," *Chemie Ingenieur Technik*, vol. 92, pp. 926-948, 2020.
- [7] L. Bolenz, F. Fischer, D. Toye, and E. Y. Kenig, "Tomographische Untersuchung der Fluiddynamik viskoser Systeme in Packungskolonnen," *Chemie Ingenieur Technik*, vol. 91, pp. 1892-1896, 2019.
- [8] T. Buzug, *Computed tomography. From photon statistics to modern cone-beam CT* vol. 39. Germany: Springer, 2008.
- [9] S. Eder, T. Linder, M. Wehrli, and M. Thommes, "High Resolution X-ray Radiographic Measurements in Structured Packings," *Chemical Engineering Research and Design (submitted)*, 2022.
- [10] J. Sohr, M. Schubert, S. Flechsig, E. Y. Kenig, and U. Hampel, "Experimental Investigation of the Froth Height in Columns with Sandwich Packings," *Chemie Ingenieur Technik*, vol. 91, pp. 139-144, 2019.
- [11] F. Fischer and U. Hampel, "Ultra fast electron beam X-ray computed tomography for two-phase flow measurement," *Nuclear Engineering and Design - NUCL ENG DES*, vol. 240, pp. 2254-2259, 09/01 2010.
- [12] S. Schug, "Untersuchung der Fluiddynamik in Packungskolonnen mittels Computertomographie," *Lehrstuhl für Thermische Verfahrenstechnik, Friedrich-Alexander-Universität Erlangen-Nürnberg*, Erlangen, 2018.
- [13] T. Kögl, T. Linder, and W. Arlt, "Spatially Resolved Tomographic Assessment of Axial Mixing in Modern Structured and Random Packings," *(submitted)*, 2022.
- [14] R. A. Brooks and G. D. Chiro, "Beam hardening in X-ray reconstructive tomography," *Physics in Medicine and Biology*, vol. 21, p. 390, 1976.
- [15] J. Seibert, J. Boone, and K. Lindfors, *Flat-field correction technique for digital detectors* vol. 3336: SPIE, 1998.
- [16] Ž. Olujić, "Effect of Column Diameter on Pressure Drop of a Corrugated Sheet Structured Packing," *Chemical Engineering Research and Design*, vol. 77, pp. 505-510, 1999/09/01/ 1999.