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MEASUREMENT OF LIQUID HOLD-UP IN CATALYTIC STRUCTURED PACKINGS: COMPARISON OF DIFFERENT EXPERIMENTAL TECHNIQUES

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Abstract
This paper presents the results of an experimental study carried out to examine liquid hold-up in 100 mm diameter catalytic structured packing Katapak-SP. Two versions of the packing have been investigated. Both global and local measurements were carried out on the very same packings, using water at ambient conditions as working liquid. To this scope, traditional methods like draining techniques were used to obtain global hold-up values. A non-intrusive high energy X-ray tomography was used for gathering information on local scale and study axial distribution of the liquid hold-up over the packed column, inside and outside the catalyst containing baskets. The total liquid hold-up of the catalytic packed bed was evaluated by averaging hold-up values obtained at different cross sections over the corresponding volume. Very good agreement was found between global values estimated with the different techniques. The experimental results are useful for supporting theoretical developments in hydrodynamic and mass transfer modeling.

Keywords: liquid hold-up, catalytic structured packing, X-ray tomography, Katapak

1. Introduction
Catalytic structured packings are used in an increasing number of reactive separation processes. The new generation packing Katapak-SP, developed by Sulzer Chemtech, is characterized by a hybrid structure made of an alternation of reactive and separation zones[1]. Figure 1 shows schematically the structure of Katapak-SP11 and Katapak-SP12, the two configurations of packing examined in the present work. The two versions differ in the ratio of catalytic baskets to corrugated sheets, which is of 1:1 and 1:2, respectively. When placed in a packed bed, the packing elements are rotated of 90° each others as schematically shown in Figure 1b.

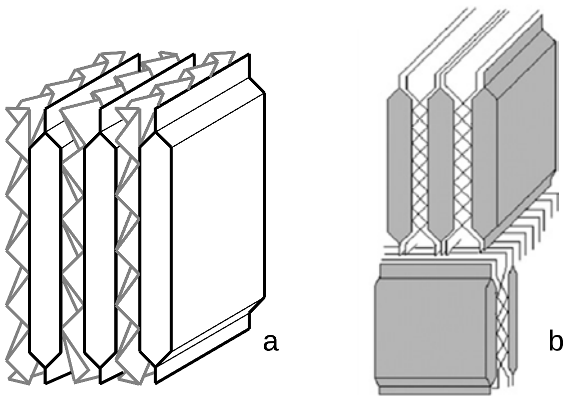

Figure 1. Schematic of the structure of Katapak-SP11 (a) and Katapak-SP12 (b) packings

The hybrid structure of the catalytic structured packing determines the flows development inside the packed bed and consequently the overall column performances. The external wire gauze of catalytic baskets allows the liquid flow penetration and prevents the gas cross-over, thus limiting the use of

Katapak-SP to those applications where the reaction takes place in the liquid phase. While the liquid hold-up inside the catalytic baskets influences the reactive performance of the packing, the liquid hold-up on the corrugated sheets is mainly responsible for the interactions with gas. Hence, knowledge of liquid hold-up is very important for the prediction of fluid dynamic related parameters, such as pressure drop, capacity, interfacial area and mass transfer volumetric coefficients. Moreover, the knowledge of the hold-up is needed for the development of accurate predictive models.

In catalytic structured packings both the static and dynamic liquid hold-up contribute significantly to the total liquid hold-up[1,2,3]. The static hold-up is the volume fraction of liquid that remains within the packed bed after complete draining and it results from the action of capillary forces that hold some liquid in the narrow sections of the packing. The dynamic hold-up is made up of the flowing liquid and strongly depends on the liquid load. Static and dynamic hold-ups are held on both corrugated sheets and catalytic baskets. Proven draining or volumetric methods and tracer based measurement techniques are commonly used to study liquid hold-up and liquid flow behaviour, thus obtaining hydrodynamic parameters referred to the bed scale, i.e. global values. Dynamic hold-up measurements have been reported for Katapak-SP12 of different nominal size, from pilot scale[1,4] of 100 mm and 250 mm to industrial scale[3] of 450 mm. As far as Katapak-SP11 is concerned, dynamic hold-up data have been measured on packings of diameter equal to 50 mm and 100 mm[5,6,7] and on the industrial packing size[3] of 450 mm. The static hold-up contribution can be mainly attributed to the catalytic baskets. The drainage of single catalytic baskets taken from dismantled packings have been investigated[6,8,9]. These studies are complementary to the measurements of the static hold-up on packing elements of Katapak-SP12[10] and Katapak-SP11[6].Properties of basket fillings (porosity, contact angle, swelling, etc.) and packing geometry strongly affect the liquid flow morphology and accordingly the liquid hold-up distribution. It is worth to note that catalytic baskets filled in packings of 250 mm and 450 mm are divided into two portions[1,3] while catalytic baskets filled in packings of 50 mm and 100 mm are made of one unique piece (as shown in Figure 1)[4,5,10].

The present work show results of liquid hold-up at both global and local scale obtained using different techniques. It is worth to emphasize that the very same packing elements and prewetting procedures were used. Two modular configurations of Katapak-SP have been investigated in the diameter size of 100 mm. The catalytic baskets were filled with glass spheres of 1 mm of diameter. Global data on dynamic and static liquid hold-up have been measured for Katapak-SP12 and compared to the values provided for Katapak-SP11 of the same dimension with similar methods. Moreover, local information on liquid hold-up inside Katapak-SP11 and Katapak-SP12 has been determined qualitatively and quantitatively by means of X-ray tomographic measurements. X-ray tomography[10,11] is a noninvasive technique capable of offering local information on the liquid hold-up distribution in the inhomogeneous packing structure[12]. The technique consists in the acquisition, reconstruction and processing of packing cross section images. Spatial variations of the liquid distribution inside the constitutive elements can be detected on irrigated packing. Quantitative information on liquid hold-up is obtainable from tomographic images when the appropriate image processing methodology is applied[10,11,12].

2. Measurements

Liquid hold-up data at both local and global scales have been obtained in a wide range of liquid loads. Water under ambient conditions was used as working liquid. Experiments were performed by varying the liquid load in the range of 3-26 $m^3/m^2/h$. As regards liquid distribution, a multiple point source distributor (approx. 4000 drip points/m^2) was used to feed the liquid at the top of the column. Table 1 summarizes geometrical properties of the investigated packings[13]. Measurements performed to collect global values using traditional techniques are grouped in the following text under the name global measurements, while tomographic experiments are called local measurements.

2.1. Global measurements

Global measurements were performed at the Department of Chemical Engineering of the University of Pisa (Italy) to determine the different liquid hold-up contributions. A standardised procedure has been set and all the results have been analysed by using a consistent measurement time, which has been fixed to 1 hour[7]. For the used configurations, results obtained at this standardised time agree with data at infinite draining time, determined by extending to catalytic packings the analysis originally proposed by Urrutia et al.[14] for trickle bed reactors. The experimental procedures and devices used for Katapak-SP11 are described elsewhere[5,6,7]. The same have been used here for Katapak-SP12.

Table 1. Geometrical properties of the tested packings

Geometrical Properties	Katapak-SP11	Katapak-SP12
packing diameter (mm)	100	100
packing height (mm)	200	200
specific surface (m^2/m^3)	210	282
packing porosity (-)	0.74	0.82
porosity of the spheres bed inside the catalytic baskets (-)	0.385	0.385
volume fraction of the catalytic baskets in the packing, ψ_{CB} (-)	0.42	0.30

In particular, the static hold-up contribution has been measured by weighing several times single packing elements while they drain after their complete immersion in water. It is defined as the volume of liquid remaining over the packing element divided by the packing volume. As already observed by Behrens et al.[8], the static hold-up is mainly due to the liquid which does not drain from catalytic baskets. Measurements on packing elements of Katapak-SP11 and Katapak-SP12 (diameter 100 mm) have provided repeatable static hold-up values, equal to 6.8 % and 5.0 %, respectively. The difference in the two values is easily attributable to the different volume fraction occupied by catalytic baskets in the two packing configurations (See ψ_{CB} in Table 1).

The dynamic liquid hold-up is the contribution which changes with operating conditions. The dependence of dynamic hold-up on the liquid load has been measured on a column filled with catalytic structured packings. It is defined as the volume of liquid which drains from the column divided by the packed bed volume. Figure 2 shows the dynamic hold-up as a function of liquid load for Katapak-SP11 and Katapak-SP12. It is worth to note that this hold-up contribution does not depend on the packing configuration. A similar behaviour was observed by Behrens[15] on 450 mm diameter packings, as shown by data reported for comparison in the same figure. For the same packing version, the size effect is on the contrary more pronounced. This is probably due to the different structure of catalytic baskets. As mentioned above, the baskets are made of unique pieces in the packing of 100 mm diameter size, whereas they are divided into two portions in the 450 mm diameter packings. The contribution to dynamic hold-up of catalytic baskets is predominant for large scale packings. Finally, from global measurements the total liquid hold-up (hl_{GLOBAL}) is estimated by summing static and dynamic hold-up contributions[2].

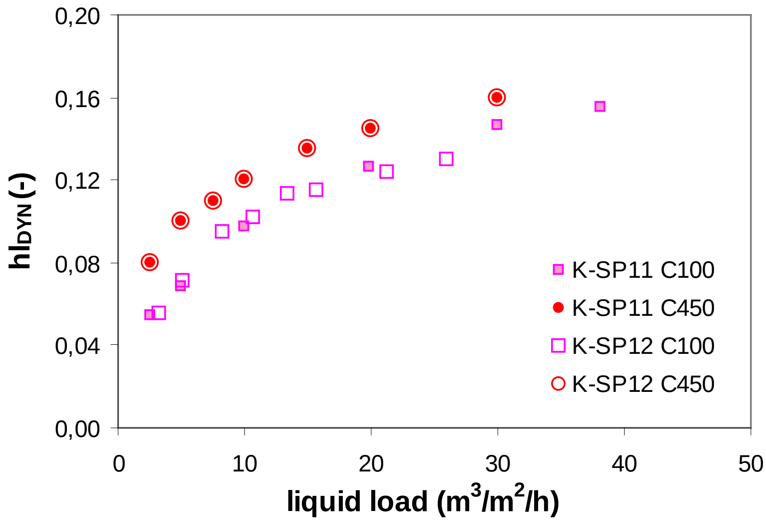

Figure 2. Dynamic hold-up as a function of liquid load for Katapak-SP11 (filled symbols) and Katapak-SP12 (empty symbols). Data on packings diameter of 100 mm (C100, square points) are from the present work, data on packings diameter of 450 mm (C450, circle points) are from Behrens[15]

2.2. Local measurements

The high energy X-ray tomograph available at the Laboratory of Chemical Engineering of the University of Liège (Belgium) was used to determine local liquid hold-up values. Tomographic measurements have been realized by scanning the packing cross sections at different heights of the column. The column was filled with four elements of catalytic packing (i.e. from the bottom to the top K1, K2, K3, K4) and four elements of conventional corrugated packing. The latter were placed three at the top and one at the bottom of the catalytic bed in order to improve the fluid distribution. The catalytic bed is placed between z=200 mm and z=1000 mm, being z the column axial coordinate. More details of the experimental setup are given in Aferka et al.[12]

The mechanical and tomographic equipment as well as the image reconstruction method are described in Toye et al.[11]. The images of the scanned cross sections provide information on the local liquid distribution between catalytic baskets and corrugated sheets. In order to obtain quantitative values of liquid hold-up in each cross section, the original methodology (thresholding, creation of masks, normalisation..) first proposed by Toye et al.[16] for plastic random packings and used in subsequent works[10,17] has been adapted to analyse images of the metallic modular packings presented here. Figure 3 illustrates reconstructed cross-section images of the Katapak-SP12 packing irrigated at two different liquid loads. Details of the geometry, such as the catalytic baskets filled by the particles, the corrugated sheets, the wall wiper and the column wall, are visible in the grey part of the images. The superimposed blue part shows the liquid distribution. Qualitatively it can be noticed that liquid is present both inside and outside the catalytic baskets and that at high liquid load the baskets are filled by more liquid. Some wetting of wall wiper is also clearly observable on tomographic images.

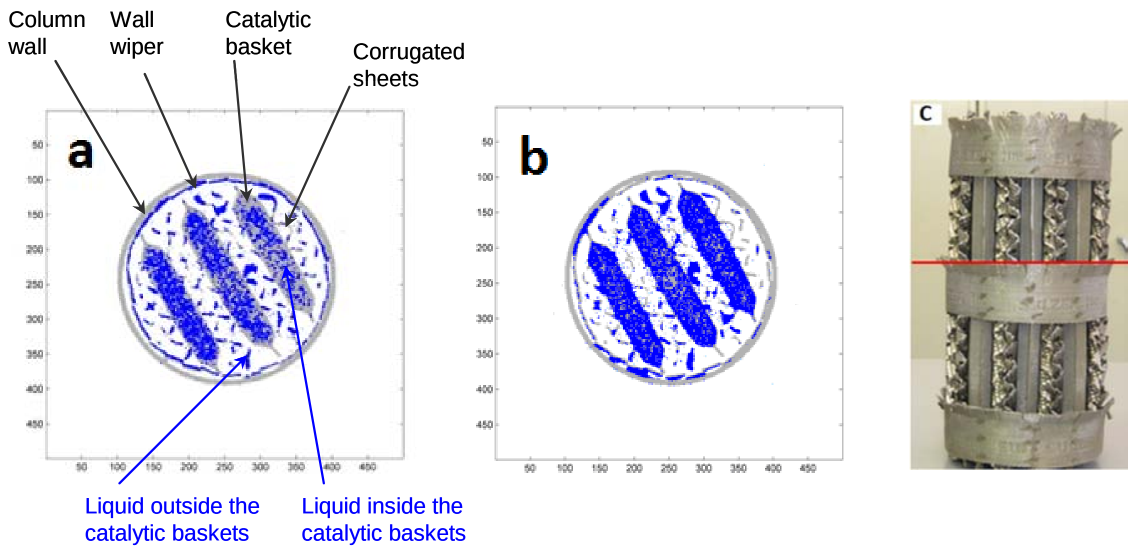

Figure 3. Reconstructed image of a cross section of Katapak-SP12 situated at a height of z=490 mm, irrigated by a liquid load of 4.62 $m^3/m^2/h$ (a) and 12.7 $m^3/m^2/h$ (b), and with no gas flowrate. In (c) the horizontal line shows the position of the cross-section with respect to the packing element height

Figure 4 shows the axial distribution along the catalytic bed height (z) of the liquid hold-up inside (hl_{CB}) and outside (hl_{OC}) the catalytic baskets. Shown data are for Katapak-SP11. In each cross-section of unit height, liquid hold-up inside the catalytic baskets, hl_{CB}, is defined as the ratio between the surface occupied by liquid inside the catalytic baskets and surface occupied by the catalytic baskets. Liquid hold-up in packing section outside the catalytic baskets, hl_{OC}, is defined as the ratio between the surface occupied by liquid outside the catalytic baskets and the surface of the column cross section. Figure 4a shows that at low liquid load the catalytic baskets are filled by liquid only for half of the height. This profile corresponds to the static hold-up behaviour observed by Aferka et al.[10] in experiments on single Katapak-SP12 packing. The hold-up values on the corrugated sheets are approximately constant except to the sections corresponding at the transitions between packings, where the contribution of catalytic baskets is absent and all the liquid is accounted for in the calculation of hl_{OC}. At high liquid load (Figure 4b) the catalytic baskets are almost saturated. The increment of liquid load corresponds also to an increment of the hold-up values outside the catalytic baskets. There are three wall wipers per packing element, i.e. at the bottom, at the top and at the half of packing height. Peaks in the hl_{OC} profile are observed at the sections corresponding to wall wipers, because the liquid flowing at the column wall is collected and redistributed inside the packing.

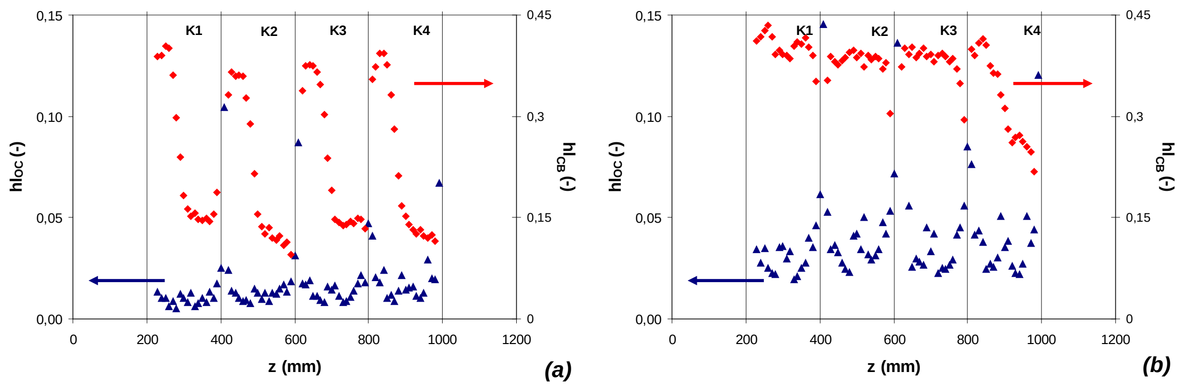

Figure 4. Axial distribution of liquid hold-up inside (hl_{CB}) and outside (hl_{OC}) the catalytic baskets of Katapak-SP11 obtained from measurements at liquid loads of 4.62 $m^3/m^2/h$ (a) and 23.1 $m^3/m^2/h$ (b)

The same behaviour is observed for the two tested configurations of modular packing. In Figure 5 the total hold-up is shown for Katapak-SP11 and Katapak-SP12 at two similar liquid loads along the column height (z). The total hold-up on each cross-section, hl, is obtained as follows. The liquid hold-up inside the catalytic baskets (hl_{CB}) is multiplied by the fraction of the surface occupied by catalytic baskets on the overall section surface measured on each section and added to hl_{OC}. This way, the hold-up data obtained by tomography are expressed as the ratio between the surface occupied by the liquid and the surface of the column cross section. Because the contribution of catalytic baskets is more significant, the liquid hold-up is locally higher for Katapak-SP11 than for Katapak-SP12. At low liquid load (Figure 5a) a strong variation in the hold-up profile is observed corresponding to the hl_{CB} trend. At high liquid load (Figure 5b) the hold-up profile is more constant along the column, because the catalytic baskets are saturated.

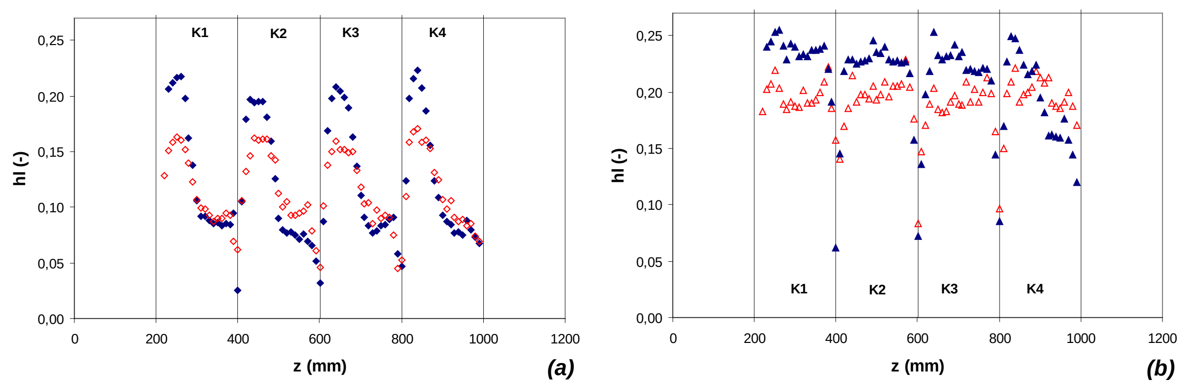

Figure 5. Total liquid hold-up measured on Katapak-SP11 (filled symbols) and Katapak-SP12 (empty symbols) at liquid load equals to 4.62 $m^3/m^2/h$ (a) and 23.1 $m^3/m^2/h$ (b)

3. Comparison and conclusions

By averaging local hold-up values obtained for different cross sections over the corresponding volume, one can determine global hold-up values which can then be compared to those obtained by using global measurements. To this end, a representative volumetric value of liquid hold-up in the packed bed is calculated for each liquid load as the average of the local (i.e. on each section) values measured on a large number of scanned sections (77 sections for Katapak-SP11 and 78 sections for Katapak-SP12). Finally, the total hold-up results obtained from tomographic measurements (hl_{TOMO}) are compared to the global values (hl_{GLOBAL}). This is shown in Figure 6, where data obtained for both packing types are presented in a parity plot. The very good agreement between the results obtained with these very different experimental techniques is extremely encouraging and demonstrates the soundness of the used experimental procedures and the image processing methodology.

In conclusion, liquid hold-up in two versions of the Sulzer catalytic structured packing Katapak-SP was investigated. Both global and local measurements have been carried out by using water at ambient conditions. New experimental results are presented which are useful in supporting theoretical developments in hydrodynamic and mass transfer modeling. In addition, results point out that high energy X-ray tomography is capable of providing quantitative liquid hold-up data and can be successfully used in combination with traditional measurements method. So if global values are needed, conventional techniques (e.g. based on draining, volumetric, RTD measurements) which are proved and less time consuming should be used. Instead if local values are required, tomographic technique is clearly the method to be chosen. The presented approach could be usefully extended to other complex devices used in integrated processes.

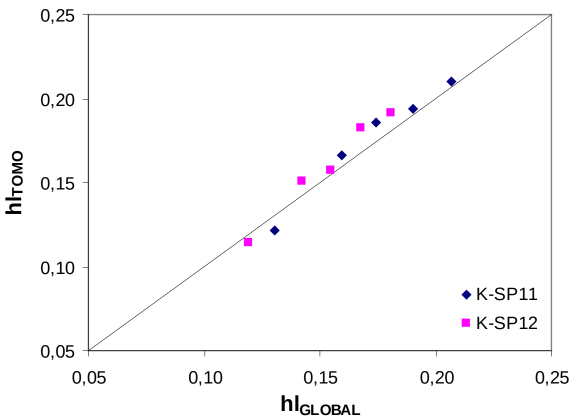

Figure 6. Parity plot of data of total liquid hold-up measured on Katapak-SP11 and Katapak-SP12 by tomographic experiments (hl_{TOMO}) and conventional draining methods (hl_{GLOBAL})

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