

Experimental analysis of the liquid hold-up contributions in catalytic packing Katapak-SP

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Abstract

Knowledge of liquid hold-up is essential for understanding reactive distillation column performances and develop fundamental models of packing behaviour. The modular structure of Katapak-SP, the novel family of catalytic structured packings manufactured by Sulzer Chemtech, provides a high degree of flexibility in process applications but complicates the design and the inner flows distribution. Therefore, for this type of packing the estimation of the different liquid hold-up contributions is a challenging task. The present investigation has focused attention on the experimental determination of this key parameter.

Keywords: catalytic structured packing, liquid hold-up, Katapak-SP, RTD

1. Introduction

The design and the reliable scale-up of reactive separation processes are strongly dependent on the hydrodynamic performance and on the transport phenomena imposed by the internals type and their geometry. The fluid dynamic behaviour of a packed column is heavily influenced by the amount of liquid, the so-called liquid hold-up, which accumulates within the packing during the operation. Many studies carried out on random and structured packings have pointed out the dependence of the liquid hold-up on packing dimensions and materials. The same approach should be applied to new catalytic structured packings because the knowledge of the liquid hold-up facilitates the prediction of fluid dynamic related parameters, such as the pressure drop, capacity, interfacial area and mass transfer volumetric coefficients.

Currently, the state of knowledge of liquid hold-up for catalytic structured packings is unsatisfactory. It may be due to the lack of experiments carried out with these new internals and also to the complexity of the geometry. Among catalytic structured internals, Katapak-SP is the new generation packing manufactured by Sulzer Chemtech. The packing consists of MellapakPlus type sheets ("separation elements") and wire-gauze catalyst bags ("reaction elements") assembled in alternate sequence

(Figure 1). This modular configuration provides a high flexibility by varying the separation to reaction zones ratio (Goetze et al, 2001) .



Figure 1. Structure of Katapak-SP 11 manufactured by Sulzer Chemtech

The geometrical structure of the packing determines the flows development inside the packed bed and consequently the overall column performances. The external wire gauze of the catalyst bags allows the liquid flow penetration and prevents the gas cross-over, thus limiting the use of this catalytic packing only to applications with liquid phase reaction. Therefore the liquid hold-up inside the catalyst bags influences the reactive performance of the internal. On the other side the liquid hold-up on the MellapakPlus layers is responsible for the interactions with the gas and for the related pressure drop in the column.

It is known that the liquid hold-up can be distinguished into a static and a dynamic contribution (Stichlmair and Fair, 1998). 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 packings. This fraction of the hold-up is characterised by a very high residence time. It is especially relevant for small random packings, e.g. Raschig rings and sphere beds, and for wire gauze structured packings, e.g. Sulzer BX and CY. On the other side, the dynamic hold-up is made up of the flowing liquid and strongly depends on the liquid load. For the metal sheets structured packing, e.g. Sulzer Mellapak, the dynamic hold-up can be considered to be the predominant part of the total hold-up (Suess and Spiegel, 1992).

In the case of structured catalytic packings, both fractions contribute significantly to the liquid hold-up and different measurements tests are required to determine them.

Usually, the static hold-up is determined by weighing the wet packing while the dynamic hold-up is obtained by measuring either the amount of liquid draining from the packed bed after stopping the liquid supply or the level reduction in the liquid feeding tank after stabilisation of the column (Brunazzi and Viva, 2006, Behrens et al., 2006). The total volume of liquid inside the internals can be measured, irrespective to the liquid state of motion, by means of tomography, e.g. high energy x-ray tomography (Aferka et al., 2006). The analysis of residence time distribution

(RTD) experiments allows determining both the liquid hold-up the axial dispersion coefficients for the packing (Kolodziej et al., 2005). The above listed techniques have been used by several authors to investigate the behaviour of catalytic structured packings.

Since unavoidable differences are connected with the nature of the experimental techniques used, the consistent analysis of the measurements of the overall hold-up and of the static and dynamic fractions is required. The consistency is especially needed for structured catalytic packings.

In this work we present a comparison of the experimental results obtained with different techniques, stressing the problem of consistency for the measurements times of liquid hold-up. Experiments have been carried out on Katapak-SP11 using an air-water system and a 100 mm inner diameter test column. The catalyst bags were filled with glass spheres with 1 mm of diameter. Some correlations proposed in the literature for the evaluation of the static hold-up have been used and the predicted values compared with the experimental results. The analysis has allowed the development of a standardised procedure that can be usefully applied to different or new catalytic structured packings.

2. Experimental apparatus and methods

2.1 Static hold-up measurements

The static hold-up was determined by using the following method. A dry packing element was firstly weighed, then it was immersed in a cylinder containing water and shaken for a while in order to remove trapped air bubbles in it. Then, the packing element was removed from the cylinder and being held just above it for some period of time to let the free water in the packing drain back to the cylinder. The packing was weighed several times during the drainage. The difference between the weight of the packing at a generic time t of the drainage period and the initial packing weight gave the amount of water captured by the packing at time t . The experiments were carried out on 4 different elements of Katapak-SP 11 and they were repeated two times to assess the repeatability of the measurements.

Due to the presence of catalyst bags and of metal sheets in the same packing, suitable investigation was carried out to find the static hold-up contribution inside the catalyst bags. One element of Katapak-SP11 packing was dismembered and two catalyst pockets were weighed at different times of drainage, after full immersion in water. During the drainage, each catalyst bag was vertically suspended and the liquid height inside was measured.

2.2 Dynamic hold-up measurements

The dynamic hold-up was determined by using two different methods, the draining method and the volumetric, also called gravimetric, method. The schemes of the test columns used to perform the experiments are presented in Figures 2a and 2b, respectively. A 100 mm inside diameter plexiglass column was filled with 10 elements

of Katapak-SP11 packing. Only the results obtained without the gas load are reported in this study.

Before a series of measurements, the column was first operated at high liquid load to ensure thorough wetting of the packed bed. Then the liquid stream was shut off and after some period of time the liquid flow-rate was set to the desired value.

In the first set of experiments the procedure was as follows (Brunazzi and Viva, 2006). After stabilisation, the liquid feed valve was closed and the liquid held up on the packing was allowed to drain down in a purposely built liquid collecting tank (D3, see Figure 2a). The time-dependent liquid dropping was measured by means of a Differential Pressure transmitter and the signal, with a data rate of 2 readings per second, stored on a PC. The dynamic free-draining hold-up was calculated as the volume of liquid per unit of column volume (the part filled with packing). The procedure was repeated for the entire range of liquid loads investigated

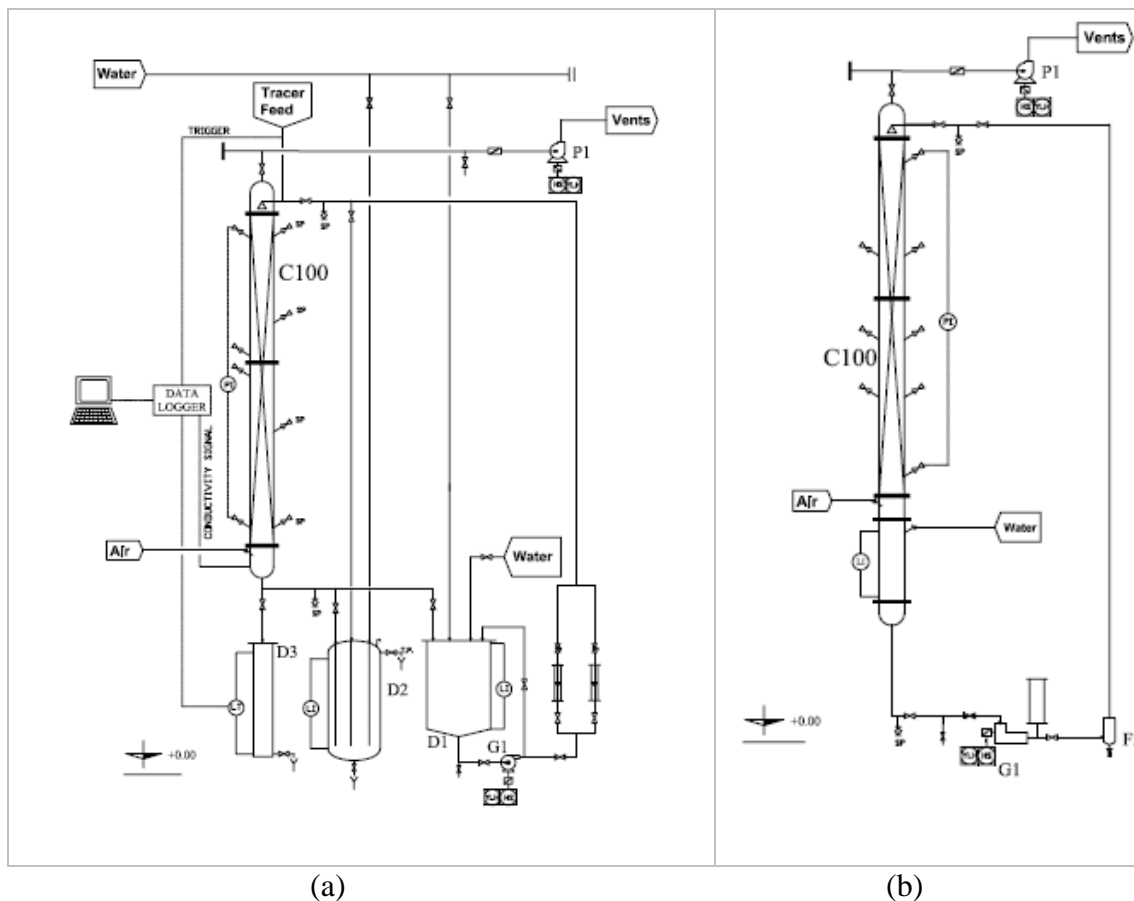


Figure 2. Schematic representation of the experimental setup.

To perform the second set of experiments the bottom structure of the column was changed. This second experimental-set up was also used to collect simultaneously pressure drop and liquid hold-up measurements as a function of the gas and of the liquid loads. As shown in Figure 2b, a calibrated section was flanged to the bottom part of the column and used as the liquid feeding tank. The liquid was supplied in

closed circuit. The liquid volume in the peripheral equipment was measured preliminarily. At steady state operation, the dynamic hold-up of the packed bed was determined by recording the liquid level change in the calibrated bottom (Brunazzi et al., 2002). The procedure was repeated for the entire range of liquid loads investigated.

2.3 RTD measurements

The same column set-up shown in Figure 2a was used for the residence time distribution (RTD) experiments. In this case, after the liquid flow stabilisation in the column, the tracer (an aqueous solution of sodium chloride) was injected via a syringe just before the liquid distributor at the top of the column. At the bottom of the column, the whole amount of liquid was collected and the tracer concentration measured in a purposely built mixing cup equipped with a conductometer flow through probe. The liquid leaving the column was collected into a separate tank (D2). The computer data acquisition system was connected both at the syringe at the top of the column and at the flow-through probe at the bottom of the column. This allowed the tracer concentration to be measured in the liquid stream leaving the column. The results were taken into consideration only if the difference between the zero moment calculated from RTD curve, which represents the mass of the tracer, agreed with the mass of the injected tracer. Liquid loads employed were up to $38 \text{ m}^3/\text{m}^2 \text{ h}$.

3. Results and discussion

The static hold-up experiments carried out on the single catalyst bags, enabled to measure the liquid height inside the bag, h_{cap} , as a function of time. The results are shown in Figure 3.

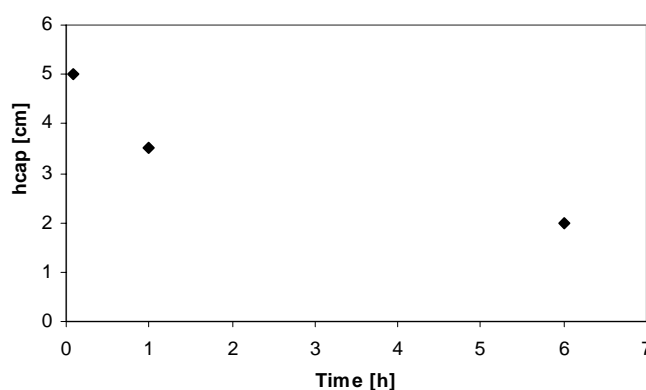


Figure 3. Height of liquid inside a single catalytic pocket as a function of the drainage time

The same approach was used on 4 different packings of Katapak-SP11 and the static hold-up measured on these is reported in Figure 4. Even though the same qualitative behaviour was found for the two experiments, a different time of drainage was observed, being the time of drainage of the packing considerably lower than that for

the single catalyst bag. This difference may be explained by considering that in the packing the metal sheet are forced against the catalyst bags, thus enhancing the liquid drainage.

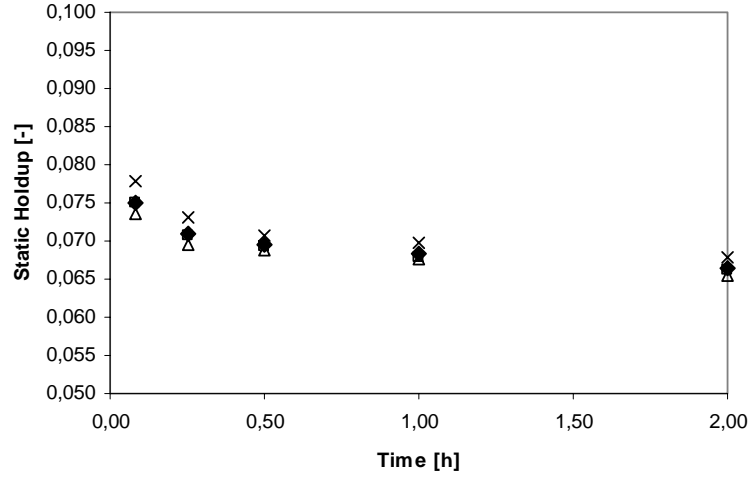


Figure 4. Static (residual) hold-up as a function of draining time measured separately for four different packing elements

The static hold-up values obtained have been analysed on the basis of the model proposed by Behrens (2006) for single catalyst pocket. According to Behrens, the static hold-up is comprised of three contributions: the pore hold-up, the capillary rise hold-up and the residual hold-up. Since the catalyst bags in our experiments are filled with glass spheres, the pore hold-up fraction is not present, therefore:

$$hl_{tot} = hl_{cap} + hl_{res}$$

The liquid height inside the catalyst pocket, which was observed and measured also in our experiments, is basically due to the capillary forces that counteract the gravity. For a vertical capillary, where no external pressures are imposed, the following correlation for the capillary rise height is proposed, taking into account the diameter of the spheres (d_p), the liquid surface tension (σ), the liquid density (ρ_L), the void fraction of the catalyst bags (ε_r), and the contact angle (θ) among the liquid and the solid spheres.

$$h_{cap} = \frac{6(1 - \varepsilon_r)\sigma}{d_p \cdot \varepsilon_r \cdot \rho_L \cdot g} \cos \theta$$

The predicted value of 4.58 cm for the capillary rise height agrees with the experimental value of 5 cm measured on the single catalyst bag after 5 min of drainage.

For the whole volume of the Katapak-SP11, the liquid hold-up due to the capillary rise can be evaluated by the following correlation:

$$hl_{cap} = \frac{h_{cap} \cdot \varepsilon_r}{h_{cb}} \varphi_{cb}$$

where h_{cb} is the height of the catalyst pocket and φ_{cb} the fraction of packing element occupied by the catalyst containing pockets. The third contribution to the static hold-up is due to the residual liquid retained by capillary forces at the contact points of the glass spheres above the capillary height after the drainage. This contribution is evaluated from the following simple model:

$$hl_{res} = 0.028 \frac{1 - \varepsilon_r}{\varepsilon_r} \frac{h_{cb} - h_{cap}}{h_{cap}} \varphi_{cb}$$

To adapt this simplified model to the whole volume of the packing, an additional contribution to the static hold-up of about 0.4% was estimated to be due to the wire gauzes of collars and seams of the catalyst bags.

We found a good agreement between the predicted value for the total static hold-up, that amounts to 6.04%, and the experimental value of 6.83% measured after 1h of drainage of the packing. This difference may be explained by considering some inevitable uncertainties in the geometrical characteristics of the packing. For example a 5% decrement of the void fraction of the catalyst bags causes an increment of more than 5% in the value of the predicted static hold-up. Moreover, a small contribution to the static hold-up may be also given by the Mellapak layers.

The same investigation on the measurements time has been used during the measurements of the dynamic free draining hold-up carried out with the DPcell level measurement. The influence of the drainage time is highlighted in Figure 5, where the liquid hold-ups measured over the range of liquid loads at different times are shown versus the values measured after 1 hour drainage from stopping the liquid.

The dynamic free draining hold-up results obtained with the two different methods described in the Section 2.2 have been compared for consistent time of measurements. The dynamic hold-up measured after 1h of drainage with the volumetric method is in very good agreement with the values obtained with the method of the liquid level variation in the bottom of the column on condition that the drainage time of 1h has been waited before taking the starting level of liquid. Increasing the drainage time, the packing inside the column gets empty and partially dry, thus requiring more liquid to reach again the proper wetting state of the catalyst bags.

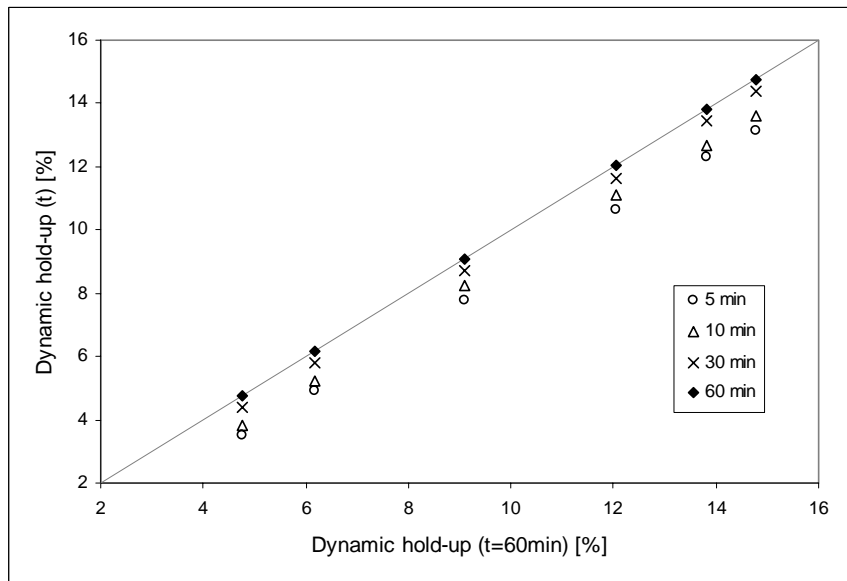


Figure 5. Liquid hold-up measured after a time t vs liquid hold-up measured after 1 hour of drainage time

The experiments carried out with the tracer for the estimation of the RTD allows the total liquid hold-up inside the column to be determined. Under the assumption of the plug flow model for the tracer material balance, the residence time distribution function $E(t)$ is demonstrated to be defined as:

$$E(t) = \frac{V^* c(t)}{\int_0^{\infty} V^* c(t) dt}$$

Where V^* represents the volumetric flow of liquid.

The first moment represents the mean residence time and is the parameter of interest for the evaluation of the liquid hold-up:

$$t_m = \int_0^{\infty} tE(t) dt = \frac{L^* h_L}{u_L}$$

where L is the packed bed height, u_L is the liquid superficial velocity and h_L is the liquid hold-up in the column.

Gorak et al (2006) studied the RTD behaviour of for the catalytic structured packing Multipak and showed that the hold-up derived from the RTD measurements agree with total hold-up from free-draining experiments.. The present experimental results for Katapak-SP11 confirm this experimental evidence. In fact, a very good agreement has been observed by comparing the hold-up derived from the RTD curve and the total hold-up given by the sum of the static and the dynamic free-draining hold-up (see Figure 6). The consistency of the addition of this two contributions has to be verified, by taking both the values measured after the same drainage time

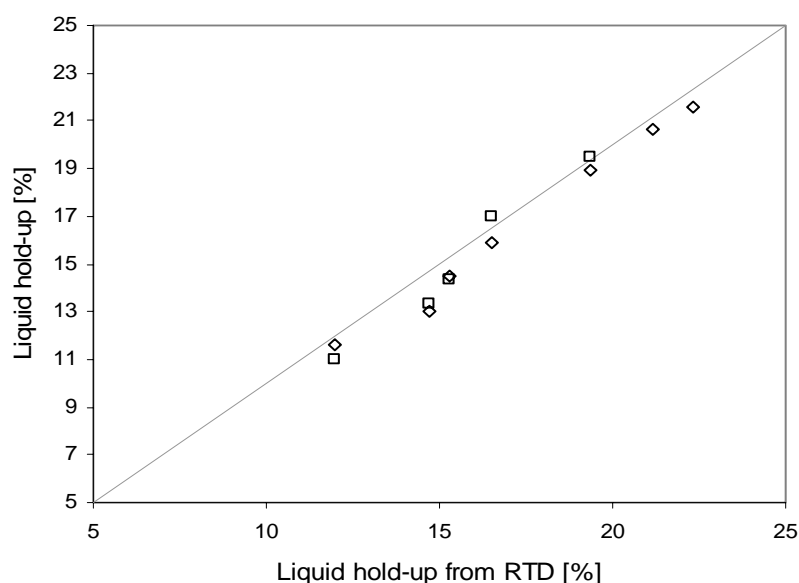


Figure 6. Liquid hold-up derived from the RTD experiments vs total hold-up given by the sum of the static hold-up and the dynamic hold-up measured with the two different methods.

4. Conclusion

The present study has demonstrated the importance of choosing consistent times for liquid hold-up experiments. In particular, due to the complex geometry of Katapak-SP packing, both the static hold-up and the dynamic hold-up contribute to the total hold-up. This fact requires different experimental methods, enabling to focus on the different scales of the problem. The hold-up was measured for one single catalyst bag, for a single packing as well as for a 2 m packed column, in order to point out the correspondence of the hold-up contribution to the different parts of the packing. The results have been compared with predicted values obtained by simple models proposed in literature. Moreover, the different methods have been analysed. The combined results obtained from the static hold-up experiments and from the dynamic hold-up measurements have been found to be in good agreement with the total hold-up evaluated from the RTD experiments. Due to the fact that higher deviation has been observed in the measured static and dynamic hold-up values at short time of measurements, a drainage time of 1h can be suggested as the suitable drainage time for all the techniques tested.

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