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The influence of modular structure on the hydrodynamics of catalytic structured packings for reactive separation processes

A. Viva, E. Brunazzi
Dipartimento di Ingegneria Chimica, Chimica Industriale e Scienza di Materiali,
Università di Pisa, Via Diotisalvi 2, 56126 Pisa, Italy.
e-mail: aurora.viva@ing.unipi.it, e.brunazzi@ing.unipi.it

Residence time distribution (RTD) and drainage experiments are used to shed light into the hydrodynamics of the catalytic structured packing Katapak-SP11. The liquid flow behaviour and the dynamic liquid holdup were derived from the RTD results and compared with the total liquid holdup derived from the drainage experiments. The liquid considered static from the viewpoint of free-draining experiments becomes dynamic during the normal column operation. The examination of the liquid drainage curves and the consistency of different techniques are discussed.

1. Introduction

The possibility of exploiting an advantageous interaction between separation and reaction has favoured the implementation of heterogeneous catalysis in reactive separation columns. Improvements in this direction are expected from a new generation of modular catalytic structured packing which allows certain flexibility with respect to the variation in the reaction to separation requirements in a single unit.
The commercially available catalytic structured packing under study is the Sulzer Katapak-SP11 (Figure 1) which is made of wire gauze envelopes filled with catalyst spheres alternated to corrugated metal sheets of the MellapakPlus family.
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 Katapak-SP only to applications with liquid phase reaction. Therefore the liquid holdup inside the catalyst bags influences the reactive performance of the internal. On the other side the liquid holdup on the MellapakPlus layers is mainly responsible for the interactions with the gas.
Hence, the knowledge of the liquid holdup represents the key input for the development of mechanistic models and for the prediction of fluid dynamic related parameters, such as pressure drop, capacity, interfacial area and mass transfer volumetric coefficients. However, for Katapak-SP11 the state of knowledge of liquid holdup is still unsatisfactory. The lack of experiments carried out with this new internal explains only

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partially this gap. It is also the complexity of the phenomena imposed by the hybrid geometry of Katapak-SP which still requires a detailed comprehensive understanding.

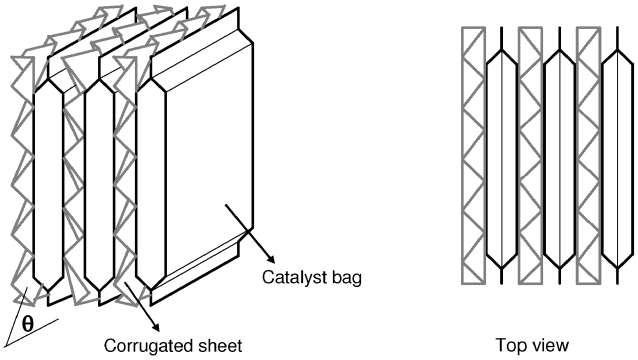

Figure 1. Structure of the modular catalytic packing Katapak-SP11.

For these reasons, the liquid holdup and flow behaviour in Katapak-SP have been evaluated by using proven gravimetric and tracer based measurement techniques. Information on the different holdup contributions is provided and the residence time distribution (RTD) analysis relates the liquid holdup in the packing to the flow behaviour.

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 holdup. The aim is to provide a standardized procedure to be usefully applied to different or new catalytic structured packings. The understanding of the liquid distribution inside the hybrid packing structure is essential as it governs the performance of the hybrid unit as a whole.

2. RTD measurements

Residence time distribution (RTD) experiments by injection of a salt tracer in the liquid flow were performed in a 100mm inner diameter column equipped with the catalytic packing Katapak-SP11. The catalyst bags were filled with glass spheres with 1 mm of diameter. The air-water system was used at ambient conditions. A detailed description of the experimental apparatus is reported in Viva and Brunazzi (2007).

The experiments allow determining the total liquid holdup inside the column. Under the assumption of the plug flow model and taking into account of the axial dispersion, the tracer material balance can be written in dimensionless form as in Equation 1.

$$\frac{\partial c}{\partial \theta} = -\frac{\partial c}{\partial Z} + \frac{D_{ax}}{u_L L}\frac{\partial^2 c}{\partial Z^2} = -\frac{\partial c}{\partial Z} + \frac{1}{Pe_L}\frac{\partial^2 c}{\partial Z^2} \tag{1}$$

where $\theta = \frac{t}{t_m}, t_m = \frac{L}{u_L}, Z = \frac{z}{L}, \; Pe_L = \frac{u_L L}{D_{ax}}$ (2)

In the above relations c is the tracer concentration, z the axial coordinate, L the packed bed height, u_L the liquid interstitial velocity, t the residence time, D_{ax} the axial dispersion coefficient and Pe_L the Peclet axial number.
Assuming a pulse function for the tracer and under the proper boundary conditions for closed system, the balance equation is solved in the Laplace domain. The residence time distribution function $E(t)$ is demonstrated to be defined as in Equation 3.

$$E(t) = \frac{V^* \cdot c(t)}{\int_0^\infty V^* \cdot c(t) dt} \tag{3}$$

where V^* represents the volumetric flow of liquid. The first moment in Equation 3 represents the mean residence time of the tracer and is the parameter of interest for the evaluation of the liquid holdup.

$$t_m = \int_0^\infty t \cdot E(t) dt = \frac{L \cdot h_{RTD}}{u_{SL}} = \frac{L}{u_L} \tag{4}$$

where h_{RTD} is the liquid holdup in the column and u_{SL} is the liquid superficial velocity.
Moreover, analysis of RTD allows identifying the variation of flux regime in the column (see previous studies on Katapak-S and Multipak by Kolodziej et al. 2005, Götze et al. 2001, Moritz and Hasse, 1999). In fact, the RTD curves point out the transition from a condition of partially wet reactive zone to a regime named liquid load point, characterized by catalyst bags completely filled with liquid. When the liquid load exceeds the Load Point, the catalyst bags rejects the exceeding liquid which flows as a by-pass over the MellapakPlus layers. This behavior is recognizable in the RTD curves because of the different residence time of liquid flowing inside the catalyst bags and over the corrugated layers. The shape of the RTD curves and consequently the distribution of the liquid inside the packing are strictly related to the modular configuration and to the geometry of the packing.
The trend derived experimentally of $E(\theta)$ as a function of the dimensionless time θ and for different liquid loads is shown in Figure 2. At low liquid loads (2.5 and 5 m^3/m^2h) the curves are narrow. The tails are due to the presence of stagnant zones inside the catalyst bags that are not completely filled with liquid. The fraction of liquid flowing outside the bags increases with liquid load, although the load point is not yet reached (10 m^3/m^2h). The shorter residence time of the liquid flowing over the Mellapak layers implies the anticipation of the RTD curve. This effect is even more visible for liquid loads above the load point. The mean residence time is reduced because of the high fraction of liquid flowing directly over the MellapakPlus layers outside the catalyst bags. In this condition, a strong back-mixing of liquid can be supposed, due to the crossing liquid from and towards the catalyst bags especially at the contact points of the wire gauze with the corrugated layers. This back-mixing generates long tails in the RTD curves and a strong deviation from the desired ideal plug flow.

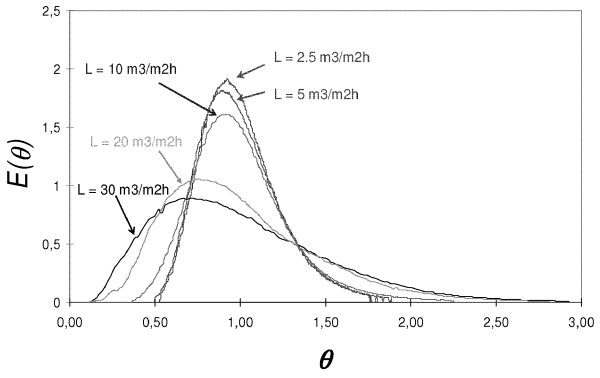

Figure 2. Dimensionless RTD curve at different liquid loads.

From the RTD curve the holdup in the column can be estimated and this holdup actively participates to mass transfer operations. For conventional packings, the RTD holdup corresponds to the dynamic holdup determined by draining experiments. For catalytic structured packing, on the contrary, the holdup derived from the RTD measurements agrees with the total holdup given by the sum of the dynamic and the static contributions derived from the draining experiments on condition that consistent measurement times are used (Viva and Brunazzi, 2007).
In the following the results obtained with the drainage method for Katapak-SP11 are reported and eventually compared with the above mentioned RTD holdup.

3. Drainage experiments

3.1 Static holdup

Viva and Brunazzi (2007) have shown that the static holdup can estimated by extending the model proposed by Behrens (2006) for single catalyst pocket to the Katapak-SP11 geometry. According to Behrens, the static holdup is comprised of three contributions: the pore holdup, the capillary rise holdup and the residual holdup. Since the catalyst bags in our test case are filled with glass spheres, the pore holdup fraction is not present. The capillary rise holdup is estimated from the balance bewteen the capillary forces and the gravity. The predicted capillary rise height value agrees with the experimental value observed by Aferka et al. (2007) on the catalyst bags holdup obtained with tomography experiments carried out on Katapak-SP12. The residual liquid is retained by capillary forces at the contact points of the glass spheres above the capillary height after the drainage. This fraction can be estimated with correlations developed for trickle beds of spheres and described in Van Hasselt (1999).
For Katapak-SP11 the predicted static holdup has found to be equal to 6%, and in good agreement with the experimental value obtained after 1 h of drainage of the packing. The details on the used correlations and the experimental procedures are reported in Viva and Brunazzi (2007).

3.2 Dynamic holdup

To measure the dynamic free-draining holdup, the liquid streams were cut off while the liquid was allowed to drain from the packing in a purposely built liquid collecting tank. Brunazzi and Viva (2006) observed a strong dependence of the volume of draining liquid on time. The dynamic free-draining holdup was calculated as the volume of liquid drained per unit of column volume (the part filled with packing). In particular a reference time of 1h was set as a proper draining time for the dynamic holdup evaluation.

In order to confirm this statement, the approach proposed by Urrutia et al. (1996) for the holdup analysis of trickle beds has been extended to Katapak-SP11. In the original model, developed under the assumption of laminar liquid film flow in a draining packed bed, it was found a linear dependence of the draining liquid volume on $t^{-0.5}$. According to Urrutia et al. (1996), the free-draining liquid holdups may be evaluated by extrapolating the dimensionless drained liquid volumes at infinite time (i.e. $t \rightarrow \infty$ or $t^{-0.5} \rightarrow 0$). For all the tested liquid loads, a linear dependence of draining liquid volume on time has been observed also for Katapak-SP11 in the present work. The drainage kinetics is evident in Figure 3 where the experimental drained liquid volumes divided by the values extrapolated at time $t \rightarrow \infty$ are plotted versus $t^{-0.5}$.

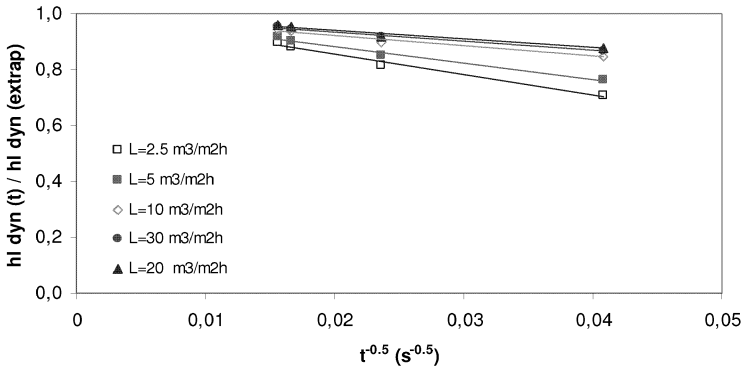

Figure 3. Dependence of the dimensionless draining liquid volume on $t^{-0.5}$.

4. Comparison and conclusions

The dynamic holdup values at infinite draining time differs from the experimental values obtained with the gravimetric method after 1h of draining for a maximum deviation of 5%. This confirms the suitability of the chosen draining reference time of 1h for the estimation of the dynamic holdup in the used column configuration. Of course for taller columns the infinite draining time method allows the reduction of the experimental draining time that otherwise would be extremely long.

Finally the static holdup has been added to the dynamic holdup at infinite time and to the dynamic experimental value at 1h of draining. The comparison between these total holdup values obtained from the drainage measurements with the total RTD holdup is

reported in Figure 4. The results and the consistency of these different techniques are therefore validated also for the complex modular structured of Katapak-SP.

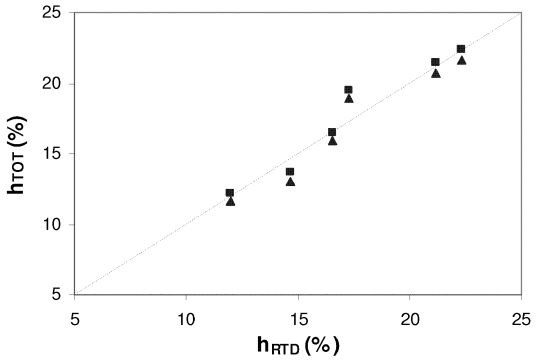

Figure 4. Total liquid holdup vs RTD holdup. Total holdup is given by the sum of the static holdup and the dynamic holdup measured after 1h of draining (▲) or the dynamic holdup from the infinite time method (■).

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