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# Reactive distillation with KATAPAK®

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## Abstract

KATAPAK-SP and -S are structured catalytic packings for reactive distillation or gas–liquid reactors available from laboratory to industrial scale. Applying the KATAPAK-SP concept, the ratio of catalyst volume fraction to separation efficiency can be varied over a wide range, therefore the design of a reactive distillation column can be further optimized to fit each reaction system best.

Pressure drop, separation efficiency, dynamic liquid hold-up and residence time distribution have been investigated for KATAPAK-SP in a diameter of 250 mm. Results for different gas and liquid loads are presented. The findings are compared with results for the structured laboratory scale packing KATAPAK-S and the industrial scale packing KATAPAK-S 170.Y.

The dehydration of tert-butyl alcohol was selected as a sample reaction to illustrate the influence of different catalytic structures on the performance of the reactive distillation column. The setups are compared with respect to dimensions and economics.

*Keywords:* Reactive distillation; KATAPAK; Pressure drop; Separation efficiency; Liquid holdup; RTD

## 1. Introduction

Reactive distillation combines chemical reaction and thermal separation of the products in one unit operation and has, therefore, been a focus of research in the chemical process industry and academia in the last years [1–3]. It is extremely advantageous for reactions that cannot be driven to total conversion in a conventional reactor due to an unfavorable chemical equilibrium. Since in a reactive distillation process the reaction products are continuously removed from the reaction mixture, chemical equilibrium limitations can be overcome and high reaction rates are maintained. This results in higher conversion compared to conventional heterogeneous catalyzed processes, in which reaction and product separation are carried out sequentially.

KATAPAK-S (S implies Sandwich) is a structured catalyst support for use in gas–liquid reaction systems such as trickle bed reactors, bubble columns or reactive distillation processes, in which catalyst granules can be embedded. It is suitable for many catalytic applications, such as esterifications, e.g. to methyl acetate [4,5], ethyl acetate [6] and butyl acetate [7], etherifications, e.g. to MTBE [8], hydrogenations, e.g. of anthrachinone [9], or alkylations [10].

Sulzer Chemtech has extended the product line with KATAPAK-SP (SP for Separation Performance) to fit the requirements of every reaction system best [11]. The advantage of this structured packing is its modular design, which allows to vary the catalyst fraction and the separation efficiency. In this work, comprehensive studies on fluid dynamics and separation efficiency of industrial scale KATAPAK-SP are reported. They are the key element for the design of reactive distillation processes using KATAPAK-SP.

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Table 1
Influence of separation layers on the catalyst volume fraction and the separation efficiency NTSM

| Type KATAPAK-SP | SP 11 | SP 12 | SP 13 |
|---|---|---|---|
| Number of catalyst layers:number of separation layers | 1:1 | 1:2 | 1:3 |
| NTSM (l/m) | 2 | 2.5 | 3 |
| Catalyst volume fraction (vol.%) | ∼40 | ∼30 | ∼20 |

## 2. Fundamentals

### 2.1. KATAPAK-SP

By combining catalyst containing wire gauze layers (catalytic layers) with layers of Mellapak, MellapakPlus or the wire gauze packings BX and CY (separation layers), reactive distillation packings with separation efficiencies up to 4 NTSM (Number of Theoretical Stages per Meter) and catalyst volume fractions up to 50% can be obtained. In KATAPAK-SP, state of the art catalyst granules (e.g. ion-exchange resins, noble metals on alumina, activated carbon supports, etc.) can be immobilized. In Table 1, the influence of the number of separation layers (compared to the number of catalytic layers) on the separation efficiency and the catalyst volume fraction of the resulting packing is shown.

For all experiments presented in this paper, KATAPAK-SP type 12 was selected. This reactive distillation packing consists of two layers of MellapakPlus per catalyst layer. The catalytic layers were

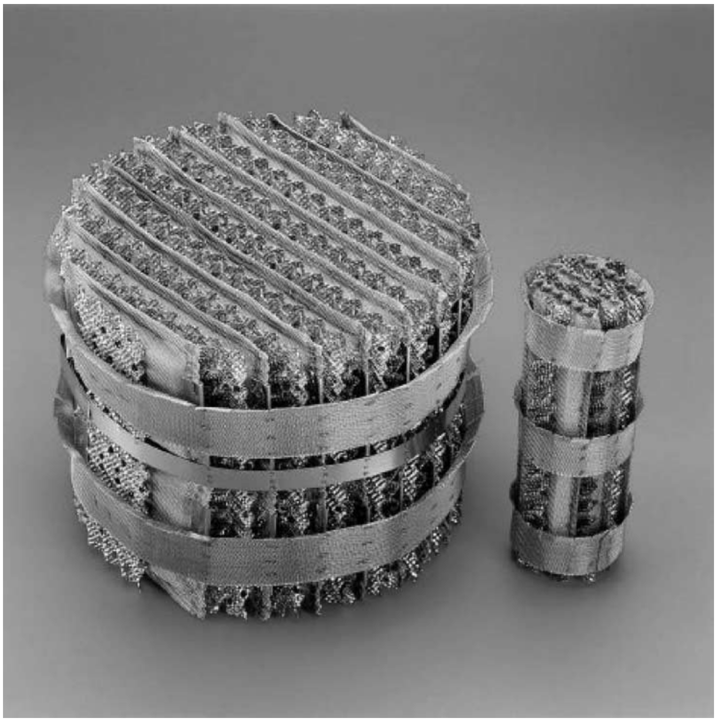

Fig. 1. Industrial and laboratory scale KATAPAK-SP 12.

Table 2
Geometric data of the KATAPAK-SP 12 industrial packing used for the fluid dynamic measurements

| | |
|---|---|
| Packing length | 200 mm |
| Tube diameter (ID) | 250 mm |
| Wire thickness | 0.25 mm |
| Mesh size | 0.5 mm |
| Catalyst layer height | 13 mm |
| Separation layer height | 6.5 mm |
| Diameter glass spheres | 1 mm |
| Volume fraction glass spheres | 0.236 |
| Number of catalytic layers | 9 |
| Number of separation layers | 18 |

filled with glass spheres. A picture of an industrial and a laboratory scale element is shown in Fig. 1, details are given in Table 2.

### 2.2. KATAPAK-S

In KATAPAK-S, the same type of catalyst granules as in KATAPAK-SP can be immobilized between two sheets of metal wire gauze, forming "sandwiches". Each of these sheets is corrugated, resulting in a structure with flow channels of a defined angle and hydraulic diameter. The sandwiches are assembled with the flow channels in opposed orientation, so that the resulting unit is characterized by an open cross-flow structure pattern. A picture of an industrial scale KATAPAK-S 170.Y is shown in Fig. 2. Details about the fluid dynamics of the laboratory scale packing KATAPAK-S can be found in [1], data about the industrial packing KATAPAK-S 170.Y and the scale-up procedure have been presented by Moritz et al. [12].

## 3. Separation efficiency

### 3.1. Experimental

Measurements of the separation efficiency of KATAPAK-SP 12 industrial packings were carried out in a 250 mm diameter distillation column using

Fig. 2. Industrial scale KATAPAK-S 170.Y.

the test system chlorobenzene/ethylbenzene at total reflux. The column was filled with 15 packing elements of 200 mm length giving a total bed length of 3000 mm. The experiments were carried out at ambient pressure. Samples from the top and bottom of the column were analyzed by gas chromatography to determine the separation efficiency, which is given in NTSM.

### *3.2. Results*

The experimental results of the separation efficiency measurements with KATAPAK-SP 12 industrial packings are shown as a function of the *F*-factor for ambient pressure in Fig. 3 together with results for KATAPAK-S 170.Y and the laboratory scale packing KATAPAK-S.

Like for KATAPAK-S, two flow regimes are found for KATAPAK-SP [13]: For liquid loads up to the load point the liquid flows mainly inside the catalytic packing structure, and for liquid loads above the load point a liquid bypass on the outer surface of the structure is observed. Up to the load point, the separation efficiency of KATAPAK-SP 12 decreases about 15% compared to the NTSM at lowest measured *F*-factors to a mean NTSM = 2.5. The decrease of the separation efficiency in a certain gas load range is typical for metal sheet packings like Mellapak or MellapakPlus, and is, therefore, also observed for KATAPAK-SP 12. The load point of KATAPAK-SP 12 at ambient pressure is reached at an *F*-factor of about 1.8 $\text{Pa}^{0.5}$. Above the load point, the separation efficiency increases rapidly due to a better vapor/liquid mass transfer of the liquid bypassing the catalytic layers between the catalytic and separation layers.

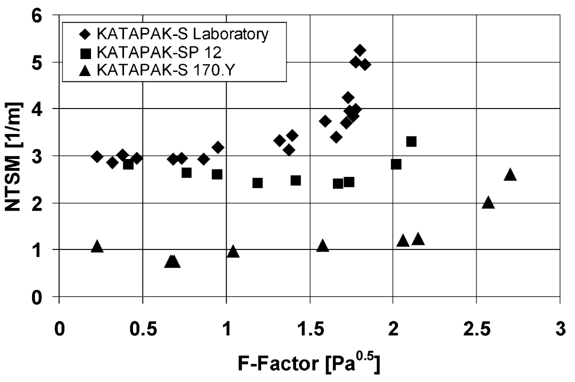

Fig. 3. Separation efficiency of KATAPAK-SP 12, laboratory scale KATAPAK-S and industrial KATAPAK-S 170.Y packings at total reflux as function of the *F*-factor at ambient pressure.

The comparison of the separation efficiency course of KATAPAK-SP 12 to both KATAPAK-S structures shows the same dependency on the gas load, but different NTSM values: For the laboratory scale KATAPAK-S, an average of NTSM = 3 and for the industrial packing an NTSM = 1 was observed. The load point for the laboratory scale KATAPAK-S is reached at an *F*-factor of 1.5 $\text{Pa}^{0.5}$ and for the industrial KATAPAK-S at about 2.2 $\text{Pa}^{0.5}$. These differences in the separation efficiency and the load points of the three catalytic packings are caused by their different surface areas, thus the structure with the highest surface area (laboratory scale KATAPAK-S) has the highest separation efficiency and the load point at the lowest liquid load.

## 4. Pressure drop and dynamic liquid hold-up

### *4.1. Experimental*

Pressure drop measurements were carried out in a 250 mm ID column for fluid dynamic experiments, equipped with 14 elements of KATAPAK-SP 12 with a total packing height of 2800 mm. The liquid flow was set by a rotameter, the gas flow was measured by a Prandtl tube. The pressure drop caused by the packings was measured with an U-tube manometer. The experiments were carried out with water as liquid and air as gas at ambient conditions.

The dynamic hold-up was determined by measuring the amount of water dropping out of the packings after stopping all feed flows (water and air).

### *4.2. Results*

In Fig. 4, the experimental pressure drop is shown for the system water/air. The liquid load was varied between 0 (dry pressure drop) and 30 $\text{m}^3/\text{m}^2\,\text{h}$, the *F*-factor was increased up to the flooding of the packings. Up to the load point, which is reached at liquid loads of about 10 $\text{m}^3/\text{m}^2\,\text{h}$, the pressure drop increases slowly over nearly the whole range of *F*-factors. For

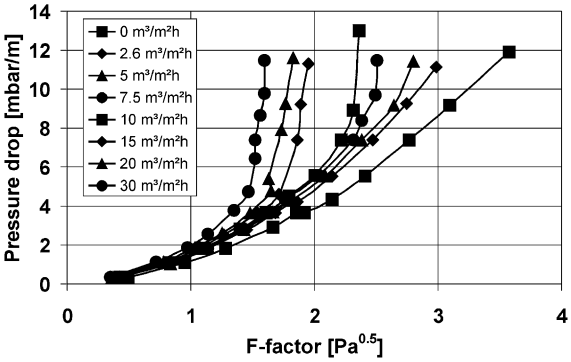

Fig. 4. Experimental pressure drop of KATAPAK-SP 12 as function of the gas and liquid load (lines smoothed).

liquid loads above the load point, a strong pressure drop increase is observed for even moderate $F$-factors. This stronger pressure drop is caused by an increased liquid flow outside the catalytic layers, which interacts with the vapor flow and thereby raises the pressure drop.

A comparison of the pressure drop for the three catalytic packings is shown in Fig. 5. The pressure drop for the laboratory scale KATAPAK-S and KATAPAK-SP 12 is comparable up to the flooding point of KATAPAK-S. KATAPAK-S 170.Y has a lower pressure drop over the whole range of $F$-factors than the other two packings because of the wider gas flow channels of this packing type. At the shown liquid load ($\sim 15\,\mathrm{m^3/m^2\,h}$) flooding of the laboratory scale KATAPAK-S starts at an $F$-factor of $1.5\,\mathrm{Pa^{0.5}}$, for KATAPAK-SP 12 at $1.9\,\mathrm{Pa^{0.5}}$ and for KATAPAK-S 170.Y at $3.3\,\mathrm{Pa^{0.5}}$.

Fig. 6 shows the experimental dynamic liquid hold-up for the three catalytic structured packings, measured with water. The laboratory scale KATAPAK-S packing has the highest liquid hold-up over the whole range of liquid loads, followed by KATAPAK-S 170.Y and KATAPAK-SP 12. The higher hold-up of the two KATAPAK-S structures is caused by the corrugation of the wire gauze, which keeps more liquid on the outside of the catalytic layers.

In additional measurements, the influence of a countercurrent gas flow on the dynamic liquid hold-up of KATAPAK-SP 12 was examined. The results are shown in Fig. 7. It can be seen, that over the whole range of $F$-factors, the hold-up is nearly constant. Only

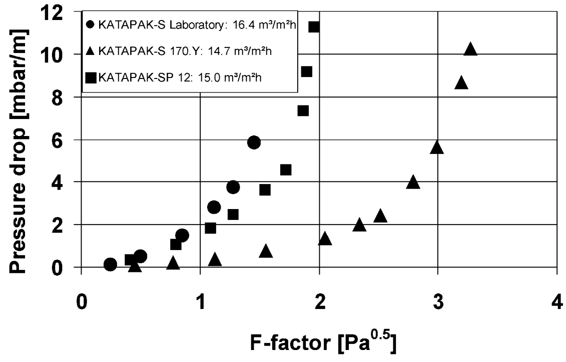

Fig. 5. Experimental pressure drop of KATAPAK-SP 12, laboratory scale KATAPAK-S and industrial KATAPAK-S 170.Y packings as function of the $F$-factor for a fixed liquid load.

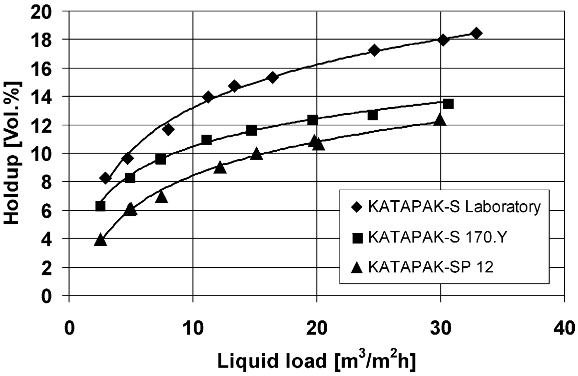

Fig. 6. Experimental dynamic liquid hold-up of KATAPAK-SP 12, laboratory scale KATAPAK-S and industrial KATAPAK-S 170.Y packings as function of the liquid load (lines smoothed).

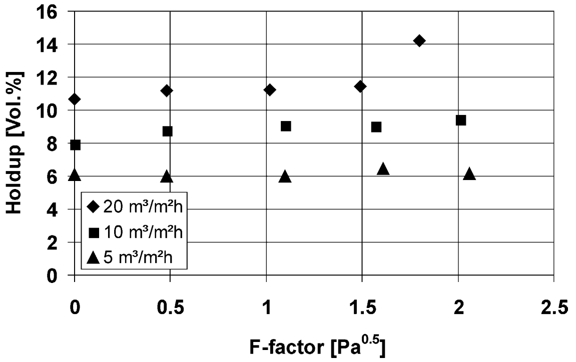

Fig. 7. Dynamic liquid hold-up of KATAPAK-SP 12 as function of the liquid and the gas load.

at $F$-factors near to the flooding point, an increase of the hold-up was found.

## 5. Residence time distribution

### 5.1. Experimental

Residence time measurements for KATAPAK-SP 12 were carried out in the same apparatus as used for the measurements of the pressure drop and dynamic hold-up. For measuring the residence time distribution, a small amount of saturated aqueous sodium chloride solution was injected as Dirac impulse at the top of the packed bed and the conductivity of the water dropping out of the lowest packing element was measured.

### 5.2. Results

According to Baerns et al. [14], the functions of the residence time distributions $E(\Theta)$ have been calculated. The dependency of the residence time behavior on the dimensionless time $\Theta$ can be seen in Fig. 8. At low liquid loads (~2 m$^3$/m$^2$ h), the residence time distribution is comparatively narrow but shows a pronounced tailing. The tailing is caused by stagnant zones, which occur when the catalytic layers are not totally filled with liquid. At higher liquid loads (~10 m$^3$/m$^2$ h), the residence time distribution is still narrow, but the tailing is strongly reduced, as the catalytic layers are now filled with fluid. At high liquid loads (~20 m$^3$/m$^2$ h), the residence time distribution becomes very broad. This is due to the fact that now excess liquid flows as liquid film at the outer side of the catalytic layers.

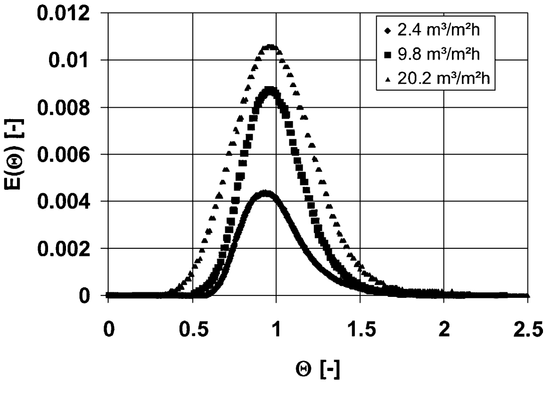

Fig. 8. Residence time distribution for KATAPAK-SP 12 at different liquid loads.

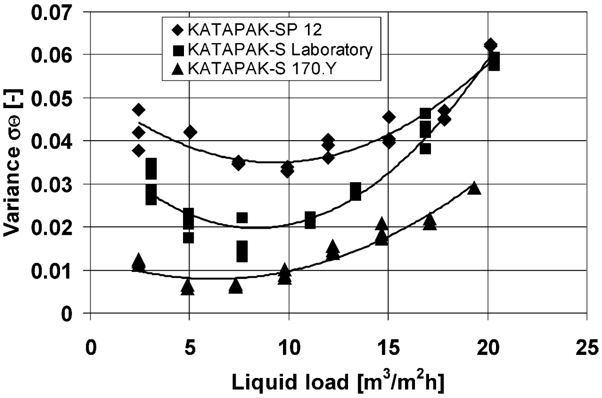

Fig. 9. Dimensionless variance $\sigma_\Theta$ of the residence time distribution of KATAPAK-SP 12, laboratory scale KATAPAK-S and industrial KATAPAK-S 170.Y packings as function of the liquid load (lines smoothed).

The variance $\sigma_\Theta$ of the experimental residence distribution was calculated by the momentum method [14]. In the following, the variance of the measured residence times (for a given liquid load) is used to characterize the spread of the distribution. Fig. 9 shows the results for the variance $\sigma_\Theta$ of the three structured catalytic packing types from experiments carried out at different liquid loads. The course of the variance is comparable for the three structures: Below the load point the variance decreases with increasing liquid load, a minimum is observed around the load point. Above the load point the variance increases with increasing liquid load. Generally, KATAPAK-SP 12 has the highest variance $\sigma_\Theta$, followed by the laboratory scale packing KATAPAK-S, while KATAPAK-S 170.Y has the lowest variance. The reason for the lower variance $\sigma_\Theta$ of the two KATAPAK-S structures is the corrugation of the wire gauze, which effects a quicker exchange of the liquid inside the catalytic layers.

## 6. Simulation results

To illustrate the effect of different catalytic packings on a reactive distillation process, the dehydration

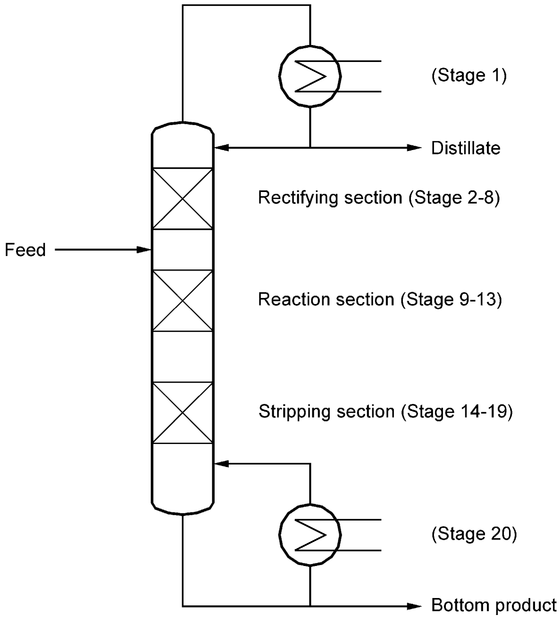

Fig. 10. Configuration of the reactive distillation column used for TBA dehydration.

of tert-butyl alcohol (TBA) was selected as a sample reaction. Information on the chemical equilibrium and the reaction kinetics are available.

The equilibrium constant for the reverse reaction, i.e. the hydration of isobutene, have been measured by Delion et al. [15] and Velo et al. [16]. In a laboratory scale reactive distillation column this reaction has been successfully tested by Abella et al. [17]. In the reaction section Amberlyst 15 in cylindrical pellet form was used for the reaction catalysis and the product separation. In some of the experiments, the maximum TBA feed flow rate for total conversion was measured. Additionally, to the column experiments, the reaction kinetics had been determined in a semi-batch reactor. These reaction kinetics were used in the simulation work for this study.

The equilibrium stage model of the process simulator Pro/II, version 5.10 of Simulation Sciences [18] has been used to simulate the reactive distillation column as shown in Fig. 10. To calculate the vapor–liquid equilibrium, the UNIQUAC model with standard Pro/II parameters was used. Further input data for the simulator are given in Table 3.

Table 3
Input data used for the simulation of the reactive distillation column (dehydration of TBA)

| | |
|---|---|
| Feed temperature (K) | 298 |
| TBA feed flow rate (kmol/h) | 10 |
| Feed stage | 8 |
| Column top pressure (bar(a)) | 1 |
| Number of rectifying stages (−) | 7 |
| Number of reactive stages (−) | 5 |
| Number of stripping stages (−) | 6 |
| Reflux ratio (reflux/distillate) (−) | 2 |
| Isobutene concentration in the distillate stream (wt.%) | 99.9 |

Table 4
Simulation results of the reactive distillation column for the dehydration of TBA

| | KATAPAK-SP 11 | KATAPAK-SP 12 | KATAPAK-S 170.Y |
|---|---|---|---|
| Relative reactive packing volume (−) | 1.00 | 1.32 | 1.36 |

In the simulations, industrial scale KATAPAK-S 170.Y and KATAPAK-SP, types 11 and 12, have been used as structured catalytic packings in the reaction section. During the calculations, the reactive packing volume and thus the catalyst amount was increased until total conversion of TBA was achieved. The results for the relative volumes of the three packings are shown in Table 4. As can be seen in the table, the lowest reactive packing volume, and therefore the economical best solution was obtained in using KATAPAK-SP 11.

## 7. Summary

The performance of structured catalyst packings depends on many parameters, most important are dynamic liquid hold-up, pressure drop, residence time behavior and separation efficiency. In the present work, detailed experimental studies on these influencing parameters of KATAPAK-SP 12, an industrial scale structured catalyst support, were carried out.

The comparison of KATAPAK-SP 12 with results for the structured laboratory packing KATAPAK-S and the industrial packing KATAPAK-S 170.Y showed differences in the separation efficiency course below the load point, a slightly increased variance $\sigma_\Theta$ and a lower hold-up of KATAPAK-SP 12. The pressure drop of KATAPAK-SP 12 and the laboratory scale

KATAPAK-S are very similar, while KATAPAK-S 170.Y has the lowest pressure drop.

The results obtained in the simulation work clarified the advantage of the flexible packing design of KATAPAK-SP: The structure can be optimized to perfectly fit the needs of each specific reaction, resulting in reduced catalytic packing volume.

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