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VAPOUR–LIQUID MASS TRANSFER PERFORMANCE OF MODULAR CATALYTIC STRUCTURED PACKING

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The mass transfer performance the Katapak®-SP has been evaluated experimentally and the Delft MCSP model, a parallel channel model is developed to predict the HETP as function of F-factor. Pilot scale (i.d. 450 mm) total reflux distillation experiments were done with an aqueous system Methanol/Water and organic system Cyclohexane/n-Heptane. With the latter, two types of Katapak-SP were tested. As reference a high capacity packing (*HCP*) is used. In the pre-loading region of operation with the MCSP-12 packing with the aqueous system shows nearly a constant HETP of 0.27 m, where with the organic system the experimental results in this region show higher HETP values (HETP $\sim$ 0.35 m). With the MCSP-11 and the organic system even higher values are obtained (HETP $\sim$ 0.5 m). The difference in this performance is mainly caused by maldistribution of the flowing liquid, namely bypassing of the vapour when the liquid flows through the catalyst filled pockets and reduced lateral spreading due to these pockets, especially in the MCSP-11.

The best performance is reached at loading in the packed beds and the model is well capable to predict the mass transfer performance.

KEYWORDS: modular catalytic structured packing, katapak, high capacity packing, mass transfer, catalytic distillation

INTRODUCTION

Catalytic distillation combines two processes, i.e. reaction with the aid of heterogeneous catalysis and thermal separation, in one unit operation. The majority of published literature focuses on the development of equilibrium and rigorous non-equilibrium models to simulate the process [1]. However, little attention has been paid to hardware design and performance. For an accurate catalytic distillation column design the hydrodynamic and mass transfer performance imposed by the internal is essential information [2]. Modular Catalytic Structured Packings (*MCSP*) combines reaction and distillation and allows the adjustment of the catalyst volume and separation performance depending on process needs [3].

Limited data on the separation performance of catalytic packings is contained in literature. Information on the performance of three basic, but geometrically different catalytic structured packing can be found, including the Bale packing [4,5], the Katapak®-S [3,6,7] and the modular catalytic structured packing like Multipak® [8,9] and Katapak®-SP [3,10].

MCSP GEOMETRY

Figure 1 shows the two configurations of MCSP evaluated in this study. In the MCSP corrugated sheets are sandwiched between catalyst containing pockets, the reaction section of the packing with two pockets per layer height. Due to the dense structure of these pockets gas cannot flow through this section. The space wherein the corrugated sheets are in the packing is the distillation section. The different MCSP types used differ mainly in the number of sheets fitted between the catalyst filled pockets.

The corrugated sheets in this MCSP are of the MellapakPlus type, which is an established high capacity packing with corrugated sheets bended to the vertical at both ends of the flow channel. This geometry is beneficial and provides a smooth transition of the phases between packing layers leading to delayed loading and flooding effects in the packed bed. In the MCSP-11 only one sheet alters with the catalyst filled pockets. Therefore in the distillation section only closed channels are present. In the MCSP-12 two sheets are placed between the catalyst filled pockets. Here in the middle of the distillation section the common crossing flow configuration is encountered as in a corrugated sheet structured packing, i.e. like in the corresponding high capacity packing (*HCP*). These channels are the open channels in this section of the MCSP. The HCP is used as reference packing in this study. The geometric data of the MCSP and HCP are given in Table 1.

DELFT MCSP MODEL

Based on the geometry of MCSP a parallel channel approach was adopted to predict the fluid dynamics and mass transfer behaviour of these packing types. As basis the Delft model, developed to predict the performance of corrugated sheet structured packing without using any empirical, packing type or size specific constants [11–13], is taken. The developed Delft MCSP Model makes a distinction between the three channel types
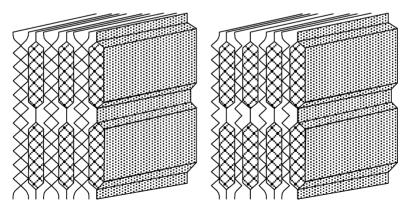

Figure 1. Schematic of MCSP, left: MCSP-12; right MCSP-11

Table 1. Geometrical packing characteristics MCSP and HCP

Property	Symbol	MCSP 12	MCSP 11	HCP
Cross-sectional fraction	Γ	0.52	0.40	1
Open channel ratio	$\times$	0.5	0	1
Void fraction	ε_p	0.7	0.55	0.975
Specific area open channels $[m^2/m^3]$	$a_{p,oc}$	132.5	0	509.5
Specific area closed channels $[m^2/m^3]$	$a_{p,cc}$	208.8	300.2	0
Corrugation angle	α_c	$41°$		
Packing element height	h_{pe}	200 mm		

that are identified; the catalyst filled pockets in the reaction section and in the distillation section, the open and closed channels.

As shown in the model framework of the Delft MCSP Model, Figure 2, the linking parameter is the liquid hold-up. Two contributions determine the total hold-up, i.e. internal and external hold-up. The internal hold-up is the amount of liquid inside the catalyst filled pockets. The maximum internal dynamic liquid hold-up described in [14] is reached at the catalytic load point, which is the point where the pockets are just full with flowing liquid. At liquid loads above the catalytic load point the excess liquid contributes to the external hold-up, which is the liquid descending in the distillation section. The excess liquid is
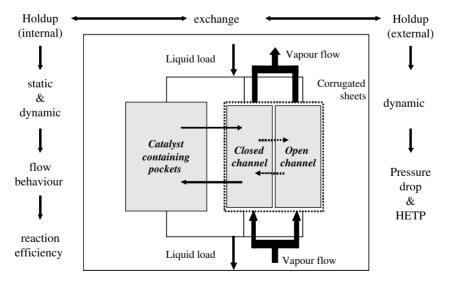

Figure 2. Delft MCSP Model framework

assumed to be evenly distributed over the channels in the distillation section of the packing.

The open channels in the distillation section, which are the channels in the middle of this section of the packing, exhibits the normal structure encountered in corrugated sheet structured HCP. Therefore these channels will be treated equally as in the reference HCP.

The closed channels are the channels next to the pockets. Both liquid and vapour are forced to follow the channel to the end and where liquid tends to flow in the form of rivulets along the lowest point of the channel. In this way a limited interface is available for contact with the ascending vapour. With respect to the geometry in case of the MCSP-11 only closed channels are present, while the MCSP-12 type has both closed and open channels.

The Delft MCSP model is a mechanistic channel model. To make a distinction between the two configurations, two relevant geometric based ratios are defined, the cross sectional ratio and the open channel ratio, respectively:

$$\Gamma = \frac{\text{cross sectional area distillation section}}{\text{total cross sectional area}} \qquad (1)$$

and

$$X = \frac{\text{number of open channels in the distillation section of MCSP}}{\text{total number of channels in the distillation section}} \qquad (2)$$

It is assumed that the flowing phases are able to fully mix at the transition between packing layers especially at higher F-factors. This consequently means that between packing layers the concentrations in both phases are uniform over the column cross section.

Packed columns are continuous differential contacting devices however the HETP is preferred to characterise the mass transfer efficiency. The HETP is related to the HTU's, i.e. the phase velocities and volumetric mass transfer coefficients, via the stripping factor, λ, which is the ratio of slopes between the equilibrium and working lines. The change in concentration is calculated based on the height of transfer units, $HTU_{L,cc}$, $HTU_{G,cc}$, $HTU_{L,oc}$ and $HTU_{G,oc}$, the corresponding HETP's and using the Fenske equation where the mole fraction at the end of the channel is written explicitly, all based on the geometry of respectively closed and open channels. At the transition between layers the average concentration is calculated by mixing of the flows according the flow contribution in each channel, i.e. taking the cross sectional and channel ratio into account. In catalyst filled pockets liquid must flow out of the upper pockets before it can enter the lower pockets and in between it mixes with the liquid flowing in the closed channel. This is taken into account in the model by intermediate mixing of liquid coming from both channels before it is redistributed.

Based on the average concentration entering and leaving a packing layer the average number of equilibrium stages per packing layer height, $N_{min,i}$, is determined with the Fenske equation. The relative volatility over a packing layer height is taken constant.

With this relative volatility also the stripping factor is calculated. This is repeated for the number of MCSP layers simulated in the column. The HETP is determined with the height of the packed bed divided by the total number of stages in the column, which is the sum of the number of stages in each packing layer.

MASS TRANSFER COEFFICIENTS

Mass transfer results from the vapour-liquid interaction at the interface, i.e. film surface, along relatively short flow channels and accompanied with considerable entrance effects. To account properly for this the vapour phase mass transfer coefficient incorporates the effective relative velocity based Reynolds number, the vapour phase Schmidt number, the vapour-liquid friction factor and the characteristic diameter to length ratio.

In the vapour phase to account for the flow regime transition, the vapour phase mass transfer coefficient, k_G, has a laminar and turbulent contribution:

$$k_{G,0c} = \sqrt{k_{G,lam,0c}^2 + k_{G,turb,0c}^2} \tag{3}$$

The liquid phase mass transfer coefficient is based on the penetration theory of Higbie where the contact, i.e. exposure time in the laminar film is defined with the characteristic length and effective liquid velocity:

$$k_{L,0c} = 2\sqrt{\frac{D_L u_{Le,0c}}{0.9\pi d_{bG}}} \tag{4}$$

with D_L the liquid side diffusion coefficient.

EFFECTIVE INTERFACIAL AREA

The effective interfacial area is determined with a, to structured packing modified Onda relation [15].

The closed channels are the flow channels next to the catalyst filled pockets. Of course similar relations for mass transfer apply as developed for the open channels, however here as function of the closed channel geometry. Since two pockets above one another are present in the reactive section of the MCSP all liquid flowing out of the upper pocket is able to mix with the liquid flowing in this channel which is accounted for in the modelling.

EXPERIMENTAL STUDIES

The mass transfer performance was experimentally determined using a pilot scale, 450 mm internal diameter, total reflux distillation column. The setup is described in detail in [17] and a simple flow diagram is shown in Figure 3. The total reflux distillation experiments were done with two systems, Methanol/Water (MeOH/Wa) and Cyclohexane/n-Heptane (CH/nH).
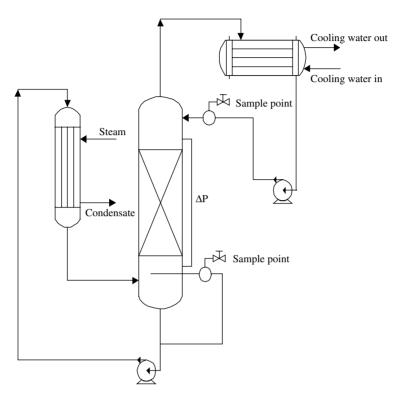
Figure 3. Process flow diagram total reflux distillation column

Also the pressure drop over the column was determined as function of gas load with two calibrated pressure difference meters, a low range one from 0 to 10 mbar and one full range for 0 to 100 mbar. The characteristic average physical properties of the two test systems are summarised in Table 2.

RESULTS AND DISCUSSION

The HETP's of the MCSP's are higher compared to the HCP and a comprehensive discussion of the latter can be found in [17]. The Delft MCSP model is used specifically to predict the performance of the MCSP. The system physical properties taken for the model calculations are summarised in Table 2. In Figure 4 the results of the MCSP model is compared with the MCSP-12 with the system MeOH/Wa. Compared to the HCP the MCSP has less installed specific geometric area and due to the MCSP geometry liquid bypasses the vapour while flowing through the catalyst filled pockets, which additionally lowers the mass transfer and this is incorporated in the model. Up to flooding the model is well capable to predict the efficiency.

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Table 2. System properties total reflux Delft parallel column model

Property	Symbol	Value	
		MeOH/Wa	CH/nH
Molecular weight [kg/kmol]	M_w	25	87
Liquid density [kg/m^3]	ρ_L	854	658
Dynamic liquid viscosity [Pa·s]	μ_L	$3.17\ 10^{-4}$	$2.87\ 10^{-4}$
Liquid diffusivity [m^2/s]	D_L	$7.25\ 10^{-9}$	$4.35\ 10^{-9}$
Surface tension [N/m]	σ_L	0.0389	0.0154
Vapour density [kg/m^3]	ρ_G	1.15	3.00
Dynamic vapour viscosity [Pa·s]	μ_G	$1.21\ 10^{-5}$	$8.09\ 10^{-6}$
Vapour diffusivity [m^2/s]	D_G	$3.70\ 10^{-5}$	$4.53\ 10^{-6}$
Liquid load at F-factor 2 [m^3/(m^{2}h)]	μ_{Ls}	9.04	18.94

The model calculations with the organic CH/nH system are compared in Figure 5 and 6 for respectively the MCSP-11 and MCSP-12. Since the model assumes uniform distribution of the phases over the column cross section at each column load the model prediction is therefore too optimistic in the pre-loading region. Regarding if the outside surface area of the pockets in the closed channels participate in V-L mass transfer due
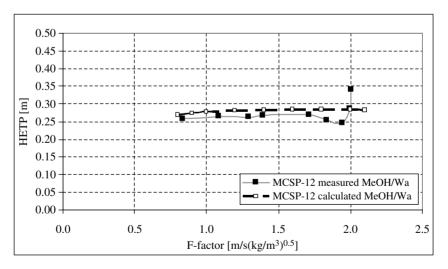

Figure 4. Model prediction and experimental results of the HETP as function of F-factor of the MCSP-12 with the system: MeOH/Water at atmospheric pressure
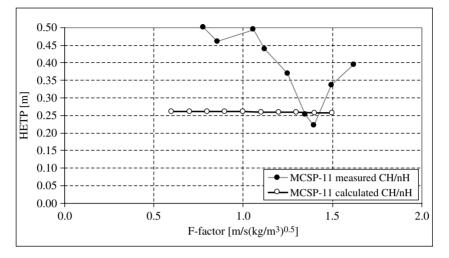
Figure 5. Model prediction and experimental results of the HETP as function of F-factor of the MCSP-11 with the system: CH/nH at atmospheric pressure
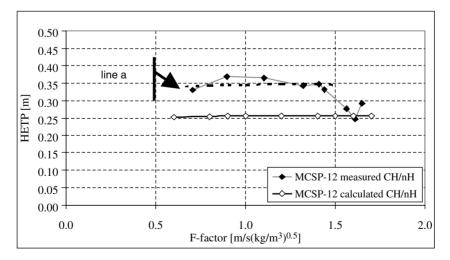

Figure 6. Model prediction and experimental results of the HETP as function of F-factor of the MCSP-12 with the system: CH/nH at atmospheric pressure

to the lower liquid surface tension, line 'a' in Figure 6 is the result of the model calculations of the MCSP-12 with the organic system where this outside area of the pockets is omitted as installed geometric area and therefore not contributing to the calculated effective interfacial area. As can be seen the model and experimental results are about on the same plateau in the pre-loading region indicating that this area is not optimally used in the MCSP.

The Parallel channel model predicts an equal HETP for both MCSP types. Because in the description of the vapour phase mass transfer coefficient is assumed that the complete channel is occupied by liquid this leads to a lower mass transfer resistance in this channel and in the model the closed channel geometry is more beneficial for mass transfer and compensates for the less installed geometric area. Around loading the model agrees with the experiments.

The most efficient mass transfer is reached at hydraulic loading where all model results agree with experiments.

The experimental results obtained in this study are comparable with data published in previous studies. For the MCSP-11 a HETP around 0.5 m at low column loads is found and this is also reported in [10] with an aqueous system. The performance with the organic system of the MCSP-12 with the reported data in [3] shows that the performance in this study with CH/nH are comparable. The same trend is observed while the column load approaches hydraulic loading of the bed where the separation efficiency increases rapidly.

CONCLUSIONS
The Delft MCSP Model was developed to predict the mass transfer efficiency of Modular Catalytic Structured Packings. The model performance was validated with experiments. These are done with two different systems and two types of MCSP.

For the MCSP-12 the HETP with the aqueous system is constant up to flooding. Here the model indicates that at the outside surface of the pockets a liquid film is developed due to high capillary forces which promote liquid spreading over the wire gauze metal surface. With the organic system higher HETP's are experimentally obtained and these values agree with model calculations where the wire gauze surface (outside surface of the pockets) is omitted to contribute to the effective interfacial area prediction. The organic system has a lower liquid surface tension and the development of a liquid film over the wire gauze surface is less promoted.

Especially with the MCSP-11 packed bed the poor performance in the preloading region is caused by liquid maldistribution due to the poor wetting of some parts of the closed channels. The assumption of uniform liquid distribution in the pre-loading region and consequently the interfacial area generation with the organic system gives too optimistic model predictions.

The Delft MCSP model is well capable to accurately predict the mass transfer performance of MCSP up to the point of onset of flooding.

ACKNOWLEDGEMENTS

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