DIFFUSIONAL AND HYDRAULIC CHARACTERISTICS OF KATAPAK-S

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ABSTRACT

The paper presents selected results of the EU-project „Intelligent column internals for reactive separations (INTINT)".

Catalytic distillation still expands its field of applications. New structured catalytic column internals have been developed in recent years. One of them, Sulzer KATAPAK-S, is a subject of the investigations presented in this paper. Important parameters of the internals have been examined experimentally in a 250mm ID laboratory column: pressure drop for dry and irrigated packing, dynamic liquid hold-up as well as mass transfer coefficients. The experimental results collected have been used to derive a set of correlation equations describing the above quantities in terms of dimensionless flow parameters. The correlations obtained are used to build a unique software package within the INTINT project for the simulation of catalytic separation processes and the behaviour of catalytic column internals.

STATE OF THE ART

Even if the concept is not new and the first application dates back to the 1920's [1,2,3,4,5], reactive distillation has become very popular in recent years. Advantages of this technology are obvious: environmental protection, reduction of investment costs, lower energy consumption. Extensive research has been carried out to fix criteria of applicability to new processes [6,7,8,9] as well as to model specific reactive distillation processes to get information about the process behaviour itself [10,11]. Models of different complexity can be found in literature to describe reactive distillation processes. The implementation of heterogeneous catalysis in reactive distillation columns led to further process improvements but also to a far greater complexity due to the existence of a third solid phase [12,13]. Different types of catalytic packing have been invented to solve the problem of countercurrent gas-liquid flow with simultaneous heterogeneous catalysis and investigations have been focussed on the flow regime within these column internals [14,15,16,17,18,19]. Mass transfer characteristics have been found to be of significant importance for the process behaviour, while the pressure drop provides information about the packing hydrodynamics [20].

In February 2000 an R&D project started entitled „Intelligent column internals for reactive separations" (INTINT) [21]. The project proposes a new way for the design of reactive distillation processes which permits a direct involvement of an end-user into the internals development. The new methodology will be based upon simultaneous use of CFD and rate-based process modelling and must be able to suggest an optimal process design including specially developed tailor-made column internals (Kenig et al., [22]). The sophisticated software developed to perform virtual experiments requires validation with a reliable set of experimental results. Some catalytic column internals have been selected and a comprehensive experimental programme has been realized including – among others – the determination of mass transfer coefficients, dynamic liquid hold-up and pressure drop. In such a way an extensive and reliable experimental database has been established for the INTINT project. One of the selected internals was KATAPAK-S.

REACTIVE PACKING INVESTIGATED

Catalytic packing KATAPAK-S-250.Y manufactured by Sulzer Chemtech, Switzerland (Fig. 1) was a subject of the present study. The packing has a sandwich structure of corrugated wire gauze sheets, interconnected to form channels filled with pellets of solid catalyst (1mm glass beads in this study). The packing characteristics are: specific surface area $a=128.2 m^2/m^3$, void fraction $\varepsilon=0.622 m^3/m^3$. The packing, manufactured as segments 200mm high, has been fitted into a test column 250mm ID. Five segments were used during the hydraulic tests (total height 1m), and 2 or 3 segments (total height 0.4 or 0.6 m, respectively) were used in the mass transfer study. The successive segments were rotated relative to one another at an angle of 90 degrees.

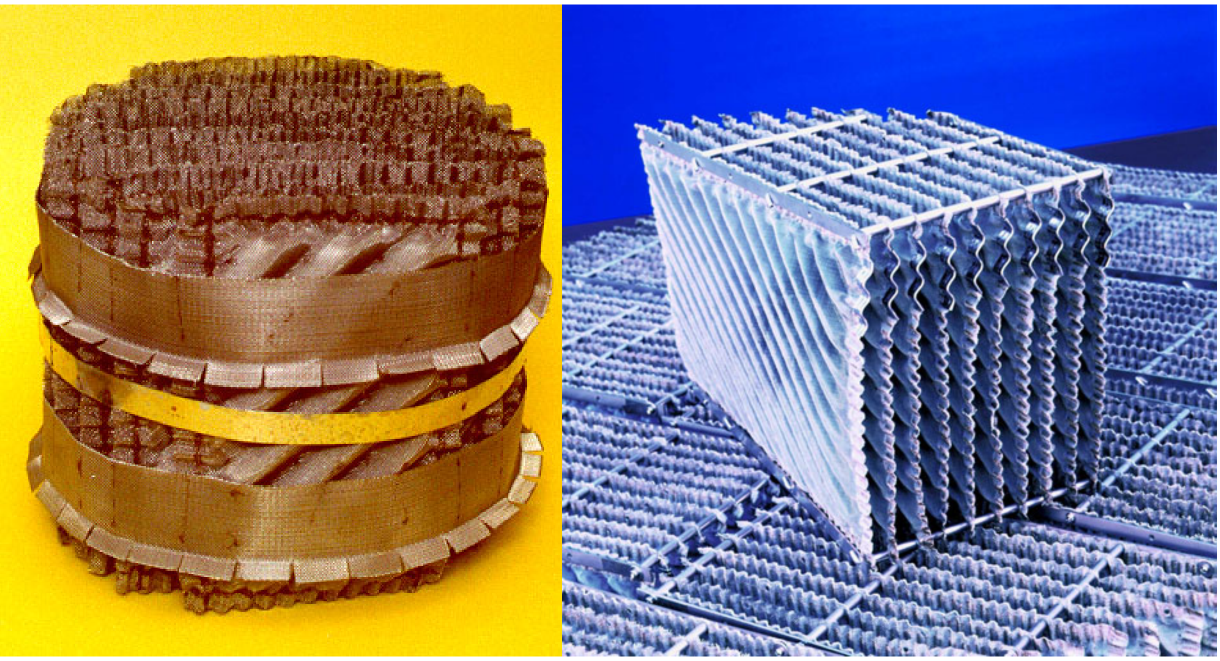

Fig. 1. The column packing KATAPAK-S-250-Y (Sulzer): laboratory scale - single segment (left) and industrial scale (right).

EXPERIMENTAL PROCEDURE

The studies were performed in a column 250mm in diameter. The air-water system was used. The studies were carried out at room temperatures and under normal pressure. The experimental set-up is presented in Fig.2 for the hydraulic and liquid-phase mass transfer investigations, and in Fig. 3 for the gas-phase mass transfer study. The liquid flowing from the tank through a pump and flowmeters was supplied into the column via the distributor. After passing through the packing layer, it was

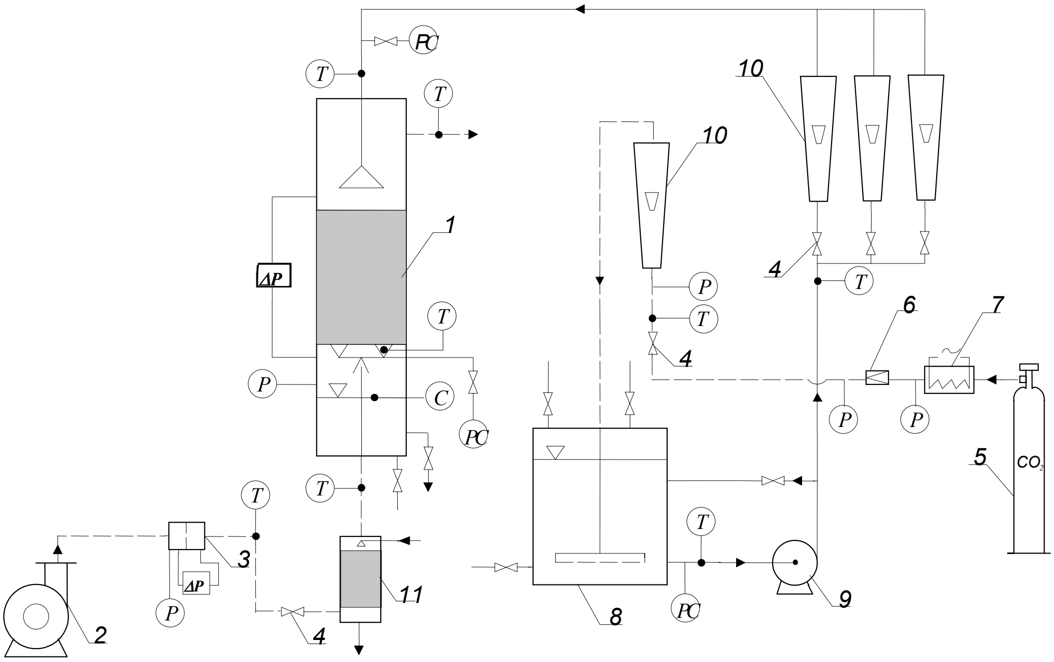

Fig. 2. Experimental setup – hydraulics and liquid phase mass transfer study. 1-packed column, 2-blower, 3-orifice flowmeter, 4-control valves, 5-CO_2 cylinder, 6-reducing valve, 7-CO_2 heater, 8-liquid tank, 9-pump, 10-flowmeters, 11-gas phase saturator

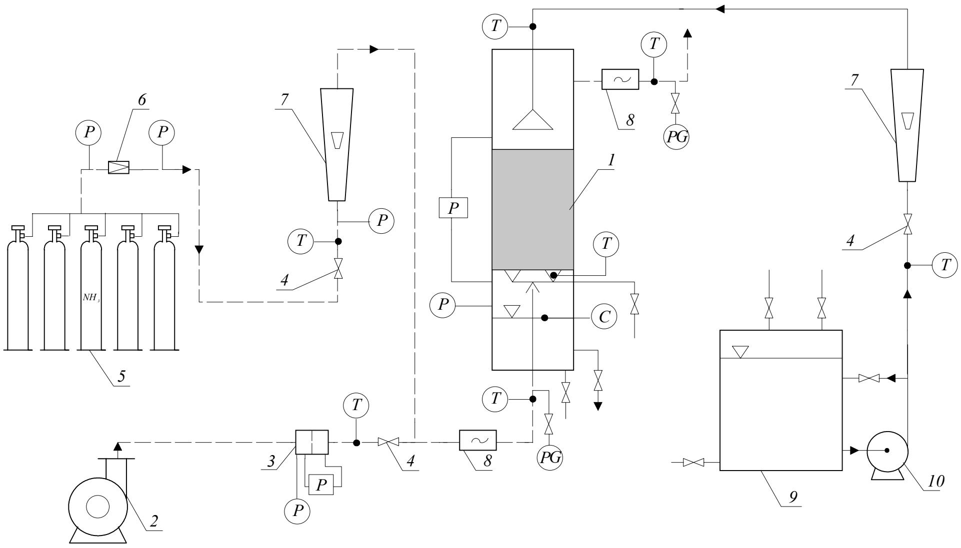

Fig. 3. Experimental setup – mass transfer coefficients in the gas phase. 1-packed column, 2-blower, 3-orifice flowmeter, 4-control valves, 5-ammonia cylinders, 6-reducing valve, 7-flowmeters, 8-static mixer, 9-liquid tank, 10-pump.

collected in the bottom of the column and withdrawn. The air stream entered the bottom of the column from a blower through an orifice flowmeter**.** Due to very low liquid flow rates in some experiments it was necessary to saturate the gas phase with water vapour using a saturator. After passing through the packing the gas was removed from the column top. Due to a very wide range of liquid flow rates during the experiments, two different capillary liquid distributors were used with various capillary diameters; both had 1,366 holes per square metre.

The study on the hydraulics of the reactive packing included the pressure drop over the dry and wetted (irrigated) packing as well as dynamic (free-draining) liquid hold-up. The pressure drop was measured using simultaneously a Recknagel micromanometer filled with ethanol (ratio from 1/2 to 1/25) and a membrane electronic manometer. For high and unstable pressures, a U-tube filled with water was used. The minimal resolution of the measurement devices was of the order of 0.2 Pa.

The liquid hold-up was measured using the volumetric method. The valves placed before and after the test column were quickly and simultaneously closed, the liquid held up on the packing drained down and its volume measured. The time of draining was 45 minutes. The hold-up was calculated as the volume of liquid divided by the volume of the column (the part filled with the packing).

During the liquid-phase mass transfer experiments the liquid in the tanks was saturated with CO_2 flowing from gas cylinders via flowmeters and bubblers, as shown in Fig.2. The desorption of CO_2 from water to the air stream occurred in the column. The concentrations of carbon dioxide in the liquid below and above the packing layer were determined by titration. For the gas-phase mass transfer experiments (Fig. 3), ammonia was supplied from cylinders through a calibrated flowmeter into the air stream entering the column. Absorption of ammonia took place inside the packing. The inlet concentration in the gas phase was determined based on the flow rates of the air and ammonia streams, while the outlet concentration – by titration, after the absorption of ammonia by bubbling the outlet gas through the acid solution.

Basic Equations: Hydraulics

The pressure drop for the dry packing can be calculated from the Darcy-Weisbach equation

$$\frac{\Delta P_{dry}}{H} = \Psi \frac{\rho_g w_g^2}{2 d_{eK}} \tag{1}$$

The real (interstitial) velocity of the gas inside the packing is related to its superficial velocity by

$$w_g = \frac{w_{0g}}{\varepsilon} \tag{2}$$

According to Maćkowiak [23]:

$$d_{eK} = \frac{4\varepsilon K}{a}, \quad K = \frac{1}{1 + 4/(aD)} \tag{3}$$

and, consequently

$$\frac{\Delta P_{dry}}{H} = \Psi \frac{a \rho_g w_{0g}^2}{8 \varepsilon^3 K} \tag{4}$$

The friction factor Ψ was assumed to be a power function of the gas-phase Reynolds number Re_{gK} which, based on the flow model adopted [24], is defined by

$$Re_{gK} = \frac{w_g \rho_g d_{eK}}{\eta_g} = \frac{4 g_{0g} K}{a \eta_g} \tag{5}$$

Basic Equations: Mass Transfer

For the liquid-phase mass transfer study the system carbon dioxide-air-water was employed. This system enabled the direct determination of the liquid-phase mass transfer coefficient as, due to the high value of the equilibrium constant (Henry's law constant), the mass transfer resistance in the gas phase could be neglected. The liquid-side mass transfer coefficient was expressed as

$$\beta_{AL} = \frac{G_{iL}(X_{A1} - X_{A2})}{A\, \Delta\pi_{Am}} \tag{6}$$

and the driving forces in two cross sections were calculated as (Hobler [25]):

$$\Delta\pi_A = \frac{(X_A - X_{A\,int})}{(1 + X_A)_m} \approx X_A - X_{A\,int} \quad (X_A << 1) \tag{7}$$

using Henry's law to evaluate the concentrations at the gas-liquid interface:

$$X_{A\,int} \approx x_{A\,int} = \frac{p_{A\,int}}{H_P} \tag{8}$$

The "active" mass transfer surface area A was assumed to be identical with the packing geometrical surface. This means that the mass transfer coefficient measured was in fact multiplied by the wetting factor of the packing surface. Such an approach seems to be more accurate than that applied in a number of former studies. There, the measured mass transfer coefficient (e.g. in the gas phase) was assumed to be dependent only on the flowrate of the phase in question. The influence of the flowrate of the other phase was described by the wetting efficiency factor (the comprehensive discussion of this problem is given by Hobler [25]). The approach presented here is strongly recommended by Burghardt and Bartelmus [26] to avoid the use of highly uncertain and often unavailable (especially for modern internals) wetting efficiency factors. However, this approach requires more experiments to cover the assumed range of both gas and liquid flowrates.

The results obtained are correlated as [26]

$$Sh_L = a\, Re_L^b Re_g^c Sc_L^{0.5}$$

(9)

The exponent at the Schmidt number was taken from literature by analogy to other investigations cited (cf. Hobler [26]) due to a small range of this number in the present study.

During the gas-phase mass transfer experiments the system ammonia-air-water was used. The overall mass transfer coefficient can be derived from the balance of the diffusing species

$$k_A = \frac{G_{ig}(Y_{A2} - Y_{A1})}{A\, \Delta\pi_{Am}} \quad \text{where} \quad \Delta\pi_A = \ln \frac{1 + Y_A}{1 + Y_A^*} \tag{10}$$

The mass transfer surface area is equal to the geometrical surface area of the packing, just as it is for the liquid phase. The gas-phase mass transfer coefficient can then be expressed as

$$\beta_{Ag} = \frac{1}{\frac{1}{k'_A} - \frac{n}{\beta'_{AL}}} \qquad (11)$$

For the absorption of ammonia in water, the liquid-phase mass transfer resistance is usually by one order of magnitude lower compared with that in the gas phase; nevertheless, the liquid-phase mass transfer coefficients were calculated from the correlation derived earlier (based on the liquid-phase experiments) and substituted into eq. (11). The results are correlated as

$$Sh_g = a\,Re_g^b\,Re_L^c Sc_g^{0.33} \qquad (12)$$

The mass transfer coefficients were determined based on the geometrical surface area of the packing. Therefore, the gas- and liquid-phase Reynolds numbers which appear in equations (9) and (12) reflect changes in the individual gas- and liquid-phase mass transfer coefficients, respectively, as well as describe the influence of the flowrates of the two phases on their interfacial surface contact area.

EXPERIMENTAL RESULTS

Pressure Drop for Dry and Irrigated Packings

The experimental data obtained for the single-phase flow are presented in Fig.4; they were correlated as follows using the Ψ parameter:

$$\begin{aligned} \Psi &= 6.275\,Re_{gK}^{-0.293} \quad \text{for } 550 < Re_{gK} < 1500 \\ \Psi &= 2.561\,Re_{gK}^{-0.171} \quad \text{for } 1500 \le Re_{gK} < 6000 \end{aligned} \qquad (13)$$

The scatter of the experimental points around the correlation does not exceed 2%.

The pressure drops for the irrigated packing are presented in Fig.5 as the specific pressure drop ΔP/H vs. gas F-factor for different liquid loads W_{0L} (the results for the dry regime are also included in this figure). The experiments cover the following ranges of the gas and liquid flowrates:

$F=0.32$ to $3.25\ Pa^{0.5}$ or $Re_{gK}=630$ to 6250;
$W_{0L}=11$ to $90\ m^3/(m^2h)$ or $Re_L=75$ to 630

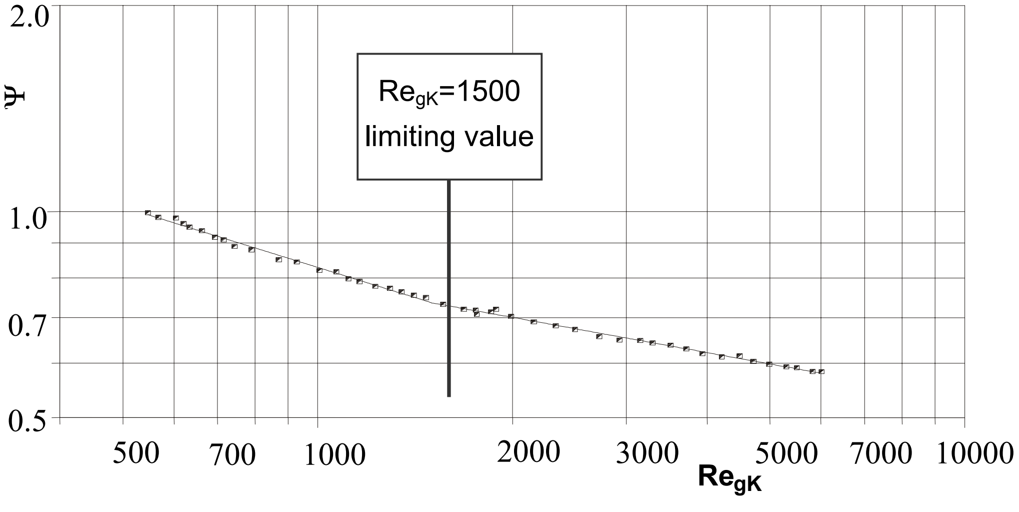

Fig. 4. Experimental results for the pressure drop over dry packing: friction factor Ψ vs. gas-phase Reynolds number.

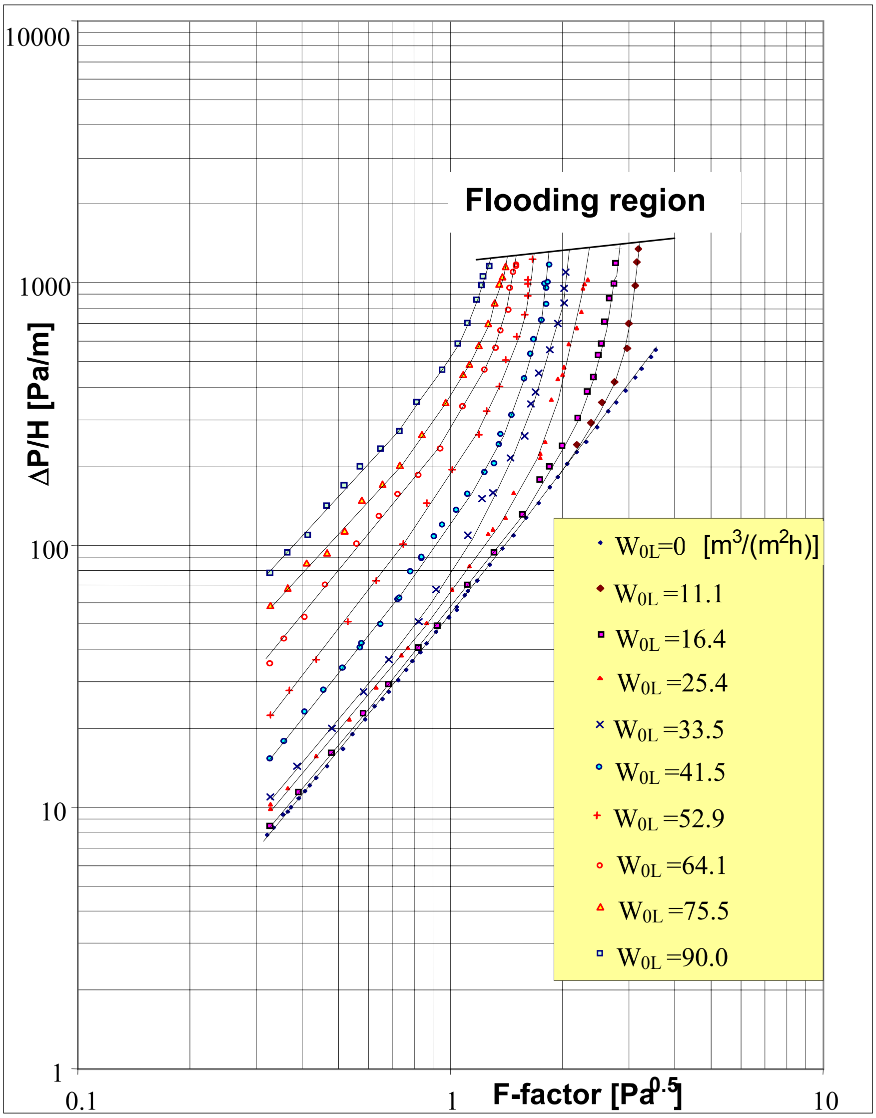

Fig. 5. Pressure drop for irrigated packing vs. gas F-factor for different liquid loads.

Liquid Dynamic Hold-up

The experimental data together with the correlation line are presented in Figure 6 as the liquid dynamic hold-up vs. liquid load. The liquid hold-up was correlated as a power function of the liquid Reynolds number:

$$h_d = 0.0273\,Re_L^{0.331} \tag{14}$$

The influence of the gas-phase load was negligible. The correlation covers the range typical for reactive distillation: Re_L=7.3 to 530; Re_g=620 to 5900. The scatter of the experimental points around the correlation is 15%.

Mass Transfer Coefficients

Representative results for the liquid-phase mass transfer are presented in Figure 7. The results obtained are correlated as

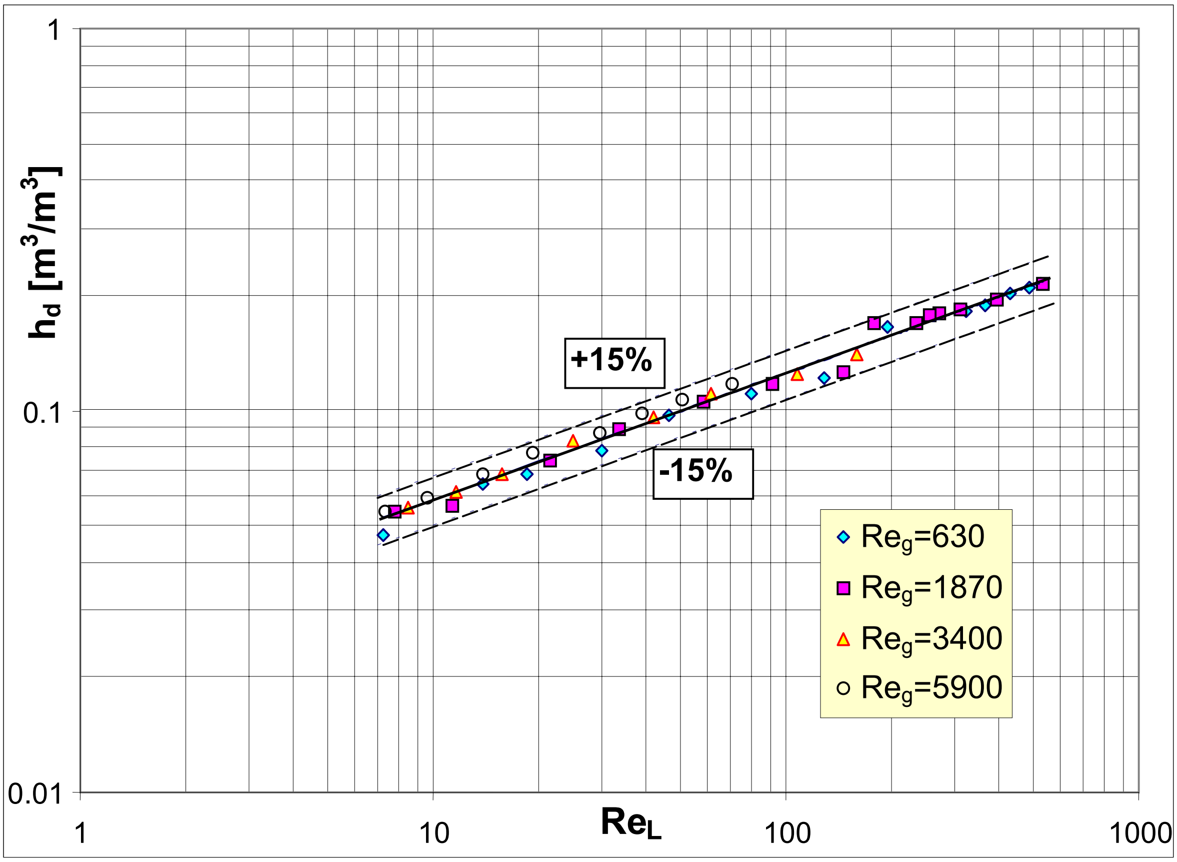

Fig. 6. Liquid dynamic hold-up vs. liquid Reynolds number. The solid line represents correlation (14).

$$\begin{aligned} Sh_L &= 3.777 \cdot 10^{-3} \, Re_L^{0.671} \, Sc_L^{0.5} \\ &\qquad \text{for } 630 < Re_g \le 1700 \\ Sh_L &= 1.235 \cdot 10^{-4} \, Re_L^{0.663} \, Re_g^{0.452} \, Sc_L^{0.5} \\ &\qquad \text{for } 1700 < Re_g \le 5900 \end{aligned} \qquad (15)$$

Figure 7 corresponds to the lower range of the gas-phase Reynolds numbers. The scatter of the experimental points around the correlation line does not exceed 15%. The exponent at the Schmidt number was taken from literature [26] by analogy to other studies, due to only a small range of this number in the present study. The correlation (15) is valid for the range of Re_L=13 to 320.

The results for the gas phase are presented in Figure 8 and are correlated as

$$Sh_g = 0.0476 Re_g^{0.736} \, Re_L^{0.229} \, Sc_g^{0.33} \qquad (16)$$

The scatter of the experimental points around the correlation line does not exceed 15%. The correlation (16) is valid for the following ranges: Re_L=15.2 to 360, Re_g=610 to 5920.

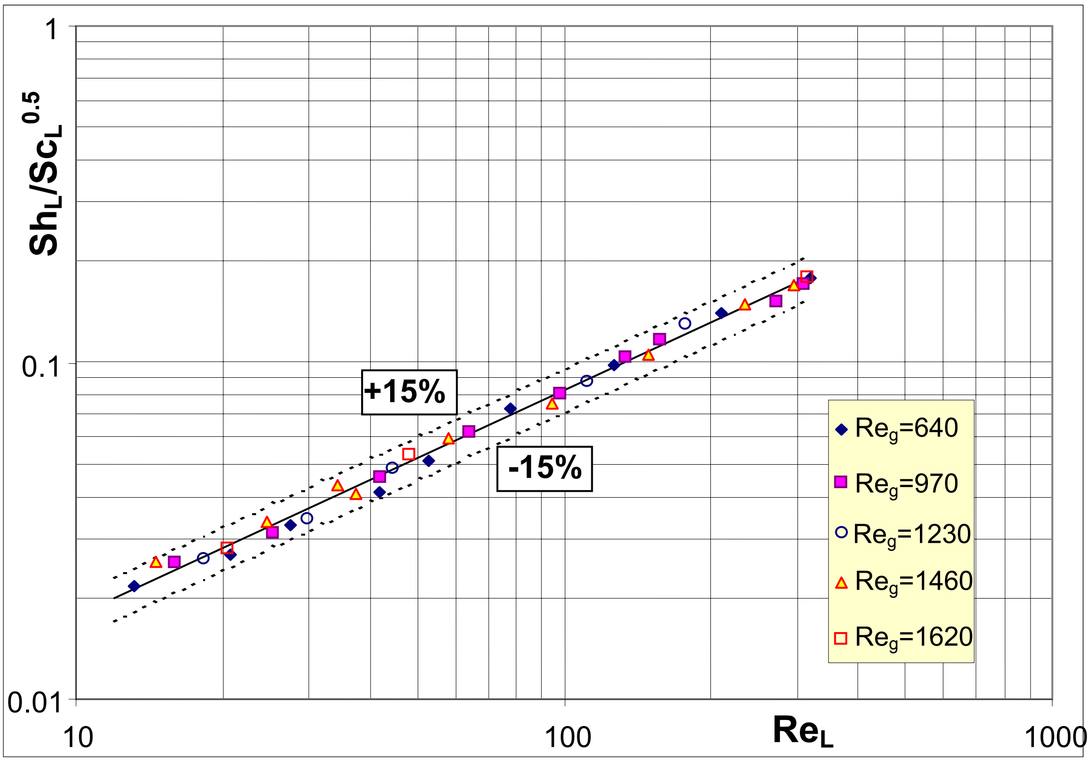

Fig. 7. Mass transfer coefficient for the liquid phase vs. liquid-phase Reynolds number. Lower range of gas-phase Reynolds numbers.

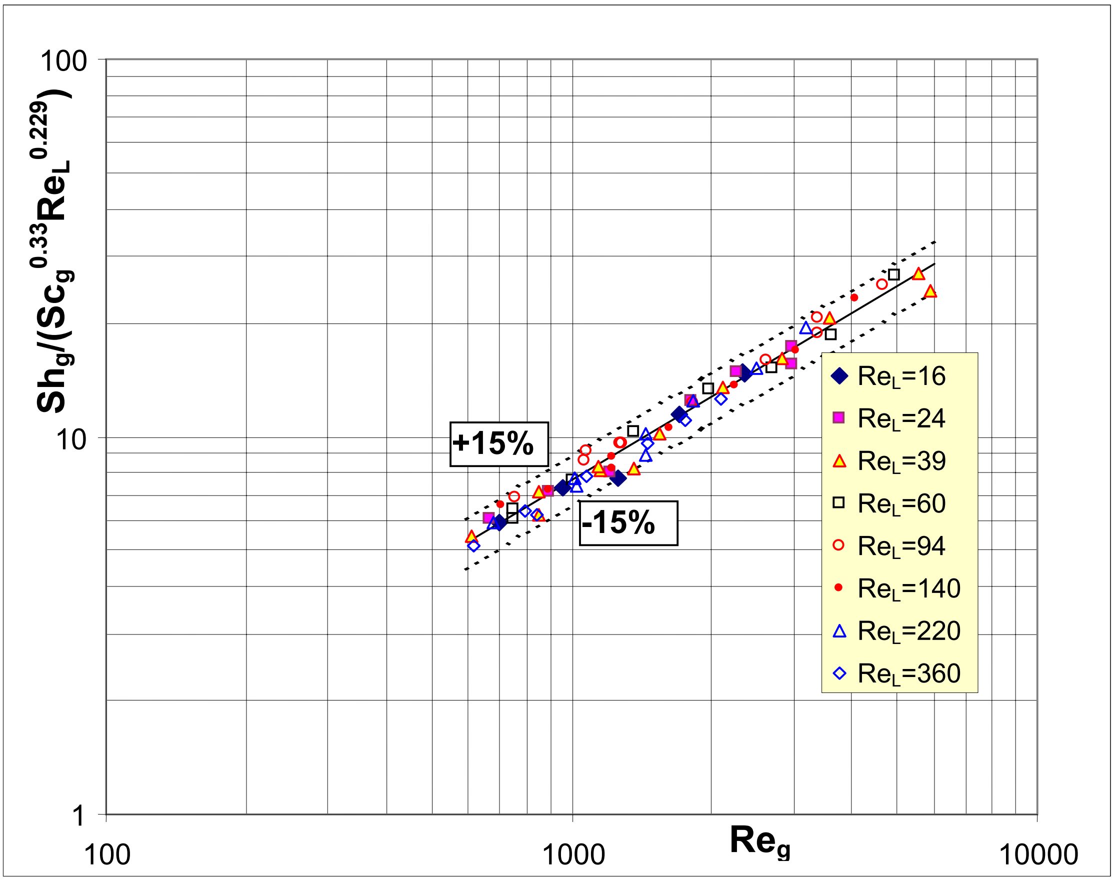

Fig.8. Mass transfer coefficient for the gas phase vs. gas-phase Reynolds number.

FINAL REMARKS.

The results of an extensive study concerning the structured catalytic packing KATAPAK-S are presented. The main hydraulic characteristics of the packing, namely the pressure drop for dry and wetted packings as well as the liquid dynamic holdup have been experimentally studied. The mass transfer coefficients in the gas and liquid phases have been determined for the whole operating range of the packing. The correlations have been derived in terms of dimensionless numbers. Several hundreds of hydraulic and mass transfer experiments have been performed to form a broad and comprehensive database, accompanied by a set of reliable correlation equations.

The significance of the derived hydraulic and mass transport characteristics is twofold: first, the data obtained are necessary to validate the new CFD software, and second, to support the reactive distillation process design. The software to be developed requires a mathematical function that describes hydraulic and mass transfer properties of the column internals used. Therefore, appropriate correlation equations have been derived for the column internals employed during the reactive experiments.

More information on the experimental programme is available at the INTINT web-page [21].

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SYMBOLS

A – surface area of mass transfer, m^2;
a – specific surface area, m^2/m^3;
D – column diameter, m;
$d_e=4\varepsilon/a$ - hydraulic packing diameter (used for mass transfer study), m;
$d_{eK}=4\varepsilon K/a$ - hydraulic packing diameter (used for hydraulics), m;
$F = w_{0g}\rho_g^{0.5}$– F-factor, $Pa^{0.5}$;
G - flow rate, kmol/s;
g_0- mass superficial velocity, $kg/(m^2s)$;
g – acceleration of gravity, m/s^2;
H –column height, m;
H_P- Henry's law constant, Pa;
$K =1/(1+4/(aD))$– wall factor, dimensionless;
k_A – overall mass transfer coefficient, $kmol/(m^2s)$;

M – molar weight, kg/kmol;
n – modified slope of the equilibrium curve, dimensionless;
W_{0L} – liquid load, $m^3/(m^2h)$;
w – interstitial velocity, m/s;
w_0- superficial velocity, m/s;
X,Y – molar ratio (liquid and gas phase, respectively), dimensionless;
x,y – mole fraction (liquid and gas phase, respectively), dimensionless;
p – partial pressure, Pa;
ΔP – pressure drop, Pa;
$Re_{gK}=g_{0g}d_{eK}/(\varepsilon\eta_g)$ - gas-phase Reynolds number used in hydraulic experiments;
$Re_g=g_{0g}d_e/(\varepsilon\eta_g)$ - gas-phase Reynolds number used in mass transfer experiments;
$Re_L=g_{0L}d_e/(\varepsilon\eta_L)$ - liquid-phase Reynolds number
Sc= $\eta/\delta M$ - Schmidt number;
$Sh_g= \beta_{Ag}d_e/\delta_{Ag}$- gas-phase Sherwood number;
$Sh_L= \beta_{AL}\vartheta_z/\delta_{AL}$ - liquid-phase Sherwood number;
β_A – individual mass transfer coefficient, $kmol/(m^2s)$;
δ_A - dynamic diffusivity, kmol/(ms);
ε - void fraction, m^3/m^3;
η - dynamic viscosity, Pa s;
$\Delta\pi_A$ - driving force, dimensionles;
$\vartheta_z=(\eta^2/(\rho^2g)^{1/3}$- equivalent linear dimension, m;
ρ - density, kg/m^3;
Ψ - friction factor, dimensionless;

Subscripts
A – diffusing component;
L – liquid phase;
dry – flow through dry packing;
g – to gas phase;
int – interface;
i – inert component;
m – mean value;
1,2 – upper or lower column cross-section, respectively;

Superscripts
* - gas-phase concentration in equilibrium with the liquid concentration.

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