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THE INFLUENCE OF THE REACTOR PRESSURE ON THE HYDRODYNAMICS IN A COCURRENT GAS-LIQUID TRICKLE-BED REACTOR

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ABSTRACT

The influence of the reactor pressure on the liquid hold-up in the trickle-flow regime and on the transition between trickle-flow and pulse-flow has been investigated in a trickle-flow column operating up to 6.0 MPa with water, and nitrogen or helium as the gas phase.
The effect of the gas velocity and gas density on the hold-up has been explained by means of the modified Galileo number $Ga\{1+\Delta P/(\rho_l g L)\}$. At the transition between trickle- and pulse-flow the liquid hold-up is - for a given value of the superficial gas velocity - nearly the same at each gas density. Therefore, at elevated gas densities the transition occurs at higher liquid throughputs. From a comparison of the experiments with water-nitrogen and water-helium it has been concluded that at an equal gas density - for given values of v_l and v_g - the hydrodynamic behaviour is the same.

KEYWORDS

Liquid hold-up; flow regime transition; elevated pressures.

INTRODUCTION

In the cocurrent gas-liquid trickle-bed reactor a gas and liquid phase flow downward over a fixed bed of catalyst particles. This type of three phase catalytic reactor is widely used in the petrochemical and chemical industry and is mostly operated at elevated pressures. In the design of a trickle-flow column the liquid hold-up and the flow regime transitions are important hydrodynamic parameters besides the pressure drop, the degree of catalyst wetting and the mixing of the fluids.
The liquid hold-up can be divided into the internal part, inside the pores of the catalyst, and the external hold-up partially occupying the void volume of the packed bed. The last-mentioned part is of main importance in describing the hydrodynamic behaviour. The flow regimes are generally distinguished into trickle-, pulse-, spray- and dispersed bubble-flow, see e.g. Gianetto *et al.*(1978). The transition between the trickle-flow and pulse-flow regime is sharp, while the transition to spray-flow and dispersed bubble-flow is more gradual. Information on the boundaries between the different regimes is essential because the pressure drop, the liquid hold-up and the especially mass transfer parameters are affected differently in each regime, see e.g. Mahajani and Sharma(1980).
In literature a number of studies has been presented on the transition between the flow regimes and on the external liquid hold-up. Both parameters are depending on the gas and liquid flow rates, the liquid properties and the packing geometry and size. To our knowledge no research has yet been carried out on the effect of the pressure of the gas phase. Hence there is still a lag between the published experimental research and the industrial practice at elevated pressures.
The objective of this study is to investigate the effect of the reactor pressure on the liquid hold-up in the trickle-flow regime and on the transition between trickle-flow and pulse-flow. Two gases of different molar mass - nitrogen and helium - are used to investigate whether at equal gas density, and hence different pressures, the hydrodynamic behaviour of the reactor is the same.

EXPERIMENTAL SET-UP AND PROCEDURES

A detailed description of the experimental installation used in this study has been given by Wammes *et al.*(1990b). Here we will only give a brief description of the set-up. A flow sheet of the installation is given in Fig.1.

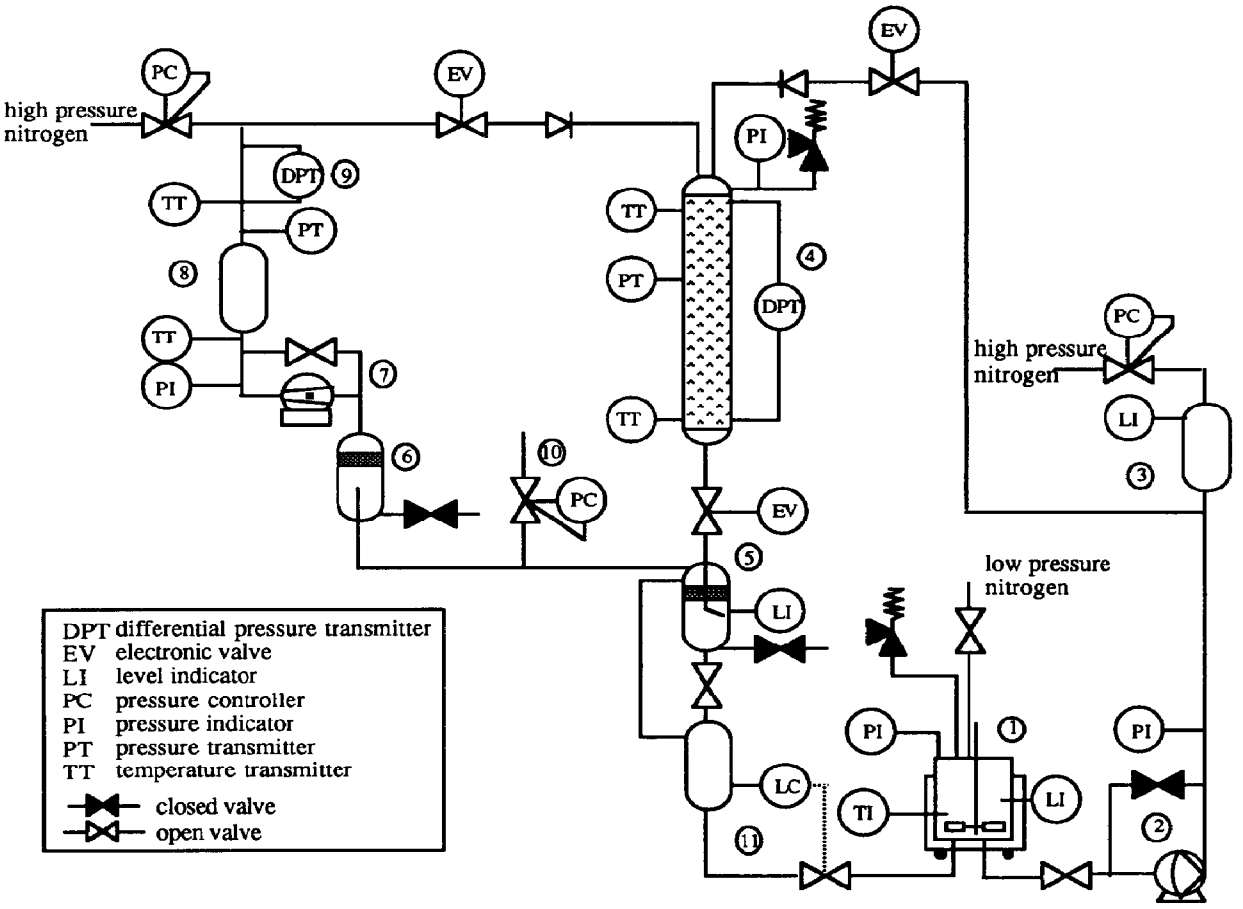

Fig.1. Flowsheet; 1 low pressure liquid storage vessel, 2 liquid pump, 3 air-chamber, 4 trickle-bed reactor, 5 gas/liquid separator, 6 demister, 7 gas booster, 8 gas buffers, 9 orifices, 10 system back pressure, 11 level controlled buffer vessel

The trickle-bed reactor(4) - the numbers refer to Fig.1 - has an inner diameter of 51 mm and has been packed with thoroughly cleaned glass spheres of 3 ± 0.5 mm diameter. The height of the packed bed is 2.62 m and it has an overall porosity of 0.39. A spray device has been located above the top of the bed in order to minimize the entrance effects by distributing the liquid phase evenly. Provisions have been made to measure the pressure and the temperature at the top and bottom of the bed and, the pressure difference over the packing. A reactor section of 1 m length has been manufactured in transparent polycarbonate material, by which we are able to observe the flow regime visually.
The installation has been designed for operating pressures up to 7.5 MPa at a temperature of 293 K. The maximum superficial liquid and gas velocity - based on the empty cross section of the reactor- are respectively 1.6 cm/s and 36 cm/s. The gas throughput is measured by means of an orifice meter (9) and the liquid flux is determined from the pump(2) calibration curves. A programmable AnalogDevices µ5000 data acquisition and control unit connected to an Apple2C computer is used for the monitoring and for continuous control of the level in the buffer vessel(11), the electronical valves, the pump piston stroke length, the pressure-, differential pressure- and temperature transmitters and the superficial gas- and liquid velocity in the reactor. The total pressure in the set-up is adjusted manually by means of a back pressure regulator(10). The physical properties of the gas-liquid systems we used in this study are listed in Table 1.

Table 1. Physical properties at 293 K of the liquids used in this study

PHYSICAL PROPERTIES	ρ_l (kg/m^3)	$\eta_l \cdot 10^3$ (Ns/m^2)	$\sigma_l \cdot 10^3$(N/m)
Water:	1000	1.0	72
40% ethyleneglycol:	1050	2.9	60
Ethanol:	790	1.2	22

Before the experiments are started the packing is prewetted by means of operating the reactor in the bubble-flow regime followed by draining.
The total external dynamic liquid hold-up is determined by the weighing method: the magnetic valves in the gas and liquid inlet and the reactor outlet are closed simultaneously. The gas-liquid separator is emptied and then its bottom valve is closed. Next the reactor outlet valve is opened so that the liquid trickles out of the column into the separator. The amount of liquid trickling out of the column is commonly called the dynamic hold-up and the amount remaining in the packed bed is indicated as the static or residual hold-up. The total external liquid hold-up is equal to the sum of the dynamic and static hold-up.
After collecting the dynamic liquid hold-up it is pressed out of the separator and weighed.
The minimum time of draining necessary to empty the column was 30 minutes for water and ethanol and 1 hour for the aqueous 40 % solution of ethyleneglycol. Data are reproducible within a relative error of 5 %.
The residual liquid hold-up measurements are performed at atmospheric pressure in a trickle-bed reactor of 50 mm in diameter, a packed bed height of 0.6 m and an overall bed porosity of 0.39. This reactor has been packed with the same packing material and the same packing procedure as has been used for the high pressure trickle-bed reactor. The reactor is flooded with the liquid phase and after the minimum time of draining the complete reactor is weighed.The residual hold-up can be calculated from the difference with the weight of the reactor with a dry packing. Data are reproducible with a relative error of maximal 10 %.
The transition between trickle-flow and pulse-flow has been observed visually in the transparent middle section of the column. In the following the expression "transition point" refers to the combination of pressure and flow rates at which pulses are observed with a frequency between 0.3 and 0.5 Hz.

RESULTS AND DISCUSSION

Total external liquid hold-up in the trickle-flow regime

The static liquid hold-up, situated around the contacting points of the particles, is independent of the gas flow, the liquid flow and the liquid viscosity, see Shulman *et al.*(1955) and Charpentier *et al.*(1968) and, in our opinion, it is also pressure independent. The experimentally determined values of the static liquid hold-up are for all three different liquids $\beta_{stat} = 0.11$. These results are well described by the diagram of Charpentier *et al.*(1968) in which the static hold-up is related to the Eötvös number, $Eö = \rho_l g d_p^2 / \sigma_l$. At low Eötvös numbers $Eö < 5$, as for our liquid-solid systems, the static hold-up reaches a maximium value of $\varepsilon\beta_{stat} \approx 0.05$, where ε is the void fraction of the packed bed.
The influence of the gas phase on the dynamic liquid hold-up will be discussed on the basis of the results for the systems water-nitrogen and water-helium. The same trends have been observed in the experiments with ethanol and aqueous 40% ethyleneglycol and with nitrogen as the gas phase. The effect of the gas phase on the liquid hold-up can be separated into the influence of the superficial gas velocity v_g and of the reactor pressure. In Fig.2 the influence of the gas velocity on the dynamic hold-up as a function of the superficial liquid velocity is shown for the system water-helium at 0.5 and 6.0 MPa. The effect of the reactor pressure at a constant superficial gas velocity is shown in Fig.3 for the system water-nitrogen. At each operating point the reactor operated in the trickle-flow regime: no pulses or gas bubbles could be observed in the transparent part of the column. The dynamic liquid hold-up values determined without gas flow, i.e. the single liquid flow operation, have also been plotted in both figures.Without gas flow the total hold-up is not affected by the pressure in the trickle-flow column, see Wammes *et al.*(1990a).

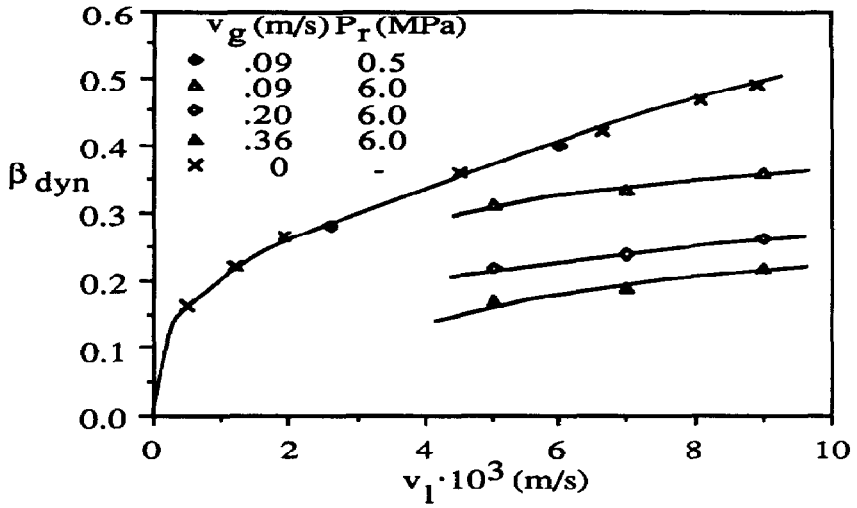

Fig.2: The influence of the gas velocity on the dynamic liquid hold-up for the system water-helium

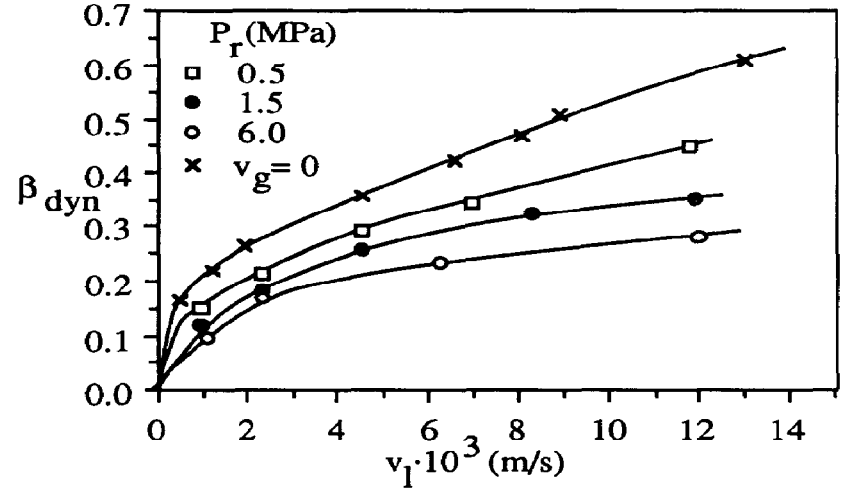

Fig.3: The dynamic liquid hold-up at various reactor pressures for the system water-nitrogen at v_g = 0.11 m/s

Fig.2 shows that when the reactor operates with helium as the gas phase at 0.5 MPa and v_g = 9 cm/s the dynamic liquid hold-up is equal to the single liquid flow operation. However, an increase of the gas velocity at 6.0 MPa results in a considerable decrease of the hold-up. The influence of the gas flow on the hold-up is more pronounced when nitrogen is used as the gas phase, see Fig.3. In case the reactor operates at 0.5 MPa and v_g = 11 cm/s, the dynamic liquid hold-up is substantially lower compared to the single liquid flow operation. A further increase of the reactor pressure at a constant gas velocity reduces the hold-up noticeably.

The influence of the superficial gas velocity and of the reactor pressure on the dynamic liquid hold-up can be explained by means of the pressure gradient over the reactor, i.e. the drag force at the gas-liquid interface. The pressure gradient depends on the gas velocity and on the gas density; it is together with the gravitational force the driving force for the liquid flow. The liquid hold-up is the result of two counteracting forces: the frictional forces at the packing surface and the driving forces acting on the liquid phase. Without gas flow the pressure drop equals zero, so the only driving force is the gravitational force, hence, the liquid hold-up is maximal. In case of both liquid and gas flow the total

driving force increases, due to the pressure gradient; nevertheless, the external liquid hold-up remains constant and only increases as soon as the pressure gradient becomes significant compared to the gravitational force. In Fig.4 the ratio between the drag and gravitational force per unit reactor volume has been plotted as a function of v_l for the same operating conditions as in Figs.2 and 3.

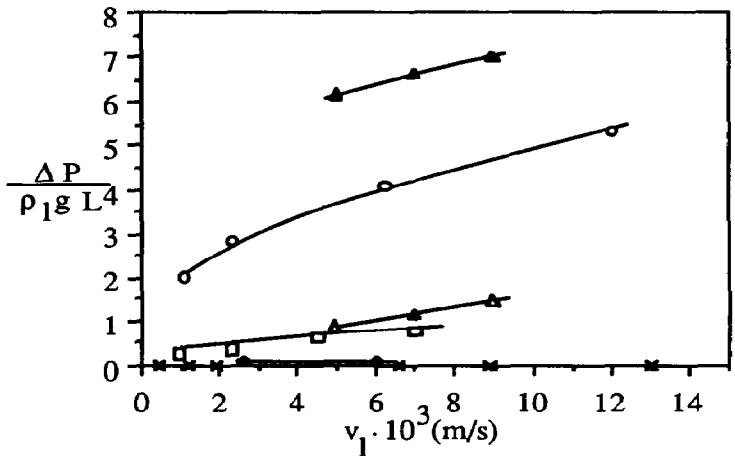

Fig.4: The value of $\Delta P/(\rho_l gL)$ as a function of v_l at different gas densities and gas velocities. The symbols are given in Figs. 2 and 3.

Comparing the figures 4 with 2 and 3 we see that for very low values of $\Delta P/(\rho_l gL)$ the dynamic liquid hold-ups are equal to those of the single liquid flow operation. But at increasing value of $\Delta P/(\rho_l gL)$ - due to an increase either of the gas velocity or reactor pressure or molar mass - the total driving force enlarges noticeably and, hence, the dynamic liquid hold-up reduces.
With our experimental data of water-nitrogen and of water-helium we have been able to compare the hydrodynamic state in the column in case of equal gas density, that is a helium pressure seven times higher than for nitrogen. From this comparison we conclude that, at a given superficial gas and liquid velocity, the pressure gradient and external liquid hold-up are the same for equal gas densities.
In literature a number of correlations have been proposed for the liquid hold-up, based on atmospheric experiments with air or nitrogen as the gas phase, see e.g. Rao *et al.*(1983). Our results, for a large range of gas densities, could be well described by means of the liquid phase Reynolds number and a modified Galileo number:

$$Ga_l^{\cdot} = Ga\left(1 + \frac{\Delta P}{\rho_l gL}\right) = \left(\frac{d_p^3 \rho_l^2 g}{\eta_l^2}\right)\left(1 + \frac{\Delta P}{\rho_l gL}\right) \qquad (1)$$

The influence of the gas velocity and gas density on the hold-up is fully taken into account by the pressure gradient.
For the effect of the type of packed bed, which has not been varied in our study, we used the variable $(a_v d_p/\varepsilon)^{0.65}$, derived by Specchia and Baldi(1977). Based on 160 experiments with helium-water, nitrogen with water, ethanol and aqueous 40% ethyleneglycol within the ranges: $2 \le Re_l \le 55$, $2 \le v_g \le 36$ cm/s, $0.32 \le \rho_g \le 70$ kg/m^3 and $0 \le \Delta P/(\rho_l gL) \le 16$ we propose the following correlation:

$$\beta_{dyn} = 3.8\left(\frac{\rho_l v_l d_p}{\eta_l}\right)^{0.55}\left(\frac{d_p^3 \rho_l^2 g}{\eta_l^2}\left(1 + \frac{\Delta P}{\rho_l gL}\right)\right)^{-0.42}\left(\frac{a_v d_p}{\varepsilon}\right)^{0.65} \qquad (2)$$

with a mean relative error of 8 %. This correlation can be applied in case the pressure gradient is negligible compared to the gravity and, moreover, when the pressure gradient is considerably larger

than the gravitational force. Finally, it is interesting to notice that Eq.2 is equal to the correlation of Specchia and Baldi(1977). They derived the correlation mainly on single liquid flow experiments, by which $\Delta P/L = 0$, with different liquids and with porous and non-porous packings of different geometry and size.

The transition between trickle-flow and pulse-flow

The transition between the trickle-flow and pulse-flow regime for the systems water-helium and water-nitrogen have been plotted in Fig.5 as a function of the superficial gas and liquid velocity at various gas densities. It has been observed visually that for each reactor pressure the first appearence of the pulses is located in the lower part of the transparent section. This has also been reported for atmospheric experiments, see e.g. Talmor(1977). After a slight increase of the gas throughput the pulses are observed over the entire transparent section. Fig.5 shows that the transition line shifts towards higher liquid throughputs with an increase of the gas density at a constant superficial gas velocity. This effect has also been found for the system aqueous 40% ethyleneglycol-nitrogen, see Wammes *et al.*(1990b).
In our opinion, the pulses originate from the collapse of liquid films which temporarily block the gas flow passages in the constrictions of the packed bed. The mean thickness δ of the liquid film on the packing particles is proportional to the total liquid hold-up:

$$\delta \approx \frac{\varepsilon \beta_{tot}}{a_v} \qquad (3)$$

The critical film thickness or liquid hold-up above which the films collapse, depends on the size of the disturbances at the liquid surface: fig.5 shows that for a given gas density and at low liquid throughputs, a high gas velocity is required to initiate pulses. On the other hand, at high liquid flow rates, low gas velocities are sufficient to form pulses.

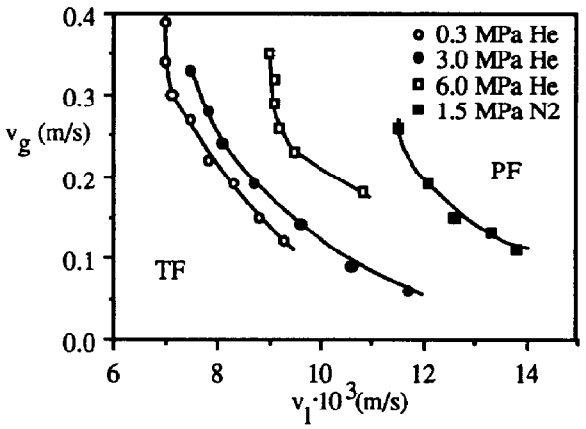

Fig.5: The transition between trickle- and pulse-flow at various gas densities

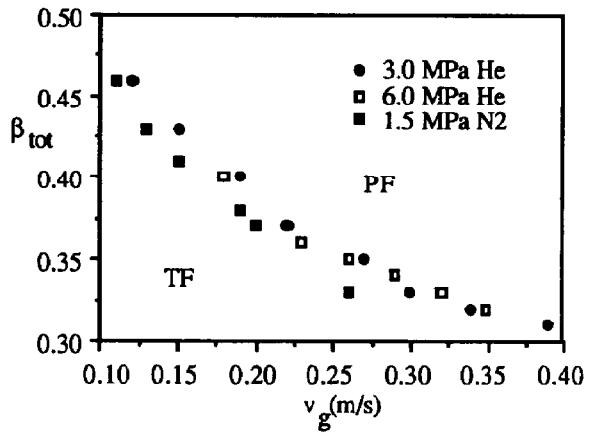

Fig.6: The total liquid hold-up at the transition as a function of the superficial gas velocity

In Fig.6 the total liquid hold-up at the transition between trickle- and pulse-flow, $\beta_{tot,tr}$, has been plotted as a function of v_g for various gas densities. It shows that at a constant superficial gas velocity and a higher gas density only a slightly lower liquid hold-up or film thickness is required for the transition between the flow regimes. The shift of the transition line towards higher v_l, see Fig.5, can be explained as follows. In case the trickle-bed reactor operates at the transition and only the reactor pressure or gas density is increased, the liquid hold-up will decrease as a result of the higher pressure gradient over the bed. The film thickness, see Eq.3, becomes too small to collapse and as a result the flow regime changes into trickle-flow. If subsequently the liquid velocity is increased, at a constant gas velocity and density, the liquid hold-up will grow. The flow regime shifts to pulse-flow again when the hold-up reaches its critical value $\beta_{tot,tr}$, corresponding to the set data of v_g and ρ_g, see Fig.6. For the system water-nitrogen and pressures above 2.5 MPa we have not been able to obtain pulses in the column. Our maximum liquid flow rate was not sufficient to reach the critical hold-up necessary for the initiation of pulses.

Comparing the transition experiments for water-nitrogen and water-helium, we can conclude that at equal gas density the transition occurs at the same combinations of the superficial gas and liquid velocities. Besides, the values of the hold-up and pressure gradient are the same.
The total external liquid hold-up line as a function of the gas velocity at the transition between trickle-flow and pulse-flow depends on the physical properties of the liquid phase, see Wammes *et al.*(1990b), and probably also on the type of the packed bed. For given liquid and packed bed properties, the transition as a function of v_l and v_g can be determined relatively easy in an atmospheric set-up. The diagram of Charpentier *et al.*(1968) and Eq.2 can be used to estimate the total external hold-up at the transition $\beta_{tot,tr}$. The resulting critical hold-up line, derived for atmospheric conditions, holds also as a first approximation for the critical hold-up at elevated pressures, because the influence of the gas density is relatively small, see Fig.6.

CONCLUSIONS

In literature not much information is available on the total external liquid hold-up and on the transition between trickle- and pulse-flow in a cocurrent gas-liquid trickle-bed reactor operating at elevated pressures. In this study, static and dynamic liquid hold-up experiments with helium-water, nitrogen and water, ethanol and aqueous 40% ethyleneglycol and transition experiments with helium-water and nitrogen-water have been discussed for reactor pressures up to 6.0 MPa. The static hold-up experiments have been restricted to low Eötvös numbers and could be well described by the diagram of Charpentier *et al.*(1968).
The gas velocity and gas density both influence the dynamic liquid hold-up. The dynamic liquid hold-up correlation as derived from the single liquid flow experiments, can still be used if we introduce the modified Galileo number, see Eq.1.
The transition between trickle-flow and pulse-flow shifts towards higher liquid throughputs in case the gas density has been increased at a constant superficial gas velocity: due to the higher pressure gradient at an elevated gas density, the hold-up or mean liquid film thickness decreases. The films become too thin to collapse and no pulses can be formed. Therefore higher liquid throughputs are required at elevated gas densities to create sufficient hold-up for the initiation of the pulses.
From a comparison of experiments with water-nitrogen and water-helium it has been concluded that at equal gas density - for a given setpoint of v_l and v_g - the hydrodynamic behaviour is the same.

ACKNOWLEDGEMENTS

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NOTATION

a_v	: specific external surface area of the packing (m^2/m^3)
d_p	: nominal particle diameter (m)
Eö	: Eötvös number defined as $\rho_l g d_p^2/\sigma_l$ (-)
Ga_l	: Galileo number defined as $d_p^3 \rho_l^2 g/\eta_l^2$ (-)
Ga_l^*	: modified Galileo number defined as $Ga_l \{1+\Delta P/(\rho_l g L)\}$ (-)
g	: gravitational acceleration constant (m/s^2)
P_r	: reactor pressure (MPa)
$\Delta P/L$	: reactor pressure gradient (N/m^3)
Re_l	: Reynolds number defined as $\rho_l v_l d_p/\eta_l$ (-)
v_g	: superficial gas velocity based on the empty reactor cross section (m/s)
v_l	: superficial liquid velocity based on the empty reactor cross section (m/s)
β	: hold-up, liquid volume in void volume (-)
δ	: mean liquid film thickness (m)
ϵ	: overall porosity of the packed bed (-)
η_l	: dynamic liquid viscosity (Ns/m^2)
ρ_l	: liquid density (kg/m^3)
σ_l	: gas-liquid surface tension (N/m^2)

subscripts

dyn	: dynamic
stat	: static
tot	: total
tr	: transition between trickle-flow and pulse-flow

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