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MODELLING OF THE HYDRODYNAMICS OF CONCURRENT GAS AND LIQUID FLOW THROUGH PACKED BED

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Abstract

The aim of the present study is to simulate concurrent gas and liquid flow through packed bed in the gas continuous flow regime (GCF) and continuity shock waves regime (CSW) using computational fluid dynamics (CFD). The application of multiple gas-liquid-solid model requires the knowledge of relationships determining interactions between phases. The exchange coefficients of these forces were defined by means of equations suggested by ATTOU et al. (1999). As a result of the computational simulation the following data were obtained: gas pressure drop in the bed, volume fraction distribution of a given phase (liquid and gas holdups) along the packing and its mean value in the reactor. The comparison of the values of the hydrodynamic parameters, both calculated and obtained experimentally in a column packed with 3 mm glass spheres, indicates that CFD model can be applied to model the hydrodynamics of concurrent gas and liquid flows through a packed bed because a good compatibility of the compared parameters was obtained.

Symbols

$d_{\min} = \left(\frac{\sqrt{3}}{\pi} - \frac{1}{2} \right) d_p$ – minimum equivalent diameter of the area [m]

d_p – particle diameter [m]

E_1, E_2 – Ergun constants

f – wetting efficiency of the bed

F_{jk} – momentum exchange coefficients between phases j and k [$\text{kgm}^{-3}\text{s}^{-1}$]

g – acceleration due to gravity [ms^{-2}]

J_0 – zero th order Bessel function

K_k – drag between phases j and k [Nm^{-3}]

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P	– pressure [Pa]
r	– radial coordinate [m]
R	– radius of column [m]
t	– time [s]
$\bar{u}$	– interstitial velocity [ms^{-1}]
w	– superficial velocity [ms^{-1}]

Greek letters

α	– volume fraction
ε	– porosity of the bed
e_L	– liquid holdup
η	– dynamic viscosity [Pas]
ρ	– density [kg m^{-3}]
σ	– surface tension [Nm^{-1}]
τ	– stress tensor [Nm^{-2}]

Subscripts

av	– denotes average
b	– denotes base
c	– denotes capillary
d	– denotes dynamic
g	– denotes gas phase
ip	– denotes impulse
j	– denotes phase other than k
k	– denotes k^{th} phase
L	– denotes liquid phase
S	– denotes solid phase
α	– L, g

Introduction

Three-phase reactors in which the liquid and gas phases flow down a fixed catalytic bed are termed trickle-bed reactors (TBR). This type of reactor is employed on an industrial scale in the processes of selective hydrogenation of various petroleum fractions, as well as in the oxidation of organic pollutants in waste waters and flue gases via reactions occurring over a bed of immobilized bacteria. Depending on the flow rates of the two phases, their physicochemical properties and geometry and size of the packing, various flow patterns can be observed of which two are of considerable industrial importance: the gas – continuous flow regime (GCF) and the pulsing flow regime (PF). The pulsing flow regime is especially attractive in the processes of mass and heat transfer as the experimental values of the transfer coefficients are by far larger than those in the GCF regime; moreover, perfect wetting of the packing is observed. However, carrying out the processes in the PF regime requires supplying the reactor with large streams of gas and liquid which results in a very short residence times of reagents in a bed and this, in turn, has an unfavourable influence on their conversion. Taking the above into consider-

ation, the idea of carrying out processes in TBRs at periodically changing feeding the bed with liquid was created (BELHOUWER et al. (1999)). Unsteady state (periodic) operations can be realized either by means of temporary stopping the stream of liquid (ON-OFF method) or by periodic change in the liquid velocity between low, but not equal zero, and high value of this parameter (BASE-IMPULSE method). Taking into account the liquid velocity in an impulse, the process can be carried out as continuity shock wave flow (CSW) or liquid induced pulsing flow (LIPF).

Periodic operations can be also grouped with respect to the criterion of duration of particular liquid cycles. Thus the processes can be carried out by means of slow or fast changing cycles of liquid modulation (SLOW MODE or FAST MODE).

In the present study, the subject of analysis is the hydrodynamics of TBR operating at GCF and CSW regimes. The key hydrodynamic parameters are pressure drop and liquid holdup. These parameters are inseparable connected and occur simultaneously in balance equations of both fluids. In the literature, a large number of papers can be found, that present, for various experimental systems, the results of experiments in which mentioned above parameters have been determined (CLEMENTS and SCHMIDT (1980), ELLMAN et al. (1990), LARACHI et al. (1991), MIDOUX et al. (1976), RAO et al. (1983), RAO and DRINKENBURG (1983), SAI and VARMA (1987), SPECCHIA and BALDI (1977), WAMMES i wsp. (1991)). The analysis of correlation equations which were developed on the basis of experimental data showed considerable differences between calculated values. Therefore, it was necessary to formulate a mathematical model describing the hydrodynamics of TBRs. A few one-dimensional models describing concurrent gas and liquid flow through packed bed of TBR operating in GCF regime can be found in literature. They are both phenomenological microscopic models (single canals in packing are taking into account) (AL-DAHMAN et al. (1998), HOLUB et al. (1992, 1993), ILIUTA and LARACHI (1999), ILIUTA et al. (2000)) and macroscopic models by SAEZ and CARBONELL (1985), GROSSER et al. (1988) or ATTOU et al. (1999), based on volume averaged balances of mass and momentum of both phases flowing concurrently through packing. Unfortunately, in the empirical equations and models uniform porosity and velocity of fluids in the bed has been assumed and the phenomena concerning complex flow, mixing and wetting of the bed are usually expressed jointly by some empirical parameters. Therefore, recently fast development of the Computational Fluid Dynamic (CFD) model, which is a quantitative formulation of the momentum balance (Navier-Stokes equations) caused a more frequent application of the CFD to simulate the dynamic phenomena in the TBR. The application of the CFD method to simulate the operation of trickle-bed reactors is much more difficult than to simulate the

processes in two-phase systems (KHAN et al. (2014), KEIR and JEGATHEESAN (2014), KUIPERS and VAN SWAAIJ (1997), SAJJADI et al. (2012), SHAIKH and AL-DAHMAN (2013), WANG et al. (2007), WANG et al. (2010)) which, among others, is due to difficulties connected with a proper description of the structure of the bed of solid particles, interaction forces between the flowing phases and the wetting efficiency of the surface of the packing (JAWORSKI (2005), JIANG et al. (2001), WANG et al. (2013)).

The aim of the present study was the simulation of concurrent flow of gas and liquid through a packed bed in GCF and CSW regimes using the CFD model. The simulations were carried out using commercial FLUENT 6.1.22 software. As the pressure gradient and average holdup are two main hydrodynamic parameters, these quantities have been chosen for comparison. The mean relative error and standard deviation of experimental and computed values of the parameters mentioned above were used as the validation criteria of the model.

Mathematical model

The Eulerian multi-fluid model was used to simulate the fluid flow through the bed of solid particles. The model treats each phase as continuous fluid having various velocities, volume fractions and physicochemical properties. The volume averaged equations of mass and momentum balances expressed by means of volume fractions occupied by each phase and their local velocities can be written as:

$$\frac{\partial}{\partial t}(\alpha_k \rho_k) + \nabla \cdot (\alpha_k \rho_k \vec{u}_k) = 0; \quad k = L, g \quad (1)$$

$$\frac{\partial}{\partial t}(\alpha_k \rho_k \vec{u}_k) + \nabla \cdot (\alpha_k \rho_k \vec{u}_k \vec{u}_k) = -\alpha_k \nabla P + \nabla \cdot \bar{\bar{\tau}}_k + \alpha_k \rho_k \vec{g}_k + \vec{K}_k; \quad k = L, g \quad (2)$$

where the viscous stress tensor is expressed by the dependency:

$$\bar{\bar{\tau}}_k = \alpha_k \eta_k (\nabla \vec{u} + \nabla \vec{u}^T) \quad (3)$$

The effect of pre-wetting of packing was also taken into consideration in the model. JIANG et al. (2002a) introduced the empirical coefficient f , characterizing the wetting efficiency of the bed, into the capillary pressure relation:

$$P_g - P_L = (1 - f)P_c \quad (4)$$

For pre-wetted bed f is set equal to one implying a zero capillary pressure and for non-wetted bed f is set to zero. Capillary pressure was described by means a function suggested in the work of ATTOU and FERSCHNEIDER (2000):

$$P_g - P_L = 2\sigma \left(\frac{\alpha_s}{1 - \alpha_g} \right)^{0.333} \left(\frac{1}{d_p} + \frac{1}{d_{\min}} \right) \left(1 + 8.1 \frac{\rho_g}{\rho_L} \right) \quad (5)$$

Capillary pressure gradient, described by relationship (5), should be introduced into the balance equation of the liquid phase in case of an incomplete wetting of the bed by means of the User Defined Function (UDF).

The application of the multiple gas-liquid-solid model requires a knowledge of relationships determining the interactions between phases. It is very significant element of uncertainty in the modelling of the processes in TBR. These forces are considerable and dominate in the momentum balance equations. The last term of this equation (2), $\vec{K}_k$, represents interaction forces between phases in the form of:

$$\vec{K}_k = \sum_{j=1}^n F_{jk} (\vec{u}_j - \vec{u}_k) \quad (6)$$

Since $u_s = 0$ and $F_{jk} = F_{kj}$ (FLUENT (2006)), then the above relationship is reduced to the form of:

– for the gas phase:

$$\vec{K}_g = -F_{gL}(\vec{u}_g - \vec{u}_L) - F_{gS}\vec{u}_g \quad (7)$$

– for the liquid phase:

$$\vec{K}_L = F_{gL}(\vec{u}_g - \vec{u}_L) - F_{LS}\vec{u}_L \quad (8)$$

The coefficients of momentum exchange between phases (F_{jk}) occurring in these equations were analysed in the work by JANECKI et al. (2016). They can be derived from one-dimensional models (slit model of HOLUB et al. (1992), permeability model of SAEZ and CARBONELL (1985)) as well as from two-phase model of interphase actions of ATTOU et al. (1999). The first two models neglect interaction forces between the gas and liquid phases due to their decay in the GCF regime (SOUADNIA et al. (2005)). However, experimental research (AL-DAHMAN and DUDUKOVIC (1994), AL-DAHMAN et al. (1997), LARACHI et al. (1991a)) shows that gas flow has a significant influence on the hydrodynamics of TBRs, especially at elevated pressure. The exchange coefficients F_{jk} descri-

bing interactions between phases in a three-phase system (gas-liquid-solid) can be derived only from the ATTOU et al. (1999) model. They have the form similar to the Ergun relations:

– for gas-liquid momentum exchange coefficient:

$$F_{gL} = A_g \left(\frac{\alpha_S}{1 - \alpha_g} \right)^{2/3} + B_g \left(\frac{\alpha_S}{1 - \alpha_g} \right)^{1/3} |u_g - u_L| \quad (9)$$

– for gas-solid momentum exchange coefficient :

$$F_{gS} = A_g \left(\frac{\alpha_S}{1 - \alpha_g} \right)^{2/3} + B_g \left(\frac{\alpha_S}{1 - \alpha_g} \right)^{1/3} |u_g| \quad (10)$$

– for liquid-solid momentum exchange coefficient:

$$F_{LS} = (1 - \alpha_S)(A_L + B_L|u_L|) \quad (11)$$

Coefficients A_g , A_L , B_g i B_L present in the above equations are expressed by the following formulas:

$$A_g = E_1 \mu_g \frac{(1 - \alpha_g)^2}{\alpha_g d_p^2} \quad (12)$$

$$B_g = E_2 \rho_g \frac{1 - \alpha_g}{d_p} \quad (13)$$

$$A_L = E_1 \mu_L \frac{\alpha_S^2}{\alpha_L d_p} \quad (14)$$

$$B_L = E_2 \rho_L \frac{\alpha_S}{\alpha_L d_p} \quad (15)$$

In the presented relationships there are parameters which determine the values of interaction forces. They are Ergun's constants (E_1 and E_2). Since by changing the values of E_1 and E_2 one can control the values the interaction forces, doubts keep appearing which values of Ergun's constants to use in the calculations. The values determined experimentally or suggested by

MACDONALD et al. (1979) ($E_1 = 180$; $E_2 = 1.8$) are most commonly applied in literature.

Description of the fixed bed structure in trickle-bed reactors

Numerous experiments showed (BENENATI and BROSILOW (1962), GOODLING et al. (1983), MUELLER (1992)) that randomly packed beds exhibit greater porosity in the vicinity of the column wall (in the region equaling to the width of 4–6 diameters of solid particles). In this region porosity exhibit strong fluctuations which are attenuated at greater distances from the wall. It makes the radial profile of bed porosity at the wall undergo strong changes which may significantly influence mean liquid holdup and gas pressure drop in the bed, especially in a reactor whose ratio of the column diameter to the particle diameter is low (laboratory reactors). Porosity values in the core of the bed show significantly lower differences and reach values close to the mean values of the parameter.

A proper description of a bed structure in a reactor should include the following elements (GUNJAL et al. (2005), JIANG et al. (2002a), LI et al. (2006)):

- the overall mean porosity,
- the axial porosity profile,
- the axially averaged radial porosity profile.

In literature one can find various correlation equations describing porosity distribution along the radius of the apparatus. Those that can be applied to model TBR were set in works by WANG et al. (2013) and VAN ANTWERPEN et al. (2010). These relationships can be divided into two groups. The first one is represented by the oscillatory correlations (MARTIN (1978), COHEN and METZNER (1981), MUELLER (1992), BEY and EINGENBERGER (1997), DE KLERK (2003)) and the other one is represented by the exponential correlations (VORTMEYER and SHUSTER (1983), HUNT and TIEN (1990) and SUN et al. (2000)). The chosen relationships describing the radial porosity profiles were analysed in detail in the work by JANECKI et al. (2014).

Experimental set-up and procedure

The experiments, being the comparative base for computational simulations, were carried out in an installation which main part was cylindrical column of 0.057 m I.D. packed with 3 mm glass spheres ($\epsilon = 0.38$, $a = 1240 \text{ m}^{-1}$) to a height of 1.35 m. The column operated at concurrent down-flow of gas (nitrogen) and liquid phases. In order to investigate the influence of the

physicochemical properties of the liquid phase on the experimental results water, two aqueous solutions of glycerol (with the viscosity of $2.5 \cdot 10^{-3}$ and $3 \cdot 10^{-3}$ Pas) and methanol solution with viscosity of $1.64 \cdot 10^{-3}$ Pas and surface tension $45.8 \cdot 10^{-3} \text{ Nm}^{-1}$ have been used.

The experiments were carried out at constant temperature and pressure (30°C , $\sim 10^5 \text{ Pa}$) for a wide range of flow rates of both phases (fully in GCF and CSW regimes). The operation of the system both in GCF and CSW regimes was discussed in detail elsewhere (BARTELMUS and JANECKI (2003), BARTELMUS et al. (2006)).

The electrochemical method was used to measure the liquid hold-up. The basic value determined during the experiments were the variations in conductivity of the two-phase gas-liquid mixture flowing through the void volume of the bed. The conductance cells placed in the bed were used to measure the conductivity of the solution; the changes in conductivity reflect the changes in the amount of gas and liquid in the mixture. Detailed description of the experimental procedure was presented elsewhere (BARTELMUS, JANECKI (2003)).

Gas pressure drop in the packing was measured by means of piezoelectric gauges (Cole-Palmer); the pressure signal, converted into electric signal, was fed into computer memory. The values of pressure drop used in calculations are the values averaged in time.

The results of experiments are discussed in detail in the earlier studies (BARTELMUS, JANECKI (2003), GANCARCZYK et al. (2014)).

Computational calculations

In order to obtain reliable results of numerical computations it is essential to select optimum grid in the system considered. It was selected as a result of numerical simulations in which the number and sizes of the cells were changed both in the core of the bed and in the layer at the wall. The way of the selection of the optimum grid was presented in detail in the work by JANECKI et al. (2014). The optimum grid, finally used in the computations, was composed of 7700 cells, out of which 4400 cells were the size of $3\text{mm} \times 5\text{mm}$ (the core of the bed) and 3300 cells forming the layer at the wall were the size of $1\text{mm} \times 5\text{mm}$. The grid was created by means of GAMBIT preprocessor.

The following boundary conditions were attributed to the generated grid: in the axis of the reactor – symmetry, at the wall – lack of slip, at the inlet – flat velocity profile, at the outlet – zero velocity gradient.

As it has been mentioned earlier, a proper description of bed structure should include: overall mean porosity, variance of porosity in axial direction

and axially averaged radial porosity distribution. In the present study the overall mean porosity of bed was determined experimentally and it was 0.38.

The algorithm of generating random numbers of the normal distribution, for a given mean value and standard deviation (5%) was used to determine the changes in porosity in the axial direction. Experimentally determined value of bed porosity ($\varepsilon = 0.38$) was used as a mean value of the parameter.

The relationships describing momentum exchange coefficients (F_{jk}), occurring in interaction forces between phases (Eqns 9–11), were implemented into a commercial Fluent software by means of the User Definition Function (UDF) and were used to calculate gas pressure drop in bed and mean liquid holdup.

To qualify the procedure of computational calculations and at the same time decrease the differences between calculated and experimental values the parametric sensitivity of CFD model was checked in the study by JANECKI et al. (2014). The following parameters, which have the greatest influence on the computed values, were analysed: relations defining the interphase momentum exchange coefficients and different radial porosity profiles.

In the present study, as a result of parametric sensitivity analysis, the classical equations of ATTOU et al. (1999) defining the friction factors (F_{jk}), with Ergun's constants calculated from neuron nets (ILIUTA et al. (1998)) ($E_1 = 235.53$; $E_2 = 1.59$) were applied in the CFD model. The radial porosity profiles were calculated from MARTIN'S (1978) correlations:

$$\varepsilon(x) = \varepsilon_{\min} + (1 - \varepsilon_{\min})x^2 \quad \text{for } -1 \leq x \leq 0 \quad (16)$$

$$\varepsilon(x) = \varepsilon + (\varepsilon_{\min} - \varepsilon) \exp\left(\frac{-x}{4}\right) \cos\left(\frac{\pi x}{0.876}\right) \quad \text{for } x \geq 0 \quad (17)$$

where:

$$x = 2 \frac{R - r}{d_p} - 1; \varepsilon_{\min} = 0.2$$

A flat velocity profiles of the liquid phase was assumed at the inlet into the column because a distributor of liquid, distributing liquid evenly on the surface of the bed, was used during the experiments. The following initial conditions were used for a reactor operating in the GCF regime: experimental values of gas (w_g) and liquid (w_L) velocities as well as mean volume fraction of the liquid phase (α_L).

However, the initial conditions for a reactor operating in the CSW regime were the following: experimental values of liquid holdup (ε_L), gas velocity (w_g),

liquid velocity in the base (w_{Lb}) and in the impulse (w_{Lip}), base and impulse durations (t_b and t_{ip} , respectively). Velocity of the solid phase was assumed to be zero. A unsteady state simulations with a time step of $t = 0.01$ s was used in the calculations.

Findings of the simulations

As a result of the computational simulations one can obtain gas pressure drop in bed, volume fraction distribution of a given phase (liquid holdup and gas holdup) along the bed and its mean value in the reactor. The comparative base for the simulations was our own data base obtained in experiments carrying out for varying flow rates of both phases and for systems of various physicochemical properties.

Experimental data as well as results of CFD simulations performer for reactor operating at constant feeding the bed with liquid and for GCF regime were illustrated, as an example, in Fig. 1. Good compatibility between calculated and obtained experimentally values of hydrodynamic parameters was obtained; the mean relative error (e_Y) for liquid holdup was 3.95% and for pressure drop was 9.4%.

The results of simulations performed for periodically changing feeding the bed with liquid phase were shown in Figs 2 and 3. They quite faithfully reproduce the changes in both hydrodynamic parameters with the changes in gas phase velocity (Fig. 2), mean liquid phase velocity and duration of base and impulse (Fig. 3). While comparing calculated and experimental values of gas pressure drop in bed and mean values of liquid holdup it has to be stated that the differences between the calculated and measured values of both parameters do not exceed a few per cent.

Fig. 4 shows a computational simulation of changes of liquid holdup in the whole column for nitrogen – 24% methanol solution system. Such result were obtained for fast changing cycles (FAST MODE) and reactor operating in CSW regime. For such low alternate liquid velocities, the impulses seen at the top of the column decay during migration down the bed, forming continuous stream of liquid (GCF regime).

A few impulses can always be seen in the column operating at fast changing cycles, however, for slow changing cycles (SLOW MODE) the duration of base and impulse may be so long that they will not be seen in the column. In such a case, there is a GCF regime in the whole column for a very short time. The results of simulation of a slow changing cycles for a water-nitrogen system and the duration of cycles of $t_b = 20$ s and $t_{ip} = 3$ s are presented, as an example, in Fig. 5. For conventional time of 0 s (the moment of switching the cycle) liquid

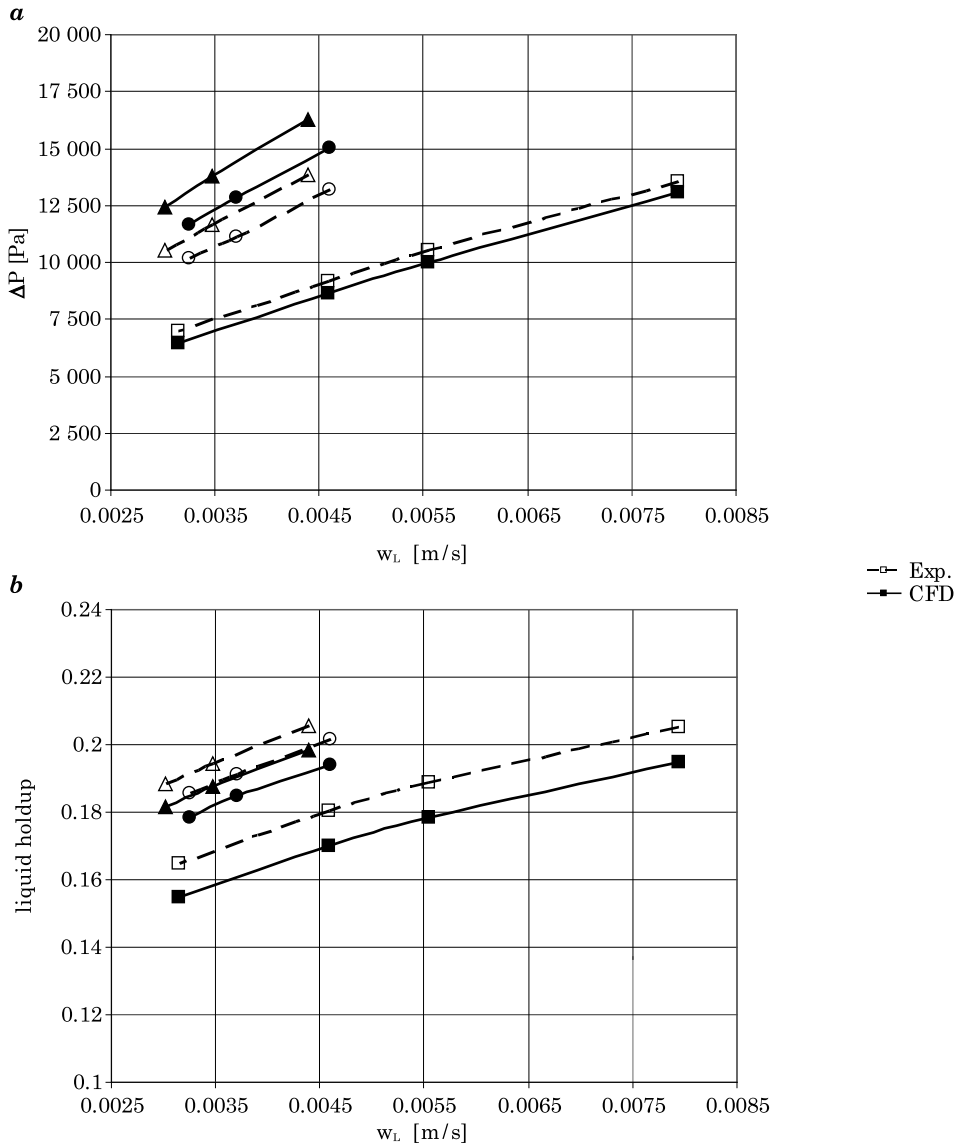


Fig. 1. Dependences of the gas pressure drop ΔP (a) and liquid holdup (b) as function of the liquid velocity w_L and phases' properties (square: water – nitrogen, circle: glycerol solution (30 wt.%) – nitrogen, triangle: glycerol solution (35 wt.%) – nitrogen); $w_g = 0.13$ m/s; GCF regime

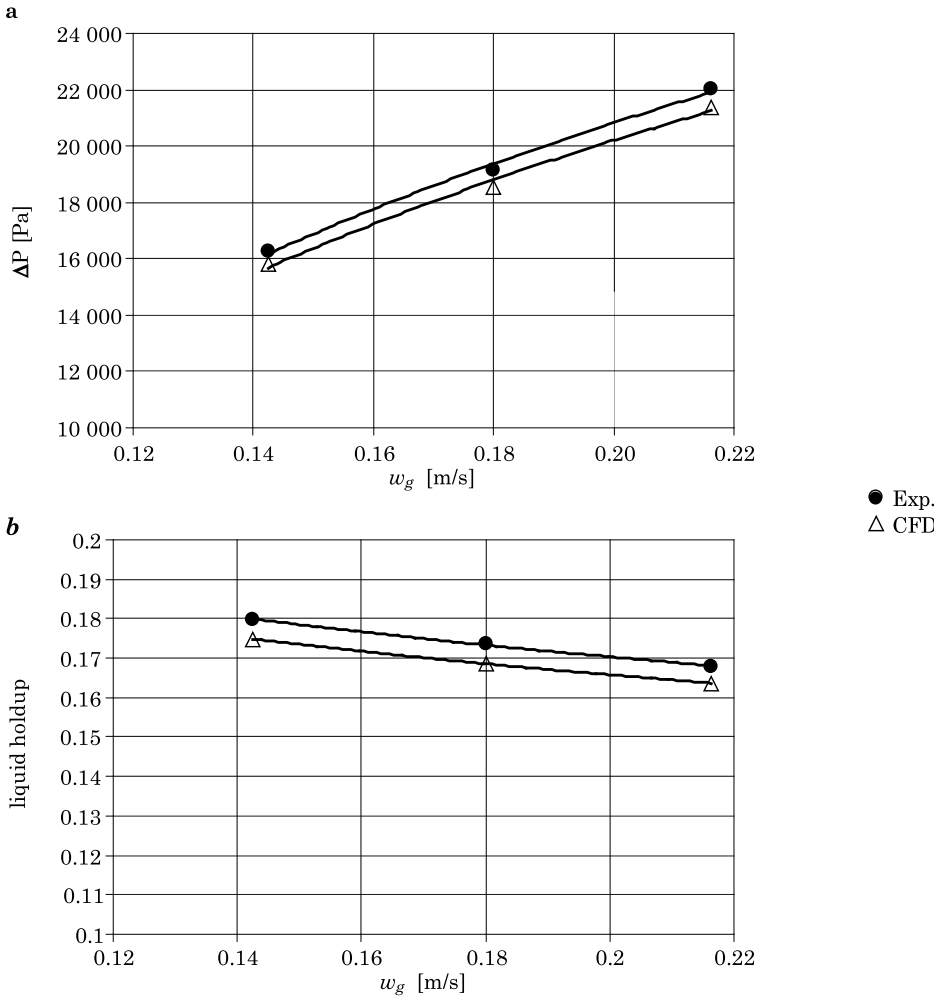


Fig. 2. Dependences of the gas pressure drop ΔP (a) and liquid holdup (b) on the gas velocity w_g ; nitrogen-water system, full symbols – experimental data, open symbols – CFD model; $w_{Lav} = 0.006 \text{ m} \cdot \text{s}^{-1}$, $t_b/t_{ip} = 3/1$; CSW regime

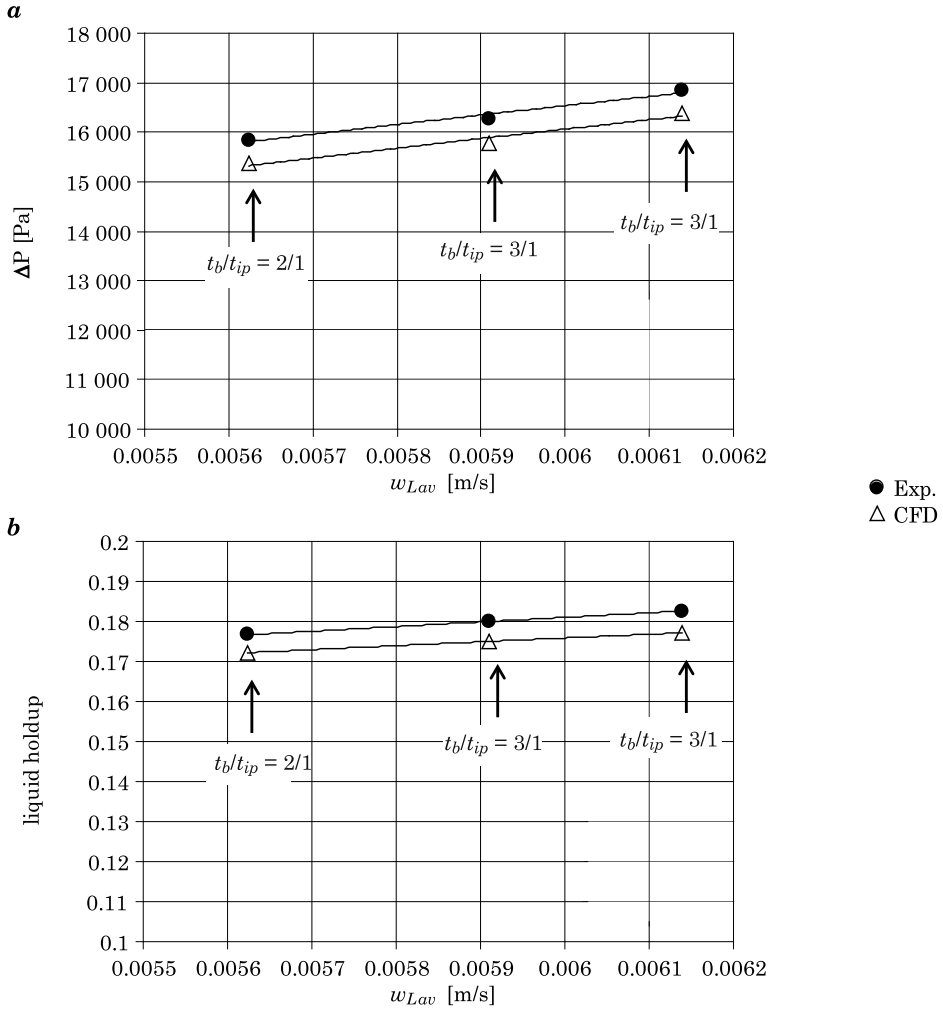


Fig. 3. Dependences of the gas pressure drop ΔP (a) and liquid hold-up (b) on the average liquid velocity w_{Lav} (or duration of the base and impulse) at various Ergun constants, nitrogen-water system, $w_g = 0.143 \text{ m} \cdot \text{s}^{-1}$, $w_{Lb} = 0.0048 \text{ m} \cdot \text{s}^{-1}$; CSW regime

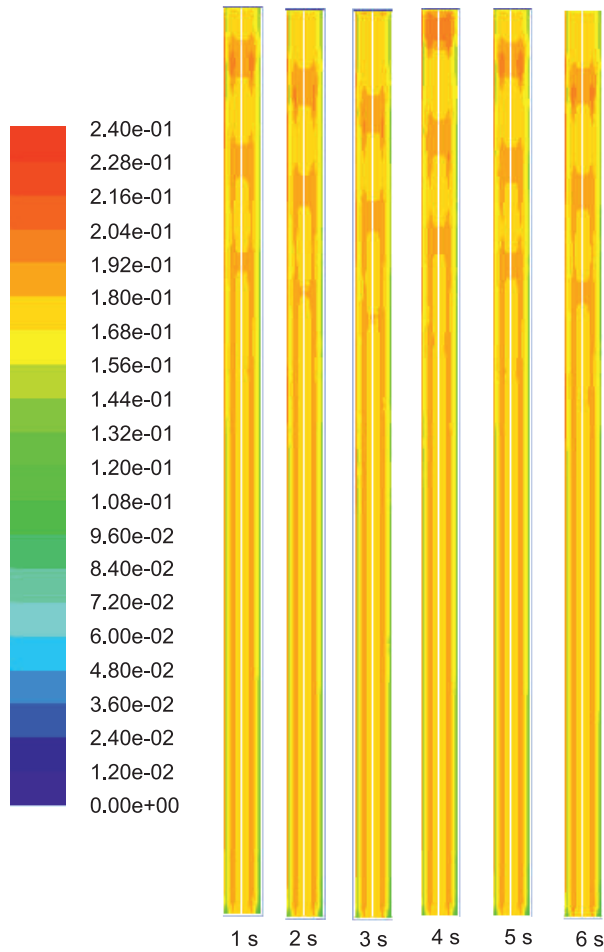


Fig. 4. Time dependences of liquid holdup along the column; nitrogen – 24% methanol solution system, $w_g = 0.1\text{ m/s}$; $w_b = 0.0025\text{ m/s}$, $w_{imp} = 0.0049\text{ m/s}$; $t_b = 3\text{ s}$, $t_{ip} = 1\text{ s}$

flows with a constant velocity (w_{Lb}). The front of impulse, which was created after 4 seconds, reaches the column bottom after about 20 s. After that time the GCF regime is again observed in the column.

Conclusions

A simulation of concurrent flow of gas and liquid through fixed bed of the column operating in GCF and CSW regimes were performed using computational fluid dynamics (CFD) model. As a result of the simulations the following

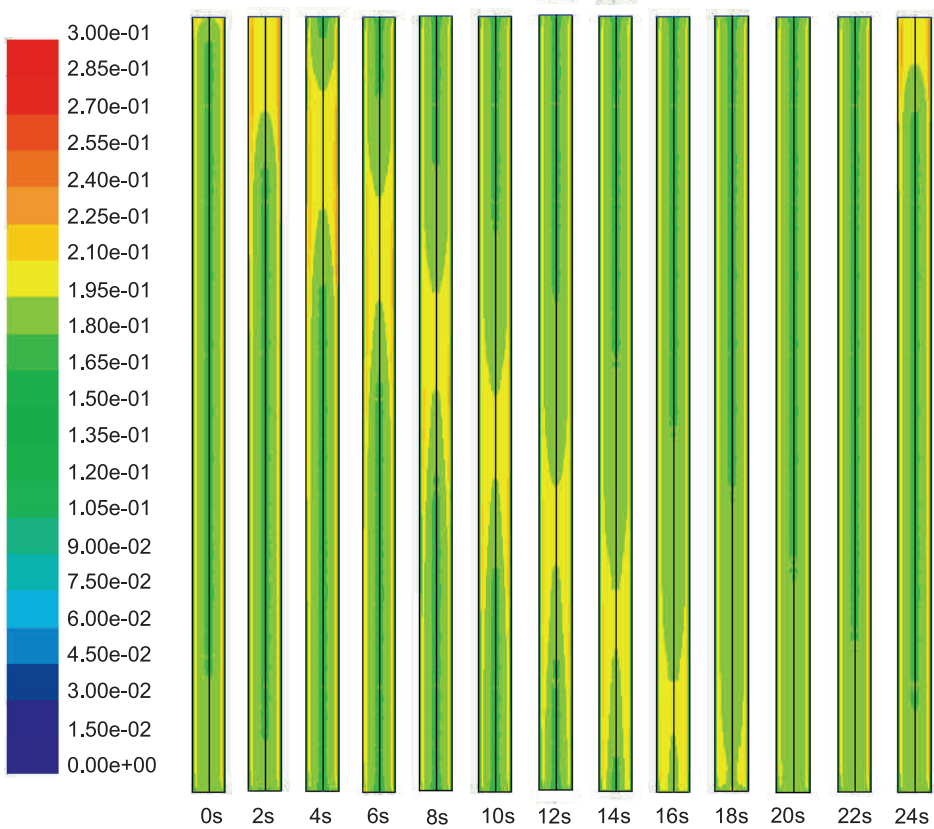


Fig. 5. Liquid hold-up variation with time along the column; nitrogen – water system; $w_g = 0.14$ m/s, $w_b = 0.0048$ m/s, $w_{Lip} = 0.0082$ m/s; $t_b = 20$ s, $t_{ip} = 3$ s

hydrodynamic parameters were obtained: gas pressure drop in the bed, volume fraction distribution of a given phase (liquid holdup and gas holdup) along packing and its mean value in the reactor. The results of the calculations reflect well both quantitative and qualitative changes of hydrodynamic parameters with the changes of: flow rates of both phases, duration of base and impulse and physicochemical properties of the liquid phase. The comparison of liquid holdup values and gas pressure drop in bed calculated and experimentally obtained shows that the CFD model applied in the present study may be used to model the hydrodynamics of concurrent flows of gas and liquid through a fixed bed, since a good compatibility of the compared hydrodynamic parameters was obtained.

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