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Hydrodynamic characteristics of a two-phase gas-liquid flow upward through a fixed bed of spherical particles

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The influence of an electrochemically generated gas phase on the hydrodynamic characteristics of a three-phase system has been examined. The two-phase fluid, (gas-liquid), in which the liquid phase is the continuous one, flows through a packed bed with glass spheres. The influence of the liquid velocity was examined, as well as the gas velocity and particle diameter on the pressure drop through the fixed bed. It was found that with increasing liquid velocity (w_l = 0.0162–0.03 m/s), the relative pressure drop decreases through the fixed bed. With increasing current density, the pressure drop increases, since greater gas quantities stay behind in the fixed bed. Besides, it was found that with decreasing diameter of the glass particles, the relative pressure drop also decreases. The relationship betweeen the experimentally obtained friction factor and the Reynolds number was established.

Keywords: two-phase flow, fixed bed, water electrolysis, pressure drop, fluidization.

INTRODUCTION

Reactors with a fixed bed are commonly used in industry, in chemical and biochemical processes, in electrometallurgy as well as in processes of environment protection.[1–3] Besides, in reactor engineering, especially in the part which refers to hydrogenation, the characteristics of three-phase systems have been examined.[4–8] Hence, it is necessary to know the hydrodynamic characteristics of a two-phase gas-electrolyte fluid through a fixed bed. The hydrodynamic characteristics of two-phase flow through a fixed bed, in which the electrolyte is the continuous phase, have been examined.[9–12] The following factors can influence the hydrodynamic characteristics: electrolyte velocity, particle size constituting the bed, gas hold-up, geometrical characteristics of the column and so on. In this work the hydrodynamic characteristics of a two-phase gas-electrolyte flow through a fixed bed of spherical particles, where gas phase was generated electrochemically, have been examined. During the electrolysis of water, oxygen evolves on the anode, whereas hydrogen evolves on the cathode. At the same time, if the cathode surface is developed, the formation of the nuclei for the development of hydrogen bubbles rising will occur on the whole surface. In such a way, the obtained

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bubbles are smaller than in the case when gas is introduced into the electrolyte through a distributor.

EXPERIMENTAL

The experimental set-up is schematically presented in Fig. 1. The vertical cylindrical column with an inner diameter of 45 mm and a height of 320 mm is made of Plexiglas. A platinum net is built in the column which serves simultaneous by as the current feeder, the bed bearer and the electrolyte distributor. The wire, which makes up the net, is 0.16 mm thick, and the number of meshes per square centimetre is 378. The total surface of the cathode is $3.154 \quad 10^{-2}$ m^2.

The height of the packed bed of particles through which the pressure drop is measured is 50 mm. The anode is placed above the bed, as it is marked in Fig. 1. It is made of a lead sheet having a cylindrical form with an outer diameter of 45 mm, height 30 mm and thickness 1 mm. The distance between the cathode and the anode is 63 mm. Because of the considerable distance between the cathode and the anode, a $3 \quad 10^3$ mol/m^3 solution of sulphuric acid was used, as it has the greatest electric conductivity. Electric conductivity of this solution is 73.88 S/m.[13] The power supply of direct current is a rectifier of alternating current equipped with voltmeter and ampermeter.

Three fractions of glass particles of 1.5 mm, 2 mm and 3 mm diameter were used as the dispersed phase.

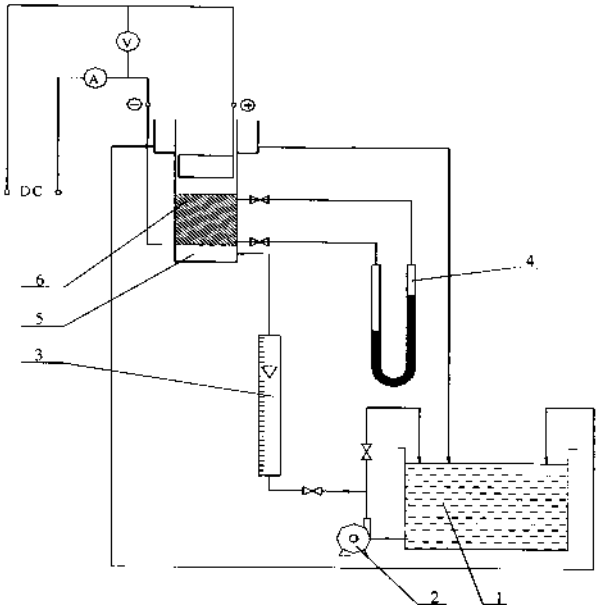

Fig. 1. Experimental set-up: 1 – reservoir; 2 – centrifugal pump; 3 – flowmeter; 4 – differential manometer; 5 – column; 6 – fixed bed.

All the experiments were performed at ambient temperature. The total volume of liquid phase for all the experiments was the same, $5 \quad 10^{-3}$ m^3. The liquid was transported by a centrifugal pump from the reservoir through a flowmeter into the column from the bottom upwards, so that the electrolyte passes through the platinum net cathode carrying the gas bubbles evolving on the cathode along with it. The liquid phase over flows from the top of the column back to the reservoir. Both the column and the reservoir are open to the atmosphere so that the gas phase, which is held-up in the liquid, goes freely to the atmosphere.

The pressure drop through the bed was measured by a differential "U-tube" manometer filed with carbon tetrachloride, the density of which is 1586.7 kg/m^3.

The experiments were performed in the following way: firstly, in the column above the platinum net electrode, *i.e.*, bed support, 0.158 10^{-3} m^3 of glass particles were introduced. The electrolyte, 3 10^3 mol/m^3 H_2SO_4, was put into the reservoir. Then the centrifugal pump was switched on and when a constant flow was achieved, the pressure drop through the bed was measured. Then both the power supply and a chronometer were switched on. The evolution of hydrogen bubbles at the platinum causes an increase in the pressure drop through the bed. This icrease was monitored every two minutes until the pressure drop reached a constant value. The centrifugal pump is remained still on, pumping the electrolyte, in order to remove the remaining bubbles from the bed. The system was then ready for the next experiment.

RESULTS AND DISCUSSION

Effect of liquid velocity on pressure drop in bed

The effect of liquid velocity on the pressure drop was examined in such a way that the velocity was varied in the interval of 0.00482 m/s – 0.0383 m/s, at a constant current density (100 A/m^2). Glass particles of 3 mm diameter were used. The results are shown in Figs. 2 – 5.

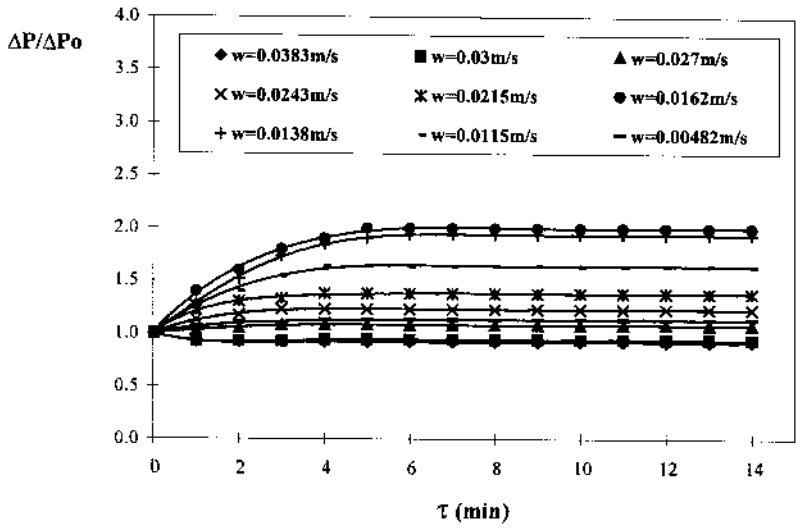

Fig. 2. Dependence of the relative pressure drop on time for different liquid flow velocities (i = 100 A/m^2; d = 3 mm).

From Fig. 2 it can be seen that the relative pressure drop increases (by w_1 = 0.0162 – 0.03 m/s) with decreasing liquid velocity. The curves showing the change p/p_0 against time have two parts: the first part corresponds to the non-steady state in which p/p_0 increases with time, and the second part corresponds to the steady state in which the pressure drop changes only slightly with time and can practically be considered as constant. The non-steady state only lasts several minutes and is shorter when the velocities of the liquid are higher. The curves, which show the dependence of p/L against time, also show the steady state, which indicates that the pressure drop along the height of the bed, after a certain time, obtains constant values. The pressure drop per unit bed height mainly depends on the liquid velocity and p/L also increases with in-

creasing liquid flow (Fig. 3). The time change of pressure with parallel gas-liquid flow through a fixed bed was examined by Krieg *et.al.*[14] For the same size of the glass particles, which make the bed (3 mm), they noticed similar changes of the pressure as were observed in this work.

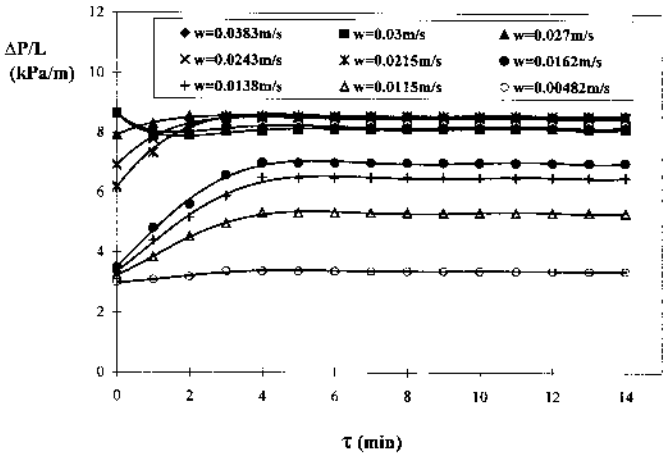

Fig. 3. Pressure drop per unit height of the bed *vs*. time for different liquid velocities (i = 100 A/m^2; d = 3 mm).

The obtained results can be explained in the following way: by switching on the current circuit, a constant current density of 100 A/m^2 passes through the liquid which leads to the electrolysis of water and the evolution of hydrogen forming a two-phase fluid which consists of a continual liquid phase and gas bubbles. The certain quantity of the evolved hydrogen is carried by the liquid flow through the fixed bed and the rest of the hydrogen is captured in the channels formed by glass particles. The hydrogen which remains in the porous bed causes a decrease of the cross-sectional area through which the liquid may flow, increasing the liquid velocity resistance which is manifested by an increased pressure drop. The greater the quantity of the hydrogen, trapped in the porous bed, the bigger the fluid flow resistance. At the same time, this results in an increase of the pressure drop through the bed, with increasing volume of hydrogen in the bed. From Fig. 2, it can be seen that the content of hydrogen in the bed increases only in the first few minutes (non-steady state). For each liquid flow rate, a maximum quantity of hydrogen can be retained in the bed and this quantity defines the plateau value of the pressure drop. It was noticed that with increasing liquid velocity, the change of the relative pressure drop through the fixed bed decreases. This means that at lower liquid velocities in the bed, there is a greater hold-up of hydrogen bubbles, which brings about a pressure drop increase. At higher liquid velocities, buoyancy forces increase the transport of hydrogen bubbles from the bed. Higher liquid velocities mean higher kinetic energy, and the transport carrying of hydrogen bubbles from the bed is easier, so the resistance to the liquid flow decreases, *i.e.*, under these conditions the change of the relative pressure drop is smaller. It can also be seen that at higher liquid velocities, the relative pressure drop does not change significantly with time. At these velocities, the appearance of fluidization has been remarked, which leads to a decrease in the total resistance to liquid

flow through the bed, and, thus, the fluidization state brings about a decrease of the pressure drop. This phenomenon was described by Stanković and coworkers,[15] and they proved that at the fluidization state the relative pressure drop is less than one.

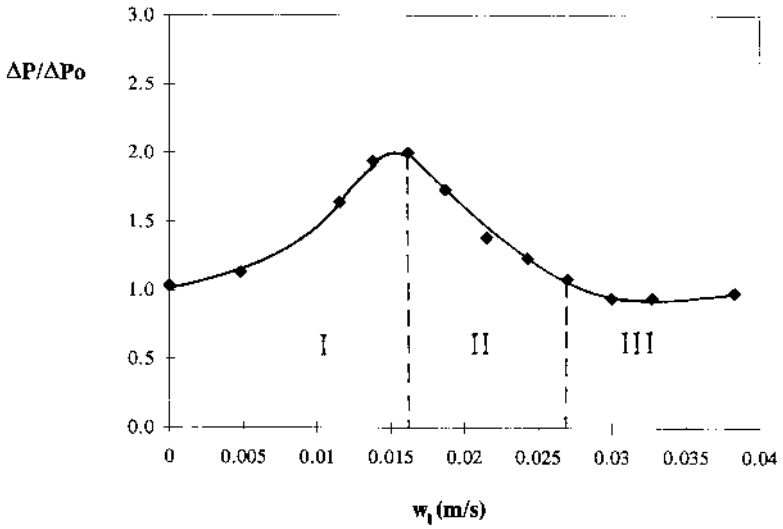

Fig. 4. Relative pressure drop values *vs*. liquid velocity (i = 100 A/m^2; d = 3 mm).

The dependence of the relative pressure drop against the liquid velocity is shown in Fig. 4. The presented values for $p/\ p_0$ are those corresponding to the steady state relative pressure drop obtained at different liquid velocities.

It is clear that the relative pressure drop increases with increasing velocity of the two-phase fluid flow in the fixed bed (1st region). This indicates that, with low liquid velocities (w_1 = 0.00482 – 0.0162 m/s), coalescence of the bubbles is reduced. When the liquid velocity is 0.0162 m/s, the pressure drop in the bed is the highest. After this value, the pressure drop in the bed decreases (2nd region). Namely, the formation of channels through the bed occurs at velocities higher than 0.0162 m/s. The two-phase fluid flows mainly through these channels and, under these circumstances, the pressure drop through the bed decreases. At a liquid velocity of about 0.027 m/s there is no change of the pressure drop with the gas phase, indicating that liquid velocity approached the minimum velocity fluidization.

In the third region, fluidization in the bed exists, and the higher the liquid velocity is, the more intensive is the fluidization of the particles and the relative pressure drop values is less than one.

A similar behaviour of the pressure drop with increasing liquid velocity was observed by Chern and coworkers.[9] Namely, in their work, the pressure drop decreases at a liquid velocity of 0.02 m/s, when the bed transforms to a fluidization state. The slightly lower minimum fluidization velocity in their work is the consequence of a smaller density of the particles constituting the bed, made of polypropylene and polyethylene, their size as well as the countercurrent flow of the gas and liquid (semi-fluidization).

The effect of current density on the pressure drop in the bed

Based on Faraday's laws, the connection between the mass of evolved gas in water electrolysis and the applied current, is given by

$$m = \frac{M}{zF} I \tag{1}$$

If the mass of the evolved gas, obtained during electrolysis, is calculated per unit of time, *i.e.*, a second, then a mass flow rate of the gas is obtained, which, if expressed through continuity equation, is the following:

$$G = Q_{\mathrm{g}} = w_{\mathrm{ig}} A_{\mathrm{c}} \varepsilon_{\mathrm{g}} \tag{2}$$

Equating Eqs. (1) and (2), the velocity of gas evolution may be expressed as folows

$$w_{\mathrm{g}} = \frac{IM}{zF \varepsilon_{\mathrm{g}} A_{\mathrm{c}}} = \frac{iM}{Fz \varepsilon_{\mathrm{g}}} \tag{3}$$

On the other hand, the relative pressure drop p/p_0 represents the measured value of the pressure drop with gas evolution p, relative to the pressure drop p_0, in the absence of the gas phase, *i.e.*, when no current (I) passes through the column.

The effect of the current density on the pressure drop was examined ty performing experiments in which the change of the pressure drop with time was measured at different current densities (32 A/m^2 – 190 A/m^2), at a constant liquid phase velocity (w_1 = 0.0162 m/s). Glass particles of 3 mm diameter were used. The obtained results are shown in Fig. 5, from which it can be seen that the curves representing the dependence of p/p_0 against time have a similar form to those obtained during the investigation of

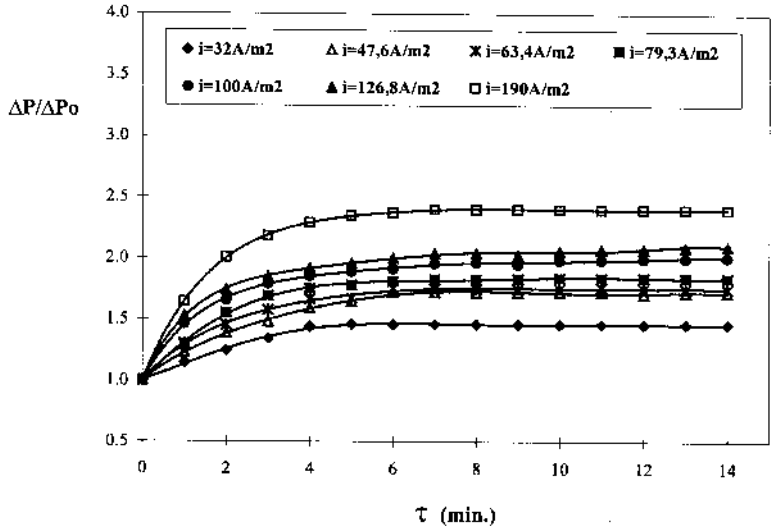

Fig. 5. Dependence of the relative pressure drop on time for different current densities (w_1 = 0.0162 m/s; d = 3 mm).

the influence of the liquid velocity on the pressure drop. From the shape of these curves a steady and non-steady part can be recognised. The steady state parts of the curves are almost horizontal and parallel to each other, and the plateau is translatory moved towards the vertical axis. The non-steady state parts of these curves have a higher inclination at higher current densities.

With increasing current density, the change of the relative pressure drop also increases. This observation can be explained in the following way: after switching on the power supply, a reduction of H^+-ions on the cathode occurs, and a two-phase gas-liquid flud forms in the continuous liquid phase. As a result of gas evolution, which in the form of bubbles is captured in the fixed bed, the resistance to liquid flow increases, and at the same time there is an increase of the relative pressure drop. In other words, the content of gas in the bed increases with time, leading to an increase of pressure drop. When the maximum content of the gas in the bed is reached, *i.e.*, when the steady state period is obtained, the change of the relative pressure drop reaches a maximum value which no longer changes with time any more. On the basis of electrochemical principles, it is well known that there will be a more intensive hydrogen evolution on the cathode at a higher current density. The greater quantities of evolved hydrogen lead to a faster saturation of the fixed bed with gas bubbles during the initial period, leading to an increase in the slopes of the p/p_0 – time curves. The increase of the relative pressure drop under steady state conditions can be attributed to an increased gas hold-up due to the higher gas evolution rate at higher current densities. This increase was observed over the whole range of applied current densities. Since the presence of bubbles in the fixed bed obstructs the liquid flow, the increased gas hold-up will directly influence the increase in the relative pressure drop.

In Fig. 6 the dependence of p/p_0 against current density under steady state conditions is shown more clearly.

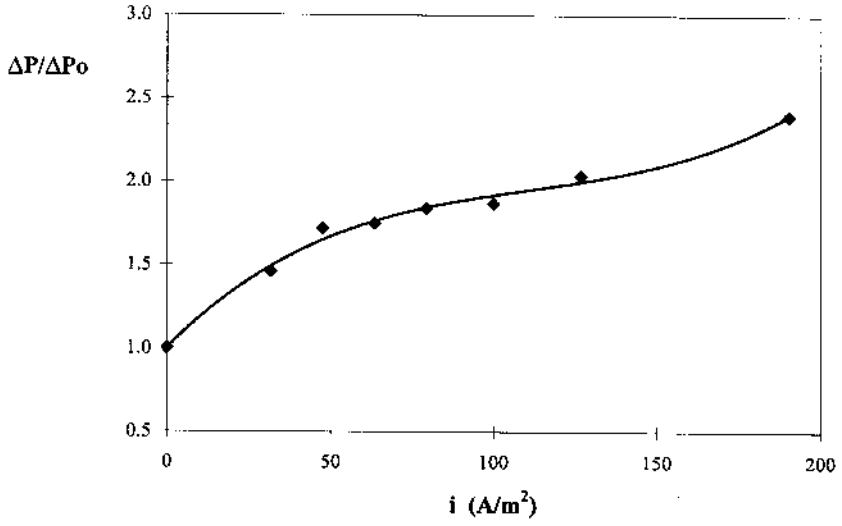

Fig. 6. Relative pressure drop *vs*. current density (w_l = 0.0162 m/s; d = 3 mm).

Lower current densities correspond to a higher change in the pressure drop to reach a certain value, after which the relative pressure drop remains almost constant. Similar behaviour was observed by Chern and coworkers[9] in a semi-fluidized bed, for the same range of both gas and liquid.

The influence of particle size on the pressure drop

Three experimental series were carried out, in which current density was kept constant (100 A/m^2), but the liquid velocity was varied, 0.0187 m/s in the first, 0.0243 m/s in the second and 0.03 m/s in the third series.

It was found that the relative pressure drop increases with increasing diameter of the particles. This indicates that, at a flow velocity between 0.0187 – 0.0243 m/s, the channels are larger in a packed bed formed from larger particles. The coalescence of smaller hydrogen bubbles in such channels is more evident than in the narrower channels which are formed in a fixed bed of smaller particles. Besides, the two-phase fluid is distributed in a better way when it passes through a bed of smaller particles, which decreases the total resistance to liquid flow. For these reasons the change of the pressure drop is higher in a bed consisting of larger particles. At fluid velocities higher than 0.03 m/s, fluidization of the particles occurs, changing the image of the investigated phenomenon. Fluidization of a bed consisting of smaller particles commences at lower liquid velocities. With the transformation of the bed, the total resistance to liquid flow through the bed decreases.

The influence of particle size on the relative pressure drop is presented Fig. 7 in which a dependence of $p/\ p_0$ (steady state values) on the diameter of the praticles is shown.

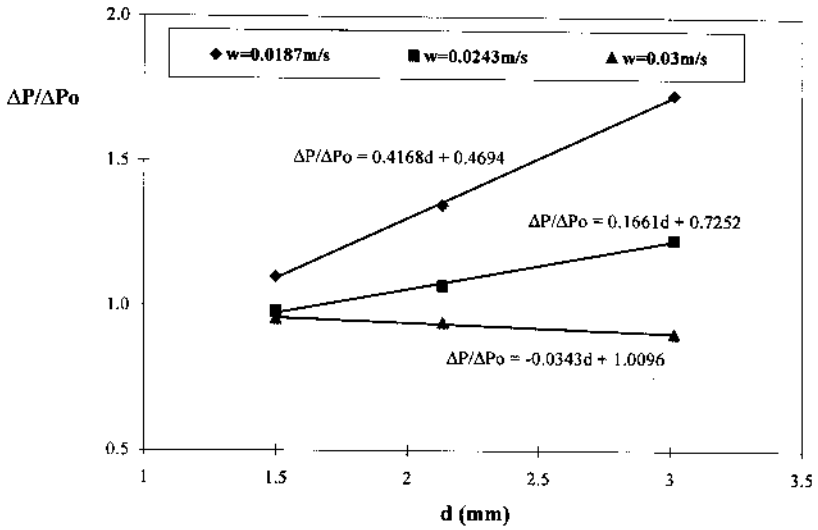

Fig. 7. Dependence of the relative pressure drop on particle diameter for different liquid velocities (i = 100 A/m^2).

The linear relationships between the relative pressure drop and the particle size for different liquid flow velocities can be seen in Fig. 7. It can also be seen that the lines

corresponding to velocities of 0.0187 m/s and 0.0243 m/s have positive slopes, indicating that with these liquid velocities, the relative pressure drop increases with increasing particle diameter. With a further increase of the liquid velocity (0.03 m/s) the slope becomes negative, indicating that at these velocities the particle size does not play a great role in the change of the pressure drop. The negative value of the slope, which was found for the velocity $w_1 = 0.03$ m/s, is the consequence of the occurrence of fluidization at this liquid velocity, where the gas phase does not increase the relative pressure drop through the bed. Stanković and coworkers[15] also remarked on a similar effect.

The friction factor

The friction factor *f*, introduced by Ergun[16] is defined by the following equation:

$$f = \frac{p}{L}\frac{d}{(w)^2}\frac{^3}{(1 \quad)} \tag{4}$$

The friction factor is a function of the Reynolds number, and when the current is off, *i.e.*, there is no gas phase in the bed, Song and coworkers[12] modified the Ergun equation[16] for the flow of a monophase through a fixed bed and defined *f* in the following way:

$$f = 0.583 + \frac{33.3}{Re} \tag{5}$$

when the Reynolds number is calculated on the basis of the physical properties of the liquid:

$$Re = \frac{d \ w_1}{} \tag{6}$$

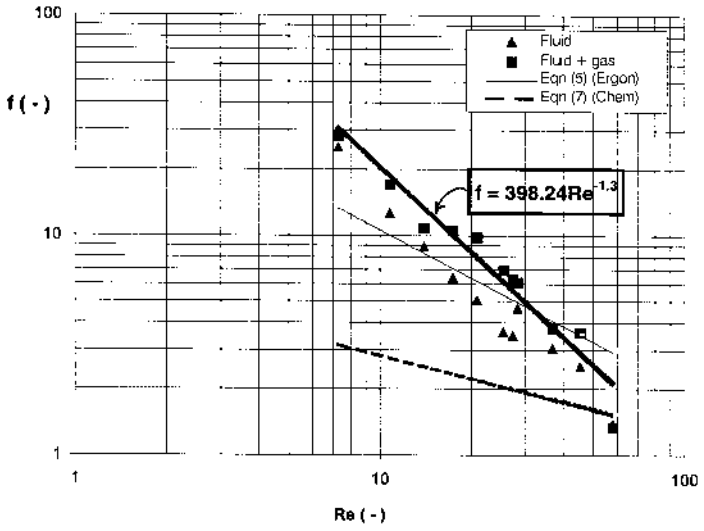

Fig. 8. Friction factor *vs*. Reynolds number ($i = 100$ A/m^2; $d = 3$ mm).

Chern and coworkers[10] suggested the following relation for two-phase gas-liquid flow upwards through a fixed bed:

$$f = 0.1075 + 6.359\, Re^{-0.368} \quad (7)$$

Here, the Reynolds number is also calculated from Eq. (6).

Experimental results, obtained in this work, have been compared to relationship (5) and (7).

The friction factor is calculated from the experimental results based on Eq. (4), and depends on the pressure drop per unit bed height, the particle size, the bed porosity, the velocity of the liquid and its density. In Fig. 8 the influence of the liquid velocity, *i.e.*, of the Reynolds number, on the friction factor for a current density of 100 A/m^2 and a particle diameter of 3 mm.

From Fig. 8, it can be seen that the dependence of the friction factor upon the Reynolds number for the liquid alone matches the Ergun Eq. (5). For two-phase gas-liquid flow, where the liquid is the continuous phase, the experimental friction factor values are greater than the values obtained on the basis of Eq. (7), which was proposed by Chern and coworkers.[10] The deviations of the experimental friction factors from the calculated values are greater at lower liquid velocities, whereas at higher velocities a satisfactory good agreement was achieved with Eqs. (5) and (7). This can be explained by the influence of the walls of the column during two-phase fluid flow. Namely, the diameter of the column in our experiments was 45 mm whereas in the work of Chern it was 76.2 mm.

CONCLUSIONS

The obtained results show the following conclusions:

1. A gas phase evolves on the electrode in the liquid flowing through the fixed bed. This contributes to the pressure drop increase. The gas bubbles decrease the existing liquid hold-up attaching to the particles and bridging the channels in the bed thus obstructing the liquid flow.

2. The relative pressure drop increases with time reaching a constant value after a certain time due to an increase of the gas hold-up with time.

3. The intensity of the gas evolution, *i.e.*, the operating current density, strongly influences the relative pressure drop in such a way that the relative pressure drop increases with increasing current density.

4. The relative pressure drop decreases with increasing liquid velocity (for w_l = 0.00162 – 0.03 m/s). Such an effect results in an increased buoyancy force enabling the bubbles to detach from the particles whereby they are transported out of the bed with the liquid flow.

5. On achieving the fluidized state, the relative pressure drop becomes equal to one or slightly less indicating that in a fluidized bed the gas phase has no remarkable influence on the pressure drop.

6. The relative pressure drop increases linearly with increasing particle size.

7. The friction factor decreases linearly with increasing modified Reynolds number in the logarithmic coordinate system in the range of the investigated hydrodynamic conditions.

LIST OF SYMBOLS

A_c – total surface of the cathode, m^2
d – diameter of a particle, m
F – Faraday's constant, 96500 A s mol^{-1}
f – friction factor
G – mass flow, kg s^{-1}
I – current, A
i – cathode current density, A m^{-2}
L – height of the bed, m
M – molar mass of the evolved gas, kg mol^{-1}
m – mass of gas obtained during water electrolysis, kg
$_P$ – pressure drop through the bed, kPa
p_0 – initial pressure drop through the bed without the gas phase, kPa m^{-1}
$\Delta p/\ p_0$ – relative pressure drop
Q – volume flow of gas, m^3 s^{-1}
Re – liquid Reynolds number
w_l – liquid superficial velocity, m s^{-1}
w – superficial mono-phase fluid velocity (liquid velocity), m s^{-1}
w_{ig} – velocity of gas evolution, m^3 s^{-1} m^{-2}
z – number of exchanged electrons

Greek letters

– bed porosity
– liquid viscosity, Pa s
$_g$ – gas density, kg m^{-3}
– liquid density, kg m^{-3}
– time realisation of the electrochemical process, s

И З В О Д

ХИДРОДИНАМИЧКЕ КАРАКТЕРИСТИКЕ ДВОФАЗНОГ ТОКА ГАС-ТЕЧНОСТ НАВИШЕ КРОЗ ПАКОВАНИ СЛОЈ СФЕРНИХ ЧЕСТИЦА

СНЕЖАНА М. ШЕРБУЛА и ВЕЛИЗАР Д. СТАНКОВИЋ

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Испитиване су хидродинамичке каратеристике трофазног система у цилиндричној колони у којој је гасовита фаза генерисана електрохемијски. Двофазни флуид, гас–течност у коме је течна фаза континуална, струји кроз пакован слој од стаклених куглица. Испитан је утицај брзине течности, брзине гаса и пречника стаклених куглица

на пад притиска кроз паковани слој. Нађено је да са порастом брзине течности (w_l = 0,0162 – 0,03 m/s) опада релативни пад притиска кроз паковани слој. С порастом густине струје повећава се пад притиска, јер веће количине гаса заостају у пакованом слоју. Поред овога, нађено је да се са смањењем пречника стаклених честица смањује релативни пад притиска. Добијена је зависност модификованог коефицијента трења од Рејнолдсовог броја за монофазни и двофазни ток.

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