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Influence of Two-Phase Liquid-Gas Flow on Permeate Flux through Ceramic Membranes

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Abstract: The impact of two-phase flow onto the flux of the permeate in microfiltration of two sorbents-Bentonite and Lewatit S1468-with and without adsorbed zinc, was examined. Two different types of membrane, single-and multi-channel, based on α-Al_2O_3, were used. In single-channel membrane application of aeration increased the permeate flux by 13 and 29% without adsorbed zinc and by 22 and 25% with adsorbed zinc for Bentonite suspension and by 4 and 5.5% both with and without adsorbed zinc for Lewatit S1468 compared to single-phase (without aeration) flow at 1.4 and 2.2 m s^{-1} of the gaseous phase rate, respectively. In multi-channel membrane the permeate flux increased not significantly without adsorbed zinc and by 22 and 25% with adsorbed zinc for Bentonite and by 12 and 13% without adsorbed zinc and not significantly with adsorbed zinc for Lewatit S1468 at 0.4 and 1.1 m s^{-1} of the gaseous phase rate, respectively.

Keywords: Ceramic Membrane, Bentonite, Lewatit S1468, Two-Phase Liquid-Gas Flow

Introduction

A liquid-gas two-phase flow is considered to be an effective way of overcoming the concentration polarization or gel layer formation in membrane separation (Lee *et al*., 1993; Li *et al*., 1998; Mercier *et al*., 1997). During the process the supplied gas (air, nitrogen, methane, etc.) is directly injected into the feed of separated substances at certain pressure and speed. The gas is released from the retentate in the storage tank afterwards. The results of published studies (Sur and Cui, 2001; Cabassud *et al*., 1997; 2001; Cui and Wright, 1996; Cui *et al*., 1997; Laborie *et al*., 1998; Cui and Taha, 2003; Cui *et al*., 2003; Campos and Guede de Carvalho, 1988; Smith *et al*., 2005) indicate a significant increase in the intensity of permeate flux by using two-phase flow in a variety of applications such as membrane bioreactors, fermentation products processing, or separation of macromolecular substances, e.g., Sur and Cui (2001) in liquid-gas two-phase microfiltration experiments of yeast suspension carried out by a multi-tubular membrane.

Three to five flow regimes of co-current liquid-gas flow in a vertical tube are distinguished by different authors. Taitel *et al*. (1980; Cabassud *et al*., 2001) introduce three basic regimes-bubble, slug and annular flows, whilst (Whalley, 1990; Cui and Taha, 2003) add churn flow. The formation of any of the flow regimes or a change of the regime type is given by the pipe diameter, flow rates of gas and liquid, their physical properties, the geometry and inclination of membrane channel, interfacial tension, etc. (Cui and Taha, 2003). A simple characteristic of the regimes follows (Cabassud *et al*., 2001; Campos and Guede de Carvalho, 1988; Whalley, 1990; Cui *et al*., 2003):

- Bubble flow can be described as gas bubbles dispersed in the form of small bubbles (less than 60% of the tube diameter). It is formed at low gas flow rates and low to medium liquid flow rate
- Slug flow can be described by alternation of gas slugs (bubbles about 60% of the tube diameter) with bullet-shaped nose and liquid slugs. They may also contain smaller bubbles inducing flow turbulence. It is formed at medium flow rates of gas and liquid
- Churn flow can be described by irregular shape and miscellaneous size. It is formed at medium liquid flow rates and higher gas flow rates
- Annular flow can be described by continuous flow of gaseous phase in the pipe centre and containing liquid droplets. The liquid phase flows on the wall. It is formed at high flow rates of gas and liquid.

Table 1. Basic division of flow regimes depending on the injection factor

Injection factor	Flow regime
$\varepsilon<0.2$	Bubble flow
$0.2<\varepsilon<0.9$	Slug flow
$\varepsilon>0.9$	Annular flow

Bubble and slug flow are the most probable flow regimes in two-phase flow modules at low applied gas and liquid flow rates. The flow regime can be estimated by the injection factor, specified as (Cabassud *et al*., 2001):

$$\varepsilon = \frac{u_g}{u_l + u_g} \quad (1)$$

In Equation 1, u_g and u_l are the flow rates of gas and liquid, respectively, determined as if circulation of each phase in the pipe was alone. The basic division is shown in Table 1 (Cabassud *et al*., 2001).

This study is concentrated on comparison of liquid-gas two-phase flow membrane filtration (microfiltration) of adsorbents-Bentonite-clay based on montmorillonite- and Lewatit S1468-a copolymer based on styrene-divinylbenzene-without and with adsorbed zinc through a single-and a multi-channel membrane. The results are also compared to the results of single-phase (no gas inlet) flow under the same conditions published previously. The change of hydrodynamic conditions by changing the inner membrane configuration (1 or 7 channels) has a significant influence on the flux as mentioned above. The previous studies concentrated either on microfiltration of organic substances (yeast, dextran, bacteria, etc.) or on different membrane modules or on theoretical modelling of the liquid-gas (-solid) flow patterns. This article focuses on comparison of one inorganic natural substance-Bentonite from Slovakia and a commercially produced organic substance-Lewatit S1468, that have not been studied in two-phase liquid liquid-gas flow microfiltration system.

Materials and Methods

Bentonite

Bentonite (Gemerská nerudná spoločnosť, a.s., Hnúšťa, Slovakia) is a layered structure clay composed of silicates. Montmorillonite (about 75%w.) is the main component that is accompanied by Fe_2O_3 (max. 3.5%w.) and Al_2O_3 (18%w.). The bentonite used in the study was natural Bentonite activated using NaOH, dried and milled (commercially sold as Bentovet K). It is a very pale off-white colored powder that swells well in water. Its ion exchange capacity and sorption capacity are high. Modified Bentonite is used in the ceramics, foundry and building industries, in removal of xenobiotics from liquids, in ion exchange, etc. The particle size distribution of Bentonite is presented in Fig. 1 (device used for determination was Mastersizer 2000, UK).

Lewatit S1468

Lewatit S1468 (Bayer Chemicals AG, Leverkusen, Germany) is a resin based on a sty-rene-divinylbenzene copolymer. It is a strongly acidic cation exchanger. It consists of same size particles of 0.6 mm. Lewatit S1468 is highly stable both chemically and osmotically. In this study finely ground form was used. It is used in chemical industry for catalysis, demineralization and water treatment and purification. The particle size distribution of Lewatit S 1468 is presented in Fig. 2 (device used for determination was Mastersizer 2000, UK).

Membranes

An asymmetrical inorganic membrane was used as single-channel membrane. It is based on α-Al_2O_3 (produced by Pall Corporation, USA). The inside surface of ZrO_2 is forming the active layer of the membrane. A 25 cm in length, 7 mm in inner diameter and 10 mm in outer diameter membrane was used in the experiments. The manufacturer indicates the average pore size of 100 nm. The area of 48.38 cm^2 is forming the active membrane area. An asymmetrical inorganic membrane was used as multi-channel membrane. It is based on α-Al_2O_3 (produced by Pall Corporation, USA). The inside surface of ZrO_2 is forming the active layer of the membrane. It contains 7 channels of (6 in mm diameter each). The area of 329.7 cm^2 is forming the active membrane area. The manufacturer indicates the average pore size of 120 nm. The intensification effect of the membrane interior layout on the flux through the membrane divided into 7 channels has been considered in an increase of the flux and turbulence of the flow. The length of the membrane was 25 cm. Both the single- and multi-channel membranes were saturated in demineralised water before the experiments.

Microfiltration

The scheme of the device used in the experiments is presented in Fig. 3.

In the experiments the mixture of adsorbent particles in different concentrations dispersed throughout demineralised water was pumped from a 4-litre-volume reservoir by a pump with frequency converter. The tubular ceramic membrane microfilter was placed in a vertical membrane module. The membrane modules for single- and multi-channel membranes were placed in two parallel modules due to their different parameters. Each segment was used separately. The feed flowed into the membrane. The gas was injected into the feed prior to the microfilter through a pipe of 1.3 cm in diameter direct. The stream of permeate from the module was gathered in a wide neck vessel put on electronic scales connected to a personal computer.

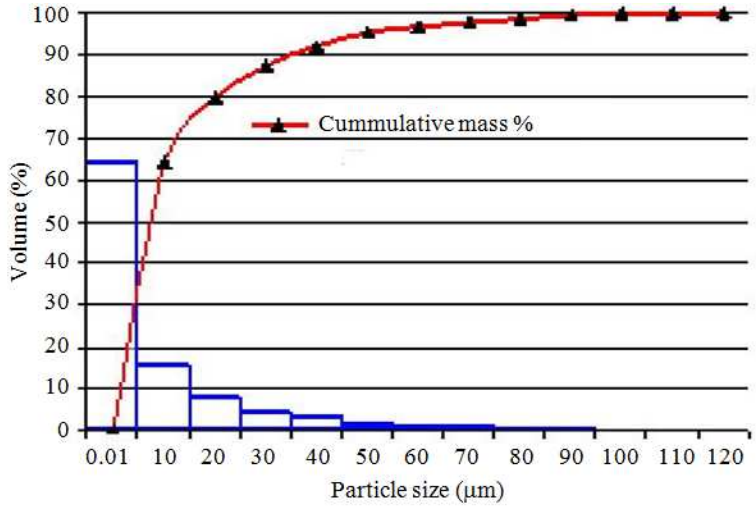

Fig. 1. Particle size distribution of Bentonite

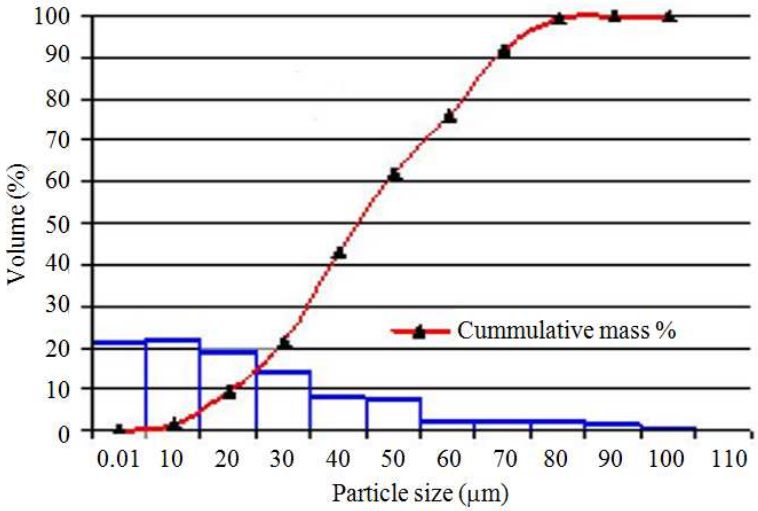

Fig. 2. Particle size distribution of Lewatit S1468

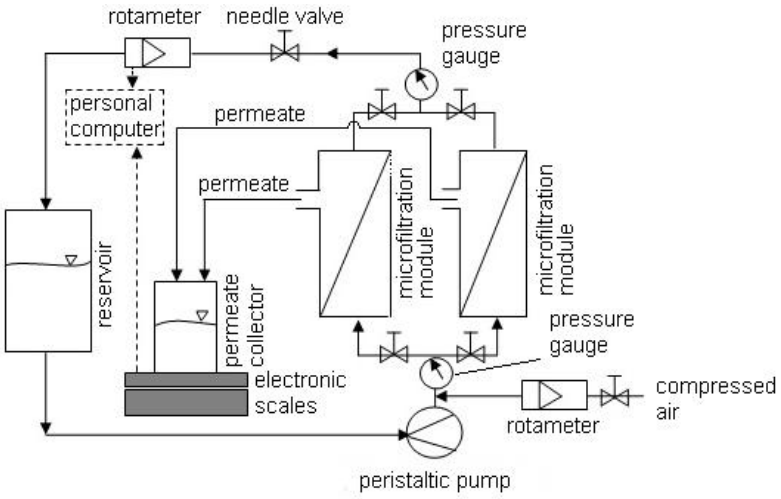

Fig. 3. Scheme of microfiltration device

Table 2. Process parameters maintained during the experiments

Memb-rane	Δp, kPa	c_a, g L^{-1}	c_{Zn}, mg L^{-1}	u_l, m s^{-1}	u_g, m s^{-1}	ε
single-channel	50	5	0	1,4	1,4	0,50
				2,2	2,2	0,50
	50	5	250	1,4	1,4	0,50
				2,2	2,2	0,50
multi-channel	50	5	0	0,7	0,4	0,36
				1,1	1,1	0,50
	50	5	250	0,7	0,4	0,36
				1,1	1,1	0,50

The stream of retentate moved out back into the reservoir. There was a pressure sensor put behind the membrane module. It measured the pressure after the membrane. The pressure was adjusted by a control valve. There was a flow metre placed in the retentate partin order to measure the flow rate in the system.

In these experiments the suspensions of the adsorbents Bentonite and Lewatit S1468 (used separately) were mixed with the solution of zinc sulphate (where appropriate) 24 h before the microfiltration experiments. The suspensions with zinc were stirred for 12 h by a magnetic stirrer. This suspension was used for microfiltration experiments while the experiments were carried out under constant pressure difference 50 kPa and suspension concentration 5 g L^{-1}. Demineralised water was added into the microfiltration apparatus and ran for 5 min before the adsorbent suspensions with zinc were added so that the final concentrations in the feed were according to the process parameters defined in Table 2. The process parameters were selected in accordance with the construction characteristics of the microfilter so that it was possible to compare the influence of two-phase flow onto the membrane separation. Measurements for both types of membranes were carried out with a constant liquid-gas two-phase flow with selected parameters.

Results

In the Fig. 4 and 5 the Bentonite suspension microfiltration through single-channel membrane without and with zinc adsorbed, respectively, are shown.

In the Fig. 6 and 7 the Lewatitu S1468 suspension microfiltration through single-channel membrane without and with zinc adsorbed, respectively, are shown.

In the Fig. 8 and 9 the Bentonite suspension microfiltration through multi-channel membrane without and with zinc adsorbed, respectively, are shown.

In the Fig. 10 and 11 the Lewatitu S1468 suspension microfiltration through multi-channel membrane without and with zinc adsorbed, respectively, are shown.

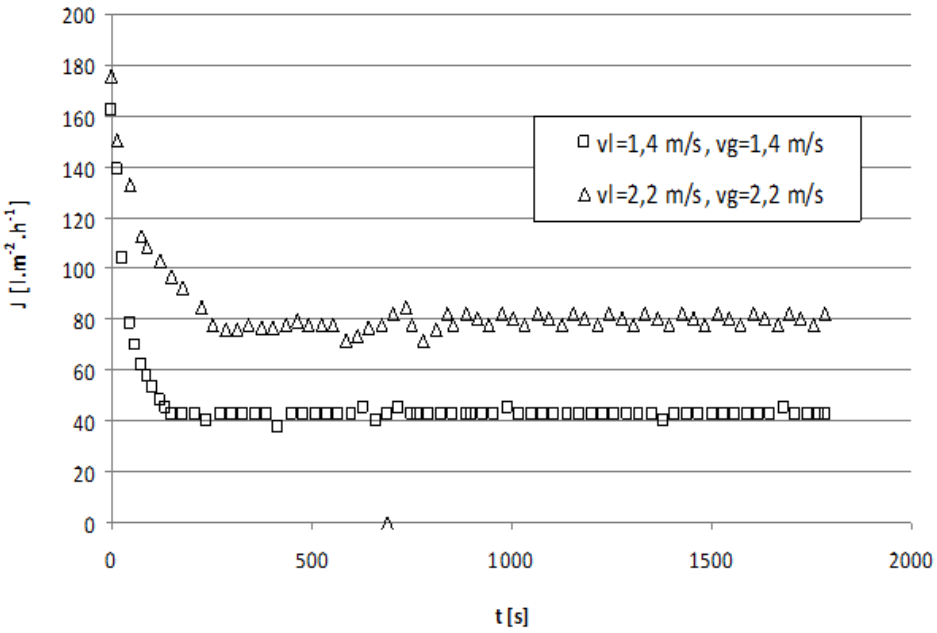

Fig. 4. Permeate flux through single-channel membrane depending on time for Bentonite suspension microfiltration with two-phase flow at adsorbent concentration 5 g L^{-1} and transmembrane pressure 50 kPa without zinc solution

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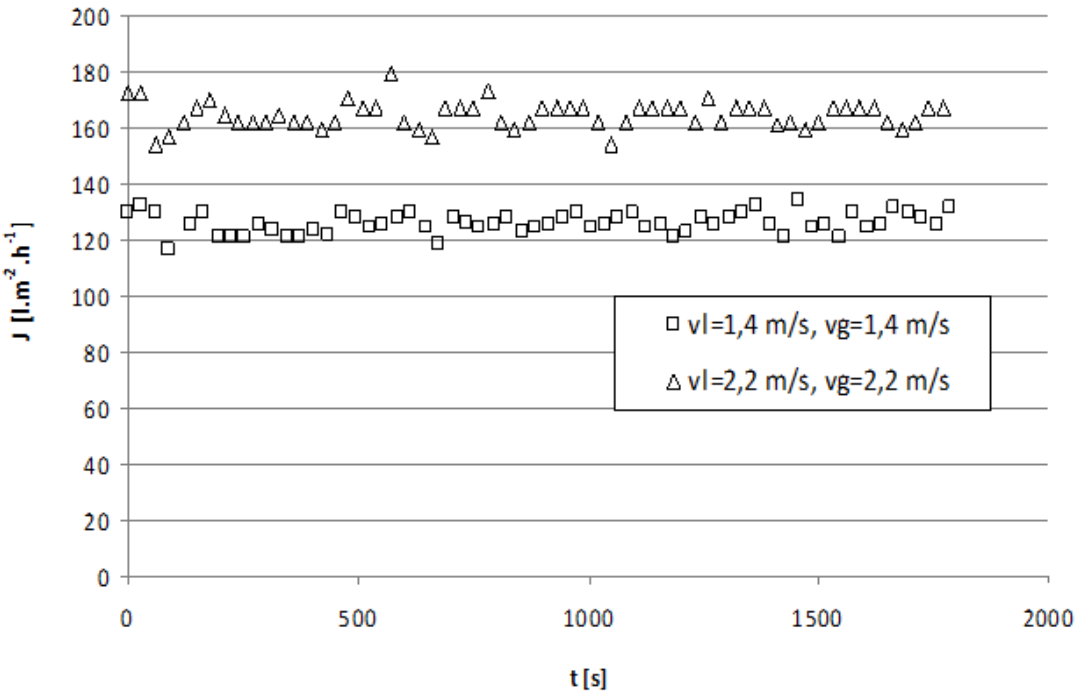

Fig. 5. Permeate flux through single-channel membrane depending on time for Bentonite suspension microfiltration with two-phase flow at adsorbent concentration 5 g L^{-1} and transmembrane pressure 50 kPa with 250 mg L^{-1} zinc solution

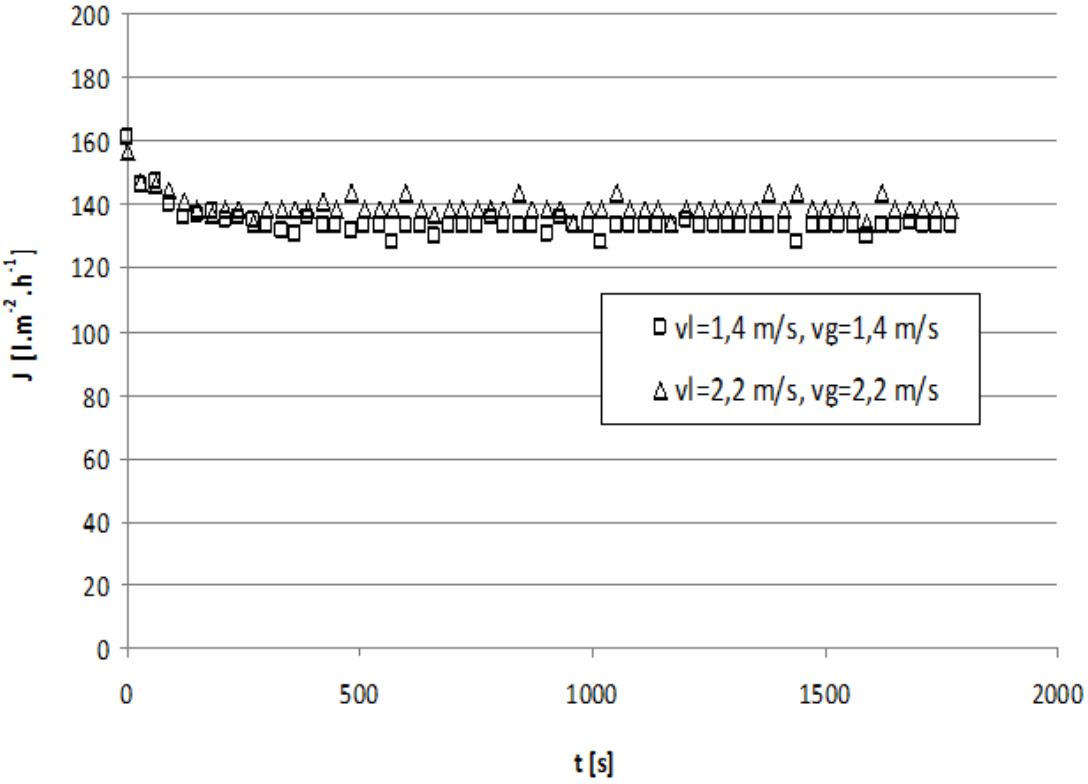

Fig. 6. Permeate flux through single-channel membrane depending on time for Lewatit S1468 suspension microfiltration with two-phase flow at adsorbent concentration 5 g L^{-1} and transmembrane pressure 50 kPa without zinc solution

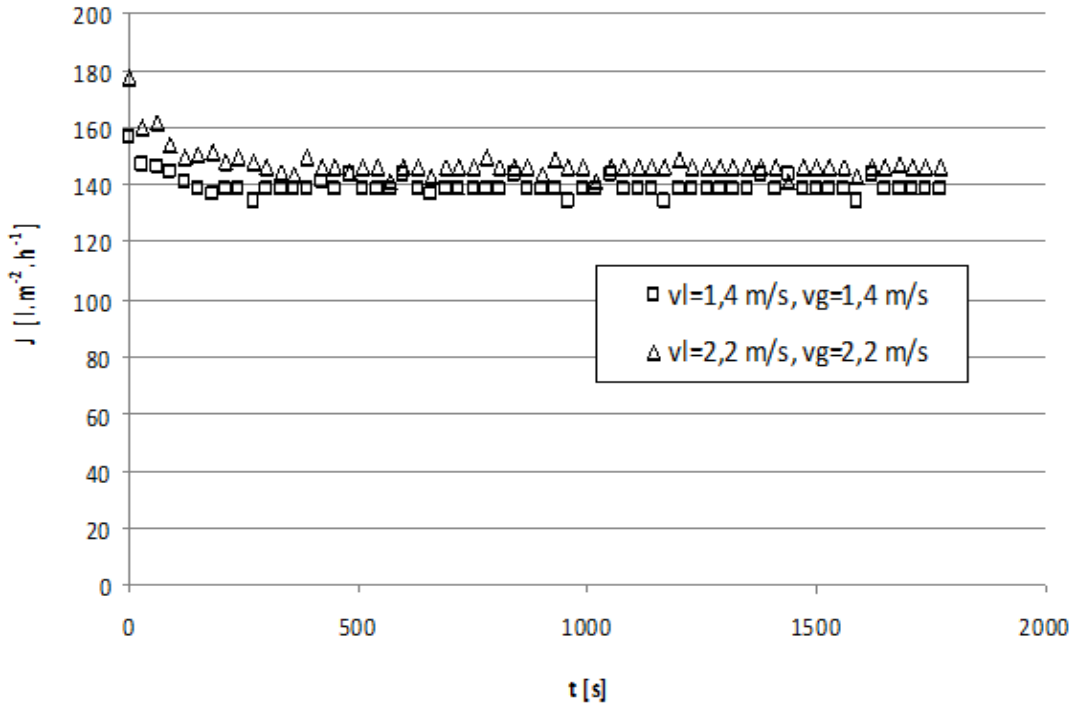

Fig. 7. Permeate flux through single-channel membrane depending on time for Lewatit S1468 suspension microfiltration with two-phase flow at adsorbent concentration 5 g L^{-1} and transmembrane pressure 50 kPa with 250 mg L^{-1} zinc solution

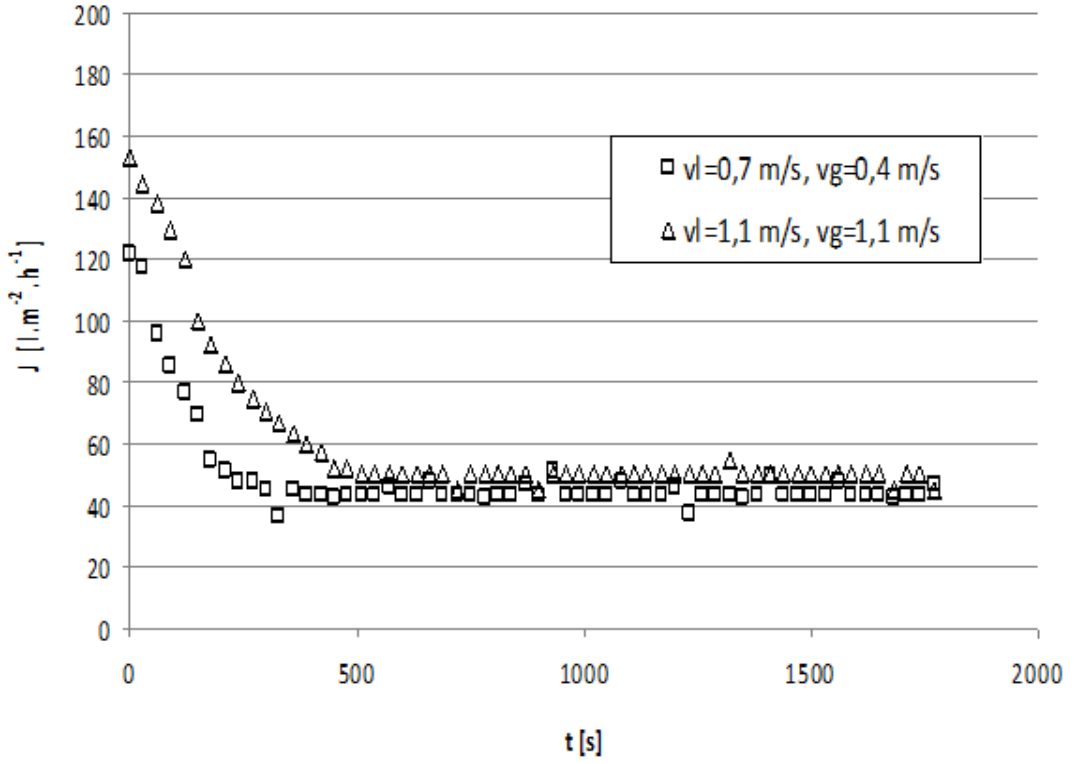

Fig. 8. Permeate flux through multi-channel membrane depending on time for Bentonite suspension microfiltration with two-phase flow at adsorbent concentration 5 g L^{-1} and transmembrane pressure 50 kPa without zinc solution

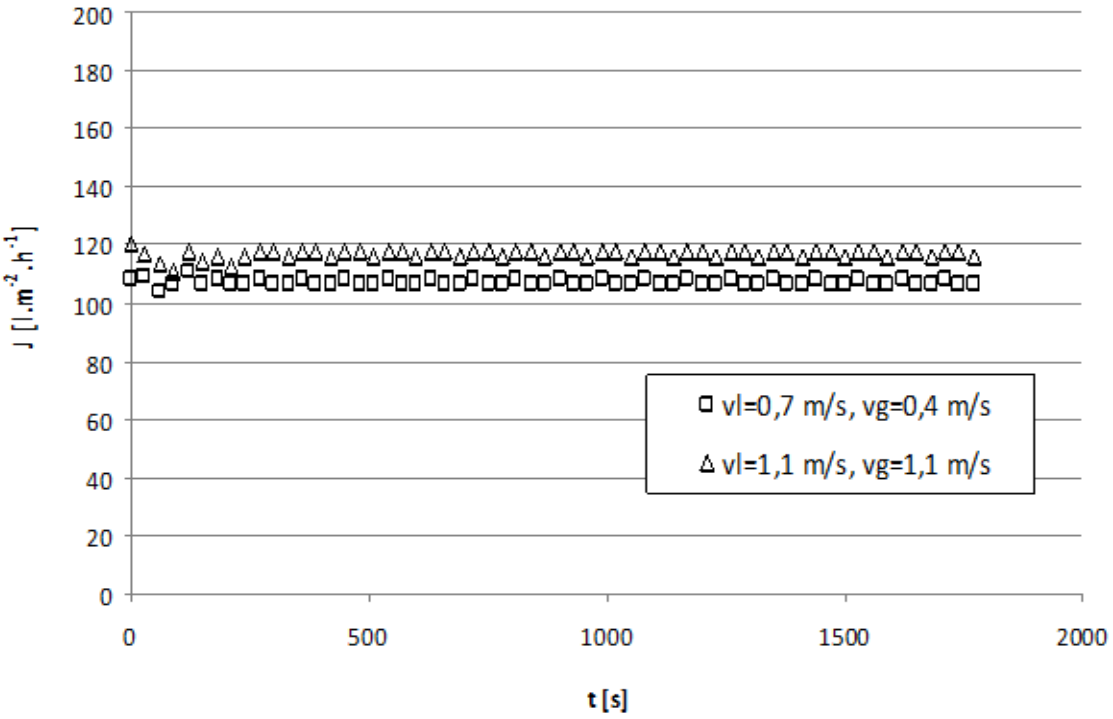

Fig. 9. Permeate flux through multi-channel membrane depending on time for Bentonite suspension microfiltration with two-phase flow at adsorbent concentration 5 g L^{-1} and transmembrane pressure 50 kPa with 250 mg L^{-1} zinc solution

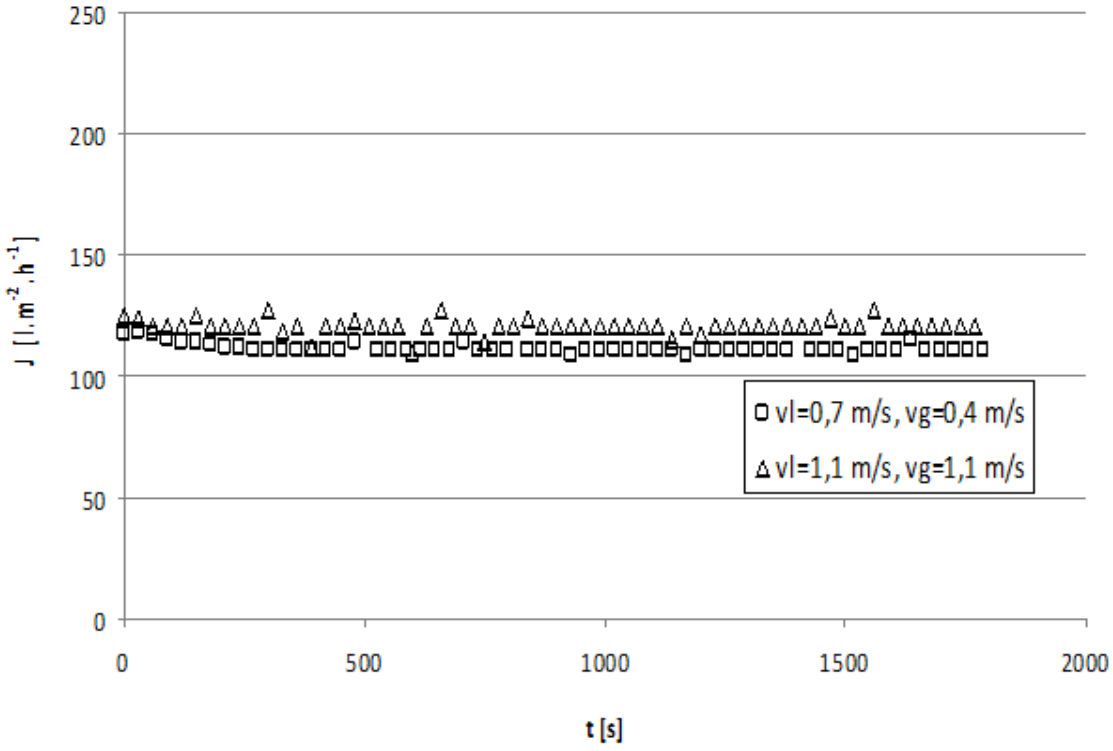

Fig. 10. Permeate flux through multi-channel membrane depending on time for Lewatit S1468 suspension microfiltration with two-phase flow at adsorbent concentration 5 g L^{-1} and transmembrane pressure 50 kPa without zinc solution

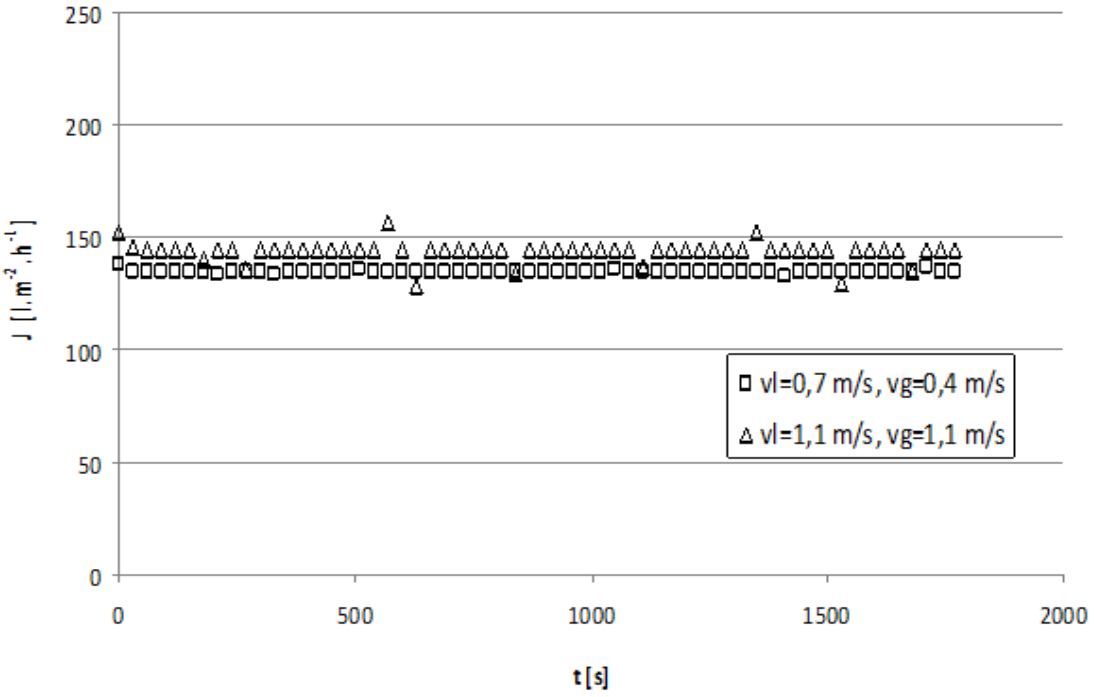

Fig. 11. Permeate flux through multi-channel membrane depending on time for Lewatit S1468 suspension microfiltration with two-phase flow at adsorbent concentration 5 g L^{-1} and transmembrane pressure 50 kPa with 250 mg L^{-1} zinc solution

Discussion

Single-Channel Membrane

An initial decrease of permeate flux was recorded during microfiltration of Bentonite suspension without Zn solution (Fig. 4). The flux decline of the two-phase flow was not as significant as for the single-phase flow, as presented in a previous study (Bakalár *et al*., 2013a; Müller *et al*., 2011).

An increase of the aeration rate also caused an enhancement of the permeate flux (Fig 6 and 7). Thus permeate flux, when the two-phase flow was applied, increased by 13 and 29% compared to the flow of single-phase at 1.4 and 2.2 m s^{-1} of the gaseous phase rate, respectively. In the experiments with suspension with adsorbed zinc there was no flux decline at the experiments with two-phase flow unlike the single-phase flow. The value was lower than the flux of clean water (Bakalár *et al*., 2013a) but higher than that of single-phase flow (Bakalár *et al*., 2013b). In this case the increase in the rate of aeration also caused an increase of the permeate flux (Fig. 5) by 13 and 16% compared with the flow of single-phase at 1.4 and 2.2 m s^{-1}, respectively.

A more significant difference was recorded in the permeate flux depending on the flow rate of liquid and gaseous phases. Compared to the single-phase flow (Bakalár *et al*., 2013a; 2013b; 2013c) the flux of the permeate achieved the steady state in longer time period but at a higher rate. The increase in two-phase flow was 4 and 5.5% compared to single-phase flow at 1.4 and 2.2 m s^{-1} of the gaseous phase rate, respectively, for both the experiments without and with adsorbed zinc. The difference in microfiltration of suspension with and without adsorbed zinc was not significant.

Multi-Channel Membrane

The decline of permeate flux and reaching the steady state lasted for a longer time in the microfiltration of Bentonite suspension through multi-channel membrane at two-than in single-phase flow (Bakalár *et al*., 2013a; Müller *et al*., 2011); however, the permeate flux was stabilized at a greater value (Fig. 8). Also there was not such a considerable growth of the permeate flux with the growth of flow rate as in the two-phase flow microfiltration through single-channel membrane. In the experiments with adsorbed zinc the situation was the same as for single-channel membrane, i.e., there was no initial permeate flux decline. The flux (Fig. 9) was higher by 22 and 25% compared to a single-phase flow (Bakalár *et al*., 2013b; 2013c) at 0.4 and 1.1 m s^{-1} of the gaseous phase rate, respectively.

There was no significant decrease of the initial permeate flux in measurements with Lewatit S 1468 suspension. The steady permeate flux was reached in approximately the same time as for the single-phase flow (Bakalár *et al*., 2013a). The permeate flux (Fig. 10) was increased by 12 and 13% at 0.4 and 1.1 m s^{-1} of the gaseous phase rate, respectively, as compared with

single-phase flow. In experiments with adsorbed zinc the initial flux decline was insignificant. There was also not a significant difference in permeate flux between the two-phase and single phase flows (Fig. 11).

The permeate flux decrease through multi-channel membrane depending on time for Lewatit S1468 suspension microfiltration with two-phase flow was not as fast as through single-channel membrane while the steady state was reached in longer time (Fig. 9 and 11). The steady permeate flux was reached at a higher value compared with single-channel microfiltration membrane by about 10 and 13% for Bentonite and Lewatit S1468, respectively. The experiments also showed an increase of the pressure drop value as a result of the membrane internal configuration. It was greater by 5.2 kPa (0.7 and 5.9 kPa for single- and multi-channel membranes, respectively).

The accuracy of the measurements has been ensured by performing the experiments in triplicate. The experimental results are a basis for further investigations expanding the range of used adsorbents as well as adsorbed metals.

Conclusion

As presented in Table 2 slug flow ($0.2<\varepsilon<0.9$) was the dominant regime in all the experiments. It is assumed that the flow followed as an alternation of gas slugs and liquid slugs containing smaller bubbles imposing flow turbulence which may significantly influence the secondary filtration cake and the gel layer formation on the top layer of the microfiltration membrane. This implies that the secondary layer formed during the process of microfiltration was washed out. This fact was also documented by the course of microfiltration experiments with adsorbed zinc as all the experiments showed no or only a minimum flux decline even after a half an hour from the start of the experiment.

The study addresses the impact of the liquid-gas two-phase flow onto the permeate flux through microfiltration membrane. In the experiments carried out the effect of the intensity of two-phase flow on the permeate flux in Bentonite microfiltration through multi-channel membrane was the most evident compared to microfiltration without aeration. Consequent increase in the intensity of permeate flow was observed with increasing aeration velocity. Generally it can be concluded that the application of aeration had an intensification effect on the microfiltration process of investigated suspensions.

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Author's Contributions

All authors of this research article have directly participated in the planning, execution and data analysis and manuscript preparation.

Ethics

This article is original and contains unpublished material. All of the authors have read and approved the manuscript and no ethical issues involved.

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