



Impact of aeration rate and zeolite concentration on the microfiltration flux

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ABSTRACT

Microfiltration is an efficient separation process in water purification. One of the drawbacks of this separation process is fouling. Several methods can be used for reduction of fouling. In this study the impact of aeration rate and the suspension concentration of two different zeolite fractions on the microfiltration flux was studied. Natural zeolite with main component of clinoptilolite with mean particle size of 20 and 50 μm and an asymmetrical inorganic membrane were used in the experiments. Microfiltration measurements were made to determine the enhancing effect of the aeration in the microfiltration system under various aeration rates and concentrations of the zeolite suspension. Higher permeate fluxes are reached at lower suspension concentrations of zeolite suspensions with mean particles of 20 μm irrespective of the applied air flow rate. At higher suspension concentrations the fluxes are similar for both the tested zeolite particle sizes. Regardless of the zeolite particle size used the applied air flow has a positive effect on the permeate flux due to the reduction of fouling of the membrane surface. The highest effect of the input air was at the flow rates of 1 and 1.5 $\text{m}^3\cdot\text{h}^{-1}$ and at high suspension concentrations. In the microfiltration experiments with applied air flow a 20–70% enhancement of the permeate flux compared to the experiments without air flow was recorded.

Keywords: Aeration; Flux; Microfiltration; Zeolite

1. Introduction

Membrane filtration using microfiltration and ultrafiltration membranes is widely used in treatment of drinking water. Plants for drinking water treatment using these methods are built in different parts of the world. The advantages of this method are high removal efficiency; flexibility – application throughout variety of materials, concentrations, pH, temperatures; no changes in phase and temperature – both permeate and retentate remaining in liquid form at the same temperature; no additional chemicals necessary; low area compared to performance; cost effectiveness; simple handling; etc. [1–3]. Despite the advantages, membrane treatment encounters several major problems. The most common are membrane fouling, insufficient removal of contaminants especially soluble organic compounds [4], etc.

The fouling of membrane is a complex process due to the adsorption of particles, particle deposition on the surface of the membrane, pores clogging and concentration polarization. The concentration polarization is irreversible damage to the membrane due to the specific physical-chemical interactions between the components of the suspension and the membrane [5,6]. The fouling of the membrane often results in reduction in permeate flow and an increase in the transmembrane pressure that causes an increase to the economic cost of the process [5,7]. Deposition of particles on the membrane surface may lead to 30 to 50% reduction in the initial flow [8,9]. The process is affected by the fouling characteristics of the membrane, operating conditions, environmental and other characteristics of solution [5].

Fouling causes a decrease in the flux; thereby reducing the separation efficiency. Much effort is put to prevent membrane fouling and improve flux, e.g. increase the filtration flow rate, gas injection into the liquid, creation of

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vortices, creation of obstacles in the membrane channel, backwashing, etc. Among these methods gas injection and backwash are economically and physically the most suitable [6–12].

Tubular microfiltration or ultrafiltration membranes are placed in a steel or plastic box with a diameter greater than 10 mm. The number of tubes in a module is usually 4–18. The feed is brought to the middle of tubes, permeate passes through the porous support, and is removed from the module. The membranes are usually ceramic. The main disadvantage of such a module is a small area per unit volume – less than $300 \text{ m}^2 \cdot \text{m}^{-3}$. The advantage is resistance to fouling due to suitable flow dynamics [13–15]. Vera et al. [16,17] in their studies proved that air sparging reduced fouling of membrane by a ferric hydroxide suspension and biologically treated wastewater and even in the case of wastewater, the solid phase was completely removed from the membrane surface. Smith et al. [18] found a 63% improvement by aeration in a hollow fibre system. Up to 320% improvement by aeration of ultrafiltration system compared to a system without aeration was reported by Cui and Wright [19]. Cui et al. [20] stated that the concept of aeration or bubbling has a positive impact on the mass transfer across the membrane surface at microfiltration and ultrafiltration and may also be useful in nanofiltration. In the study by Bakalár et al. [21] the effect of aeration on fouling in single- and multi-channel microfiltration membranes by suspensions of Bentonite and Lewatit S1468 – with and without adsorbed zinc was studied and the results showed an increase in the flux by 12–29%. Smith et al. [22] studied the process of ultrafiltration in ceramic membranes and confirmed the improvement of permeate flux by 47.2 to 131.2% using an aeration system. Injection of air into the microfiltration apparatus in order to reduce fouling of the membrane surface has been studied by several authors. In this study the emphasis was placed on the effect of the three-phase flow (gas-liquid-solid) on the results of microfiltration. Sur and Cui [23] also proved a flux enhancement in liquid-gas two-phase microfiltration experiments of yeast suspension carried out by a multi-tubular membrane. The results of published studies [23–33] indicate an 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. To reduce the adverse impact of fouling and concentration polarization on the microfiltration process, other methods have also been proved [29,30].

In our study the properties and behaviour of the suspension in a microfiltration apparatus run under changing conditions was studied in order to enhance the flux by reducing fouling in tubular ceramic microfiltration membrane. Our study concentrates on study of two commercially available fractions of Zeolite for possible use in a hybrid adsorption and microfiltration process for removal of selected heavy metals from aqueous solutions. This paper deals with one part – microfiltration which is studied separately due to the properties of Zeolite – small particle size and large active surface area that assumes the use of microfiltration for the removal of solid particles from treated solution and related flux decline due to particle deposition on membrane wall.

2. Materials and methods

2.1. Zeolite

Natural zeolite with main component of clinoptilolite (Zeocema.s. Bystré, Slovakia) was used in the experiments. Two different fractions were used – with mean particle size of 20 and 50 μm . These fractions were selected as they are commercially available.

Zeolites are naturally occurring minerals – aluminosilicates with an open crystal structure occupied by cations and water molecules. These components can be varied within large cavities, allowing ion exchange. Zeolite is composed of three-dimensional lattices of SiO_4 and tetrahedrons of AlO_4 . Natural zeolites are relatively cheap, safe, and environmentally friendly adsorbents. They also have a large surface area. Zeolites have great potential for many applications, for example molecular sieves, catalysts, adsorbents, surfactants, and for removal of cations from acid mine water and industrial waste water. They have a strong affinity for heavy metals [22,34–37].

2.2. Membrane

An asymmetrical inorganic membrane based on $\alpha\text{-Al}_2\text{O}_3$ (produced by Pall Corporation, USA) was used in the experiments. The inside surface of ZrO_2 forms 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 membrane module, i.e. an area of 54.98 cm^2 representing the active membrane area. The manufacturer indicates the average pore size of 100 nm.

2.3. Microfiltration device

The outline of the membrane instrument used for microfiltration experiments is presented in Fig. 1. In the experiments the mixture of adsorbent dispersed in demineralised water was pumped from a 10-L-volume reservoir by a pump with frequency converter. The tubular microfiltration membrane was placed in a vertical membrane module. The gas was injected into the feed before the membrane at a flow rate of u_g through a pipe of 1.3 cm in diameter. Permeate flowing from the membrane at a flux of J was gathered in a wide neck vessel. The retentate moved out back into the reservoir. The pressure P was adjusted by a control valve behind the membrane module. A pressure sensor connected to a personal computer was placed behind the membrane module. A flow metre was placed in the retentate part.

2.4. Experimental

Prior to each measurement a blank experiment was performed using demineralised water in order to insure the same conditions at the beginning of the experiment. Before the microfiltration experiments the Zeolite suspensions were stirred for 12 h to fully hydrate the solid phase. Also, prior to the experiments, distilled water was drawn from the reservoir into the microfiltration module and ran for 20 min. After stabilisation of the apparatus the Zeolite suspension was added to the reservoir so that the final suspension concentration (c_s) in the feed was as required. The

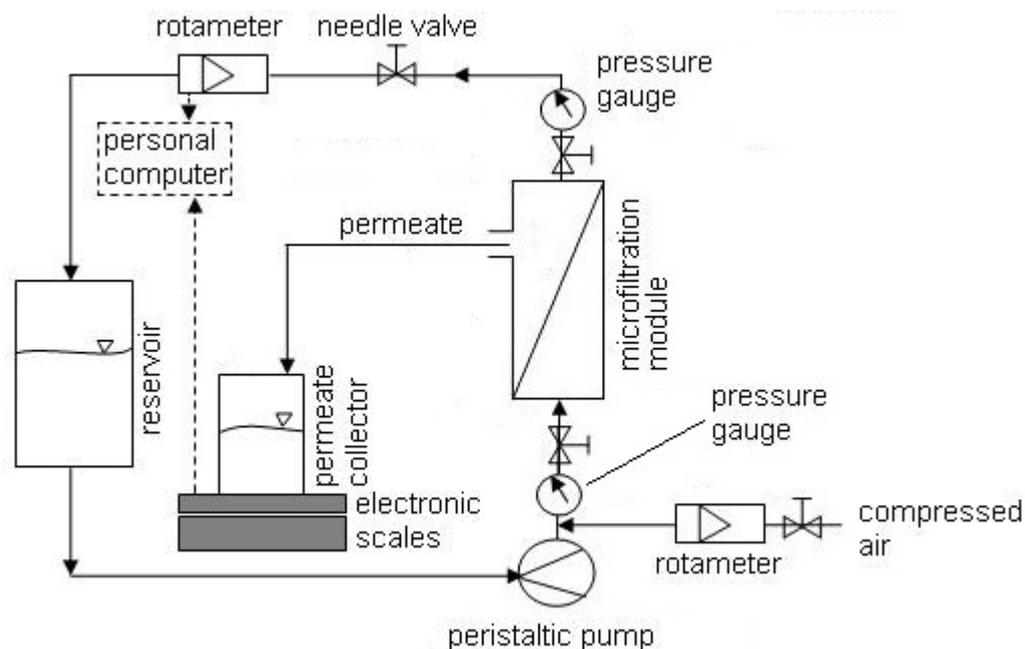


Fig. 1. Scheme of microfiltration device.

microfiltration experiments were carried out under constant pressure difference of 50 kPa, flow rate of 300 l·h⁻¹, and suspension concentrations of 1, 3, 6, 9, 15, 20, 25, and 30 g·l⁻¹. Measurements were carried out with a constant solid-liquid-gas three-phase flow with selected parameters. All the suspensions were subjected to experiments under gas flow rates of 0, 0.5, 1, and 1.5 m³·h⁻¹. Distilled water was added to the reservoir on regular bases in order to keep the suspension concentration and volume at the constant value of 4 L. The weight of permeate was recorded in regular intervals (*t*) and the flux was counted based on the weight.

3. Results and discussion

Microfiltration measurements were made to determine enhancing effect of aeration in the microfiltration system under various conditions i.e. under various aeration rates and concentrations of zeolite suspension. The experiments were done in triplicate and the average values were used in the results (Fig. 2) for easier differentiation. The reproducibility is more than 90%.

In the bar graphs permeate fluxes for last 50 min of microfiltration experiments are averaged.

3.1. Zeolite suspension with mean particle size of 20 μm

Plot of flux as a function of the time is shown in Fig. 3 as an example at various gas flow rates. At low zeolite suspension concentration the increasing aeration rate increases the flux but at the highest aeration rate (1.5 m³·h⁻¹) the flux drops. The course of the experiments with suspension concentrations of 3, and 6 g·dm⁻³ is similar; though the aeration rate of 6 g·dm⁻³ is influencing the flux considerably. Even more considerable is the influence of aeration at suspension

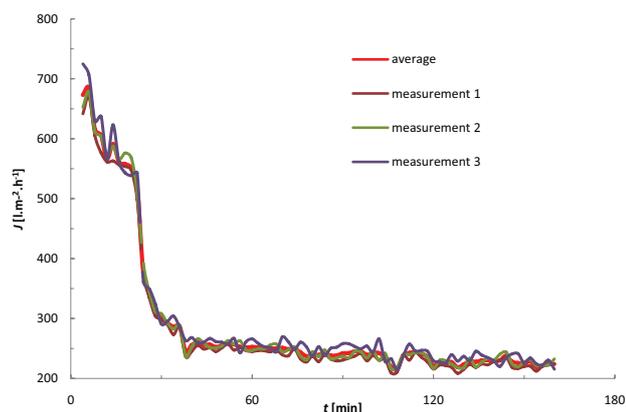


Fig. 2. Example of permeate flux as a function of the time – three parallel measurements and their average for Zeolite microfiltration with mean particle size of 20 μm, concentration of suspension 1 g·dm⁻³ and gas flow rate of 1 m³·h⁻¹.

concentration of 9 g·dm⁻³ while there is only a slight increase at lower aeration rate; however at higher aeration rates of 1 and 1.5 m³·h⁻¹ the increase of flux is much higher – 26, and 20%, respectively, compared to the flux without applied aeration. Again the flux drops at the highest aeration rate. Gradual increase of flux was recorded for the experiments with suspension concentrations of 15, 20, 25, and 30 g·dm⁻³ and the recorded increase of the flux was 50%, 35%, 55%, and 57%, respectively.

The course of experiments at suspension concentration of 20 g·dm⁻³ is different from the others as the difference in the flux for aeration rates 0 and 0.5, and 1 and 1.5 m³·h⁻¹ is very low though still keeping its growing order. At higher suspension concentrations there is greater impact of the

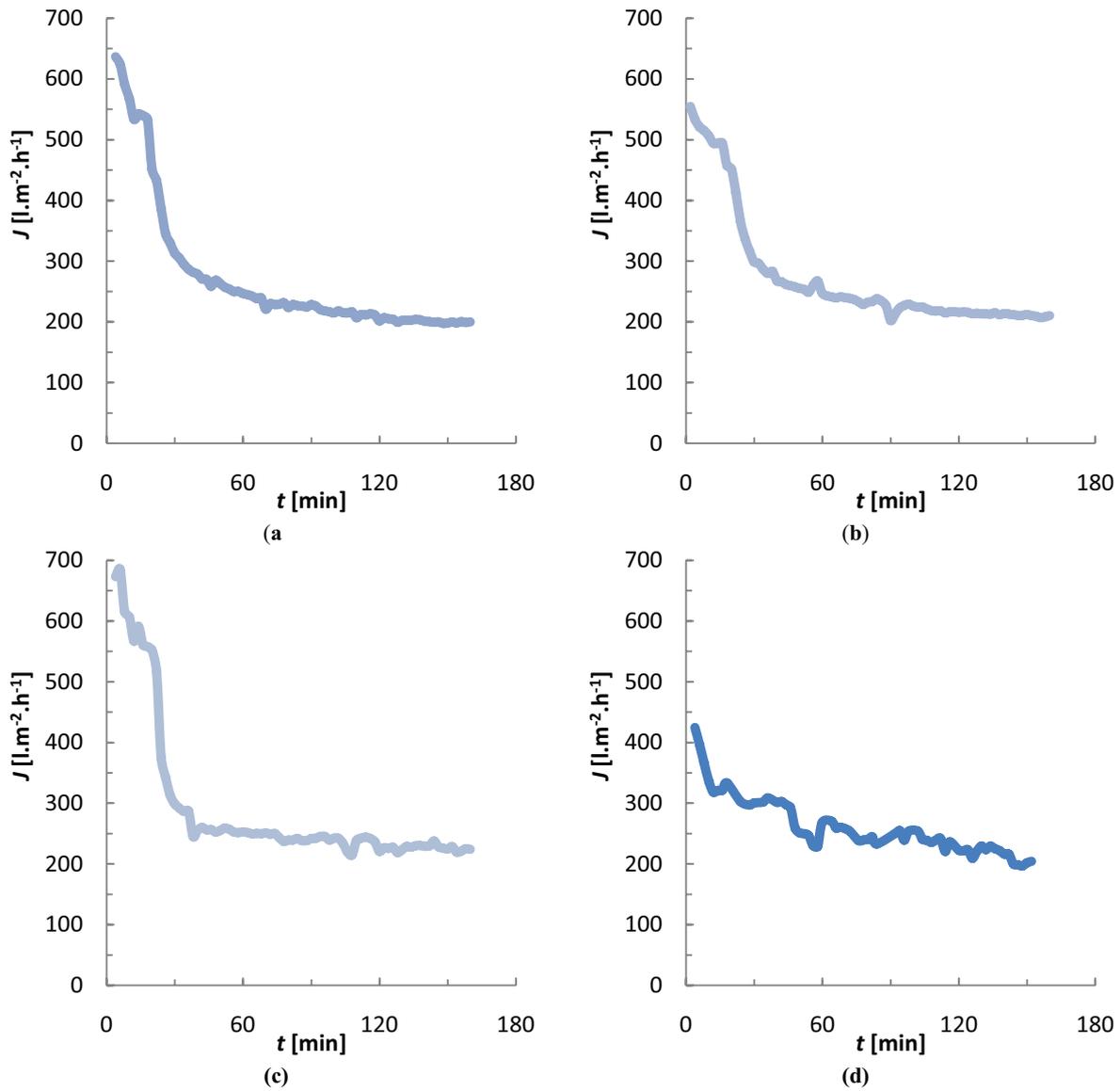


Fig. 3. Permeate flux for concentration of suspension $1 \text{ g}\cdot\text{dm}^{-3}$, with mean particle size of $20 \text{ }\mu\text{m}$. Gas flow rate: (a) $0 \text{ m}^3\cdot\text{h}^{-1}$, (b) $0.5 \text{ m}^3\cdot\text{h}^{-1}$, (c) $1 \text{ m}^3\cdot\text{h}^{-1}$, (d) $1.5 \text{ m}^3\cdot\text{h}^{-1}$.

aeration rate on the flux and therefore the improvement of the microfiltration conditions; though at low concentrations higher permeate fluxes are reached even at lower aeration rates. The results of the microfiltration experiments for suspensions of Zeolite with mean particle size of $20 \text{ }\mu\text{m}$ are presented in Fig. 4. At all the aeration rates the flux is lowering with increasing suspension concentration. On the contrary, with a few exceptions, the flux is increasing with increasing aeration rate at all suspension concentrations. The highest increase is at high suspension concentrations.

3.2. Zeolite suspension with mean particle size of $50 \text{ }\mu\text{m}$

Plot of flux as a function of the time is shown in Fig. 5 as an example at various gas flow rates. At suspension concentration of $1 \text{ g}\cdot\text{dm}^{-3}$ the flux is increasing from 91 to 156

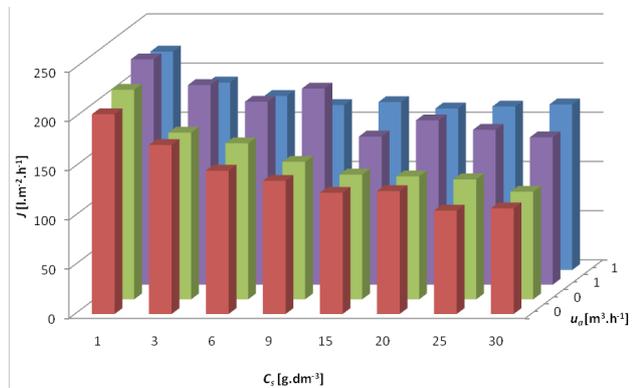


Fig. 4. Effect of suspension concentration with mean particle size of $20 \text{ }\mu\text{m}$ and the aeration rate on the permeate flux.

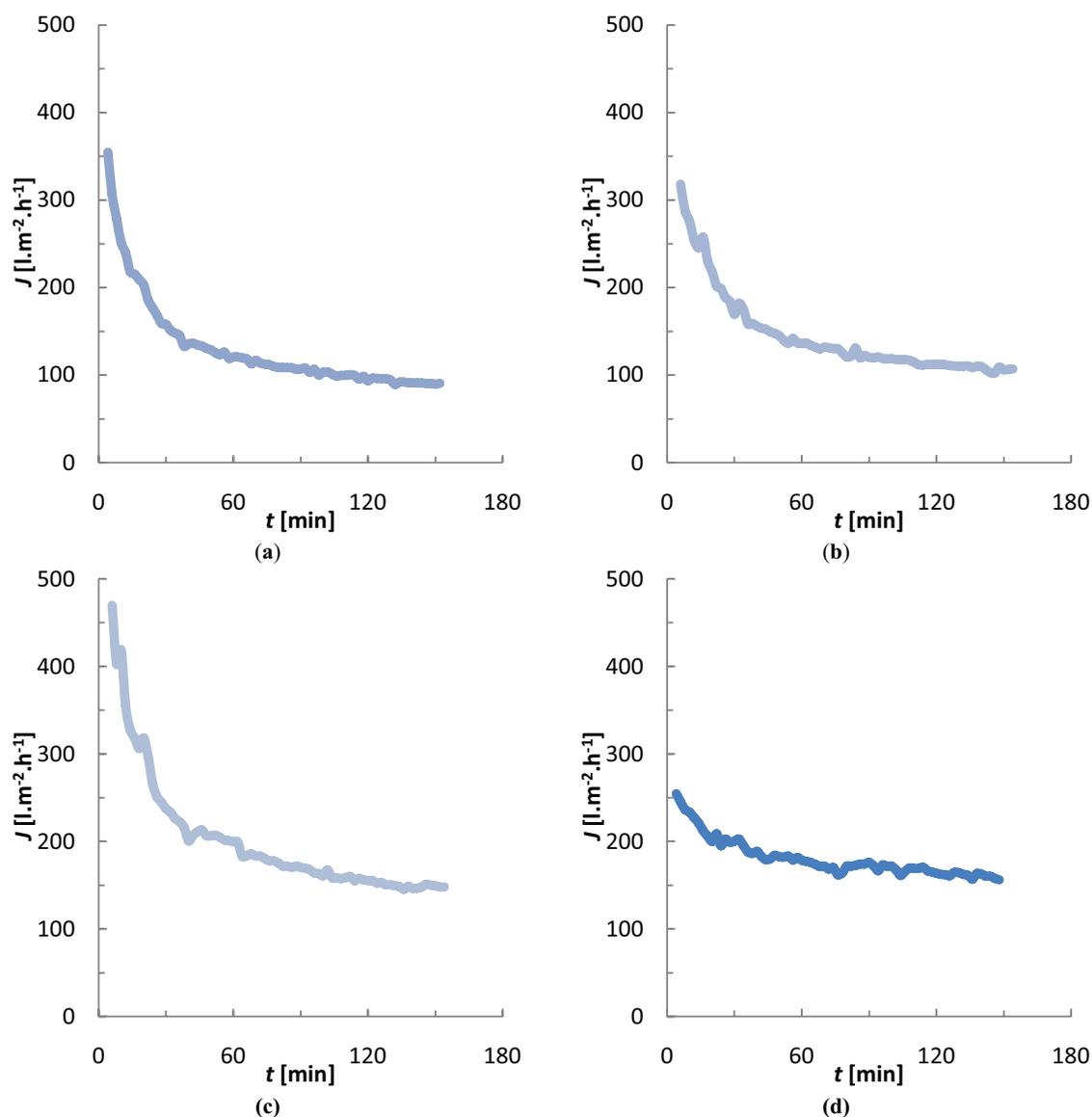


Fig. 5. Permeate flux for concentration of suspensions $1 \text{ g}\cdot\text{dm}^{-3}$, with mean particle size of $50 \mu\text{m}$. Gas flow rate: (a) $0 \text{ m}^3\cdot\text{h}^{-1}$, (b) $0.5 \text{ m}^3\cdot\text{h}^{-1}$, (c) $1 \text{ m}^3\cdot\text{h}^{-1}$, (d) $1.5 \text{ m}^3\cdot\text{h}^{-1}$.

$1\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ with increasing aeration rate from 0 to $1.5 \text{ m}^3\cdot\text{h}^{-1}$, respectively; hence the flux improved of 73% . At suspension concentration of $3 \text{ g}\cdot\text{dm}^{-3}$ the flux is also increasing with increasing aeration rate but the flux is lower at suspension concentration at $1.5 \text{ m}^3\cdot\text{h}^{-1}$ than at $1 \text{ m}^3\cdot\text{h}^{-1}$. The flux is increasing with increasing aeration rates for all the other suspension concentrations of $6, 9, 15, 20, 25,$ and $30 \text{ g}\cdot\text{dm}^{-3}$ by $63, 56, 37, 28, 27,$ and 33% , respectively, compared to the flux without aeration.

The results of the microfiltration experiments for suspensions of zeolite with mean particle size of $50 \mu\text{m}$ are presented in Fig. 6. Unlike the experiments with zeolite with mean particle size of $20 \mu\text{m}$ (Fig. 4), the flux is increasing with the increasing suspension concentration at all the applied aeration rates with the exception of the highest used suspension concentration ($30 \text{ g}\cdot\text{dm}^{-3}$) for which a flux

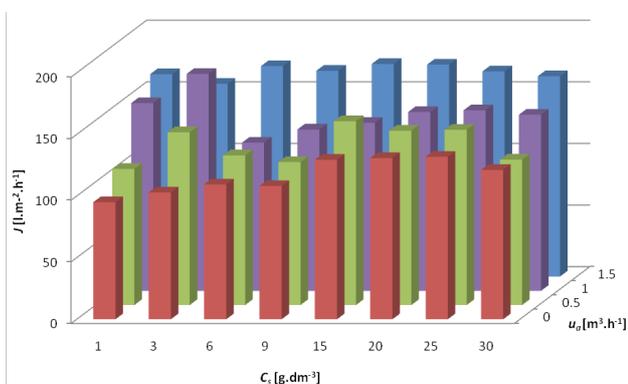


Fig. 6. Effect of suspension concentration with mean particle size of $50 \mu\text{m}$ and the aeration rate on the permeate flux.

decline was observed. At the same time an intensification of the microfiltration process was observed by the application of aeration and therefore the increase of flux was high, especially for the aeration rate of $1.5 \text{ m}^3\cdot\text{h}^{-1}$.

Our results confirm the results of the most comprehensive study of effects of operating parameters on flux in bubble enhanced cross flow microfiltration of baker's yeast using 5 mm diameter tubular membranes performed by Sur and Cui [23]. At more concentrated suspension the main reason for flux decrease is cake formation [10]. The increase of flux caused by applied aeration is the highest when cake formation is more severe thus the results suggest that an increase to the flux is caused by disruption of the cake formed on the membrane wall. It was also found that at any suspension concentration air injection has an effect on the permeate flux. The aeration increases the flux to suspensions with mean particle size of both 20 and 50 μm but in the case of 20 μm fraction the highest enhancement is between gas flow rates 0.5 and $1 \text{ m}^3\cdot\text{h}^{-1}$ while in the case of 50 μm fraction the highest enhancement is between gas flow rates 1 and $1.5 \text{ m}^3\cdot\text{h}^{-1}$. This implies that in the case of 20 μm fraction not only cake formation is the cause of flux decrease but also pore blockage may occur and in the case of 50 μm fraction only cake formation is the cause of flux decrease.

The effect of aeration during microfiltration process can make a notable contribution to process streamlining. In the microfiltration experiments with applied air flow a 20–70% enhancement of the permeate flux compared to the experiments without air flow was recorded.

Our results of the study on the effect of aeration on the microfiltration of two different fractions of natural zeolite (Table 1) confirmed the results of other authors in various applications, such as membrane bioreactors, fermentation products processing, or separation of macromolecular solutions.

4. Conclusion

Aeration can have a considerable effect on membrane fouling reduction; therefore, it can increase permeate flux.

Table 1
Maximum percentage of flux enhancement by aeration compared to the flux with no applied aeration for particular suspension concentrations

Particle size	20 μm		50 μm	
	Increase of flux [%]	Rate of air flow [$\text{m}^3\cdot\text{h}^{-1}$]	Increase of flux [%]	Rate of air flow [$\text{m}^3\cdot\text{h}^{-1}$]
Suspension concentration [$\text{g}\cdot\text{dm}^{-3}$]				
1	12	1	73	1.5
3	15	1	78	1
6	29	1	63	1.5
9	42	1	56	1.5
15	42	1.5	37	1.5
20	35	1	28	1.5
25	56	1.5	27	1.5
30	57	1.5	33	1.5

The impact of the gradual increase of air flow rate on permeate flux in two fractions (20 and 50 μm) of zeolite suspension was studied. The transmembrane pressure was 50 kPa. In addition to air flow rate ($0\text{--}1.5 \text{ m}^3\cdot\text{h}^{-1}$), the concentration of used suspension was also varied from 1 to $30 \text{ g}\cdot\text{dm}^{-3}$. According to the results of experiments higher permeate fluxes are reached, in particular, at lower suspension concentrations of zeolite suspensions with particles of 20 μm irrespective of the applied air flow rate. At higher suspension concentrations ($>15 \text{ g}\cdot\text{dm}^{-3}$) fluxes are similar for both the tested zeolite suspension particle sizes. Regardless of the zeolite suspension used (20 or 50 μm) the applied air flow has a positive effect on the permeate flux due to the reduction of fouling of the membrane surface. The highest effect of input air was at flow rates of 1 and $1.5 \text{ m}^3\cdot\text{h}^{-1}$ and at high suspension concentrations. The results clearly point out that the negative effect on flux in microfiltration membranes caused by cake deposition of fine particles on the membrane surface can be reduced by aeration. Thus the application of aeration can increase yield, cut back the frequency of cleaning which might prolong the membrane lifespan and result in lower costs and energy requirements. Further research will focus on combination of a hybrid adsorption and microfiltration process using Zeolite for removal of selected heavy metals incorporating the results of presented study.

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