Investigations of the mechanism of the fouling in microgranular adsorptive filtration

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Abstract

The application of microgranular adsorptive filtration (μGAF) has been successfully used in conjunction with membrane filtration. It proves to be efficient not only in removal of natural organic matter (NOM) but also it significantly reduces the extent of fouling. There are a few mathematical models evaluated to understand the underlying mechanism of fouling. This paper describes a method of predicting filtration capacities using constant flow datasets collected when μGAF was applied. The results suggest that the behaviour of fouling in microgranular adsorptive filtration system varies between different filtration operations and natural water quality. In analysed case the mechanism of pores blocking can be described by the complete blocking mechanism.

Key words: *constant flow fouling models, membrane filtration, membrane fouling, pore blocking*

INTRODUCTION

The filtration process is widely used, particularly in water supply but also in industrial and municipal wastewater treatment plants. The filtration through the membrane is susceptible to fouling, resulting in significant reduction of the filtration efficiency and increase the costs of the process. Fouling is consider to be connected with clogging of the pores, and the formation of the cake layer, as well as the concentration polarization on the membrane surface.

The modelling of filtration process is difficult due to influence of various factors [MARCINKOWSKI *et al*. 2016; MIODUSZEWSKI *et al*. 2014; SCHALLER *et al*. 2006]. For example, the mechanism in ultrafiltration (UF) is not clearly explained mainly because of the diversity of its application in different arrangements. Moreover, the filtration models are usually divided onto two main groups: dead-end mode and cross-flow mode. Usually to describe the mechanism of permeation two flow models are commonly used: pore-flow model (permeates are transported by pressure-driven convective flow) and solution-diffusion model. The

pore-flow model is the most common and it describes the flux across the membrane based on Darcy's law:

$$J = \frac{\Delta P}{\eta(R_M + R_c)} \tag{1}$$

where: ΔP = transmembrane pressure, Pa; η = the solution viscosity, Pa-s; R_M = the hydrodynamic resistance of clean membrane, $1 \cdot m^{-1}$; R_c = the resistance of the concentration polarization layer, $1 \cdot m^{-1}$.

The difference between the solution-diffusion and pore-flow mechanisms lies in the relative size and permanence of the pore [BAKE 2004]. Mathematical mechanism of membrane fouling based on blocking filtration law was described by the Hermia's models [ARYANTI *et al*. 2016; HERMIA 1982; KONIECZNY, RAFA 2000; RAJCA 2011]. The equations come with their limits, though. Both dead-end and constant pressure filtration are required to use them. Moreover, Hermia's models are based on the assumption that single fouling mechanism dominates in given system [HERMIA 1982; BENJAMIN, LAWLER 2013]. In general, four fouling mechanisms can be observed: com-

plete pore blocking, internal pore blocking, partial pore blocking, and cake filtration. In 1993, HLAVACEK and BOUCHET [1993] modified Hermia's for porous membranes equations for constant flowrate condition.

When particles are larger than the pore size, they completely block pores and in such case the complete blocking model can be applied. The intermediate blocking model is used when particles smaller than the pore size enter the pores and get either adsorbed or deposited onto the pore walls. And when particles get accumulated inside the membrane on the pore walls and the resulting constrictions of pores reduce the membrane's permeability the standard blocking model can be used [FIELD 2010; SAMPATH *et al.* 2014]. These mechanisms have been used individually as well as in combination to explain experimental various observations [HALE, DANIELS 1961; HO, ZYDNEY 2000; SAMPATH *et al.* 2014].

Fouling is affecting the hydraulics by decreasing the flux, when fixed TMP (Transmembrane Pressure) is used, or increasing TMP for a fixed flux. At the same time, it contributes to the change of the effective pore size distribution.

Many studies were devoted to mitigating membrane fouling and enhancing membrane process performance. Among others the pretreatment of water is most common technique. There are different approaches for pretreatment like pre-filtration through granular media or coarser membranes, or applying coagulation/adsorption and pre-oxidation [LIU 2015]. One of the newest approaches is raw water pretreatment by micron-sized heated aluminium oxide particles (HAOPs) prior to membrane filtration, so called micro-granular adsorptive filtration (μGAF). The process integrates granular media filtration and packed bed adsorption [CAI *et al.* 2008; KIM *et al.* 2007; 2008; 2010; LIU 2015; MALCZEWSKA 2016; MALCZEWSKA, BENJAMIN 2016; MALCZEWSKA *et al.* 2015]. The aim of this paper was to estimate the fouling parameters using the experimental data for micro-granular adsorptive filtration.

MATERIALS AND METHODS

Lake Union, located in Seattle, WA, was used as the source of natural water. The detailed description of experimental setup is presented in the publication MALCZEWSKA and BENJAMIN [2016]. Briefly, the stainless steel mesh (#316) had a 200 × 1400 twilled Dutch weave pattern with nominal 10-μm openings (Howard Wire Cloth Corp., Hayward, CA) was used as the adsorbent (μGAF) support media. The tubes of mesh were set vertically and operated with dead-end and inside-out filtration mode. After HAOPs were deposited on the tube inside wall, the water filtration started with the flux set up as 150 $dm^3 \cdot m^{-2} \cdot h^{-1}$ (commonly referred as LMH) and then 250 $dm^3 \cdot m^{-2} \cdot h^{-1}$. Then, the water samples were analysed on a UV-Vis spectrophotometer at a wavelength of 254 nm. UV254 nm was measured with a dual-beam Lambda-18 spec-

trophotometer with a 1 cm quartz cell (Lambda-18, Perkin-Elmer, USA).

In this study, filtration experiments were operated under two different constant flowrates and dead-end filtration, therefore Hlavacek and Bouchet's models were chosen for fitting the experimental data.

RESULTS AND DISCUSSION

Figure 1 shows the TMP that is required to maintain the constant flux of 150 $dm^3 \cdot m^{-2} \cdot h^{-1}$ and 250 $dm^3 \cdot m^{-2} \cdot h^{-1}$, as a function of filtration time. HAOPs pretreatment removed substantial amounts of NOM from tested samples and significantly mitigated fouling by all them regardless of increasing flux.

Presented results confirm existing reports that passage of feed through a thin layer of pre-coated HAOPs removes NOM and membrane foulants and substantially removes the majority of the UV254 from the raw water [CAI *et al.* 2008; KIM *et al.* 2007; 2008; 2010; LIU 2016; MALCZEWSKA 2016; MALCZEWSKA, BENJAMIN 2016; MALCZEWSKA *et al.* 2015].

The blocking filtration law models for constant flowrate condition is expressed in the term of ΔP (an effective or transmembrane pressure (Pa)) and *Vsp* (defined as the cumulative permeate volume per unit of membrane area**)** as shown in Figure 2.

The Table 1 provides membrane fouling models for constant flowrate condition and estimated values of parameters for both analysed flux. The estimation of mathematical models of fouling mechanisms using filtration blocking laws was investigated to predict the main fouling mechanisms when natural water passed through a thin layer of HAOPs (Tab. 1). The best fit model for the measurement data is the complete blocking law, with linear regression coefficients R^2 of 0.94–0.96, the standard blocking gave the R^2 between 0.93 and 0.94; and in case of the intermediate blocking law the R^2 was between 0.92 and 0.93. The complete blocking of the pores of μGAF is the most adequate for fouling description. The particles/solutes accumulate on the HAOPs layer and they do not overlap to other solute that has been previously deposited on the surface.

ARYANTI *et al.* [2016] studied the principles and fouling analysis of konjac glucomannan discovered that the fouling mechanism was controlled by complete blocking and cake/gel formation, respectively. This results are supported by GIJIU *et al.* [2012] they reported when combined complete pore blockage-caking filtration model is the most suitable model for their experimental data. On the other hand, SU *et al.* [2015] when studied the effect of hydrophobic and hydrophilic fractions on membrane fouling. They discovered that hydrophobic fractions exhibited a cake layer as the major fouling mechanisms, while the intermediate blocking was caused by the hydrophilic fraction. Others argued that standard blocking of the pores of ultrafiltration (UF) and microfiltration (MF) membranes is usually the most adequate [LE-CLECH
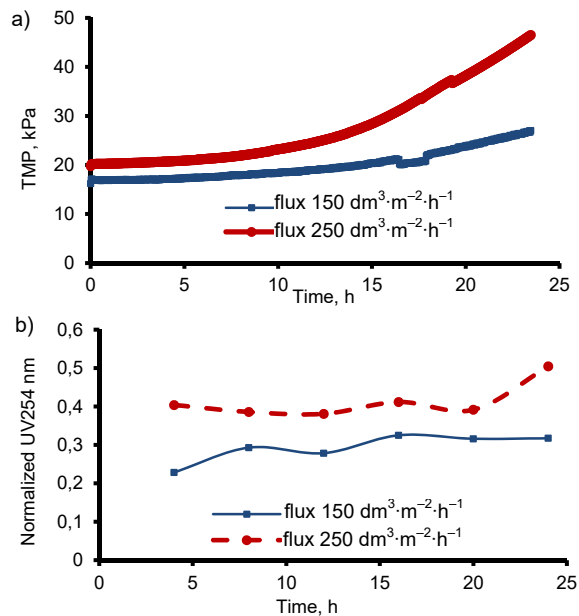
Fig. 1. Experimental data generated for constant flow rate during the operation period: a) transmembrane pressure (TMP) profiles during membrane filtration, b) normalized UV 254 nm (permeate/feed) whereas samples were collected at pre-selected intervals for analysis of UV absorbance; source: own study
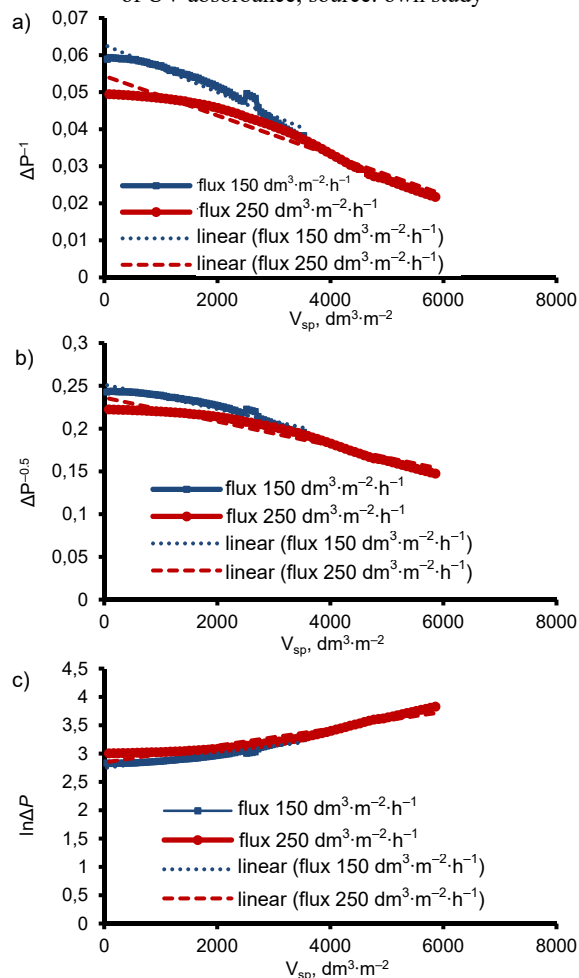

Fig. 2. Blocking filtration law analysis of μGAF: a) complete blocking, b) standard blocking, c) intermediate blocking law; source: own study

Table 1. The fouling models for constant flowrate and estimated values of parameters for both analysed flux

Parameter	Complete blocking $\dfrac{1}{\Delta P} = a + bV$	Standard blocking $\dfrac{1}{\sqrt{\Delta P}} = a' + b'V$	Intermediate blocking $\ln \Delta P = a'' + b''V$
Flux 150 dm³·m⁻²·h⁻¹			
a	0.0627	0.2517	2.7475
b	−0.000006	−0.00001	0.0001
R^2	**0.944**	0.9314	0.9173
Flux 250 dm³·m⁻²·h⁻¹			
a	−5E–06	0.2364	2.846
b	0.0544	−0.00001	0.0001
R^2	**0.959**	0.9444	0.9257

Source: own study.

et al. 2006; POLYAKOV *et al*. 1999]. LIU [2015] evaluated fouling of the μGAF and modeled it by Hermia's intermediate fouling model when cartridge system was used. On the other hand, this study conducted on the tubular system showed that best model describes the phenomenon of fouling is the complete blocking law.

CONCLUSION

Fouling is a major problem and the most challenging obstacle in the application of the membrane technology. There are a few mathematical models evaluated to understand the underlying mechanism of fouling.

In this case, the blocking filtration law models for constant flow rate based on Hlavacek and Bouchet's model were used. The results suggest that the behaviour of fouling in micro-granular adsorptive filtration system varies between different filtration operations and natural water quality.

This suggests that the mathematical description of fouling is more complex than a model assuming one of its components constant.

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Beata MALCZEWSKA

Badanie mechanizmu zatykania porów membrany (foulingu) w przypadku zastosowania filtracji µGAF

STRESZCZENIE

Mikro-ziarnowa adsorpcyjna filtracja (µGAF) jest z powodzeniem stosowana w połączeniu z filtracją membranową. Metoda ta jest skuteczna nie tylko w usuwaniu naturalnych substancji organicznych (NOM), ale również znacznie zmniejsza stopień zatkania się podczas filtracji porów membrany (proces foulingu). Aby zrozumieć zjawisko foulingu, opracowano kilka modeli matematycznych. W niniejszej pracy opisano mechanizm blokowania porów podczas µGAF filtracji. Uzyskane wyniki dają podstawy do przypuszczeń, że w zależności od zastosowanego systemu oraz właściwości chemicznych i fizycznych filtrowanej wody różne modele matematyczne mogą mieć zastosowanie. W analizowanym przypadku mechanizm blokowania porów najlepiej opisuje model całkowitego zatykania porów.

Słowa kluczowe: *filtracja membranowa, fouling, model zatykania porów ze stałym przepływem, zatykanie porów*