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가압식 멤브레인 수처리에서 수리학적 세정이 파울링 기작에 미치는 영향

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Hydraulic Cleaning Effect on Fouling Mechanisms in Pressurized Membrane Water Treatment

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요 약: 멤브레인 파울링은 지표수를 처리하는 저압 멤브레인 기술 적용의 확장에 있어 큰 장애가 된다. 따라서 파울링 제어를 위한 주기적인 수리학적 세정기술의 최적화는 매우 중요하다. 주기적인 수리학적 세정과 이와 연관된 파울링 현상에 관한 올바른 이해는 멤브레인 세정 전략을 최적화하기 위해 매우 유용할 수 있다. 실험적으로 측정한 투과도와 전통적인 Hermia 파울링 모델 예측 치의 비교를 통해, 본 연구에서는 합성 탁도유발 시료를 처리하는 가압식 멤브레인 공정에서 30분 여과와 정세정/역세정이 포함된 1분 세정조건을 바탕으로 6번의 운전사이클을 통해 발생하는 파울링 현상을 분석하고 이를 통해 지배적인 파울링 기작을 정량적으로 이해하고자 하였다. 단독 세정에서, 첫 번째 운전사이클에서 발생하는 파울링은 완전공극막힘 현상에 의해 주로 지배되었고 마지막 운전 사이클에서는 케이크 형성이 지배적인 파울링 기작으로 관찰되었다. 정세정과 역세정이 혼합된 경우, 파울링 속도는 감소하였으나 전반적으로 케이크 형성이 주 파울링 기작으로 관찰되었다.

Abstract: Membrane fouling is the main issue hindering the expansion of low pressure membrane processes for surface water treatment. Therefore, applying periodic hydraulic cleaning for fouling control should be well optimized. Better understanding of membrane fouling associated with periodic hydraulic cleaning would be useful to optimize membrane cleaning strategies. By comparing experimental permeability data with the classical Hermia blocking laws, this study aims at analyzing membrane fouling and understanding dominant fouling mechanisms occurring when filtering a synthetic surface water solution with a pressurized membrane process during six filtration cycles of 30 min each, separated with cyclic cleaning of 1 min by backwashing and forward flushing separately and combined. When applying single cleaning technique, membrane fouling during the first cycles was controlled by complete blocking mechanism while the last cycles were dominated by cake formation. Nevertheless, when combining cleaning technique better membrane regeneration was obtained and fouling was mainly due to cake formation.

Keywords: *Pressurized membrane, Hydraulic cleaning, Modelling approach, Fouling mechanism, Surface water treatment*

1. Introduction

Interest in low pressure driven membrane processes for surface water treatment has kept increasing due to high removal efficiency of pathogens, viruses and bacteria as well as its ability in removing considerably colloids, suspended particles and natural organic matters[1-2]. Nevertheless, membrane fouling is the main issue and it always hinders membrane applications [3-5]. The main foulants identified when treating surface water were the particulate/colloidal materials, proteins like substances as well as humic and fluvic acids [6-7]. To alleviate fouling, a periodical cleaning by relaxation, backwashing and/ or forward flushing is often

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applied[8]. Nevertheless, determining the optimal cleaning strategy in terms of cleaning technique, for example, cleaning duration and frequency, is still a big challenge, since it depends on feed water characteristics, membrane material and module configuration[9-10]. Moreover, the cleaning technique applied to the membrane can be related to the fouling mechanisms[11]. Membrane relaxation is applied to allow the diffusion of the foulants deposited on the membrane surface to the bulk solution according to a concentration gradient. Forward flushing is applied to increase the shear intensity in the vicinity of the membrane surface to remove the foulants deposited on the membrane surface. Backwashing is mainly recommended to mitigate pore blocking by removing the foulants trapped within the membrane pores[12]. A better understanding of the fouling phenomenon would be useful to optimize the cleaning strategy. Furthermore, modelling has been shown to be a helpful way to assess fouling and its mechanisms[13-14].

This study aims to identify the fouling mechanisms occurring when applying backwashing and forward flushing during the filtration of high turbidity surface water using a pressurized membrane module. The fouling mechanisms will be identified using a modelling approach based on the classical blocking laws developed by Hermia[15].

2. Modelling Approach

An approach based on the blocking laws developed by Hermia, is used to identify the fouling mechanisms responsible of membrane permeability decrease[15]. The classical blocking laws consist in four mathematical models expressing the permeate flux variation due to four different fouling mechanisms, the pore constriction, the complete blocking, the intermediate blocking and the cake formation (Table 1). Even if developed to describe fouling in a dead end filtration mode at a constant trans-membrane pressure TMP, in this study Hermia models are considered to study fouling obtained at constant permeate flux. Chang et al. compared fouling occurring at constant permeate flux and constant TMP and showed that similar fouling was obtained when particulate solution is filtered by an ultrafiltration membrane, which is similar to the case studied in this work where a solution of SiO_2 particles was filtered[16]. Based on the permeability expression, Hermia models where modified to describe the permeability variation instead of the permeate flux.

$$L_p = \frac{J}{TMP} \tag{1}$$

Table 1. Hermia Blocking Filtration Laws

Fouling mechanism	Expression
Pore constriction	$L_p = \frac{4.L_{p,0}}{(K_{pc}.L_{p,0}.t+2)^2}$ (2)
Complete blocking	$L_p = L_{p,0}\,exp(-K_{cb}.t)$ (3)
Intermediate blocking	$L_p = \frac{L_{p,0}}{K_{ib}.L_{p,0}.t+1}$ (4)
Cake formation	$L_p = \frac{L_{p,0}}{(2.K_{cf}.L_{p,0}^2.t+1)^{\frac{1}{2}}}$ (5)

Where L_p is the membrane permeability ($m^3 \cdot m^{-2} \cdot s^{-1} \cdot Pa^{-1}$), $L_{p,0}$ the initial membrane permeability ($m^3 \cdot m^{-2} \cdot s^{-1} \cdot Pa^{-1}$), t the filtration time (s), K_{pc} the pore constriction parameter (m^{-1}), K_{cb} the complete blocking parameter (s^{-1}), K_{ib} the intermediate blocking parameter (m^{-1}), K_{cf} the cake formation parameter ($s \cdot m^{-2}$).

The determination of the predominant fouling mechanism is based on identifying which of these four fouling mechanisms best fits the permeability experimental data. The parameters corresponding to each model including K_{pc}, K_{cb}, K_{ib} and K_{cf}, have been optimized to adjust each model on experimental data using least squares method on Matlab software. This method is based on optimizing the model parameters permitting the minimization of the Least Squares (LS) function (Eq. 6). The model allowing the lowest LS value corresponds to the dominant fouling mechanism.

$$LS = \sum(L_{p_{experimental}} - L_{simulation})^2 \tag{6}$$

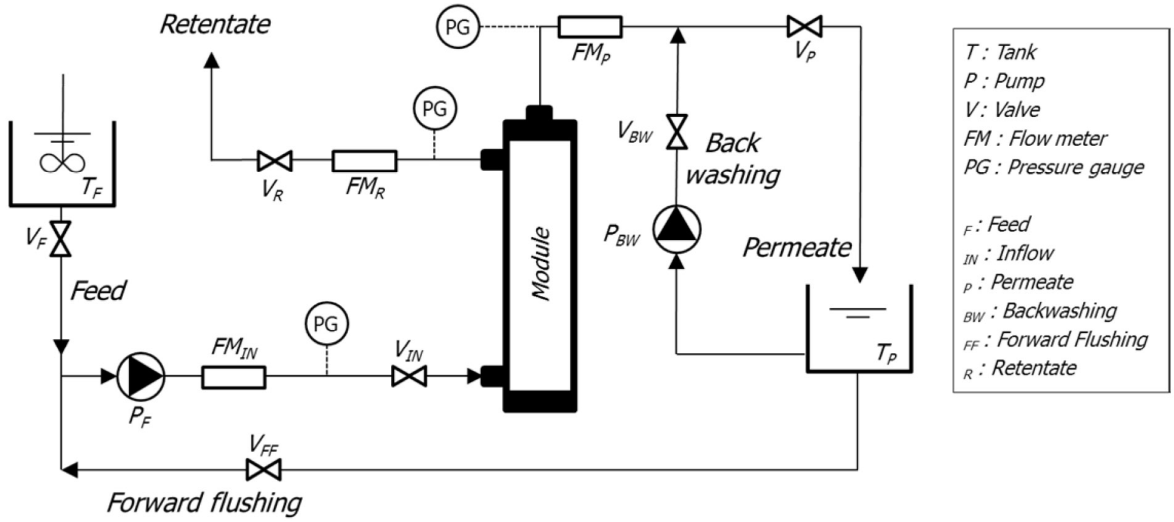

Fig. 1. Experimental set-up of pressurized UF membrane system.

3. Experimental

3.1. Experimental set-up

A laboratory scale pressurized ultrafiltration (UF) membrane setup shown in Fig. 1 was used. A hollow fibers polysulfone membrane module of 100 fibers, with a total area of 0.11 m^2, a mean pore size of 0.05 μm and an ID/OD of 0.9/1.4 mm was used for this study. The membrane water permeability at 28 ± 0.9°C was 594 ± 94 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$. Set-point permeate flux was maintained by micro gear pump (WT3000-1JA, Longerpump, China). Flow rates and pressures were measured by impeller flow meter (FHK G1/4, Digmesa, Swiss) and digital pressure gauge (PSAN-L1CA, Autonics, Korea), respectively. System control and data registration were realized by PLC installed customized software. The system is operated continuously. The driving force of the filtration process is ensured by the feed pump, which enables feed solution to flow through the membrane matrix. A flow sensor downstream the membrane detects the permeate flux decrease due to membrane fouling and sends a signal to the feed pump controller to increase pump speed in order to maintain a constant permeate flux.

3.2. Membrane operation

A synthetic high turbid surface water with turbidity of 10 NTU was used. High turbidity was ensured by adding 50 $mg \cdot L^{-1}$ of SiO_2 (Samchun chemical, Korea) with a mean particle size of 3 μm, to simulate the particulate colloidal matter. To prevent sedimentation of SiO_2 particle, the feed solution was stirred in the feed tank. To measure the turbidity of retentate and permeate solution, turbidity meter (2020we, LaMOTTE, USA) was used. A dead end filtration experiments in outside-in mode were conducted at constant permeate flux of 100 LMH. To ensure the dead end filtration mode, the discharge valve was closed during filtration process. Every 30 min of filtration, a periodical physical cleaning of 1min by backwashing and/or forward flushing was applied using the permeate solution. The backwashing was performed by passing permeate from inside to outside of the fibers, which is reverse to filtration. Forward flushing consists in flowing the permeate solution along the membrane surface to remove foulants deposited on the membrane. Forward flushing flux was equal to the considered permeate flux, while backwashing flux was 2 times higher than the permeate flux. The different cleaning frequencies and durations are detailed in Table 2.

Table 2. Hydraulic Cleaning Condition in this Study

(FF : Forward Flushing, BW : Backwashing)

Cleaning Condition	Filtration	FF	BW	FF
FF	30 min	60 s		
BW			60 s	
FF/BW		30 s	30 s	
BW/FF			30 s	30 s

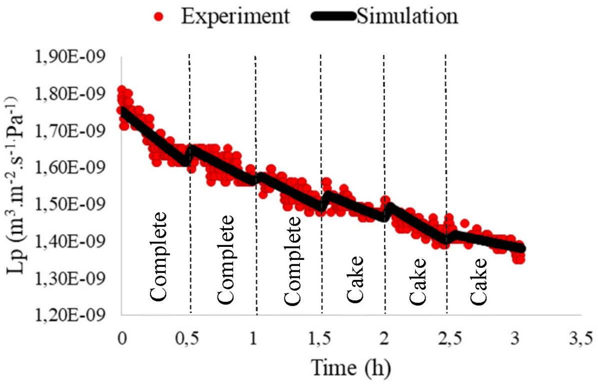

Fig. 2. Comparison of Hermia models with experimental data obtained when applying only forward flushing.

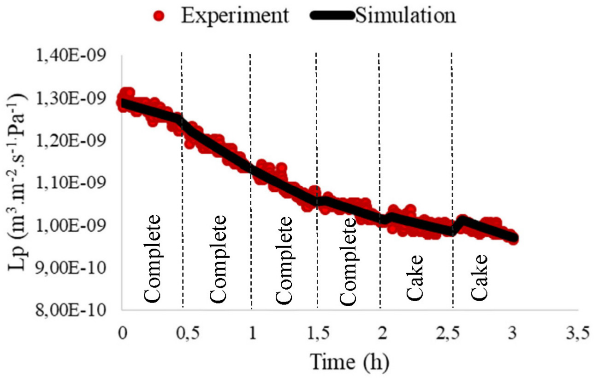

Fig. 3. Comparison of Hermia models with experimental data obtained when applying only Backwashing.

4. Results and Discussion

4.1. Fouling analysis when applying one cleaning technique

To better understand the effect of different hydraulic periodical cleaning on the fouling mechanism responsible of membrane permeability decrease, Hermia models were compared to the experimental permeability data for each filtration cycle. Table 3 displays the values of least squares obtained for each applied model. The model showing the lowest least squares value corresponds to the dominant fouling mechanism. Fig. 2 shows the fitting of the predominant fouling mechanism model with the experimental data obtained when applying only forward flushing for 1 min every 30 min of filtration. Hermia models showed a satisfactory fitting with experimental data. The first 3 cycles were controlled by the complete blocking mechanism and were followed by 3 cycles controlled by cake formation. The values of identified model parameters are displayed in Table 4. The experimental data as well as the model showed that the cleaning strategy applied was not effective to totally regenerate the membrane permeability. An irreversible fouling is accumulated through the filtration cycles. The filtered solution contained only silica particles which particle size distribution analysis showed that even if a majority of particles had higher size than the membrane pores, a considerable fraction had similar size with the membrane pores, which would foster the complete blocking mechanism. The enhanced irreversible complete blocking during the first cycles leads to decrease the membrane porosity which allowed the cake formation mechanism.

Similar analysis of membrane fouling was applied in the case where only backwashing for 1 min was applied every 30 min of filtration. The least squares values for each model are presented in Table 3. Fig. 3 shows the results of fitting between the predominant fouling mechanism model and the permeability experimental data in every filtration cycle. Hermia models described well the permeability decrease. The fitting results shows that complete blocking was the

Table 3. Least Squares Values Obtained when Fitting Hermia Models to Experimental Data Obtained when Applying Forward Flushing or Backwashing Only

Cleaning Cycle	Model	Forward Flushing	Backwashing
		Least squares	Least squares
1st	Cake	9.80×10^{-20}	2.21×10^{-20}
	Complete blocking	8.74×10^{-20}	2.18×10^{-20}
	Pore constriction	9.01×10^{-20}	2.20×10^{-20}
	Intermediate blocking	9.78×10^{-20}	2.20×10^{-20}
2nd	Cake	9.16×10^{-20}	1.33×10^{-20}
	Complete blocking	1.22×10^{-19}	6.19×10^{-19}
	Pore constriction	6.98×10^{-20}	1.32×10^{-20}
	Intermediate blocking	9.28×10^{-20}	1.34×10^{-20}
3rd	Cake	7.18×10^{-20}	2.56×10^{-20}
	Complete blocking	6.08×10^{-20}	2.46×10^{-20}
	Pore constriction	6.10×10^{-20}	2.48×10^{-20}
	Intermediate blocking	6.12×10^{-20}	2.51×10^{-20}
4th	Cake	2.44×10^{-20}	1.11×10^{-20}
	Complete blocking	2.49×10^{-20}	1.09×10^{-20}
	Pore constriction	2.47×10^{-20}	1.11×10^{-20}
	Intermediate blocking	2.46×10^{-20}	1.10×10^{-20}
5th	Cake	3.66×10^{-20}	2.06×10^{-20}
	Complete blocking	3.75×10^{-20}	2.08×10^{-20}
	Pore constriction	3.72×10^{-20}	2.07×10^{-20}
	Intermediate blocking	3.70×10^{-20}	2.07×10^{-20}
6th	Cake	1.63×10^{-20}	1.04×10^{-20}
	Complete blocking	1.66×10^{-20}	1.06×10^{-20}
	Pore constriction	1.66×10^{-20}	1.06×10^{-20}
	Intermediate blocking	1.65×10^{-20}	1.05×10^{-20}

Table 4. Values of Identified Parameters Obtained when Applying Only One Cleaning Technic

Cleaning	Forward flushing		Backwashing	
Cycle n°	Fouling mechanism	Identified constant	Fouling mechanism	Identified constant
1st	Complete	$K_{cb} = 4.7 \times 10^{-5}$	Complete	$K_{cb} = 1.9 \times 10^{-5}$
2nd	Complete	$K_{cb} = 3.2 \times 10^{-5}$	Complete	$K_{cb} = 4.5 \times 10^{-5}$
3rd	Complete	$K_{cb} = 3.3 \times 10^{-5}$	Complete	$K_{cb} = 4.0 \times 10^{-5}$
4th	Cake	$K_{cf} = 1.21 \times 10^{13}$	Complete	$K_{cb} = 2.7 \times 10^{-5}$
5th	Cake	$K_{cf} = 1.95 \times 10^{13}$	Cake	$K_{cf} = 2.03 \times 10^{13}$
6th	Cake	$K_{cf} = 7.30 \times 10^{13}$	Cake	$K_{cf} = 2.98 \times 10^{13}$

dominant mechanism during the first 4 cycles, while cake formation controlled fouling for the 4th and 5th cycles. The values of identified model parameters are presented in Table 4. The cleaning strategy applied in this case seems also inefficient to totally regenerate the membrane permeability. Compared to the case applying

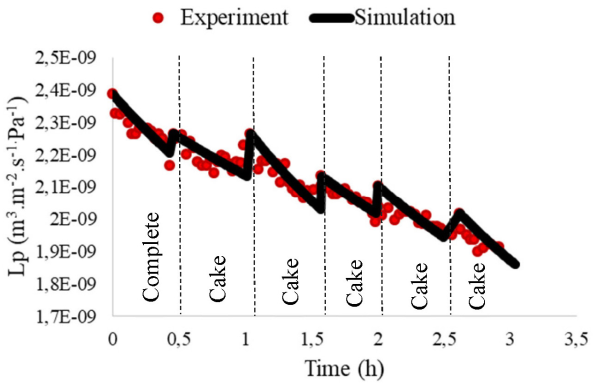

Fig 4. Comparison of Hermia models with experimental data obtained when applying forward flushing followed by backwashing.

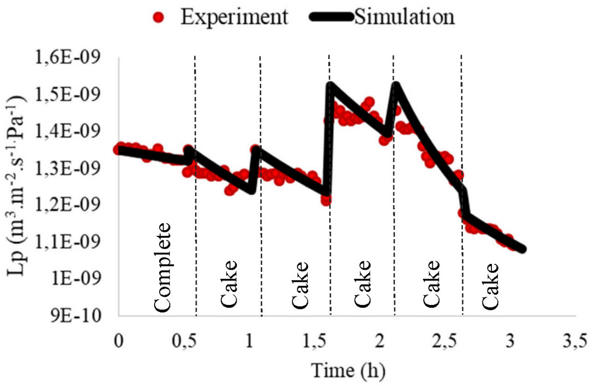

Fig. 5. Comparison of Hermia models with experimental data obtained when applying Backwashing followed by forward flushing.

only forward flushing cleaning, lower permeability was obtained. Moreover, the fouling analysis shows that longer period was controlled by complete blocking which would lead to lower membrane porosity and consequently lower membrane permeability. Applying only forward flushing seems more efficient in membrane regeneration than backwashing. This would be due to the fact that the fouling is mainly a surface fouling and cleaning by forward flushing would be more efficient to mitigate external fouling. In a previous study, Schulz et al., and Heijman et al. reported that inorganic colloidal foulants reduced permeability recovery by backwashing[17-18].

4.2. Fouling analysis when combining cleaning techniques

Fouling mechanisms analysis was also conducted on experimental data registered when combining periodical hydraulic cleaning by Forward Flushing for 30 s and Backwashing for 30 s, every 30 min of filtration. Similarly to the previous cases, the dominant fouling mechanisms were defined based on the least squares values displayed in Table 5. Fig. 4 shows the comparison between predominant fouling mechanism models and the membrane permeability experimental data obtained when applying forward flushing before backwashing. Hermia models display satisfactory fitting with experimental data. While the first cycle experimental data fitted well with the complete blocking model, the 5 following cycles fitted with the cake formation model. The values of identified model parameters are presented in Table 6. Higher permeability was obtained when applying forward flushing followed by backwashing, compared to applying only one technique. This result was observed by Kennedy et al. who estimated that forward flushing would be effective for cake removal while backwashing is effective for pore blocking mitigation, and combining both of them would be more effective for fouling mitigation[11]. Nevertheless, in the studied case this strategy was not effective to totally regenerate membrane permeability since cleaning duration and frequency should be further optimized. The experimental results were confirmed by the theoretical fouling analysis showing cake formation as the main mechanism controlling fouling which leads to lower permeability decrease than the complete blocking mechanism.

Fouling mechanisms analysis was conducted also on permeability data obtained when applying backwashing followed by forward flushing. The least squares values obtained when fitting models with experimental data are displayed in Table 5. The analysis results displayed in Fig. 5, show satisfactory fitting between Hermia models and experimental data. Similarly to the previous case, the first filtration cycle was controlled by complete blocking mechanism while the 5 following cycles

Table 5. Least Squares Values Obtained when Fitting Hermia Models to Experimental Data Obtained when Combining Forward Flushing and Backwashing Only

Cleaning		Forward flushing/Backwashing	Backwashing/Forward flushing
Cycle	Model	Least squares	Least squares
1st	Cake	1.97×10^{-19}	3.57×10^{-20}
	Complete blocking	1.87×10^{-19}	3.54×10^{-20}
	Pore constriction	1.95×10^{-19}	3.56×10^{-20}
	Intermediate blocking	1.92×10^{-19}	3.56×10^{-20}
2nd	Cake	2.81×10^{-19}	1.46×10^{-19}
	Complete blocking	3.21×10^{-19}	1.54×10^{-19}
	Pore constriction	3.21×10^{-19}	1.52×10^{-19}
	Intermediate blocking	3.21×10^{-19}	1.50×10^{-19}
3rd	Cake	3.88×10^{-19}	1.93×10^{-19}
	Complete blocking	4.11×10^{-19}	2.02×10^{-19}
	Pore constriction	4.05×10^{-19}	2.00×10^{-19}
	Intermediate blocking	3.99×10^{-19}	1.97×10^{-19}
4th	Cake	8.90×10^{-20}	2.60×10^{-19}
	Complete blocking	8.99×10^{-20}	2.69×10^{-19}
	Pore constriction	8.97×10^{-20}	2.67×10^{-19}
	Intermediate blocking	8.94×10^{-20}	2.64×10^{-19}
5th	Cake	2.69×10^{-19}	3.10×10^{-19}
	Complete blocking	2.77×10^{-19}	3.74×10^{-19}
	Pore constriction	2.75×10^{-19}	3.56×10^{-19}
	Intermediate blocking	2.73×10^{-19}	3.40×10^{-19}
6th	Cake	1.83×10^{-19}	1.88×10^{-19}
	Complete blocking	1.94×10^{-19}	2.26×10^{-20}
	Pore constriction	1.91×10^{-19}	2.23×10^{-20}
	Intermediate blocking	1.88×10^{-19}	2.20×10^{-20}

Table 6. Values of Identified Parameters Obtained when Applying Only One Cleaning Technic

Cleaning	Forward flushing/Backwashing		Backwashing/Forward flushing	
Cycle n°	Fouling mechanism	Identified constant	Fouling mechanism	Identified constant
1st	Complete	$K_{cb} = 5.2 \times 10^{-5}$	Complete	$K_{cb} = 1.2 \times 10^{-5}$
2nd	Cake	$K_{cf} = 6.14 \times 10^{13}$	Cake	$K_{cf} = 2.87 \times 10^{13}$
3rd	Cake	$K_{cf} = 1.24 \times 10^{13}$	Cake	$K_{cf} = 2.72 \times 10^{13}$
4th	Cake	$K_{cf} = 8.69 \times 10^{13}$	Cake	$K_{cf} = 2.62 \times 10^{13}$
5th	Cake	$K_{cf} = 1.07 \times 10^{13}$	Cake	$K_{cf} = 5.95 \times 10^{13}$
6th	Cake	$K_{cf} = 1.43 \times 10^{13}$	Cake	$K_{cf} = 4.10 \times 10^{13}$

were controlled by cake formation. The values of identified model parameters are presented in Table 6. Even if not effective to regenerate membrane permeability throughout the whole experiment, higher cleaning efficiency was ensured when combining cleaning techniques and especially when starting by backwashing.

5. Conclusion

A theoretical method using the classical blocking laws was considered to identify the fouling mechanisms controlling membrane permeability loss in a pressurized membrane process treating synthetic surface water, where periodic hydraulic cleaning by backwashing and forward flushing is applied. When applying only one cleaning technic, the first filtration cycles were controlled by complete blocking mechanism while the following cycles were controlled by cake formation. However, when combining both cleaning technics, permeability decrease is found due mainly to cake formation. Moreover, higher regeneration efficiency was obtained.

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