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Yinghong Gao

Backpulsing technology for membrane fouling mitigation in produced water treatment

Doctoral thesis

Yinghong Gao

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Trondheim, July 2022

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Abstract

Produced water from oil and gas industry poses an immense threat to the environment due to its substantial volume and complicated composition. Membrane filtrations especially with ceramic membranes can effectively remove emulsified oil and suspended solids and meet the requirements for discharge or reinjection. However, membrane fouling is severe in the treatment of produced water and is the largest obstacle to the wide application of membrane technology. Physical cleaning can mitigate membrane fouling, reduce the frequency of chemical cleaning, thus prolong membrane lifetime, and reduce operational costs. Backpulsing is a promising physical cleaning method, which is induced by periodically reversing the transmembrane pressure (TMP) for a very short duration (typically less than 1 s). It has a transient effect that can effectively mitigate membrane fouling.

In this doctoral work, backpulsing technology applied in microfiltration (MF) and ultrafiltration (UF) processes was reviewed while the experimental rig was built up in the lab. After the setup was finished, commissioned, and optimized, various experiments were carried out to investigate the significance of backpulsing parameters and their interactions on backpulsing efficiency, membrane fouling situations in the filtration of different types of produced water, and fouling mitigation by backpulsing compared with backwashing.

The backpulsing review addresses the fundamentals of backpulsing, applications of backpulsing in different fields and results of pilot- and commercial-scale operations. Factors affecting backpulsing efficiency are illustrated, including feed properties, membrane properties and operating parameters. Mathematical models of backpulsing are overviewed, which could predict membrane productivity or provide a perspective to evaluate backpulsing performance in fouling mitigation. Finally, the existing challenges and outlook are discussed.

Experiments using with a 2^3 full factorial design were carried out to investigate the effect of backpulsing parameters (amplitude, duration and frequency) and their interactions on membrane performance. Results based on Al_2O_3 membranes show that backpulsing was efficient to mitigate membrane fouling. However, the cleaning efficiency varied between different backpulsing conditions. Amplitude was the most crucial variable for fouling removal and final specific flux, while frequency was the most significant one for membrane net yield. Further experiments were conducted to find the optimum backpulsing frequency at a sufficient amplitude (0.5 bar) and a moderate duration (0.6 s).

Ceramic membranes with three types of selective layers (TiO_2, Al_2O_3 and ZrO_2) were tested. For the same type of feed water and under the same filtration conditions, the optimal backpulsing frequencies of the three membranes were in a range of 11 – 15 s. A backpulsing frequency of 12 s was selected for the later backpulsing experiments. Later, ten types of produced water, including two based on treated real produced water, were used in the crossflow microfiltration with the three ceramic membranes mentioned above. Pure fouling experiments were carried out without any cleaning. Fouling behavior due to different compositions of produced water was investigated. Moreover, four out of the ten types of produced water were selected for longer experiments to compare the efficiency of fouling mitigation by backpulsing and backwashing. Backpulsing was in general much better than backwashing in terms of the net yield during the 12 h filtrations. Nevertheless, the trend of flux recovery after each backpulse/backwash was not always backpulsing better than backwashing. The flux recovery was depended on the fouling situation in each specific case. ZrO_2 membrane showed the best performance in the three membranes, which was in accordance with that ZrO_2 membrane had the narrowest membrane pore size distribution.

Preface

This thesis is submitted to the Norwegian University of Science and Technology (NTNU) for partial fulfilment of the requirements for the degree of Philosophiae Doctor (PhD). In accordance with the guidelines of the Faculty of Engineering, the thesis comprises an introduction to the scientific work and four scientific papers/manuscripts.

The work presented in this thesis was carried out at the Department of Civil and Environmental Engineering, NTNU, in Trondheim, Norway. This research was funded by Siemens Energy Inc. It is a sub-project “Advanced treatment of produced water with ceramic membranes” under Siemens-NTNU joint project. Prof. Stein Wold Østerhus at NTNU has been the main supervisor. Prof. Zhiwei Wang from Tongji University, China and Mrs. Andrea Larson from Siemens Energy Inc., USA have been the co-supervisors.

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I am sincerely grateful to Siemens Energy for the financial support. Siemens and NTNU has offered a great platform for me to carry out an interesting industrial project that is a good start for my carrier in industry. Anngjerd Pleym, Chad Felch and Morten Grønli are acknowledged to have given me useful suggestions, support, and guidance.

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List of publications

Journal papers

1. Yinghong Gao, Jie Qin, Zhiwei Wang, Stein Wold Østerhus. *Backpulsing technology applied in MF and UF processes for membrane fouling mitigation: A review*. Journal of Membrane Science, 587 (2019) 117136.
2. Yinghong Gao, Yeqing Zhang, Marcin Dudek, Jie Qin, Gisle Øye, Stein Wold Østerhus. A multivariate study of backpulsing for membrane fouling mitigation in produced water treatment. Journal of Environmental Chemical Engineering, 9 (2021) 104839.
3. Yinghong Gao, Yasser K. Abdelhamed, Marcin Dudek, Junli Wan, Xiaoyang Guo, Gisle Øye, Stein W. Østerhus. Application of ceramic membranes for produced water treatment. Part I: Effect of feed compositions on membrane fouling. In preparation.
4. Yinghong Gao, Yasser K. Abdelhamed, Marcin Dudek, Xiaoyang Guo, Junli Wan, Gisle Øye, Stein W. Østerhus. Application of ceramic membranes for produced water treatment. Part II: Comparison of backpulsing and backwashing for membrane fouling mitigation. In preparation.

Conference presentations

1. Yinghong Gao, Stein Wold Østerhus. *Investigation of backpulsing technology to mitigate membrane fouling*. The 8th IWA-MTC Conference, Singapore, 5 – 9 September 2017. Oral presentation.
2. Yinghong Gao, Yasser K. Abdelhamed, Marcin Dudek, Xiaoyang Guo, Junli Wan, Gisle Øye, Stein W. Østerhus. *Comparison of backpulsing and backwashing for membrane fouling mitigation in produced water treatment*. The 12th ICOM, online, 7 – 11 December 2020. Poster presentation.

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Symbols and Abbreviations

API	American petroleum institute
CFV	Crossflow velocity
CFU	Compact flotation unit
CMF	Crossflow membrane filtration
CPI	Corrugated plate interceptor
DGF	Dissolved gas flotation
IGF	Induced gas flotation
LMH	Liter per square meter per hour
MF	Microfiltration
NCS	Norwegian continental shelf
NSF	Nutshell filter
SG	Specific gravity
TMP	Transmembrane pressure
UF	Ultrafiltration

Chapter 1 Introduction

This chapter gives a brief introduction to the background, problem statement, and scope and objectives of the doctoral work. An overview of the thesis structure is also included.

1.1. Background and problem statement

Produced water is difficult to handle and poses a big threat to the environment if not treated properly. It is a by-product generated during the production of oil and gas and is the largest waste stream by volume. Its characteristics change considerably depending on the geographic location of the field, the type of hydrocarbon product being produced, the lifetime of the reservoir, and the chemical additives used during production process [1, 2]. On the Norwegian Continental Shelf (NCS), 1583 tons of oil was discharged to the sea, from which 1487 tons of oil was from produced water [3]. Reducing the risks from produced water has been focused on the dispersed oil content [4]. With the stringent requirement for produced water reinjection [5] and more strict requirement for discharge [6], membrane filtration has been considered as a potential technology due to the efficient oil removal even though the feed water may have a broad compositional range [7]. Membrane filtration is capable to remove the smallest (< 10 μm) and most stable oil droplets from produced water [8, 9]. However, the key obstacle that hinders widespread application of membrane technology for produced water treatment is membrane fouling. The mechanisms of membrane fouling caused by emulsified oil remain poorly understood [7]. Oil droplets that can both deform and coalesce are unique foulants.

Backpulsing technology is a promising technology and has the transient effect that can knock off foulants from the membranes [10, 11]. It is conducted every few seconds for a very short duration (typically less than 1 s) [12, 13]. The fundamental difference between backpulsing and backwashing is the speed and force utilized to dislodge foulants from the membrane. In backpulsing, reverse cleaning occurs every few seconds at high pressure for very short time, while the flow reversal lasts for 5 – 30 s once every 30 min to one hour [14].

The previous project at our department ("TOP water" project, Petromaks program, NFR 2005-2010) investigated produced water treatment with ceramic membrane microfiltration [15]. High frequency back-pulsing was found to be an efficient strategy for membrane fouling control for all the three membranes (0.1 µm, 0.2 µm and 0.5 µm Al_2O_3 membranes) tested in the filtration of feed with different properties. Besides, the permeate quality was also improved when backpulsing technique was employed [16].

Although backpulsing has been tested to be an effective method for fouling mitigation since 1989 when Victor Rodgers found significant improvement of permeate flux in the ultrafiltration of a single solute (1% albumin), and various studies have covered the topics including theory of backpulsing, application of backpulsing in different fields, factors affecting backpulsing efficiency, and backpulsing modeling to predict optimum conditions and understand the mechanisms, there has not been a comprehensive summary of the state-of-the-art of this technology. Similar reviews have been published for backwashing technology at least twice. One reviews backwashing as a fouling control strategy used for membrane bioreactors in wastewater treatment [17]. The other focuses on backwashing for low-pressure membranes in drinking water treatment [18]. Besides, there has not been any research focusing on the comparison of backpulsing and backwashing on membrane fouling mitigation under the same amplitude and accumulated reverse cleaning time. The assumptions on fouling mitigation mechanisms have not been fully investigated and proved by experiments.

1.2. Scope and objectives

This research belongs to the industrial project of 'Advanced treatment of produced water by membrane filtration', which was sponsored by Siemens. The project was based on the practical problems of severe membrane fouling occurring in the treatment of produced water in the oil and gas industry. The main objectives of the doctoral work are to investigate and understand

1) the state-of-the-art of backpulsing technology applied in MF and UF processes for membrane fouling mitigation (Paper I).
2) the effect of backpulsing parameters and their interactions on membrane fouling mitigation (Paper II).
3) membrane fouling phenomena caused by different produced water properties (Paper III).
4) the comparison of backpulsing and backwashing on membrane fouling mitigation (Paper IV).

1.3. Structure of thesis

Chapter 1 provides an introduction to this doctoral thesis, including background and problem statement, scope and objectives, and structure of thesis.

Chapter 2 reviews the background of produced water treatment and membrane fouling mitigation using backpulsing and backwashing technology. Knowledge gaps are identified, and research questions are raised.

Chapter 3 describes materials to carry out experiments, membrane filtration setups used for different experimental purposes, analytical methods to investigate water quality, and modeling to assist experimental design and understand membrane fouling.

Chapter 4 summaries the main findings of each research paper and discusses on how different papers are related to each other and how different papers answers the research questions.

Chapter 5 draws the conclusions from this thesis and outlines some recommendations for future work.

Appendix A encloses research papers.

Appendix B shows the experimental results on optimal backpulsing frequency.

Chapter 2 Background

This chapter reviews the background and knowledge status of the topics addressed in this thesis. Knowledge gaps are identified, and research questions are raised.

2.1. Crude oil

2.1.1. Classification of crude oil

Crude oil is a mixture of hydrocarbons that exist in the underground reservoirs. The American Petroleum Institute (API) gravity expresses the density of crude oil and measures how heavy or light a crude oil is compared to water [19]. API gravity is calculated using the specific gravity of crude oil as follows:

$$\text{API gravity} = (141.5/SG) - 131.5 \qquad (1.1)$$

where Specific Gravity (SG) is determined at 60 °F (15.6 °C), kg/m^3.

Crude oil is classified as light, medium, or heavy according to its API gravity, as the weight of an oil is the largest determinant of its market value. API gravity of water is 10.0°.

- **Light oil**: API > 31.1° (SG < 870 kg/m^3)
- **Medium oil**: 22.3° < API < 31.1° (870 < SG < 920 kg/m^3)
- **Heavy oil**: 10.0° < API < 22.3° (920 < SG < 1000 kg/m^3)
- **Extra heavy oil**: API < 10.0° (SG > 1000 kg/m^3)

2.1.2. Main components of crude oil

The composition of crude oil is complicated and highly dependent on the geographic regions and oil fields. It normally contains liquids, solids, and some dissolved gases. The liquids consist of saturates, aromatics, and resins, and the most dominant solid component is asphaltene. These four components are grouped together as the main divisions of a crude oil, SARA composition. SARA analysis is based on the polarity and solubility of the components and performed using chromatography technology [20]. Demonstration of molecular structures of the SARA groups is shown in Figure 2-1.

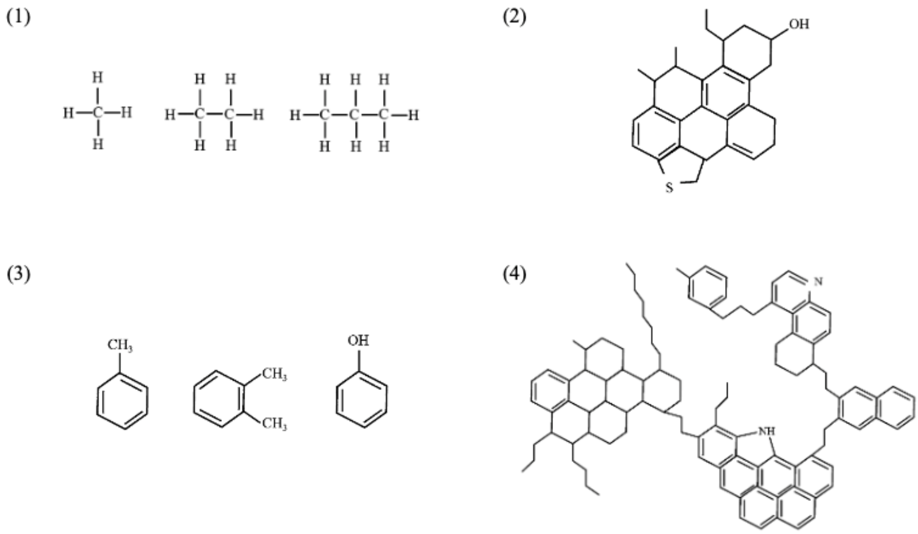


Figure 2-1. Examples of molecular structures of (1) saturates, (2) aromatics, (3) resins, and (4) asphaltenes. Adapted from [21].

- **Saturates** are saturated hydrocarbons with carbon atoms bonding to the maximum allowable hydrogen. They can be straight, branched, or cyclic configurations, and account for the main components of a crude oil.
- **Aromatics** contain one or more aromatic rings and sometimes heteroatoms. They are the second main components of a crude oil and are slightly more polarizable than saturates.
- **Resins** are more complex in structure than the first two groups and have more polar moieties. Resins are important for the stabilization of asphaltenes in crude oils [22].
- **Asphaltenes** have different structures and molecular makeup, which makes them among the most complex components of oils [21]. They are generally classified as insoluble in n-alkanes [23]. Asphaltenes are highly polar. Resins have both polar and nonpolar sides and thus function as a bridge that connects the nonpolar hydrocarbon compounds to the highly polar asphaltenes [24].

2.2. Produced water

2.2.1. Classification of oil in water by size

Oil droplets in water can be divided into four categories according to the size [25-27].

- **Free oil**: d > 150 µm. The API defines free oil as oil droplets larger than 150 µm in diameter which are large enough to be separated efficiently from water by gravity difference in open style separation chambers, such as API gravity separator (skimmer).
- **Dispersed oil**: 5 µm < d < 20 µm. Historically, dispersed oil are the oil droplets that do not separate readily by gravity and lie between free oil and emulsified oil. With the development of the corrugated plate interceptor (CPI), practical removal of oil droplets lager than 20 µm became feasible via coalescence and gravity separation. As coalescing separators are widely used, it is common for droplets of 20 µm and larger to be considered as free oil, leaving droplets with size between 5 and 20 µm as truly dispersed oil.
- **Emulsified oil**: d < 5 µm. Droplets with size smaller than 5 µm exhibit Brownian movement and almost never separate by gravity or coalescence. They are normally made mechanically by disperser or chemically by surfactant.
- **Dissolved oil**: droplet size of nanometer. Dissolved components in produced water are mainly organic acids, BETX (benzene, toluene, ethylbenzene, and xylene), phenols, and some PAH (polycyclic aromatic hydrocarbon) [3].

2.2.2. What is produced water?

In oil and gas reservoirs, the natural rocks normally contain both petroleum hydrocarbons (liquid and gas) and water. The water is often referred to as “connate water” or “formation water”, which has been in contact with the hydrocarbons for thousands of years. Produced water is the water that is brought to the surface together with crude oil and/or natural gas [2]. Sources of produced water can be the original connate water/formation water and the injected fluids including additives resulting from production activities.

Produced water is the largest volume waste stream generated during the production of oil and gas. The water to oil ratio is different from well to well, depending on the geological conditions and the age of the well. In general, 3 barrels of produced water are produced for each barrel of oil extracted worldwide [1]. On the NCS, the water/oil volume ratio increased from about 0.2 in 1993 to near 2.0 in 2018 [3]. The global annual produced water volume was estimated to increase over years [28].

The physical and chemical properties of produced water are complicated, site-specific and changing over time [29], depending on the geographic location of the reservoir, the geochemistry of the formation, the type of hydrocarbon being produced, as well as the lifetime of a producing well [2]. Produced water typically contains dispersed oil, dissolved organic compounds, inorganic compounds, production chemicals, solids, and heavy metals [30, 31]. Many of these components can pose a great threat to the environment, if not treated properly.

2.3. Produced water treatment

2.3.1. Offshore treatment goals

There are two main approaches for disposal of produced water offshore: discharge into the ocean or reinject back to the reservoir [4, 32]. On the NCS, the current regulatory threshold of oil content for discharge is 30 mg/L monthly average [33]. A new guideline for hydrocarbons exploration and production was published by European Commission that oil companies in Europe were pushed to minimize discharges of dispersed oil to < 15 mg/L [6]. Reinjection of treated water to a disposal or production reservoir can sustain the production pressure of the reservoir [34] and improve oil recovery [32], which is regarded as a means to reduce harm to the environment caused by produced water [35]. On the NCS, produced water reinjection increased substantially from 14% in 2003 to 22-24% in recent years [3]. However, this process carries several risks, such as reservoir souring [36], formation damage due to reduced injectivity [37], or clog of pores in the reservoir because of the presence of suspended solids and dispersed/emulsified oil [38-40]. More stringent reduction of both oil content and suspended solids is required for reinjection option in restrictive reservoirs [41].

2.3.2. Overview of produced water treatment technologies

A general overview of the common treatment technologies in platforms is shown in Figure 2-2. The techniques for produced water treatment are typically divided into three categories.

- **Primary**: predominantly gravity-based separation sometimes with the help of coalescence, targeting the three-phase separation of solids, water, and oil (free oil and much of the dispersed oil).
- **Secondary**: further removal of dispersed oil.
- **Tertiary**: water polishing step, focusing on the removal of emulsified oil and dissolved components.

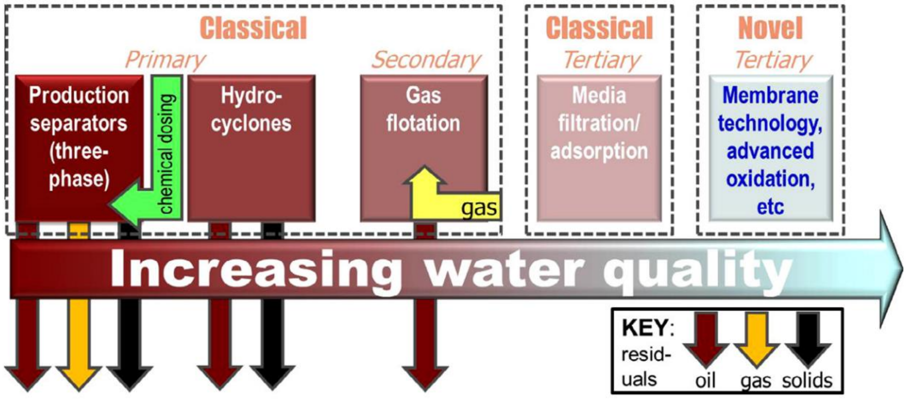


Figure 2-2. Overview of offshore produced water treatment, source from S. Judd et al, 2014 [27].

A summary of the physical and chemical units for produced water treatment is shown in Table 2-1, including both onshore and offshore techniques. Note that the data in this table are more related to industrial applications, as they are captured mainly from manual books and industrial resources (technology suppliers, consultants, contractors, and end users). The oil removal efficiency listed in Table 2-1 is a general range and is affected by inlet oil concentration, oil droplet size, and operating conditions.

Table 2-1. A summary of single industrial units for produced water treatment. Adapted from [27]

Technology	C_{in} (mg/L)	C_{out} (mg/L)	Oil removal (%)	Min. drop dia. (µm)
Primary treatment				
API gravity separator	5 000 – 20 000	50 – 100	80 – 90	150
CPI	5 000 – 20 000	40	90 – 98	20 – 40
Hydrocyclone	100 – 20 000	20 – 80	90 – 95	12 – 20
Secondary treatment				
IGF	200 – 500	25 – 50	90 – 95	10 – 25 10 with optimized chemicals [42]
CFU	200 – 500	15 – 25	90 – 95	10 – 25
DGF	200 – 500	15 – 25	95 [43]	10 – 25 3 with optimized chemicals [42]
Tertiary treatment				
NSF	20 – 50	2 – 5	99	2
CMF	20 – 50	< 1	99	< 1

Abbreviations: CPI, corrugated plate interceptor; IGF, induced gas flotation; DAG, dissolved gas flotation; CFU, compact flotation unit; NSF, nutshell filter; CMF, crossflow membrane filtration.

2.3.3. Membrane filtration

As is shown in Table 2-1 that membrane filtration is a promising technique for the stricter requirement of produced water treatment. Its distinct advantages over conventional treatment methods are efficient separation of oil/water mixture, small footprint, and ease of operation [5, 44, 45]. Recently, there has been a growing interest of using ceramic membranes for the filtration of oily wastewater due to its high mechanical, chemical and thermal stability, as well as the good tolerance to high oil content and other foulants [46, 47]. Besides, the typical disadvantage of high cost for ceramic membranes can be compensated by their longer lifetime, robustness and better performance compared to polymeric membranes [5]. Various studies of microfiltration and ultrafiltration have been carried out aiming at the removal of dispersed and emulsified oil.

Nevertheless, membrane fouling is the main obstacle to the more widespread application of membrane technology in produced water treatment. This is more complicated for the filtration of oil-in-water emulsions, as it is distinct from the fouling by other rigid foulants in terms of deformation and coalescence of oil droplets [48]. A continuous oil layer may be formed and cover the membrane surface after some time filtration [8]. The fouling degree in the filtration of produced water is strongly affected by the feed properties. It is shown in many studies that membrane flux decreased with increasing feed oil concentration and increased with increasing temperature [26, 49]. In the microfiltration of four different oil-in-water emulsions made by four oil types (namely hexadecane, soybean oil, fish oil and crude oil), Tanudjaja and Chew [50] found that the rate of surface coverage increase and the initial transmembrane pressure (TMP) increase was the fastest for crude oil at a constant flux of 125 LMH. pH is also an important factor, as the charges of membrane surface and oil droplets are influenced by the feed pH, which affects the interactions between membrane surface and oil droplets [51]. All oil and gas reservoirs produce some solids along with produced water [52]. Fine particles passing through pretreatment process and entering membrane filtration contributes to membrane fouling. However, the mechanisms of membrane fouling caused by the different constituents of produced water remained poorly understood due to the complexity of the water itself and lack of experience [7, 8]. **(Knowledge gap for Paper III)**

2.4. Backpulsing technology

To control membrane fouling, physical cleaning methods are prioritized before chemical methods, as they are cost-efficient and environmentally friendly.

2.4.1. What is backpulsing?

Backpulsing is a promising physical cleaning method. It is induced by periodically reversing the TMP for a very short duration (typically less than 1 s [12, 13, 53] and used in conjunction with surface tangential flow [54]. The deposited foulants on the membrane surface or in the membrane pores are dislodged by backpulses and swept away by the tangential flow, such as crossflow in crossflow filtrations [12] and air sparging in immersed membrane systems [55]. It is also named backshocking [56-59], high-frequency retrofiltration [10, 11, 60, 61], or transmembrane pressure pulsing [62-67] in literature. There are three basic parameters associated with backpulsing: amplitude, duration and frequency. Amplitude is defined as the absolute value of the negative TMP during each backpulse. Duration is the time each pulse lasts. Frequency is defined as the inverse of the sum of backpulsing duration and forward filtration time [12, 13]. It indicates the interval of two consecutive pulses. Besides, backpulsing volume is also of interest, which means the amount of clean water consumed for each backpulse.

The backpulsing efficiency towards membrane fouling mitigation is strongly affected by the setting of backpulsing parameters. Literature shows that backpulsing frequency [13, 68, 69] and amplitude [12, 54, 70] were important parameters for the permeate flux, while duration seemed to play different roles in different studies [71]. However, the studies in literature were all based on one-variable-at-a-time methods. There is no systematic investigation on the importance of the three backpulsing parameters and their potential interaction between each other. (**Knowledge gap for Paper II**)

There has been an increasing number of publications on backpulsing technology, including various topics, such as backpulsing performance in different applications, optimization of operating parameters, as well as modeling for flux prediction and mechanism investigation. However, there has not been any literature review on this technology. (**Knowledge gap for Paper I**)

2.4.2. Backpulsing vs. backwashing

Both backpulsing and backwashing are reverse flow cleaning and aim at hydraulically reversible fouling. However, they are two different techniques, in terms of names, definitions, and fouling removal mechanisms. The fundamental difference is the utilized speed and force [72, 73]. Backpulsing happens in a fraction of a second every few seconds [74], while backwashing usually lasts for a few seconds or minutes every few minutes or an hour [18]. Theoretically, backpulsing has a transient effect that can knock off foulant from membrane surface before the fouling layer becomes more compact over longer filtration [53], instead of only relying on the shear force generated by backwashing.

Backwashing is commonly used in all kinds of membrane applications. Backpulsing is also getting to know by industry as companies like Atech Innovations, Pall, and Novasep supply membrane systems with backpulsing equipment. However, there has not been any study comparing the performance of backpulsing and backwashing on membrane fouling mitigation with the same downtime and amplitude, let alone the recommendations of using backpulsing/backwashing for specific cases. (**Knowledge gap for Paper IV**)

Note that the background on backpulsing technology has been comprehensively reviewed in Paper I. Knowledge gaps have been fully described in each paper/manuscript, see Appendix A. They are only briefly mentioned in this chapter.

2.5. Research questions

The following research questions were formulated for the work:

Q1: What is the effect of backpulsing parameters (amplitude, frequency, and duration) and their interactions on membrane fouling mitigation?

Q2: What are the main fouling components to membranes in produced water treatment? What is the membrane fouling situation in produced water treatment?

Q3: What is the key factor(s) of membranes that affect membrane performance in the treatment of produced water?

Q4: What is the difference between backpulsing and backwashing, in terms of fouling formation and fouling mitigation? How does backpulsing/backwashing perform in produced water treatment?

Chapter 3 Methods

This chapter presents the experiments that were carried out to address the main objectives of the doctoral work. Materials including membranes and produced water, experimental setup for the membrane filtration systems, analytical methods for the measurement of feed and permeate quality, and modeling to help analyze experimental data are briefly presented in this chapter. More detailed descriptions are in the Paper II to Paper IV.

3.1. Materials

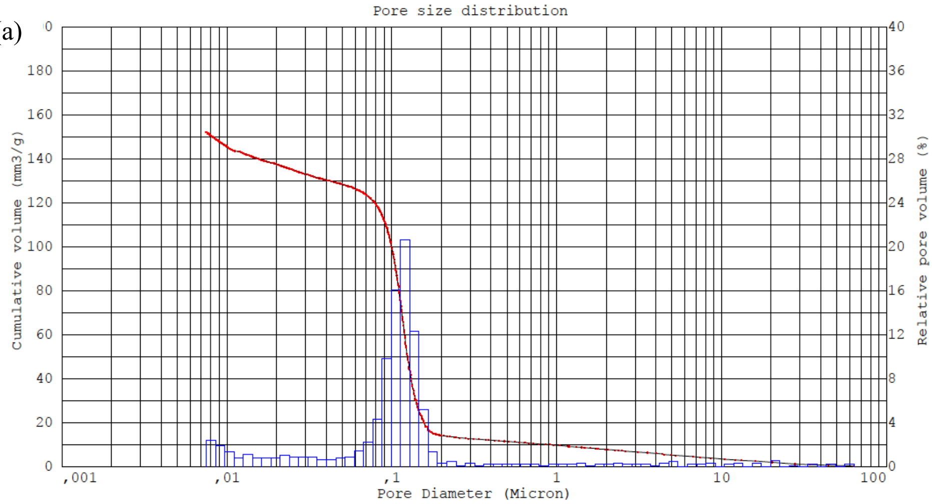
3.1.1. Membranes and modules

Ceramic membranes used in this doctoral work were commercially available mono-tubular membranes. MF membranes with a nominal pores size of 0.1 µm were chosen mainly due to the efficient oil removal, high productivity, and low fouling tendency [41, 73]. A summary of membranes used in this thesis is listed in Table 3-1. Although the nominal pore sizes of the three membranes used in Paper III, Appendix B and Paper IV were the same, the pore size distributions of different membranes were different, as shown in Figure 3-1.

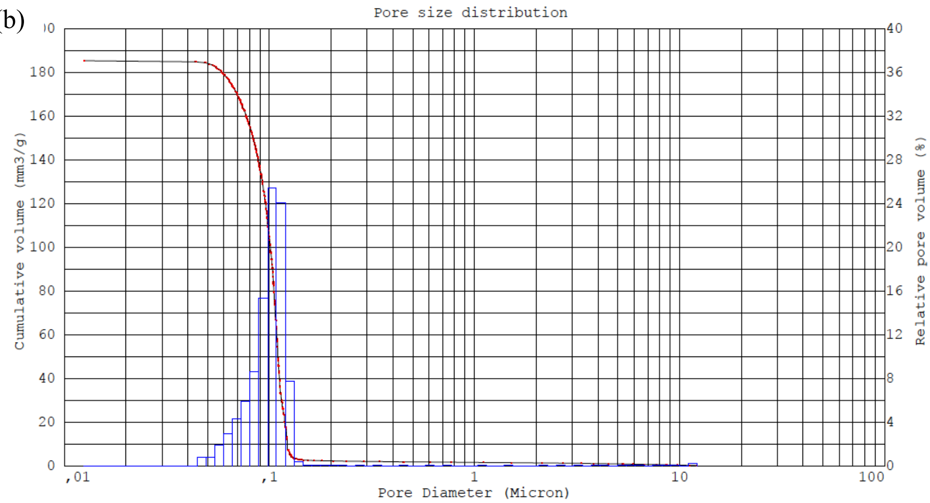
Table 3-1. Ceramic membranes used in this thesis.

	Paper II	Paper III, Appendix B & IV
Nominal pore size (µm)	0.1	0.1
Inner/Outer diameter (mm)	8/11	6/10
Length (mm)	340	340
Selective layer material	α-Al_2O_3	TiO_2, α-Al_2O_3, ZrO_2
Selective membrane area (cm^2)	85	60
Supplying company	ECO-Ceramics, the Netherlands	Atech Innovations GmbH, Germany

(a)


Pore size distribution
Cumulative volume (mm3/g)
Relative pore volume (%)
Pore Diameter (Micron)
,001
,01
,1
1
10
100

(b)


Pore size distribution
Cumulative volume (mm3/g)
Relative pore volume (%)
Pore Diameter (Micron)
,01
,1
1
10
100

(c)

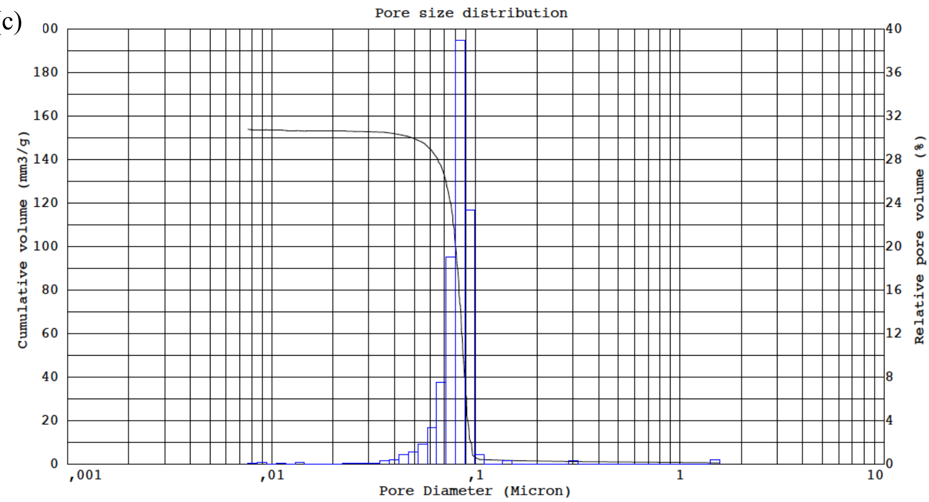


Figure 3-1. Pore size distribution of membranes. (a) TiO_2 membrane, (b) Al_2O_3 membrane, (c) ZrO_2 membrane.

Photos of the membranes and modules used in this thesis is shown in Figure 3-2. The three modules are mounted together. They share the same inlet and retentate outlet by using flange connection. Each membrane module has one separate permeate outlet. This design allows to run parallel experiments under the same operational conditions (i.e., TMP, CFV, temperature) and using the same feed water.

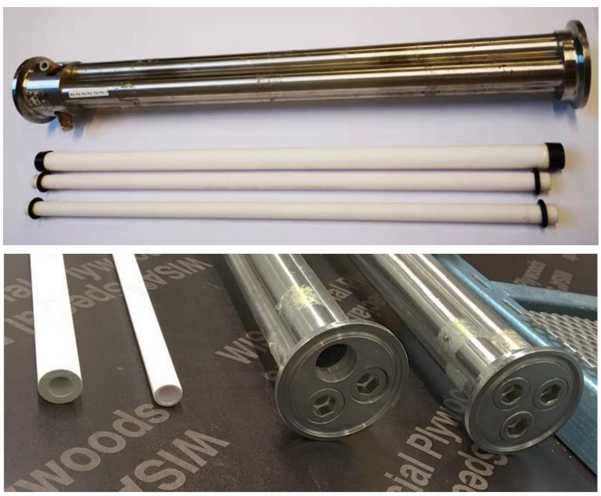


Figure 3-2. Photos of membranes and modules used in this doctoral work.

3.1.2. Produced water

Produced water after pretreatment and before entering membrane filtration contains generally hydrocarbons mainly in the form of emulsified oil, formation water from reservoir, particles, and other chemical additives. Eleven types of produced water were prepared and tested in this doctoral work, as shown in Table 3-2. They consisted of different crude oils, brines, and particles. The detailed information of the two types of crude oils from the NCS is listed in Table 3-3. Crude oil 1 was a lighter oil with an API gravity of 35.2°. Crude oil 2 was heavier and with an API gravity of 23.0°. A non-ionic surfactant, Tween 80, was used to help emulsify oil. Two types of brines were to simulate salinity conditions similar to the produced water from the NCS which was reported to have comparable salinity as seawater [75]. One brine was Na-Brine prepared by dissolving 35 g/L NaCl (VWR) into tap water. The other is NaCa-Brine which was made by mixing 32.35 g/L NaCl and 1.75 g/L $CaCl_2$ (VWR) into tap water. Both brines had the same ionic strength (I = 0.6 M). Extra particles were added to PW6 – PW10. One type was kaolin microparticles (K7375, Sigma-Aldrich) with a particle size distribution of 0.1 – 4 µm. The other type was silica nanoparticles (Aerosil 200, Evonik Resource Efficiency GmbH) with an averaged particle size of 12 nm. PW9 (PW_{Real1}) and PW10 (PW_{Real2}) were to mimic real situations based on a type of treated real produced water from the NCS. The treated real produced water was after the treatment of separation and filtration, containing a residual oil concentration of about 10 mg/L and 500 mg/L corrosion particles due to improper storage in jerry cans. Total dissolved solids (TDS) of the treated real produced water were 44 200 mg/L, while those of Na-Brine and NaCa-Brine were 34 500 and 34 300 mg/L, respectively.

Oil emulsions were generated by using a homogenizer (Ultra-Turrax S25N-10G, IKA, Germany) at 10 000 rpm for 7 min. The homogenizer can emulsify max 2 L liquid per batch. 16 L (8 batches) produced water was prepared for each experiment.

PW0 with a higher concentration of Tween 80 was tested in Paper II as it is important to keep the feed properties constant and stable for all the experiments with different backpulsing conditions. PW1 – PW10 were tested in Paper III where membrane fouling caused by different feed properties was investigated. PW1, PW3, PW6 and PW9 were used in Paper IV for the comparison of backpulsing and backwashing on membrane fouling mitigation.

Table 3-2. Compositions of the eleven types of produced water used in this thesis.

Name	Oil	Surfactant	Brine	Particles
PW0	Crude oil 1 (254 mg/L)	Tween 80 (25 mg/L)	Na-Brine	No
PW1	Crude oil 1 (250 mg/L)	Tween 80 (10 mg/L)	Na-Brine	No
PW2	Crude oil 1 (250 mg/L)	Tween 80 (10 mg/L)	NaCa-Brine	No
PW3	Crude oil 2 250 mg/L)	Tween 80 (10 mg/L)	NaCa-Brine	No
PW4	Crude oil 1 (250 mg/L)	Tween 80 (10 mg/L)	Na-Brine (pH=9)	No
PW5	Crude oil 1 (250 mg/L)	Tween 80 (10 mg/L)	Na-Brine (pH=4)	No
PW6	Crude oil 1 (250 mg/L)	Tween 80 (10 mg/L)	NaCa-Brine	Kaolin (50 mg/L)
PW7	Crude oil 1 (250 mg/L)	Tween 80 (10 mg/L)	NaCa-Brine	Silica Nanoparticles (50 mg/L)
PW8	Crude oil 2 (250 mg/L)	Tween 80 (10 mg/L)	NaCa-Brine	Silica Nanoparticles (50 mg/L)
PW9 (PWReal1)	Crude oil 1 (240 mg/L)	Tween 80 (10 mg/L)	Treated real PW + Kaolin (10 mg/L)	
PW10 (PWReal2)	Crude oil 2 (240 mg/L)	Tween 80 (10 mg/L)	Treated real PW + Kaolin (10 mg/L)	

Abbreviations: PW, produced water.

Table 3-3. Physiochemical properties and compositions of crude oils used in this thesis.

Parameter	Crude oil 1	Crude oil 2
API gravity (°)	35.2	23.0
Density at 20 °C (g/cm^3)	0.847	0.911
Viscosity at 20 °C (mPa•s)	11.05	74.4
TAN (mg of KOH/g of oil)	< 0.1	2.7
TBN (mg of KOH/g of oil)	0.9	1.1
SARA composition [% (w/w)]		
Saturates	74.81	64.9
Aromatics	20.92	26.3
Resins	4.25	8.4
Asphaltenes	0.02	0.4

Abbreviations: TAN, total acid number; TBN, total base number.

3.2. Membrane filtration system

3.2.1. Experimental setup for Paper II

Membrane filtration system for Paper II was under constant-flux mode to test backpulsing experiments with various backpulsing conditions. The schematic flow diagram is shown in Figure 3-3. Two membrane modules were running at the same time and under the same operational conditions. Crossflow velocity (CFV) was 2 m/s generated by a centrifugal pump with a speed regulator. Permeate fluxes were kept constant by peristaltic pumps at 100 L/(m^2h) (LMH) by automatically adjusting transmembrane pressure (TMP). Each experiment ran 4 800 s (1.3 h). Pressure values were recorded at a frequency of 2 Hz. Experiments ran at room temperature. Permeate fluxes were corrected to 20 °C considering the changes in water viscosity under various temperatures.

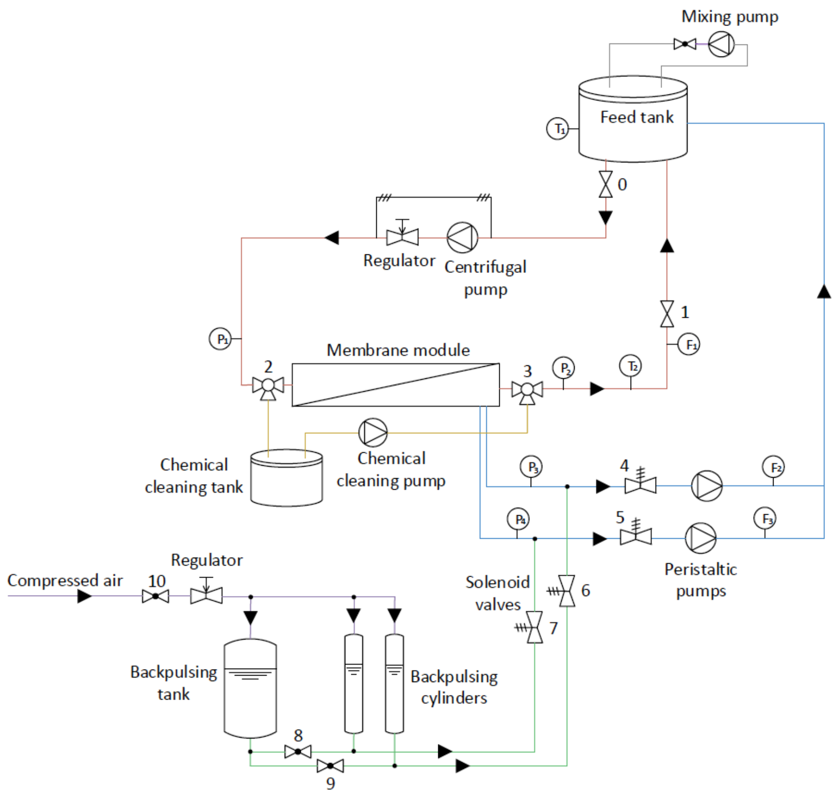


Figure 3-3. A schematic flow diagram of experimental setup for Paper II.

Backpulsing conditions were set according to a 2^3 full factorial design with center points. This is described in section 3.4.1. After each experiment, membranes were chemically cleaned with 1%-v of Derquim+ (PanReac AppliChem, Germany) for 1 h at 60 °C and 2.8 m/s CFV. More than 90% flux recovery was achieved. If not, chemical cleaning was repeated.

3.2.2. Experimental setup for Paper III

Membrane filtration system for Paper III was under constant-pressure mode without any reverse flow cleaning, see Figure 3-4. Three membrane modules were running at the same time and under the same operational conditions. CFV was 2 m/s and TMP was kept constant at 0.5 bar by adjusting the feed pump speed and the gate valve 2. Experiments were under room temperature. The permeates were collected into vessels on electronic balances (Model Navigator®-NV2101, Ohaus, USA) interfaced with a computer that recorded values of permeate mass at 1 s intervals. Permeate fluxes were averaged by 12 s and corrected to 20 ºC. All the experiments were carried out for 7200 s (2 h). New membranes were used in each filtration experiment. Clean membrane fluxes were measured with distilled water for 120 s (2 min) before each filtration run.

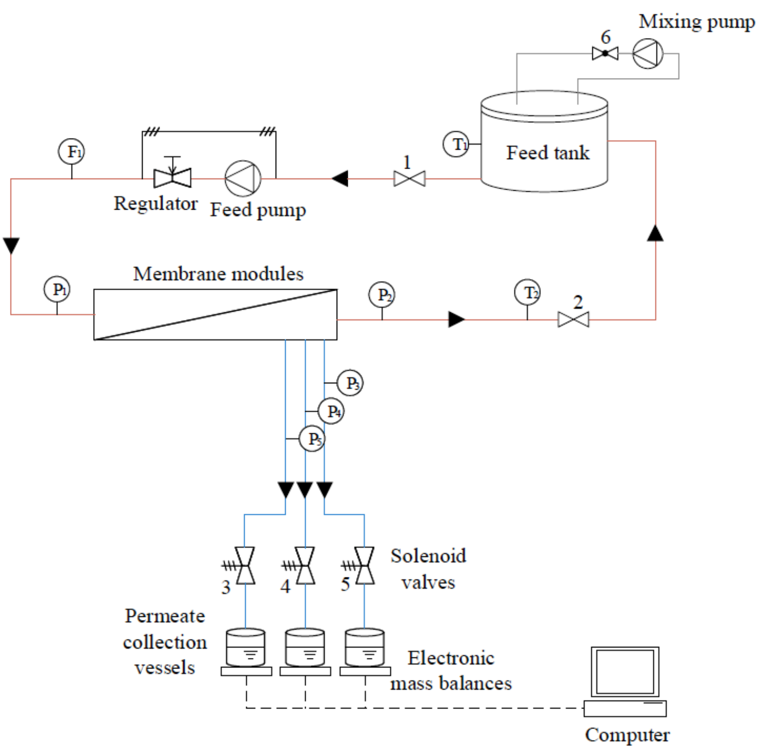


Figure 3-4. A schematic flow diagram of experimental setup for Paper III.

3.2.3. Experimental setup for Appendix B and Paper IV

Membrane filtration system for Appendix B and Paper IV was under constant-pressure mode with backpulsing/backwashing (Figure 3-5 and Figure 3-6). New membranes were used in each filtration experiment. The forward filtration conditions were the same as for Paper III that CFV = 2 m/s, TMP = 0.5 bar, and room temperature. Time breaks were set between the switch of valves to avoid any potential disturbance between backpulsing and filtration. A 1 s break was set after forward filtration and before backpulsing started. There was a 0.5 s break after backpulsing and before next forward filtration.

The experiments for Appendix B were carried at to find the optimal backpulsing frequency at a fixed backpulsing amplitude of 0.5 bar and a fixed backpulsing duration of 0.6 s. Each experiment ran 10 000 s (2.8 h). For Paper IV, each experiment was extended to 12 h to investigate the effect of backpulsing/backwashing on membrane fouling mitigation. The forward filtration conditions were the same as for Paper III. Backpulsing was carried out every 12 s with a duration of 0.6 s and an amplitude of 0.5 bar. backwashing was conducted every 600 s with a duration of 30 s and an amplitude of 0.5 bar.

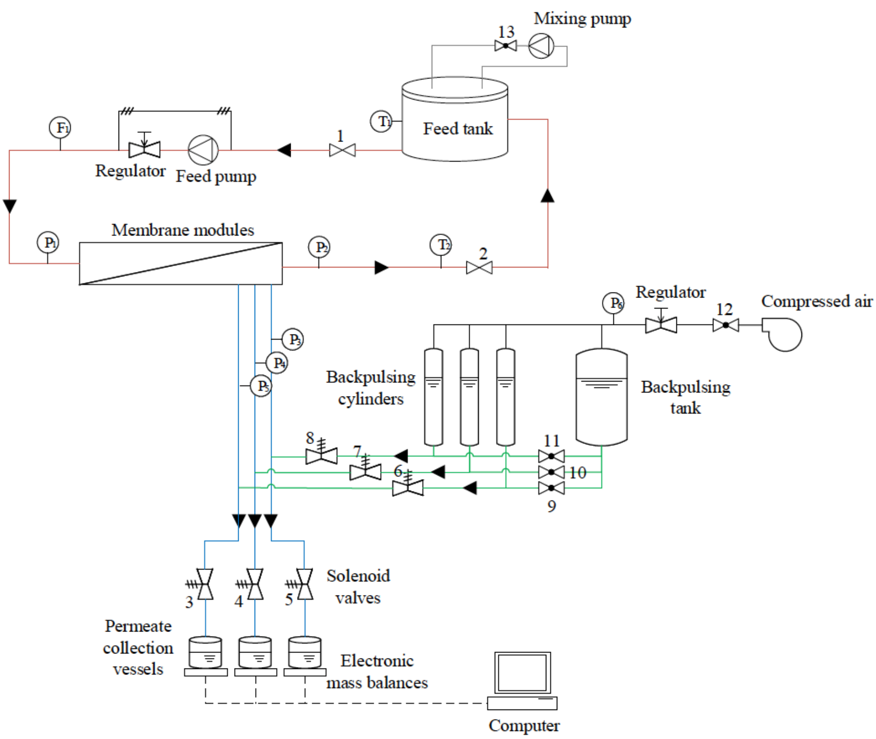


Figure 3-5. A schematic flow diagram of experimental setup for Paper IV.

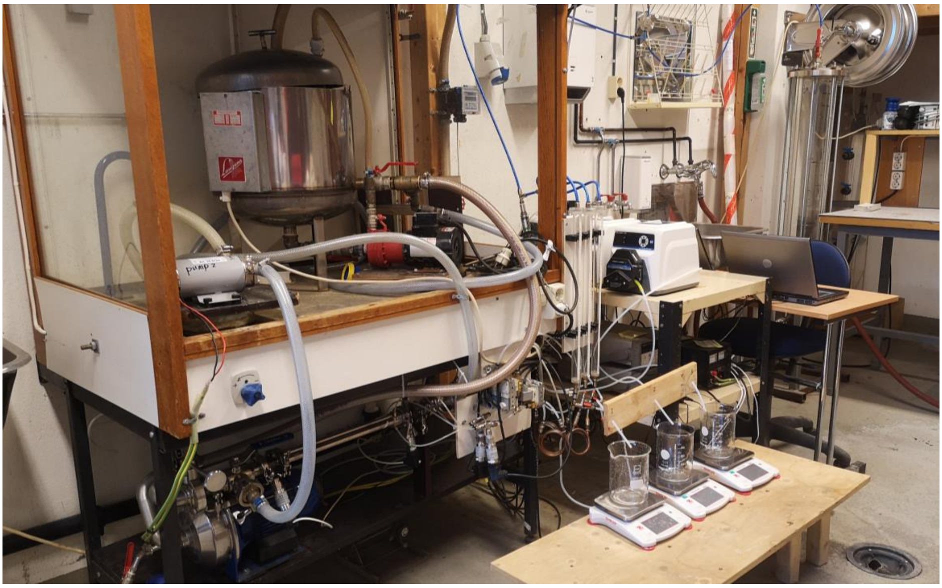


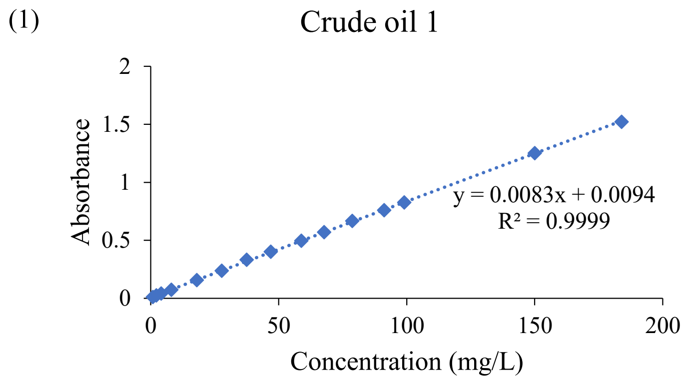
Figure 3-6. A picture of experimental setup.

3.3. Analytical methods

Three samples (at the beginning, in the middle, and at the end) were taken for feed water and permeates in the short-term experiments for Paper II and Paper III. Four samples (at 0 h, 4 h, 8 h, and 12 h) were taken for the feed and permeates in the long-term experiments for Paper IV.

3.3.1. Total oil concentration

Oil in water samples was extracted with dichloromethane (HiPerSolv CHROMANORM for HPLC, VWR) and separated by separation funnels. The total oil concentration in water samples was measured based on the UV absorbance of the organic phase at 259 nm using a UV-vis spectrophotometer (Lambda 650, PerkinElmer, USA). UV absorbance at 259 nm and oil concentration had a linear relationship. The calibration curves for crude oil 1 and crude oil 2 are shown in Figure 3-7 (1) and (2), respectively.



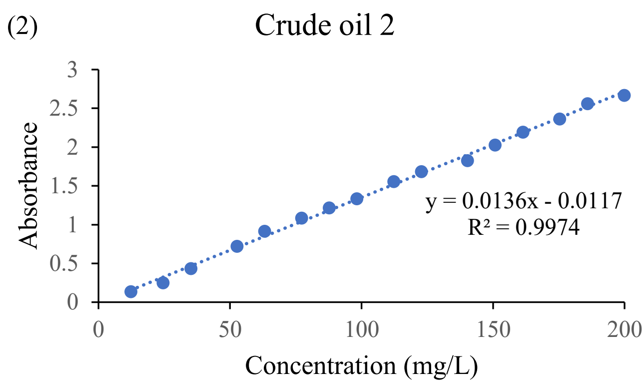


Figure 3-7. Calibration curves for (1) crude oil 1 and (2) crude oil 2, respectively.

3.3.2. Particle/droplet sizes

The particle/droplet sizes in the feed water were analyzed using two instruments: a microscope (Nikon LV 100D, Japan) and an optical particle detector (PN3000 XPT-C, Postnova Analytics GmbH, Germany). The initial samples after homogenization were analyzed using the microscope. 10 pictures were captured for each sample. Image-Pro Plus 5.0 software was used to determine particle/droplet size distributions. This method provided intuitive information of the particles and emulsions in the feed water. The optical particle detector has a LED light source, a CCD camera and an image analysis system and is coupled with a flow system. It can follow the dynamic changes of *in-situ* particle/droplet sizes during the experiment.

3.3.3. pH

The pH of the feed water was measured by a pH meter (SevenEasy pH, Mettler-Toledo, Greifensee, Switzerland).

3.4. Modeling

3.4.1. Full factorial design for Paper II

A 2^3 full factorial design was used to identify the importance of backpulsing amplitude, duration and frequency, and the interactions between them [76, 77]. The high and low levels of each parameter (see Table 3-4) were selected based on literature summary in Paper I, capacity of the setup and economic considerations. Two responses were investigated: J_{sf} (averaged specific flux of forward filtration at the final cycle, LMH/bar) and Y (net permeate yield during the whole experiment period, L).

2 blocks were used in the factorial design as experiments ran with 2 replicates. 3 center samples were added to each block to check error, curvature and reproducibility [78]. In total 22 experiments were performed. The experiments were carried out in a random order to avoid system errors [79]. Minitab 19 was used for the design of experiments and data analysis.

Table 3-4. Levels of factors in the 2^3 full factorial design.

Factors	Coded symbol	Values of coded levels	
		Low level (-1)	High level (+1)
Amplitude (bar)	A	0.1	1.0
Duration (s)	B	0.1	0.5
Frequency (Hz)	C	0.0167	0.05

3.4.2. Pore blocking modeling for Paper III

The analytical model proposed by Field et al. [80] was applied to describe the fouling mechanisms during the crossflow microfiltration of different produced waters under a constant-pressure mode. The characteristic equation is shown in Eq. ().

$$-\frac{dJ}{dt} = k(J - J^*)J^{2-n} \tag{1}$$

where the parameter n characterizes the predominant fouling law with n = 2 for complete pore blocking, n = 1.5 for internal pore blocking (also called standard pore blocking), n = 1 for partial pore blocking, and n = 0 for cake filtration. J is flux, t is time, n and k are constants specific to the type of blocking law, and J^* is the limiting flux for the specific blocking law. The physical meaning of J^* is that below the limiting flux J^*, for a constant-pressure filtration, no fouling of this type of blocking law would occur [81]. In this study, J^* was taken to be the steady-state flux [82].

To avoid the need to differentiate, integral analysis of the model is used, and Eq. (1) is then re-written as [82]:

$$\int_{J_0}^{J} \frac{-dJ}{J^{2-n}} = K \int_0^t (J - J^*)dt = K\,(v - J^*t) \qquad (2)$$

where v is the volume of permeate per unit area. K is the constant including the area term, rather than k.

The fit of the fouling models to the experimentally obtained flux data was determined by the correlation coefficient R^2. The model with the highest R^2 corresponds to the dominant fouling mechanism.

Chapter 4 Results and discussion

This chapter presents the main outcomes from the four scientific papers/manuscripts and one set of unpublished results. How the papers/results are connected to each other and how the papers/results answer research questions are discussed later. The full version of the selected papers and the unpublished data is provided in Appendix A.

4.1. Main results

4.1.1. Paper I

Backpulsing technology applied in MF and UF processes for membrane fouling mitigation: A review

The state-of-the-art of backpulsing technology applied in low-pressure membrane filtrations is reviewed.

Firstly, the fundamentals of backpulsing are overviewed. Backpulsing removes hydraulically reversible fouling. Evaluation of backpulsing efficiency depends on the improvement of membrane permeation/production compared to filtration without backpulsing. For mechanism study purpose, it is significant to look at the membrane performance before and after each backpulsing. The theory of backpulsing is investigated from two perspectives. One is to focus on the behavior of a single foulant during backpulsing that is affected by hydrodynamic forces, diffusions, and intermolecular interactions, as shown in Figure 4-1. The other is to consider the fouling system as a continuum and look at the mass transfer in the system.

Secondly, applications of backpulsing are summarized. Backpulsing has been used in all kinds of membrane configurations and for both polymeric and ceramic membranes. Research of backpulsing covers many industrial fields: water and wastewater treatment, food industry, biotechnology application and other industries. Pilot- and industrial-scale applications has been tested in the purification of radioactive wastewater, the separation of dispersed substances in thermomechanical pulp process water, and the filtration of produced water.

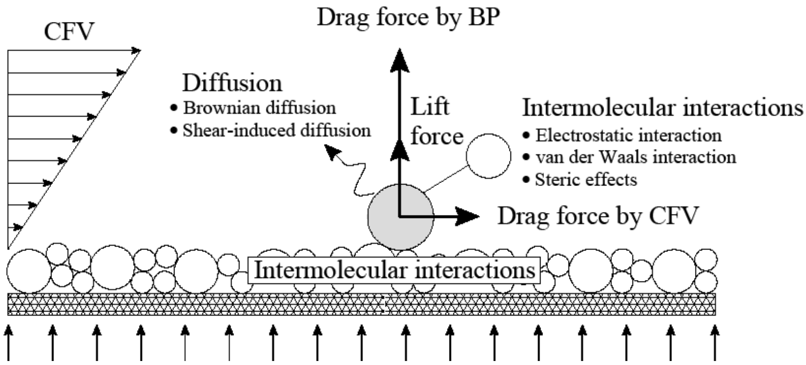


Figure 4-1. Paper I [83]: A schematic of the acting forces on a deposited foulant at the beginning of backpulsing. The foulants are not drawn in scale. Adapted from [84, 85].

Moreover, factors affecting backpulsing efficiency are discussed. As is shown in Figure 4-1, to dislodge the deposited foulant from the membrane, the drag force by backpulsing associated with inertial lift force and back diffusion needs to overcome the attractive interactions between foulant and membrane and/or between foulant and foulant. These forces are affected by feed properties, membrane properties, and operating conditions. Backpulsing is more efficient to remove external and non-adhesive fouling. The types of foulants in the feed solution directly determine the types of membrane fouling. The more concentration of foulants, the more severe membrane fouling and the less cleaning efficiency of backpulsing. Modification of membrane surfaces and membrane pore sizes are corelated to the types of membrane fouling and affects backpulsing efficiency. Optimum conditions of backpulsing (i.e., amplitude, duration, and frequency) and filtration (TMP/flux and CFV) favor the backpulsing efficiency significantly.

Furthermore, modelling is developed to predict the optimum conditions of backpulsing and explain the fouling situation during the process of backpulsing. Six analytical models have been developed based on cake layer formation assumption (see Figure 4-2). They are qualitatively precise to predict the optimum backpulsing interval/frequency. By analyzing critical parameters, such as τ_1 and β, membrane performance under different backpulsing conditions or in different membrane filtrations can be evaluated.

Finally, the existing challenges and outlook are discussed. To further improve the implementation of backpulsing in commercial applications, several aspects on backpulsing need more work: further development of modelling, combination of backpulsing and backwashing, membrane ageing and damage caused by backpulsing, as well as more long-term and large-scale experience.

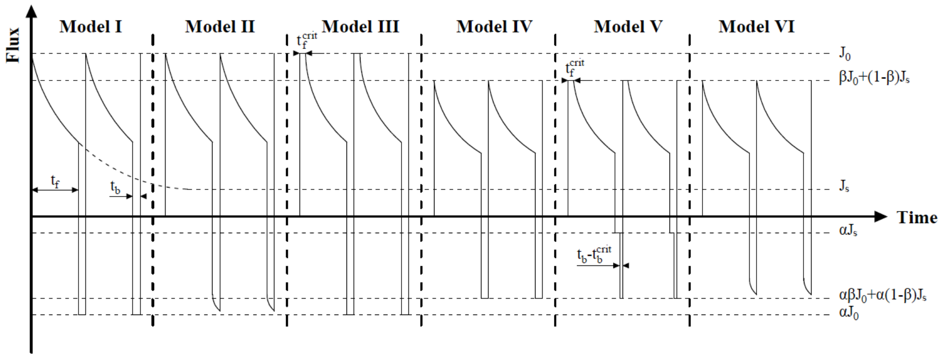


Figure 4-2. Paper I [83]: Theoretical forward and reverse fluxes for six analytical models. J_0 is clean membrane flux, J_s is steady-state flux, $\alpha = \Delta P_b/\Delta P_f$, β is cleaning efficiency, t_f is forward filtration duration, t_b is backpulsing duration, t_f^{crit} and t_b^{crit} are the time delay during forward filtration and backpulsing, respectively. Adapted from [60].

4.1.2. Paper II

A multivariate study of backpulsing for membrane fouling mitigation in produced water treatment

Backpulsing efficiency on membrane fouling mitigation in produced water treatment was investigated via various backpulsing conditions. Graphic abstract is shown in Figure 4-3. The composition of produced water in this study is shown as PW0 in Table 3-2. A 2^3 full factorial design was used to study the effect of the three backpulsing parameters (amplitude, duration, and frequency) and their interactions on membrane performance. Final specific flux and net permeate yield were chosen as responses to indicate the situation of membrane fouling and membrane productivity, respectively. Two membranes were tested in parallel as two replicates in the statistical design. Three center points were added to help check error, curvature, and reproducibility of the design [78].

Microfiltration with a nominal pore size of 0.1 μm was effective to remove oil in produced water from 199.6 mg/L to less than 5 mg/L averagely. In the filtration without backpulsing, the normalized specific fluxes dropped dramatically in the initial 1000 s and continued decreasing to 11.2% at 2400 s for both membranes. When backpulsing was applied, the membrane fouling was greatly mitigated. However, the efficiency of backpulsing varied significantly between different backpulsing strategies. After running 4800 s, the normalized specific fluxes were at the highest 72.0% and 70.1% and at the lowest 11.9% and 13.0% for the two membranes, respectively.

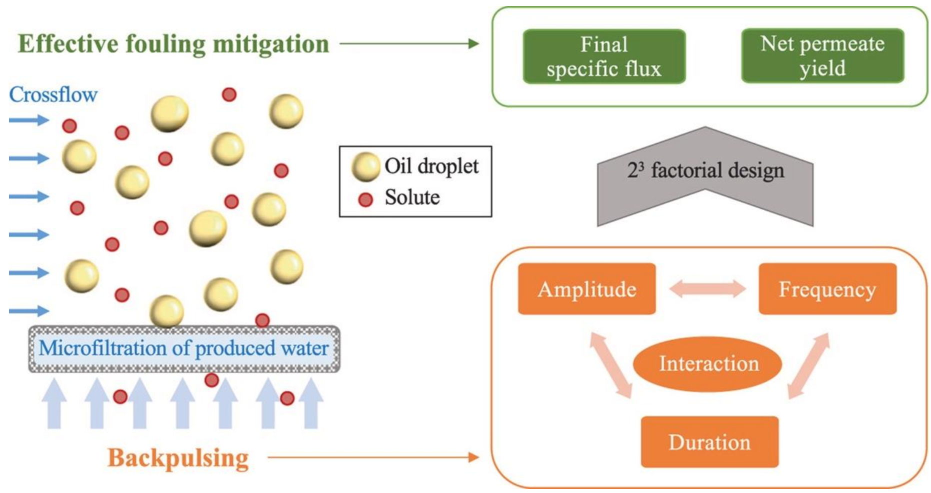


Figure 4-3. Graphic abstract of Paper II [86].

The statistical results on the significance of the main terms for final specific flux and net yield are shown in Figure 4-4 (a) and (b), respectively. Within the selected levels all the three backpulsing parameters were important for the two responses. Amplitude was the most influential factor for final specific flux as it affects the reverse force to dislodge foulants. There was a minimum threshold of amplitude for an effective backpulsing, after which the improvement of cleaning efficiency was not obvious. Frequency was the most significant one for net yield. It plays a role on the situation of fouling formation. If frequency was too low, membrane was not cleaned timely, then permeate flux was low. However, at an extreme high frequency, net permeate was also low because of the excessive loss of clean water and filtration time. Besides, membrane filtration with backpulsing is a dynamic process [13], and frequency is a very important factor for the dynamic characteristics and stability of the system. Duration was important to both final specific flux and net yield too, but not as prominent as the other two parameters. Increasing any of the three backpulsing parameters led to an increase in the final specific flux but caused more water loss and thus a lower net yield. Interactions of amplitude × duration and amplitude × duration × frequency were important for final specific flux. All two-way interactions between amplitude, duration and frequency were important for net yield. However, the two-way interactions on the two responses performed differently. Increasing duration/amplitude reduced the effect of amplitude/duration on final specific flux, while increasing any of the parameters promoted the effect of the other parameter on net yield.

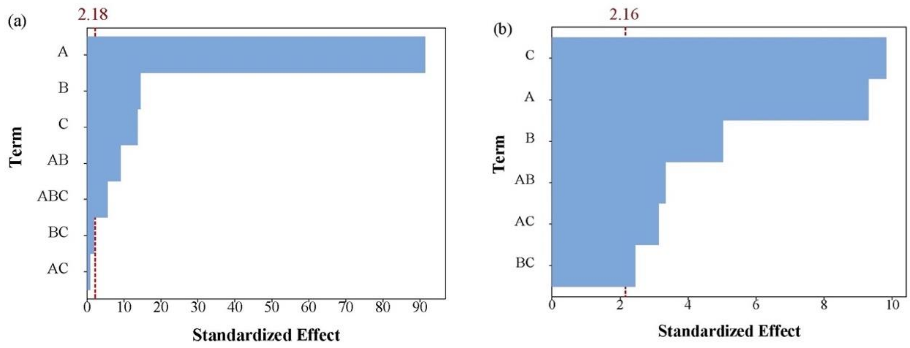


Figure 4-4. Paper II [86]: Pareto chart of the standardized effects for (a) final specific flux and (b) net yield. Coded symbol: A, amplitude; B, Duration; C, Frequency; AB, interaction between amplitude and duration, and so on.

4.1.3. Paper III

Application of ceramic membranes for produced water treatment. Part I: Effect of feed compositions on membrane fouling

Ten types of produced water were treated using three types of ceramic membranes. The effect of feed properties on membrane fouling was investigated. Graphic abstract is shown in Figure 4-5. The compositions of the ten types of produced water are shown in Table 3-2 from PW1 to PW10.

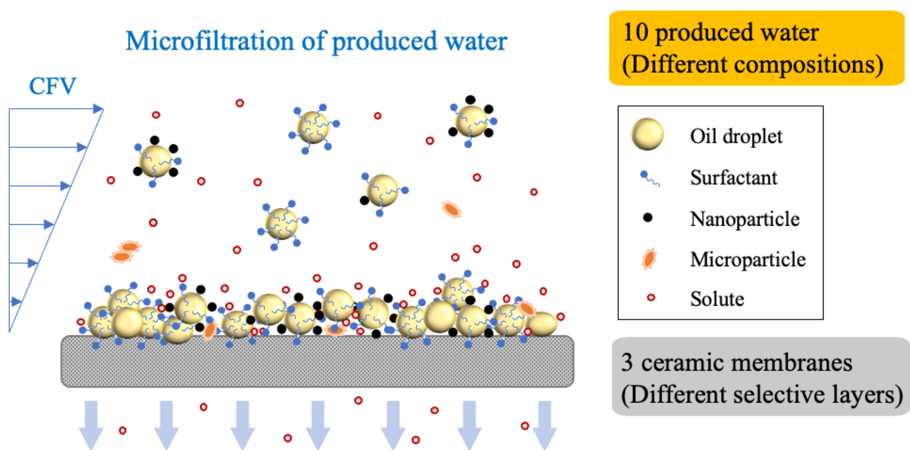


Figure 4-5. Graphic abstract of Paper III.

The three membranes had different selective layers of TiO_2, Al_2O_3, and ZrO_2, respectively. Although their nominal pore sizes were all 0.1 µm, the pore size distributions were different, see Figure 3-1. TiO_2 membrane had the widest pore size distribution, ranging from 0.009 to 60 µm. Al_2O_3 and ZrO_2 membrane had similar and narrower pore size distributions, and ZrO_2 had the narrowest between 0.01 and 1.5 µm. Results show that microfiltration was effective to remove emulsified oil despite different feed compositions. However, fouling situation was different for different membranes mainly due to different membrane pore size distributions, instead of different membrane selective layers. In general, although the clean water fluxes of TiO_2 membrane were more than three times larger than those of Al_2O_3 and ZrO_2 membrane, ZrO_2 membrane showed the best membrane performance in the three membranes in terms of the slowest flux decline in the initial filtration period, the highest steady-state flux, and the highest total yield.

Crude oil, particles, surfactant, and brine were used to synthesize produced water with a homogenizer to make emulsions. The oil concentrations in the feed were between 197.7 and 239.9 mg/L. The oil droplets had average sizes between 6.56 and 9.68 µm by volume. Two out of the ten types of produced water were synthesized based on a treated real produced water from the NCS. The effect of different types of crude oils, particles, brines, and real produced water on membrane fouling was compared and studied. Particles and crude oils had a significant effect on the membrane fouling formation, while brines with different pH and valency did not affect the fouling tendency much. Adding 50 mg/L nanoparticles to the synthetic produced water greatly mitigated membrane fouling. The dominant membrane fouling for all the three membranes changed from different pore blockings in the filtration of PW2 and PW3 to cake formation in the filtration of PW7 and PW8 due to the aggregation of nanoparticle and oil droplets. Produced waters synthesized by different crude oils led to different fouling tendencies. In the filtration of PW3 that contained a heavier crude oil than PW2, the permeate flux declined more dramatically in the initial filtration period, but the steady-state fluxes were higher for all the three membranes. Compared with the filtration of pure oil-in-water emulsions (PW2), adding 50 mg/L microparticles in PW6 slowed down the fouling rate slightly in the beginning 1200 – 1500 s filtration, however, led to a continuous fouling afterwards. The steady-state fluxes of Al_2O_3 and ZrO_2 membrane in the filtration of PW6 were the lowest in the ten produced waters. The effect of brine pH on membrane fluxes was different for different membranes. However, the flux decline curves and flux changing time were similar, as the crude oil in PW1, PW4 and PW5 was the same. Membrane showed the worst performance in the filtration of alkaline produced water (PW4) compared with neutral (PW1) and acidic (PW5) conditions in terms of oil removal, steady-state flux, and total yield. The effect of brine valency on membrane fluxes was negligible for all the three membranes, although involving divalent ion (Ca^{2+}) increased the averaged size of oil droplets. At the end, crossflow microfiltration was use for the tertiary treatment of real produced water with oil concentrations in the feed increased to a comparable level in this study by adding crude oil 1 (PW9) and crude oil 10 (PW10), respectively. In the filtration of PW9 and PW10 with more complicated compositions, the oil removal by the three membranes decreased slightly, however, the permeate yield and steady-state fluxes increased in general.

4.1.4. Appendix B. Additional results

Results on optimal backpulsing frequency

Three types of ceramic membranes, the same as used for Paper III, were used for the set of experiments to find out the optimal backpulsing frequency in the filtration of PW1 whose composition is shown in Table 3-2. Although the optimal frequencies were slightly different for different membranes, the optimal backpulsing frequencies for the three membranes was generally in the range between 10 s to 15 s. Backpulsing interval of 12 s was selected as an optimum backpulsing frequency and used in the following experiments in Paper IV.

4.1.5. Paper IV

Application of ceramic membranes for produced water treatment. Part II: Comparison of backpulsing and backwashing for membrane fouling mitigation

PW1, PW3, PW6, and PW9 (see Table 3-2) were carried out for the 12 h filtration experiments with backpulsing and backwashing using three types of ceramic membranes. The graphic abstract is shown in Figure 4-6. PW1 and PW3 were pure oil-in-water emulsions and were reproducible for different filtrations. Oil droplets coalesced, and large droplets floated to the membrane surface over time. PW6 and PW9 contained both oil and particles and were less stable than PW1 and PW3. PW6 contained both oil droplets and kaolin particles. Oil droplets and particles coalesced more in the filtration with backwashing than in the filtration with backpulsing, which greatly reduced the fouling tendency in the later period of filtration with backwashing. PW9 was made based on treated real produced water from the NCS whose composition changed greatly over time. As the different filtration experiments were carried out at different days, there was a big difference in the initial feed properties of PW9 in different filtrations. The initial fouling tendency of PW9 was filtration with backwashing < filtration with backpulsing < normal filtration.

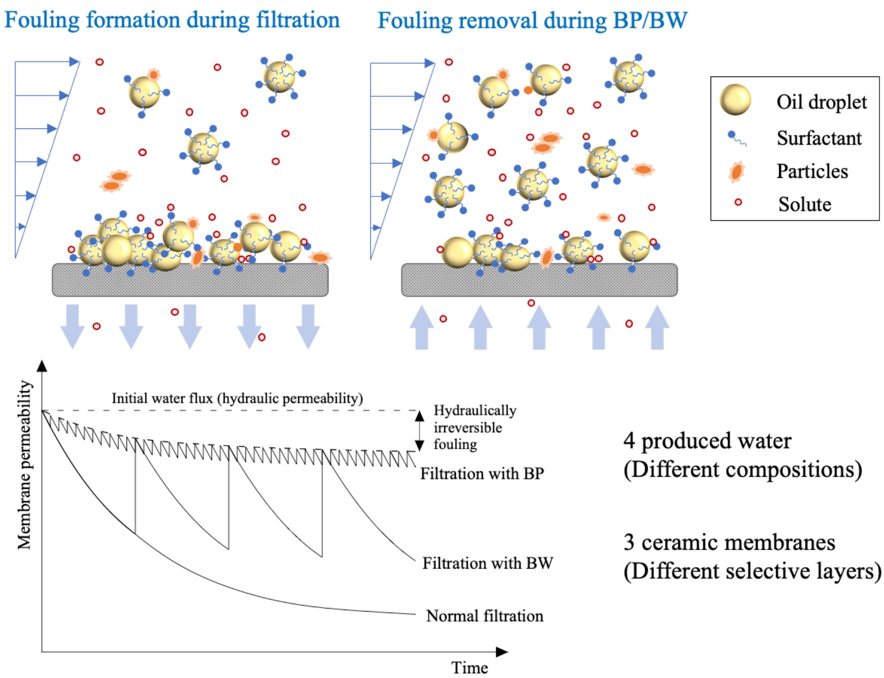


Figure 4-6. Graphic abstract for Paper IV.

The reverse flow cleaning could effectively mitigate membrane fouling by increasing membrane fluxes and filtration time without decreasing the permeate quality. The efficiency of backpulsing or backwashing on membrane fouling mitigation was affected by feed water properties and membrane properties. The performance stability of membranes in different produced water were in accordance with the pore size distributions of the membranes. ZrO_2 membrane has the narrowest pore size distribution, the behavior of which is the most stable within different feed solutions.

Backpulsing was more effective than backwashing in general even with some difference in the feed water that was beneficial to the filtration in backwashing experiments. Although backpulsing consumed more clean water than backwashing, the net yields of backpulsing experiments were higher than those of backwashing experiments, and the 12 h end fluxes in backpulsing experiments were higher than those in backwashing experiments. The advantage of backpulsing over backwashing is more obvious in applications where more permeate per membrane area is desired such as in offshore applications. Optimization of the setting of backpulsing/backwashing parameters was case specific and affected by the feed and membrane properties. A combination of backpulsing and backwashing could be a good solution to applications where higher flux recovery requires longer duration.

4.2. Discussion

Discussion on the work is divided into four sections, see Figure 4-7. As has been reviewed Paper I, factors of feed properties, membrane properties, and operating parameters have influence on backpulsing efficiency, the discussion on this thesis will be carried out from these aspects including backpulsing conditions, feed properties, membrane properties and the comparison of backpulsing and backwashing. The discussion on the four parts also answers the research questions raised in section 2.5.

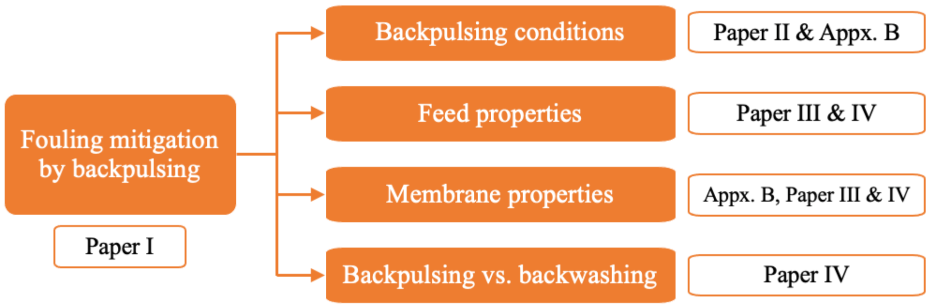


Figure 4-7. Structure of the discussion on this thesis work.

4.2.1. Backpulsing conditions (Q1)

In membrane filtration, the fouling situation is strongly affected by the filtration conditions (e.g., TMP under constant pressure mode/flux under constant flux mode, CFV, and temperature), while the fouling removal by backpulsing is closely related to the setting of backpulsing parameters, i.e., amplitude, duration, and frequency.

The statistical results in Paper II show that for the responses of both final specific flux and net yield, all the three backpulsing parameters and some of their interactions were important. This indicates that to find the optimal backpulsing conditions statistical methods including the effect of the interactions between parameters would be more accurate than the one-variable-at-a-time method. Individual effect of each parameter was also investigated. There was a threshold of amplitude to dislodge foulants from the membrane, and in this study it was between 0.1 and 0.55 bar, probably close to 0.55 bar. Similar results were also obtained by Mores et al. [87] in the microfiltration of washed bacterial cells where maximum fraction of the membrane cleaned by backpulsing increased with increasing amplitude up to 5 psi (0.34 bar), above which the cleaning efficiency leveled off. In the microfiltration of light non-aqueous phase liquids, McAlexander and Johnson [54] found the minimum amplitude was 0.6-0.85 bar for effective membrane cleaning. Frequency was the most significant parameter for net yield, which has also been indicated by Edmundo et al. [68] who studied the effects of several process variables on the microsieve performance during the filtration of whole milk. Compared with inlet-outlet pressure gradient and backpulsing amplitude, TMP and backpulsing frequency influenced the permeate flux the most.

The effect of frequency on the net yield was further investigated. Results are shown in Appendix B. The selected backpulsing amplitude was taken 0.5 bar which should be close to the amplitude threshold. The selected backpulsing duration was taken 0.6 s which was in the middle range of 0.1 s and 1 s. Frequency varied from 0.0048 Hz (every 210 s a backpulse) to 0.11 Hz (every 9 s a backpulse). The optimal frequency was slightly different for different membranes, but within the range of 11 s to 15 s. In the later backpulsing experiments, amplitude was kept at 0.5 bar, duration was kept at 0.6 s, and frequency was using 0.083 Hz (every 12 s a backpulse).

4.2.2. Feed properties (Q2)

More aggregation of between oil droplets, particles, or both led to larger droplet/particle sizes in the feed water, which resulted in less membrane fouling and favored the membrane permeate flux. This is because the size difference between droplets/particles in the feed water and membrane pores increased, so that membrane fouling tended to shift from pore blocking to cake formation. In Paper III for all the three membranes, the permeate flux curves in the filtration of PW7 and PW8 stood out the flux curves in the filtration of other produced water, which is due to the aggregation of oil droplets and nanoparticles with the assistance of surfactant, so that the dominant fouling mechanisms were all cake formation from the beginning of filtrations. The fluxes in the initial 30 min filtration of PW9 were much higher compared to the filtration of PW2, PW3, and PW9. This also owes to the aggregation of a different crude oil with particles in the treated real produced water. This aggregation in PW9 was more obvious in the experiments in Paper IV. The composition of real produced water changed over time. The experiments of normal filtration, filtration with backpulsing, and filtration of backwashing were carried out at different days. For PW9, the normal filtration experiment was carried out earlier than filtration with backpulsing, followed by filtration with backwashing. The mean droplet/particle size of PW9 in different filtrations were filtration with backwashing > filtration with backpulsing > normal filtration. The initial fouling situation was then filtration with backwashing < filtration with backpulsing < normal filtration.

Crude oil had a dominant effect on the physio-chemical properties of oil droplets/emulsions. In Paper IV, although different particles were involved in PW6 and PW9, the shapes of the droplet/particle size distribution were similar for PW1, PW3 and PW6 by having one major peak at around 5 – 6 μm, while PW3 had two major peaks. The reason is that PW3 was made of a different type of crude oil and produced water properties were greatly affected by the crude oil [39, 73].

Salinities/ionic strengths of the produced water studied in this work was high, which screened the surface charge of the membranes, as the ion density was high and electrical double layer was compressed. In Paper III, the effect of brines including the effect of divalent ion and the effect of pH were studied. The permeate fluxes of the three membranes were just slightly affected by these factors.

4.2.3. Membrane properties (Q3)

Three types of ceramic membranes, TiO_2, Al_2O_3, and ZrO_2 membrane, were used in Appendix B, Paper III and Paper IV. ZrO_2 membrane had the most stable and in general best flux behavior in different kinds of filtrations. The performance of Al_2O_3 membrane was a bit less stable than ZrO_2 membrane, but still rather similar. The flux of TiO_2 membrane showed big variations in different filtrations. The stability of flux behavior for the three membranes was consistent with their pore size distributions. The narrower the membrane pore size, the more stable and better the membrane flux. In Appendix B, the standard deviation of net yields in different backpulsing intervals of 9 s to 210 s was 3.2 kg for TiO_2 membrane, 1.0 kg for Al_2O_3 membrane, and 0.9 kg for ZrO_2 membrane. In Paper III the filtrations without cleaning, steady-state fluxes were achieved after 30 min filtration for most types of produced water, except PW7 and PW8 where no steady-state fluxes were achieved during 2 h experiments. The averaged steady-state fluxes of the 8 types of produced water were 66, 84 and 93 for TiO_2, Al_2O_3 and ZrO_2 membrane, respectively. In Paper IV, with the same setting of backpulsing and backwashing parameters, the permeate flux of TiO_2 membrane varied a lot in the filtration of PW1, PW3, PW6 and PW9. While for ZrO_2 membrane, the permeate fluxes during backpulsing experiment were almost the same in the filtration of PW1, PW3 and PW6, and the permeate fluxes of the four types of produced water during backwashing experiments were quite similar. Al_2O_3 membrane again had similar performance as ZrO_2 membrane.

The surface properties due to different membrane selective layers were not so important to the membrane fouling in this work. In Paper III, the permeate fluxes of the three membranes were not affected much when pH of the produced water changed from 4 to 9. Besides, as irreversible fouling usually occurs in the filtration of oily wastewater [88], the membrane surface properties tend to be changed due to the irreversible membrane fouling [89].

4.2.4. Backpulsing vs. backwashing (Q4)

As is reviewed in Paper I, backpulsing may seem like backwashing with a very short duration, however, they are fundamentally different, both with respect to operational conditions, and with respect to the mechanisms of fouling formation and mitigation. Backpulsing has a dynamic and transient effect to remove membrane fouling that is not found in backwashing [13]. It is thought that the deposition of foulants is removed from the membrane before it is fully formed and compacted when backpulsing is applied [53].

In Paper IV, despite some difference in the PW6 and PW9 that favored the filtration in backwashing experiments, the net yields during the 12 h filtrations were backpulsing > backwashing in each specific case. This is because backpulsing happened so frequently that the membrane fouling was not fully compact and the permeate fluxes were kept at relatively high levels, instead of reaching and remaining at steady-state fluxes. However, when looking into details, the flux recovery by backpulsing was not always higher than that by backwashing. The level of flux recovery by backpulsing/backwashing was closely related to membrane fouling and was case specific. In 7 out of the 12 applications (4 types of produced water * 3 types of membranes), the flux recovery by backpulsing was completely higher than that by backwashing, as the curve of backpulsing max flux was totally above that of backwashing max flux. This is probably due to the transient effect of backpulsing that knocked off the foulants from the membranes. Besides, the fouling in these backpulsing experiments was probably easier to remove, as it could not firmly form in such short filtration periods. In 3 applications, the flux recovery of backpulsing was higher than that of backwashing in the first one-third to half period of filtration time, afterwards the flux recovery of backwashing was higher. This indicates when membrane fouling became more severe after some time filtration, backpulsing with a shorter cleaning duration was not so effective to dislodge foulants as backwashing, although the cleaning frequency of backpulsing was higher. Similar results were also observed in our previous experiments in Paper II that the significance of cleaning duration increased when membranes fouled more severely [86]. The permeate flux curve of the filtration with backpulsing using a longer duration and a lower frequency crossed that of the filtration with backpulsing using a shorter duration but higher frequency and became higher in the later period of filtration. In the filtration of PW3 using ZrO_2 membrane and the filtration of PW6 using TiO_2 membrane, backwashing could recover more flux than backpulsing from the beginning. This is probably because in these two cases backpulsing with a duration of 0.6 s could not efficiently knock off the foulants from the membranes. Fraga et al. supposed that backpulsing was mainly to loosen and detach the trapped foulants via inertia excitation, while backwashing was to provide constant shear and drag force and wash foulants away from the membrane surface and membrane pores [90]. The two technologies function differently and are suitable for different cases. A combination of the two technologies could bring a synergistic effect on fouling mitigation, as has been tested in literature before [72, 91].

Chapter 5 Conclusions and recommendations

5.1. Conclusions

This doctoral work consists of two parts of work: literature review on backpulsing technology and lab-scale experiments on produced water treatment with backpulsing for fouling mitigation.

- Paper I critically reviews the development of backpulsing technique in microfiltration (MF) and ultrafiltration (UF) processes. Backpulsing is a promising physical cleaning method, which can effectively mitigate external and non-adhesive fouling. It has been applied in many industrial fields. Some were in pilot- and commercial-scale operations. The backpulsing efficiency towards fouling mitigation is affected by feed properties, membrane properties and operating parameters. Analytical models, semi-analytical models and simulation models have been developed to simulate backpulsing process in different applications, which can help predict membrane productivity and evaluate the performance of backpulsing in fouling mitigation.
- Membrane filtration had very stable permeate quality. Microfiltration with nominal pore size of 0.1 μm was efficient to remove oil concentration from around 200 mg/L to less than 10 mg/L in all the experiments. These experiments included feed water with different compositions (different crude oil, different particles, and different brines) and under different filtration conditions (normal filtration without cleaning, filtration with backpulsing, and filtration with backwashing). (Paper II - IV)
- All the three backpulsing parameters were important for the development of membrane fouling and net permeate yield. Increasing backpulsing amplitude, duration or frequency decreased the membrane fouling formation, but caused more clean water loss. Amplitude was the most critical variable for fouling removal and final specific flux, while frequency was the most significant one for membrane net yield. The interactions between parameters were also important, meaning the setting of one parameter affected the effect of another parameter on the fouling situation/net yield. (Paper II)
- Membrane fouling was strongly affected by the compositions of produced water. Crude oil determined the physio-chemical interactions between oil droplets and membranes. However, when the oil droplets were stabilized by nanoparticles and

surfactants, the fouling mechanisms were changed from pore blocking to cake formation, which resulted in distinct improvement of permeate flux. (Paper III)

- Membrane surface charge was less important for the membrane fouling in this study, as the salinity in the feed water was very high. Although the membranes had the same nominal pore size, the membrane pore size distributions were different. ZrO_2 membrane had the most similar flux behaviors in the filtration of four different types of produced water in both backpulsing and backwashing experiments, which is because ZrO_2 membrane had the narrowest pore size distribution of the three membranes. (Paper IV)
- Backpulsing showed higher net yields than backwashing in all the filtrations during the 12 h experiments. In most cases, the average permeate flux of backpulsing was completely above the corresponding average flux of backwashing. However, the flux recovery was not always backpulsing better than backwashing. Flux recovery after each reverse cleaning was affected by the fouling situation during the forward filtration period in each specific case. (Paper IV)

5.2. Recommendations for future work

Some topics have been identified for future work in this section since they could not be carried out within this scope of study or were only partially involved.

- **Verification and improvement of existing models**. The flux data for Appendix B, Paper III, and Paper IV in this study were all recorded at the frequency of 1 Hz and in relatively high resolution. This makes it possible to verify the basic equations and assumptions of existing fouling models and backpulsing models.

  Fouling models: The models to study fouling mechanisms have been developed relatively well under constant-pressure mode [80, 82, 92] and lately also under constant-flux mode [93]. However, most models simplify the fouling of the whole filtration period with one dominant fouling type. As is known that fouling phenomena are complicated, there must be different fouling types happen simultaneously or consecutively. The combined models [94, 95] can still be further developed. High-resolution experimental data make it possible to improve the existing models, especially combine models.

  Backpulsing models: The six analytical backpulsing models and three simulation/semi-analytical models proposed by Vinther et al. [96-99] have not been verified by high-resolution flux data yet. Besides, backpulsing models are less developed than the fouling models. All the existing models as reviewed in Paper I are under constant-pressure mode. Further development of models suitable for constant-flux mode are desired, especially for practical applications.

- **Long term and large scale running**. The experience of backpulsing available from literature nowadays is mainly short-term and lab-scale experience. However, backpulsing is operating in a much higher frequency than backwashing, which increases the failure risk of an operating system, such as failure of valves. Besides, surface properties of membranes can be changed during long-term filtration because of the irreversible membrane fouling [89]. Therefore, although it is shown both experimentally and theoretically that backpulsing is an efficient way to mitigate membrane fouling, long-term and large-scale experience from practical applications is necessary for a wider application of backpulsing technology.
- **Membrane ageing and damage caused by backpulsing.** The replacement of membranes accounts for 25-40% of the total cost in a membrane plant [100]. Membranes' lifetime depends on the degree of membrane ageing and damage. Pressure differentials during backpulsing induce a degradation of membrane material,

especially for polymeric membranes. However, literature relating to the fatigue behavior under mechanical stresses is very limited [101].

- **Combination of backpulsing and backwashing.** Fraga et al [90] supposed that backpulsing was mainly to loosen and detach the trapped foulants via inertia excitation while backwashing was to provide constant shear and drag force and wash foulants away from the membrane surface and membrane pores. Backpulsing and backwashing can be combined to obtain a synergistic effect on membrane cleaning [90, 91]. In addition, when the membrane surface is frequently exposed to backpulsing, more internal fouling is resulted [102, 103]. This is because backpulsing removed the cake layer or gel layer on the membrane surface which often acts as a secondary dynamic membrane [104, 105]. The combination of backpulsing and chemically enhanced backwashing is therefore envisioned to be a promising solution.

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Appendix A. Selected papers

Paper I

Backpulsing technology applied in MF and UF processes for membrane fouling mitigation: A review

Yinghong Gao, Jie Qin, Zhiwei Wang, Stein W. Østerhus.

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Backpulsing technology applied in MF and UF processes for membrane fouling mitigation: A review

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ABSTRACT

Membrane cleaning is crucial to maintain the permeability and selectivity of membranes. Physical cleaning can mitigate membrane fouling, lower the frequency of chemical cleaning, thus prolong membrane lifetime, and reduce operational costs. Backpulsing is a promising physical cleaning method, which can effectively mitigate external and non-adhesive fouling and has been used in many industrial fields. However, a comprehensive understanding of backpulsing and the optimization of this technology is still lacking. This paper critically reviews the development of backpulsing technique in microfiltration (MF) and ultrafiltration (UF) processes. Firstly, the fundamentals of backpulsing are addressed. Secondly, applications of backpulsing are summarized according to the applied fields. Results of pilot- and commercial-scale operations are presented. Moreover, factors influencing backpulsing efficiency are illustrated, including feed properties, membrane properties and operating parameters. Furthermore, mathematical models of backpulsing are overviewed. The models not only predict membrane productivity, but also provide a perspective to evaluate the performance of backpulsing in fouling mitigation. Finally, the existing challenges and future outlook are discussed.

1. Introduction

Commercial scale microfiltration (MF) and ultrafiltration (UF) membranes are available since 1960s [1,2]. Nowadays membrane filtrations have been used in a wide range of industrial applications with the purpose of removal of impurities, concentration of valuables or permeation purification of valuables [3]. However, membrane fouling is consistently the obstacle that restricts process efficiency by reducing membrane permeability, deteriorating membrane selectivity and increasing operating and maintenance costs [4,5]. Various techniques have been tested and applied to remove membrane fouling. Physical cleaning, although is not efficient in removing irreversible fouling, has several advantages compared with chemical cleaning. Physical cleaning is cost-efficient because it does not require the filtration plant to be shut down for longer periods, and it is environmentally friendly since there is no chemical waste generated. Besides, effective physical cleaning is essential to mitigate membrane fouling, in order to maintain high productivity of the membranes, reduce the frequency of chemical cleaning and prolong membrane lifetime [6]. The common physical cleaning methods are crossflushing, backwashing, vibration, air sparging and sponge ball cleaning, etc [4,7].

In MF and UF systems, especially in a dead-end filtration mode, the most adopted method for membrane fouling control is reverse flow cleaning, in general backwashing/backflushing [8,9]. After certain time of filtration, a flow of clean water is pumped back through the membrane from the permeate side, thereby lifting foulants from the membrane surface and reducing concentration polarization near the membrane surface.

Abbreviations: AA, acrylic acid; AN, anopore; BSA, bovine serum albumin; CA, cellulose acetate; CFD, computational fluid dynamics; CFV, crossflow velocity; CML, carboxylate modified latex; DCP, dynamic crossflow pulse; DMAEMA, dimethyl aminoethyl methacrylate; *E.coli*, *Escherichia coli*; GGM, galactoglucomannan; LMH, liter per square meter per hour; MF, Microfiltration; MLSS, mixed liquor suspended solids; NVP, N-vinyl-2-pyrrolidinone; PA, polyamide; PE, polyethylene; PEG, polyethylene glycol; PES, polyethersulfone; PP, polypropylene; PS, polysulfone; PVC, polyvinyl chloride; PVDF, polyvinylidene fluoride; SiC, silicon carbide; TMP, transmembrane pressure; TSS, total suspended solids; UF, ultrafiltration

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Backpulsing is also a kind of reverse flow cleaning. It is induced by periodically reversing the transmembrane pressure (TMP) for a very short duration (typically less than 1 s [10–20]) and used in conjunction with surface tangential flow [21]. The deposited foulants on the membrane surface or in the membrane pores are dislodged by backpulses and swept away by the tangential flow, such as crossflow in crossflow filtrations [10] and air sparging in immersed membrane systems [22].

However, backpulsing is different from backwashing/backflushing. The fundamental difference is the utilized speed and force [23]. Short duration and high frequency are the most obvious characteristics of backpulsing. Backpulsing usually performs in a fraction of a second every few seconds [15], while backwashing normally lasts for a few seconds or minutes every several minutes or longer [12]. Unlike backwashing, backpulsing introduce no interruption to the process of membrane filtration. The feeding in crossflow filtrations need to be stopped before using backwashing, while it is not interrupted when using backpulsing. Besides, the fouling/fouling mitigation mechanisms associated with backpulsing is different from backwashing. Backpulsing causes a dynamic and transient effect that is not found in conventional backwashing [11]. An illustration of the decline of membrane permeability in the filtration with and without backpulsing or backwashing is shown in Fig. 1. It is thought that the deposition of foulants is removed from the membrane before it is fully formed and compacted when backpulsing is applied [12]. Cakl et al. [24] found that in the crossflow microfiltration of an oil-in-water emulsion, the efficiency of the reverse flow cleaning was higher with a shorter cleaning duration. Backpulsing with a duration of 0.2 s showed the highest net flux. Backwashing with a duration longer than 5 s could hardly improve the net flux. When the forward filtration time was higher than 100 s, neither backpulsing nor backwashing was efficient to obtain a net flux higher than the steady-state flux.

The cumulative number of publications about backwashing or backpulsing used in MF/UF processes is shown in Fig. 2. Since 1990, there has been numerous publications involving backwashing, 1056 publications until 2018. Two review papers on backwashing were published in 2016 and 2017, respectively. One reviews backwashing as a fouling control strategy used for membrane bioreactors in wastewater treatment [27]. The other focuses on backwashing for low-pressure membranes in drinking water treatment [28]. However, reviews on backpulsing has not yet been published. On the other hand, there is an increasing number of publications on backpulsing, 110 publications until 2018. Various topics, including backpulsing performance in different applications, optimization of operating parameters, as well as modelling for flux prediction and mechanism study, have been covered by these publications. Therefore, a review of backpulsing is needed. This review summarizes backpulsing technology applied in MF and UF membrane processes from the following aspects: 1) Fundamentals of backpulsing, including overview and theory of backpulsing. 2) Applications of backpulsing, including lab-scale experiments and applications in pilot- and industrial-scale. 3) Factors affecting backpulsing efficiency, illustrated from three categories: feed properties, membrane properties and operating parameters. 4) Mathematical modelling to predict membrane filtration and investigate backpulsing performance in fouling mitigation. 5) Existing challenges and future research efforts. This review will provide a comprehensive understanding of backpulsing, help to find out its optimum operating conditions, and promote a wider implementation of backpulsing in industrial applications.

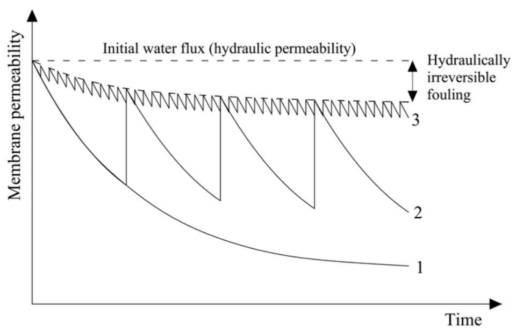

Fig. 1. An illustration of the effect of backpulsing and backwashing on the decline of membrane permeability: 1) filtration without backpulsing or backwashing; 2) filtration with backwashing; 3) filtration with backpulsing. It is assumed that both backpulsing and backwashing are capable to remove hydraulically reversible fouling completely and both cleaning methods run under reasonable conditions. Adapted from Refs. [25,26].

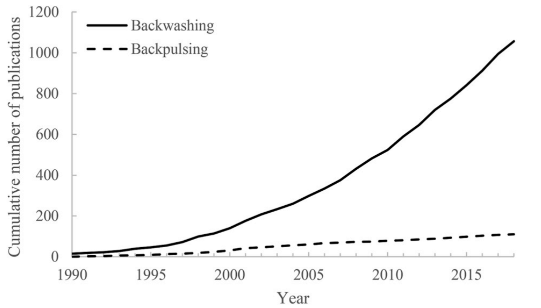

Fig. 2. Publications with themes involving backwashing and backpulsing in MF/UF processes from 1990 to 2018. Data were taken from http://apps.webofknowledge.com in January 2019, satisfying the search criteria "MF/UF and backwashing or backpulsing" in the theme.

2. Fundamentals of backpulsing

2.1. Overview of backpulsing

Backpulsing is also named backshocking [29–32], high-frequency retrofiltration [33–36], or transmembrane pressure pulsing [37–42] in literature. There are three basic parameters associated with backpulsing: amplitude, duration and frequency. Amplitude is defined as the absolute value of the negative TMP during each backpulse. Duration is the time each pulse lasts. Frequency is defined as the inverse of the sum of backpulsing duration and forward filtration time [10,11]. It indicates the interval of two consecutive pulses. Besides, backpulsing volume is also of interest, which means the amount of clean water consumed for each backpulse.

2.1.1. Backpulsing performance on membrane cleaning

An illustration of backpulsing performance on membrane cleaning is shown in Fig. 3. Backpulsing is effective in removing hydraulically reversible fouling and reducing concentration polarization (Fig. 3). Hydraulically reversible fouling [43] includes external fouling and non-adhesive fouling. External and internal fouling is classified according to the mechanisms of fouling formation [7,44]. Cake formation is normally external fouling, while fouling caused by pore constriction or pore blocking is usually internal fouling. Non-adhesive and adhesive fouling is defined based on the types of fouling substances [45]. Particulate and inorganic fouling is generally non-adhesive fouling, whereas biofouling, colloidal and organic fouling is typically adhesive fouling. Concentration polarization affects membrane performance significantly in reverse osmosis (RO) or nanofiltration (NF) where scaling is typically a big problem; while in UF that is discussed within the range of this review, colloidal solutes or macromolecules (e.g., proteins and polysaccharides) are the main contributors to it [46].

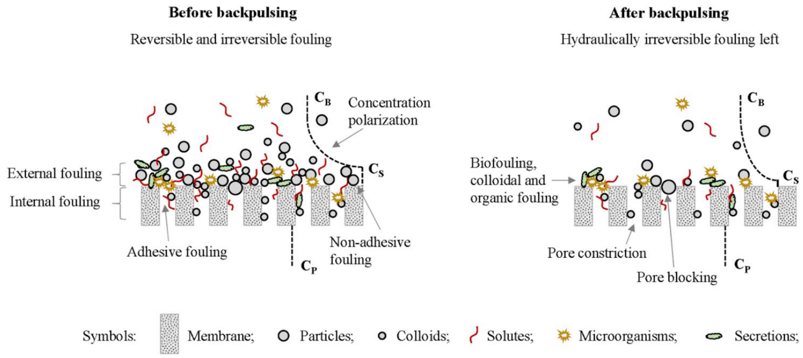

Fig. 3. Overview of membrane before and after backpulsing in MF and UF processes. Where C_B, C_S and C_P are the solute concentration in the bulk, near the membrane surface and in the permeate, respectively.

Because of the large size of these solutes, their diffusion rate from the membrane surface to the bulk is low. Thus, the solute concentration at the membrane surface C_S could reach 20–50 times of the solute concentration in the bulk C_B [47]. This leads to a resistance for solvent flux to go through the membrane due to an osmotic pressure offsetting TMP [48]. In the membrane filtrations using backpulsing, concentration polarization can be decreased by the release of positive TMP and the reverse flow from the permeate side [39,41,42].

2.1.2. Evaluation of backpulsing efficiency

Backpulsing will significantly lower the net permeate flux if it is applied for a large fraction of the filtration cycle. Stronger amplitude, longer duration or higher frequency of backpulsing causes more permeate loss. While on the other hand, backpulsing with too weak amplitude, too short duration, or too low frequency is not effective to remove membrane fouling. The optimization of backpulsing conditions not only results in higher permeate flux, but also reduces the operating cost required to achieve the desired production rate. The optimal operating conditions of backpulsing should result from practical experiments because the optimization is system dependent [49].

To evaluate the efficiency of backpulsing to a membrane system (with backpulsing vs. Without backpulsing) or to compare membrane performance by using different backpulsing strategies, there are two aspects to consider. One is membrane permeation, e.g., flux, TMP, resistance and permeability. The other is production quantity, e.g., net permeate flux/volume at certain filtration time [20] and filtration time to produce certain permeate volume [50]. For mechanism study, it is also important to evaluate the efficiency of each backpulse (comparison of membrane performance before and after each backpulse). These evaluation parameters are similar to the ones for backwashing, which have already been summarized by Chang et al. in their review about backwashing in 2017 [28].

2.1.3. Types of backpulsing

There are two ways to induce backpulsing: water backpulsing and gas backpulsing [51]. Water backpulsing is carried out by forcing clean water back through the membrane. While for gas backpulsing, compressed gas is introduced directly to the permeate side and forces out permeate trapped in the filtration chamber and membrane pores. Depending on the duration of each backpulse, permeate retained in the filtration chamber might be cleared entirely, but difficult to be removed from membrane pores due to membrane capillary forces [16]. Although air backpulsing could avoid excess water loss during backpulsing, it can lead to embrittlement and membrane integrity problems [28]. Therefore, very few studies focused on gas backpulsing. It is also shown that water backpulsing is more effective than gas backpulsing to recover membrane permeability. Ma et al. [16] compared the effect of water backpulsing and gas backpulsing on the net permeate flux in the crossflow MF of 0.1 g/L carboxylate modified latex (CML) particles with polypropylene (PP) membranes of 0.3 μm nominal pore diameter. The water backpulsing experiments were performed at a reverse TMP of 6.9 kPa (1.0 psi) for 0.15 s after every 4 s of forward filtration. For gas backpulsing, nitrogen was used and the operating parameters are almost the same as water backpulsing except for the pulse duration of 0.2 s. The results showed that for the long-term fluxes at the end of 1 h filtration, the enhancement over filtration rate without backpulsing was 3.7- and 3.2-fold for water backpulsing and gas backpulsing, respectively. Similar results about backwashing were also demonstrated by Matsumoto et al. [52] in the crossflow MF of yeast suspensions. Backwashing with permeate, supplied either by compressed gas or by a suction pump, gave a higher permeate flux than filtration with gas backwashing.

2.1.4. Backpulsing setup

For gas backpulsing, backpulses are generated by directly inducing pressurized gas into the permeate side through the control of a series of solenoid valves. For water backpulsing, the commonly used technique is either based on a series of solenoid valves or a piston device that moves back and forth and causes reverse flow through the membrane [29]. Fig. 4 demonstrates and compares different types of backpulsing setup.

Backpulsing controlled by solenoid valves is operated by switching the valves in the production line and the backpulsing line according to a certain program controlled by a computer or a timer. The difference of water backpulsing and gas backpulsing controlled by valves is whether there is a clean water tank before membrane module or not, as is shown in Fig. 4(a) and (b). During forward filtration, the valve in the permeate line is open and the one in the backpulsing line is closed. While during backpulsing period, the valve in the permeate line is switched off and the valve in the backpulsing line is switched on. Backpulses generated by controlling valves can have a wide range of operating parameters. The minimum time of backpulsing duration depends on the reaction time of valves. In the setup of Heran et al. [33], the solenoid valves could operate within 0.03 s. The shortest backpulsing duration could be 35 ms and the fastest frequency could be 10 Hz.

In the filtration with piston-controlled backpulsing, a backpulsing unit is attached on the permeate side of the membrane housing. Fig. 4 (c) illustrates the schematic of a membrane filtration with piston-controlled backpulsing. Backpulses are initiated by an actuator. By operating a solenoid valve pressurized with nitrogen gas or other gas, a gas pulse is directed to the piston [53]. With the expanding of the piston, a permeate flow is reversed through the membrane. During the following forward filtration, the piston cylinder is then filled with permeate for the next backpulse. In the setup of Sondhi et al. [10], backpulsing

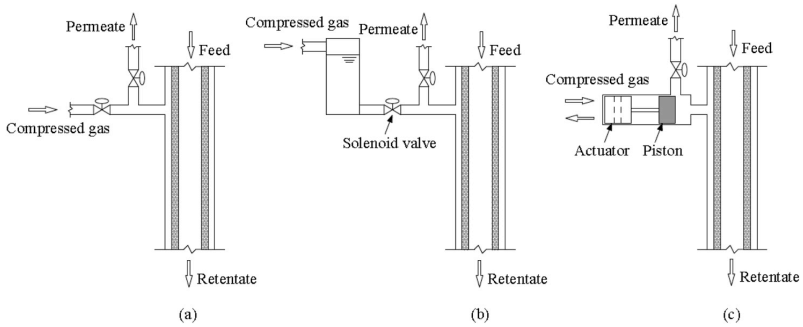

Fig. 4. Schematic of different types of backpulsing setup in a crossflow membrane filtration: (a) Gas backpulsing controlled by solenoid valves; (b) Water backpulsing controlled by solenoid valves; (c) Water backpulsing controlled by a piston device. Adapted from Refs. [24,50].

interval could be as short as 30 s and the backpulsing duration could be down to 0.5 s.

These two common ways to generate backpulsing are summarized and compared in Table 1.

It is easy and common to increase the pressure on the permeate side to get a reverse flow. Nevertheless, a reverse flow can also be generated by regulating the pressure in the feed side. Koh et al. [14] applied a dynamic crossflow pulse (DCP) unit in the feed side to generate backpulses by fluctuating the feed pressure. The DCP unit contained a rotating shut-off valve. As it rotated, the flow in the feed side was temporarily interrupted, causing fluctuated feed pressure. There are two advantages of this design. One is that there is no concern about the contamination of membrane backside caused by reverse flow, since the fluctuated pressure is regulated from the feed side. The other is that the device can generate backpulses with very high frequency (up to 50 Hz) and very short duration (between 5 and 200 ms). The possible disadvantage is that backpulsing parameters are related to each other and cannot be varied independently in this device.

2.2. Theory of backpulsing

There are two approaches to look into the theory of backpulsing. One is to focus on the behavior of a single foulant during backpulsing, the other is to consider the fouling system as a continuum and look at the mass transfer in the system [59].

2.2.1. Acting forces on a deposited foulant during backpulsing

Acting forces on a deposited foulant during the forward filtration has been discussed in literature [59]. Fig. 5 is a schematic of the acting forces on a deposited foulant on the membrane surface at the beginning of backpulsing. After a period of forward filtration, foulants are accumulated and deposited on the membrane. Backpulsing can lift the reversible foulants from the membrane and the foulants can be swept away by crossflow. The forces acting on a deposited foulant during backpulsing period are generally caused by Refs. [59,60].

- Hydrodynamics: drag force by backpulsing, drag force by crossflow velocity (CFV), inertial lift force [61,62];
- Foulant diffusion: Brownian diffusion, shear-induced diffusion;
- Intermolecular interactions (foulant-foulant and/or foulant-membrane): electrostatic interaction, van der Waals interaction, steric effects.

Foulants will leave membrane surface when the exerted drag force by backpulsing associated with inertial lift force and back diffusion is sufficient to overcome the attractive interactions between foulant and foulant and/or between foulant and membrane. Foulants may also flow or roll along the membrane surface when the drag force by CFV is greater than the forces tangential to the membrane surface in the other direction (e.g., forces from intermolecular interactions and diffusion). The combination of all the forces determines the motion of foulants, and thus determines the efficiency of backpulsing on fouling removal. The acting forces are different from foulant to foulant. For large foulants (i.e., foulant size larger than 1 μm), backpulsing is mainly controlled by the hydrodynamics; while at submicron foulant sizes, a further consideration of diffusion and foulant-foulant/foulant-membrane interactions is necessary [59]. In general, these acting forces on foulants are affected by feed properties, membrane properties and operating parameters. The performance of the affecting factors on backpulsing efficiency is discussed in detail in section 4.

2.2.2. Mass transport during backpulsing

The mass transport during forward filtration and before backpulsing is shown in Fig. 6. The rate of substances transporting towards the membrane due to the pressure-driven flow is balanced by the rate of substances penetrating through the membrane plus the diffusive substance rate back into the bulk solution [59]. The mass transport during backpulsing is regarded as pure solvent transport. That is, the solvent flux towards the membrane is equal to the solvent flux passing through the membrane.

The solute concentration profiles in the bulk of an incompressible Newtonian fluid can be described by the governing equations of motion: the continuity equations of solvent and solute as well as the momentum equation [65,66].

The continuity equation for the solvent is

$$\nabla \cdot \boldsymbol{v} = 0 \tag{1}$$

where **v** is flow velocity.

The continuity equation for the solute is

$$\frac{\partial c}{\partial t} + \boldsymbol{v} \cdot \nabla c = D \nabla^2 c \tag{2}$$

where c is the concentration of the solute, D is the diffusion coefficient of the solute.

The momentum equation is

$$\frac{\partial \boldsymbol{v}}{\partial t} + (\boldsymbol{v} \cdot \nabla) \boldsymbol{v} - \frac{\mu}{\rho} \nabla^2 \boldsymbol{v} = -\frac{\nabla P}{\rho} + \frac{S_i}{\rho} \tag{3}$$

where μ is the permeate viscosity, ρ is the density of the solvent, P is pressure, S_i is the source term for the i th (spatial coordinates) momentum equation.

S_i is a source term used to describe the added resistance of the membrane. The flow through the membrane is described using porous media model. The source term is composed of two parts, a viscous loss term (the first term on the right-hand side of Eq. (4)) and an inertial loss term (the second term on the right-hand side of Eq. (4)) [67]:

Table 1
Summary of two common ways of generating backpulsing.

Control Mode	Description	Advantages	Disadvantages	References
Valves	Backpulses are generated by the fast switch of valves in the filtration and backpulsing lines. Compressed gas or water is applied in the backpulsing line.	Flexible; A wide setting range of backpulsing parameters	Backpulsing volume is not under control.	[18,34,54,55]
Piston apparatus	Backpulses are generated by the action of piston, which is actuated by compressed gas.	Easy operation; Known amount of backpulsing volume	Limited options of backpulsing parameters; Frequency cannot be very high due to the refilling time of the piston.	[10,53,56–58]

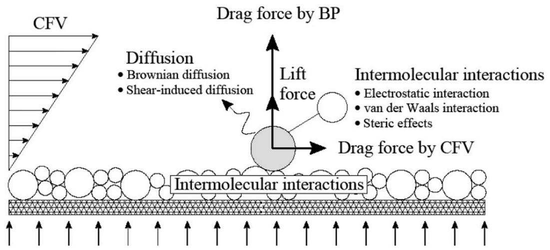

Fig. 5. A schematic of the acting forces on a deposited foulant at the beginning of backpulsing. The foulants are not drawn in scale. Adapted from Refs. [59,63].

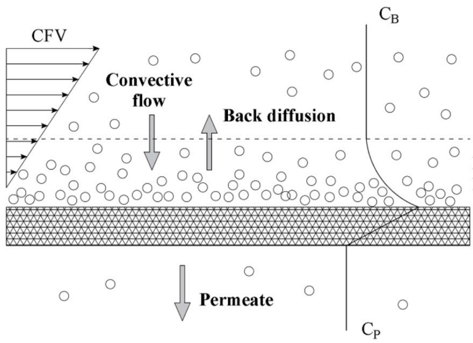

Fig. 6. A schematic representation of mass transport during forward filtration (before backpulsing). Adapted from Ref. [64].

$$S_i = -\left(\sum_{j=1}^{3} D_{ij}\mu v_j + \sum_{j=1}^{3} C_{ij}\frac{1}{2}\rho|v|v_j\right) \quad (4)$$

where $|v|$ is the magnitude of the solvent velocity. D_{ij} and C_{ij} are prescribed material matrices consisting of the porous media resistance coefficients. In laminar flows through porous media, the pressure drop is typically proportional to velocity and the inertial resistance constant C can be considered to be zero. At high flow velocities, the constant C provides a correction for inertial losses in the porous media.

Considering forward filtration and backpulsing period individually, flows through the membrane are under laminar flow conditions. Therefore, the existing models on backpulsing (section 5) are based on Darcy's law and ignoring inertial effects. In constant-pressure filtration mode, the permeate flux J_v can be evaluated in terms of the effective pressure driving force [66,68].

$$J_v = L_p(\Delta P_m - \sigma\Delta\Pi) \quad (5)$$

where L_p is the hydraulic/solvent permeability constant, depending on the membrane structure (i.e., pore size distribution and porosity) and permeate quality (i.e., viscosity) [69]. σ Is the osmosis reflection coefficient. ΔP_m is the hydraulic pressure drop across the membrane, which is forward TMP during forward filtration and reverse amplitude during backpulsing. ΔΠ is the osmotic pressure difference across the membrane, which may be important to consider during forward filtration due to the high concentration of retained solutes in the boundary layer near the membrane surface, while is negligible during backpulsing period.

The momentum equation can also be simplified to

$$\rho\left(\frac{\partial \boldsymbol{v}}{\partial t} + \boldsymbol{v}\cdot\nabla\boldsymbol{v}\right) = \nabla\boldsymbol{\Pi} \quad (6)$$

Where **Π** is the total stress tensor.

Considering backpulsing and forward filtration together, it is a dynamic process with frequent flows through the membrane back and force. The flow regime of concern contains accelerations and decelerations of the fluid through the membrane pore space. Therefore, the inertial loss term in Eq. (4) should be taken into consideration [70]. However, it is not implemented into backpulsing models yet.

2.2.3. Rapid/high-frequency backpulsing

During forward filtration, the feed flow transports substances towards the membrane due to the positive TMP. A concentration gradient or boundary layer of the rejected particles in MF or solutes in UF forms near the vicinity of the membrane (Fig. 6). As the concentration at the membrane surface reaches the maximum packing or gel concentration (the convective flux towards the membrane and the diffusive flux away from the membrane are in balance), a stagnant cake or gel layer develops which offers a significant hydraulic resistance to the permeate flow. In rapid/high-frequency backpulsing, a primary goal is to prevent the formation of the stagnant layer. That is, rapid backpulsing/high-frequency backpulsing should start as soon as the concentration at the membrane surface reaches the maximum packing or gel concentration [71]. Where early removal of cake and gel layer is less important, there is less need for rapid/high-frequency backpulsing.

The transient development of the boundary later during rapid/high-frequency backpulsing is calculated using Eq. (2), under the hypotheses of a Newtonian fluid with constant density and viscosity and complete rejection of particles or solutes. The permeate flux during the boundary layer remains equal to the initial flux J_0, the boundary-layer thickness is $\delta = D/J_0$ and the time for the development of boundary-layer/short residence time of the particles or solutes in the vicinity of the membrane is $t_c = D\ (C_C\text{-}C_B)/(J2\ 0C_B)$. This time approximation is used in Model III and Model V in section 5.1. In the case of particles with a diameter of 5 μm in MF where shear-induced diffusion is dominant and macromolecules with a Stokes-Einstein diameter of 10 nm in UF where Brownian diffusion is dominant, the time scales for the boundary-layer formation are both approximately 1 s, indicating that the high frequency on the order of 1 Hz is needed to prevent cake or gel formation.

3. Applications of backpulsing

3.1. Lab-scale experiments

Backpulsing was first investigated by Victor Rodgers in 1989 for protein UF [72]. Significant improvements were achieved when backpulsing was used. The permeate flux produced by the UF of a single solute (1% albumin) with backpulsing was twice the limiting flux for the non-pulsed case. The increase in the net flux due to backpulsing was equivalent to that in the conventional filtration by increasing the crossflow rate as much as 200 times [42]. In the UF of binary solutes (1% albumin and 0.3% gamma-globulin), both the solute flux and the solvent flux increased significantly with backpulsing. Mathematical models revealed that backpulsing was effective in reducing both the fouling resistance and the concentration polarization resistance in such systems [11,41,42]. Since then, increasing studies on backpulsing have been carried out to mitigate membrane fouling in various membrane configurations, see Fig. 7(a). Most of the backpulsing tests were carried out with tubular (44.59%) and flat-sheet (35.14%) membranes. Experiments of backpulsing in hollow-fiber (16.22%) [21,50,54,73–75] and spiral-wound (4.05%) [76,77] membrane modules also showed good performance on enhancement of membrane permeability. Studies of backpulsing have also been conducted in many application fields, such as water and wastewater treatment (48.44%), food industry (23.44%), biotechnology applications (23.44%) and some other industries (4.69%), see Fig. 7 (b). A list of detailed lab-scale experiments about backpulsing is shown in Table 2. Besides the results showing how efficient backpulsing is to mitigate fouling, the characteristics of feed solutions, membranes, as well as (optimal) backpulsing conditions are

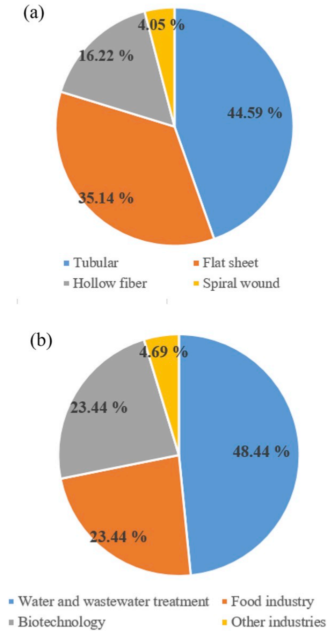

Fig. 7. The ratios of backpulsing used in (a) different membrane configurations and (b) different application fields.

also listed in the table for a better understanding.

3.2. Pilot- and industrial-scale applications

Backpulsing technology shows good performance in membrane fouling mitigation not only in lab-scale experiments, but also in pilot- and industrial-scale tests. Following are examples of backpulsing applications in the treatment of industrial wastewater.

3.2.1. Effect of backpulsing on the purification of radioactive wastewater

Membrane technology applied in the treatment of wastewater from nuclear facilities has one more concern than fouling problem because of the exposure to radiation. A decrease of membrane lifetime and membrane performance can occur because the radiation may alter membrane structure, especially for polymeric membranes [98,99]. The more deposition of the radioactive substances, the higher radiation dose rate on the membrane. The effect of backpulsing on removing the deposition of radioactive substances was studied in a pilot plant [100]. The membranes were silicon carbide (SiC) membranes with an average pore size of 70 kDa and a total surface area of 0.3 m^2. Backpulsing with time intervals of 5 and 10 min were tested. Results showed that backpulsing was effective to reduce the deposition of radioactive substances on the membrane and the deposition was lower for backpulsing with interval of 5 min.

3.2.2. Effect of backpulsing on the separation of valuables and dispersed substances in thermomechanical pulp process water

The process streams during the production of thermomechanical pulp contain several valuable substances. Galactoglucomannan (GGM) is a hemicellulose that attracts great interest as a value-added chemical. Krawczyk and Jonsson [101] used MF to remove dispersed substances and recover GGM in the permeate from pulp mill process water. They carried out filtrations with a membrane element that is common for large-scale applications, a multi-channel alumina ceramic membrane with a length of 1.0 m and an area of 0.13 m^2. Backpulsing had a

Table 2
A summary of backpulsing performance in lab-scale applications.

Feed solutions	Membrane properties	(Optimal) backpulsing conditions	Results	References
Water and wastewater treatment				
Olive mill wastewaters	SiC, 0.04 µm, TM, CF	ΔP_b = 3 bar, t_b = 0.8 s, t_i = 10 min	When the filtration tests were running at a constant flux of 67 LMH, the TMP increase during 24 h test was reduced from 0.53 bar to 0.48 bar by using backpulsing.	[78]
Industrial wastewaters with high concentration of heavy metals	ZrO_2, 210 kDa, TM, CF	P_b = 7 bar, t_b = 1 s, t_i = 2 min, V_b = 3 mL	Backpulsing was used to enhance the efficiency of chemical cleaning.	[79]
Real produced water from Arabian Gulf	SiC, 0.04/0.5 µm. TiO_2, 50 kDa/2 µm. TM, CF	P_b = 3 bar, t_b = 0.8 s, t_i = 330 s	Both backpulsing and backflushing were used to mitigate membrane fouling and SiC membranes offered a higher overall permeability than TiO_2 membranes.	[80]
NaCl solutions	PA, nanofilter, TFC, CF	ΔP_b = 6 bar, t_b = 0.9 s, t_i = 150 s	The recovery of permeate flux when using backpulsing increased clearly with increasing salt concentration in the feed. A more than 37% improvement of permeate flux was observed in the maximum feed concentration.	[81]
Real and synthetic produced water	ZrO_2, 0.1 µm, TM, CF	P_b = 7 bar, t_b = 0.5 s, t_i = 5 min	A 36% improvement of permeate effective flux could be achieved when both backpulsing and backwashing technologies were applied.	[82]
Synthetic produced water	α-Al_2O_3, 0.1/0.2/0.5 µm, TM, CF	ΔP_b = 0.4 bar, t_b = 0.2 s, t_i = 5 s	Backpulsing could avoid and reduce further deposition of foulants on the membrane surface. The fouling rate could be reduced significantly for all the membranes.	[55]
Dextrin solutions	PS, 100 kDa, SW, CF	ΔP_b = 38 kPa, t_b = 0.17 s, t_i = 1 s	The saturation flux with backpulsing was 82% of the clean water flux and 3.9 times the saturation flux without backpulsing.	[76]
Olive mill wastewater	ZrO_2–TiO_2, 0.1 µm, TM, CF	ΔP_b = 1–3 bar, t_b = 1 s, t_i = 1 min	The permeate flux of about 100 LMH was observed and was around 10% higher than flux obtained without backpulsing. In the long term backpulsing made it possible to maintain a constant permeate flux for 10 days.	[83]
Biologically treated wastewater	ZrO_2, 20/50 nm, TM, CF	P_b = 7 bar, t_b = 0.5 s, t_i = 1 min	9% and 17% improvements of permeate flux were obtained using backpulsing for membranes of nominal pore size 20 nm and 50 nm, respectively.	[84]
Phenol-containing wastewater	ZrO_2–TiO_2, 0.1 µm, TM, CF	ΔP_b = 0.8, 2.8 bar, t_b = 1s, 2 s, t_i = 30 s, 45 s, 1 min	Backpulsing could increase the permeate flux by 10% and maintain a high constant flux over several weeks.	[85]
Electroplating wastewater containing $Cr(OH)_3$ suspensions	α-Al_2O_3, 0.05/0.2/0.8 µm, TM, CF	ΔP_b = 170 kPa, t_b = 0.5 s, t_i = 30 s	The steady-state fluxes without backpulsing were 2100, 2500 and 2000 LMH for 0.05, 0.2 and 0.8 µm membranes, respectively, while fluxes with backpulsing were 4700, 6250 and 9500 LMH, respectively	[57]
Board industry wastewaters	α-Al_2O_3, 0.1 µm, FS, CF	P_b = 4 bar, t_b = 1 s, t_i = 1 min	Backpulsing improved the permeate flux by 20–57 LMH (11–28%).	[17,86]
A bentonite suspension, biologically treated wastewater and an activated sludge suspension	α-Al_2O_3, 0.2 µm, TM, CF	ΔP_b = 2 bar, t_b = 0.05 s, f = 0.2 Hz	A 4 – 8-fold improvement of permeate flux was obtained with backpulsing than without backpulsing for the filtration of bentonite suspension. No significant improvement of permeate flux was observed for biologically treated wastewater or activated sludge suspension.	[36]
Oil/water emulsion	ZrO_2, 0.1 µm, TM, CF	ΔP_b = 400 kPa, t_b = 0.2, 0.5, 1 s	Backpulsing could maintain permeate flux at a level which was nearly 3-fold over the long-term flux without backpulsing.	[24]
Oil/water emulsion	ZrO_2, 0.1 µm, TM, CF	ΔP_b = 3 bar, t_b = 0.7 s, t_i = 1 min, V_b = 3 mL	The steady-state flux without backpulsing was 450 LMH and the steady-state flux with backpulsing was 1000 LMH.	[87]
A primary municipal sewage effluent	Ceramic, 0.35 µm, TM, CF	Tb = 0.3 s, t_i = 30 s	A 65% higher flux was effectively maintained with backpulsing.	[88]
Clay suspensions and dilute oil-in-water dispersions	Al_2O_3, 0.2/0.8 µm, TM, CF. PS, 0.2/0.45 µm, HF, CF	ΔP_b = 1.4 bar (20 psi), t_b = 0.2, 0.5 s, f = 0.01–0.1 Hz	Backpulsing enhanced the long-term permeate flux in the filtration of bentonite clay suspensions from 3 to 10-fold over the flux without backpulsing. For the experiments of dilute oil-in-water dispersions, the permeate flux was increased up to 25 times.	[19]
Food industry				
Skim milk with a fat content below 0.1%	α-Al_2O_3, 1.4 µm, TM, CF	P_b = 6 bar, t_b = 0.2 s, t_i = 1 min, V_b = 3 mL	At a constant flux of 350 LMH, experiment with backpulsing lasted for over 11 h without any sign of fouling, while filtration without backpulsing lasted only around 5.4 h. Backpulsing efficiency and the optimal backpulsing conditions were dependent on settings of fluxes.	[89–91]
Whole milk	Silicon nitride microsieves, 0.8 µm, FS, CF	ΔP_b = 15 kPa, f = 15 Hz	High fluxes (5000 up to 27 000LMH) could be maintained for more than 2 h using backpulsing.	[49]
Bovine and ovine milk	Ceramic, 1.4 µm, TM, CF	P_b = 7 bar, t_b = 1 s, t_i = 1 min, V_b = 3 mL	No permeate was observed without backpulsing, while a permeate flux of 410 LMH was obtained using backpulsing.	[92]
Beer	α-Al_2O_3, 0.5 µm, TM, CF	ΔP_b = 1.7, 1.2, 2.3, 3 bar, t_b = 0.5, 0.3, 0.2, 0.1 s, t_i = 1, 0.5, 0.25, 0.125 min	24 h average flux was increased to 21.6 $kg \cdot h^{-1} m^{-2}$ by using multi-stage backpulsing, which is 442% improvement of the flux from filtration without backpulsing.	[93]
Skim milk	α-Al_2O_3, 1.0 µm, TM, CF	P_b = 1 bar, t_b = 0.022 s, t_i = 0.33 s	Backpulsing increased the permeate flux by 100% and was possible to maintain filtration for 13 h without other cleaning.	[30]

(continued on next page)

Table 2 (*continued*)

Feed solutions	Membrane properties	(Optimal) backpulsing conditions	Results	References
Biotechnology				
BSA solution	Silicon nitride microsieves, 1.2 µm, FS, CF	ΔP_b = 5, 8 kPa, t_b = 20 ms, f = 3.3, 5, 6.7 Hz	Fouling rate with backpulsing reduced almost half that without backpulsing.	[94]
Unconjugated polysaccharide	PS, 0.1/0.2 µm, HF, CF	ΔP_b = 69–83 kPa (10–12 psi), t_b = 0.8 s, t_i = 1 min	A 3.5–10-fold reduction in buffer usage and a 3 – 8-fold reduction in cycle time were accomplished by using backpulsing. Removal of unconjugated polysaccharides was also increased by backpulsing, thus increased the efficiency of membrane.	[50,54]
Yeast cell suspension	AN, 0.2 µm, FS, CF. CA, 0.2 µm, FS, CF	AN: ΔP_b = 14 kPa, t_b = 0.5 s, t_i = 10 s. CA: ΔP_b = 21 kPa, t_b = 0.4 s, t_i = 10 s	The steady-state flux was increased twice with backpulsing under optimum conditions for AN membrane. The steady-state flux with backpulsing was almost 4 times the long-term flux without backpulsing for CA membrane.	[95]
E. coli bacteria suspensions	CA, 0.22 µm, FS, CF	ΔP_b = 69 kPa (10 psi), t_b = 0.5 s, t_i = 10 s	The steady-state flux without backpulsing was 40 LMH and the steady-state flux with backpulsing was 250 LMH.	[18]
Yeast homogenate suspensions	Ceramic, 0.2 µm, TM, CF	ΔP_b = 0.5 bar, t_b = 0.1 s, f = 1 Hz	Backpulsing could increase the transmission of proteins during the MF of yeast homogenate suspensions at high concentrations. An up to 6-fold increase in the mass flux of a target enzyme was observed by using backpulsing.	[56]
Bacterial suspensions and whole bacterial fermentation broths	CA, 0.2 µm, FS, CF	ΔP_b = 69 kPa (10 psi), t_b = 0.1, 0.2, 1 s, t_i = 0.2–40 s	An approximately 10-fold higher net permeate flux was achieved under optimum backpulsing conditions for the washed bacterial suspensions and a 2-fold improvement of the flux was obtained for the fermentation broths.	[20]
Bacterial lysates	CA, 0.22 µm, FS, CF	ΔP_b = 69 kPa (10 psi), t_b = 0.09 s, f = 2.5 Hz	A 10-fold improvement in permeate flux and 100% protein transmission was obtained under optimal backpulsing conditions for the filtration of a very dilute solution.	[96]
Other industries				
Waste slurry containing insoluble solids	Stainless steel, 1–3 µm, TM, CF	ΔP_b = 4.8 bar (70 psi), t_i = 30 min	Averaged permeate flux was much higher when backpulsing was applied. However, the irreversible fouling increased during backpulsing operations because the recovered flux was lower with backpulsing.	[97]
Light non-aqueous phase liquids	PE, 0.1 µm, HF, DE	ΔP_b = 60 kPa, t_i = 500 s	Backpulsing was able to mitigate pore constriction and cake layer formation. It improved the recovery rate about 30% over a 15 h extraction period.	[21]

Note: ΔP_b, backpulsing amplitude; P_b, backpulsing pressure (pressure applied in the permeate side); t_b, backpulsing duration; f, backpulsing frequency; t_i, backpulsing interval (t_i = 1/f); V_b, backpulsing volume. AN, anopore; CA, cellulose acetate; PA, polyamide; PE, polyethylene; PS, polysulfone; SiC, silicon carbide; TFC, thin-film composite. TM, tubular membrane; HF, hollow fiber; FS, flat sheet; SW, spiral wound. CF, crossflow; DE, dead-end. BSA, Bovine serum albumin. LMH, liter per square meter per hour.

positive effect on both flux and permeability of GGM at CFV of 3 m/s. Almost 40% higher flux and 9% higher permeation of GGM was obtained by backpulsing with a duration of 0.25 s and forward filtration time of 5 s.

3.2.3. Effect of backpulsing on the filtration of produced water

Chen et al. [102] used chemical pretreatment and backpulsing techniques to enhance the performance of the crossflow MF of real produced water from an offshore platform in the Gulf of Mexico, which contained oil and grease of 28–108 mg/L and total suspended solids (TSS) of 100–290 mg/L. Multichannel ceramic membranes were tested in the pilot-plant studies with a filtration area of 0.2 m^2 and an average pore size of 0.5 μm. Backpulsing was carried out with an amplitude of 5.5 bar (80 psi) and a duration of 0.5 s every 2 min. Without feed pretreatment and backpulsing, the filtration could only last for less than 4 h at a constant flux of 1538 L/(m^2·h) (906 gal/(ft^2·D)). Whereas, with backpulsing and the pretreatment of chemical coagulation, the filtration could last 66 h or longer without chemical cleaning at an even higher flux of 2237 L/(m^2·h) (1318 gal/(ft^2·D)).

Weschenfelder et al. [82] conducted a long-term test of synthesized produced water with oil and grease concentration of about 100 mg/L using a pilot-scale membrane filtration. The membranes were multichannel zirconia membranes with 3.4 m^2 filtration area and 0.1 μm average pore size. Different filtration procedures of backwashing and backpulsing were tested. The backwashing lasted 1 min at 2.0 bar every 30/60 min and the backpulsing had a duration of 0.5 s at the amplitude of 7.0 bar every 5 min. A clear increase in permeate flux was observed when backwashing/backpulsing were applied. Applying backpulsing only, the effective net permeate flux was 16% higher than that without backpulsing/backwashing. When backwashing was used, applying backpulsing could still increase the permeate flux by 5%. When both backwashing and backpulsing were used, the filtration of the synthetic PW could generate a net flux of 243 L/(m^2·h) for 100 h under ΔP_f = 2.0 bar, CFV = 2.0 m/s.

Furthermore, long-term flux stability was achieved on a commercial-scale installation equipped with backpulsing devices in the treatment of produced water containing 10–100 mg/L oil and suspended solids. The filtration system used Membralox® membranes with an average pore size of 0.8 μm and a total filtration area of approximately 115 m^2. The operation was extremely stable with a continuous flow rate of about 850 m^3/day and an average flux of 300 L/(m^2·h) over several months [57].

4. Factors affecting backpulsing efficiency

Backpulsing mainly involves the detachment of substances from membrane surfaces and membrane pores. Factors affecting fouling formation (the way substances attach to the membrane) as well as the backpulsing conditions (how to remove the attachment from the membrane) have influence on the efficiency of backpulsing. In general, the affecting factors can be divided into three groups: feed properties, membrane properties and operating parameters.

4.1. Feed properties

4.1.1. Types of foulants

The effect of backpulsing on fouling mitigation depends on the types of membrane fouling that is mainly determined by the nature of the feed solutions. As is mentioned in 2.1.1, backpulsing is efficient to remove external and non-adhesive fouling. Sondhi et al. [10,57,103] studied the effect of backpulsing on the MF of synthetic electroplating wastewater containing mainly inorganic compounds. It is observed that all the permeate fluxes recovered 100% with backpulsing. The steady-state net fluxes were 2–5 times of the fluxes without backpulsing [57]. Whereas in the filtration of biologically treated municipal wastewater, the permeate flux improved slightly with backpulsing. The highest enhancement of the steady-state net flux due to backpulsing were only 9.13% and 17% for membranes of 20 nm and 50 nm average pore size, respectively [84]. Similar results were obtained by Barrios-Martinez et al. [85]. They achieved only a 10% increase of permeate flux when backpulsing was used in the filtration of a biological wastewater containing a high mixed liquor suspended solids (MLSS) concentration of 10 g/L.

Ma et al. [104] investigated the effect of backpulsing on the crossflow MF of different feed solutions in the same filtration system and operating conditions. 5-Fold and 1.3-fold permeate volume enhancement were obtained for the filtration of bentonite clay and crude oil, respectively. The recovered fluxes after water backwashing were approximately 80% and 30% of the initial fluxes for bentonite suspensions and oil emulsions, respectively, which confirms that membrane fouling is more difficult to remove in the filtration of adhesive substances.

4.1.2. Feed concentration

Backpulsing is less effective for feed water with a higher concentration because of possible more and denser fouling. Kuberkar et al.'s [20] carried out the filtration of a washed bacterial suspension with a dry cell weight of 1.2 g/L, the steady-state net flux obtained with backpulsing increased almost 12.5-fold over the flux obtained without backpulsing. Whereas in the filtration of a suspension with higher bacterial concentration (10 g/L dry weight), the steady-state net flux with backpulsing was 7-fold greater than the flux of the normal crossflow operation. The cake formation and flux decline were faster for the suspension with a higher concentration. Parnham et al. [96] demonstrated that backpulsing was inefficient in the improvement of the steady-state flux when feed cell lysate concentration exceeded 10 g/L, although backpulsing was able to enhance the net flux up to 10-fold for 2.5 g/L feed concentration. Mores et al. [18] set up a mathematical model to predict the optimum backpulsing conditions in the filtration of a bacteria suspension. The maximum fraction of the cake that could be removed by backpulsing was found to drop from 0.4 to 0.6 for 0.01 g/L bacteria in the feed to 0.1–0.2 for 1 g/L bacteria in the feed, indicating that backpulsing was less effective when feed concentration increased.

4.2. Membrane properties

4.2.1. Membrane material

When backpulsing is used in polymeric membranes, a minute membrane motion is observed, which is not noticed for inorganic membranes. The vibration caused by rapid pressure changes from the permeate side can shake fouling layers off the membrane surface and decrease concentration polarization. Czekaj et al. [58] evaluated the influence of infrasonic pulsing on fouling layer removal and flux improvement. The so-called infrasonic pulsing refers to the process similar to backpulsing but with low reverse pressure that is lower than the TMP. Permeate is obtained during both the filtration and the backpulsing period, which means foulants deposited on the membrane surface and in the membrane pores could only be removed by membrane vibration, and not by the reverse flow of permeate. The results showed that the membrane vibration was efficient in removing a portion of fouling layer. A 4-fold improvement of the steady-state net flux was obtained compared to that obtained in normal filtration during the MF of a model turbidity suspension with a polyvinylidene fluoride (PVDF) flat-sheet membrane. 2.4-fold and 2.1-fold enhancement of the net fluxes were obtained during the filtration of wine and beer samples, respectively, with a PS hollow-fiber membrane [58]. In addition, as shown in experiments of Rodgers and Sparks [42], the concentration polarization boundary layer was also altered by the minute but significant membrane motion, resulting in a significant improvement of the solute flux during the filtration of a 1% albumin solution. Similar results were observed by Girones et al. [105] that relative constant fluxes could be achieved at a constant TMP by using polyether sulfone (PES) polymeric microsieves with backpulsing when filtering protein solutions, skimmed milk and white beer. However, such backpulsing effectiveness and membrane permeability was not obtained by silicon nitride microsieves that were more rigid and had little vibration.

Note that mechanical stresses caused by frequent changes of pressure transition is a big inducement to membrane ageing and damage. Huisman and Williams [106] found that PS hollow-fibers used in a cosmetics company for wastewater treatment were mainly damaged by mechanical forces (high local shear forces or vibrations caused by pressure shocks) rather than chemical agents. In research of Zondervan et al. [75], of the four ageing factors: fouling status of membrane, cleaning agent concentration, magnitude of backpulsing and number of backpulses, fouling status in combination with the number of applied backpulses were significant ageing factors for their PES hollow-fiber membranes. Compared with ceramic membranes, polymeric membranes are more sensitive to the mechanical stresses (pressure differentials) during backpulsing [107]. In order to use backpulsing process to mitigate membrane fouling, mechanical fatigue of membranes should be taken into consideration and fatigue analyses should be conducted.

4.2.2. Surface properties

The surface properties of membranes, e.g., hydrophilicity and surface charge, affect the tendency of fouling formation during filtration, thus indirectly affecting the cleaning efficiency of backpulsing.

Membranes with higher hydrophilicity produce higher water fluxes because of the affinity to water. Kim et al. [108] modified a polyvinyl chloride (PVC) membrane with N-vinyl-2-pyrrolidinone (NVP) monomer to increase the surface hydrophilicity. The permeate volume of the backpulsed MF of activated sludge using unmodified membrane was 4.16 times that without backpulsing. The permeate volume of the filtration combined backpulsing and membrane modification was 6.33 times that of the unmodified membrane without backpulsing. Similar results were also reported by Ma et al. demonstrating the synergistic effect of backpulsing and surface modification for the filtration of crude oil emulsions [104] and bacterial suspensions [16]. The higher permeate improvement obtained from backpulsing and membrane modification is attributed to the weaker hydrophobic-hydrophobic interactions between foulants and the modified membrane surface. In the filtrations of bovine serum albumin (BSA) solutions and skimmed milk, Girones et al. [109] also observed reduced fouling and enhanced flux using polyethylene glycol (PEG)-modified microsieves in combination with backpulsing, as PEG-based compounds are protein- or cell-resistant.

Surface charge of membrane has an effect on membrane permeability and backpulsing efficiency. Ma et al. [16] used modified and unmodified PP membranes for the filtration of CML particles that are hydrophilic and negatively charged. Without backpulsing CML fouling was not strongly dependent on membrane surface chemistry. With backpulsing, however, a significant higher permeate volume collected in 1 h was obtained for the membrane modified with acrylic acid (AA) (hydrophilic and negatively charged), and a lower volume was acquired for membrane modified with dimethyl aminoethyl methacrylate (DMAEMA) (hydrophilic and positively charged), compared to that of the unmodified PP membrane (hydrophobic and neutral). When membrane has the same negative charge as CML, higher membrane permeability with the help of backpulsing was observed due to the electrostatic repulsive forces. This electrostatic interaction was then found to be affected by ionic strength. When ionic strength increased, both the permeate flux and the recovered water flux decreased. Because the electric double layer of the CML particles decreased and the repulsion between CML particles and the AA-modified membrane surface decreased, causing CML particles easier to deposit on the membrane surface.

4.2.3. Membrane pore sizes

There is a close relationship between membrane pore sizes and fouling types. If foulant sizes are smaller than or similar to membrane pore sizes, pore constriction and pore blocking tend to be the dominate fouling. While if foulant sizes are larger than membrane pore sizes, there will be more external fouling like gel or cake formation. Therefore, membrane pore sizes also influence the efficiency of backpulsing on the membrane performance. In the work of Sumihar Silalahi [110] on the MF of synthetic produced water, ceramic membranes with different average pore sizes of 0.1, 0.2 and 0.5 μm were tested. The results showed that the fouling of the tighter membrane (average pore size of 0.1 μm) was dominated by external fouling. Thus, a lower backpulsing amplitude (0.25 bar) was needed to remove the fouling deposition. While membranes with more open pore sizes (0.2 and 0.5 μm) were more prone to internal fouling because of more adsorption of foulants into the membrane pores. Backpulsing with a higher amplitude (0.5 bar) was then needed. The effect of backpulsing duration and frequency showed a similar tendency as backpulsing amplitude. Krawczyk and Jönsson [101] used tubular ceramic membranes with average pore sizes of 0.2, 0.4 and 0.8 μm for the MF of thermomechanical pulp process water to recover a kind of hemicellulose. The membrane with the smallest pore size (0.2 μm) had the highest permeate flux and lowest flux decline. This is due to severe pore blocking in the membranes with bigger pores [111]. In addition, the pure fluxes of the more open membranes could not recover after several cleaning cycles, which confirms irreversible fouling of these membranes.

However, other researchers observed different results. Zhao et al. [112] investigated the filtration of TiO_2 particles from acid solutions, where the dominant fouling mechanisms were cake layer formation. The flux restoration for 1.0-μm membrane was higher than that for 0.2-μm membrane by the same backpulsing operation. There were two possible reasons for this phenomenon. One was that the flux decline for 0.2-μm membrane was lower than that for 1.0-μm membrane, thus the flux recovery for 0.2-μm membrane was not as evident as that for 1.0-μm membrane. The other was that the membrane and fouling resistance of 0.2-μm membrane was larger than that of 1.0-μm membrane. Therefore the resistance for backpulsing was also higher for 0.2-μm membrane. Similar results were obtained by Sondhi et al. [10,57] where removal of $Cr(OH)_3$ suspensions with crossflow MF were investigated. The bigger the pore sizes of the membrane were, the lower cleaning time (backpulsing duration) was needed, which means the more effective the backpulsing was.

4.3. Operating parameters

4.3.1. Backpulsing parameters

4.3.1.1. Amplitude. Backpulsing amplitude is a prerequisite to the success of backpulsing. A minimum amplitude is required for an effective backpulsing to occur. McAlexander et al. [21] verified that backpulsing frequency and duration was not the parameters to control the degree of cleaning by backpulsing. Only when backpulsing amplitude was big enough could optimum membrane cleaning be performed. The minimum amplitude was 60–85 kPa in their dead-end MF of a light non-aqueous phase liquid obtained from a refinery site. Sondhi et al. [10] found a minimum amplitude of 175 kPa to initiate cleaning by backpulsing in their experiments filtering $Cr(OH)_3$ suspensions through porous alumina ceramic membranes. In cases where external fouling is the main fouling mechanism, membrane cleaning efficiency increases with increasing amplitude to a certain point, above which the cleaning efficiency levels off [10,112]. Mores et al. [18] found that the pressure inflection point was 34 kPa (5 psi) for their MF of bacterial cells.

It should be noted that for backpulsing applied in polymeric membrane systems, the highest limit of the reverse TMP is an important factor to be concerned. If the backpulsing amplitude is too high, the membranes will be damaged. The upper limit of reverse pressure for the hollow-fiber cartridge used in the experiments of Meacle et al. [50] was less than 69 kPa (10 psi) over the feed side at any point of the membrane. Moreover, high intensity backpulses can also break up particles with loose or fragile structures in the feed, resulting in more irreversible fouling. Increased irreversible fouling of the membranes was observed in the filtration of yeast suspensions when the amplitude was 21 kPa (3 psi) or more, which was caused by debris and intracellular matters of the ruptured cells [113].

4.3.1.2. Frequency. Frequency is an important parameter in a backpulsing process. Edmundo et al. [49] studied the effects of several process variables on the microsieves performance during the filtration of whole milk. Compared with inlet-outlet pressure gradient and backpulsing amplitude, TMP and backpulsing frequency influenced the permeate flux the most.

In theory, backpulsing with higher frequency prevents deposition on the membrane to be fully formed and compacted. If a noticeable decrease of flux occurs, more frequent backpulsing will be required [114]. However, in some cases, higher frequency does not improve the membrane productivity. In the experiments of Wilharm and Rodgers [38], backpulsing showed little improvement of the net flux or even a decrease of flux when filtering a binary protein solution through UF membranes. This was because of the extra loss of permeate and less forward filtration time due to the high frequency backpulsing (0.5 Hz). There exists an optimum setting of backpulsing frequency. The effect of frequency on the net permeate flux and the sieving coefficient was investigated during the diafiltration of Pneumoconjugate vaccine serotypes with backpulsing duration of 0.1 s [50]. The interval between backpulses ranged from 5 s to 25 min. It is clear to see that backpulsing with the interval of 30 s had the best performance (the highest permeate flux and the highest sieving coefficient). Membrane damage because of backpulsing in this experiment was after 30 h diafiltration with the highest backpulsing frequency (interval of 5 s).

4.3.1.3. Duration. The effect of duration on backpulsing efficiency varies from applications to applications. It depends on the types of membrane fouling and the mechanisms of fouling removal. Meacle et al. [50] studied purification of a polysaccharide-protein conjugate vaccine using a hollow-fiber cartridge with PS membranes. Backpulsing duration (ranged from 0.1 s to 15 s) was not a significant parameter to improve the sieving coefficient of unreacted polysaccharide. It is thought that the removal of membrane foulants was due to a wave propagation caused by membrane motion instead of the reverse flow through the membrane. Mores et al. [18] developed a model to predict the optimum backpulsing duration and frequency for the filtration of washed bacterial cells with a flat-sheet CA membrane. They declared that it is more important to select an appropriate duration for a given frequency than a proper frequency for a given duration.

4.3.2. Backpulsing strategies

The effect of individual operating parameters has been discussed previously. It is apparent that all the parameters have an influence on the efficiency of backpulsing and that there exists an optimum value for each parameter for a given system. Moreover, these parameters are related to each other. For instance, the extra loss of permeate due to increasing backpulsing amplitude can be compensated by decreasing pulse duration. In the experiments of Sondhi et al. [10], the requirement of backpulsing duration reduced from 3.2 s to 2.4 s when the backpulsing amplitude increased from 180 kPa to 300 kPa.

Mores and Davis [15] studied different backpulsing strategies during the crossflow MF of yeast cells. The cleaning effectiveness was compared under various conditions. Under conditions of high shear rate and high amplitude, short backpulsing of 0.1 s was most effective in fouling removal. With the same accumulative duration time, backpulsing using a greater number of shorter backpulses was more effective than using a lower number of longer backpulses. However, under conditions of high amplitude low shear rate and low amplitude high shear rate, backpulses of 0.2 s and 0.4 s showed the best performance respectively.

As has been illustrated, if the backpulsing with too high amplitude, too long duration or too fast frequency, there will be excessive loss of permeate. In some extreme cases, the loss of time due to backpulsing could be up to 50% of the total filtration time [14]. In order to minimize the permeate loss, Gan et al. [93,115] combined gas backpulsing and water backpulsing in their beer clarifying experiments. By using CO_2 as a complimentary backpulsing media, permeate loss reduced 40% compared to only water backpulsing and the cleaning effectiveness was similar. To control irreversible fouling over time, a multi-stage backpulsing strategy was developed with regard to the flux at different filtration stages. In early filtration stages, backpulsing with lower amplitude, longer duration and slower frequency was used. More frequent backpulses with higher amplitude and shorter durations were used later on to control more severe fouling. The net permeate during the 10-h filtration with the customized backpulsing strategy was increased by 400% compared to the permeate without backpulsing [116].

4.3.3. Other filtration parameters

Not only the three backpulsing parameters, but the filtration parameters have influence on backpulsing efficiency, e.g., TMP/flux and CFV.

The difference in TMPs under constant-pressure mode or fluxes under constant-flux mode means the difference of filtration intensity. During intense filtrations, the vertical drag force towards the membrane is stronger. Foulants are pressed more strongly on the membrane surface as well as into the membrane pores, which is more difficult to be removed by backpulsing. Wen et al. [54] investigated the purification of a polysaccharide conjugate vaccine from its unconjugated polysaccharide. Backpulsing enhanced the performance of the purification by increasing both the permeate flux and the sieving coefficient of the unwanted substances. However, the rate of the enhancement was reduced with increasing TMP. At high TMP values of 69 and 103 kPa (10 and 15 psi), backpulsing had almost no improvement on the permeate flux. Similar tendency was observed by Barrios-Martinez et al. [85]. Wang et al. [117] used an interfacial model and direct microscopic observation to study the microbial adhesion to the polymeric membranes during the UF of yeast cell suspensions. When fluxes were below the critical flux, deposited cells were almost completely removed by backpulsing, even with the lowest intensity backpulsing. Whereas at supercritical fluxes, most cells were irreversibly deposited.

Backpulsing and crossflow address the same fouling problem [50]. A synergistic effect exists between them. Both Krawczyk and Jonsson [101] and Baruah et al. [118] found that backpulsing is more effective at low shear rates and less effective at high shear rates in their experiments. This is because the positive effect of backpulsing could not offset the loss of permeate and filtration time due to backpulsing, when most deposits had already been removed by high crossflow. Even though in some cases normal filtrations with high CFV can have similar performance to the filtration with the reverse flow cleaning steps [119], the application of backpulsing can help reduce the requirement of CFV and decrease the energy consumption of crossflow [34]. Arkell et al. [90] combined crossflow MF and backpulsing to remove bacterial content and spores in a milk solution. Under the optimum conditions of backpulsing, CFV reduced from 5.8 m/s to 4.5 m/s without significant deterioration in membrane performance, which reduced energy consumption by 50% in their system.

5. Modelling of backpulsing

Membrane filtration with backpulsing is a complex and dynamic process. Modelling helps to obtain useful information, especially on fouling and cleaning situations [33]. The efficiency of backpulsing depends on various factors. A reliable modelling can reduce the time and cost associated with running tedious experiments in order to find the optimized backpulsing conditions [69]. This section summaries the development and applications of the main backpulsing models.

The most interesting aspect of modelling a membrane filtration is flux prediction under constant-pressure mode. The existing backpulsing models are based on lab-scale filtrations under constant-pressure mode and predict the global average flux under different backpulsing conditions. The so-called global average flux is calculated by the total fluid collected through filtration minus the total fluid consumed by backpulsing over the entire period of experiment per unit membrane surface area [35]. In the modelling, it is assumed that the flux behavior between pulses is the same during the entire period, thus the global average flux is predicted from the net permeate flux J during one cycle of forward filtration and backpulsing [71]:

$$J = \frac{\int_0^{t_f} J_f(t)dt - \int_{t_f}^{t_f+t_b} J_b(t)dt}{t_f + t_b} \tag{7}$$

where $J_f(t)$ and $J_b(t)$ are the forward flux and the reverse flux of respective forward filtration duration t_f and backpulsing duration t_b. Note that in the existing backpulsing models, J is dependent variable and t_f is independent variable. T_f is related to the basic backpulsing parameters as $t_f = 1/f - t_b$, where f is the backpulsing frequency.

5.1. Analytical models

5.1.1. Theory

For a fixed backpulsing duration, an optimum forward filtration time is expected which maximizes the net permeate flux. In order to predict net permeate fluxes at different forward filtration times and find the maximum net flux, six analytical models have been developed based on Darcy's law [66] and Blake-Kozeny equation (for cake resistance prediction) [120]. Theoretical forward and reverse fluxes over time for the six different models are shown in Fig. 8. Each model is illustrated by two cycles of forward filtration and backpulsing. The six backpulsing models are further described below.

5.1.1.1. Model I. In Model I [35], it is assumed that membrane fouling is instantly and completely removed by backpulsing, thus the permeate flux after a backpulse immediately reaches the clean membrane flux J_0. The forward flux is assumed to decline due to cake formation and can be approximated by the dead-end filtration equation [120]:

$$J_f(t) = J_0/(1 + t/\tau_1)^{1/2} \tag{8}$$

where τ_1 is the time constant for permeate flux decline due to cake formation and t is the elapsed time since the backpulse. Besides the dead-end filtration approximation, Cakl et al. proposes a semi-empirical equation to describe the forward flux [121]:

$$J_f(t) = \frac{J_0 - J_s}{(1 + t/\tau_1)^n} + J_s \tag{9}$$

where n and τ_1 are regression constants, J_s is the steady-state flux. For $J_s = 0$ and $n = 0.5$, this model changes to the model of dead-end filtration.

Time constant τ_1 is given by Murkes and Carlson [122] according to standard cake filtration theory:

$$\tau_1 = \frac{(C_C - C_B)\Delta P_f}{2\gamma\mu C_B J_0^2} \tag{10}$$

where C_B and C_C are the solute concentration in the bulk and in the cake, respectively, ΔP_f is the forward TMP, γ is the specific cake resistance per unit depth. τ_1 Can be experimentally determined by plotting $(J_0/J)^2$–1 versus t during the flux decay in conventional cross-flow filtration without backpulsing [123].

During backpulsing period, it is simplified that there is only membrane resistance. Thus, $J_b(t)$ is equal to the flux through the cleaning membrane:

$$J_b(t) = \frac{\Delta P_b}{\mu R_m} = \frac{\Delta P_b}{\Delta P_f} J_0 = \alpha J_0 \tag{11}$$

where ΔP_b is the backpulsing amplitude, R_m is the clean membrane resistance, α is the ratio of the reverse and forward TMP, $\Delta P_b/\Delta P_f$.

Substituting Eqs. (8) and (11) into Eq. (7), the corresponding net permeate flux is easily derived by integration:

$$J = J_0 \frac{2\tau_1[(1 + t_f/\tau_1)^{1/2} - 1] - \alpha t_b}{t_f + t_b} \tag{12}$$

5.1.1.2. Model II. Model II [124] assumes that the cake removal during backpulsing is non-instant.

The forward flux follows Eq. (8). For the reverse flux, it is assumed that the cake removal is delayed similar to the forward filtration. Hence, the reverse flux is given by:

$$J_b(t) = \alpha J_0[1 - (1 + t/\tau_2)^{-1/2}] \tag{13}$$

where τ_2 is the time constant for reverse flux increase due to cake removal. Then the net flux is

$$J = J_0 \frac{2\tau_1[(1 + t_f/\tau_1)^{1/2} - 1] - \alpha t_b + 2\alpha\tau_2[(1 + (t_f + t_b)/\tau_2)^{1/2} - (1 + t_f/\tau_2)^{1/2}]}{t_f + t_b} \tag{14}$$

Note that there is a minor discrepancy between Eq. (14) and the equation in Ref. [124]. The equation in Ref. [124] was missing a coefficient τ_2, which is corrected in this review.

5.1.1.3. Model III. Model III [71] considers a delay of cake formation in the beginning of each filtration cycle, which is due to small time (t_{crit}^f) for the backpulsed clean liquid to be refiltered and for the subsequent

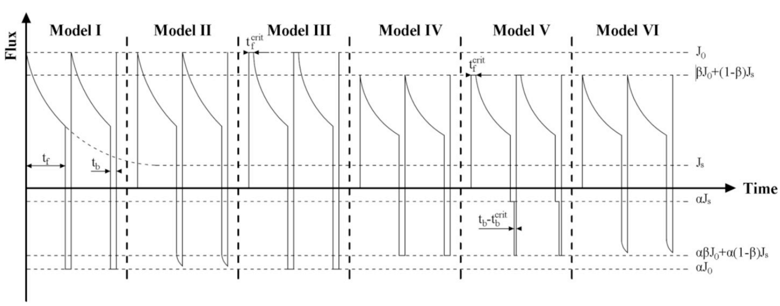

Fig. 8. Theoretical forward and reverse fluxes for six analytical models. J_0 is clean membrane flux, J_s is steady-state flux, $\alpha = \Delta P_b/\Delta P_f$, β is cleaning efficiency, t_f is forward filtration duration, t_b is backpulsing duration, t_{crit}^f and t_{crit}^b are the time delay during forward filtration and backpulsing, respectively. Adapted from Ref. [33].

development of a gel or cake layer that reduces the flux. The time delay cannot be determined directly from experiments. Redkar et al. [71] calculated t^{f}_{crit} by solving the convection-diffusion equations for the concentration polarization and depolarization during the cycles of forward filtration and backpulsing:

$$t_f^{crit} = D(C_C - C_B)/(J_0^2 C_B) \tag{15}$$

It is further explored that t^{f}_{crit} increases with increasing t_b, increasing α, increasing shear rate for short t_b, and decreasing solute concentration in the bulk. T^{f}_{crit} is not known a priori and is expected to vary with t_b [71].

The forward flux including the delay time in the beginning of forward filtration is

$$\begin{cases} J_f(t) = J_0 & when\ t \le t_f^{crit} \\ J_f(t) = J_0/(1 + (t - t_f^{crit})/\tau_1)^{1/2} & when\ t > t_f^{crit} \end{cases} \tag{16}$$

The reverse flux follows Eq. (11) in model I. Then the net flux is modified from Eq. (7) to

$$\begin{cases} J = J_0 \dfrac{t_f - \alpha t_b}{t_f + t_b} & when\ t_f \le t_f^{crit} \\ J = J_0 \dfrac{t_f^{crit} + 2\tau_1 \left[\left(1 + \left(t_f - t_f^{crit} \right) / \tau_1 \right)^{1/2} - 1 \right] - \alpha t_b}{t_f + t_b} & when\ t_f > t_f^{crit} \end{cases} \tag{17}$$

5.1.1.4. Model IV. In Model IV [20], irreversible fouling and non-complete cleaning of membrane during backpulsing is taken into account. The parameter β is defined as cleaning efficiency, the ratio between cleaned membrane surface area and total filtration surface area. It is assumed that only the cleaned portion of membrane has the clean membrane resistance, through which the flux is J_0. Whereas the uncleaned membrane parts have the long-term resistance for filtration without backpulsing, through which the flux is J_s.

The forward filtration flux and the reverse backpulsing flux are composed of fluxes through both the cleaned and fouled fractions of membrane:

$$J_f(t) = \beta J_0/(1 + t/\tau_1)^{1/2} + (1 - \beta) J_s \tag{18}$$

$$J_b(t) = \alpha \beta J_0 + \alpha (1 - \beta) J_s \tag{19}$$

Substituting Eq. (18) and Eq. (19) into Eq. (7), the corresponding net flux is:

$$J = \beta J_0 \frac{2\tau_1 [(1 + t_f/\tau_1)^{1/2} - 1] - \alpha t_b}{t_f + t_b} + (1 - \beta) J_s \frac{(t_f - \alpha t_b)}{t_f + t_b} \tag{20}$$

5.1.1.5. Model V. The assumptions in Model V [20] are more comprehensive and more complicated. It is not only assumed that the fouling is non-completely cleaned by backpulsing as in Model IV, but also assumed that there is a delay in both cake formation at the beginning of forward filtration (t^{f}_{crit}) and cake removal at the start of backpulsing (t^{b}_{crit}).

The equations for the forward filtration flux are

$$\begin{cases} J_f(t) = \beta J_0 + (1 - \beta) J_s & when\ t \le t_f^{crit} \\ J_f(t) = \beta J_0/(1 + (t - t_f^{crit})/\tau_1)^{1/2} + (1 - \beta) J_s & when\ t > t_f^{crit} \end{cases} \tag{21}$$

The equations for the reverse backpulsing flux are:

$$\begin{cases} J_b(t) = \alpha J_s & when\ t \le t_f + t_b^{crit} \\ J_b(t) = \alpha \beta J_0 + \alpha (1 - \beta) J_s & when\ t > t_f + t_b^{crit} \end{cases} \tag{22}$$

It is of course required that $t_b \ge t^{b}_{crit}$ if backpulsing is to be effective in fouling removal. Considering these aspects, Eq. (7) yields:

$$\begin{cases} J = [\beta J_0 + (1 - \beta) J_s] \dfrac{(t_f - \alpha t_b)}{t_f + t_b} \\ when\ t_b \ge t_b^{crit},\ t_f \le t_f^{crit} \\ J = \beta J_0 \dfrac{t_f^{crit} + 2\tau_1 \left[\left(1 + \left(t_f - t_f^{crit} \right) / \tau_1 \right)^{1/2} - 1 \right] - \alpha (t_b - t_b^{crit})}{t_f + t_b} + (1 - \beta) J_s \dfrac{(t_f - \alpha t_b)}{t_f + t_b} - \beta J_s \\ \dfrac{\alpha t_b^{crit}}{t_f + t_b} \\ when\ t_b \ge t_b^{crit},\ t_f > t_f^{crit} \end{cases} \tag{23}$$

5.1.1.6. Model VI. Model VI [18] considers that the partial cake removal during backpulsing is not instantaneous and assumes that the cleaning efficiency versus time during backpulsing can be described by a simple exponential rise:

$$\beta(t) = \beta_{\max} \left[1 - e^{-(t - t_f)/\tau_2} \right] \tag{24}$$

where β_{max} is the maximum possible fraction of the membrane that can be cleaned physically. At the end of backpulsing duration t_b, the fraction of cleaned membrane is

$$\beta_b = \beta_{\max} \left(1 - e^{-t_b/\tau_2} \right) \tag{25}$$

The forward filtration flux is similar as that in Model IV, but using β_b instead of β:

$$J_f(t) = \beta_b J_0/(1 + t/\tau_1)^{1/2} + (1 - \beta_b) J_s \tag{26}$$

For easy illustration, β_b is still shown as β in Fig. 8. The reverse flux during backpulsing is:

$$J_b(t) = -\alpha \{ \beta(t) J_0 + [\beta_b - \beta(t)] J_0/(1 + t_f/\tau_1)^{1/2} \} - \alpha (1 - \beta_b) J_s \tag{27}$$

The net flux is:

$$J = \left(\begin{array}{l} J_0 \left\{ \begin{array}{l} 2\tau_1 \beta_{\max} (1 - e^{-t_b/\tau_2}) [(1 + t_f/\tau_1)^{1/2} - 1] \\ -\alpha \beta_{\max} \left[t_b - \tau_2 (1 - e^{-t_b/\tau_2}) + \frac{\tau_2 - (\tau_2 + t_b) e^{-t_b/\tau_2}}{(1 + t_f/\tau_1)^{1/2}} \right] \end{array} \right\} \\ + J_s [1 - \beta_{\max} (1 - e^{-t_b/\tau_2})] (t_f - \alpha t_b) \end{array} \right) \div (t_f + t_b) \tag{28}$$

5.1.2. Results

There are two kinds of applications of these models for membrane filtrations with backpulsing. One is to predict net permeate flux and the optimum forward filtration time. The other is to understand the processes involved in fouling formation during forward filtration and fouling removal during backpulsing.

The six models were applied to 21 experiments from different literature [16,19,20,33,35,71,125]. The parameters used in the six models are listed in Table 3. This table includes cleaning duration $t_b \le 2$ s. It is because the review on backwashing published in 2017 [28] includes the washing duration from 2 to 600 s. The common parameters in the six models are J_0, J_s, τ_1, α and t_b. Among them, α and t_b were controlled by the experimental setup. J_0, J_s, and τ_1 were measured by experiments. The critical times t^{f}_{crit} and t^{b}_{crit} cannot be determined experimentally and they were therefore adjusted. Except in experiment No. 7–9 where t^{f}_{crit} were calculated by Redkar et al. [71] using Eq. (15), t^{f}_{crit} in other experiments were taken equal to t_b because t^{f}_{crit} mainly varies with t_b [20,33,71]. T^{b}_{crit} was simplified to be equal to 0 for all the experiments. Because t^{b}_{crit} should be very short and showed to have little influence on the net flux in experiments of Heran and Elmaleh [33]. Special parameters, like τ_2 in Model II, β in Model IV and Model V, as well as τ_2 and β_{max} in Model VI were difficult to obtain from experiments. They were fitted by minimizing the sum of the squared errors between the measured and predicted fluxes [18,33,124]. The range for τ_2 to be fitted is $[0.01\tau_1, 3\tau_1]$ [18] with a step of $0.01\tau_1$.

Table 3
A list of parameters in the six backpulsing models and the prediction of Model V.

Gr.	No.	Feed solutions	Measured/simplified paremeters						Fitting parameters						Experiments		Prediction of Mod.V		$J_{max,\ Mod.V}/J_s$	Ref.
			All six models						Mod.II	Mod.IV	Mod.V	Mod.VI								
			J_0 (LMH)	J_s (LMH)	τ_1 (s)	α	t_b (s)	t^f_{crit} (s)	τ_2 (s)	β	β	τ_2 (s)	β_{max}	β_b	J_{max} (LMH)	t^{opt}_f (s)	J_{max} (LMH)	t^{opt}_f (s)		
A	1	Yeast susp. (3% vol.)	6300	101	0.67	0.5	2	2	2.010	1	1	0.482	1	0.98	2772	7	2524	5.5	25	[35]
	2	Yeast susp. (0.3% vol.)	6300	137	6.9	1	2	2	1.311	1	1	0.345	1	1	4176	9/20	3578	16.5	26	[35]
	3	Yeast susp. (3% vol.)	6300	101	0.67	1	2	2	2.010	1	1	0.690	1	0.94	2664	7	1829	9.3	18	[35]
	4	Yeast susp. (6% vol.)	6300	72	0.32	1	2	2	0.960	1	1	0.960	1	0.88	1584	9	1390	8.3	19	[35]
	5	Yeast susp. (3% vol.)	6300	101	0.67	1	1	1	2.010	1	1	0.241	1	0.98	3672	5	2311	5.3	23	[35]
	6	Yeast susp. (3% vol.)	6300	101	0.67	1	0.5	0.5	1.601	1	1	0.107	1	0.99	4284	3	2837	3.1	28	[35]
	7	Yeast susp. (2.6 g/L)	792	94	4.1	1	0.3	1	0.041	1	0.93	0.041	1	1	690	5	569	3.9	6	[71]
	8	Yeast susp. (2.6 g/L)	792	94	4.1	1	0.2	0.67	0.041	1	1	0.041	1	0.99	684	3	630	3.1	7	[71]
B1	9	Yeast susp. (2.6 g/L)	792	94	4.1	1	0.1	0.29	0.041	0.8	0.77	0.041	0.87	0.79	655	1.5	529	2.1	6	[71]
	10	Latex susp. (0.1 g/L)	780	140	117.7	0.25	0.2	0.2	1.177	0.63	0.63	1.177	1	0.16	544	4	522	11.8	4	[16]
	11	Clay susp. (0.04 g/L)	1950	270	380	1	0.5	0.5	3.800	0.94	0.94	3.800	1	0.12	2020	10	1760	40.7	7	[19]
	12	Clay susp. (0.2 g/L)	1950	125	82	1	0.5	0.5	0.820	0.61	0.61	0.820	1	0.46	1300	50/20	1120	20.1	9	[19]
	13	Clay susp. (2.0 g/L)	1950	89	9	1	0.5	0.5	0.090	0.16	0.16	0.090	0.16	0.16	410	10	299	8.8	3	[19]
	14	Clay susp. (2.0 g/L)	2070	200	10	1	0.5	0.5	0.100	0.18	0.17	0.100	0.18	0.18	610	10	418	10.3	2	[19]
	15	Clay susp. (0.2 g/L)	1950	125	82	1	0.2	0.2	0.820	0.52	0.52	0.820	1	0.22	1134	5	1008	12.4	8	[19]
	16	Bentonite susp. (1 g/L)	1800	154	3.85	1	0.05	0.05	0.039	0.44	0.44	0.039	0.6	0.43	850	2–5	763	1.5	5	[33]
	17	E.coli susp. (0.14 g/L)	1100	70	14.3	0.6	0.2	0.2	0.143	0.17	0.16	0.143	0.22	0.17	250	5	209	5.6	3	[125]
	18	E.coli susp. (1.2 g/L)	4680	12	0.039	1	0.1	0.1	0.0004	0.07	0.05	0.003	0.07	0.07	139	0.2	79	0.5	7	[20]
	19	E.coli susp. (10 g/L)	4680	9	0.005	1	0.1	0.1	0.015	0.01	0.05	0.015	0.04	0.04	68	0.5	36	0.4	4	[20]
B2	20	E.coli susp. (1.2 g/L)	4680	12	0.039	1	1	1	0.002	0.19	0.03	0.0004	0.19	0.19	49	40	25	5.5	2	[20]
	21	Yeast susp. (3% vol.)	6300	101	0.67	0	2	2	0.007	0.67	0.46	0.429	0.68	0.67	1620	9	1652	3.5	16	[35]

Note: *E.coli*, *Escherichia coli*; susp., suspensions; Gr., group; No., number; Mod., Model; Ref., references; LMH, liter per square meter per hour.

The range for β is [0, 1] with a step of 0.01. β_b in Model VI was calculated by τ_2 and β_{max} using Eq. (25). The 21 experiments are separated into two groups by the value of β. In Group A, β in Model IV/V is equal or close to 1, which means that the fouling during filtration is reversible and that the original flux can be obtained after each backpulse. In Group B, $0 < \beta < 1$, which means that the fouling is not completely removed by backpulsing. Group B is divided into two sub-groups by backpulsing duration t_b: Group B1 ($t_b < 1$ s) and Group B2 ($t_b \geq 1$ s).

J_0 is the clean membrane flux, J_s is the steady-state flux (long-term flux without backpulsing), τ_1 is time constant for cake formation, $\alpha = \Delta P_b/\Delta P_f$, t_b is backpulsing duration, t^f_{crit} is the time delay during forward filtration, τ_2 is time constant for cake removal, β is the cleaning efficiency, β_{max} is the maximum cleaning efficiency, β_b is the cleaning efficiency at the end of one backpulse, J_{max} is the maximum net flux, t^{opt}_f is the corresponding optimum forward filtration time for J_{max}.

α, t_b, J_0, J_s, and τ_1 were from experiments. T^f_{crit} was simplified to be equal to t_b, except in No. 7–9 where t^f_{crit} was calculated by Eq. (15). T^b_{crit} was taken equal to 0 for all the experiments. τ_2, β and β_{max} were fitted parameters. β_b was calculated by τ_2 and β_{max} using Eq. (25) in Model VI.

Normalized net flux J/J_0 can be used to compare results from different experiments by using the same scale. The normalized experimental fluxes from 21 experiments [16,19,20,33,35,71,125] versus the normalized net fluxes predicted by Model I − VI are shown in Fig. 9, respectively. The models are calculated in MATLAB. The legends are from the numbering and grouping in Table 3.

Fig. 9 compares the accuracy of each model on predicting the net permeate fluxes. Diagonal lines are added to the plots as reference lines. The closer the data points are to the reference lines, the more accurate the model predictions are. The difference between the modelling- and the experimental results can also be indicated by the mean absolute deviation (MAD):

$$MAD = \frac{1}{n}\sum_{i=1}^{n}\left|\frac{J_{model,\,i} - J_{exp.,\,i}}{J_{exp.,\,i}}\right| \tag{29}$$

where n is the number of data points. The smaller the MAD is, the closer the modelling results are to the experimental results, and the more accurate the model is. The values of MAD from the six models are summarized in Table 4. Note that the MAD values are only comparable within the same row, which numerically demonstrates the different prediction between models.

In Group B2, MAD value for Model IV is 0.1680, MAD value for Model VI is 0.1678.

As is shown in Fig. 9 and Table 4, in Group A (β → 1) Model V is the same as Model III and shows the best prediction, which indicates that when fouling is completely reversible, t^f_{crit} is significant to take into consideration. In Group B ($0 < \beta < 1$) where both reversible and irreversible fouling are present, the predictions of Model IV − VI are pretty similar and are much better than Model I − III, which illustrates that it is important to include β in the modelling. In general, for all the 21 experiments Model V considering both t^f_{crit} and β works the best according to Table 4 and Fig. 9. The predictions of maximum net fluxes (J_{max}) and the corresponding optimal forward filtration times (t^{opt}_f) by Model V are listed in Table 3. They are in good agreement with experimental results. The predicted net fluxes obtained with backpulsing were 2–28 times the long-term fluxes in the absence of backpulsing (See Table 3 column $J_{max,\ Mod\ V}/J_s$).

As shown in Fig. 9, Model I underestimates most of the net fluxes in Group A, as the red points are mostly under the diagonal line; while it overestimates almost all the fluxes in Group B, as almost all the blue and green points are above the reference line. Model II predicts similar results as Model I does in general (shown in both in Fig. 9 and Table 4), which is reasonable because Model II only makes a slightly change in the reverse flow during backpulsing (see Fig. 8).

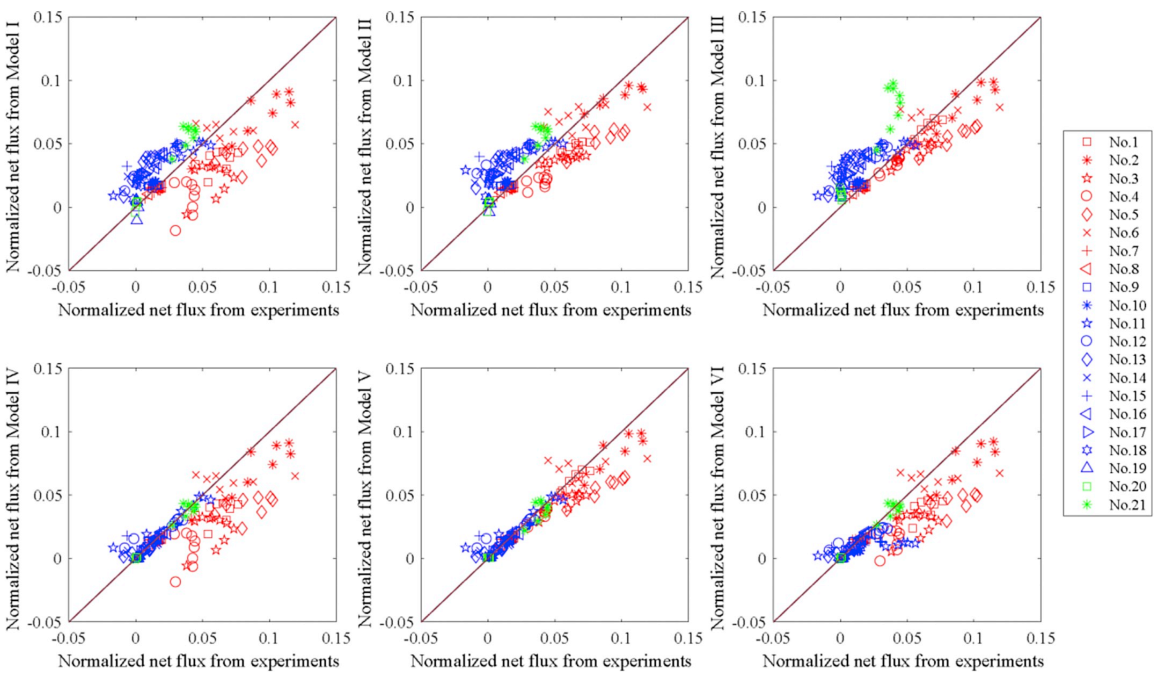

Fig. 9. Normalized net flux from experiments versus that calculated from Model I − VI, respectively. Data are collected from Refs. [16,19,20,33,35,71,125] and analyzed in MATLAB. The legends use the experiment numbering in Table 3. Data points of Group A are in red, Group B1 in blue, and Group B2 in green.

Table 4
A list of MAD values from the six models applied in different groups of data points.

	No. Of data points	Model I	Model II	Model III	Model IV	Model V	Model VI
Group A	67	0.402	0.272	**0.179**	0.402	**0.179**	0.354
Group B	91	3.820	4.013	5.909	**0.601**	0.633	0.625
Group B1	75	4.187	4.421	5.529	0.693	**0.673**	0.723
Group B2	16	2.099	2.102	7.689	0.168	0.445	**0.168**
All 21 exp.	158	2.371	2.427	3.479	0.517	**0.441**	0.510

Note: The best predictions in each row are marked in bold.

Model III improves the prediction of the fluxes in Group A remarkably compared to Model I, but overestimates the fluxes in Group B (especially Group B2) even more than Model I does (see Fig. 9). This is because compared with Model I, Model III includes a time delay of cake formation in the beginning of forward filtration (see Fig. 8), which makes the forward flux and the net flux even larger than those predicted by Model I.

Model IV is a big improvement to Model I as it involves cleaning efficiency β in the modelling. It is the same as Model I when $\beta = 1$. Therefore in Table 4 Model IV shows the same MAD value as Model I for Group A. However, when $0 < \beta < 1$, Model IV improves the prediction of net fluxes dramatically. All the blue and green points in Fig. 9 are pretty close to the reference line.

Model V considering both t_{crit}^{f} and β is the best model, since all the data points locate near the reference line in Fig. 9. However, it is doesn't show the best prediction for all the experiments, especially data in Group B2 where $t_b \geq 1$ s (see Table 3). It is because of the error made by the simplification that t_{crit}^{f} is equal to t_b. When β is very low in case No. 20 or $\alpha = 0$ in case No. 21, very little cake removal occurs during backpulsing, thus a smaller value of t_f^{opt} is needed to rebuild the cake layer during forward filtration [71].

Model VI is theoretically more correct than Model IV as β is considered to be varied with elapsed time and the cake removal during backpulsing is not instant. However, this refinement does not improve Model IV much. In some cases (No. 10, 11 and 15) Model VI predicts worse results than Model IV does (detailed results are not shown here), which is because τ_2 is not chosen correctly due to the limited range of the fitting parameter τ_2.

In general, Model IV − VI improves Model I − III dramatically by taking β into consideration. Model IV and Model VI are slight worse than Model V on the prediction of net fluxes, which indicates that t_{crit}^{f} is significant to be included besides β. However, the accuracy of Model V highly depends on the selection of a correct value of t_{crit}^{f}. The arbitrary simplification of $t_{crit}^{f} = t_b$ is not accurate enough. A better calculation of t_{crit}^{f} could refer to literature [71].

Heran and Elmaleh [33] proposed experimental methods to measure transient fluxes and calculate cleaning efficiency β for the first time. Experiments were run by using bentonite in tap water (1 g/L) as a suspension of inorganic and non-compressive particles and under conditions of $\Delta P_f = \Delta P_b = 1$ bar and $t_b = 0.05$ s. Model I, III, IV and V were compared to predict the net flux and the optimum forward filtration time by not using any fitting parameters, shown in Fig. 10. The theoretical predictions of Model I and III are overlapped. So as Model IV and V. This indicates that the time delay parameters in Model III and V had little influence on the net flux, which is reasonable because t_{crit}^{f} in model III and V is simplified to be equal to t_b that is very short (only 0.05 s) and t_{crit}^{b} in model V is simplified to be 0. Model I and III largely overestimate the experimental net fluxes. The reason is probably that the backpulsing duration was very short or there was irreversible fouling, resulting in a fouling layer that was not completely removed by each back pulse, which is against the main assumption of Model I and III. Model IV and V underestimate the experimental results, but fit the experimental data much better than Model I and III do, which confirms the incomplete cleaning by backpulsing. The error between experimental results and the modelling results from Model IV and V is mainly due to the error of finding the correct β. β Measured by experiments is 0.35, while the best fitted β for both Model IV and V is 0.44, as is shown in Table 3 (experiment 16). In addition, all the four models predicted the same optimum forward filtration time of 2 s, which agrees with the experimental value of 2–5 s. If only the optimum forward filtration time is to be predicted, all the four models may be satisfactory. If both the maximum net flux and the corresponding optimum forward filtration time are to be predicted, Model IV and V are better choses than Model I and III.

The other application of these models is to investigate and compare membrane fouling in different filtration processes or in different stages of a certain filtration process. Analyzing critical parameters provides insight into the mechanism of backpulsing and fouling behavior.

Time constant τ_1 indicates the situation of cake formation during the forward filtration. In the same membrane filtration system, the smaller τ_1 is, the smaller the flux is and the faster the flux declines, which implies faster cake formation/more severe fouling. As is shown in experiments No. 2–4, No. 11–13 and No. 17–19 in Table 3, τ_1 is decreasing with increasing feed concentrations. τ_2 Is the time constant during reverse filtration and reflects the situation of cake removal. However, there is no obvious tendency of τ_2 with different feed concentrations in the same membrane system, which shows that τ_2 has a weak function of feed concentration.

In Model IV and V, fitting parameter β describes an average sense of cleaning efficiency by backpulsing. In Kuberkar et al. [20], backpulsing experiments were run at the duration of 1.0 s and 0.1 s, respectively, in the filtration of a washed *Escherichia coli (E. coli)* suspension with a dry cell weight of 1.2 g/L. The best-fit value of β was 0.19 for t_f between 5 and 80 s when backpulsing duration was 1.0 s. While the best-fit β was 0.075 for $t_f < 1$ s and 0.028 for $t_f > 1$ s with a backpulsing duration of 0.1 s. It is obvious that backpulses with a very short duration (0.1 s) was inefficient to remove membrane fouling. Besides, the longer the forward filtration time was, the lower the cleaning efficiency of backpulsing became.

In Model VI, fitting parameter β_{max} shows the maximum cleaning efficiency by backpulsing. Mores et al. [18] used Model VI to analyze the results from the filtration of washed *Escherichia coli* (*E.coli*) suspensions. Both single backpulsing experiments and standard backpulsing experiments were tested. The so-called single backpulsing experiments were designed to remove the pre-deposited cake layer. After filtrating 1 L 0.01 g/L E. *coli* suspension in a dead-end filtration, a single backpulse or more was used until the recovered flux did not increase with further additional backpulses. The so-called standard backpulsing experiments were the normal backpulsing in the crossflow filtration of a 1 g/L E. *coli* suspension. The values of the best-fit model parameters were $\tau_2 = 0.15$ s and $\beta_{max} = 0.48$ in single backpulsing experiments, while $\tau_2 = 0.2$ s and $\beta_{max} = 0.11$ in standard backpulsing experiments.

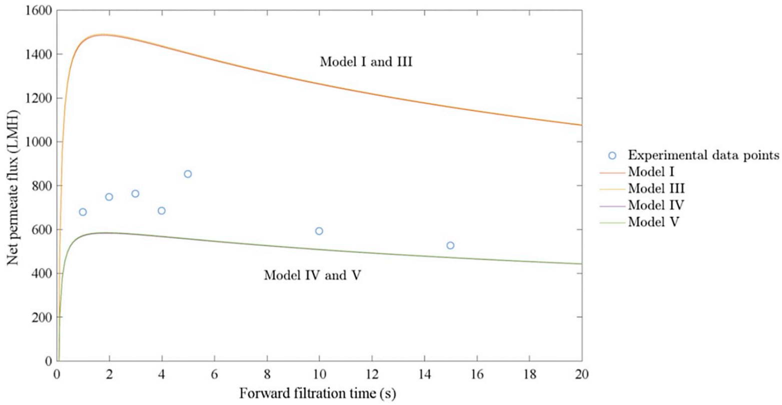

Fig. 10. Net permeate flux vs. Forward filtration time for the filtration of a bentonite suspension under conditions of $\Delta P_f = \Delta P_b = 1$ bar and $t_b = 0.05$ s. Adapted from Ref. [33].

The two τ_2 were similar in the two groups of experiments with different feed concentrations, which is in accordance with the conclusion above. β_{max} was much lower in standard backpulsing experiments indicated that the combination of higher bacteria concentration and the presence of crossflow led to more irreversible fouling.

5.2. Other approaches

Vinther et al. have proposed two simulation models and one semi-analytical model recently for backpulsing in an UF with hollow-fiber membranes under laminar crossflow conditions.

The simulation models are based on computational fluid dynamics (CFD) methods. The first simulation model [126,127] solves the continuity equation Eqs. (1) and (2) with assumptions on solute velocity in the two-dimensional domain near the membrane surface. The second simulation model [128] is a mathematical model based on solving the Navier–Stokes equation Eq. (6) along with the continuity equations, Eqs. (1) and (2), for both the solute and the solvent.

The semi-analytical model [129] is able to estimate optimal backpulsing duration under the hypothesis that the solute pathline from the beginning of the membrane surface should end at the end of the membrane surface after one backpulse and the following forward filtration. The optimal backpulsing duration can be calculated by

$$T_{bs} = \sqrt{\frac{2L_m}{kL_p \Delta P_b (1 + \Delta P_b/\Delta P_f)}} \tag{30}$$

where k is a coefficient represents velocity gradient. The optimal backpulsing frequency and the maximal net flux can also be found base on the streamlines and pathlines during a backpulsing cycle and the forward flux without backpulsing [129].

Although the results from the two simulation models on optimal backpulsing duration/frequency were in good agreement with the estimated results from the semi-analytical model, there has not been any direct experimental results to verify the accuracy of the three models yet.

6. Future outlook

Backpulsing, a physical cleaning method for membranes, is shown to be effective and energy-efficient in many research fields. Although various studies have been carried out to understand backpulsing and make good use of it, a number of issues worth further investigation.

- **More fundamental studies of the effect of feed properties on backpulsing efficiency.** Huang et al. [130] found that the feed water source was more important for irreversible fouling than hydrodynamic conditions (e.g., forward and reverse flux). Besides, varies studies have shown that a low fouling potential does not necessarily result in a high backwashing efficiency [67,131]. The hydrophilicity/hydrophobicity of natural organic matters, molecular weight distribution of the feed, and the change of feed water properties after pretreatments make difference to backwashing efficiency [28]. As is mentioned in section 4.1.1, different types of foulants have different effect on backpulsing efficiency. However, there are only limited studies about it. More research of the effect of feed properties on backpulsing efficiency is needed, which can help understand more about the fundamentals of fouling removal by backpulsing.
- **Further development of modelling.** The existing analytical models could predict optimal operating conditions to some extent. However, the accuracy and applicability need to be improved. First of all, inertial effects should have pronounced influence on the mass transport in the process of filtration with backpulsing, since it is associated with frequent and transient changes of pressure gradients across the membrane [132]. The inertial resistance coefficient in Eq. (4) could be determined empirically by estimation from membrane parameters (e.g., permeability and porosity) [70,133]. Secondly, the existing models are based on constant-pressure mode because it is easy to operate in lab-scale experiments. However, for most industrial applications, constant-flux is what the customers prefer. New models based on constant-flux mode are of great interest.
- **Membrane ageing and damage caused by backpulsing.** The replacement of membranes accounts for 25–40% of the total cost in a membrane plant [134]. Membranes' lifetime depends on the degree of membrane ageing and damage. Pressure differentials during backpulsing induce a degradation of membrane material, especially for polymeric membranes. However, literature relating to the fatigue behavior under mechanical stresses is very limited [107].
- **More long-term and large-scale experience.** The experience of backpulsing available from literature nowadays is mainly short-term and lab-scale experience. However, backpulsing is operating in a much higher frequency than backwashing, which increases the failure risk of an operating system, such as failure of valves. Besides,

surface properties of membranes can be changed during long-term filtration because of the irreversible membrane fouling [135]. Therefore, although it is shown both experimentally and theoretically that backpulsing is an efficient way to mitigate membrane fouling, long-term and large-scale experience from practical applications is necessary for a wider application of backpulsing technology.

- **Combination of backpulsing and backwashing.** Fraga et al. [78] supposed that backpulsing was mainly to loosen and detach the trapped foulants via inertia excitation while backwashing was to provide constant shear and drag force and wash foulants away from the membrane surface and membrane pores. Backpulsing and backwashing can be combined to obtain a synergistic effect on membrane cleaning [78,136]. In addition, when the membrane surface is frequently exposed to backpulsing, more internal fouling is resulted [16,125]. This is because backpulsing removed the cake layer or gel layer on the membrane surface which often acts as a secondary dynamic membrane [137,138]. The combination of backpulsing and chemically enhanced backwashing is therefore envisioned to be a promising solution.

7. Conclusion

This paper presents a comprehensive review of backpulsing technology applied in low-pressure membrane filtrations for the purpose of mitigating membrane fouling. Fundamentals of backpulsing (i.e., definition, classification, setup and theory) are illustrated. Backpulsing has been used in all kinds of membrane configurations: flat sheet, tubular, hollow fiber and spiral wound. Research of backpulsing also covers many industrial fields: water and wastewater treatment, food industry, biotechnology application and other industries.

Although experiments demonstrate that backpulsing is efficient in reducing concentration polarization and mitigating membrane fouling, the cleaning efficiency of backpulsing depends on several factors:

- Feed properties. The types of foulants in the feed solution directly determine the types of membrane fouling. Backpulsing is more efficient to remove external and non-adhesive fouling. The more concentration of foulants, the more severe membrane fouling and the less cleaning efficiency of backpulsing.
- Membrane properties. Because of the elasticity of polymeric membranes, the cleaning capability of backpulsing could result from the membrane vibration caused by rapid reverses of TMP instead of the reverse flow. Modification of membrane surfaces can alter the types of fouling formation, making backpulsing more effective. Moreover, membrane pore sizes are correlated to the types of membrane fouling and backpulsing efficiency.
- Operating parameters. It is important to carry out backpulsing at the optimum conditions, such as amplitude, duration and frequency. It is also beneficial to study the three backpulsing parameters together as a backpulsing strategy. Besides, conventional filtration parameters, TMP/flux and CFV, have also influence on backpulsing efficiency.

Modelling is developed to predict the optimum conditions of backpulsing and explain the fouling situation during the process of backpulsing. Six analytical models have been developed based on cake layer formation assumption. They are qualitatively precise to predict the optimum backpulsing interval/frequency. By analyzing critical parameters, such as τ_1 and β, membrane performance under different backpulsing conditions or in different membrane filtrations can be evaluated. Three alternative approaches have emerged recently. However, there is no direct experimental verification of them yet.

Finally, in order to further improve the implementation of backpulsing in commercial applications, several aspects on backpulsing need more work: further development of modelling, combination of backpulsing and backwashing, membrane ageing and damage caused by backpulsing, as well as more long-term and large-scale experience.

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Paper II

A multivariate study of backpulsing for membrane fouling mitigation in produced water treatment

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Research paper

A multivariate study of backpulsing for membrane fouling mitigation in produced water treatment

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ABSTRACT

Produced water contains a large amount of oil droplets and suspended solids that need to be removed before discharge. Research has been focused on membrane technology because it can effectively remove dispersed oil and provide stable effluent of high quality. To mitigate severe membrane fouling in the microfiltration of synthetic produced water, backpulsing with different conditions was investigated in this study. Results showed that backpulsing was efficient to mitigate membrane fouling. However, the cleaning efficiency varied between different backpulsing conditions. The effect of backpulsing parameters (amplitude, duration and frequency) and their interactions on membrane performance were studied by a 2^3 full factorial design. Final specific flux and net permeate yield were chosen as responses to indicate the situation of membrane fouling and membrane productivity, respectively. Besides, mechanisms of backpulsing on fouling mitigation and membrane productivity were investigated. Within the selected levels all the three backpulsing parameters were important for the two responses. Amplitude was the most crucial variable for fouling removal and final specific flux, while frequency was the most significant one for membrane net yield. The effect of interactions on both responses were significant but performed in a different way. Amplitude and duration showed a trade-off interaction effect on final specific flux, whereas the three parameters showed synergistic interaction effect between each other on net yield.

1. Introduction

Produced water is the largest byproduct in the production of oil and gas. It typically consists of dispersed oil, dissolved organic compounds, inorganic compounds, production chemicals, solids and heavy metals [1,2]. It is the most important source of oil discharged to the Norwegian Continental Shelf (NCS) [3]. The current regulation for oil content discharged in the North Sea Region is less than 30 mg/L [3]. In order to further protect the environment on the NCS, Norwegian authorities established a 'Zero Discharge' target for the petroleum activities in 1997 [4]. The main rule is that no environmentally harmful substances may be released, neither added chemicals nor naturally occurring chemical substances. Compared to conventional treatment technologies, such as gravity separation, air flotation and hydrocyclones, membrane filtration (microfiltration and ultrafiltration) has several advantages [5]. For instance, no extraneous chemicals are needed, has a smaller foot print, can remove the most stable dispersed oil droplets (<10 μm) in water [6] and maintain a uniform permeate regardless of influent fluctuation [7,8]. However, membrane fouling is the largest obstacle to membrane application, which leads to a low permeate flux and increased operating cost [9]. Blocking filtration laws, consist of four fouling mechanisms: complete blocking, standard blocking, intermediate blocking and cake layer formation [10,11], have been widely used in the microfiltration and ultrafiltration of particles and colloids [12]. During produced water treatment, Dickhout et al. [6]. proposed an additional fouling mechanism, which is formation of a continuous oil layer on the membrane surface due to coalescence of oil droplets without the presence of particles.

Backpulsing technology is an in-situ method to mitigate membrane fouling in membrane microfiltration and ultrafiltration. It is induced by periodically reversing the transmembrane pressure (TMP) for a very short duration (typically <1 s [13–15]). Reversible foulants accumulated on the membrane surface or inside the membrane pores are dislodged and flushed away from the membrane, as most of the backpulsing schedules are carried out in conjunction with crossflow filtration. Backpulsing may seem like backwashing with a very short duration.

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However, they are fundamentally different, both with respect to operating conditions, and with respect to the mechanisms of fouling formation and mitigation [16]. Backpulsing has a dynamic and transient effect to remove membrane fouling that is not found in backwashing [15]. It has shown to be an efficient method to mitigate membrane fouling in many applications fields, e.g., wastewater treatment, food industry, biotechnology, and other industries [16]. Many studies also showed that backpulsing was effective in fouling mitigation in membrane filtrations of oily wastewater. In ultrafiltration of an unstable secondary oily wastewater, the steady-state flux with backpulsing was more than twice the steady-state flux without backpulsing [17]. Similar results were also reported in the microfiltration of an oil-in-water emulsion [18] and the treatment of olive mill wastewater by a membrane bioreactor (MBR) system [19,20]. Silalahi and Leiknes [21] found that membrane fouling rates were significantly reduced and the effect of feed water characteristics on the permeate flux was also reduced when backpulsing was applied in the microfiltration of produced water.

The efficiency of backpulsing is a function of multiple parameters, especially the three basic backpulsing parameters, amplitude, duration and frequency [16]. Many studies agreed that frequency was an important parameter for the optimization of permeate flux [15,22,23] and there existed an optimal frequency for a fixed duration [24,25]. If the frequency was too low and cake or gel layer had formed and consolidated, the membrane would not be efficiently cleaned. If the frequency was too high, although membrane could be well cleaned, excessive water might be lost unnecessarily. Redkar et al. [26] and Kuberkar et al. [27] found that shorter backpulsing durations yielded higher net permeate flux. However, experiments of Sondhi et al. [14] showed that both frequency and duration did not show a significant effect on permeate flux. Instead, backpulsing amplitude was found to be the most important parameter and increasing amplitude led to a better membrane cleaning. Moreover, there might be a relationship between the parameters of backpulsing. In ultrafiltration of binary protein mixtures, Wilharm and Rodgers found that permeate fluxes and membrane resistances under backpulsing with high amplitude and short duration were similar to those provided by backpulsing with low amplitude and long duration. However, there is no systematic study on the three backpulsing parameters and their interactions.

In this study, backpulsing technology was applied to mitigate membrane fouling in the filtration of synthetic produced water. A 2^3 full factorial design was used to study the significance of the three backpulsing parameters, amplitude (0.1–1.0 bar), duration (0.1–0.5 s) and frequency (0.0167–0.05 Hz), and their interactions on the responses of final specific flux and net permeate yield. Furthermore, the effect of backpulsing on membrane fouling mitigation was investigated and the mechanisms of backpulsing on membrane performance were discussed.

2. Materials and methods

2.1. Apparatus

α-alumina ceramic membranes (ECO-Ceramics, the Netherlands) with an averaged pore size of 0.1 μm were used for filtration experiments, considering the relatively high productivity and low irreversible fouling tendency [21,28]. Membranes were 340 mm in length with an internal diameter of 8 mm and an outer diameter of 11 mm. The selective membrane area was 85 cm^2.

Experiments were performed using a crossflow membrane system, as shown in Fig. 1. The membrane unit had a pair of modules which allowed two parallel experiments under the same experimental

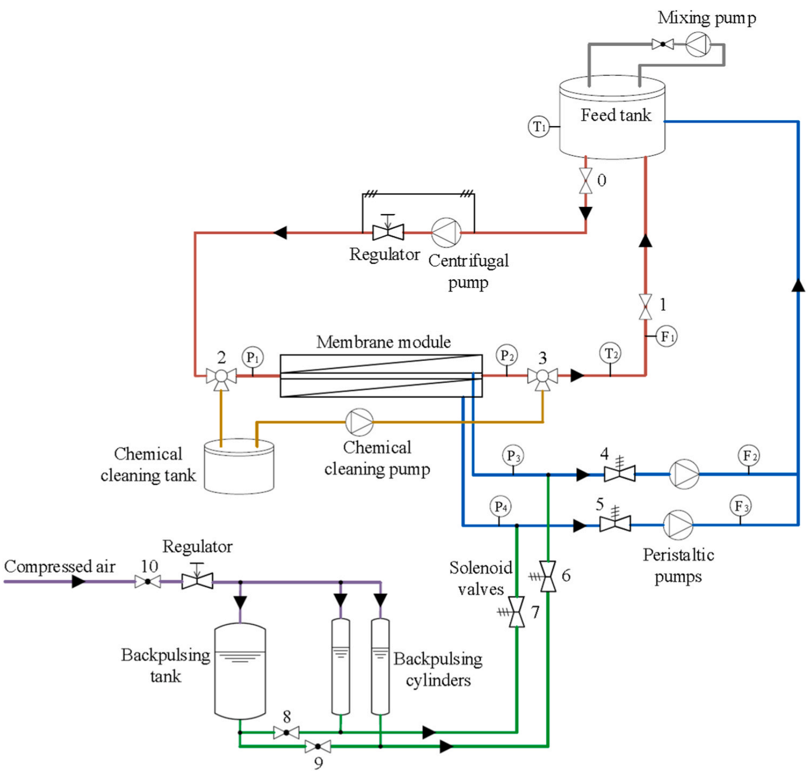

Fig. 1. Schematic flow diagram of the crossflow filtration system.

conditions. The feed tank was a 20 L container with a mixing pump to maintain stable produced water with constant particle size distribution (PSD) and oil concentration during the entire filtration. Crossflow was generated by a centrifugal pump with a pump speed regulator. Pressure in the membrane channel and crossflow velocity (CFV) could be regulated by controlling the feed pump speed and gate valve 1. During forward filtration, solenoid valves 4 and 5 in the permeate lines were open and valves 6 and 7 in the backpulsing lines were closed. While during backpulsing, the solenoid valves 4–7 were switched. Constant permeate flow was controlled by peristaltic pumps (Masterflex 07528-30, Cole-parmer, USA). Compressed air with needed pressures was exerted on clean water tanks. Distilled water was used as backpulsing liquid. In our setup, the volume of each pulse was calculated by the level change in the backpulsing columns that had an inner diameter of 20 mm and a length of 500 mm. Distilled water in the backpulsing columns was replenished by opening valves 8 and 9 from the backpulsing tank (10 L). Online chemical cleaning could be operated by switching three-way valves 2 and 3. Pressures before and after membranes were monitored by pressure transmitters 1–4. Crossflow was measured by flowmeter 1 and permeate flows were monitored by flowmeters 2–3. All data were recorded by a data acquisition unit (Agilent 34970A, USA) at real time. The forward filtration and backpulsing system were controlled by a Python program.

2.2. Preparation and characterization of produced water

The characteristics of real produced water change with time. Synthetic produced water was used in this study because the feed properties could be maintained the same and constant for all experiments. Each batch of produced water was made by mixing crude oil (254 mg/L, detailed information is shown in Table 1), salt (35 g/L NaCl, VWR), surfactant (25 mg/L Tween 80, Sigma-Aldrich) and tap water (2 L) with an Ultra-Turrax S25N-10 G homogenizer (IKA, Germany) at 10 000 rpm for 7 min 16 L (8 batches) synthetic produced water was prepared for each experiment.

Oil in the feed and permeates was extracted with dichloromethane (HiPerSolv CHROMANORM for HPLC, VWR) and measured by UV spectrophotometer (Lambda 650, PerkinElmer, USA) [29]. Oil concentration was calculated from the calibration curve of the absorbance at 259 nm. Droplet size distribution of the feed water was measured by XPT-C particle analyzer (Prozesstechnik GmbH, Germany).

2.3. Operating conditions

All experiments ran at room temperature and a constant-flux mode. The retentate and permeate were circulated back to the feed tank to maintain a constant oil concentration in the feed. As backpulsing brought clean water into the system, the same amount of permeate were discharged. Although a small amount of oil deposited on the membranes, the quantity was negligible compared to the large amount of feed water. Samples for the measurement of oil concentration in the feed and permeates were collected and analyzed after experiments. Samples for droplet size distribution in the feed were measured immediately to avoid coalescence and separation of oil droplets.

All experiments were performed at a constant permeate flux of 100 L/(m^2 h) (LMH)±1% by adjusting TMP [30]. The filtrations with backpulsing were set for 4800 s while the filtration without backpulsing only lasted for 2400 s due to severe membrane fouling. CFV was 2 m/s. The average pressure before membrane $(P_1+P_2)/2$ was set to be 0.5 bar.

TMP during filtration is calculated by

$$TMP = \frac{P_{in} + P_{out}}{2} - P_{per.} \tag{1}$$

Backpulsing amplitude is the reverse TMP calculated by

$$Amplitude = P_{per.} - \frac{P_{in} + P_{out}}{2} \tag{2}$$

where P_{in} is the crossflow inlet pressure, P_{out} is the crossflow outlet pressure, and $P_{per.}$ is the permeate pressure. The pressures involved in the above Eqs. (1) and (2) were measured by pressure transmitters at a frequency of 2 Hz. Specific flux J_s was calculated by flux divided by the averaged TMP between two consecutive backpulses. Flux was corrected to 20 °C by considering changes in water viscosity with temperature.

The cleaning procedures of membranes after each experiment consisted of water flushing, chemical cleaning and water flushing. 1%-v of Derquim+(PanReac AppliChem, Germany) was used for chemical cleaning for 1 h at 60 °C and CFV of 2.8 m/s. The permeate lines were closed during the first and last 20 min and opened in the middle 20 min. More than 90% of flux recovery was achieved by this cleaning method. If not, chemical cleaning would be repeated.

2.4. Factorial design methodology

A 2^3 full factorial design was used to identify the importance of backpulsing amplitude, duration and frequency, and the interactions between them [31,32]. The high and low levels of each parameter were defined in Table 2. The ranges of the factors were selected based on literature summary [16], capacity of the setup and economic considerations. 2 blocks were used in the factorial design as experiments ran with 2 replicates. 3 center samples were added to each block to check error, curvature and reproducibility [33]. In total 22 experiments were performed. The experiments were carried out in a random order to avoid system errors [34]. Minitab 19 was used for the design of experiments and data analysis.

Two responses were investigated: J_{sf} (averaged specific flux of forward filtration at the final cycle, LMH/bar) and Y (net permeate yield during the whole experiment period, L). The two responses were normalized for comparison between experiments by Eqs. (3) and (4), respectively,

(Normalized) final specific flux J_{nsf}:

$$J_{nsf}(\%) = \frac{J_{sf}}{J_{si}} \times 100 \tag{3}$$

where J_{si} is the initial specific flux of the membrane, LMH/bar.

(Normalized) net yield Y_n:

Table 1
Physiochemical properties and compositions of crude oil.

Crude oil from NCS	
Density at 20 °C (g/cm^3)	0.847
Viscosity at 20 °C (mPa s)	11.05
TAN (mg of KOH/g of oil)	<0.1
TBN (mg of KOH/g of oil)	0.851
SARA composition [% (w/w)]	
Saturates	74.81
Aromatics	20.92
Resins	4.25
Asphaltenes	0.02
Water content (ppm)	1112.1
IFT at pH 6 (mN/m)	
Na-Brine	22.7 ± 0.1

Abbreviations: TAN, total acid number; TBN, total base number; IFT, interfacial tension.

Table 2
Levels of factors in the factorial design.

Factors	Coded symbol	Values of coded levels	
		Low level (−1)	High level (+1)
Amplitude (bar)	A	0.1	1.0
Duration (s)	B	0.1	0.5
Frequency (Hz)	C	0.0167	0.05

$$Y_n(\%) = \frac{Total\ amount\ of\ permeate - Total\ BP\ water\ consumption}{Total\ amout\ of\ permeate} \times 100 \quad (4)$$

3. Results and discussion

3.1. Effect of backpulsing on membrane performance

The design matrix and the results of the responses are shown in Table 2. The conditions of the synthetic produced water for each experiment are also listed in the table, with an average oil concentration of (199.6 ± 15.6) mg/L and an averaged droplet size of (4.6 ± 0.1) μm based on number distribution. Stable feed conditions ensure reliable comparison of the effect of different backpulsing strategies on membrane performance. Particle size distribution by number of the influent is shown in Fig. 2. The majority of droplets (68.1%) were smaller than or equal to 5 μm.

As shown in Table 3, the oil concentrations of permeates for all experiments were less than 5 mg/L and oil removals were all above 97%. Besides, the quality of permeates were slightly improved when backpulsing was used, except experimental run 3. This is because oil emulsions are deformable and are likely to pass more through membrane pores under continuous TMP than TMP with intermittent release, even though the size of oil emulsions are mostly larger than membrane pore size. Similar results were observed by Silalahi and Leiknes [21] that in various experiments testing different membrane pore sizes and different feed water properties, oil removal by microfiltration all increased when backpulsing was used compared to without backpulsing.

The effect of backpulsing on fouling mitigation was demonstrated by the decline of the normalized specific flux under different backpulsing strategies, as shown in Fig. 3 (a) and (b). In filtration without backpulsing, the normalized specific flux dropped sharply in the initial 1000 s and continued decreasing to 11.2% at only 2400 s for both membranes. When backpulsing was used, membrane fouling was much less. All the backpulsing experiments could run the entire experimental period (4800 s). However, the efficiency of fouling mitigation varied a lot between different backpulsing strategies. The behavior of fouling mitigation by backpulsing under different conditions is discussed in detail in Section 3.3. Note that permeate is normally used to backflush the membrane. Distilled water was used in this study due to the limitation of the setup.

As is shown in Fig. 3 and Table 3, the most effective backpulsing was under conditions (+1,+1,+1) and the final specific fluxes (J_{nsf}) were 72.0% and 70.1% for membrane #1 and #2, respectively. However, the corresponding net yields (Y_n) were very low, 74% and 72.8% for membrane #1 and #2, respectively (see Table 3). The optimal backpulsing conditions should be found by optimizing both responses overall. The best run in this study was backpulsing with (+1,−1,−1) conditions. Note that the optimal backpulsing conditions vary with different operating conditions (e.g., CFV and flux) for a given feed water and a given membrane. Because backpulsing efficiency is dependent on membrane fouling situation which is influenced by the setting of operating conditions. In this study, CFV was chosen to 2 m/s which is within the typical range of 0.2–5 m/s CFV reported in different studies of the microfiltration/ultrafiltration of produced water [8]. Constant flux was set to be 100 LMH considering that the threshold flux in a similar study was 88 ± 6 LMH, above which rapid membrane fouling was observed [35].

These experiments were done at a certain constant flux, so membrane surface area costs are fixed. The lower the specific flux after a run implies more frequent chemical cleaning (greater cleaning costs) and more frequent membrane replacement (greater membrane replacement costs). If a certain normalized specific flux was considered for a CEB (chemically enhanced backwash) or a CIP (clean in place), for example 80% in Fig. 3 (a) and (b), both membranes could only run about 200 s in the filtration without backpulsing. Filtration with (−1,−1,−1) backpulsing could extend the experiment to around 400 s, while filtration with (+1,+1,+1) backpulsing could prolong the experiment to more than 2900 s for both membranes. Therefore, a proper design of backpulsing conditions is important not only to the mitigation of membrane fouling but also to the minimization of operating expenses.

3.2. Presentation of key statistical results

Analysis of variance was used to analyze results from the full factorial design. Two blocks were used to represent the two individual membranes. The P-values of blocks for both responses were larger than 0.05, meaning that blocks were insignificant, which confirms that all the experiments were repeatable. Empirical models with significant terms were used for two responses at a 95% confidence interval respectively:

$$J_{nsf} = 44.14 + 21.41A + 3.39B + 3.21C - 2.12AB + 1.30ABC \quad (5)$$

$$Y_n = 90.54 - 4.61A - 2.49B - 4.86C - 1.65AB - 1.55AC - 1.22BC \quad (6)$$

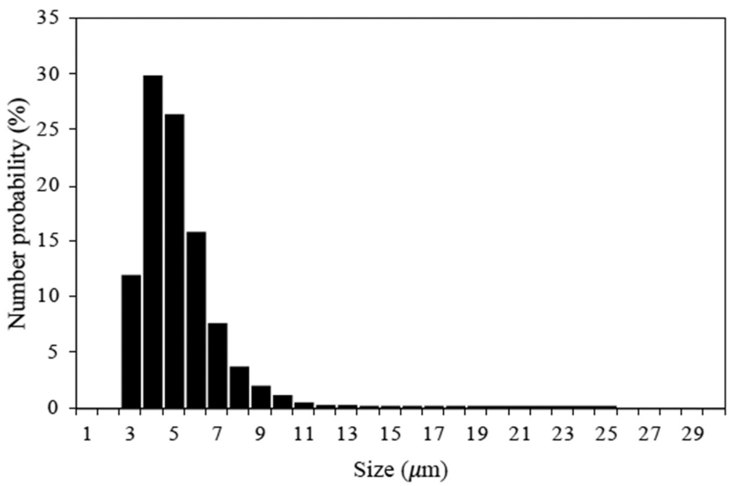

Fig. 2. Particle size distribution by number for the synthetic produced water.

Table 3
Experimental design and results.

Run test order	Feed		Permeates		Membranes		Coded values of factors			Responses			
	Avg. oil conc. (mg/L)	Mean size (μm)	Avg. oil conc. (mg/L)		J_{si} (LMH/bar)		Amp.	Dur.	Freq.	J_{nsf} (%)		Y_n (%)	
			#1	#2	#1	#2				#1	#2	#1	#2
10	180.9	4.6	2.5	1.7	509	626	+1	+1	+1	72	70.1	74	72.8
2	226.1	4.8	3.4	3.1	525	608	+1	-1	+1	67	66.7	82.3	88.8
9	210	4.6	3.3	2.3	582	541	+1	+1	-1	62.6	62.6	90.1	90.2
4	191.1	4.5	4.1	3.7	515	649	+1	-1	-1	61.5	62	94	95.1
3	195.4	4.6	4.8	4.4	637	565	0	0	0	60.6	61.5	93.4	93.4
5	188.9	4.5	4.4	3.3	517	595	0	0	0	63	62.6	93.2	93.9
11	185	4.5	2.4	2.4	553	520	0	0	0	60.9	62	93.9	94.8
8	190.1	4.9	4.1	2	513	568	-1	+1	+1	29.9	29.1	86.5	94.5
6	227	4.5	4.5	2.2	639	617	-1	+1	-1	27.9	26.1	97.5	98.7
7	199.8	4.6	4.4	2.4	549	592	-1	-1	+1	23.1	20.9	93	93.3
1	189.1	4.7	3.9	4.4	526	608	-1	-1	-1	11.9	13	98.7	98.9
0	212.1	4.6	4.5	4.6	539	578	Filtration without backpulsing			11.2	11.2	100	100
Best run	191.1	4.5	4.1	3.7	515	649	+1	-1	-1	61.5	62	94	95.1

Abbreviations: Avg., average; conc., concentration; Amp., amplitude; Dur., duration; Freq., frequency.

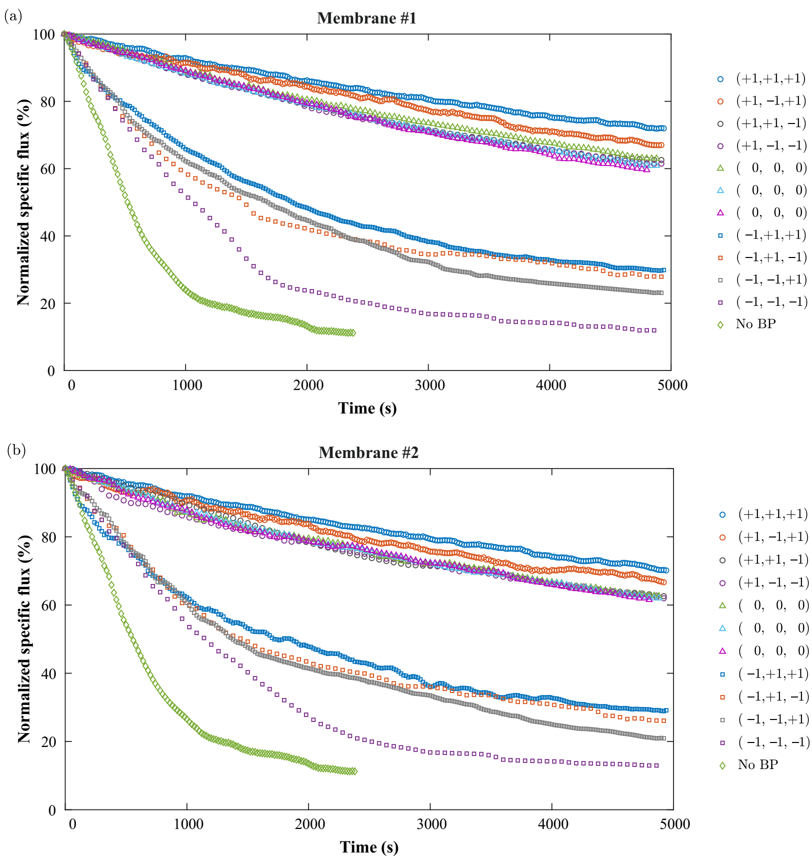

Fig. 3. Normalized specific flux versus time of experiments without backpulsing and experiments using different backpulsing strategies. (a) Membrane #1; (b) Membrane #2.

where A, B and C are amplitude, duration and frequency, respectively. Note that the equations and coefficients were only valid to the coded units of terms. The coefficients of determination (R^2) of the two models were 99.88% and 95.07% respectively, and the adjusted ones (Adj-R^2) were 99.80% and 92.03% respectively, indicating that the two models explained the process well.

Fig. 4 presents the relative magnitude of the terms and their statistical significance based on student's *t*-test. The standardized effects are absolute values of the terms' T-values. Reference lines are at critical T-values, which was $T_{0.025,\ 12}$=2.18 for final specific flux and $T_{0.025,}$

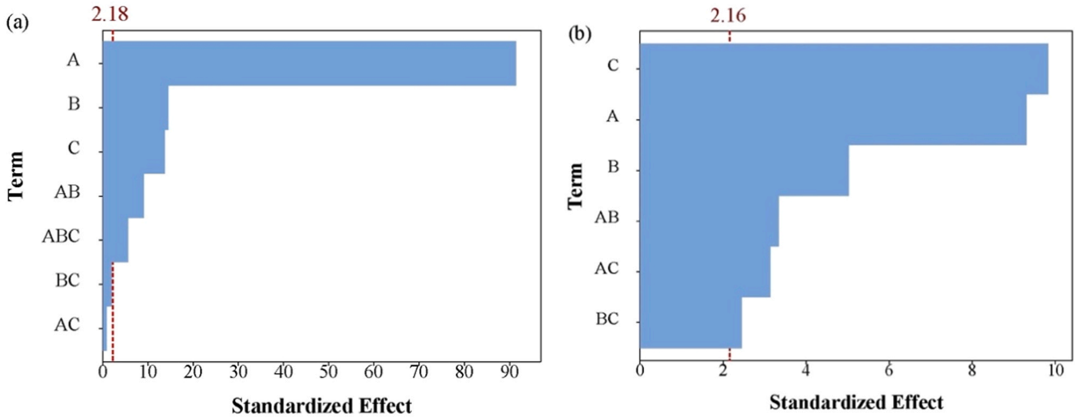

Fig. 4. Pareto chart of the standardized effects for (a) final specific flux and (b) net yield.

$_{13}$=2.16 for net yield at a 95% confidence interval in current study. Terms with bars that cross the reference lines are statistically significant. The relative importance of main factors and their interactions are easily displayed: A >B>C>AB>ABC for final specific flux and C>A>B>AB>AC>BC for net yield.

Plots of sample means give more visual information on interactions, as is shown in Fig. 5. If the dashed and solid lines are far from parallel in an individual squared plot, the interaction between the two factors is important for the response. In the upper left plot in Fig. 5 (a), when duration was at the low level (see the blue solid line), final specific flux changed from 17.2% to 64.3% (47.1% in difference) along with amplitude from −1 to +1. While when duration was at the high level (see the green dashed line), final specific flux changed from 28.2% to 66.8% (38.6% in difference) with the changing of amplitude from −1 to +1. The variation of duration levels changed quite a bit the effect of amplitude on final specific flux, which means the interaction between amplitude and duration was important. Similar significant interactions were also shown in interactions of amplitude × duration, amplitude × frequency, and duration × frequency for net yield in Fig. 5 (b). If there is a difference between the two-way interactions, then there is a three-way interaction [36]. For instance, for final specific flux, the two-way interaction of amplitude × duration was important, while amplitude × frequency was not significant. Therefore, three-way interaction between amplitude × duration × frequency was important.

The analysis of a factorial design assumed that the experimental data come from a normal distribution. The normality was checked by plotting a normal probability plot of residuals at a 95% confidence interval, as shown in Fig. 6 (a) and (b), respectively. All the data points fell fairly close to the straight lines and within the confidence interval ranges, thus it is 95% confident that the data were normally distributed and reliable.

3.3. Mechanisms of backpulsing on membrane performance

Combining the performance of different backpulsing strategies on produced water microfiltration in Section 3.1 and statistic results in Section 3.2, mechanisms of backpulsing on fouling mitigation and membrane productivity were further investigated in this section.

Amplitude, duration and frequency all showed a positive effect on final specific flux and a negative effect on net yield, as shown by the signs of the coefficients in Eqs. (5) and (6), respectively. This is reasonable because higher amplitude exerted stronger reverse force to dislodge deposited foulants, longer duration provided longer reverse flow for cleaning and higher frequency helped prevent membrane fouling to be fully formed or compact. Higher amplitude and longer duration meant more cleaning and higher frequency meant less fouling, which all resulted in less fouling accumulation and a higher membrane flux. However, increasing in amplitude, duration or frequency inevitably led to higher consumption of clean water and therefore a lower net yield.

Amplitude played a major role on the success of backpulsing for fouling mitigation. Fig. 4 (a) showed that amplitude had a profound effect on final specific flux, which far surpassed the other terms. In Fig. 3, when amplitude was at −1 level (0.1 bar), backpulsing could mitigate membrane fouling to some extent, however, apparent drops of normalized specific flux were still observed. When amplitude increased to 0 level (0.55 bar), no obvious drops of normalized specific flux were shown, instead, normalized specific flux declined slowly and steadily from the beginning of filtration. However, further increase in amplitude did not show much improvement in normalized specific flux. This means that a minimum amplitude existed for an effective backpulsing and it was between 0.1 and 0.55 bar in current filtration conditions. Similar results were also obtained by Mores et al. [37] in the microfiltration of washed bacterial cells that the maximum fraction of the membrane cleaned by backpulsing increased with increasing amplitude up to 5 psi (0.34 bar), above which the cleaning efficiency leveled off. In the microfiltration of light non-aqueous phase liquids, McAlexander and Johnson [38] found the minimum amplitude was 0.6–0.85 bar for effective membrane cleaning. Therefore, the dominated significance of amplitude on final specific flux was probably because that the range of amplitude was across the minimum effective pressure.

Frequency was the most essential factor for net yield, as is demonstrated in Fig. 4 (b) where frequency had the highest standard effect. Various backpulsing experiments in literature also showed that frequency was a significant parameter to optimize net permeate flux [23–27]. If frequency was too low, membrane was not cleaned timely, then permeate flux was low. However, at an extreme high frequency, net permeate was also low because of the excessive loss of clean water and filtration time. Besides, membrane filtration with backpulsing is a dynamic process [15], and frequency is a very important factor for the dynamic characteristics and stability of the system. In Fig. 3, there are fluctuations shown in the specific fluxes and more instability is shown in experiments at +1 level frequency than in experiments at 0 or −1 level frequency.

Duration was an important parameter for both responses too, but not as prominent as the other two parameters, as is shown by the standardized effect bars in Fig. 4. The importance of duration increased when membrane fouling became more severe. As is shown in Fig. 3, when amplitude was at −1 level and membrane fouling could not be removed effectively, the final specific fluxes in experiments with a longer duration were higher than those in experiments with a higher

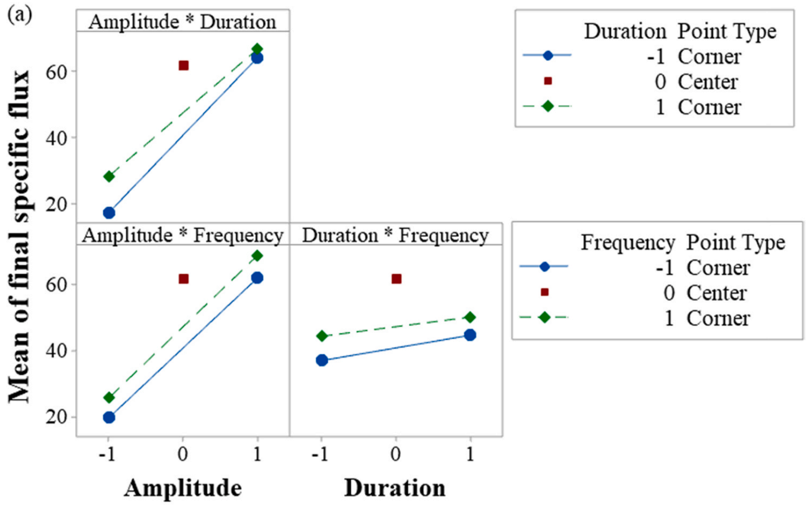

Fig. 5. Interaction plots for (a) final specific flux and (b) net yield. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article)

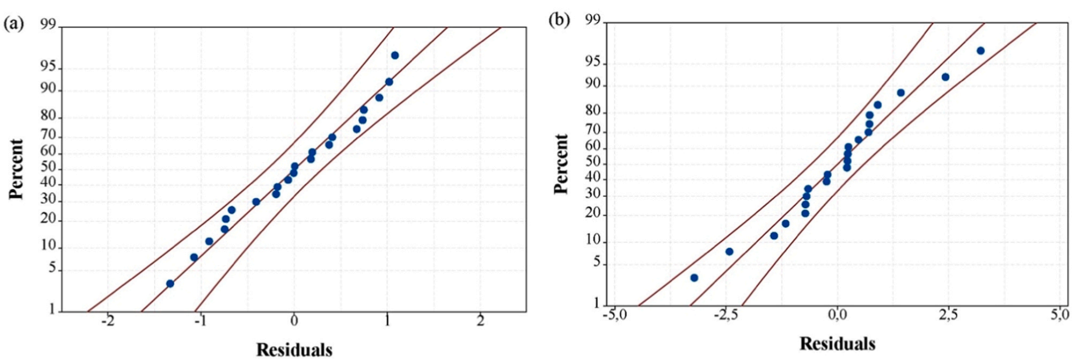

Fig. 6. Normal probability plots of the residuals for (a) final specific flux and (b) net yield.

frequency, while it was opposite when amplitude was at +1 level. This is also evident by the cross-over observed between specific flux data of runs with levels of (−1,+1,−1) and (−1,−1,+1) in Fig. 3 (a) and (b), respectively. At the later stage of filtration, backpulsing with (−1,+1,−1) conditions became more effective than backpulsing with (−1,−1,+1) conditions. This is probably because when membrane fouling accumulated, backpulsing with a longer duration although a lower frequency gave a better cleaning to the membrane than backpulsing with a shorter duration but a higher frequency.

The importance of interactions among backpulsing parameters for both responses could not be ignored, which has not been figured out in any other literature before. Actually, there are also limited studies of interactions among hydraulic parameters of membrane filtrations [39]. Three-way interaction is too complicated, which gives little information for understanding the mechanisms of backpulsing and will not be discussed here, but two-way interactions are important to investigate. For final specific flux, only the two-way interaction of amplitude × duration was important, which is reasonable as amplitude and duration both affect the efficiency of fouling removal from the membrane, while frequency affects more on the fouling formation. However, the sign of the coefficient was negative, opposite to the coefficient signs of the three main factors, as shown in the empirical model in Eq. (5). That is when duration/amplitude was at the low level, final specific flux increased more as amplitude/duration increased than when duration/amplitude was at the high level, as is shown in Fig. 5 (a). Such a trade-off interaction between amplitude and duration is because backpulsing could only remove the hydraulically reversible fouling. For net yield, the three two-way interactions were all important and the signs of the interaction coefficients in Eq. (6) were all negative, the same as the three main factors, which means that all the three factors promoted the effect of each other on net yield, similar to a so-called synergistic effect. Therefore, to investigate the effect of backpulsing parameters on membrane performance or to find the optimal backpulsing conditions, it is not reliable to focus on one parameter at a time without taking the interactions between parameters into account.

4. Conclusion

The results of present work showed that microfiltration with an averaged membrane pore size of 0.1 μm was effective to remove dispersed oil down to less than 5 mg/L with an averaged oil concentration of 199.6 mg/L in the influent. Membrane fouling in filtration without cleaning was severe. Backpulsing mitigated membrane fouling. The cleaning efficiency varied a lot with the different backpulsing parameters. Through a 2^3 full factorial design, the effect of the backpulsing parameters and their interactions on membrane performance was investigated systematically, and the fundamental understanding of fouling mitigation by backpulsing was further improved. All the three backpulsing parameters were found to be important for the two responses of final specific flux and net yield. Increasing any of the three backpulsing parameters led to an increase in the final specific flux but caused more water loss and thus a lower net yield. Amplitude affects the reverse force to dislodge foulants and was the most influential factor for final specific flux. There was a minimum threshold of amplitude for an effective backpulsing. Frequency plays a role on the situation of fouling formation and was the most significant one for net yield. Duration represents the time for each cleaning was important to both final specific flux and net yield too, but not as prominent as the other two parameters. Interactions of amplitude × duration and amplitude × duration × frequency were important for final specific flux. All two-way interactions between amplitude, duration and frequency were important for net yield. However, the two-way interactions on the two responses performed differently. Increasing duration/amplitude reduced the effect of amplitude/duration on final specific flux, while increasing any of the parameters promoted the effect of the other parameter on net yield. Further investigation on backpulsing parameters and the optimization of backpulsing conditions should take the interactions between parameters into consideration.

CRediT authorship contribution statement

Yinghong Gao: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Writing - original draft. **Yeqing Zhang**: Data curation, Formal analysis. **Marcin Dudek**: Data curation, Writing - review & editing. **Jie Qin**: Software, Investigation. **Gisle Øye**: Resources, Supervision, Writing - review & editing. **Stein Wold Østerhus**: Conceptualization, Validation, Funding acquisition, Supervision, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Paper III

Application of ceramic membranes for produced water treatment. Part I: Effect of feed compositions on membrane fouling

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Manuscript ready to submit

This paper is awaiting publication and is not included in NTNU Open

Paper IV

Application of ceramic membranes for produced water treatment. Part II: Comparison of backpulsing and backwashing for membrane fouling mitigation

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Manuscript ready to submit

This paper is awaiting publication and is not included in NTNU Open

Appendix B. Additional results

Results on optimal backpulsing frequency

Introduction

It is shown in various publications that backpulsing frequency is critical to net permeate flux [1-5]. Besides, in our previous work on investigating the importance of backpulsing parameters, frequency was found to be the most significant parameter for net permeate yield [6]. Therefore, experiments with a wide range of frequency were carried out to study the effect of frequency on membrane filtration. The backpulsing frequency varied from 0.0048 Hz (every 210 s a backpulse) to 0.11 Hz (every 9 s a backpulse).

Methods

The experimental setup, membranes, and filtration conditions were the same as used in Paper IV. New membranes were used in each experiment. Backpulsing amplitude was 0.5 bar and duration was 0.6 s. PW1 in Paper III with the composition of a light crude oil, tween 80 surfactant and NaCl brine was used as the feed water.

Results and discussion

A group of experiments was carried out to find the optimum backpulsing frequency with the backpulsing interval between 9 s and 210 s. New membrane flux for each membrane in each experiment is shown in Table 1. The new membrane fluxes of TiO_2 membrane were much higher than those of Al_2O_3 membrane and ZrO_2 membrane. This is similar to the tests in Paper III.

Table 1. New membrane fluxes in each experiment.

BP Interval	New membrane flux (LMH)		
	TiO_2	Al_2O_3	ZrO_2
9s	2766	929	771
10s	3091	800	922
11s	3092	839	914
12s	3024	864	864
13s	2880	895	855
15s	3074	868	731
30s	3045	787	688
60s	2772	720	-
210s	2451	734	778
Average	2911	826	815

Gross yields and backpulsing consumptions were measured after experiments, as shown in Table 2. Comparing the three membranes, TiO_2 membrane produced much more permeate than Al_2O_3 membrane and ZrO_2 membrane in general during the 10000 s tests, as the averaged gross yield of TiO_2 membrane was 12.27 kg and those of Al_2O_3 membrane and ZrO_2 membrane were 6.93 kg and 6.57 kg, respectively. The averaged clean water consumption due to backpulsing was also the highest for TiO_2 membrane (1.24 kg), compared with 0.61 kg for Al_2O_3 membrane and 0.47 kg for ZrO_2 membrane. Gross permeates generated by TiO_2 membrane had the biggest difference at different backpulsing intervals, while ZrO_2 membrane has the smallest difference, as is shown by the standard deviations of gross yields for the three membranes. Backpulsing consumptions of the three membranes followed similar tendencies. This is probably because the pore size distribution of TiO_2 membrane was the widest of the three membranes, and ZrO_2 membrane had the narrowest but similar to Al_2O_3 membrane.

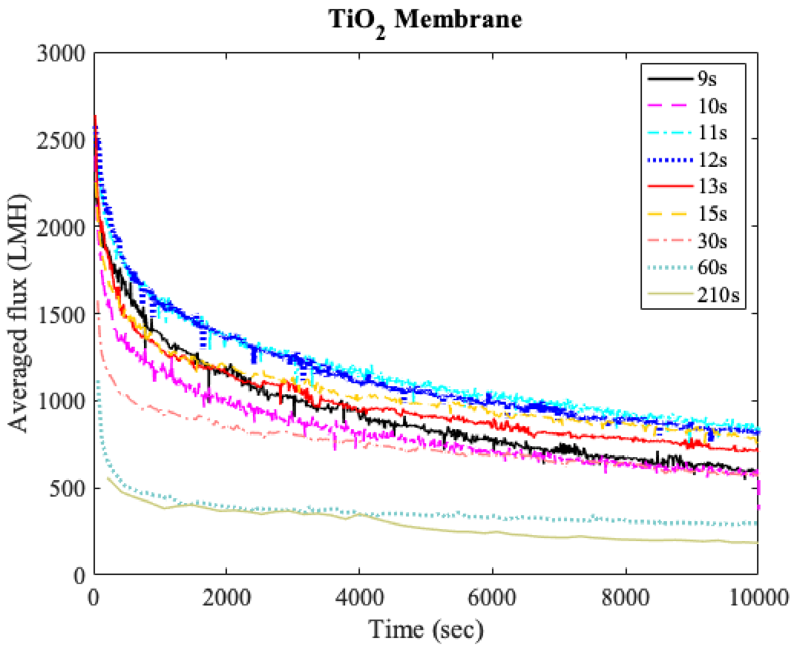
Table 2. The gross yields and backpulsing consumptions of different membranes at experiments with different backpulsing intervals.

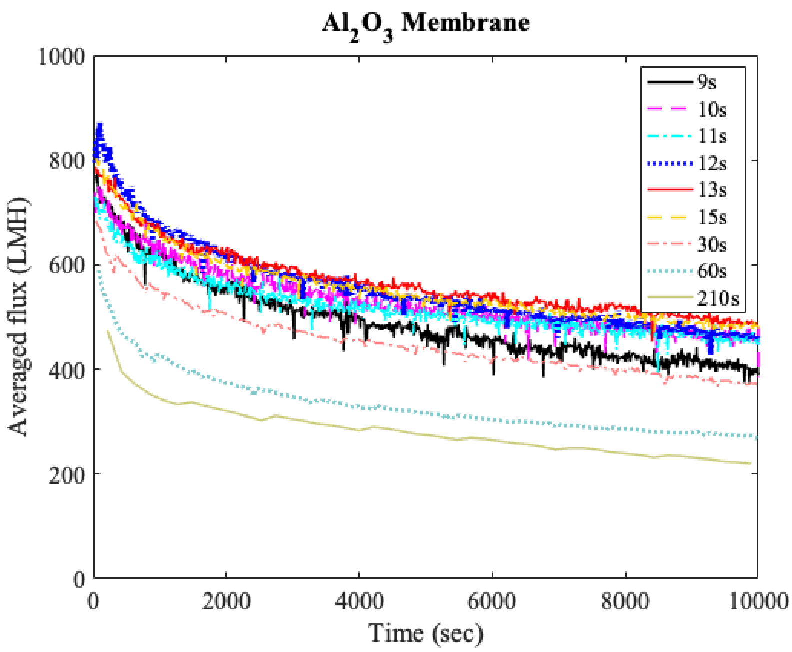
BP Interval	Gross yield (kg)			Backpulsing consumption (kg)		
	TiO_2	Al_2O_3	ZrO_2	TiO_2	Al_2O_3	ZrO_2
9s	12.25	6.67	5.17	1.63	**0.95**	**0.67**
10s	11.61	**7.34**	**7.66**	**1.72**	**0.90**	**0.75**
11s	**15.96**	**7.31**	**7.02**	**1.91**	**0.92**	**0.68**
12s	**16.21**	**7.80**	**7.85**	**1.81**	0.68	0.64
13s	**14.20**	**8.03**	**7.46**	1.61	0.73	0.54
15s	**15.34**	**8.20**	6.34	1.40	0.67	0.43
30s	11.99	7.02	5.64	0.59	0.38	0.20
60s	7.17	5.31	-	0.39	0.12	0.32
210s	5.74	4.71	5.41	0.10	0.11	0.04
Average	12.27	6.93	6.57	1.24	0.61	0.47
STDEV	3.73	1.20	1.07	0.69	0.33	0.24

Note: higher values are marked in bold. Abbreviation: STDEV, standard deviation.

Detailed permeate flux curves that corresponds to gross yields are shown in Figure 1. Experiments with the flux curves staying in the upper area of each plot and the end fluxes of higher values generate higher gross yields. As shown in Figure 1, experiments with backpulsing interval of 11 – 15 s generally produced the highest permeates for TiO_2

membrane that are also marked in bold in Table 2. Backpulsing intervals of 10 – 15 s are good options for Al_2O_3 membrane referring only to the gross yield. For ZrO_2 membrane the optimal backpulsing intervals are between 10 – 13 s. Experiments with more frequent backpulsing would not generate too much permeate is obvious known to be due to the less filtration time. Moreover, Figure 1 shows that backpulsing with very short interval of 9 s was not efficient to remove membrane fouling, as the fluxes declined fast during the initial filtration period and the end fluxes were at relatively low level for all the three membranes. This is a bit surprising as the common sense would be that the more frequent the backpulsing cleaning is, the less compact the fouling would generate and the more efficient the cleaning is [7]. The reason here may be that backpulsing with too high frequency (every 9 s a backpulse) generated too much turbulence in the filtration process that caused more irreversible fouling than backpulsing with medium range frequencies (e.g., 11 s to 15 s). Clean water consumptions due to backpulsing were also affected by membrane fouling situation. It is known that the more frequent the backpulsing is, the more time a membrane is under reverse cleaning and the more accumulated clean water consumption for backpulsing is. Nevertheless, the accumulated backpulsing consumptions were not always the highest for the experiment with backpulsing interval of 9 s (see Table 2). This is probably also because of severe fouling and the resulting low reverse fluxes. Experiments with less frequent backpulsing (interval of 60 s or 210 s) could not produce much permeate either, as it is apparent from Figure 1 that the permeate fluxes dropped dramatically to a low level and could not be recovered efficiently by backpulsing. This is mainly due to the severe fouling generated during long forward filtration time and the inefficient cleaning of backpulsing.


TiO2 Membrane
Averaged flux (LMH)
Time (sec)
9s
10s
11s
12s
13s
15s
30s
60s
210s
0
500
1000
1500
2000
2500
3000
2000
4000
6000
8000
10000


Al2O3 Membrane
Averaged flux (LMH)
Time (sec)
9s
10s
11s
12s
13s
15s
30s
60s
210s
0
200
400
600
800
1000
2000
4000
6000
8000
10000

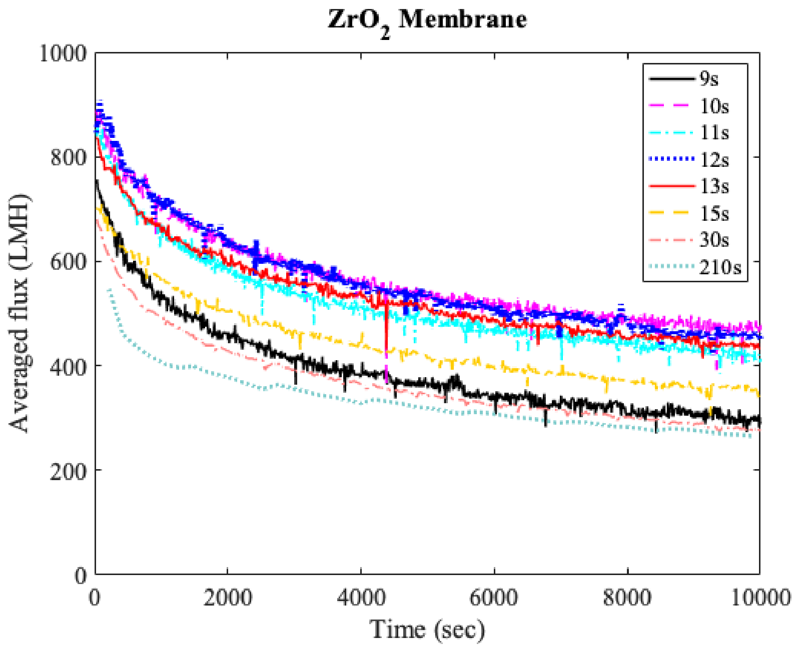


Figure 1. Flux decline curves at different backpulsing intervals. (1) TiO_2 membrane; (2) Al_2O_3 membrane; (3) ZrO_2 membrane.

To find the optimum backpulsing intervals, net permeate yields should be considered by using gross permeates subtracting backpulsing consumptions. Net yields of experiments with different backpulsing intervals are shown in Figure 2. In Figure 2 (1) for TiO_2 membrane, experiment with backpulsing interval of 12 s generated the highest net yield of 14.4 kg, followed by experiments with backpulsing interval of 11 s and 15 s. In Figure 2 (2) for Al_2O_3 membrane, experiment with backpulsing interval of 15 s generated the highest net yield of 7.5 kg, followed by experiments with backpulsing interval of 13 s and 12 s. In Figure 2 (3) for ZrO_2 membrane, experiment with backpulsing interval of 12 s generated the highest net yield of 7.2 kg, followed by experiments with backpulsing interval of 13 s and 10 s.

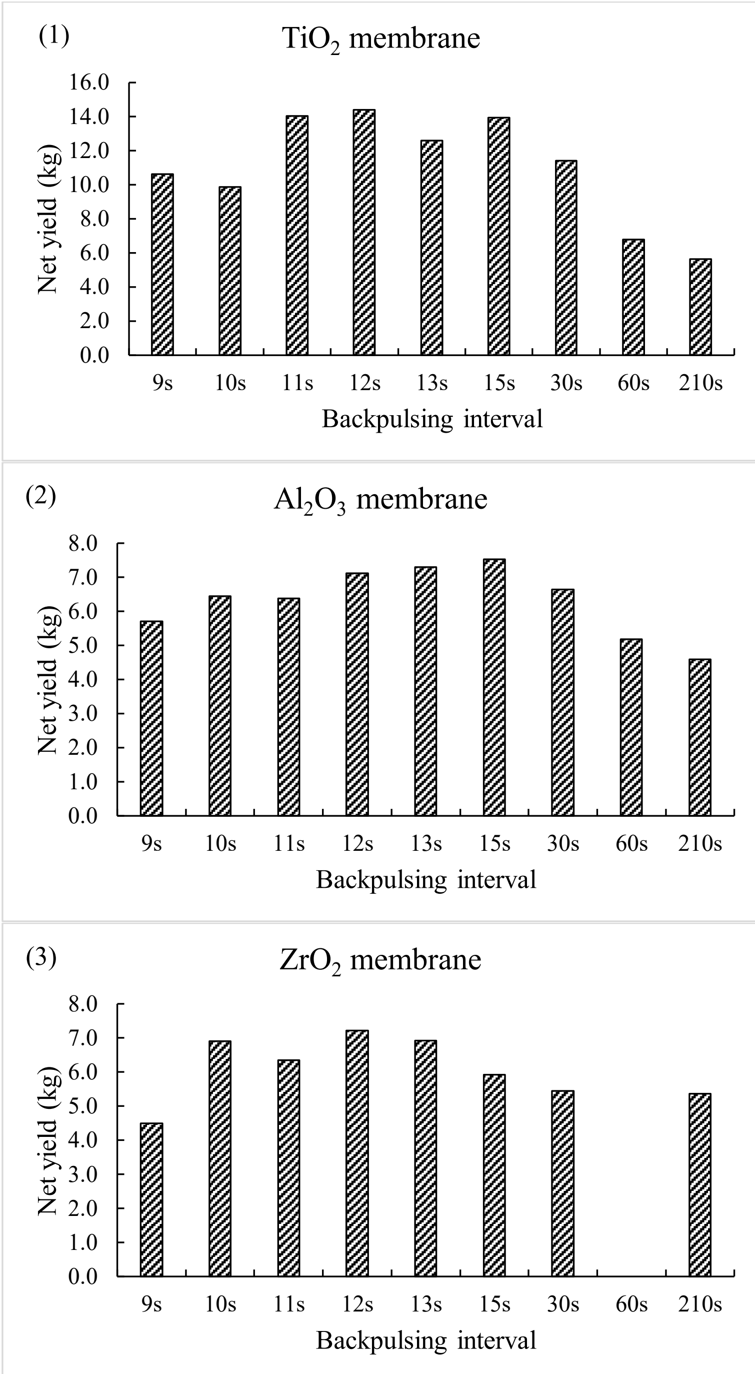


Figure 2. Net yields at different backpulsing intervals. (1) TiO_2 membrane; (2) Al_2O_3 membrane; (3) ZrO_2 membrane.

Conclusion

The experiments with a high backpulsing frequency (every 9 s per backpulse) did not generate high net yields for all the three membranes. This is not only because backpulsing consumptions were high, but also because membranes had relatively low permeate fluxes compared with membranes under other backpulsing conditions. The experiments with low frequencies (every 60 s or 210 s per backpulse) generated low net yields. Although backpulsing consumptions were quite low, the gross permeates were also low due to hydraulically irreversible fouling. The optimal backpulsing frequencies were between 11 s and 15 s for the three membranes in general.

In the three membranes, TiO_2 membrane had the biggest new membrane fluxes, gross yields, backpulsing consumptions, net yields, and variations between experiments with different backpulsing frequencies. While ZrO_2 membrane had the smallest productions, consumptions and variations between experiments, but similar to Al_2O_3 membrane. This is in correspondence with the pore size distributions of the membranes that TiO_2 membrane had the widest pore size distribution, while ZrO_2 membrane had the narrowest and similar to Al_2O_3 membrane.

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Appendix C. Co-author statements

Encl. to application for assessment of PhD thesis

Faculty of Engineering

NTNU

STATEMENT FROM CO-AUTHOR

(cf. section 10.1 in the PhD regulations)

Yinghong Gao applies to have the following thesis assessed:

Backpulsing technology for membrane fouling mitigation in produced water treatment

*) The statement is to describe the work process and the sharing of work and approve that the article may be used in the thesis.

*) Statement from co-author **Stein W. Østerhus**:

I hereby declare that I am aware that the work in the articles entitled as follows, of which I am co-author, will form part of the PhD thesis by the PhD candidate Yinghong Gao who made the major contribution to the work in the design, experimental phase, data analysis and writing phase.

- Backpulsing technology applied in MF and UF processes for membrane fouling mitigation: A review
- A multivariate study of backpulsing for membrane fouling mitigation in produced water treatment
- Application of ceramic membranes for produced water treatment. Part I: Effect of feed compositions on membrane fouling
- Application of ceramic membranes for produced water treatment. Part II: Comparison of backpulsing and backwashing for membrane fouling mitigation

Trondheim 4/4-22
Place, date

Stein W. Østerhus
Signature co-author

*) Statement from co-author **Gisle Øye**:

I hereby declare that I am aware that the work in the articles entitled as follows, of which I am co-author, will form part of the PhD thesis by the PhD candidate Yinghong Gao who made the major contribution to the work in the design, experimental phase, data analysis and writing phase.

- A multivariate study of backpulsing for membrane fouling mitigation in produced water treatment
- Application of ceramic membranes for produced water treatment.
  Part I: Effect of feed compositions on membrane fouling
- Application of ceramic membranes for produced water treatment.
  Part II: Comparison of backpulsing and backwashing for membrane fouling mitigation

Trondheim 30/3-22
Place, date

Gisle Øye
Signature co-author

*) Statement from co-author **Marcin Dudek**:

I hereby declare that I am aware that the work in the articles entitled as follows, of which I am co-author, will form part of the PhD thesis by the PhD candidate Yinghong Gao who made the major contribution to the work in the design, experimental phase, data analysis and writing phase.

- A multivariate study of backpulsing for membrane fouling mitigation in produced water treatment
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  Part I: Effect of feed compositions on membrane fouling
- Application of ceramic membranes for produced water treatment.
  Part II: Comparison of backpulsing and backwashing for membrane fouling mitigation

Sandefjord, 01.04.2022
Place, date

Marcin
Signature co-author

*) Statement from co-author **Yasser K. Abdelhamed**:

I hereby declare that I am aware that the work in the articles entitled as follows, of which I am co-author, will form part of the PhD thesis by the PhD candidate Yinghong Gao who made the major contribution to the work in the design, experimental phase, data analysis and writing phase.

- Application of ceramic membranes for produced water treatment.
  Part I: Effect of feed compositions on membrane fouling
- Application of ceramic membranes for produced water treatment.
  Part II: Comparison of backpulsing and backwashing for membrane fouling mitigation

Cairo, 03.04.2022
Place, date

Signature co-author

*) Statement from co-author **Zhiwei Wang**:

I hereby declare that I am aware that the work in the articles entitled as follows, of which I am co-author, will form part of the PhD thesis by the PhD candidate Yinghong Gao who made the major contribution to the work in the structure, data analysis and writing phase.

- Backpulsing technology applied in MF and UF processes for membrane fouling mitigation: A review

ZhiWei Wang

Shanghai, March 30, 2022
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Place, date Signature co-author

*) Statement from co-author **Jie Qin**:

I hereby declare that I am aware that the work in the articles entitled as follows, of which I am co-author, will form part of the PhD thesis by the PhD candidate Yinghong Gao who made the major contribution to the work in the design, experimental phase, data analysis and writing phase.

- Backpulsing technology applied in MF and UF processes for membrane fouling mitigation: A review
- A multivariate study of backpulsing for membrane fouling mitigation in produced water treatment

Nanjing, March 30, 2022
……………………... ……………………………………
Place, date Signature co-author

*) Statement from co-author **Yeqing Zhang**:

I hereby declare that I am aware that the work in the article entitled as follows, of which I am co-author, will form part of the PhD thesis by the PhD candidate Yinghong Gao who made the major contribution to the work in the design, experimental phase, data analysis and writing phase.

- A multivariate study of backpulsing for membrane fouling mitigation in produced water treatment

Shanghai, March 30, 2022
……………………... ……………………………………
Place, date Signature co-author

*) Statement from co-author **Junli Wan**:

I hereby declare that I am aware that the work in the articles entitled as follows, of which I am co-author, will form part of the PhD thesis by the PhD candidate Yinghong Gao who made the major contribution to the work in the design, experimental phase, data analysis and writing phase.

- Application of ceramic membranes for produced water treatment.
  Part I: Effect of feed compositions on membrane fouling
- Application of ceramic membranes for produced water treatment.
  Part II: Comparison of backpulsing and backwashing for membrane fouling mitigation

万俊力

Changsha, April 03, 2022
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Place, date

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Signature co-author

*) Statement from co-author **Xiaoyang Guo**:

I hereby declare that I am aware that the work in the articles entitled as follows, of which I am co-author, will form part of the PhD thesis by the PhD candidate Yinghong Gao who made the major contribution to the work in the design, experimental phase, data analysis and writing phase.

- Application of ceramic membranes for produced water treatment.
  Part I: Effect of feed compositions on membrane fouling
- Application of ceramic membranes for produced water treatment.
  Part II: Comparison of backpulsing and backwashing for membrane fouling mitigation

Trondheim 04 April, 2022
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Place, date

郭晓洋
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Signature co-author