TALLINN UNIVERSITY OF TECHNOLOGY

SCHOOL OF ENGINEERING

Department of Civil Engineering and Architecture

Environmental Engineering and management

FOULING, AGING AND FAILURE OF LOW-PRESSURE MEMBRANES IN WATER PURIFICATION

MADALRÕHUMEMBRAANIDE UMMISTUMINE, VANANEMINE JA TÕRKED VEEPUHASTUSES

MASTER THESIS

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Environmental Engineering and Management

THESIS TASK

Student: Sohail Kordmirza Nikouzad, 194380EABM

Study programme, EABM, Environmental Engineering and Management

Supervisor(s): Programme Director (Environmental Engineering and Management), Professor Karin Pachel, 6202504

Thesis topic:

(in English) *Fouling, aging and failure of low-pressure membranes in water purification*

(in Estonian) Madalrõhumembraanide ummistumine, vananemine ja tõrked veepuhastuses

Thesis main objectives:

Based on scientific research and publications to investigate and assess:

1. Membrane technology principles, characterization, classification, and applications in water treatment
2. Fouling and aging of low-pressure membranes and mitigation methods
3. Failure prediction of membrane systems in the water treatment process

Thesis tasks and time schedule:

No	Task description	Deadline
1.	Literature review; membrane technology overview and MF/UF applications in the water treatment process	15 February
2.	Literature review; different approaches of fouling and aging assessment for MF/UF membrane in the water treatment process	01 March
3.	Literature review; holistic failure characterization of membrane systems in the water treatment process	15 March
4.	Review of the studies' results to propose the conclusion, recommendations, and future trends	01 April
5.	Thesis writing	30 April

Language: English **Deadline for submission of thesis:** ".26.".May 2021a

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PREFACE

I wish to express my deepest gratitude to my kind supervisor Prof. Karin Pachel for her great support and guidance right through my Master's Degree thesis. I have been hugely fortunate to have a supervisor who paves the ground for me to work on the topic.

This study is an in-depth review of membrane failure investigation in the water purification process due to membrane fouling and aging. The main focus in this matter is exclusively on widespread low-pressure membranes including microfiltration (MF) and ultrafiltration (UF). Herby, the study follows the hierarchy of operational conditions as causes and effects of final membrane system failure.

In this regard, there are summarized preliminary data of the membrane technology followed by the mechanism, classification, and applications mostly achieved from handbooks and regarded published works. Followed by that, MF/UF integrated system in the water treatment process is justified as the base of the study. The study core is respected to MF/UF fouling, aging, and failure studies. In each mentioned issue, a comprehensive background causes and effects, mechanisms, mitigation methods, and evaluation of approaches are scrutinized.

Based on the author's viewpoint, the mentioned assessment has been done by using extracted data from the newest published kinds of literature in the field, then, the classification has been applied to form a conclusion in each section from the author point of view. A significant share of reviewed data is rooted in the Environmental Systems Engineering (ESE), UBC, Canada, which is at the frontline of research studies on the topic.

Moreover, there exist novel methods per proposed conclusions which are assessed at the end of chapters in the form of a case study or recommendations as to the future trend.

Keywords

Water treatment; MF/UF membranes; Membrane fouling and aging; Membrane failure

LIST OF ABBREVIATION

AST	Acoustic sound test
BGA	Beta-lactoglobulin
BSA	Bovine serum albumin
CF	Concentration factors
CP	Concentration Polarization
DAF	Diffusive air flow
DAF	Dissolved air flotation
DBP	Disinfection by-products
FEA	Finite Element analysis
GDM	Gravity-driven membrane
HAs	Hydrophilicity Add-ons
IMS	Integrated Membrane System
LRV	Log removal value
MBR	Membrane Bio-reactor
MF	Microfiltration
MTBF	Mean time between failures
MTTF	Mean time to failure
MWCO	Molecular weight cut-off
NF	Nanofiltration
NOM	Natural organic material
OVA	Ovalbumin
PDT	Pressure decay test
PGM	Passive Gravity-driven membrane
PMR	Photocatalytic Membrane Reactors
PVDF	Polyvinylidene difluoride
RCM	Reliability-centered maintenance
RO	Reverse osmosis
SEM	Scanning electron microscope
TFC	Thin-film composite
THM	Trihalomethane
UF	Ultrafiltration
VDT	Vacuum decay test

LIST OF SYMBOLS

C (CF)	Concentration
C_f	Concentration of feed flow (ppm)
C_p	Concentration of permeate flow (ppm)
C_r	Concentration of reject flow (ppm)
C*t	Relativity of concentration and time factors
Da	Dalton or unified atomic mass unit
lmh	Permeate of the membrane (liter/the membrane surface area/hour)
Q_f	Feed flow rate (l/m^2h)
Q_p	Permeate flow rate (l/m^2h)
Q_r	Reject flow rate (l/m^2h)

1. INTRODUCTION

Water is the most valuable and renewable element of life on earth. The human population in the last century has become three times more and will increase by 50% in the next 50 years [1]. This increase will demand more urbanization and freshwater consequently. Therefore, water would become scarce even in the areas that they have a rich source of it currently. That is why it is a must-have to investigate new methods or modify the existed approaches for water purification. Methods that can eliminate pollutants such as suspended solids, organics, biological materials, and chemicals from water and make it drinkable based on the regulations which are becoming tighter.

The continuous demand for freshwater leads to Figuring out more eco-friendly methods for water purification, besides reusing, recycling, and finding new water resources. Membrane technologies are in the front line of high-quality water production efficiently and environmental-friendly. In this way, membranes are defined as a sustainable method for water treatment due to;

- Decreasing the plant footprint
- Adaptable and lack of moving sections
- Lower occupation area (lower land use)
- Working in ambient temperature (mostly)
- Lower relative environmental impacts (than the conventional method)
- Lower demand for chemicals
- High surface per volume rate, etc. [2][3].

Thus, Membrane technology can address problems related to water shortage resulted from economic and environmental issues. Membrane applications in water treatment practices have been increased in the last 20 years [1]. Mentioned development is due to two main reasons, the decrement in costs associated with the membrane implement and the change in the regulations demanding specific drinking water quality.

In this venue, firstly, general membrane technology definition, classification, and characterization are summarized to magnitude applications of membranes in the water treatment process.

Then, different approaches towards membrane systems consist of the industrial unit design of membranes are assisted to justify exact applications of MF/UF membranes in the water treatment plant.

Furthermore, followed by characterizing the fouling and concentration polarization (CP) issues precisely, various mitigation approaches are explored. In that respect, by assessing mentioned approaches, the final recommendation is provided based on scrutinizing fouling mechanisms regards to the feed water compositions and specific plant conditions. A case study is considered as a novel membrane system to address the negative impacts of membrane fouling.

Knowing fouling issues are important due to the identification of the membrane aging mechanism. Therefore, the study depicts inclusive approaches that can be focused on lab-scale methods to characterize the membrane aging to mimic the practical circumstances in full-scale water treatment plants as an existed knowledge gap. Thus, a case study that follows the better experimental attitude regards to the membrane aging assessment, is evaluated.

Last but not the least, membrane failure is defined and characterized by mentioning reasons and consequences. The membrane failure can be classified to propose mitigation methods. In this regard, considering membrane failure threats to the whole operation, the risk assessment of the plant can be evaluated with the help of resilience, reliability, and availability terms.

Altogether, mentioned indexes can issue the holistic view of membrane failure way-outs. Since the main reason for the membrane failure is identified as membrane aging, a thorough review of applied aging monitoring tools, directly and indirectly, is proposed. Ultimately, the final recommendation can state a useful novel method related to the aging monitoring for the membrane failure anticipation more efficiently.

It worth mentioning that, conclusion and recommendation sprang from three backgrounds as reported advantages of each reviewed approach or their minor conflicts and the own perspective as an Environmental Engineering student.

2. MEMBRANE TECHNOLOGY

2.1 Membrane Definition

A membrane is defined as a bar that can let specific materials pass selectively. Membranes separate materials physically between two homogenous media and can be a substitute for chemical methods such as adsorption, extraction, distillation, etc. [4]. Semipermeable membrane filters, in liquid or gas phase feed, are capable to sieve materials classified in Figure 1;

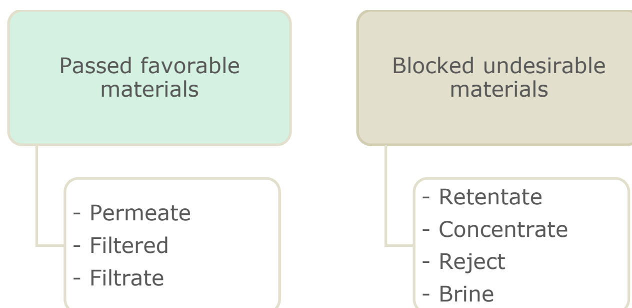

Figure 1 Membrane filtration by-products [5]

The symbolic process sketch of a membrane package is shown in Figure 1 [6]. The primary and general mass balance of a membrane is reported below equation [6].

$$Qf.Cf = QrCr + QpCp$$

In which, Q and C stand for flow rate and target material concentration in the feed, plus, f, c, and p suffixes stand for feed, reject and permeate water (Figure 2).

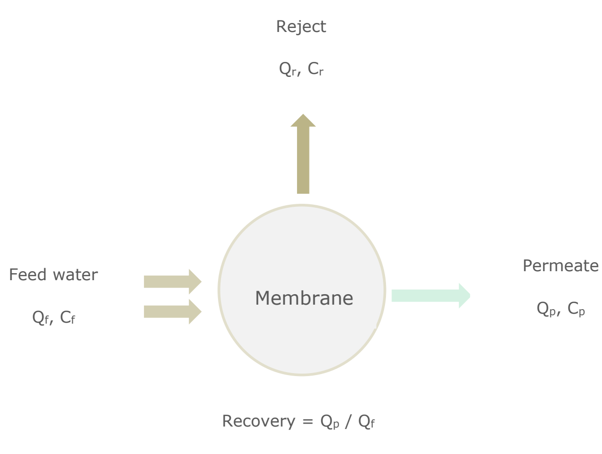

Figure 2 Membrane filtration schematic [6]

2.1.1 Membrane Operation Flow Modes

The operation flow mode of the membrane can be categorized into two modes of dead-end and cross-flow. In the dead-end mode (or batch mode), the feedwater flow is perpendicular to the membrane surface and rejected materials would be reduced by forming a cake layer on the membrane surface. On the other hand, in the cross-flow mode, the feed water moves in a diverging direction, and rejected materials can be collected from the identical side [7]. Figure 3 can illustrate the difference of modes schematically.

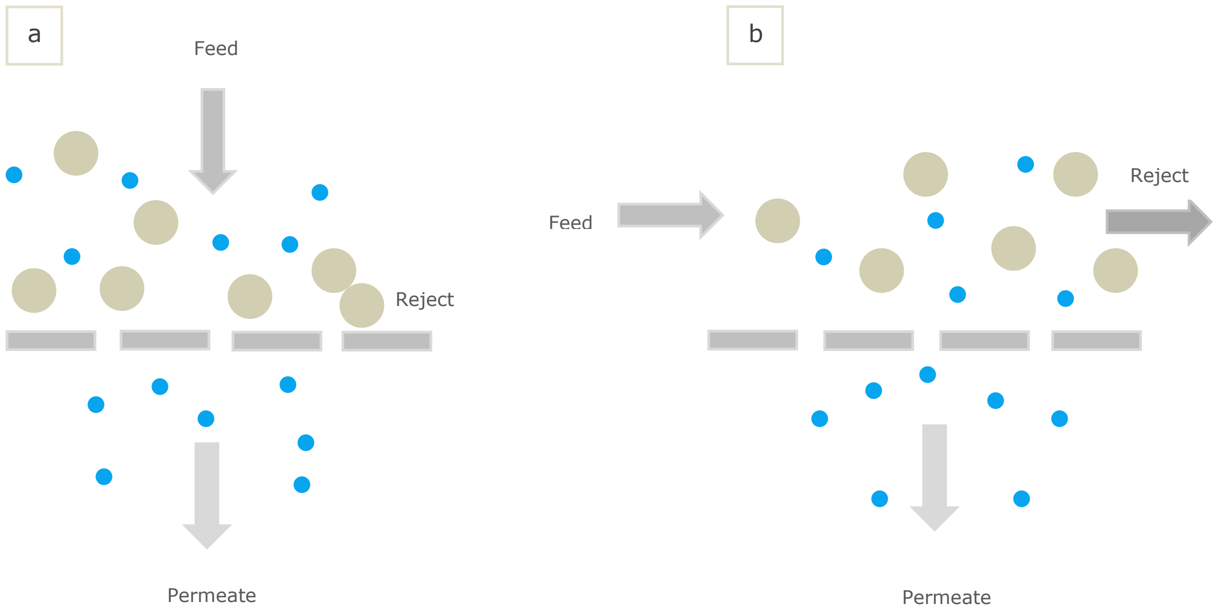

Figure 3 Membrane filtration operation flow modes, a) Dead-end mode b) Cross-flow mode [7]

2.1.2 Membrane Configurations

In the case of performing the separation process by membrane packages in a full-scale plant, there would be a need for a high level of an area. The needed area would be occupied by membranes that need to be linked together. It is the point that membrane modules can optimize the membrane packages and linkages in a high level of performance. There exist different kinds of modules applied in different processes with variable costs. To name a few, they can be classified as Figure 4 [7] and shown in Figure 5.

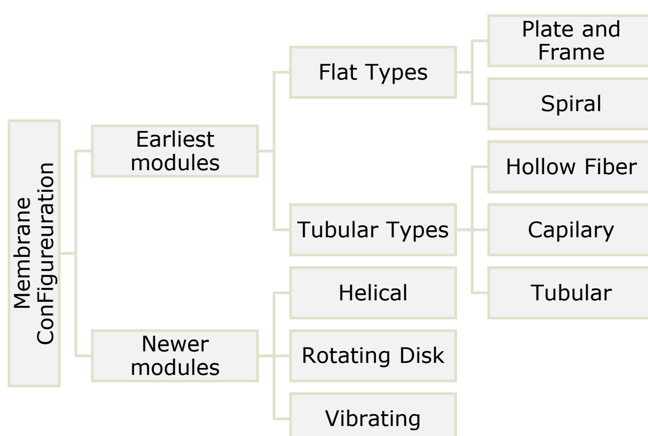

Figure 4 The classification of membrane configurations [7]


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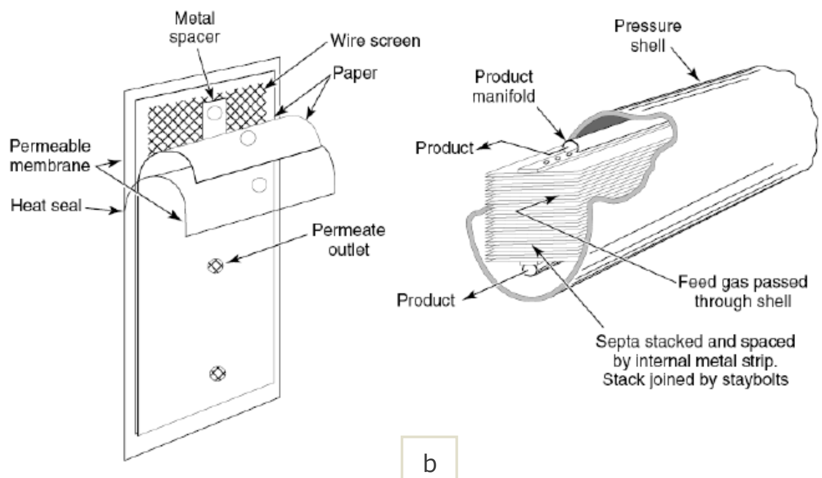

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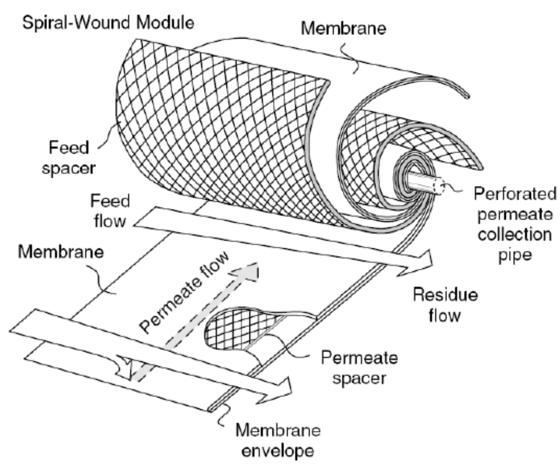

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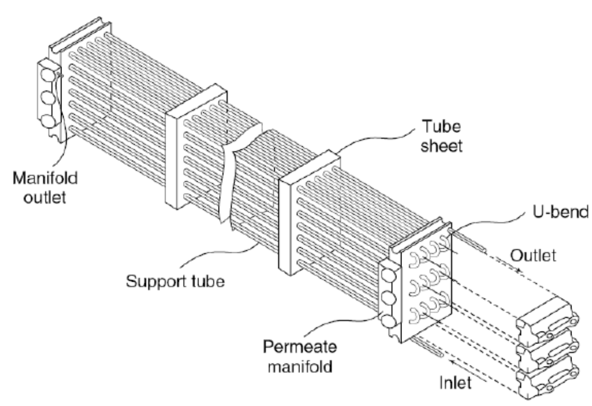

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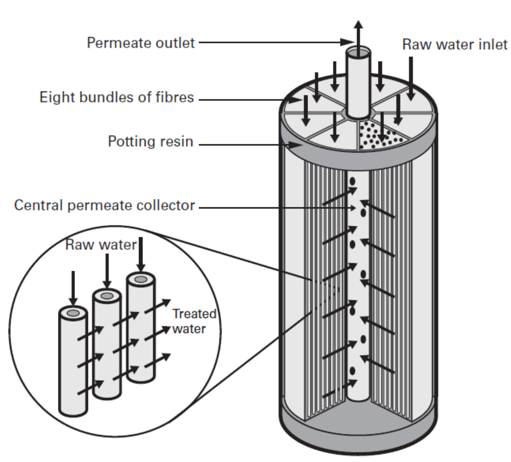

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Figure 5 Widespread membrane configurations; a) General textures shapes and commercial view of the membrane system, b) plate and frame, c) spiral wound, d) tubular and d) hollow-fiber schematic operation diagrams [8][7][9]

Table 1 compares the most important indicators to assess the right choice of membrane configuration. In the table, the advantageous and disadvantageous features are symbolized as + and -, respectively. Most of the indicators will be defined and discussed in further sections [10].

Table 1 The assessment of different membrane configurations [10]

Membrane conFigureuration	Indicator				
	Pre-treatment	Recovery rate	Cleaning scenario	Module size	Cost
Tubular	+++	+	+	-	-
Hollow-fiber	+	+++	+++	++	+++
Plate-frame	+	+	-	+	+
Spiral wound	-	++	+	++	+++

2.1.3 Membrane parameters

To understand further explanations of the membrane technology, it is useful to describe some permeation parameters briefly as below.

Molecular Weight Cut-Off (MWCO) is one of the most widespread assessment parameters in membrane technologies that demonstrate the permeation capability of a membrane. It is described as the molecular weight of the solute in which the membrane can filter 90% of the solute. MWCO relation with the pore size follows a logarithmic trend and the unit of MWCO is Da (Dalton or unified atomic mass unit). Da is a mass unit that is equal to the mass of the subject in one mole (g/mole) in general speaking [11].

Log removal value (LRV) is another assessment parameter in membrane technology to evaluate the capability of a membrane to filter pathogen microorganisms specifically. The logarithmic equation is as;

$$LRV = Log\ (Feed\ pathogen\ concentration)/(Permeate\ pathogen\ concentration)$$

As it can be realized from the LRV equation, if the membrane removes 90% of pathogens, the LRV is 1 [12].

2.2 Membrane Types

In this section, the membrane can be categorized based on the membrane driving force for the filtration. There are different membranes by this classification listed in Table 2 [5].

Table 2 Different membrane types [5]

Membrane system	Driving force gradient
Micro-filtration (MF)	Hydrostatic pressure
Ultra-filtration (UF)	
Nano-filtration (NF)	
Reverse osmosis (RO)	
Membrane distillation (MD)	Vapor pressure by - Temperature - Downstream
Pervaporation (PV)	Vapor pressure by - Downstream

Electrodialysis (ED)	Electric potential
Reverse Electrodialysis (RED)	
Osmotic distillation (OD)	Vapor pressure by - Concentration
Forward osmosis (FO)	Concentration
Pressure retarded osmosis (PRO)	

2.2.1 Pressure-driven Membranes

As the focus of the study is water purification, therefore, pressure-driven membranes are the focus of further discussions.

Regarding pressure-driven membranes, in the first place, considering the extent of pressure which membranes need to operate, they can be classified as two groups of low-pressure membranes include MF and UF, and high-pressure membranes include NF and RO. Then, pressure-driven membranes are distinguished based on the permeated material size and are not normally related to the membrane pore size [15].

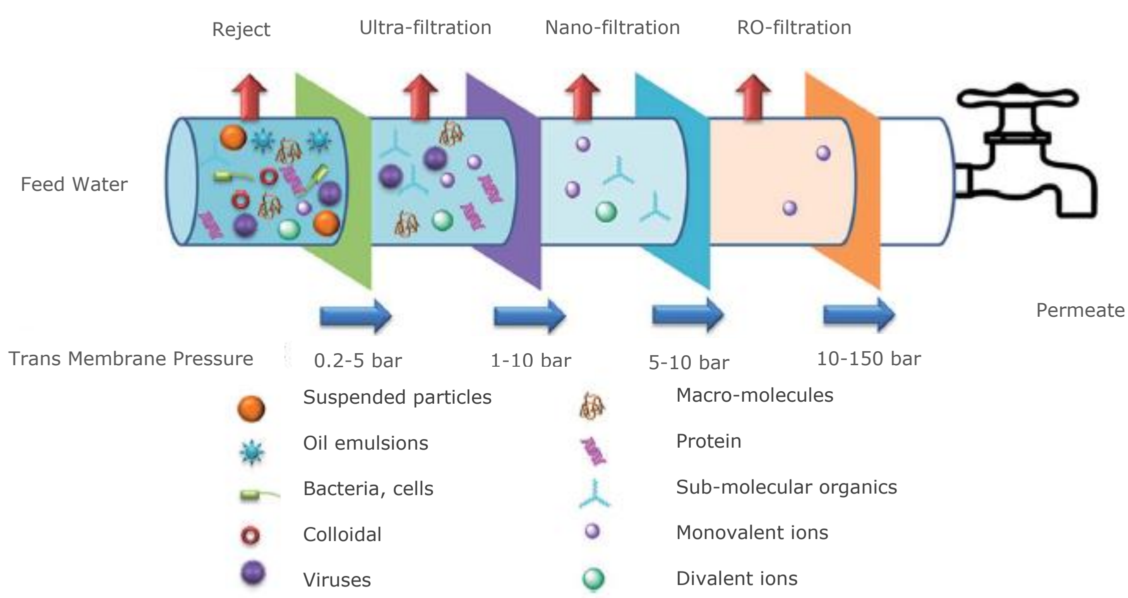

Figure 6 Pressure-driven membranes [13]

To find out the exact materials and particle sizes for passing the membrane and the needed pressure of each membrane filtration approach, Figure 6 can illustrate the differences of methods schematically [4].

Table 3 summarizes the characteristics of four membrane types for better comparison [7].

Table 3 Performance level of pressure-driven membranes [7]

Parameter	Micro-filtration	Ultra-filtration	Nano-filtration	RO-filtration
Pore size (µm)	0.1-10	0.001-.0.1	0.0005-0.002	< 0.0005
MWCO (Da) (90% salt rejection)	> $5*10^5$	2-$5*10^5$	$5*10^2$-$2*10^3$	< $5*10^2$
Pressure (bar)	0.2-5	1-10	5-10	10-150

Removal targets	- Suspended and colloidal particles - Bacteria	- Viruses - Micro molecules - Proteins	- Sub-molecular organics - Divalent ions - High molecular weight molecules	- Monovalent ions - Low molecular weight molecules
Separation mechanism	- Sieving (Based on particle size)	- Sieving (Based on particle size)	- Sieving - Diffusion - Donnan exclusion	- Diffusion
Type	Symmetric - Polymeric - Ceramic	Asymmetric - Polymer composite - Ceramic	Asymmetric - Polymer composite - Ceramic	-

Low-pressure Membranes include MF and UF membranes.

MF Membranes besides UF membranes are the most applicable membranes in water purification. Pore sizes of MF membranes are in the range of 0.1-10 µm. MF membrane application is to be placed followed by sand or cartridge sieve in the treatment process in both dead-end and cross-flow configurations. Also, in the RO filtration system, MF needs to be placed as a pretreatment filter. MF membranes (Figure 7) are significantly porous and the pore size dispersal is notably narrow.

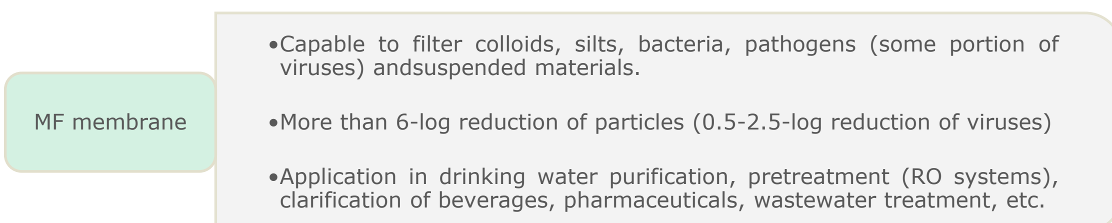

Figure 7 Performance of MF membranes [14][6][4]

UF Membranes are one the most widespread membranes condemned infiltration processes (Figure 8). Pore sizes of UF membranes are in the range of 0.001-.0.1 µm. In this way, UF membranes are capable to filter macromolecules of organic materials [13]. Although MF and UF follow the same operational and process regulations, more asymmetric UF membranes have higher hydrodynamic steadiness compared to MF membranes [15] [16]. The associated costs of UF operation compared to the conventional methods have been estimated about three times more for 1000 m^3 treated water [15].

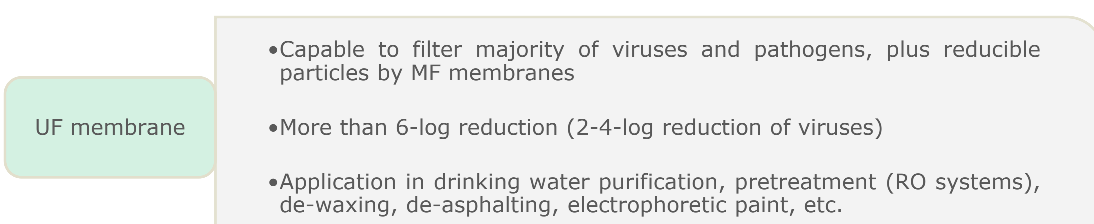

Figure 8 Performance of UF membranes [6][4][5]

Regarding MF and UF Structural and Configuration characters, as the main focus of this study is on MF and UF membranes, there is the need to know more details about their differences other than the pore size and target material for removal. By comparing the MF and UF membranes, it can be realized that the upper layer of UF membranes is denser with a lower level of pore sizes and porosity compared to MF membranes. The denser upper layer of UF can bring about to have better hydrodynamic resistance.

MF and UF membranes can be applied in both dead-end and cross-flow configurations based on the scale of the process. However, the dead-end mode is more common towards MF/UF membranes. The reason is that in the cross-flow mode, there is the need for high flow to mitigate fouling which in this case it demands higher energy for filtration [4].

High-pressure Membranes include NF and RO membranes.

NF Membranes have a pore size in the range of 0.0005-0.002 μm [4]. NF membranes are capable to filter large organics (mostly multivalent ions than monovalent ions), hardness, turbidity, microorganisms, and some portion of dissolved salts. Regarding salt filtration, an example of NaCl filtration by different membranes can clarify the matter better. In this way, NF membranes can filter NaCl salt in the range of 20-80%. The filtration progress for RO can be above 90%, however, compared to UF membranes with the filtration degree of lower than 5%, NF membranes (Figure 9) show good performance in this matter [6][4][5].

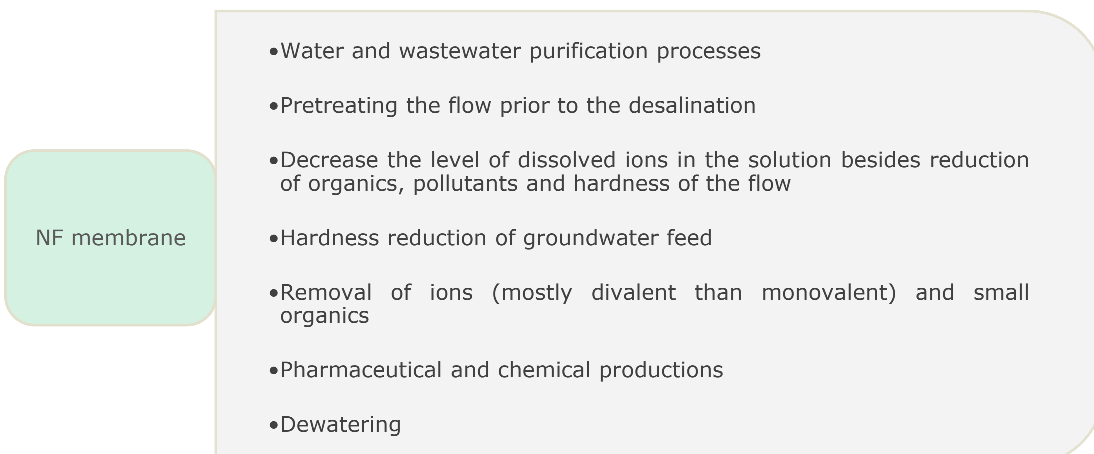

Figure 9 Performance of NF membranes [6][4][5]

Regarding RO Membranes The osmotic pressure is a natural transfer of the solvent through the membrane from a low concentrated solution to a higher concentrated solution, based on decreasing the Gibbs free energy as the driving force.

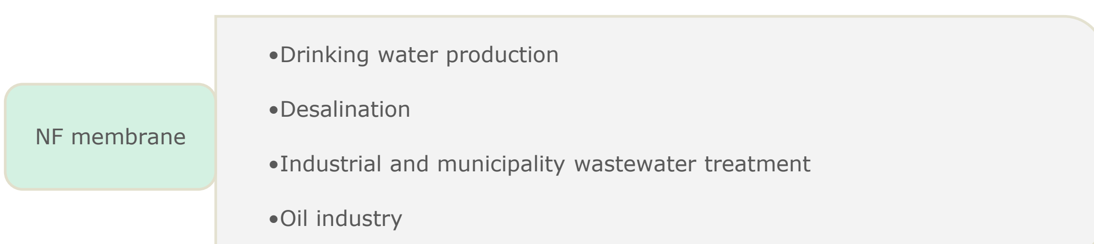

Figure 10 7 Performance of MF membranes [6][4][5]

In the reverse osmosis (RO) membranes (Figure 10), it is possible to filter particles by prevailing over the osmotic pressure and consequence backward movement than the natural one. RO membranes have a pore size less than 0.0005 μm. RO membranes can eliminate dissolved materials, monovalent ions, organics, pharmaceuticals, pollution compositions from cosmetics, resins, plastics, etc., also they can efficiently reduce monovalent ions [6][4][5].

2.3 Hybrid Membrane Systems

2.3.1 Membrane Bio-reactor (MBR)

There exist numerous reasons that recycling water is demanded, for instance, economical interests, environmental issues, water scarcity, etc. The mentioned reasons can justify the reuse of water released from irrigation, household wastewater, industrial wastewater, etc. Membrane bioreactors are the response to purify water by using membranes and other processes in form of a reactor. Advantages of MBR are being novel and economical, with the not-complex operation, and able to perform one-step treatment in a compact area.

As it seems from the name of MBR, the system includes biological sludge process by biomass and MF/UF membranes for biodegrading of wastes and treated water separation, respectively. In the MBR method, membranes are replaced to sedimentation step of the conventional biological process leading to produce high-quality water [16]. One of the applications of the hybrid coordination of MF/UF membranes is in the membrane bioreactor (MBR) using in wastewater treatment [17].

2.3.2 Electrodialysis Membranes

The electrodialysis process includes the reduction of materials by the assist of their different potential charges. The system includes membranes that filter materials when they want to join the opposite pole leading to treat feed water. There exist several applications of Electrodialysis such as desalination, protein separation, etc. Electrodialysis has been applied in different processes since the 1950s, however, it is steel under more attention and development in recent years, as it is reported the Electrodialysis with UF/NF membranes can perform the separation with significant efficiency [18].

2.3.3 Photocatalytic Membrane Reactors (PMR)

Photocatalysis is a science regards to assess advanced oxidation processes (AOP) by using a photocatalyst, a semiconductor, and benefit from its activation under the irradiation of light. Besides numerous advantages of photocatalysis which make it one of the top-charted pioneer methods to apply in different processes, it is capable of coupling with other physical and chemical methods.

The benefit of adding membrane technology to photocatalysis is to solve photocatalyst regeneration. In this way, it can lead to performing the process continues as far as used photocatalysts are regenerated by membranes and enter in the photocatalytic reaction. The coupled package is named a photocatalytic membrane reactor (PMR). In the face of water purification, it is proved that photocatalysis is promised method to degrade a wide range of pollutants efficiently. Thus, PMR can more effectively produce high-quality water by applying both photocatalysis and MF/UF/NF technologies [19].

2.4 Specific Separation Targets

The specific substance's regulation comes from the tighter regulations toward some evolved materials in water resources and the need for distinctive separation processes for them.

Membrane technologies can address the reduction of special cases of materials listed in Table 4.

Table 4 Membrane technologies for specific material separation [4]

Material		Proposed solution by membrane technologies
Arsenic	- Organic and inorganic arsenic components - WHO criteria less than 0.01 mg/l - Higher exposure leads to cancer	Separation mostly by NF membranes
Pesticides	- Release in natural water due to the inevitable use in the agricultural industry	Separation by NF membranes effectively depends on the Mw
Disinfector By-product (DBP)	- Disinfectors using for biological neutralization in water treatment react with NOM existed in natural water leading to DBP - Harmful for consumers	Membranes can be arranged and controlled to remove NOM efficiently and prevent undesirable DBP
Pharmaceuticals	- Release from different sources to natural water would have impacted the drinking water quality	Membrane systems coupled with conventional methods (ozonation and carbon active) to remove pharmaceuticals efficiently

2.5 Membrane Characterization

Membranes' compositions can include a wide range of materials. The most available and common approach is to use both single and blend of different stable polymers. In addition, ceramics, glasses, etc. are used in membrane production. Membranes can be fabricated, amended, and enhanced from mentioned materials by different methods listed in Table 11.

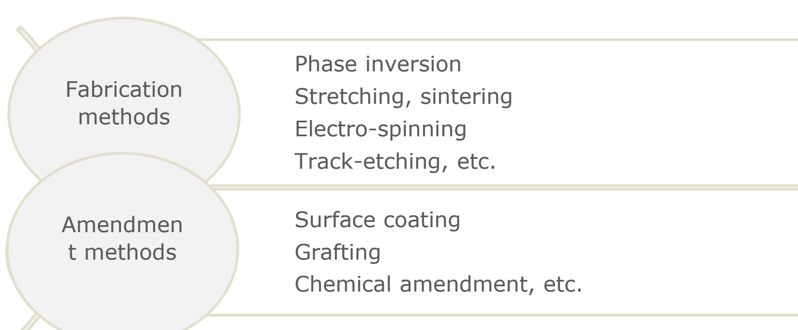

Figure 11 Fabrication and amendment methods of low-pressure driven membranes [5]

In this section, an overview of materials applied in the membrane preparation is described. The goal of this is to know more information regards to the membrane material and the future destiny of the membrane coupled with its material (e.g. membrane failure).

2.5.1 Polymer-based Membranes

There exist three different characterization approaches in this venue as below.

Mixed Matrix Membranes include mixing materials and methods to enhance the performance and steadiness by composing secondary materials to the membrane matrix. Regarding the mentioned mixing materials, polymers and nano-materials can be implemented in this matter. The final membrane would be considered as hybrid membranes with different performances depending on the application, target pollution, etc. There exist different classifications for mixed-media membrane preparing [1].

Polymer blending is the most common method by adding additives that can alter the membrane features. The main reason to use this method is the polarity and stability of the membrane. Polymers like PVDF and PES have enough steadiness but vulnerable to fouling and lower permeate because of their hydrophobicity characters. In contrast, PVA and PAN are vice versa. Therefore, blending the materials can result in the design of an efficient and well-steady composite membrane [21]. Polymeric UF membranes originated from Polyvinylidene difluoride (PVDF) has a significant capacity to reduce target pollutions in the drinking water purification process [12].

Regarding Thin-film Composite Membranes, it needs to mention the membrane porosity and steadiness. Membranes with less porosity, named as dense membranes, have a lower level of permeate but higher selective character and vice versa. For enhancing the porosity of a dense membrane and increase the permeate rate, it is needed to decrease its thickness. However, that action can compromise the physical steadiness of the membrane. Thus, Thin-film composite (TFC) membranes that include two layers can be introduced. TFC is made of a crest layer of the dense selective part and a porous sub-layer as the support. In this way, the upper part carries the high permeation and selectivity responsibility, whereas, the sub-layer addresses the physical and chemical steadiness of the membrane [1].

Nano-improved Membranes are in accordance to enhance the membrane technology efficacy in a way that inorganic nanoparticles can be used in the membrane polymer matrix. In this venue, varieties of nanoparticles have been implemented to prepare nano-improved membranes listed in Table 5 besides the bold impacts of each type of nano-improved materials on the membrane. Based on the reported features of the membrane impacted by nano-tubes, there are increases in performance and flux and also anti-fouling characteristics due to the higher hydrophilic surface of membranes.

Table 5 Nano-improved polymer-based membranes [1][22][23]

Nano-improved membrane	Feature
Carbon Nanotubes (CNTs) Single-walled (SW) Double-walled (DW) Multi-walled (MW)	More hydrophilic and anti-fouling High surface area Higher chemical stability Higher antibacterial properties
Clay Nanoparticles Unmodified montmorillonites (Na_MMT) Cloisite grades (modified MMT)	Higher performance Hydrophilic features of layered silicate Higher thermal stability
Silver Nanoparticles	Strong bacteriostatic nature of silver ions Environmentally friendly Non-toxic and non-allergic Enhanced virus reduction
TiO_2 Nanoparticles	High stability Commercially cheap More hydrophilic and anti-fouling High porosity Photocatalytic bacteriostatic features
ZnO Nanoparticles	Anti-bacterial and bacteriostatic Cheaper than TiO_2 and Al_2O_3 membranes More hydrophilic Higher stability Anti-fouling surface and in-pores
Al_2O_3 Nanoparticles	High stability in severe conditions More hydrophilic Anti-fouling

Fe_3O_4 Nanoparticles	Paramagnetic features Higher stability More hydrophilic
SiO_2 nanoparticles	More hydrophilic High stability
ZrO_2 Nanoparticles	High stability than TiO_2 and Al_2O_3 membranes More hydrophilic High surface porosity

2.5.2 Nanostructured Membranes

The classification of membranes is based on the size of particles that can be rejected by membranes, not the pore size in the membrane structure. By that definition, most NF and UF which can reject particles by the diameter of 1-100 nm can be categorized as nanostructured membranes.

In this venue, adding a different amount of another agent into the casting solution of the membrane or/and applying specific preparing methods may result in a UF membrane with modified morphology and performance. The final prepared membrane would achieve nanostructured pores with enhanced hydrophilic character, physical steadiness, permeate and rejection rate, and antifouling feature [24].

2.5.3 Bio Membranes

Natural phenomena can be inspirations to make the technology more efficient and address the difficulties of current processes. In this venue, biomaterials (biopolymers) can act as an applicable agent in membrane preparation with enhanced characters. Another merit of using biopolymers in this matter is to condemn almost natural conditions of preparing such as aqueous media, regulated temperature and pressure, and natural pH. For instance, Membranes consist of an upper layer with inspired-bio-channels into the membrane matrix-assisted by the hollow fiber polymeric support can propose the nano-scale porous membrane technology.

In this way, bio-structured membranes have issued a wide range of applications with a high level of permeate rate and antifouling character in the water purification process. Besides the separation features, there exist interesting characteristics to study such as self-cleaning and –healing followed by bio-structured membranes [25].

2.5.4 Ceramic Membranes

By comparing the capital cost of polymeric membranes and ceramic membranes, the lateral ones need greater investment. However, by the start of the operation, MF and UF ceramic membranes show more mechanical steadiness and higher fouling resistance. The organic essence of the ceramic-based membrane provides the hydrophilic character form of them which is favorable to be anti-fouling. Also, in the case of the cleaning procedure, ceramic membranes need a more simple cleaning operation since they can face strong chemicals with high temperatures without dropping the performance. Therefore, in the general view, ceramic membranes have lower costs of operation. As a result, they can stand as a rival substitute for polymeric membranes [17].

Ceramic Membranes for the water purification Process are not widely used. All mentioned superior operational advantages led to the use of MF and UF ceramic membranes in many processes such as enzyme purification, fermentation, juice clarification, and oily wastewater. However, they are not nominated and developed for water purification due to the higher capital cost of production compared to polymeric membranes.

Followed by the minor share of ceramic membranes in water treatment, they would be demanded based on the feed wastewater or water that contains oil or strong chemicals. In this way, the use of ceramic membranes in a harsh environment can overweigh the high initial costs compared to the polymeric membranes which are sensitive towards strong chemicals and fouling [17].

3. MF/UF MEMBRANES IN WATER PURIFICATION

3.1 MF/UF System and Market

3.1.1 Package System

Using MF and UF membranes in the water purification process is a turning point as the technology has commercialized since the 60s [17]. The MF or UF package system normally includes MF or UF membrane modules' attachments. Membrane fibers form a unit module, and multi modules are supported and connected to shape an integrated unit. Units of MF or UF membranes are fixed together to respond to the demanded flux.

Membrane separation technology has achieved attention based on issuing new tighten water regulations which demand higher standard criteria for drinking water. Thus, the superior membrane separation technology found its place as a substitute for adsorption, sand sieving, and ion exchange possesses in water purification, besides its other applications in food, beverage, biotechnology, etc. [4]. In terms of the drinking water purification process and MF/UF membranes, the membrane technology can be used in the below sections [17].

3.1.2 Pretreatment for MF/UF

Normally, if the feed water carries a high level of solids, the dissolved air flotation (DAF) or clarification step can be placed before MF/UF membranes to decrease operational costs. Generally, there is no strict pretreatment needed for MF/UF membranes to achieve the highest available water quality, however, to address fouling issues, different pretreatment scenarios are investigated in other sections [6].

3.1.3 MF/UF Membrane Market

As an estimation to perceive the global market investing for MF and UF membranes, it is reported that each year there would be an increase of 10% for the market for MF membranes, while it was recorded as 1.6 billion dollars in 2013. Also, for the UF membrane, only the US market recorded 1.2 billion dollars in 2015 with rising of 5.7% annually [13]. The growing market of cross-flow membranes is predicted to reach 15.9 billion dollars in 2021 [26].

3.2 Sustainable Membrane Technology in Water Treatment

The general full-scale conventional process of water purification consists of initial screening, chemical addition, coagulation/flocculation/sedimentation, fine and natural sieving, disinfector addition, and then storage (Figure 13).

Based on Figure 13, at the first, the coarse sieve can reduce large particles. Then the water would be exposed to chlorine as a disinfector. After that, water needs to be clarified by flocculation and hardness removal (lime can be used). Again the sand media sieve can filter the water to make the water ready for pH regulation to decrease the alkalinity. Chemicals can be added to water to play a role in preventing scaling in water systems.

Then, activated carbon filters or sodium bisulfate would eliminate free chlorine. In the next step, UV radiation is condemned to sterilize water. Finally, cartridge sieves can reduce suspended materials. In non-conventional treatment, MF/UF membrane technologies can be participated in the process differently listed in Figure 12.

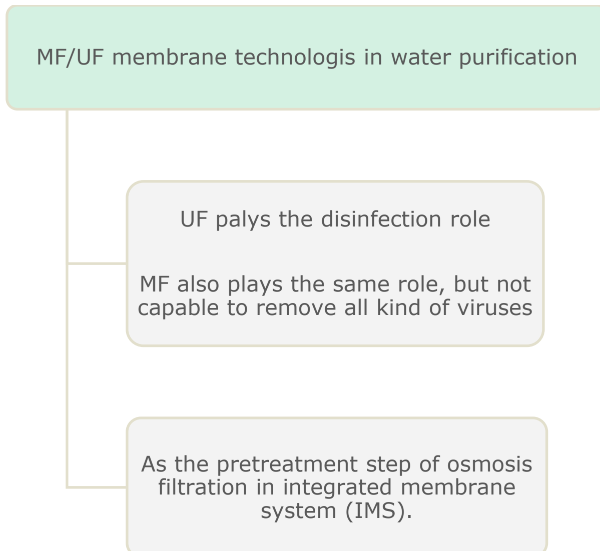

Figure 12 MF/UF in the water purification process [4]

3.2.1 Integrated Membrane System (IMS)

Based on the membrane properties and operation parameters, MF/UF membranes are efficiently up to filter organics, suspended particles, and colloids, and they let the dissolved particles pass the filter. In contrast, NF and RO membranes have efficient performance for filtering dissolved particles and cannot stand for colloids filtration. Although materials that can be filtered by an MF or UF membrane are filterable by NF and RO, it is not a good approach to filter all materials with different sizes with a sole type of membrane such as RO. As NF and RO membranes are hesitant against colloidal and large materials, MF/UF can be as pretreatment steps before NF membranes to preserve NF performance and fouling control [6].

In other words, the membrane technology consists of different types of membranes to filter each kind of target materials part by part with different pore sizes of MF, UF, NF, and RO membranes. In this way, since the NF and RO membranes are so delicate towards fouling by larger materials, MF and UF membranes can be condemned before increase efficiency. The designing of the configuration of different membranes to minimize fouling effects is famous as IMS (Figure 14) [4].

The justification reason of the IMS for the separation is to design a low-pressure membrane system first and then completing the treating process by high-pressure membranes. If in any case there would be only a possibility to use low-pressure membranes, the dissolved materials of the feed should be transformed to colloids before MF/UF membranes [6].

Figures 13 and 14 illustrate a general process flow for surface water purification plants in both conventional and membrane systems. It should be considered in each approach steps can be different (other or fewer steps need) based on the feed water quality (river, ground, etc. feed) or demanded water quality requirements. Plus, even in some plants there would be a need for coagulation/flocculation for the membrane system too, which this study investigates in further sections. Therefore, the diagram is just a general simplified scheme.

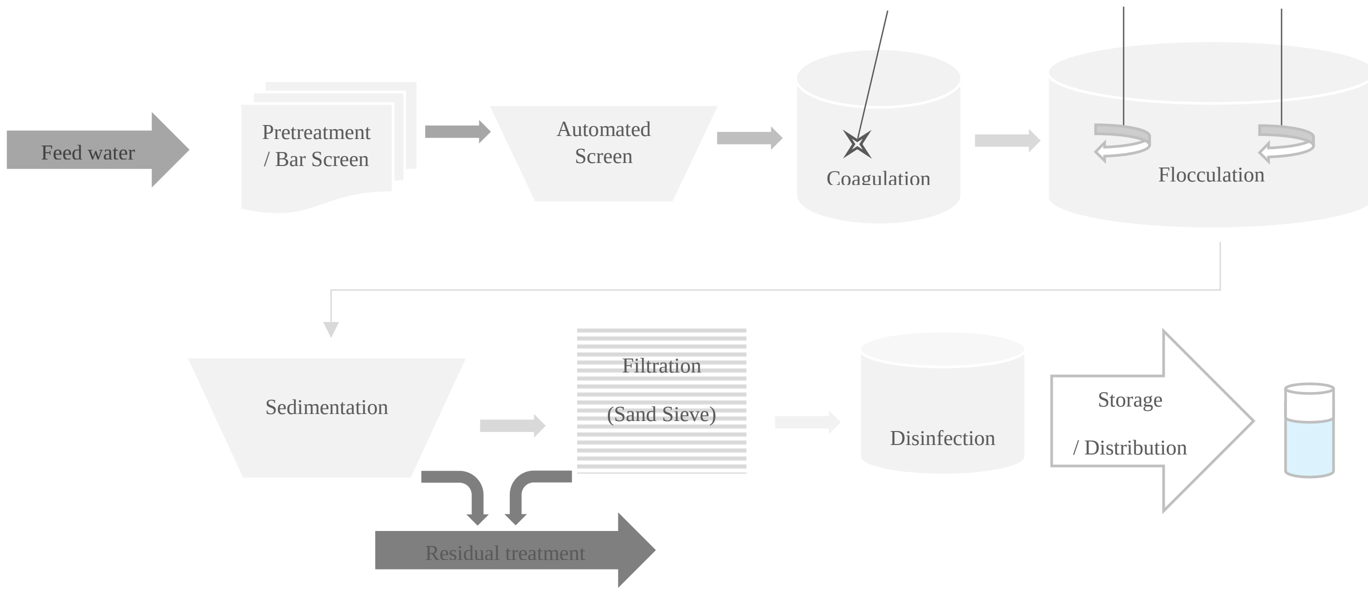

Figure. 13 Schematic of conventional drinking water purification process [4]

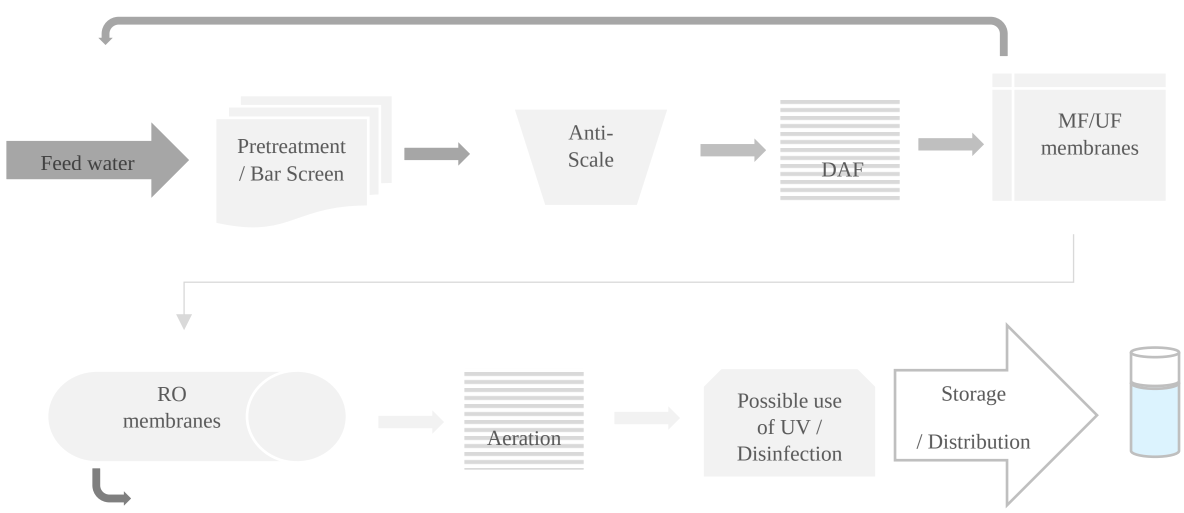

Figure. 14 Schematic of Membrane system in drinking water purification process [4]

In this way, in comparison to the conventional method of water purification, IMS has major differences listed in Figure 15.

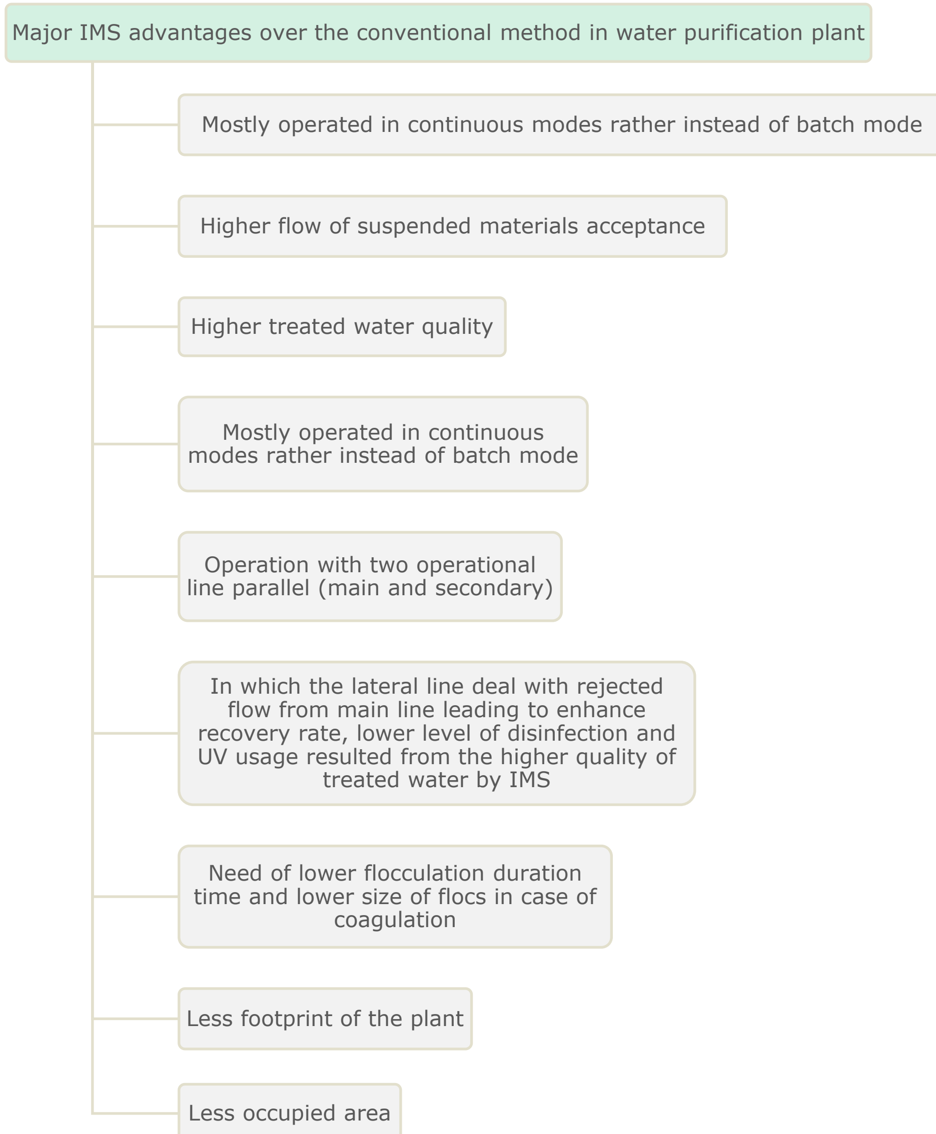

Figure 15 IMS advantages over the conventional water treatment process [16]

Regarding the design of IMS, it has so tendency to feed water characterizations, for instance, the flow rate of suspended materials, expected treated water quality and quantity, site-specific circumstances, etc. As a case in point, one of the important decisions which can be made in the pre-assessment of IMS design is the membrane configurations (Figure 16).

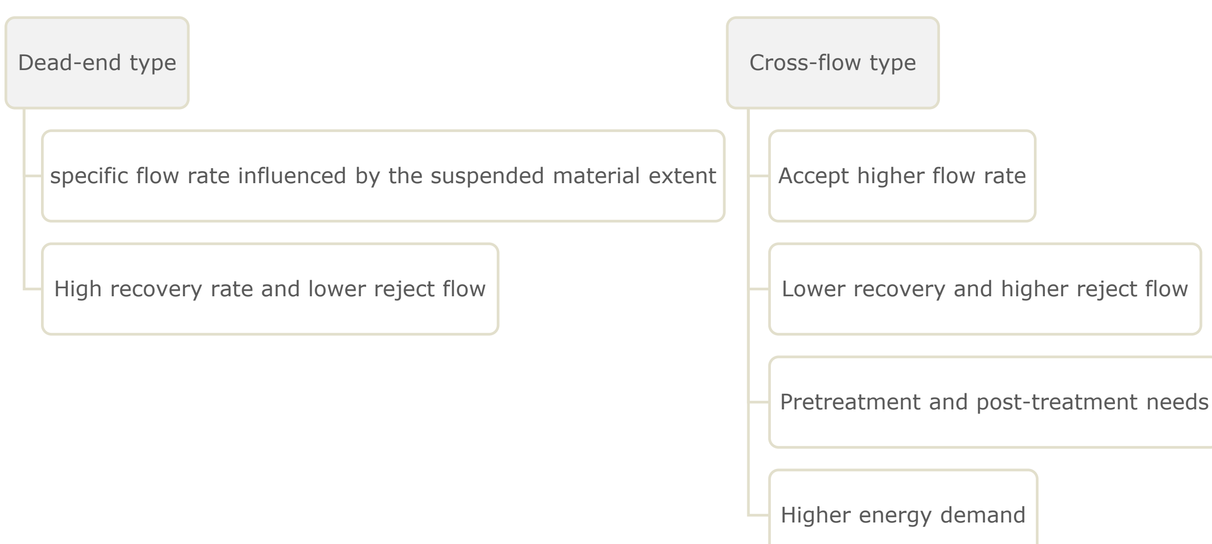

Figure 16 Performance of IMS with different membrane configurations [6]

3.2.2 Sole MF/UF System

Low-pressure membranes, MF/UF can be an efficient substitute for coagulation, flocculation, and sedimentation, and even also suspend the use of chemical disinfectors in the conventional water purification process. By removing suspended and colloidal materials, bacteria, and viruses, high-quality water can be produced with fair capital cost [16]. MF/UF membranes can treat the water by introducing the filtered water with low turbidity and a safe level of microorganisms, NOM, chemicals, bacteria, and viruses.

Figure 17 shows hollow fiber type UF membrane modules manufactured by Lenntech with the maximum capacity of 100 m^3/h. The spec data of the mentioned UF system is listed in Table 6.

Figure 17 UF membrane modules manufactured by Lenntech [10]

Table 6 Spec data of the UF membrane modules manufactured by Lenntech [10]

Parameter	Surface feed water	Ground feed water	Sea feed water	Unit
Flux	45	75	60	lmh (l/membrane surface area/h)
Turbidity	< 50	< 5	< 20	NTU
Recovery/day	85	96	94	%

Therefore, MF/UF membranes can be used as a sole system or as part of the treatment process (IMS) based on the feedwater characteristics. The sole system works when the source of feed water quality is in the status with no need of NF or RO membranes for treatment. However, for drinking water, the output of the treated water needs to be following water regulations towards the limit of suspended solids, micro-organisms, and chemicals [17].

3.2.3 Portable Purification Unit

The portable or are designed for different applications such as recycling of water for remote/small communities which do not have access to the main purified water, and high-tech surviving equipment for hikers and campers, and all in all the situations that human health would be in danger by using unsafe water [17]. The compacted high-tech consecutive membranes (Figure 18) can be shaped in different equipment such as pocket-size straw shapes with or without storage, batch or continuous water jugs and pitcher, etc.

As a case in point, one of the pioneer manufacturers in this industry is LifeStraw Co. illustrated in Figure 18(a). The extreme light products of the company can filter bacteria and parasites up to 99.999999%. The reported life span of filters is roughly 4.5-118 m^3 depends on the product type [27].

Figure 18 Portable water purifier using membrane systems manufactured by a) LifeStraw and b) Vero [27][28]

3.2.4 Decentralized water treatment system

Decentralized water purification is defined as placing the on-site or mobile small-scale treatment plant in where the feed water is supplied and drinking water is demanded. This approach is getting attention and importance due to the need for drinking water access in any far-fetched areas detached from healthy water distribution. The possible applicable hosts and advantages of this approach are listed in Figure 19 [29][30][31].

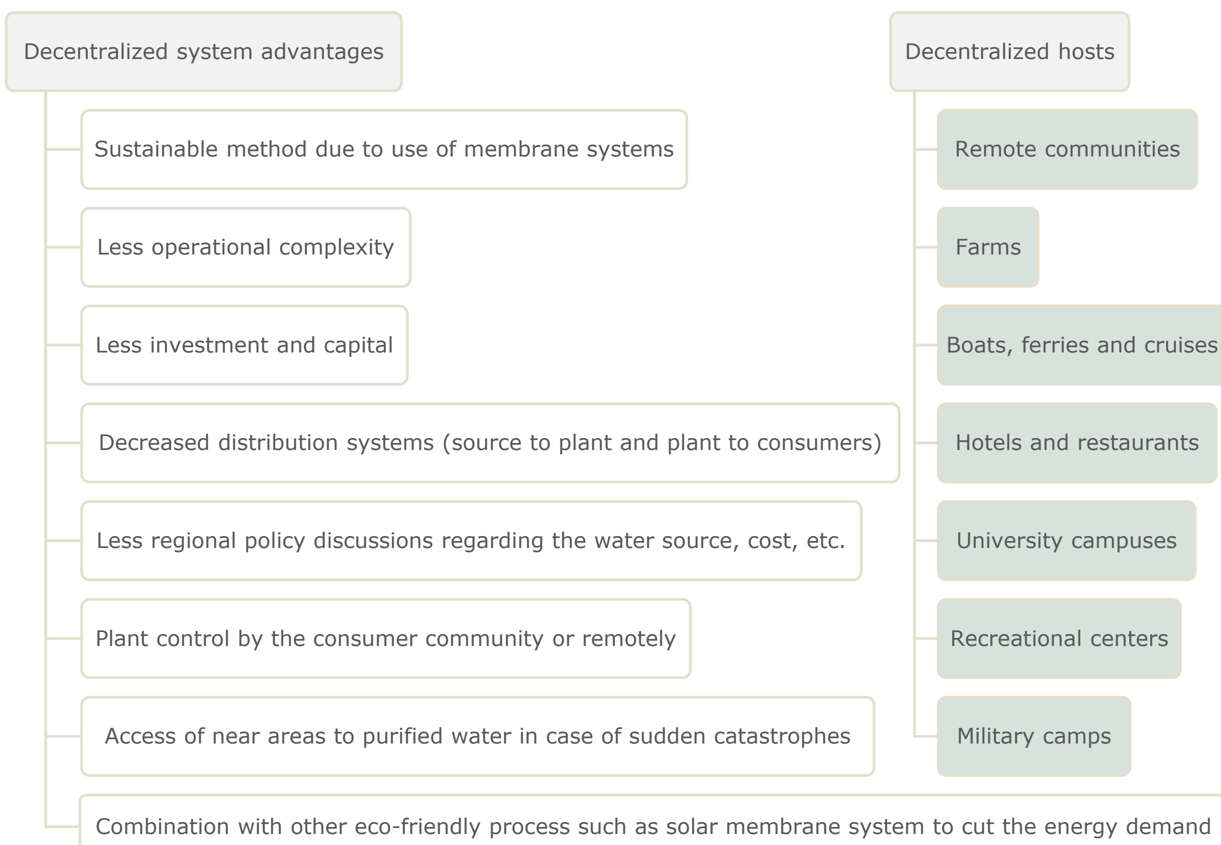

Figure 19 Applicable hosts and advantages of decentralized water treatment units [29][30][31]

There are several pioneer companies to produce portable centralized treatment units benefiting solar power as the source of energy (Figure 20).

Figure. 20 Solar power attached to decentralized membrane in water treatment [32][33]

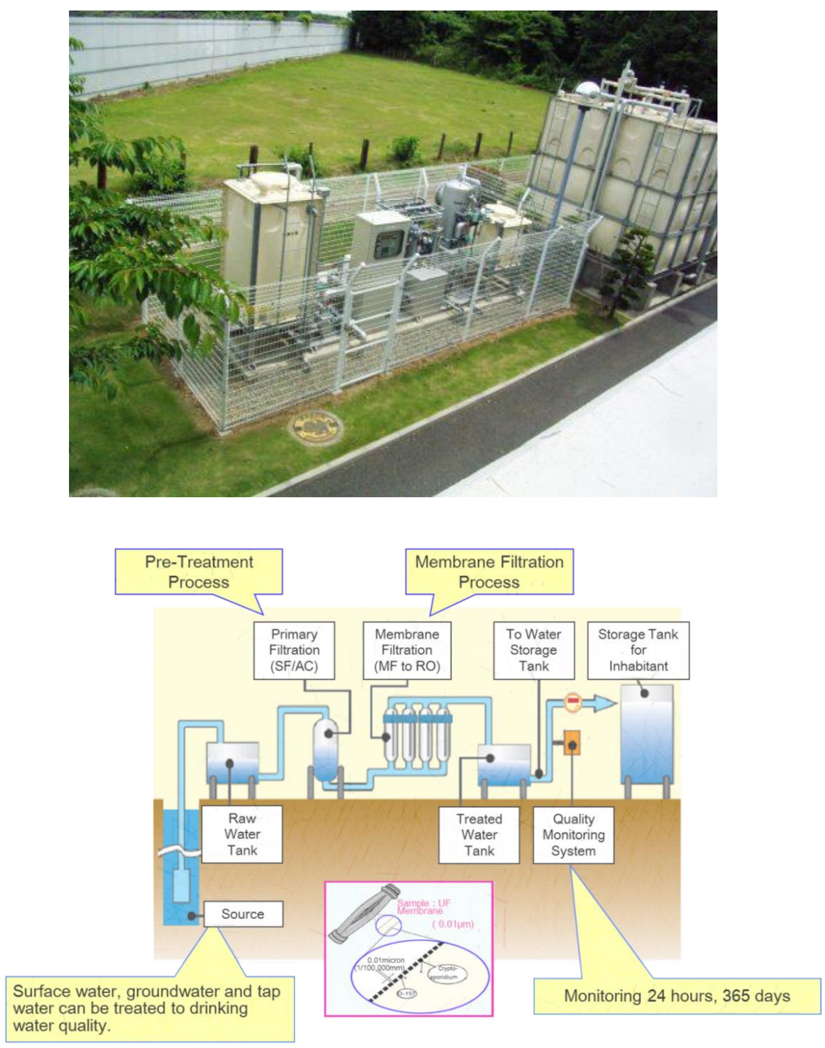

Figure 21 On-site compact membrane systems in decentralized water treatment manufactured by Mitsubishi Chemical Aqua Solutions Co. [29][3]

In this matter, one of the on-site compact membrane systems manufactured by Mitsubishi Chemical Aqua Solutions Co. is worth to be mentioned which has the process diagram in Figure 10. This system is distinguished due to the capability to be monitored, sampled, and controlled fully automatic and remotely. All the operational and performance data can be achieved by the system itself and send to different control rooms to places outside of the site [29].

4. FOULING OF MF/UF MEMBRANES

4.1. MF/UF Technology Science Assessment

To find the obstacle gaps in applying MF/UF membranes, it needs to list and classify possible issues towards the technology. In this way, it is possible to find out where problems are rooted more fundamentally and propose the enhanced mitigation method. In this case, knowing the advantages of the technology would also help for mitigation solutions. Therefore, before explaining several problems facing the MF/UF operation, Table 7 lists the advantages and disadvantages of the science of MF/UF membrane filtration in water treatment.

Table 7 Advantages and disadvantages of the MF/UF membrane technology in water treatment [1][5][6][34][35]

<table>
<tr><th colspan="2">Overall advantages and disadvantages of the MF/UF membrane technology in water treatment</th></tr>
<tr><td rowspan="8">Advantages</td><td>Ongoing novel developments in
• Hybrid membrane materials and structures
• Characterization methods for specific applications
• Hydrodynamic enhancement of membranes
• Different modules and configuration
• Fouling issue</td></tr>
<tr><td>High capacity level to scale-up</td></tr>
<tr><td>Lower relative concentrate for further treatment</td></tr>
<tr><td>Reliable, simple, eco-friendly
• Applicable in different ranges of temperature, pressure, and pH.
• having same output quality in case of pollution, hydraulic and fluctuation changing of feed water
• Expandable modules
• Remote control and monitoring</td></tr>
<tr><td>Low relative capital and energy needed, environmentally friendly, and</td></tr>
<tr><td>Effective removal capability towards a wide range of materials</td></tr>
<tr><td>the lower level of process footprint
• Lower need of chemicals applying
• Single-stage operation
• The lower level of produced sludge
• Less use of post-treatments like UV</td></tr>
<tr><td>The flexible process to combine with other processes such as;
• De-carbonator system for CO2 level decrement and pH control
• Prior clarification
• Prior AOPs to degrade organics
• Before applying dissolved air flotation (DAF)
• Chemicals adding for coagulation, flocculation, pH modifying</td></tr>
<tr><td rowspan="7">Disadvantages</td><td>Fouling issues
• High cleaning resistance
• Low cleaning rate
• High Susceptibility to breach
• Low Infrastructure reliability</td></tr>
<tr><td>High chemical sensitivity</td></tr>
<tr><td>Extending prior applications to prevent fouling</td></tr>
<tr><td>concentration polarization (CP)</td></tr>
<tr><td>Low filtration rate of non-ionized ions</td></tr>
<tr><td>Low filtration rate of dissolved gas</td></tr>
<tr><td>Membrane aging and Failure</td></tr>
</table>

Based on the mentioned disadvantages, further, the study will scrutinize all background reasons and mechanisms leading to failure of the MF/UF membranes.

4.2 Fouling

MF/UF membranes have universal applications in the water purification process. However, fouling as one of the main drawbacks of MF/UF membranes makes it less efficient and takes its application under question [36]. In other words, same as all technologies, it also has obstacles in operation that can limit the application such as fouling. Thus, comprehensive studies have been focused to address the fouling issue [1].

When the permeation transport drag force through the membrane is significantly higher than contrary lift forces, the material accumulation in both the membrane surface and pores is predictable. The importance of this accumulation mechanism, namely as fouling, is the forming resistance to the permeation. Mentioned fouling phenomena followed by material retention and accumulation can happen either in the membrane pores or its surface [37][56][38]. Fouling is one of the main disadvantages of using UF membranes which is the most impacting agent to threaten the membrane performance. [14].

Figure 22 is illustrated how the fouling trend look likes from the standard (reversible form) rate to severe emergency circumstances (irreversible forms). Since fouling agents would root from different sources, schematic molecular forms of colloidal, scaling, organic, inorganic, and bioagents are demonstrated in Figure 23 [14].

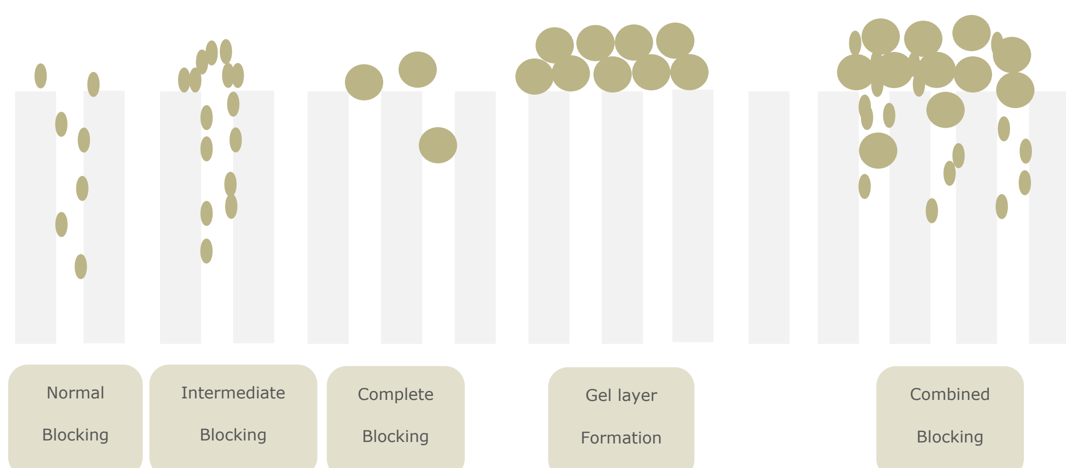

Figure 22 Different membrane fouling type and mechanisms [37][56][38]

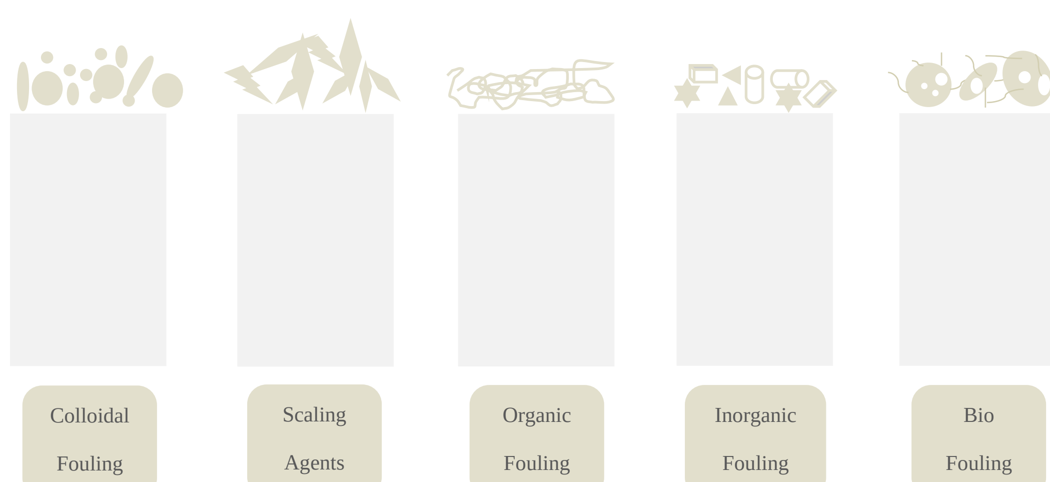

Figure 23 Different fouling source materials and their molecular shapes [14]

Summarized significant fouling consequences can be listed in Figure 24. As it can be seen from the cyclic listed diagram, impacts can be rooted or resulted from other impacts.

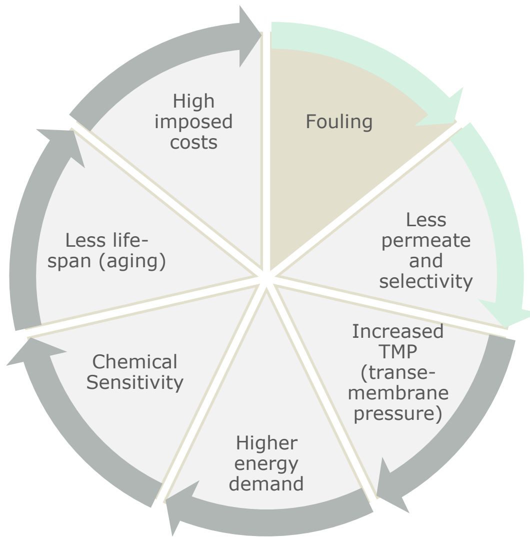

Figure 24 Significant fouling consequences

4.2.1 Fouling Assessment Parameters

Fouling could lead to perform lots of efforts to explore the main reasons for the high operational maintenance in UF filtration and then to propose the mitigation scenarios. In this way, since fouling is the main agent to reduce primary high efficiency, the fouling control methods hold the principal share in the costs [39].

Also, there exist indexes to be measured in membrane performance to that fouling rate can be evaluated in fouling studies. In this way, the mentioned assessment tools for assessing the membrane performance declines are listed in Figure 25.

Fouling Rate	•The fouling progress level which shoes the membrane age
Clean Resistance	•The more demand of pressure for operation due to fouling
Cleaning Rate	•The performance retrieval level of the membrane after cleaning
Susceptibility to Breach	•The vulnerability of the membrane to rupture
Infrastructure Reliability	•The site infrustracturess hazard for the membrane rupture
HAs content	•The level of hydrophilic content of the membrane (being anti-fouling)

Figure 25 Membrane fouling assessment tools [40]

The fouling Rate index can evaluate the resistance increment infiltration periods led by fouling. As a ground fact, it is established that increase in the age of membrane results in the increase of fouling rate. Also, there is an assumption in fouling studies bringing up the reason for the more vulnerability of aged membranes for fouling. The mentioned reason is clarified as aged membranes would lose their hydrophilic characters (loss of the hydrophilic add-ons by aging) and being more hydrophobic leading to be more favorable for fouling [36].

The clean Resistance factor is based on the applied pressure in membrane filtration. MF/UF membranes need low pressure for filtration. On the other hand, an obstacle like fouling or as a general term aging can make the process demand more pressure for achieving the setup permeate. Thus, an increase or decrease in clean membrane resistance index as it is the amount of pressure needed to keep up initial permeate flow, informs the fouling progress in membranes [36].

The cleaning Rate index characterizes the extent of the performance retrieval of the membrane by chemical cleaning. The mentioned index has significant importance in fouling studies. It is due to that the fact that by decreasing the index, the chemical cleaning procedure time increases. Therefore, more needed time results to make the overhaul time more than before. In addition, based on the lower efficiency of chemical cleaning for aged membranes, therefore, longer chemical cleaning and exposure escalates the required chemical dosage [36].

Susceptibility to Breach (rupture) of the membrane due to aging brings the emergency and halts the whole process. It is a fact that an aged membrane can be breached more easily than a less aged one. The mentioned occurrence is not a frequent incident and the vulnerability of the membrane to be ruptured is not a direct performance factor. But, physical characteristic assessment of the membrane can be checked as the substitute of performance factors to assess the membrane breach [36].

Infrastructure Reliability Besides factors related to membrane aging, the extent of the infrastructure's trustworthiness can make a hazardous situation leading to membrane deterioration or breakage. The mentioned assisting agents are not directly related to the membrane composition and the level of trustworthiness of them depends on the manufacturer [36].

Hydrophilicity Add-ons (HAs) Content is important since the more a membrane surface becomes hydrophobic, the greater would engage with fouling. Thus, the fouling study of MF/UF membranes can be assessed by the HAs parameter to evaluate the level of hydrophilicity, and consequently, the five mentioned fouling indexes above [34].

4.3 Concentration Polarization (CP)

In contrast to the fouling resulted from the accumulation of substances on the membrane surface and pores, concentration polarization (CP) is the formation of reduced materials away in bulk by the membrane, near the membrane surface. In this way, another important index affecting the permeate resistance is CP. However, the nature of resistance resulted from CP is different from fouling [7][41].

4.3.1 Negative CP Mechanism

To elaborate the mechanism of the CP, two forms of flow are needed to be considered. Firstly, materials have a convection flow (bulk transport) into the membrane, and secondly, retained substances would follow a diffusion flow back to the bulk. The equilibrium point of those mentioned flows which can reach the steady-state in the first minutes of the filtration characterizes the CP. CP can have an influence on permeate rate followed by Figure 26 [7][41]. Also, the detail of CP position on the membrane surface relative to membrane fouling and forces applied on a filtered particle is shown in Figure 27 [37].

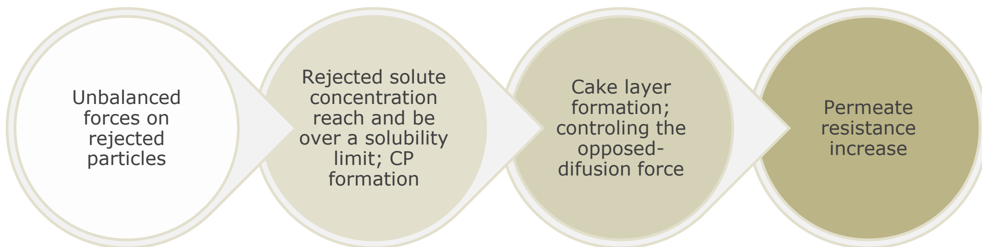

Figure 26 CP influence mechanism on the membrane permeate [7][41]

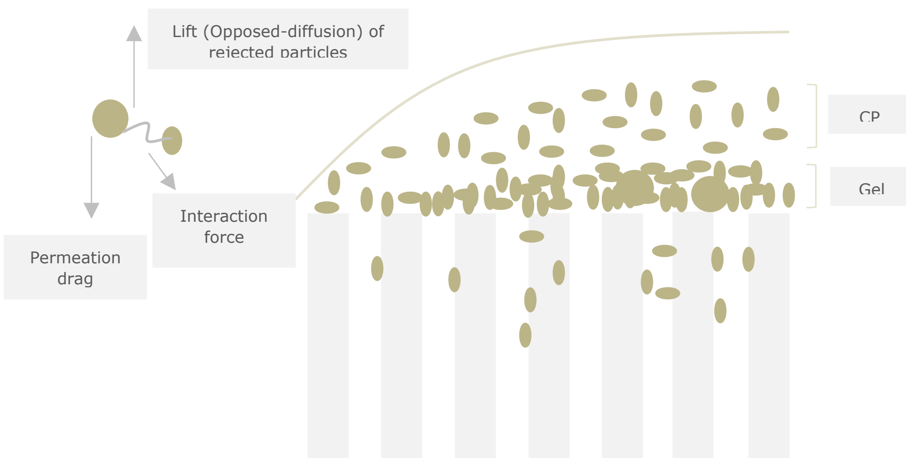

Figure 27 CP position and applied forces on particles in the filtration [37]

4.3.2 Natural Organic Materials (NOM) role in CP

Natural organic materials (NOMs) can be the source of CP. The permeate resistance by CP is mostly due to the low Mw NOM, in contrast to fouling phenomena (due to the high Mw NOM). In this matter, the Stoke-Einstein equation proves that the reverse diffusion flow to the bulk is impacted by the size of the substance. The size of NOM enhances by ionic strength and drops by pH in the range of hundreds to more than 20,000 Da [7].

Therefore, regarding low MW NOM, an increase in the feed concentration and decrease in the MWCO of membranes can lead to the more escalated CP level or even cake layer formation. It is also worth mentioning that the cross-flow membrane configuration has a more efficient response towards continuous CP mitigation, in contrast to the widespread dead-end type [37].

4.4 Fouling Mitigation Assessment

The major problems which MF/UF membranes would face in operation are fouling and CP leading to flux and performance reduction. In this venue, there exist different approaches to mitigate the fouling (Figure 28) which will be scrutinized in this study to understand the advantages and disadvantages of each of them [5].

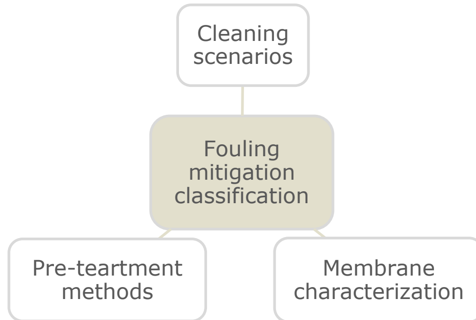

Figure 28 General membrane fouling mitigation methods [5]

It is worth mentioning that the assessment of each mitigation approach is impacted by other factors like the feed water properties and experimental method which may issue deviant results (Figure 29).

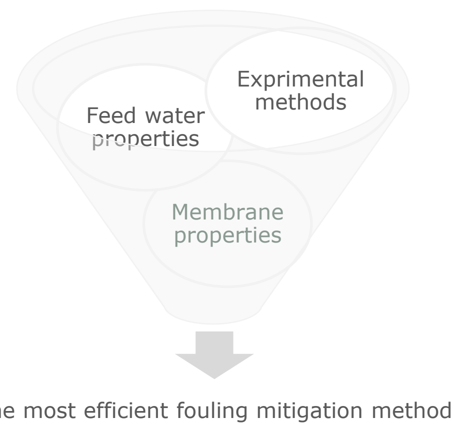

Figure 29 Affecting considerations to assess mitigation methods [5]

4.4.1 Natural Organic Materials (NOM)

Regarding feed water assessment in the water treatment process, among materials evitable to make fouling in membranes; NOM is one of the main precursors. NOM presence in drinking water can drastically deviate the regulated drinking water quality parameters. Besides the change of odor, taste, and color, NOM can be a key reason for chlorine demand increasing. Furthermore, it would lead to forming disinfection by-products (DBP) and the regrowth of microbes in distribution systems [42].

Humic Substances has a share in NOM existed in feed water. It has been proven that the degree of fouling by NOM is not related to the amount of material that existed in the water source, but it is relevant to fractions of specific NOM in the feed. There is the consent that humic material (Figure 30) is the important fraction of NOM in the water regarding the fouling rate. In this matter, other materials also can impact the fouling degree by NOM followed by the below mechanism as a case in point [37].

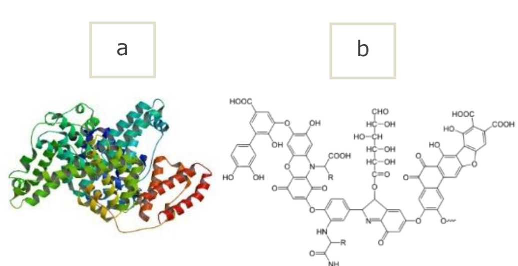

Figure 30 a) Bovine serum albumin crystal structure and b) General chemical structure of humic acid [43][44]

4.4.2 Biopolymers (Proteins)

Biopolymers (macromolecules), specifically proteins also carry the principal role for physically irreversible fouling in UF membranes in feed water for drinking water purification process followed by the mechanism in Figure 31 [45][46].

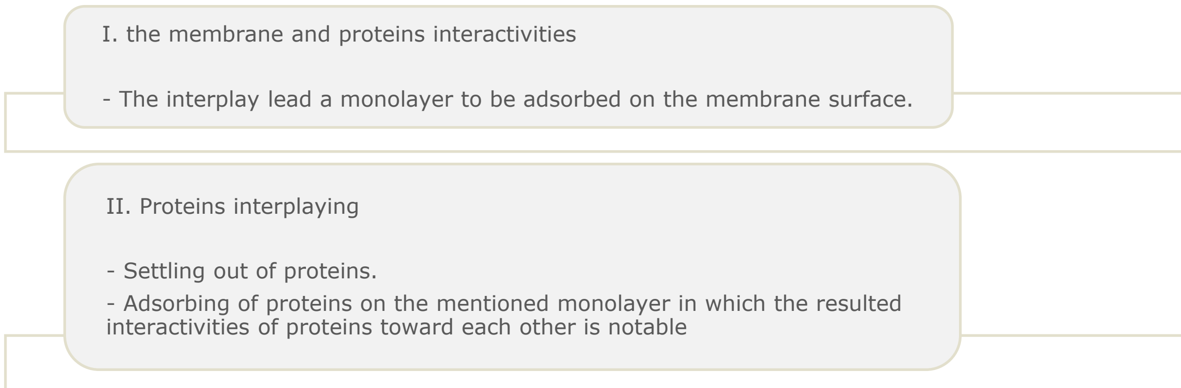

Figure 31 Membrane fouling mechanism by proteins [45][46]

4.4.3 Fouling Agents Recognition

It can be established that both biopolymers (like proteins) and humic substances that existed in feed waters play significant roles in fouling. Both humic materials and biopolymers in the feed water are not considered but the resulted fouling even by the minimum presence of them is hard to mitigate by backwashing. In this way, humic substances and biopolymers hold the responsibility of the fouling mostly.

In this matter, other materials also can impact the fouling degree by NOM followed by the mechanism in Figure 32 as a case in point [47].

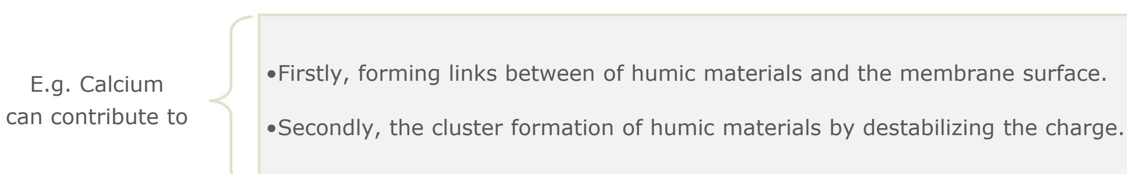

Figure 32 Calcium impact on membrane fouling by NOM [47]

As a general fact, fouling processes can be drawn by a mix of some major occurrences rivalry listed in Figure 33.

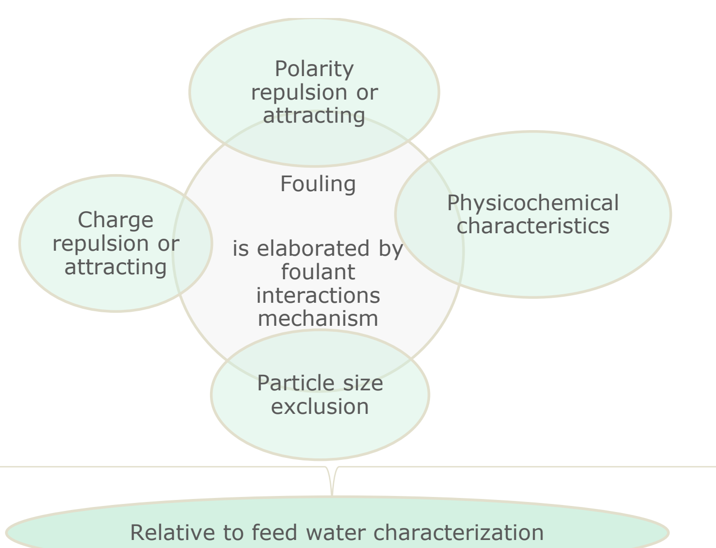

Figure. 33 Major factors impacting on fouling mechanism [47]

The study of the aforementioned features and mechanisms may spring to a bright horizon in fouling characterization which might propose mitigation scenarios consequently.

Feed Water Recognition Has Crucial Importance. Above mentioned features and mechanisms will be studied further in this study through fouling mitigation methods and case studies. Besides, it should be a ground fact that foulant interactions in fouling would not follow a unique algorithm due to the different feed water and membrane types.

As a case in point, the interesting contrast of the newer studies is that humic materials and proteins in the face of polarity are hydrophobic and hydrophilic, respectively.

It can be concluded, the characterization of the specific feed water introducing to the filtration is a must-have to investigate fouling and different fouling agents may be identified. Thus, by that assessment of predominant agents of fouling, the mitigation methods can be condemned more efficiently [48].

4.5 Fouling Mitigation Methods

4.5.1 Cleaning Scenarios

For setting the desired efficiency back and to prevent permeate reduction due to fouling, cleaning scenarios are way outs depends on the type of fouling. In this matter, fouling can be categorized as physically reversible and irreversible, and chemically irreversible.

As mentioned before, cleaning scenarios are characterized based on the type of fouling. To begin with the classification, different physical methods can be applied to reduce the formed reversible fouling – fouling that can be reversed to the status of the particle before their accumulation [49].

Backwashing (reverse flow) of membranes is the most applicable physical reversible cleaning method towards physical reversible fouling leading to make the reverse moving of foulants from the membrane pores and surface. In this matter, air sparging can also apply shearing or cross-flow forces to develop the cleaning and detached particles transportation out of the membrane surface.

Backwashing (Figure) has important disadvantages listed in Figure 34 [50].

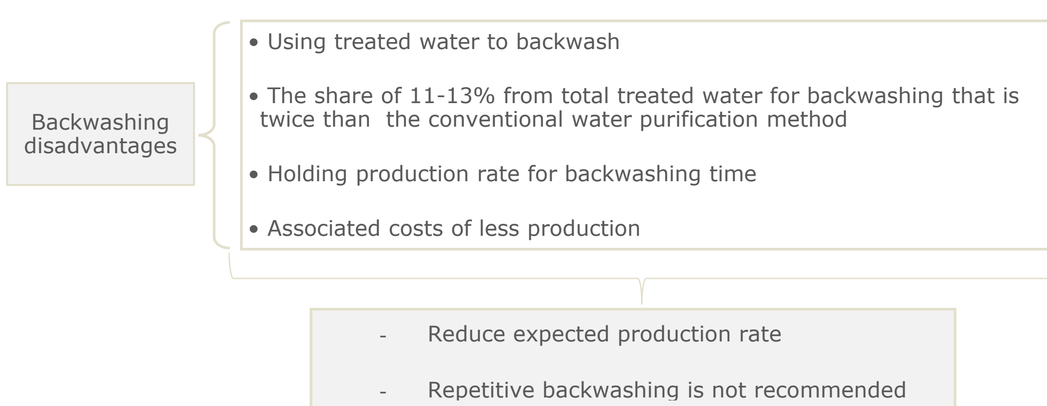

Figure. 34 Negative consequences of backwashing [50]

Air Sparging is another physical fouling mitigation method. Applying air Sparging periodically and at the same time of backwashing (due to decrease its added costs) can reduce the physical reversible fouling significantly. Air Sparging can scrub the material deposition on the membrane surface and decrease the CP and fouling.

Full-air Sparging is applying the sparging process continually in operation. some lab-scale studies have investigated the possibility of the sparging during treating water and backwashing continuously (full air sparging) based on covering up the final costs related to the electricity demand by enhanced filtration efficiency due to the less fouling. However, established results show that, in contrast to wastewater treatment, for water purification systems, full air sparging may lead to the negative reactivation of the fouling layer. This reactivation includes ejection and shifts the fouling layer from the membrane surface to membrane pores and leads to blocking them. In this way, it is revealed that full sparging cannot enhance the fouling mitigation [50].

Chemical Cleaning is another widespread form in cleaning scenarios. After ongoing filtration even by condemning periodic physical cleaning in a while, there would be the evolving of materials that are adsorbed onto the membrane holes or surface. The mentioned physically irreversible fouling would not be impacted by physical scenarios and demand chemical cleaning.

In this venue, the used oxidative agent chemicals are alkali and acidic solutions (specifically for inorganic fouling) like sodium hypochlorite and citric acid, respectively [3]. The most widespread chemical agent for the chemical cleaning scenario is sodium hypochlorite (NaClO) [38][48][34]. The simplified schematic process flow of cleaning scenarios can be illustrated in Figure 35.

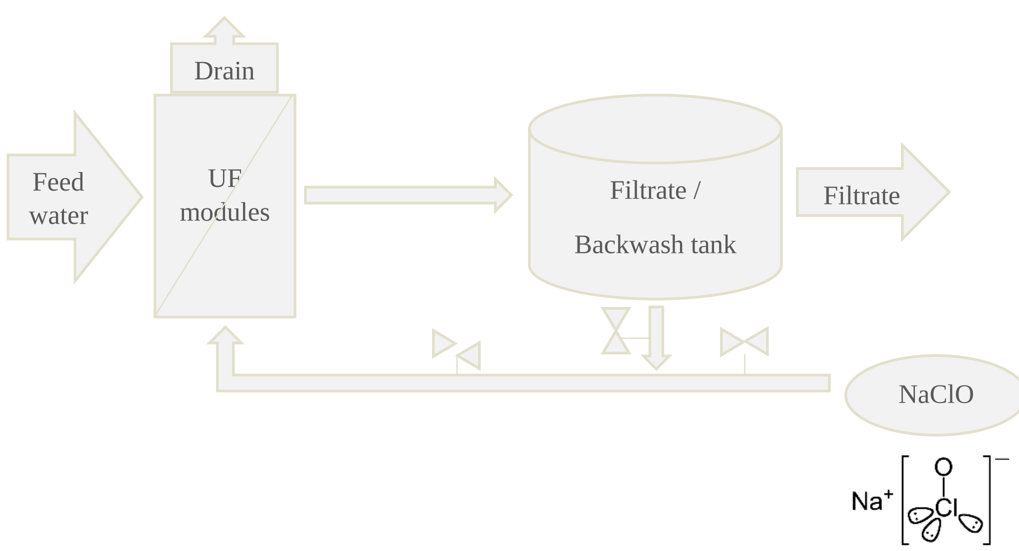

Figure. 35 Chemical (NaClO [52] cleaning process flow in the membrane system [38][48][34]

Ultimately, a long-term formed fouling and frequent cleaning procedures result in the deterioration of membrane properties and the consequent decline of the filtration efficiency irreversibly. Irreversible impacts are the point that is referred to as membrane aging [38][48]. The summarized hierarchy of the cleaning scenarios is illustrated in Figure 36.

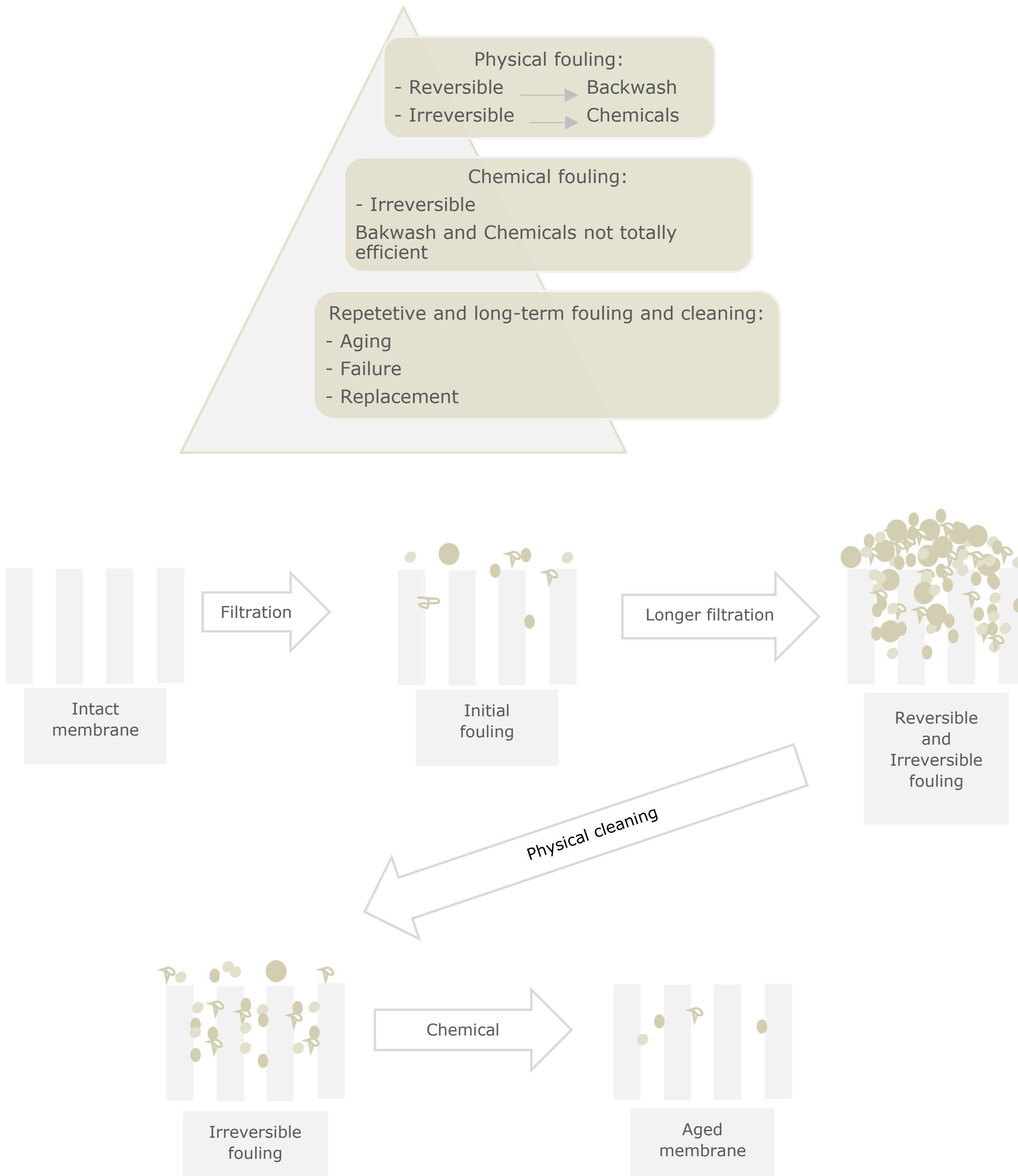

Figure. 36 The hierarchy of the cleaning scenarios for MF/UF membranes in water purification process [34][38]

As a general approach both fouling and concentration polarization (CP) levels are the foremost features to address the better membranes in this matter. However, in some cases, they would play a two-edged sword role. To name a few, an example is listed in Figure 37. As a result, each system needs to pick the membrane type depends on the feed water quality, operational conditions, and exclusive demanded result [37].

Spiral wound type membranes	•Backwashing is not efficient for fouling reduction •Need of higher trans-membrane pressure •Thus, sunstantial need of pre-treament phase • But, better CP reduction rate
Hollow fiber type membranes	•More efficient regards to Spiral wound type disadvantages • But, lower CP reduction rate

Figure 37 Different membrane types towards CP and fouling mitigations [37]

Membrane Cumulative Life-span is a considerable factor in cleaning scenarios. Reasonably, repeated using sodium hypochlorite can lead to change features and consequently the performance of membranes to a great extent progressively over time. That is why even a membrane would have a specific maximum cumulative life span at the time of purchasing from the producer. The mentioned index shows the membrane capacity to be effective after facing specific chemical agent dosages through cycles of cleaning.

For instance, the reported indexes towards sodium hypochlorite exposing for PVDF UF membranes are normally between 3-10 years. In this way, after passing that dosage of exposing the membrane with chemicals over time, the membrane would be regarded as aged one which will be alongside performance dropping significantly. Thus, the approach towards an aged membrane would be the replacement [38].

4.5.2 Membrane Characterization

The most developed approach towards fixing the fouling issue is to consider it at the time of membrane design and production. The newly designed membrane with the amended surface would have better resistance towards chemical agents and consequent rotting [53].

There exist extensive studies which have been scrutinized novel materials and methods to produce membranes with better efficiency as can be seen in Figure 38. In this matter, many studies have focused on polymeric membranes (the most share in water treatment) to suggest methods to make membranes more hydrophilic (anti-fouling), lower surface roughness, and escalate the cross-linking degree of the polymeric top layer to address the fouling issue. Moreover, depends on fouling agents, providing the membrane surface with opposite charge can make it more reluctant to absorb fouling [1].

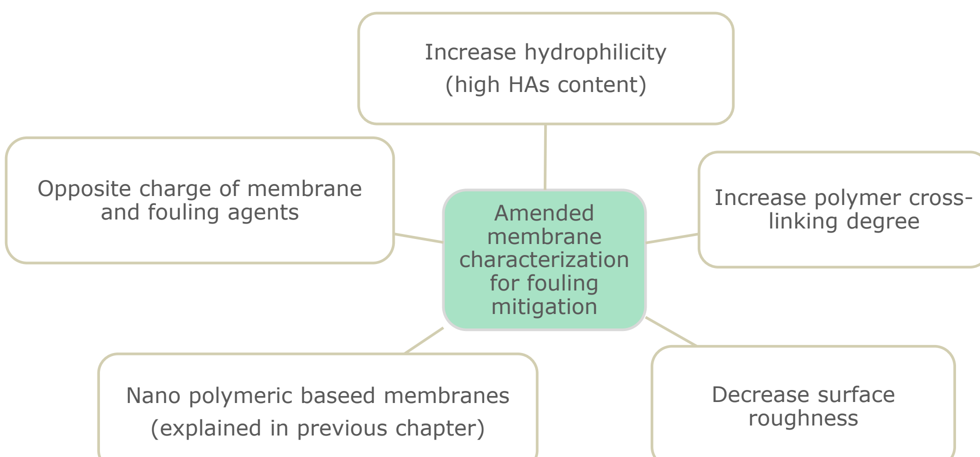

Figure 38 Membrane characterization tactics for fouling mitigation [1][53]

As a case in point, to show different membrane character responses to different fouling degrees, Figure 39 shows significant changes in SEM photos of different UF membranes manufactured by Microdyne Nadir, Germany. The membrane characteristics are Cellulose-based UF (UC) with MWCO of 5, 10, and 30 kDa, and polyethersulfone-based UF (UP) with MWCO of 5, 10, and 20 kDa [54].

SEM photos illustrate the surface of membranes before and after fouling by different types of macromolecules such as biopolymers. In SEM photos of fouled membranes, there exist obvious surface increased surface roughness of membranes.

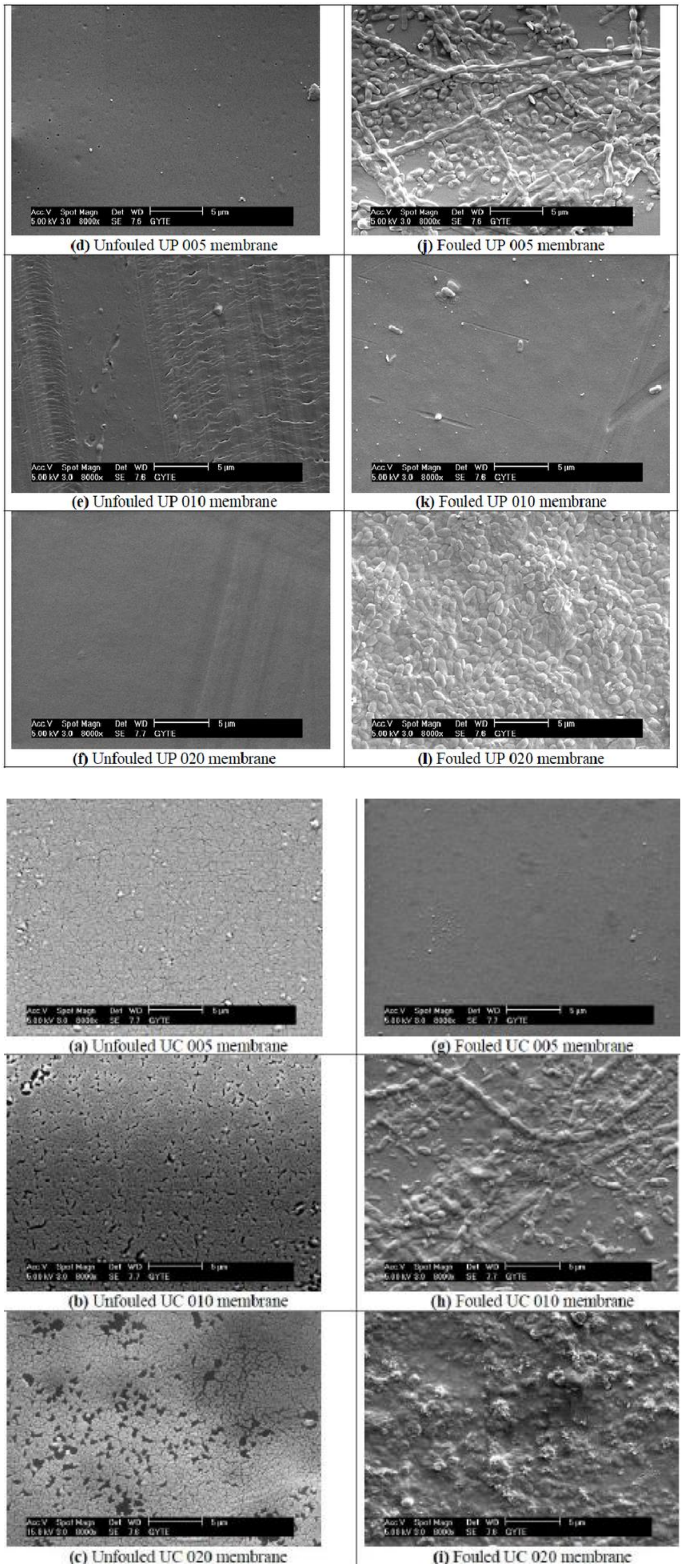

Figure 39 SEM photos of different surface UF membranes before and after of fouling [56]

Bio-fouling needs Membrane Amendment. It is reported that there is a distinction between bio-fouling and other forms of fouling. The difference is that bio-fouling is rooted in microbes that can reproduce themselves and ultimately will be the host of bacteria. Figure 40 is a schematic that shows bio-fouling consequences and mitigation ideas [53].

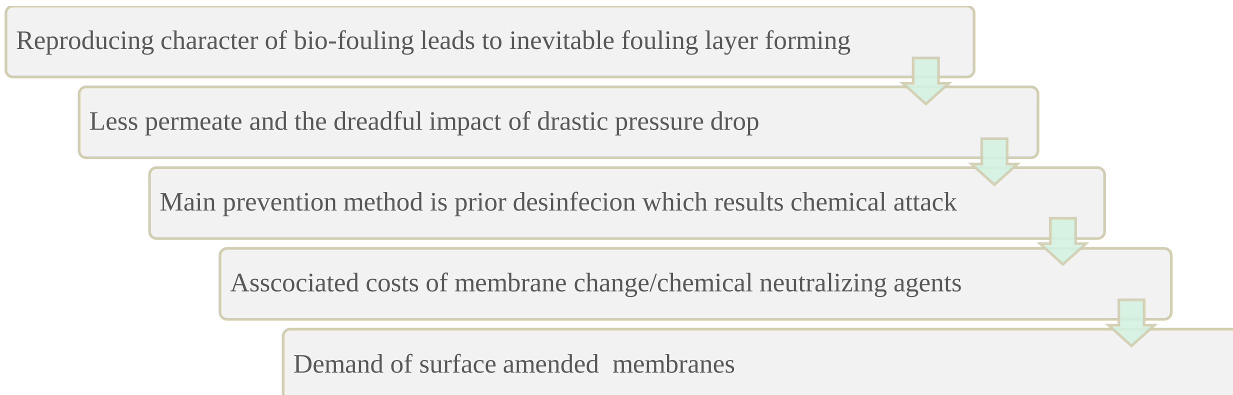

Figure 40 Schematic of bio-fouling consequences and mitigation ideas [53]

4.5.3 Pretreatment Methods

The feed water introduced to the water purification plant needs to pass pretreatment steps before membrane systems to mitigate possible fouling issues. The steps can be designed depends on the feed water quality and the desired treatment result. Each step has the responsibility to provide acceptable water for the next processes and make sure the flow would not create operational problems for the next levels.

In this venue, pretreatment processes can play a significant role in the further organic, bio, and colloidal fouling in membranes. In addition, pretreatment might impact directly to prevent scaling of water systems. Normal feed water depends on the source, can contain materials listed in Figure 41. Different approaches can be briefly described below [13].

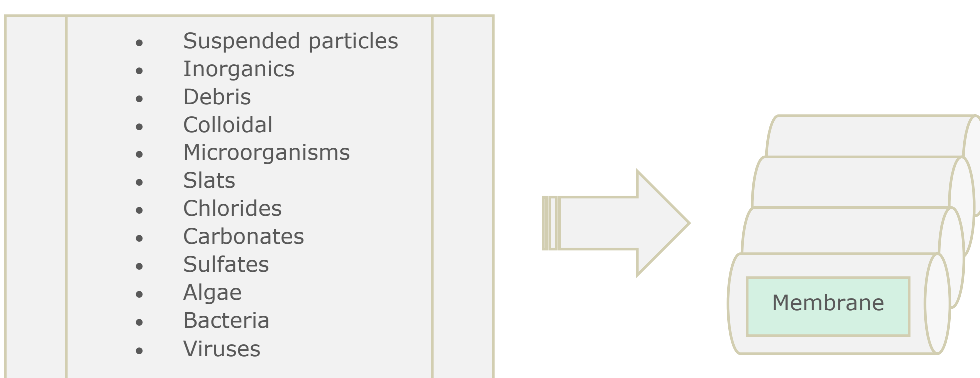

Figure 41 Normal feed water pollutants [4]

Chemical Conditioning is the first approach in this section. There exist numerous applications of chemical conditioning agents includes pH regulating, bio-fouling mitigation, scale prevention, disinfection, and using as coagulants. Chemical conditioning usage is the normal practice before membrane systems. For instance, RO/NF membrane producers suggest applying the mentioned chemicals to hinder scaling by soluble salts formation which can damage a membrane. Regarding the MF/UF, coagulants or lime mainly are used to increase the membrane performance, applying the settlement step can improve the permeate flux. The principal rule in using any sort of chemical conditioning is that the chemical agent should be picked and designed following the specific membrane material that existed in the process [4].

Adsorption is applied to mitigate specific fouling agents. Adding an adsorption step can be due to NOM presence in feed water. NOM can make a wide range of impacts on the fouling but the adsorption step objective is to adsorb specifically trihalomethane (THM) formed in the disinfection step [4]. THM is the product of the reaction of NOM and chemical disinfectors which cannot be filtered by membranes. Thus, the adsorption process can reduce NOM and THM in the flow before membranes.

The Ion Exchange process can be applied to soften water [4]. In this way, dissolved salts or cations may not exist to scale the membrane surface (Figure 42). The process includes using cationic resins in which Na^+ can be substituted by calcium cations and prevent further insoluble salt formations by calcium and carbonate ions. This process is an efficient approach in RO systems since the RO retentate can be used to regenerate cation exchange resins [4].

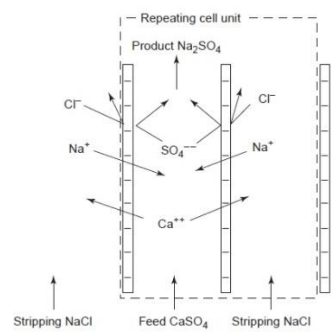

Figure 42 A softening ion exchange process schematic [55]

Prior Coagulation/Flocculation/Sedimentation can be applied to the membrane system. Coagulation is applied during traditional water purification to neutralize the charges of the particles to increase dissolved organic reduction and turbidity. However, the mentioned type of coagulation before UF would need a different dosage of coagulant (e.g. diatomite) to only pick out specific groups regards to fouling (e.g. Biopolymers) In this matter, the reachable floc size need to be adjusted since it can make irreversible fouling by going to the membrane pores. The disadvantages of the coagulation are residuals and adding solids which lead to a decrease in the membrane permeate flux [48].

Prior Coagulation/Flocculation/Sedimentation Mechanism can be defined before extending the approach. It is confirmed that using the coagulation step before UF membranes can collect and make clusters of NOM, biopolymers, and biodegradable materials leading to make low-resistance fouling. The more fouling is low- resistance, the more fouling is physically reversible which results in a lower total fouling degree. The coagulation impact can be observed on the lower degree of both pore choking and cake layer. Thus, physical cleaning (backwashing) can be more effective [48].

For instance, the coagulation mechanism regards to NOM is shown in Figure 43 schematically.

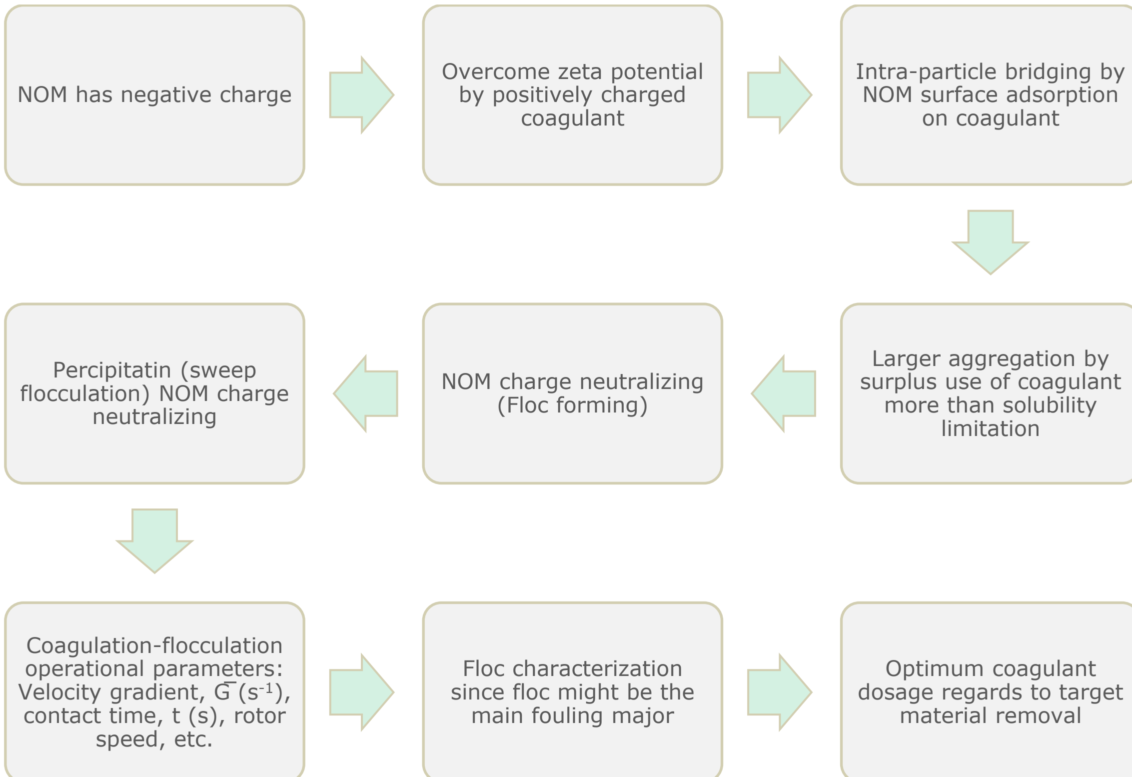

Figure 43 Coagulation mechanism as a fouling mitigation method regards to NOM in feed water [48]

In-line Coagulation is one of the choices in the face of membrane systems that can decrease piping and equipment installation and areas [57]. Some types of in-line coagulation are seen in Figure 44. This method includes adding the coagulant to water without reduction of the coagulated materials by the settlement phase. The advantage of this method over the normal coagulation is that the coagulated materials without settling can be compressed by the membrane working and its pressure which results in decreasing the gel layer resistance forming on the membrane surface. In this way, the permeate flow quality and performance would be enhanced [4].

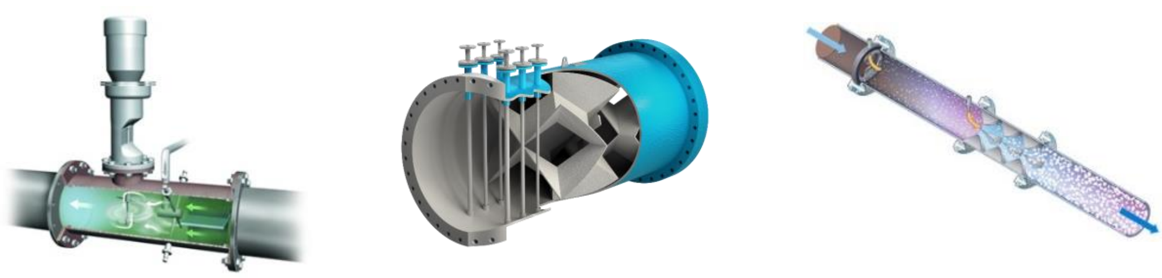

Figure. 44 In-line coagulation schemes [56][57][58]

The Associated Cost of Coagulation is an important factor for applying coagulation. In this venue, the amount of coagulant becomes a factor contributed to costs mainly related to the sludge disposing. Thus, the coagulant usage during whole permeation cycles is not favorable. Regarding optimum use of coagulant, membranes could sustain the primary permeate rate by using coagulant for the first half of each treating cycle [3]. In other words, using phased coagulation can propose the same fouling control rate with no extra irreversible fouling increment in comparison to the continual coagulation. In this matter, studies can prove and propose gradual (not continual) coagulation as an effective and positive method for fouling control with minimum operation modifying and optimal sludge disposal costs [50].

4.5.5 Coagulation/Flocculation/Sedimentation approaches

Considering many studies that have reported applying specific setups for pretreatment before MF/UF membranes including coagulation, flocculation, and sedimentation, results can be categorized by classifying the methods as Figure 45.

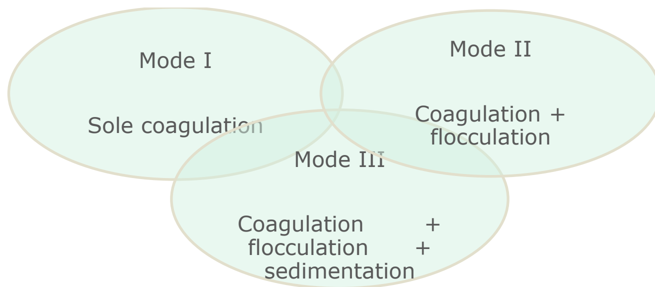

Figure 45 Three applied different modes of coagulation/flocculation/sedimentation [48]

The further assessment trend may spring to issue a broad perception of an optimum pretreatment configuration design before MF/UF filtration in drinking water purification plants [48]. There exist numerous studies applying each mentioned mode [48]. Figure 46 is illustrated the schematic process flow of defined modes. The review conclusion of different studies for each mentioned mode is as below.

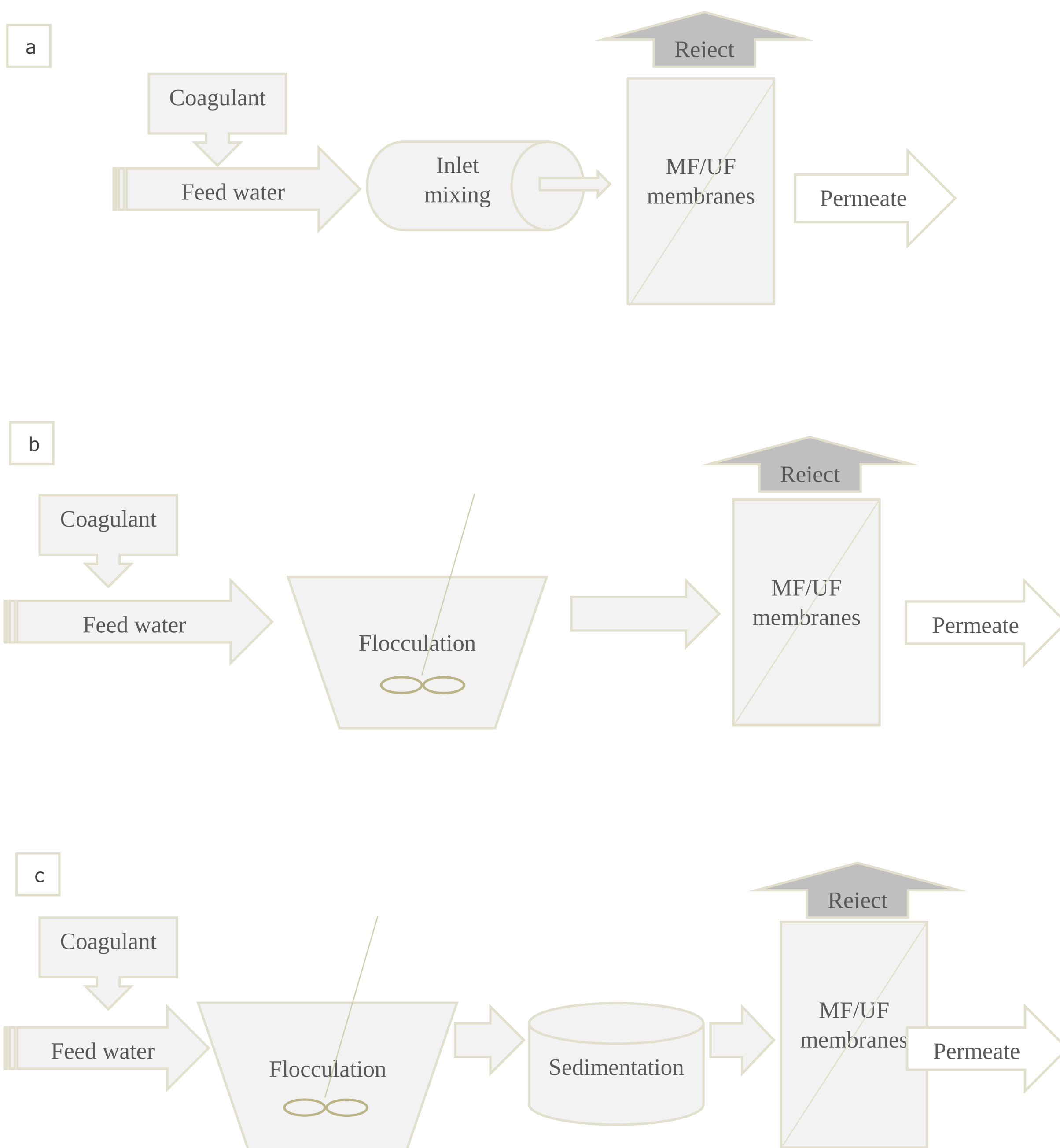

Figure 46 Schematics of mode I, II and III in a, b and c, respectively [48]

Firstly, sole coagulation would not be an optimum approach compared to other modes. That is because of the small floc by lacking the flocculation step. Therefore, there is no settling-out step for colloids which may contribute to fouling by blocking membrane pores and decrease permeate flow rate [4].

Secondly, it is established that flocculation creates floc with a large size which might be more porous and favorable for cleaning. However, the pile-up of large floc due to the lack of settling step leads to escalating fouling layer. On the other hand, a lower fouling rate of coagulants after passing flocculation and sedimentation would make thin layer fouling but in a more irreversible form. Moreover, the needed contact time is higher in the order of mode III > mode II > mode I [48].

In conclusion, coagulant dosage plays a crucial matter for all modes which needs to be between lower to higher of a range and required to be characterized based on the membrane pore size and feed water properties. Therefore, it is recommended to characterize a promising dosage for each operational plant and mode [48].

Prior Coagulation in Face of Proteins can be scrutinized in more detail. Proteins are hydrophilic compared to the hydrophobic surface of PVDF/UF membranes. There is evidence of the low-dosage coagulant impact on a conformation altering in proteins which changes the polarity of proteins and makes them more hydrophilic than the case without coagulation. In this way, the first proteins formed a monolayer on the membrane surface shows a reluctant reaction towards other proteins. This level of reluctance can be considered as the fouling decreasing mechanism. On the contrary, it is reported that using a low dosage of coagulant may lead to a harsh fouling rate by applying bovine serum albumin (BSA) as the fouling agent. The reason proposed for that high level of fouling has been introduced as particle size altering by coagulation to the extent near to the diameter of membrane pores [49].

Therefore, investigating the coagulant optimum dosage to make the fouling reduced in UF in the water purification process in the face of different proteins as major precursors of fouling is crucial [49]. To issue a general guideline based on the reviewing of studies, Table 8 can be beneficial.

Table 8 Recommended coagulant dosage for coagulation of proteins and consequences [49]

Protein type	Recommended coagulant dosage	Result
Ovalbumin (OVA)	< 0.3 mg/l metal	Decrease the physically reversibly and irreversible fouling
Bovine serum albumin (BSA)	< 0.3 mg/l metal	Decrease the physically reversibly and irreversible fouling
Beta-lactoglobulin (BGA)	No impact of coagulant	Revealing the complication of proteins composition in different feed water

Generally, mitigation methods focus on turning the irreversible fouling to the reversible phase for preparing the ground for fouling reduction by physical reducing like backwashing. This trend is based on the fact that the more the fouling layer has porosity, the more it is vulnerable to consider as reversible fouling. In contrast to that trend, applying prior low-dosage coagulation includes a different mechanism in which the mitigation is followed by increasing the rate of proteins transmitting [49]. The mentioned transmission would influence the next processes after the membrane system.

4.6 Case Study; Novel GDM Membrane

Among different membrane types, gravity-driven membrane (GDM) is a filtration method by using weak hydrostatic pressure which creates the force of filtration (Figure 47). The system itself benefits from MF or UF membranes and can be applied in the water purification process. In this case, the filtered flow would be lower than other systems, however, as a significant advantage, there is no need for fouling mitigation and control as much as other types need. Thus, in GDM, physical and chemical cleaning scenarios are normally minimized and not repetitive demanded [59][60].

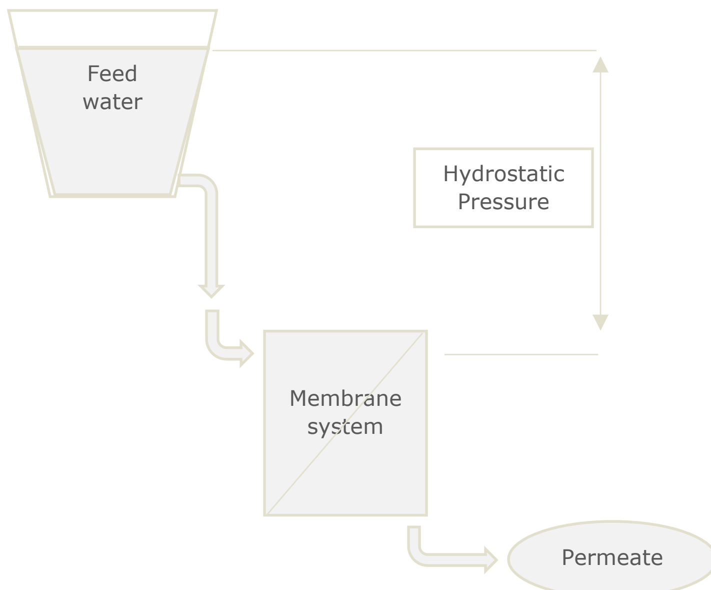

Figure 47 Scematic process flow of GDM system [59][60]

4.6.1 Passive Gravity-driven; Cleaning Scenario for GDM

Since it is mentioned about the lower filtered flow rate of GDM, in case of any physical fouling existence, the filtered flow would make it lower and not favorable. So applying backwashing or air scrubbing, and sometimes draining can easily keep up the standard flow rate. Even by using the mentioned fouling controls, it is established that these implementations are minimal due to the simplicity of those passive physical scenarios compared to other types and their cleaning scenarios. In this venue, the fouling control of GDM is named PGM referred to apply a passive gravity-driven membrane process.

One of the main merits of the GDM/PGM process is to propose a simple process with fewer operation sophistications. The mentioned advantage makes the GDM/PGM favorable for compact water purification system needed by small and remote living places which are detached from treated water distribution systems [60].

Figure 48 is illustrated the lab-scale GDM filtration in the water purification laboratory of Prof. Pierre Bérubé at the University of British Columbia, Canada [61].

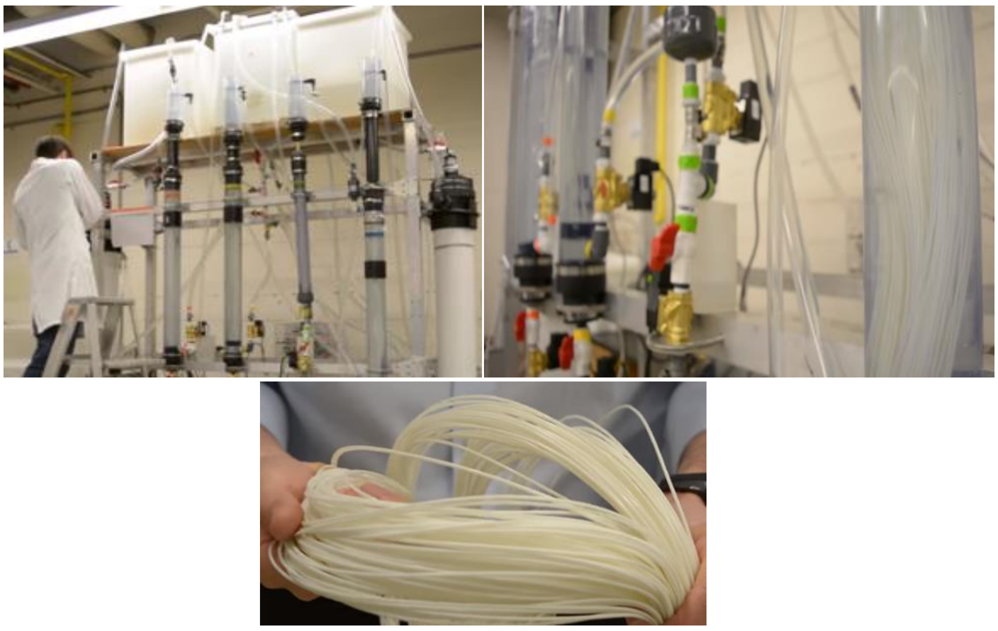

Figure. 48 Lab-scale GDM system in University of British Colombia, Civil Engineering Dept. [61]

4.6.2 GDM/PGM Anti-fouling Character and Mechanism

The advantageous performance of the GDM/PGM system is rooted in forming a biofilm layer in the membrane surface leading to keep a stable permeate rate. Continuing filtering operation results to that mentioned layer transform biologically to heterogeneous compositions with channel web works. That transformation can ease the flow rate by resistance reduction. On the other hand, irreversible fouling may escalate the resistance. Thus, the final stable status can result from the interaction of those two opposite affecting elements towards the flow resistance [60].

GDM/PGM system can benefit from the impact of the formed biofilm layer to escalate the NOM reduction. In this way, the wide range of fouling agents in the water purification process can be removed [9]. It is worth mentioning that formed biofilm layer can have the same virus reduction ability as formed gel layer (gel layer follows the mechanism of size exclusion, electrostatic repulsion, and adsorption). Thus PGM/GDM contributes to virus elimination significantly [60].

Table 9 is summarized the comparison of filtration parameters for PGM/GDM and traditional membrane systems.

Table 9 Performance comparison of GDM system with conventional membrane system in water treatment [60]

Index	PGM/GDM	Traditional membrane system	Unit
Operation cycle time	24	1	h
Permeate	5	40	l/m^2h
Halting time after the cycle for backwash and aeration start	1	-	h
Backwash trend	20	40	m^3/m^2min
Aeration trend	2	2	m^3/m^2min
Chemical cleaning	-	3	Per week

4.6.3 GDM/PGM Anti-aging Assessment

Pressure decay tests (PDTs) assessment can approve the biofilm layer can minimize the breaching of membranes. The mechanism of that action is to cover up breaches by the biofilm layer and make another sort of obstacle against pollutions. In this way, the biofilm layer would deny the movement of pollutions into breaches. It is also reported that a higher level of virus elimination resulted from the biofilm layer, may not be affected by filtration flow through the biofilm layer and applying periodic PDTs [9]. However, PDTs assessment cannot be beneficial effects to evaluate the level of virus elimination by the biofilm layer in GDM/PGM systems. It means there exists still room for an amended PDT method in that venue [60].

5. MEMBRANE AGING

5.1 Membrane Aging Definition

There exist shreds of evidence regarding the decrease in the membrane performance after long-term using which is referred to as membrane aging (Figure 49). The membrane aging resulted from fouling formation in membranes and consequently the repetitive applying of cleaning scenarios leading to make a membrane aged and resist responding to the cleaning. Therefore, it would result in membrane substitution in the final point. In this way, the more aging reasons and consequences are realized and investigated, the more anticipation and mitigation methods would assist to increase the performance and reduce the economic burden of membrane aging [38][51].

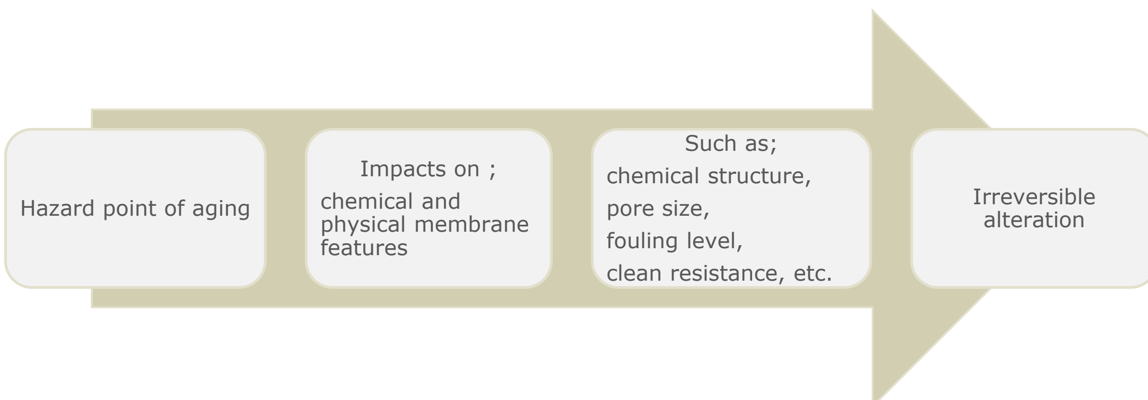

Figure 49   The hierarchy of membrane aging and consequences [38][51]

5.1.1 Membrane Aging Assessment

One of the more important features of using membrane technologies is the better level of process controlling and monitoring. It means in membrane technologies, it is possible to scan the inlet and outlet quantity and quality easier. However, the membrane replacement prediction is still considered a possible issue. The mentioned prediction has importance because the wrong prediction may jeopardize and halt the operation flow. In this venue, membrane producers mostly issue a working guarantee, for instance, in the range of 3-10 years, but operational circumstances can change the lifespan of membranes to even outside of that range. Figure 50 justifies the membrane aging assessment.

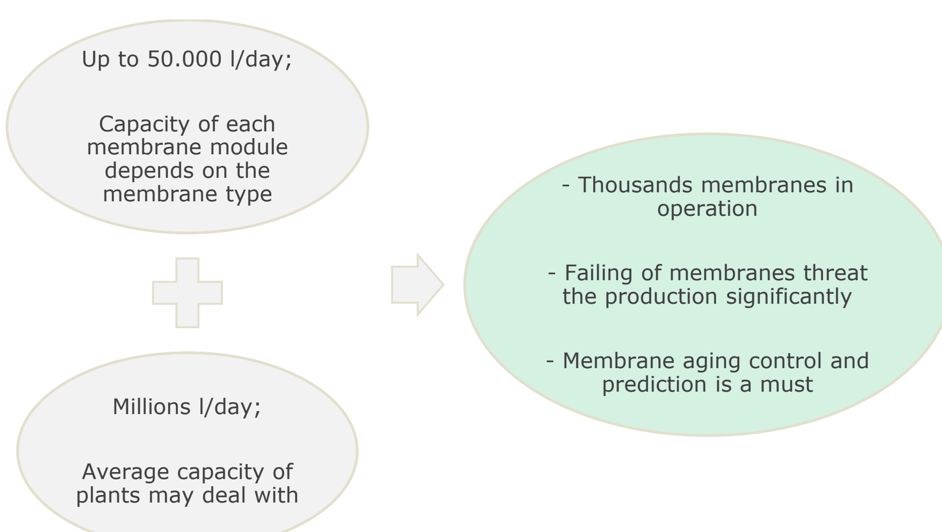

Figure 50   The justification of membrane aging assessment [53]

5.2 Membrane Aging Assessment Considerations

The general method to assess membrane aging is to age different membrane setups by chemical cleaning agent exposure and compare performance parameters of the filtration by membranes and the intact (not aged) membrane. In this matter, there are different approaches for chemical exposure and exposure concentration to mimic the full-scale conditions as below.

5.2.1 Lab-scale Membrane Aging Set-ups

Single-step Immersing of the Membrane is the most popular form of performing the membrane aging tests in a reasonable timeline by immersing the membrane in a concentrated chemical solution illustrated in Figure 51 (a). This is because to imitate the long-term exposure of the membrane to chemicals and aging by the passing of time [51].

It is reported by several studies that single-step aging would show a decrease in the membrane permeation resistance which is a controversial result compared to the aged membrane in the full-scale plant in the long term. This contrast can be justified by pore enlargement in single-step aging in labs [51].

Cycled aging of the Membrane is another form of aging set-ups. As it can be seen in Figure 51(b), there still exist studies that have another lab-scale approach by applying cycling aging (repetitive filtration and cleaning periods) like the full-scale situation [39].

For this mode, there are in contrast HAs results reported regards to the performance parameters. The atypical results bring this idea that there might be other factors other than HAs contributing to membrane efficiency [51].

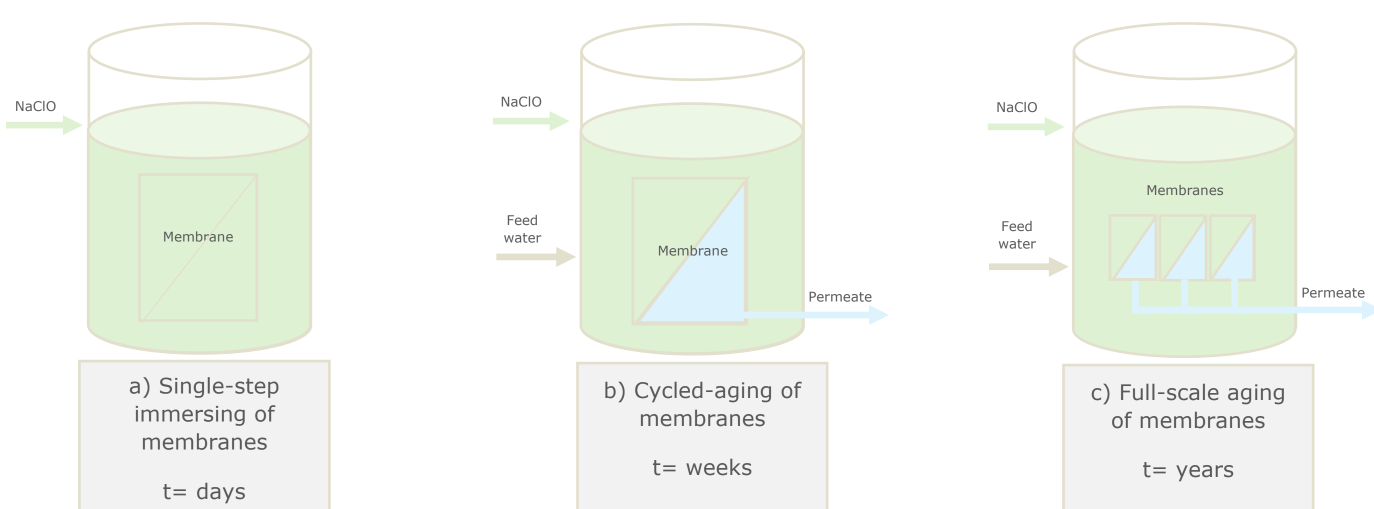

Figure 51 Schematic process flow of different lab-scale aging approaches and full-scale condition [51]

In both forms of lab-scale studies still using high chemical agent concentration is a strong disadvantage. It is reported the normal amount of mentioned concentration in lab-scale studies is 10-1000 times more than the common concentration applying in the full-scale mode of water treatment plants due to the timeframe limitation of the membrane aging studies [51].

5.2.2 Chemical Agent Concentration and Dosage

The cleaning agent dosage which makes the irreversible deterioration in the long-term is considered as a concentration in long-term time. The most important parameter in the assessment of the chemicals exposing effects on aging is the concentration of the chemical solution.

Moreover, for having a better approach towards the aging trend led by sodium hypochlorite, exposing dosage can change the aging trend for membranes. Generally, in full-scale plants, there is an optimum point regarding the level of dosage and concentration for the prevention of aging [62]. Thus, for stimulating the full-scale conditions in the lab-scale tests of aging, the chemical dosage needs to be regarded besides chemical.

5.2.3 Chemical Concentration-time (C*t)

Due to the mentioned deficiencies to consider only chemical concentration solution for aging the membrane, novel approaches have proposed parameter which is relative to the chemical concentration and aging time collaboratively, known as C*t (Figure 52). C*t is the realistic exposer parameter that happened in the full-scale plant which reflects the exposure over the long-term time [36].

C*t has been defined as the concentration multiply by the exposure time. It is applicable to assess the effect of the chemical agent on membrane aging more practical. There is a vital proven result by applying the C*t parameter that the aging of the membrane is more dependent on the exposure time (t) compared to the chemical agent concentration (C) [36].

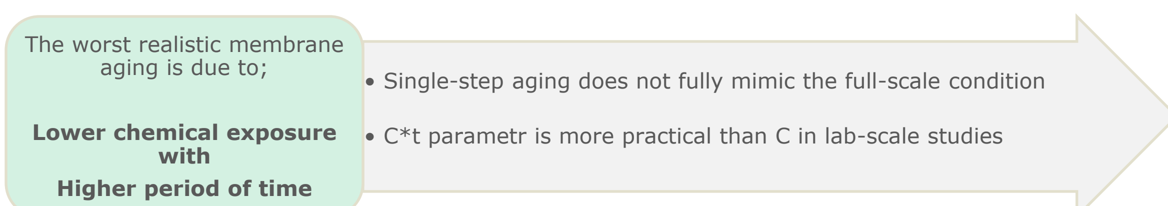

Figure 52 C*t concept justification [36]

The mentioned result proposes that most lab-scale studies which have used single-step concentrated solution for membrane aging would have not predicted the aging effects on the membrane efficiency rigorously [38][62]. On the other hand, aging assessment during years in treatment plants might demand extensive workloads and control [51].

5.2.4 Number of Tested Lab-scale Membranes

Numerous studies have focused on fouling reasons for a sole membrane in the lab-scale tests, while, the achievements cannot be applied for a practical situation with parallel membranes. In this venue, the reason can be identified as the different feature essence of a sole membrane that has been studied and the characteristics of the membranes in a full-scale plant.

As cases in point, the difference in specific surface area and packing density or higher concentration factors (CF) of membranes implied in a water treatment plant compared to the lab-scale one might not have a similar response to the mitigation methods. For more elaboration, it can be implied that CF is relevant to the existed components in the feed water, the membrane type (dead-end or cross-flow), and applying turbulence by air sparging. Herby, considered CF value in most of the studies around 1 in the lab-scale operations has a different and specific value for varieties of feed water introduced to the plants which use cross-flow membranes or air sparging. The CF only can be considered approximately equaled for lab and full-scale operations if the membrane would be the dead-end type. As a result, the CF influence on fouling in lab-scale studies that can reflect the same configurations as the practical circumstances is a crucial step that can prepare the ground to compare the results to full-scale systems [63].

5.2.5 Operational Parameters

In the lab-scale studies, performance is simulated in a way other than the full-scale unit to induce the same circumstances. It results to achieve outputs that are not matched with the full-scale parameters due to the elimination of real complex operational status in treatment plants. The mentioned on purpose reduced complexities for lab-scale experiments are rooted in the natural used feed water, cleaning processes, and extra parameters added by hydraulic circumstances in the plant [38]. The disadvantages of lab-scale studies concerning operational parameters are listed in Figure 53.

Lab-scale operational parameter disadvantages	A good approach features
• Measuring only limited performance factors	• Evaluation of performance factors and membrane characteristics beyond permeability
• Short timeframe parameter measuring	• Approach of multi-year timescales
• Only considering one full-scale membrane not testing different membranes from multiple plants	• Data from a variety of water treatment facilities

Figure 53   Disadvantages of operational parameters in lab-scale aging studies [38]

5.2.6. Ageing Assessment Methodology

The assessment of the aging effects on MF/UF membranes that have been exposed to sodium hypochlorite earlier is important to predict the failure time. The evaluation base always is to consider the cleaning performance and final filtered permeate of chemically exposed membranes in comparison to a not used and –affected membrane by chemicals [62].

It is reported that the increase in age may have an impact on the permeate level through chemical recovery. It means that the more age of membranes increases, the more liking of them to have fouling escalates. As a result, it can be recommended to apply to observe and control permeate recovery changes the method in the chemical cleaning procedure. The mentioned method suggests a way to be a continuous implement for the membrane age assessment with no making interruptions in the operation. Thus, it can give a promising perspective and anticipation for the membrane substitution time [62][53]. As result, the aging evaluation parameters are shown in Figure 54.

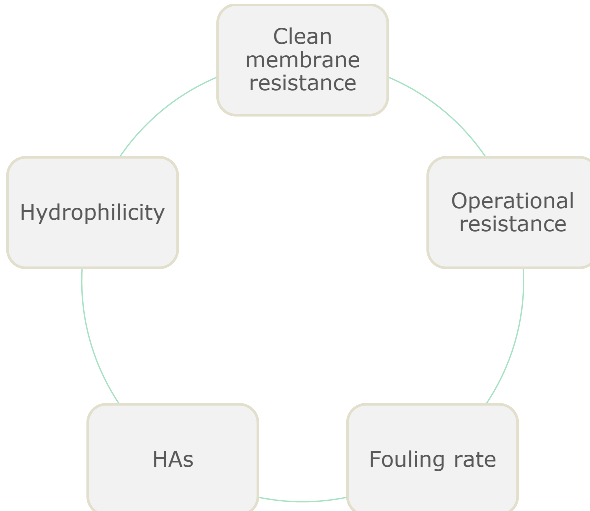

Figure 54 Membrane aging evaluation parameters [62]

The impact trend of aging on the mentioned parameter can be described (Table 10) based on the reported data for three different approaches of experiments including single-step immersing, cycling aging, and full-scale aged membranes [51].

Table 10 Membrane aging assessment for three aging procedure approaches [51]

Evaluation parameter	Single-step immersing	Cycled-aging	Full-scale	Result
Clean membrane resistance	High decrease	Decrease	Increase	- Irreversible fouling in the full-scale process is more extensive
Operational resistance	-	Increase	Increase	- Harmonized respond approves this index for the full-scale aging study
Fouling rate	Increase	Increase	Increase	- Polarity exchanging from hydrophilicity to hydrophobicity - Indexes can anticipate the real aging procedure in full-scale mode - In single-step aging, there is more advanced hydrophilicity with porosity and pore size of elevations due to high concentrated chemical solution - In cycled aging, there is an increase in pore size but a decrease in porosity. The porosity decrease would result from the irreversible fouling pile-up. - It can be anticipated that, the porosity lowering of aged membranes in full-scale can be a sign of higher clean resistance parameter due to irreversible fouling effects
HAs	Decrease	Decrease	Decrease	
Hydrophilicity	Increase	Increase	Increase	

Considering the results of the above table, as expected the membrane resistance alteration is rooted in two mechanisms in different rate levels including pore expansion and pilling up of irreversible fouling in the below manner (Figure 55).

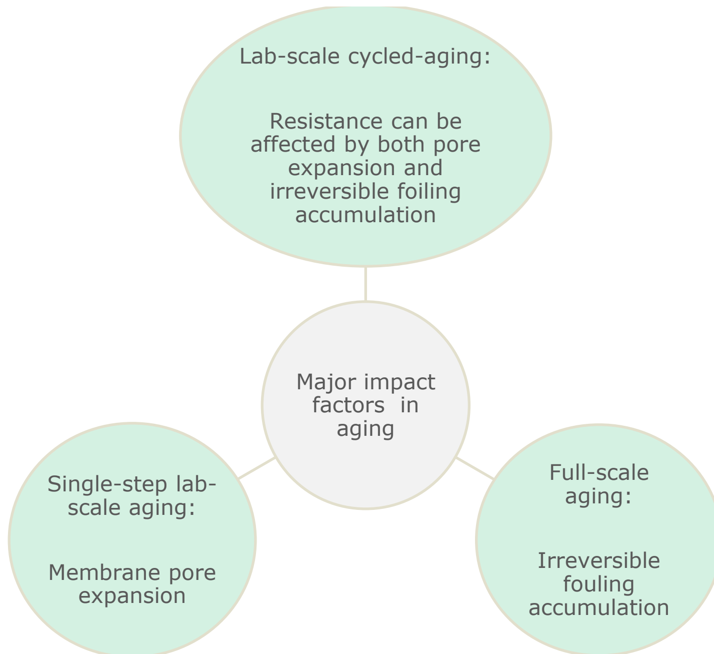

Figure 55 Impact factors of membrane aging for three tested aging approaches [51]

As mentioned earlier, typically for all approaches, HAs decreasing during the membrane aging results in higher hydrophobicity. The more hydrophobicity level brings about the greater fouling rate. As an established recommendation rose from results of comparing three aging approaches, there is a need for the new trend towards cleaning process in full-scale operation. The suggestion is that unscheduled cleaning even by more duration time would contribute less to the membrane aging less than scheduled repetitive plans of cleaning. It means the extensive chemical cleaning scenario can be applied just at the time of need not on the frequent plan.

Aging Methodology Evaluation needs to be condemned too. In this way, another anticipated result of the above comparisons is to find out how lab-scale aging approaches are beneficial for full-scale aging. With comprehensive studies, it is reported that although the cycled-aging method would have better and adjacent results to the full-scale operation compared to the single-step method, none of the lab-scale approaches are reflected and estimate the aging process happening in the full-scale plant. As proof, none of both lab-scale can have irreversible fouling level and consequently, the higher clean resistance recorded for a full-scale aged membrane. On the other hand, mentioned lab-scale methods can have a beneficial approach to intimate chemical structure-oriented parameters in the full-scale operation to a good extent. As a case in point, the polarity assessment via HAs index shows in common trend for all methods [10].

5.3 Case Study; Canadian water purification plants

In this section, the study reviews the assessment of the membrane aging in full-scale UF water purification treatment plants in a long-term period by following up all mentioned required notifications. The evaluation data originated from different aged membranes from eight different treatment plants (Canada) applying the performance tests in lab-scale setup followed by the below-listed considerations (Figure 56) [38].

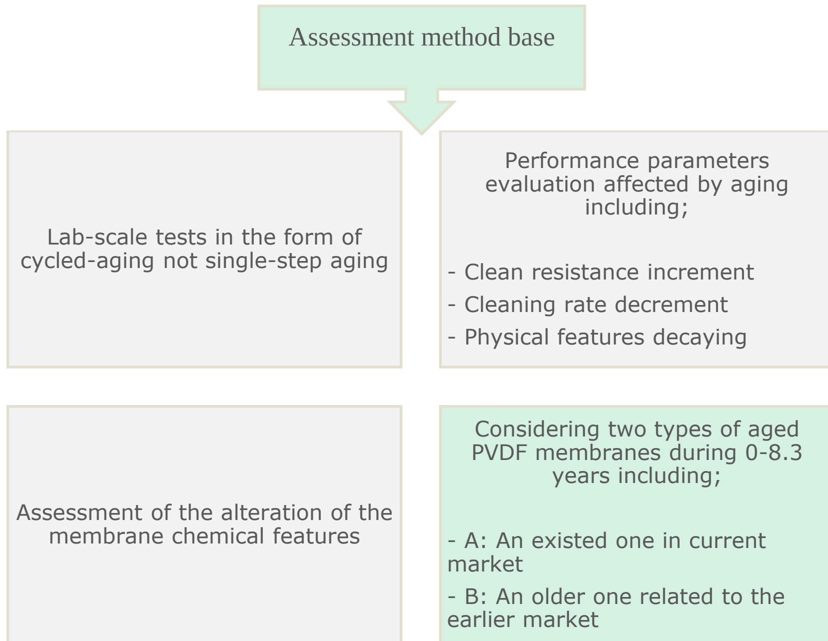

Figure 56 Experimental considerations to study membrane aging of eight Canadian water plants [38]

Based on the above-mentioned assessment methods, the results of the membrane aging evaluation can be issued regards to two types of condemned membranes and performance parameters, listed in Table 11.

5.3.1 Affected Chemical Features

The results illustrate that membrane-type A does not have a considerable performance decline in terms of chemical features. Thus, results are mainly issued for the membranes type B and there would be more time to test the membranes type A and detect parameters' alteration.

On the other hand, the membrane-type B with more age shows the HAs content reduction. It suggests that evaluation of the older membrane of that type is needed since there would the possibility of performance change during long-term activity. In this way, the single-step immersing tests of the membrane type A with a high concentration of chemical cleaning agent prove the HAs decreasing. Thus, it is anticipating that the older and more aged membrane-type A might experience the same deterioration [38].

For the evaluation of the chemical features of the aged membrane-type B, the bulk and surface of the membrane tests show chemical alteration in both of them. In this venue, HAs content is reduced over time and makes the membrane more hydrophobic and consequently vulnerable to the increase of fouling (Table 11) [38].

Table 11 Chemical, mechanical and fouling rate trend of membrane-type B by aging [38]

Factor	Trend by age	Trend by clean membrane resistance
Fouling Rate	↑	↑
Surface HA	↓	↓
Bulk HA	↓	↓
Elongation	↑	↑
Maximum Stress	↓	↓
Young's Modulus	↓	↓

5.3.2 Affected Clean Resistance

In terms of the clean resistance factor, there is an identified increase for the membrane type B which has been aged during 5 years of filtration listed in the below table. This result is anonymous with those lab-scaled test results that applied aging via cycling (filtration and cleaning periods) not the single-step immersed membrane aging tests. This comparison is proof of the matter that is mentioned earlier as; clean resistance factor increases for single-step immersing of the membrane due to the enlargement of membrane pores by aging, in contrast, it should decrease in full-scale or full-scale simulation operation due to the irreversible fouling formation [38].

5.3.3 Affected Physical Features

Regarding physical features, since there is no available data for type A with more than 5 years of age, the more aged membrane-type B only is assessed in this matter. The results of factors are shown in Table 24. It can be concluded that the constant and decrease in physical feature parameters besides the irreversible escalation of the clean resistance factor can alarm the fouling rate and vulnerability of the membrane type B for breakage by the passing of time [38].

5.3.4. C*t Parameter Result

Considering the C*t parameter, if selected membranes have experienced aging by a constant and not high concentration of the cleaning agent, the use of that parameter would not be needed. It is reported that for a selected membrane from a plant that used a high concentration of chemicals, the age of the membrane compared to others is higher than the same operational years of performance. It means it is strongly recommended for the treatment plant not to apply a high level of concentration of chemicals in cleaning due to the rapid aging. But in any case that a plant has been used different dosages (from low to high) of chemicals in the cleaning, applying the C*t parameter is recommended to achieve a better understanding of aging characterization [38].

6. MEMBRANE FAILURE

6.1 Membrane Failure Definition and Reasons

Membrane failure is categorized into two types including failure in operation and failure in commissioning as can be seen in Figure 57 [53].

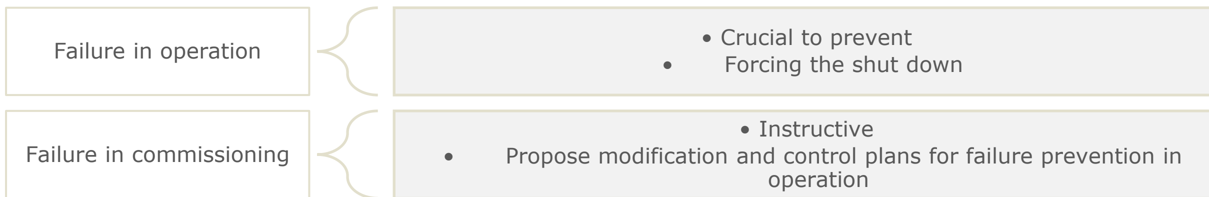

Figure 57 Membrane failure classification [53]

Table 12 has listed a review of the causes and effects of major common failure types.

Table 12 Cause and effect of common failure types and [53]

Failure type	Position	Reason	Phase	Impact
Manufacture error	-Membrane - Module	Incidental	- Installation - Operation	- Low reduction level - Pressure deviation
Fracture/breakage	Module	- Hydraulic disturbance - High pressure or shear disturbance - Increased backwash force	- Initiation - Operation - Shut down	- Pressure deviation
Sealing error	Module	Incidental	Operation	- Low reduction level - Pressure deviation
Physical injury	Module	Incidental	- Installation - Operation	- Pressure deviation
Operative layer injury	Membrane	- Pre-treatment error - Polishing materials - Crystal-shape materials	Operation	- Odd high permeate flow - Low reduction level
Oxidative injury	Membrane	Powerful oxidative agents	Long-term operation	- Odd high permeate flow - Low reduction level
- Colloidal fouling - Inorganic fouling	Membrane	Pre-treatment error	Long-term operation	- Permeate flow decrease - Transmembrane pressure increase
Organic fouling	Membrane	Increased Organics in feed	Long-term operation	- Permeate flow decrease - Transmembrane pressure increase
Lumen fouling	Hollow fiber Membrane	Long-term operation	Long-term operation	- Permeate flow decrease - Transmembrane pressure increase

Crack and breakage	Hollow fiber Membrane	- Hydraulic disturbance - High pressure or shear disturbance	- Initiation - Operation - Shut down	- Odd high permeate flow - Low reduction level
Operative layer injury	Flat Sheet Membrane	Increased backwash force	- Operation - Shut down	Low reduction level
Compressing	Flat Sheet Membrane	- High pressure - Increased pressure shock	Operation	Permeate flow decrease

There is distinguish between membrane aging and failure, while the membrane aging makes the grounds to lowering permeate rate and separation level, and the consequent membrane failure. Aging would be rooted in two main sources including before and during filtration [64].

The summarized reasons for the membrane aging and the consequence membrane failure mode [65], based on the mentioned classification, are shown in Figure 58.

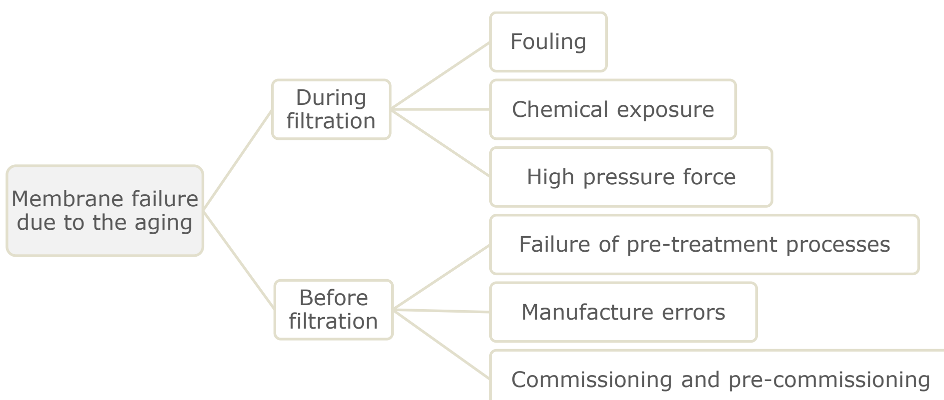

Figure 58 Classification of membrane failure reasons [65]

6.2 Failure mechanism

Regarding mentioned membrane failure reasons, some of the important ones can be scrutinized briefly in terms of the failure mechanism as Table 13.

Table 13 Membrane failure reasons, agents, mechanisms, and impacted positions in the system [53]

Failure reason	Agent	Mechanism	Impact position
Chemical exposure	ClO^- - Chemical cleaning by NaClO - Long-term exposure - High concentration exposure - Side-products - Interaction in feed water	$NaClO + H_2O = HClO + NaOH$ $HClO = H^+ + ClO^-$ - HClO is a powerful oxidative acid, low pH can bring about appropriate for ClO^- generation	Membrane physical and chemical change
Module design	High pressure and wrong; - Membrane type - Membrane packing method - Sealing method	High pressure makes stress leading to: - Detached membrane layers - Membrane cause breakage - Module seal deterioration	- Casing - Sealing

Operational Disturbance	Stress force by air scrubber (applied for wiping foulants)	A greater force of scrubbing - Leads to vibration	Membrane
Bio-fouling	High pressure backwashing	- Reaction of chlorine residual (used as disinfector) and organics in feed water - As the solution, applying high-pressure backwash for bio-fouling reduction	- Module - Sealing sides

Chemical exposure	•Applying chemical cleaning by strongly exclusive study of side-products depends on each feed water
Module design	•Characterization of the exact amount of stress level in membrane module design
Operational disturbance	•Considering already hydrostatic pressure on the membrane by feed water and modify scrubber pressure
Bio-fouling	•Applying $NaHSO_3$ prior to the membrane to neutralize chlorine leftovers •Considering producer recommendations for backwash force circumstances

Figure 59 Membrane failure mitigation approaches [53]

Failures rooted in any disruption of other prior processes, such as pretreatment procedures, are not considered in the section due to the numerous not-straight impacting factors. Moreover, there exist mitigation considerations regards to the mentioned important failure cases as Figure 59 [53].

6.3 Resilience

Membrane technologies are supposed to do the filtration continuously by entering the feed and introduce sustain filtered water, however, even in a continuous process, there is a need of times that the membrane would go out of services. The mentioned times are rooted in different situations which can be wanted or unwanted like the need for equipment maintenance. In the study of membrane failure, the study of membrane resilience can help to control, anticipate and manage production holding times since the failure is the most unwanted reason for that [66].

In this way, resilience generally refers to the operation potential for bear interruptions and coming back again in the normal performance. As a result, resilience is a key agent to guarantee demanded continuous pure water quality and quantity based on regulations. In this matter, reliability and maintainability are pivotal factors to consider.

6.3.1 Reliability

Reliability defines as a parameter that explains how much the treated water quality provided by the membrane system follows expected thresholds. In other words, reliability gives the promise to reach the expected water quality by applying membrane technology. For instance, the extent that the membrane technology can guarantee a satisfying level of virus reduction in a water plant is based on the reliability study.

In this way, reliability can be achieved in the process design to catch the regulation limitations for filtered water [66]. In the reliability assessment, some parameters which are listed and defined in Table 14 be evaluated. Also, the failure rate trend named as the bathtub diagram, and three categorized phases are illustrated in Figure 60.

Table 14 Membrane reliability parameters and definitions [66]

Reliability parameters		
Mean failure time	Mean time to failure (MTTF)	The system cannot work by failure MTTF = Tot operational time of system / No. of equipment
	Mean time between failure (MTBF)	The system can be restored by repair MTBF = Tot operational time of system in service / No. of failure
Failure rate	Failure rate increase = reliability decrease	Phase 1. Immature tiring • The high number of failure due to trial and error, and insufficient control in pre-commissioning and commissioning • Leads to identification and control failure agents
	Estimation based on the bathtub diagram (Figure)	Phase 2. Inherent accidental • No considerable fluctuation • Led by operation mistakes; staff or pre-treatment errors
	Including three phase	Phase 3. Exhausting • Physical and chemical membrane exhaustion in long-term operation • End of life-span • Ultimately, LRVs decrease and membrane breach
Availability	the practical and optimum time that a system is ready to operate with an acceptable performance	Restoration time = holding system time; • Planned maintenance times for risk reduction • Interrupted repair needs happening in the operation
	Operational time of system / (operational time + restoration time)	Availability = MTBF / (MTBF + MTTR)
	Design	E.g. forming the system from numerous small units, modules and membranes lead to an optimized restoration plan

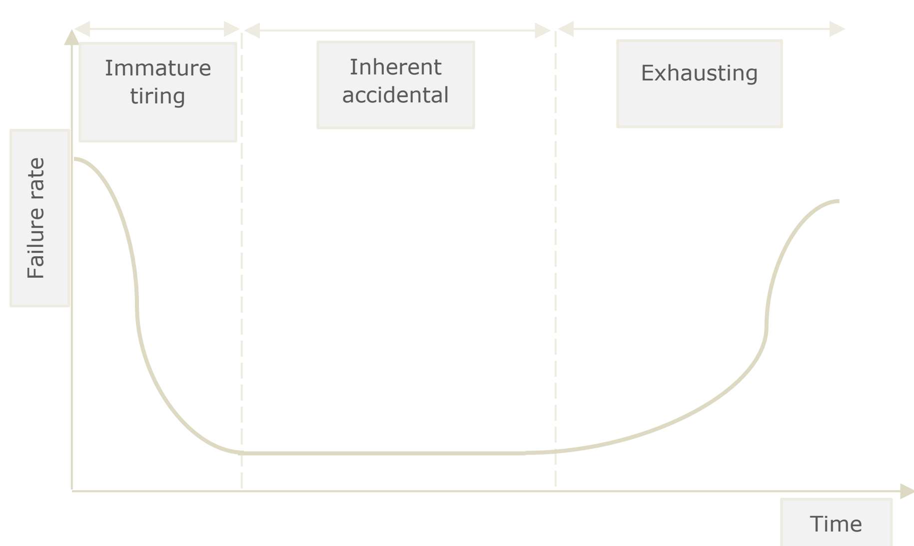

Figure. 60 bathtub diagram; Membrane failure trend and phases [53]

6.3.2 Maintainability

The level of maintainability is an important factor in membrane technologies due to the forcing of the overhaul time into the process [66]. Maintainability assessment can be assigned as the possibility of the membrane restoration back to an operational performance by special care. It can be classified as three sub-majors which are defined in Figure 61 beside the implementation tool of reliability-centered maintenance (RCM) [53].

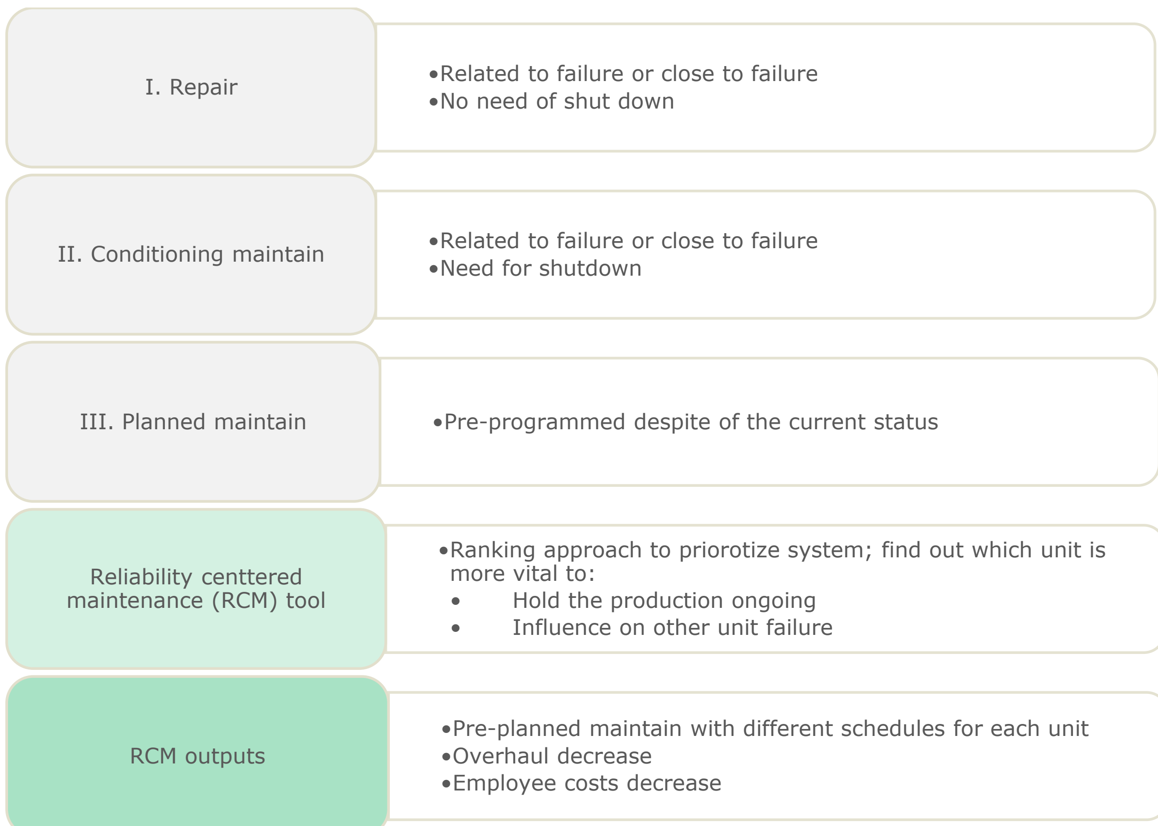

Figure 61 Membrane maintaining approaches and RCM tool considerations [53]

6.4 Membrane Aging Monitoring

As mentioned earlier, membrane aging can alter membrane properties. This phenomenon would impact directly and negatively on the filtered water flow even after cleaning and physical stability of the membrane. Thus, monitoring practices are developed to evaluate effecting reasons in membrane aging [62].

There exist different ways to evaluate membrane aging and failure monitoring. Based on different researches regards to membrane features, they are classified depends on the membrane features such as filtration properties to structural properties and listed as below Figure 62.

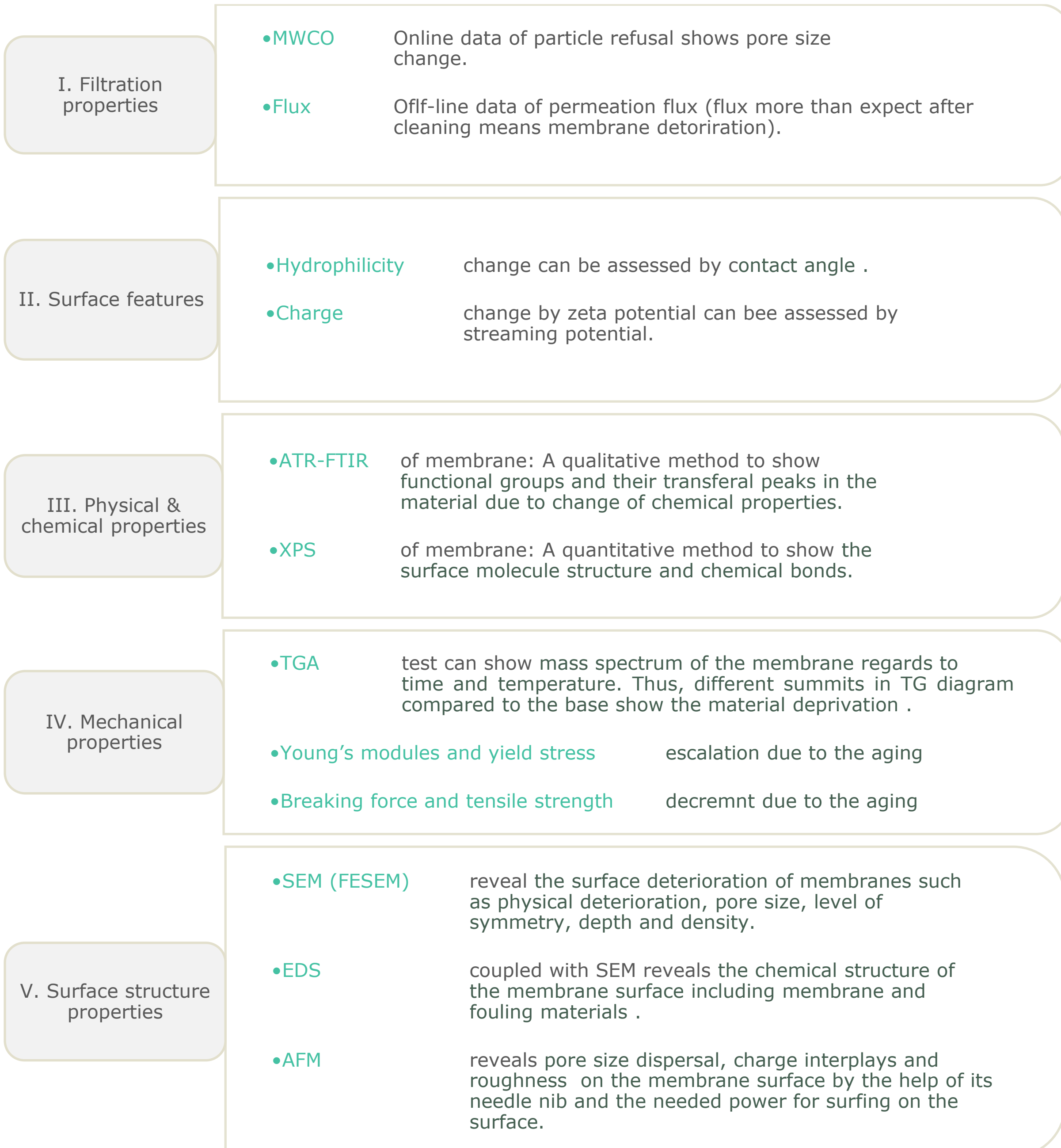

Figure 62 Tools, tests, and parameters for membrane failure and aging monitoring [53]

6.5 Membrane Unity Assessment

As expressed earlier, in the case of failure happening to the membrane system, the membrane unity will be violated. Therefore, filtered water products would be polluted by contaminants such as viruses, and thread the target consumers. As a result, it is essential to scan and control the membrane unity continuously and make sure about the product quality based on the dictated thresholds. In this venue different direct and indirect membrane unity evaluation methods can be applied. The mentioned methods can be explained below.

6.5.1 Direct Assessment

This category of the membrane unity assessment brings the clearest results originated from methods straightly applied on membranes [53]. Direct tools can be explained and compared in Figure 63.

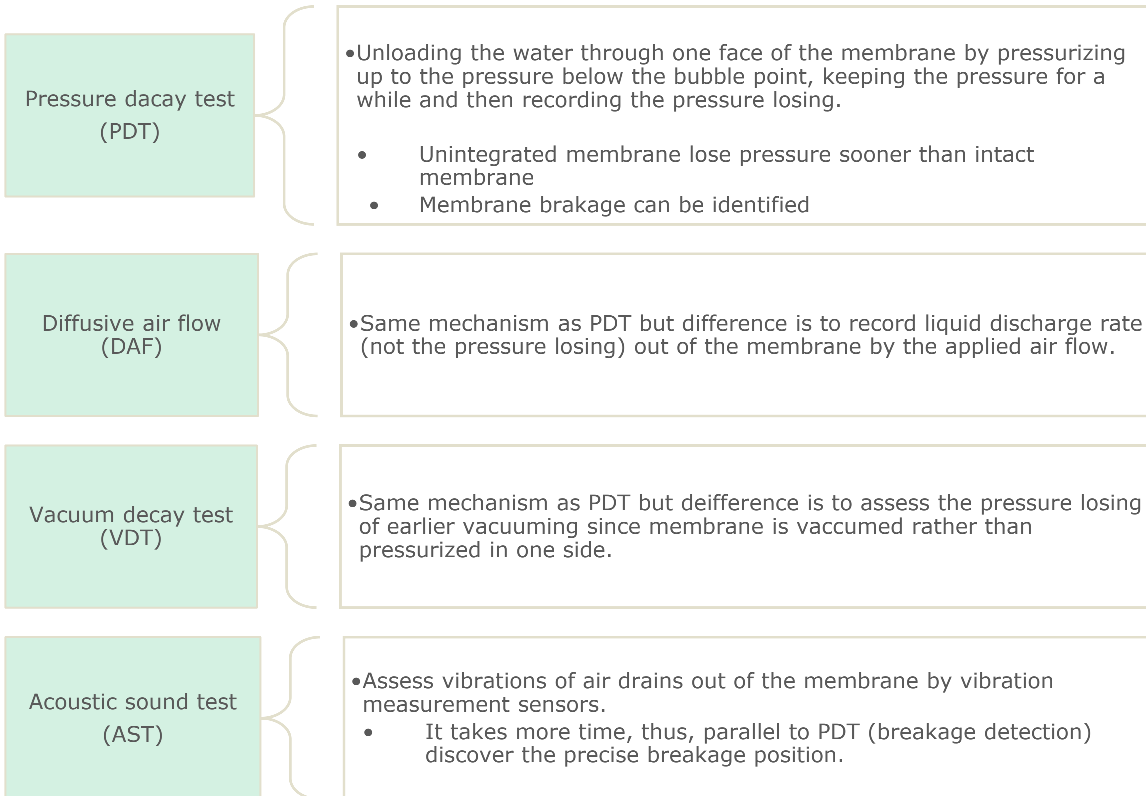

Figure 63 Direct assessment tests for membrane integrity evaluation [53]

By the mentioned mechanisms of the direct membrane integrity tests, Table 15 has listed the advantages and disadvantages of each method for more efficient comparison.

Table 15 Advantages and disadvantages of direct membrane integrity assessment methods [53]

Direct unity assessment method	Cons	Pros
PDT	. Good accuracy . Widespread and accepted . Low repairing needs . Simple performing . Not impacted by the feed quality	. High-pressure need . Off-line test (holding operation time) . Aged or not moisture membrane breakage possibility . Restricted by bubble point of the membrane . Not issuing the permeate quality data
DAF	. More accurate than PDT . Simple performing . Not impacted by the feed quality	. Off-line test (holding operation time) . Less applicable in full-scale due to extra tube lines . Not issuing the permeate quality data
VDT	. More accurate than PDT . Less harmful (no air inducing) . Not impacted by the feed quality	. Off-line test (holding operation time) . Tough performing in large-scale . Not issuing the permeate quality data
AST	. Highest accuracy . Simple performing . Online test . Not harmful for operation	. Conditional to environment noise . Conditional on superposing of waves . Need of trained performer . Unpleasant sound making . Not issuing the permeate quality data

6.5.2 Indirect Assessment

Indirect methods benefit from the assessment of a distinctive index in treated water to evaluate the membrane unity level. Thus, the condemned assessment of the index needs to be precise which detects any tiny change in the index leading to guarantee the demanded quality for the product. Different approaches in this matter are defined in Figure 64.

Particle quantity	•The qauntity exploration of particel numbers by the light scattering laser method for a determined size of water before/after filtration.
Particle quality	•The quality exploration of particel numbers by the light scattering laser method for a determined size of water before/after filtration.
Turbidity	•The membrane performance assessemnt by NTU number since the mebrane is responsibel for suspended particle removal
Phage and Spore tests	•The membrane performance assessment based on microbe removal by releasing surrogates like bacteriophages in feed water and monitor level of microbes in filtered water
Powdered Activated Carbon (PAC) Test	•Same as phage and spore test but uses PAC instead of surrogates
Fluorescent / Magnetic Particles Tests	•Same as phage and spore test but uses magnetic or fluorescent forms of surrogate

Figure 64 Indirect assessment tests for membrane integrity evaluation [53]

Considering the mechanisms of the indirect membrane integrity tests, the advantages and disadvantages methods can be assessed followed by Table 16.

Table 16 Advantages and disadvantages of indirect membrane integrity assessment methods [53]

Indirect integrity assessment method	Cons	Pros
Particle quantity	. Online test . Simple performing . Not impacted by membrane configurations	. Low accuracy . Impacted by the feed quality and operation . Impacted by bubbled air in filtered water . Rather expensive to apply . Accuracy depends on particle size or its dispersal status . Decrease lower-limit of particle counting to make it cheaper
Particle quality	. Lower price than quantifying . More accurate than turbidity . Followed by particle quantifying	. Unpopular in water plants . Followed by particle quantifying . Low accuracy (working with a tiny range of numbers compared to membrane numbers; e.g. changing hundreds of membrane fibers, changes less than twenty-thousandths NTU)
Turbidity	. Popular / accepted in water quality assessment . Followed by particle quantifying	. Followed by particle quantifying
Phage and Spore	. Online test . High accuracy . Issuing real LRVs .	. Time-taking microbe cultivation and assessment . Not reflect current integrity assessment . Impacted accuracy when microbe removal is related to deactivation biologically, absorbing on the membrane surface, or piling up in the filtered water pipes. . Fouling impacts
PAC test	. Online test . High accuracy	. Fouling effects The high required amount of PAC . Impacted by same-sized PAC particles and particle size dispersal
Fluorescent / Magnetic Particles Tests	. Online test . High accuracy . Virus-scale fracture identification . Not impacted by the feed quality	. Not well-founded method . Fouling effects . Not applied in full-scale

6.5.3 Major Disadvantage of the Membrane Integrity Tests

The earlier described methods for membrane unity detection and control benefit from different in-site scanning. However, the result of tests would be issued after off-site assessment such as particle counting. The matter is that even by make the off-site assessments shorter and easiest, the detection philosophy is based on the fact that the membrane deterioration would have happened earlier than the test result [53]. In other words, the membrane unity tests would not notify future integrity breakage but they would detect the breakage which is already happened.

Therefore, besides all mentioned disadvantages the scanning tests suffer from not being online and continuously applied in different steps of the membrane system. In this matter, as a recommendation, there exist rooms to develop not-harmful and may be computerized-based programs to assess the membrane age continuously in operation.

6.5.4 Novel Finite Element analysis (FEA) Method

Based on the previous recommendation, a promising method for the membrane unity evaluation would have the capability to anticipate future membrane degradation precisely. In this venue, it is reported that finite element analysis (FEA) can be a helpful implementation [53].

FEA is a famous modeling program method in engineering. This method is significantly beneficial for structural assessment in different fields. The advantageous FEA method can break complex structures into small units and provide different sub-equations regards to physical structure. Then FEA can create a generalized assessment for the whole system (an example of FEA visual outputs as the stress distribution for an airplane in Figure 65).

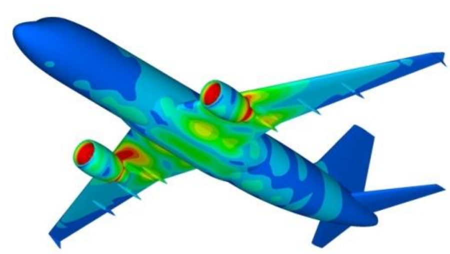

Figure 65 Examples of FEA visual outputs [67]

Moreover, FEA can simulate the operational circumstances and operate hypothetically based on prior aged and failed membranes. As a result, FEA can suggest that which part of the membrane system would be more vulnerable to failure and make the ground for the membrane modification scenarios. In a nutshell, FEA method development in membrane systems can be the most novel complementary tool besides other surrogate-based tests for more enhanced membrane failure detection and control.

6.5.5 Membrane Failure Strategies

As expressed earlier, the membrane failure can be rooted in combined or individual agents in/out the system sourced. For instance, membrane aging is the most responsible reason which results from fouling and operational errors mostly. Therefore, a promising method to prevent membrane failure would be in common with fouling and aging mitigations explained in earlier sections.

In this matter, the membrane amendment to make the membrane flexible against aging agents is on the numerous research studies to introduce modified membrane structures.

Regarding the plant design view, there still one process concept which is classified between mitigation methods and membrane substitution. The approach is the spare design which can prevent halting the production [17].

The Membrane Spare Design Approach Figure 66 can be classified, justified, and assessed spare approach briefly [53].

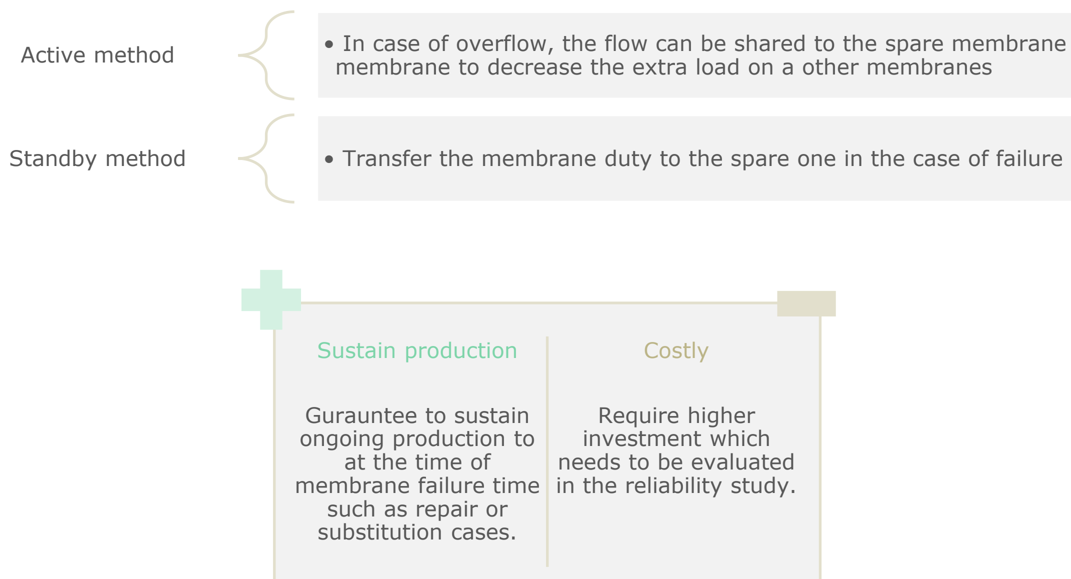

Figure 66 Membrane spare design classification and assessment [53]

The membrane spare design approach can be defined in the membrane system design to locate parallel spare membranes to share or transfer the duty from one membrane to another one.

SUMMARY

Global water scarcity is anticipated to be widespread at a fast rate. Water treatment and recycling are environmental solutions in essence that can be addressed more efficiently by applying sustainable water purification technologies such as membrane systems due to significant approved eco-friendly separation methods.

The first objective of the current study has been nailed by introducing and comparing different membrane modules, configurations, and fabrication methods. In this regard, polymeric membranes with the amended surface by nanotubes can show the anti-fouling capacity and specific character depending on the application. This would make the membrane more efficient with a higher life-span, and fewer associated costs lower than ceramic membranes with lower fouling but higher cost. Among membrane configurations, hollow fiber type was recognized promising compared to plate and frame, tubular, and spiral wound types, due to lower need of pre-treatment considerations, significant higher recovery rate and better response to cleaning scenarios, lower demanded area, and capital cost. However, the process feed and plant-specific design are essential for the membrane type choosing.

The further goal of the study was achieved by exploring MF/UF membranes that can filter suspended and colloidal particles, bacteria, micro molecules, and proteins (LRVs more than 6), plus, majority of viruses and pathogens (LRVs 2-4), with a lower level of energy demand relatively to NF/RO systems. Furthermore, compact MBR was introduced as a hybrid approach of biological and membrane methods with high efficiency by using MF/UF membranes. Also, hybrid PMR can benefit from membranes to regenerate photocatalysts and attached solar cell systems are fetching to cut the water treatment energy demand.

MF/UF membranes are widespread due to their vast applications in water treatment by working solely or in IMS concept prior to NF/RO systems. Thus, the next output of the study was to discuss major IMS advantages over the conventional method besides design considerations. Based on the thesis author view, several MF/UF roles and advantages in portable and decentralized water treatment have been promised an excellent solution for the remote-controlled system in far-fetched areas which are detached from pure water distribution.

By reviewing the overall advantages and disadvantages of MF/UF membranes, fouling and CP were recognized the most limitation factors that make the membrane vulnerable to be aged and substituted in the failure case. Thus, the core objectives of the study were to scrutinize fouling, aging, and failure processes including identification of reasons, impact factors, indicators, controlling and mitigation methods alongside advantages and disadvantages of proposed solutions.

The study has been investigated the water feed water constituents and their impacts on the fouling degree in-depth. Fouling and aging causes and effects have been scrutinized comprehensively. It has been established and examined that NOM, specifically humic substances, and biopolymers (proteins) control key roles in the fouling degree and fouling mechanisms have been covered.

The assessment tools for fouling and aging level were part of the study objectives. Therefore mentioned tools such as cleaning rate, clean resistance, hydrophilicity, HAs, etc.

have been identified and elaborated. Regarding fouling mitigation methods variant cleaning scenarios such as backwashing, air sparging (cycled or full), chemical conditioning, membrane modification, etc. have been assessed by the mentioned fouling indicators.

Based on the authors' viewpoint, firstly, coagulation pretreatment might have better efficiency for fouling control if it is combined by flocculation and sedimentation steps (other than sole coagulation or coagulation with flocculation). However, the key role in this matter is the floc size control influenced by the membrane character (pore size) and fee water constituents.

Secondly, the membrane surface amending by fabrication of more hydrophilic membrane (anti-fouled, since the majority of fouling agents, are hydrophobic), prepare the surface membrane with the opposite charge of foulants, decrease surface roughness, etc. might be a more environmentally friendly method than adding pre-treatment steps to membranes.

Thirdly, novel membrane designs like GDM/PGM system have the excellent capability to be anti-fouled by the impact of the formed biofilm layer on the membrane surface and less need for energy by using weak hydrostatic pressure. However, the permeation flow in GSM/PGM is lower than in traditional membrane systems.

With regards to the aging assessment purpose, repetitive chemical exposure due to the cleaning of irreversible fouling of membranes is recognized as the crucial major. By defining the aging indicators, it has been concluded that the cycled-aging method can mimic the full-scale conditions in lab-scale researches more than the single-step aging method. However, none of them can stimulate a significantly real aging mechanism efficiently since real aging happens during years in full-scale plants. For instance, it has been proved that lower concentrated chemical exposure of membranes in the long-term is more harmful than high concentrated exposure in a short time (lab-scale) due to the explained concept of C*t. In addition, skipping whole affecting operational parameters and the real number of membranes in lab-scale aging studies, still limit a clear aging anticipation algorithm through different studies.

The side-objective of the study was exploring the case study of aged membranes from eight different Canadian treatment plants with more than 5 years of age. Mentioned membranes study has alarmed the possible breakage and deterioration soon by the fouling rate, low surface, and bulk HAs, and deficient mechanical steadiness trends.

The last goal of the study to explore membrane failure resulted from numerous agents and their mechanisms have been identified through inside or outside the membrane system. In this regard, terms of reliability (how the membrane system is reliable to operate continuously), availability, and maintenance have been formulated and explored. An experimental bathtub curve that shows the failure rate during the time from the membrane commissioning to the end of the life span was helpful for further failure control like pre-planned maintenance planning. In this venue, the RCM tool can prioritize different sections in the system regards to their priority to go under repair procedures. Also, active or standby spare membranes can be one of the failure prevention strategies.

To the author's view, since any actions towards the membrane failure need to be forced after the membrane integrity detection, among varieties of defined direct tools, AST coupled with PDT had the more accurate anticipation result. Also, in respect of indirect tools, Fluorescent/Magnetic particle tests are more precise than other investigated methods.

However, the whole currently applied integrity methods are suffered from aging detection after the incident not as online predictor approaches.

As a result, the most novel method required to be developed has been introduced as FEA which can hypothetically use computerized simulation of any structure in the operation including membrane units. Therefore, it can forecast the membrane failure prior to the failure and provide better design and preventive procedures effectively.

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