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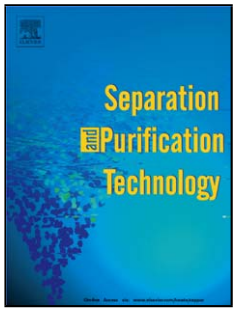

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Chemical cleaning of potable water membranes: a review

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Abstract

The literature on chemical cleaning of polymeric hollow fibre ultrafiltration and microfiltration membranes used in the filtration of water for municipal water supply is reviewed. The review considers the chemical cleaning mechanism, and the perceived link between this and membrane fouling by natural organic matter (NOM) – the principal foulant in municipal potable water applications. Existing chemical cleaning agents used for this duty are considered individually and their cleaning action described, along with the most commonly applied cleaning protocols (i.e. the cleaning conditions, cleaning sequence and method of cleaning agent application).

It is concluded that chemical cleaning is poorly understood and not extensively investigated, in marked contrast to the much more widely studied area of membrane fouling generally, for which there are thousands of published studies. Studies of chemical cleaning specifically have instead been generally limited either to qualitative measurements, such as the use of surface or other analytical tools to characterise membrane foulants and record their removal, or incidental permeability recovery recorded from cleaning events during pilot or full-scale trials. It is proposed that a chemical cleaning index is needed, analogous to the recently proposed general membrane fouling index, based on empirical data to inform cleaning protocols for specific duties and feedwater quality.

Keywords: Chemical cleaning; microfiltration; ultrafiltration, potable water

1. Introduction

There is considerably less literature dedicated to membrane cleaning than to fouling [1,2] particularly regarding either theoretical development or municipal applications; this is possibly because membrane cleaning is considered a low-cost activity (compared to, for example, pre-treatment). A review of the scientific and technical literature on *Scopus* reveals that of the 1200-or-so papers on microfiltration or ultrafiltration identified when these terms are combined with those relating to drinking water supply ("potable", "drinking", "freshwater" and "raw water"), 279 included the subject of fouling with 80 of these including the word "fouling" in the article title. Using the same terms and substituting the word "cleaning" for fouling the title search yielded only 9 results. Whilst this is far from

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comprehensive, with many key papers (Table 1) left unidentified by such a "blind" search, the outcomes are none-the-less indicative of the lack of attention paid to membrane chemical cleaning in the municipal sector when compared to fouling.

The earliest particle fouling modelling studies were based on colloidal particle trajectory analysis with Darcian relationships and Hagen-Poiseulle flow [3]. These models assumed homogeneous fouling and pore size and distribution characteristics and did not relate well to field observations. Fane and Fell [4] discussed the properties of ultrafiltration (UF) and microfiltration (MF) membranes and related these to flux decline behaviour, but the discussion of fouling control mechanisms was not oriented towards development of practical options such as chemical cleaning. With uptake of potable membrane water treatment plants increasing from the late 1990s, researchers began to relate permeation to the characteristics of natural organic matter (NOM), a key component of most source waters, with many feed water characterisation studies aimed at identifying and understanding the behaviour of major NOM foulants [5-7]. This work has extended through prediction of fouling [8,9] and amelioration through pretreatment [10], and membrane material surface modification [11].

Early modelling investigations revealed inconsistencies between fouling from natural raw source waters and that from model studies [12]. It has since become apparent that not only NOM but also extracellular polymeric substances, or EPS [13,14] are possible key foulants; the complexity of organism behaviour and refouling tendencies makes cleaning science development critical to operation. With increasing numbers of membrane installations, and commensurately more related studies providing more information, modelling based on fouling mechanisms has become more sophisticated, incorporating observed impacts of membrane morphology on heterogeneity of the foulant deposition layers [15] and the morphology of fouling itself [16]. Such models have mainly been based on laboratory work using analogue foulants, dedicated to either a fundamental understanding of fouling and/or the development of fouling resistant membranes [17], or through mitigation of adsorption properties of known problem foulants [18,19]. From the turn of the millennium there has been increased study of patterns of foulants with variability in cleaning and backwashing. Combinations of operational and material conditions, such as the impact of surface modification of membrane materials [20], have been investigated aimed at predicting fouling behaviour.

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Across the entire range of UF and MF, studies of cleaning have been secondary to developing an understanding of fouling; cleaning studies themselves have been largely limited to physical cleaning (i.e. backflushing, or reversing the flow back through the membrane, and relaxation, or temporary cessation of permeate flow). Notwithstanding the greatly improved understanding of fouling, there remains a challenge both to develop the techniques and regimes for their effective and efficient removal; the same level of understanding for cleaning as exists for fouling needs to be attained.

2. Membrane chemical cleaning

2.1. Definition

The simplest definition of cleaning is a procedure applied to a material to relieve it of all non integral substances which are generally termed "foulants" [21]. Fouling is thus the general term for the decline in flux, or more accurately permeability, with time. The foulants may be reversible or irreversible, the latter causing permanent changes in the membrane performance [22], and cleaning thus aims to remove all foulants either by physical, chemical or biochemical means [23].

2.2. Mechanism

Membrane chemical cleaning can be envisaged as a 6-step process [24,25]:

1. bulk reaction (hydrolysis and other) of cleaning reagents as the CIP is introduced,
2. cleaning agent is transported to membrane surface,
3. cleaning agent transits through foulant layers to membrane surface,
4. cleaning reactions solubilise and detach foulants,
5. waste cleaning agent with suspended foulants transported to interface, and finally
6. transport of waste matter to the bulk solution from retentate side of membrane.

A *de facto* accepted conceptual model for cleaning and fouling was presented by Liu et al. [26], which links the major fouling and cleaning forces. The foulant and membrane interact mainly through hydrophobic attraction; the mechanism for cleaning is primarily electrostatic repulsion. An understanding of the foulant chemical properties expedites the selection of cleaning chemicals, since chemical effects are largely understood: fouling by potable waters is increased at higher foulant molecular weights and charge ratios, the presence of divalent cations in the water and membrane hydrophobicity [16,26]. An increase in the cleaning

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medium electrostatic potential through charge density, polarity or pH thus suppresses the attraction forces and increases cleaning efficacy.

2.3. Plant operation and design.

Cleaning is applied routinely via a chemically enhanced backflush (CEB) or a clean in place (CIP). A chemical reagent is introduced into the permeate side of the membrane; the lumens for in-out capillary tube (CT) or the module shell for out-in hollow fibre (HF) membranes. For a CEB, the chemicals are introduced with the routine backflush. CEB is used for stabilising/maintaining the permeability (and is often referred to as "maintenance" cleaning) by preventing foulant build up, and is automatically scheduled daily to weekly with no manual intervention. The CEB reagent concentration is relatively low, soak times relatively short and application temperatures normally ambient, compared to a CIP where the objective is to recover the flux and thus requiring longer soak times and higher cleaning agent concentrations and soak temperatures. CIPs are normally applied on an ad-hoc basis in response to excessive permeability decline, and can involve a sequence of cleans using different reagents (normally basic, oxidative and acidic).

Studies of cleaning efficacy across 21 existing full-scale potable water MF/UF plants [27,28] revealed differing sustainable fluxes according to the level of pretreatment, backflush cycle time, chemical cleaning cycle time, cleaning temperature and, in particular, design flux. It is now generally recognised that the capital cost advantage offered by higher-flux operation is more than offset by the greater diligence demanded with cleaning, the reduced cleaning cycle time, and performance and life of the membrane [29]. In a more recent and extensive survey of 87 plants [30], very diverse cleaning practices were revealed. Backflush frequencies varied from 5 to 96 per day with a median of 32 and lasted from 10 seconds to 10 minutes with a median of 77 seconds. Less than half the plants surveyed carried out maintenance cleaning on average more than once a week. The CIP cleaning frequency ranged from 0.2 to 50 per year with a median of 4 per year. Results were skewed by the under sizing of some plants, confirming findings of water industry wide research [2] regarding flux sustainability.

2.4. Cleaning reagent performance.

The primary barrier to effective chemical cleaning is thought to be mass transfer, the second step in the cleaning mechanism (Section 2.2.), where the chemical is prevented from reaching the foulant at high enough concentrations to overcome the attraction forces [26]. Figure 1 shows this electrostatic model schematically. Since, in the electrostatic equilibrium model,

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forces retaining the foulant at the membrane are reduced during cleaning to allow its physical removal, it follows that the most appropriate cleaning agent will depend upon the nature of the foulant, i.e. organic/inorganic, acidic/basic, and the charge state. It is possible to envisage the transport mechanisms using a simple stage model shown in Figure 2. The accepted physico-chemical mechanisms attributable to the most commonly available potable membrane cleaning agents are summarised in Table 2.

2.4.1. Caustic soda

Caustic soda (sodium hydroxide, NaOH) solutions are applied mainly at pH levels of 11-12, or less if membrane chemical resistance is an issue (as is the case with polyvinylidene difluoride, or PVDF). Hydroxide encourages dissolution of weakly acidic organic matter, generally with carboxylic and phenolic functional groups, and promotes cleavage of polysaccharides and proteins into smaller sugars and amides [31,32,33]. Hydroxide also expands NOM molecules, allowing enhanced mass transfer of the cleaning agent solution to the membrane surface [34]. It may also be effective for removing inorganic colloids and silicates where charge effects and ionic strength increase the solubility and electrostatic repulsion [35]. Permeability recovery tends to increase with NaOH concentration up to a threshold value which varies for different foulants and membrane materials [26,33,35,36], application [37] and the degree of fouling [22].

2.4.2. Oxidants

Oxidants include hydrogen peroxide (H_2O_2) and sodium hypochlorite (NaOCl), of which the latter is by far the most common. However hypochlorite is not universally applied since (a) some polypropylene (PP) membranes are not chlorine tolerant [23], and (b) chlorinated organics are generated which can have health and environmental impacts which then may restrict the use of chlorine based reagents. Oxidation degrades the NOM functional groups to carboxyl, ketonic and aldehyde groups, which makes them more susceptible to hydrolysis at high pH levels [38]. This may explain why the combination of oxidant with alkaline cleaning agents has been shown empirically to be more effective than oxidant cleaning agents alone, especially where organic foulants dominate [34,39-41].

In a comparison study between alkaline H_2O_2 and NaOCl cleaning of NOM-fouled PES UF membranes, NaOCl was found to provide better removal of organics through oxidation of

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aromatic humic substances at elevated pH levels [39]. This was not repeated for peroxide. The halogenation of the aromatic rings is known to be a common mechanism for the breakdown of humic materials [42], furthermore, the Strugholtz study [39] appears to suggest that the oxidative properties of the hypochlorite may be more significant than the hydrolysis catalysing effect of the hydroxide. Liu et al.'s study (2001) [26] indicates that caustic promotes a more open fouling layer that allows the chlorine to reach the membrane surface, which may explain the increased efficacy of hypochlorite at higher pH levels. Huang et al., (2008) [43] showed that free chlorine affects oxidation of organic colloids while the soak time governs their diffusion from the membrane surface. Chlorine is thought to contribute to membrane swelling, which generally enhances mass transfer of the cleaning agent to the foulant at the membrane surface [22].

Clearly, it is advantageous to establish the optimum concentration of caustic soda and hypochlorite for membrane cleaning, since excessive concentrations of hypochlorite are undesirable on the basis of cost and loss of membrane integrity. At concentrations above 150 $mg.L^{-1}$ and low-neutral pH levels, chlorination of the C-S bonds in PES has been observed [44], and the alkaline tolerance of PVDF membranes is limited to pH levels below 11. However, the variation in permeability recovery from these studies indicates that cleaning efficiency cannot be estimated solely on the basis of the chemistry of the foulant, cleaning agent and the membrane material - a conclusion arrived at from a recent survey of plants in the UK [45].

2.4.3. Acids

Acid cleaning aims to remove multivalent cationic species such as in hardness salts and metal hydroxides [21,22]. Mineral acids, predominantly hydrochloric (HCl) and sulphuric (H_2SO_4), are commonly used, due to their low cost, and are effective for both CEB and CIP down to pH levels of 1.0 for PES and PVDF, below which integrity problems may arise. As well as catalysing acid hydrolysis, acids are mildly oxidative for NOM, forming soluble aromatic aldehydes and acids at NOM functional groups [38], but are generally used for removal of mineral scaling.

Whilst strong mineral acids can solubilise deposits, organic acids, such as citric and oxalic, are more effective for formation and transportation of organo-metallic foulants from the

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membrane to the bulk solution [39]. Citric acid provides buffering and has good chelating abilities, making it effective and easy to use with a lower risk of pH damage compared with mineral acids [29]. Along with NaOCl, it is reported to be the most used chemical cleaner for MF/UF membranes [30]. Citric acid can disrupt biofilm formation by removing minerals from foulant layers [46], disrupting enzymes that allow their establishment [47,48]. Iron readily forms complexes [23] but their limited solubility inhibits their transport from the membrane surface [22]. The combination of citric and oxalic acid has proved effective in practice where multiple organometallic complex species can form [49], supporting observations from cleaning effectiveness studies where combinations of cleaning agents in multi stage applications proved more effective and high permeability recovery more attainable despite incomplete foulant removal [48].

Phosphoric acid (H_3PO_4) is another chelating ligand providing reasonable removal of inorganic scale but, in common with most chelates, has poor removal of elemental sulphur and colloidal silicates [47]. As with citric acid, its buffering capability and sequestering ability make it potentially useful for routine automated cleaning or even CEB where the risk of overdose and damage are lessened. Its use has been reported in very few MF/UF cleaning papers; it is recommended only for some proprietary industrial membranes [21] and, most usually, for cellulose acetate RO membranes fouled with metal oxides [33]. It is apparently less effective than other cleaning chemicals against NOM [41].

2.4.4. Others

Cleaning additives such as EDTA (ethylenediamine tetraacetic acid) provide additional chelation capacity for metals such as calcium and the dispersal minerals in general [22,26,41] EDTA is often blended into proprietary cleaning agents as an enzyme disruptor (such as *P3 Ultrasil 11*) and studies have demonstrated they inhibit biofilm reformation [25]. Enzyme cleaning agents have been applied on some PP membrane-based US plants where they are shown to function at temperatures up to 45°C [27]. However, its general use in potable water is constrained both by cost and potable water legislation.

Ammonium bifluoride (NH_4HF_2) has also been used to assist the removal of silica deposits [29], though silica remains one of the most challenging of foulants since it forms inert silicate colloids with high surface attraction forces. Silica is highly insoluble and does not readily

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hydrolyse [50]. It is destabilised only by fluoride, which can displace the counter ion but may also cause damage to some membranes. Ammonium bifluoride is occasionally dosed to break up silica deposits [29]. It is favourable to remove silica through pretreatment [31].

3. Cleaning processes

3.1. Effect of cleaning agent sequence

Cleaning sequence application is known to affect the degree of permeability recovery [22]. Studies where membrane cleaning sequences have been tested consistently indicate that an alkali followed by an acid clean is more effective than the reverse for membranes treating surface water [21,26,33-37,43,51]. This could be explained by the effect of the charge on the membrane surface and foulants (associated with the cleaving of the foulant molecules) following the alkaline clean, along with swelling of both the foulant layer and the membrane at higher pHs. This then enhances the cleaning reagent mass transfer for the subsequent step. However, an acid-alkali cleaning sequence is commonly specified for groundwater sources, since the acid removes inorganics such as metal oxides and carbonates which may otherwise form precipitates at high pH [29]. A reduced temperature sensitivity has been reported for removal hydrophobic NOM, with the acid-alkali sequence [39]. Industrial process waters treating proteinaceous waters routinely employ an acid-alkali sequence to prevent gel layer formation [2] or mineral precipitation [21].

The choice of cleaning sequence is governed by the feed water and the type of fouling. From the literature it appears that a predominance of hydrophobic NOM favours alkali followed by acid, whilst predominantly inorganic scaling or metal hydroxide precipitates favour acid-alkali. Given that both these materials are present in biofilms in varying proportions, and that biofouling can be considered ubiquitous in potable water membrane plants, selection of the most appropriate cleaning sequence is not always apparent on the initial process technology and system selection.

3.2. Other cleaning factors

Increased temperatures decrease the reliance on turbulence to overcome mass transfer limitations, since diffusive mass transfer is increased, and this is seen as an important qualitative cleaning model factor [36]. Foulant solubility is also increased at higher temperatures, as is reactivity of the NOM functional groups [42], and also swelling and

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mechanical destabilisation of organic foulant or biofilm layers [52]. However extremes of temperature and pH can also solidify and crystallise biofilm deposits, and specifically the scalant components of these (calcium carbonate, ferric hydroxide, silicates, etc), making them harder to remove [36]. Membrane integrity also limits the range of pH and temperature, although the latter is determined by the potting material used in the membrane modules rather than the membrane material itself.

3.3. Alternative processes

More benign but expensive alternative cleaners, such as enzymes, are employed more usually for industrial and food waste treatment [18] where added value is higher than for municipal applications: enzymic cleaning is not considered appropriate to bulk potable applications. Ultrasonic cleaning has been used for treating flat sheet membranes in laboratory tests [53] and for membranes fouled from dairy processing operations [54], though this treatment can cause membrane damage and incurs a high energy demand [55]. In this regard there is evidence that some membranes are more susceptible to integrity failure than others: PES materials were shown to fail after 5 minutes of exposure to the ultrasound (at f = 47 kHz). However, more recent studies demonstrated some potential for ultrasonic cleaning in lower flux crossflow systems fouled with dairy wastewater [54]. There is evidently further research needed to establish whether ultrasonically-assisted cleaning has a place in potable water membrane filtration where integrity is critical.

3.4. Cleaning process studies

Given the stated variables influencing cleaning efficacy it is clear that rigorous assessment of cleaning efficacy is challenging, given the heterogeneous and sometimes quixotic nature of fouling. Comprehensive modelling of membrane cleaning demands a large number of experiments to cover possible ranges of membrane fouling conditions from natural waters. An early attempt by Bartlett et al. to quantify cleaning [36] studied the permeability recovery response to cleaning factors for flat sheet membranes fouled with dairy waste. This study examined concentration, temperature, alkali/acid combinations and cross flow effects on different surface concentrations of the same foulant, and concluded that the optimum cleaning response could be reached using different regimes.

Chen et al. [56] carried out the first reported factorial study experiment on a suite of membrane operating parameters, using analysis of variance (ANOVA) on their operational impact. The experiments employed HCl and NaOH to clean spiral wound RO and UF fouled

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with municipal wastewater secondary effluent. Factors considered were NaOH concentration, temperature and recirculation duration, HCl cleaning time and the use of forward backflush and normal backwash after chemical cleaning. The relationships between the resulting recoveries were explored. For this application it was seen that the temperature and concentration of the NaOH were most critical in optimising recovery in combination with the application of a backflush.

This factorising of chemical cleaning variables has been advanced in the research [45, 46, 52, 57] and there also exists possibilities for automating and optimising the cleaning and backwash cycles, as demonstrated for UF plants [58]. The need for controlling cleaning has led to the proposal of dynamic models for predicting optimum soak times [59]. These models have been developed for given specified conditions and mainstream cleaning research remains generally restricted to specific treatment problems using empirical and heuristic data [51]. These should not be conflated with recently suggested generic fouling models [60]. Recent factorial studies of a number of membranes show that simple optimisation based on multiple factors varies between and within the different types and configurations [45] and this suggests cleaning performance optimisation demands an ongoing systems approach.

The poor predictability of full scale systems in practice has led researchers to study neural networks, such as for predicting flux decline in MF/UF systems [61,62]. This approach has been developed further by Strugholtz et al. (2008) [63], where ceramic UF systems were optimised using neural networks incorporating genetic succession algorithms based on the initial operational and raw water parameter states.

4. Discussion and conclusions

Foulant studies, particularly NOM fouling, have dominated research into cleaning but are best described as “a single frame of a moving picture” [38]. It is clear that membrane material choice affects the cleaning agent systems performance but these are prescribed by suppliers from limited information and can therefore serve to limit the scope for ongoing optimisation. Cleaning studies which have evolved from simple mass transport models, based on attachment/detachment force equilibrium models and where the cleaning factors have followed foulant characterisation, have the potential to inform cleaning practice. A variety of tools are becoming more available for characterising systems and the empirical/heuristic

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approach has led to differing interpretations of the relationship between fouling and MF/UF plant design and operation. It has therefore been proposed that unified fouling index (UFI) be developed [60]. A UFI would create indices based on quantification of the fouling potential of commercial membranes by standardised bench tests, and offers advantage(s) of being universally applicable to low pressure (i.e. ultrafiltration and microfiltration) membrane systems for generic potable water types.

Permeability recovery alone is itself insufficient to characterise changes in membrane fouling in response to cleaning [64]. The development of dynamic cleaning models provides a systems approach to cleaning, whereby input variations, surface modification and ageing effects can be accommodated. A quantitative understanding of how the cleaning efficacy changes under different loading and fouling conditions is also required. Advanced use of statistical methods, such as response surface models [65], can be used to generate polynomials based on the key cleaning design parameters (cleaning agent concentration and temperature, CEB backflush rate, cleaning exposure time, etc.), and have been recently employed to optimise membrane cleaning [1, 46]. Whilst the coefficients generated by this method appear to be site specific [45], they may provide a basis for developing a unified cleaning index (UCI) from pilot or continuing bench scale tests alongside the UFI currently being proposed. With amalgamation of such bench tests and real time plant data, heuristic models would be possible that would expand the development of a practical membrane cleaning science. Given that UFI is currently in its infancy, it would seem that there is a considerably more development required in generating a usable UCI for informing chemical cleaning protocols.

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Table 1: Major potable water treatment membrane studies by synopsis

Topic and synopsis	***Papers***
Cleaning review- Industrial applications	[4,18, 21,35 and 37]
Optimisation and control (cleaning scheduling)	[41, 53, 59]
Cleaning reagent review - single plant	[18, 32, 39 and 51]
Cleaning control factor model development	[36, 43]
Cleaning and foulant interactions	[60]
Cleaning - ultrasound	[54,55]
Cleaning and membrane damage	[44]
Cleaning and pre-oxidation	[38]
Membrane cleaning performance	[26]

Table 2: Generic chemical cleaning agents with usual reactions

Cleaning agent	*Chemical*	*Reactions*
Base	Caustic Soda, (NaOH)	Hydrolysis and solubilisation, saponification.
Oxidant	Hypochlorite, (HOCl), Hydrogen Peroxide (H_2O_2)	Oxidation and disinfection
Acid	Hydrochloric (HCl), Sulphuric Acid (H_2SO_4), Nitric Acid (HNO_3)	Solubilisation
Acid Chelate	Citric	Chelation
Alkaline Chelate	EDTA	Chelation
Surfactants	Proprietary	Emulsifying, dispersion and surface conditioning

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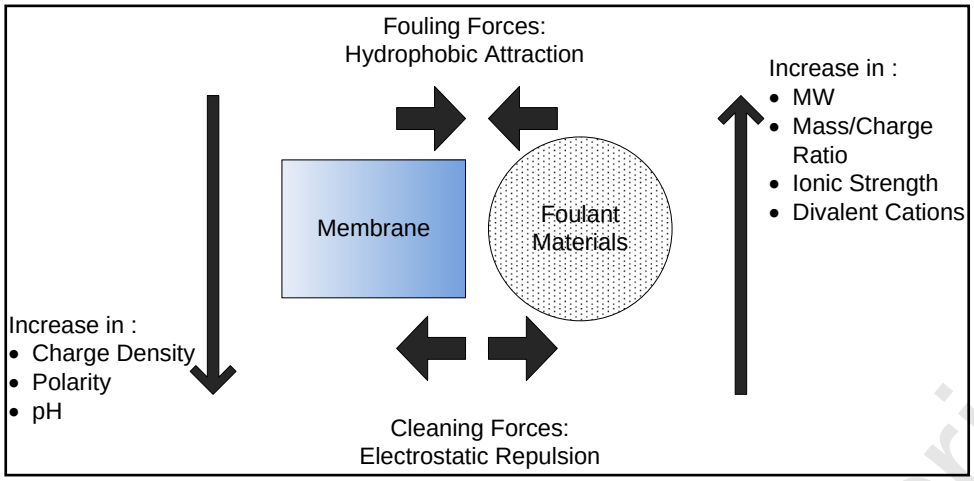

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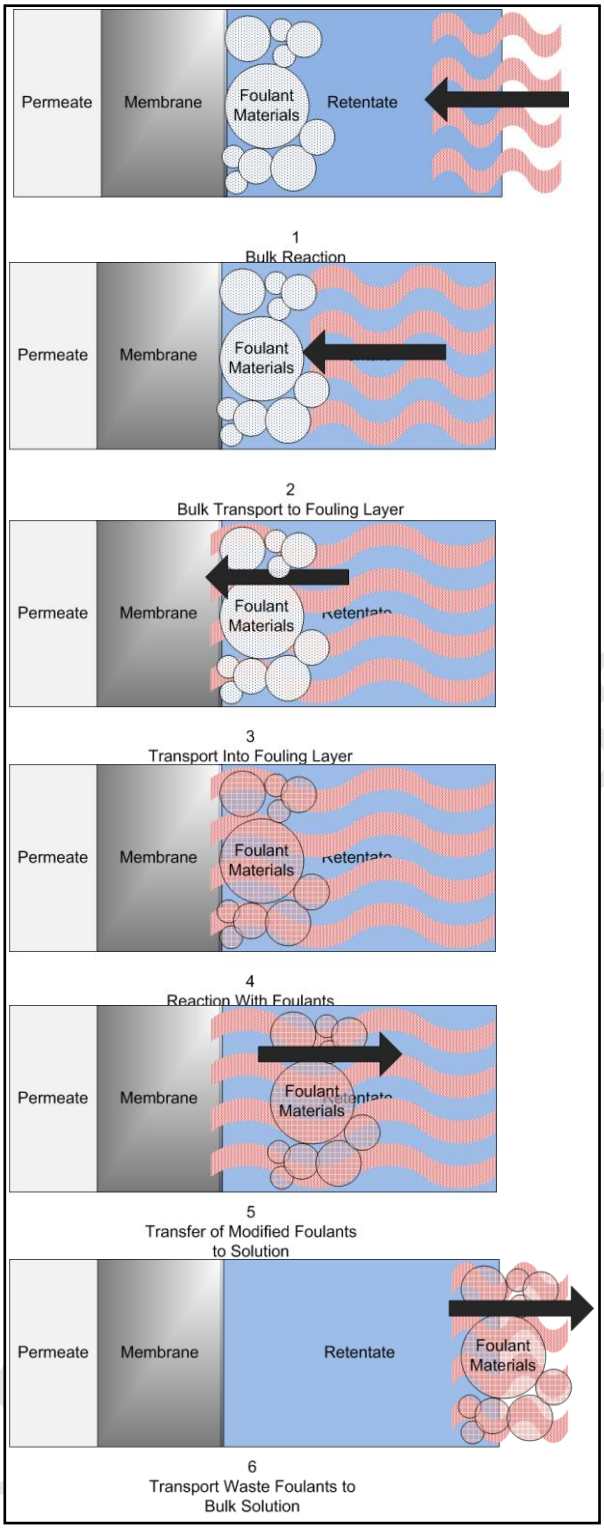

Figure 1: Conceptual electrostatic equilibrium model for membrane cleaning [26]

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Figure 2: Schematic stage model for solution transport

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