On the validation perspectives of the proposed novel dimensionless fouling index

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ABSTRACT

Fouling constitutes one of the main issues challenging the application of membranes in reverse osmosis processes. Serious results of fouling are decrease of the permeation flux through time, decreased lifetime of membranes, more usual washings, and therefore more important use of chemical products and extra energy costs to balance out the decline in membrane permeability. A short discussion of investigations realized on fouling parameters and experiments established is given. This work aims to attract the attention on a dimensionless grouping of a restricted number of parameters that may ultimately play a role of a fouling index upon a wide span of working situations. This is done by a convenient non-dimensionalization of the equation of Ruth. The dimensionless number which is attained is named *dimensionless fouling index* (DFI) and may be explained as the ratio of the membrane resistance to that of the cake due to the concentration of the raw water. The validation of DFI requires doing tests subject to different conditions of membrane type and an extent of particulate and/or dissolved material. Unfortunately, the lack of appropriate equipment and the lack of all needed data to realize the necessary transformations using experimental results found in the literature make this step unattainable at this stage.

1. Introduction

One of the serious difficulties challenging the usage of membranes in reverse osmosis (RO) processes is fouling (Ait Messaoudene and Naceur, 2014; Fane et al., 2009). Fouling is known as the phenomenon conducting to the damage of membrane efficiency (Khirani, 2007). It is generated by the aggregation of matter at the membrane surface (Hong et al., 2009). When a membrane system works at constant trans-membrane pressure, the most intricate repercussion of fouling is the decrease of the permeation flux through operation (Park et al., 2006). Fouling may be considered like an identical extra resistance to run across the membrane. It may as well affect the separation capacity of a membrane performing as a supplementary obstacle which is layered on the membrane. Material aggregation may happen at the surface of a membrane. Particles may obstruct the pores and, in the instance of a membrane with pores wide enough comparative to the matters in the water to be removed, there may be an adsorption and/or deposition of fouling substances on the internal wall of pores. In the fouling models suggested by some researchers and discussed later, it will be shown that scientists usually think that there is primarily a blockage/obstruction of pores, joined by the germination of a cake at the surface of the membrane. This cake is distinguished by a porosity of its own, a specific resistance opposite to flow through the membrane, cohesion, a specific density, and a more or less homogenous granulometry (Tamas, 2004).

As a result, it may be confirmed that fouling is the principal restriction for membrane separation. It is consequently required to understand the different processes implied in addition to the relationships

and models that let characterizing them. It is as well crucial to discuss the state of the art concerning the solutions suggested to that degree to expect, restrict or rectify fouling if practical (Ait Messaoudene and Naceur, 2014).

Fouling has been the focus of countless research investigations on modeling of fouling (Jaffrin et al., 1997; Roehl et al., 2018), fouling indexes (Schippers and Verdouw, 1980; Boerlage et al., 2000, 2002, 2003a, 2003b), or fouling mechanisms (Belfort et al., 1994; Koo et al., 2013; Crittenden et al., 2012).

Successful monitoring of fouling needs a convenient identification of the fouling types that are existent in a particular technique. Natural waters carry a diversity of constituents. The size, nature and physicochemical features of these constituents control the fouling capacity of particular water. The function of a fouling indicator is to merge all these parameters into an only one factor.

Fouling measures expanded heretofore are built on experiments with microfiltration or ultrafiltration (UF) membranes. Nevertheless, a number of interrogations come to light concerning the efficiency of these parameters to anticipate fouling: (1) Can the identical fouling parameter include a large variety of water type? (2) Is the fouling capacity of water an inherent feature of that water or is it reliant on the membrane employed? (3) In the identical view, if the experiment for ascertaining the fouling parameter employs filtration on a specific membrane, to what range is it feasible to expect the efficiency of different membrane? (4) If the experiment is employed to evaluate the fouling parameter employs a dead-end cell, to what level is it practicable to anticipate the efficiency in tangential flow filtration? (Ait Messaoudene and Naceur, 2014).

Even if it is not simple to give a complete answer to these interrogations, an effort is performed to show a list of experiments established and employed yet to examine both their advantages and their restrictions.

2. Survey on fouling index research

In order to better anticipate fouling, Boerlage et al. (2002) have worked largely on the establishment of the modified fouling index-UF (MFI-UF). This technique, employing UF membranes, considers fouling attributed to solids whose diameter is less than 0.05 μm. They examined membranes of two materials (polysulfone (PS) and polyacrylonitrile (PAN)) with cutoff extending from 1 to 100 kDa. The filtration is performed at fixed trans-membrane pressure (1 bar) with an acquisition of filtered volume every minute. They have focused attention on the effect of the experiment temperature, which impacts the viscosity and may importantly modify the experimental findings. With the working procedure explained, the value of the MFI-UF is evaluated for the PAN membranes after 20 to 50 hours of checking. The MFI-UF is not reached in these periods for the PS membranes though.

The values achieved by this protocol change between 2,000 and 13,300 s/L^2 while the existing modified fouling index (MFI$_{0.45}$) (which is based on cake filtration, and uses a 0.45 μm microfiltration membrane to measure the particulate fouling potential of feed water) changes from 1 to 5 s/L^2 for the tap water used. The extremely big values of the MFI-UF illustrate the inefficiency of conventional microfiltration membranes for evaluating the proper fouling potential. These elevated values of the MFI-UF are interpreted by the retention of particles of very small diameter. In fact, the MFI value is inversely proportional to the square of the diameter of the particles forming the cake. The MFI-UF turns out to be weakly dependent on the cutoff of membranes used. Values of 2000 and 4500 s/L^2 respectively were obtained for the PAN and PS membranes with cutoff ranging between 3 to 100 kDa. This conclusion is valid within the range of cutoffs tested and for the type of water used in the tests. Following these tests, a PAN membrane of 13 kDa cut off is proposed as a reference membrane for determining the MFI-UF. The limiting parameter is the duration of the test (over 20 hours).

Another method of determining the MFI-UF at constant filtration flux is also developed. This method is used to determine the MFI-UF in a shorter period (1-5h) (Boerlage et al., 2004). The results also highlight the fact that the value of the MFI-UF at constant flux is stable over a short period of time relative to the MFI-UF at constant pressure.

Using the same assumptions as Boerlage et al. (2002) and Roorda and van der Graaf (2005) defined the normalized MFI-UF, which is calculated with the same experimental formula as that used by Boerlage et al. (2002). The only difference is that they normalized the MFI values with respect to a membrane area of 1m ^{2} and a trans-membrane pressure (TMP) of 1 bar. Instead of following the evolution of the filtrate volume at constant pressure, it is the evolution of the TMP at a constant flow rate which is monitored. The MFI-UF values obtained at constant TMP are higher than those obtained at constant flow rate; this can be explained by the compressibility of the cake.

Khirani et al. (2006) have used NF membranes at constant TMP in order to improve the representativeness of MFI in NF/RO. These researchers showed that dissolved organic matter is responsible for fouling in NF/RO and must be taken into account when measuring the fouling potential. Thus, the NF-MFI allows better reflecting fouling by organic matter and the effectiveness of pretreatment. Determining the fouling index is performed within even shorter time in this case (about 1h).

Choi et al. (2009) have developed a more complex fouling index called *combined fouling index*. This index is based on the implementation of a model in the form of a linear relationship involving different MFI expressions. This is intended to represent various types of membranes (UF and microfiltration) for selective separations of different types of fouling

materials. This fouling index is used to measure the impact of each type of fouling material on the FDR (flux decline rate) in RO/NF as contributing factors to overall fouling involved in the model describing the combined fouling index.

Yu et al. (2010) have developed a new approach to evaluate the fouling potential of water supply in RO and NF. Multiple membranes array system (MMAS), in which MF, UF and NF membranes are connected in series (the selection of membranes is based on the theory of cake filtration), is designed for separating and targeting the different types of fouling matters in water and assessing their fouling potential. Thus, particulate, colloidal and organic matters are separated sequentially by MF, UF and NF membranes respectively. The MFI is determined for each separation resulting in three measurements: particle-MFI, colloid-MFI and organic-MFI. After an optimization of the series of MF, UF and NF membranes configurations, the results show that the evaluation of fouling potential of different types of water (raw seawater, pre-treated seawater) by the MMAS method is more precise. This method gives more information compared to conventional methods (silt density index (SDI), single MFI) (Alhadidi et al., 2011). Fouling potential indexes determined by the MMAS method accurately reflect changes in terms of water quality by the various pretreatments (sand filtration, microfiltration and UF). Traditional measures, on the other hand, are not sensitive enough to detect these changes. It is also shown that fouling potentials evaluated by MMAS are better correlated with the rate of flow decrease determined by RO pilots. This allows stating that the MMAS provides a better prediction of fouling and a better indication for the selection of the appropriate pre-treatment.

A fouling index called Cross Flow Sampler Modified Fouling Index in UF (CFS-MFIUF), measured at constant flow is developed by Sim et al. (2011). This method is capable of capturing particles while simulating the hydrodynamic conditions of tangential flow in RO. Traditional fouling indices such as the MFI are measured during filtration and hydrodynamic conditions of RO are not taken into account. The results of humic acids (Ju et al., 2015) and silica have shown that the CFS-MFI$_{UF}$ is not only sensitive to the presence of colloids in solution but also to organic matter. Indeed, a linear relationship between this indicator and the concentration of humic acids added to the silica is obtained. On the other hand, the comparison between measurements of the CFS-MFI$_{UF}$ and the MFI-UF constant flux (MFI in UF at constant flow) shows that the conventional MFI-UF constant flux tends to predict a larger fouling compared to the CFS-MFI$_{UF}$. The CFS-MFI$_{UF}$ allows a better detection of the improvement in water quality after pre-treatment compared to the MFI.

The CFS-MFI$_{UF}$ predicts a PTM profile in agreement with the actual behavior in RO with only 11% deviation in the absence of dissolved salts. This allows saying that the CFS-MFI$_{UF}$ is a more realistic approach to the determination of fouling potential in RO. By cons, in the presence of dissolved salts, the PTM profile predicted by the CFS-MFI$_{UF}$ corresponds to the formation of a cake added to the osmotic pressure generated by the amount of salts in solution. This is not consistent with the actual behavior in RO and indicates the importance of the effect of the presence of osmotic pressure on the cake formation in RO fouling. This contribution must therefore be taken into account in the measurement of CFS-MFI$_{UF}$ (Sim et al., 2010; Sim, 2011).

Present-day fouling indices usually used in RO practices, like SDI and MFI, have been criticized considerably due to their incapacity to anticipate de facto fouling capability firstly because of the inaccurate explanation of fouling processes (Jin et al., 2015). Jin et al. (2015) intelligibly established that the impact of pore blocking must be omitted throughout fouling index quantifications to model actual RO usages. Therefore, Jin et al. (2015) suggested novel notion of cake fouling index (CFI) with view to minutely calculate correct fouling cake layer resistance. Precisely, the CFI was determined during successive filtration experiments upon subtracting the flux decrease of the secondary filtration from that of the primary one to remove the impact of pore blocking. The findings established that CFI better anticipates the level of fouling rate in RO tests than MFI. It was as well demonstrated that it may be employed as a helpful instrument for detecting and determining the fouling mechanisms. By comparing MFI and CFI, it was illustrated that pore blocking was improved as much as cake generation when pH diminished; whereas divalent cations (Ca^{2+}) augmented only cake generation on the membrane surface.

At a full-scale UF/RO seawater desalination plant, Jin et al. (2017) assessed the pertinence of multiple MFIs (i.e., MFI$_{0.45}$, MFI-UF$_{100\ kDa}$ and MFI-UF$_{10\ kDa}$). During one year, MFIs were evaluated with RO feed water and the findings were consistent with increases in differential pressure (DP) of RO method. They performed regression analysis with view to discover if the fouling index may expect RO efficiency impairment. The spotted MFIs distinctly illustrated linkage with DP variation; whereas SDI and water quality parameters (such as turbidity and UV$_{254}$) presented to a certain degree weak correspondence. Specifically, MFI-UF$_{100\ kDa}$ was the most responsive, showing that colloidal fouling is the principal source of fouling in SWRO desalination. The regression findings were asserted again during particular incidents where feed water quality was seriously varied because of red tide or ship movement at intake. Such incidents presented unclassical feed water features; however, identical tendency was noticed that MFI-UF$_{100\ kDa}$ was most responsive.

3. Context of the proposal of a new fouling index

During the previous Section, we abstracted some main important works performed heretofore with a view to enhance the anticipation of fouling.

Nevertheless, it is obvious that the applicability of these procedures in real conditions is controversial because of: (1) Dissimilarity in filtration way, (2) Cutoff of membranes not typical of the molecular weight distribution of the different constituents found in water.

Consequently, it is obligatory to attempt to emerge additional techniques of quantification that would better anticipate the capacity of membrane fouling in RO and Nano Filtration (NF). These techniques would be employed on the other hand to experiment the performance of various pretreatment processes by evaluating the fouling capacity of the pretreated water.

The MFI-UF has been the target of plenty of disapproval with regard to its performance in anticipating fouling in membrane filtration factories. It is in this way that numerous research investigations have been performed in view to treat defect in the MFI, suggesting correction factors (deposition factor, compressibility factor, etc.) to enhance its extrapolation and applicability in actual situations. In the identical fashion, fouling indicators obtained from the primary MFI were also applied (such as the MFI-UF at constant flow, the CFS-MFI$_{UF}$, the NF-MFI, etc.) so as to enhance the anticipative potential of this index.

In the written works, there is a large span of empirical estimations of MFI-UF depending on working situations, the membranes employed and the purity of the filtered water. The explanation of these findings and their influence on fouling in RO or NF is complicated since fouling mechanisms rely on the filtering process employed. It is for this cause that guiding values of the MFI-UF may be suggested for ascertaining an admissible degree in matter of fouling prior to the RO/NF functioning.

Our method is to suggest a fouling indicator following from the nondimensionalization of the Schippers and Verdouw equation (model of cake type fouling). The justification for this procedure is associated to the known advantages of dimensionless equations.

The characterization of the system by an equation reassembling numerous dimensional parameters and variables is decreased upon nondimensionalization to an equation carrying a sole dimensionless number. The benefit of a dimensionless equation is that it represents all identical physical systems, letting changing from one scale to another (scale up-down). A dimensionless number may usually give important rough calculations of value; the magnitude of these evaluations is linked to the selection of the scale. In the main, a dimensionless number possesses a physical explanation that takes part in the physical comprehension of the process. Therefore, in our situation, the usage of a dimensionless fouling index (DFI) is explained by the problems encountered in the interpretation and utilization of the values of the dimensional MFI-UF.

The objective of this work is to decrease the number of parameters required to explain the estimates of the primary MFI-UF acquiring evaluations for a sole index founded on some parameters separately. Two parameters are especially searched: the membrane type and the concentration of raw water.

In the end, examination of the findings acquired by tests under various situations of membrane type and concentration of water and their conformity with the targets will allow us to reach deductions concerning the establishment of this manner. Unluckily, the absence of requisite equipment and the deficiency of all required data to perform the needed transformations employing experimental findings discovered in the writing works render this task unreachable at this level. The authors are completely conscious of this restriction. However, the results of the present investigation are shown to the scientific community with the deliberate aim of sharing the dimensionless index which is reached in the aspiration of discerning it confirmed by accessible or subsequent empirical research.

4. Derivation of the new dimensionless fouling index (DFI)

The cake filtration model is largely employed. It explains the way of filtration when a cake is gradually and regularly generated on the surface of the membrane. The fundamental hypothesis of this model is that the amount of matter put down on the surface of the filter is proportional to the filtered volume of water. The model conducts to a simple equation obtained from the linearization of Ruth's equation and is especially crucial for the expansion of fouling indicators. Founded on this procedure, Schippers and Verdouw (1980) suggested a fouling indicator known as "Modified Fouling Index" (MFI) that takes into account the mechanisms which operate fouling.

Founded on the underlying equations of filtration and considering the linearization of Ruth's equation, the cake filtration model conducts to a dimensional equation implying the variables (t, V) and the parameters (μ, r, C, ΔP, S, R_m):

$$\frac{t}{V} = \frac{\mu r C}{2\Delta P S^2} V + \frac{\mu R_m}{S \Delta P} \tag{1}$$

with

$$\frac{\mu r C}{2\Delta P S^2} = MFI \tag{2}$$

where V is the filtrate volume and t is the filtration time. ΔP is the applied transmembrane pressure, μ the water viscosity, R_m the membrane resistance, S the membrane surface area, r the cake specific resistance and C is the concentration of the bulk.

The MFI relationship is in the form of a group of dimensional parameters, the dimension of the MFI is (time/length^6) or in (s/L^2) if liter is used as the volume unit.

In order to nondimensionalize Eq. 1, reference parameters (scales) require to be involved. These

parameters should be typical estimates of the variables (V, t) in Ruth's equation. To perform this, the best selection of reference scales is explained by the following estimates:

Volume scale (V_f)

$$V_f = \frac{R_m S}{rC} \tag{3}$$

This volume was suggested by Ruth. It is employed to take into account the participation of the membrane resistance by equating it to a fictional cake layer with equivalent resistance. This fictitious layer is assumed to have generated before the real beginning of the filtration operation and correlates with the flow of a fictitious volume (V_f) of filtrate.

$$t_{ref} = \frac{1}{R_m J_0} \tag{4}$$

with

$$J_0 = \frac{\Delta P}{\mu R_m} \tag{5}$$

J_0 being the clean water flux (CWF) of the new membrane at the pressure of the test. Therefore, the reference time t_{ref} relies exclusively on the primary features of the membrane.

The selection of these parameters is not random; indeed, their values rely on the primary features of the membrane and the water to be filtered. Considering all these facts, the following dimensionless variables are presented:

$$V^* = \frac{V}{V_f} \Rightarrow V = V^* V_f \tag{6}$$

$$t^* = \frac{t}{t_{ref}} \Rightarrow t = t^* t_{ref} \tag{7}$$

from Eq. 1, we may obtain:

$$\frac{\mu rC}{2\Delta PS^2}V^2 + \frac{\mu R_m}{S\Delta P}V = t \Rightarrow V^2 + \frac{2R_m S}{rC}V = \frac{2\Delta PS^2}{\mu rC}t \tag{8}$$

considering Eq. 8 and Eq. 3, we may find:

$$V^2 + 2V_f V = \frac{2\Delta PS^2}{\mu rC}t \Rightarrow V + 2V_f = \frac{2\Delta PS^2}{\mu rC}\left(\frac{t}{V}\right). \tag{9}$$

Based on Eq. (9) and taking into account Eq. (6), the nondimensionalization procedure is then performed as follows:

$$V^* V_f + 2V_f = \frac{2\Delta PS^2}{\mu rC}\left(\frac{t}{V^* V_f}\right) \tag{10}$$

Dividing Eq. 10 by V_f, Eq. 11 is found:

$$V^* + 2 = \frac{2\Delta PS^2}{\mu rC}\left(\frac{t}{V^* V_f^2}\right) \Rightarrow V^* + 2 = \frac{2\Delta PS^2}{\mu rC V_f^2}\left(\frac{t}{V^*}\right). \tag{11}$$

Then, by introducing t_{ref} from Eq. 7 in Eq. 11, Eq. 12 is obtained:

$$V^* + 2 = \frac{2\Delta PS^2}{\mu rC V_f^2}\left(\frac{t^* t_{ref}}{V^*}\right) \Rightarrow V^* + 2 = \frac{2\Delta PS^2 t_{ref}}{\mu rC V_f^2}\left(\frac{t^*}{V^*}\right) \tag{12}$$

Replacing V_f and t_{ref} by their respective expressions from Eq. 3 and Eq. 4 in Eq. 12, Eq. 13 is obtained:

$$V^* + 2 = \frac{2rC\Delta P}{\mu R_m^3 J_0}\left(\frac{t^*}{V^*}\right) \tag{13}$$

Combining Eq. (13) and Eq. (5), Eq. (14) is obtained:

$$V^* + 2 = \left(\frac{2rC}{R_m^2}\right)\left(\frac{t^*}{V^*}\right) \tag{14}$$

This dimensionless equation can then be transformed into a form which is identical to Ruth's linearization as follows:

$$\frac{t^*}{V^*} = \frac{R_m^2}{2rC}V^* + \frac{R_m^2}{rC} \tag{15}$$

where $\frac{R_m^2}{2rC}$ is a dimensionless number. This number may be called "dimensionless fouling index" (DFI) by analogy to Eq. 1. In other words:

$$DFI = \frac{R_m^2}{2rC} \tag{16}$$

Consequently, taking in consideration Eq. 16, Eq. 14 may be formulated in the following relationship:

$$\frac{t^*}{V^*} = DFI(V^* + 2) \tag{17}$$

Comparing the two formulas (dimensional, i.e. Eq. (1), and dimensionless, i.e. Eq. 15), it seems that the dimensionless formula is transformed into a simple linear relationship that is a function of an only one parameter which is the DFI as described in Eq. 16. It seems that DFI relies only on R_m, r and C. However, one should keep in mind that it is no longer t/V which is written as a function of V but their dimensionless counterparts. As a result, the impact of the remaining parameters (i.e., μ, ΔP, S) is tacitly taken into consideration.

The DFI is a group of dimensional parameters that can be explained as the ratio of the membrane resistance to that of the cake due to the concentration of raw water. Consequently, the larger it is, the larger is the potential of the membrane to fouling when treating the given water.

5. Conclusion

Fouling constitutes one of the main issues challenge the usage of membranes in RO processes. In this research, a brief review of studies performed on fouling indicators is presented. Fouling indicators established yet are found on experiments of frontal filtration with MF or UF membranes and are presented in the form of dimensional coefficients depending on several operating parameters. Moreover, their values take cover very wide ranges, which does not let attributing obvious indications concerning guideline values signaling the onset of fouling or strong fouling potential.

The aim of the present work was to give a dimensionless grouping of a limited number of parameters that could eventually serve as a fouling index under a large range of operating conditions. This is performed by an appropriate nondimensionalization of the equation of Ruth's equation and the following conclusions are made:

- The dimensionless number which is obtained is termed dimensionless fouling index (DFI) and can be explained as the ratio of the membrane resistance to that of the cake due to the concentration of raw water.
- It is hoped that the proposed number will allow formulating appropriate fouling potential indicators falling in relatively moderate ranges under different operating conditions. This would confer a more "universal" character to the guideline values inferred and would certainly be of great convenience for industrial applications.
- It is perfectly clear that the validation of the present approach requires performing experiments under different conditions of membrane type and a range of particulate and/or dissolved material. Unfortunately, the lack of adequate equipment and the lack of all needed data to make the necessary transformations using experimental results found in the literature make this step unattainable at this stage.
- The authors are perfectly aware of this limitation. Nevertheless, the findings of the present study are presented to the scientific community with the deliberate intent of sharing the dimensionless index which is obtained in the hope of seeing it validated by available or future experimental work.

Abbreviations

C	Concentration of the bulk (mg/L)
CFI	Cake fouling index
DFI	Dimensionless fouling index (Eq. 16)
J_0	Clean water flux (CWF)
MFI	Modified fouling index (Eq. 2, (s/L^2))
MFI-UF	Modified fouling index-ultrafiltration
NF	Nanofiltration
PAN	Polyacrylonitrile
PS	Polysulfone
r	Cake specific resistance
R_m	Membrane resistance
RO	Reverse osmosis
S	Membrane surface area (m^2)
SDI	Silt density index
t	Filtration time (s)
t^*	Dimensionless time (Eq. (7))
TMP	Trans-membrane pressure (Pa)
t_{ref}	Reference time (s)
UF	Ultrafiltration
V	filtrate volume (m^3)
V_f	Fictitious volume of filtrate (Eq. 3), m^3
V^*	Dimensionless volume (Eq. 6)

Symbols

ΔP	Applied transmembrane pressure (Pa)
μ	Water viscosity (Pa.s)

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