



# Quantitative determination of the electrical properties of RO membranes during fouling and cleaning processes using electrical impedance spectroscopy



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## HIGHLIGHTS

- Electrical impedance spectroscopy (EIS) used for in-situ fouling detection in RO.
- Impedance spectra of membrane process consist of four Maxwell–Wagner elements.
- EIS parameters: capacitance and conductance were quantified and studied.
- EIS data is used to understand fouling and cleaning behavior of silica/alginate.

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## ABSTRACT

Electrical impedance spectroscopy (EIS) has been used as a non-invasive means of characterising the electrical properties of a reverse osmosis (RO) membrane during fouling and cleaning processes. The crossflow RO unit used in this study was equipped with suitable electrodes and used in conjunction with a high resolution impedance spectroscopy that allowed changes in the electrical properties of the membrane and foulant layer to be monitored. The internal structure of the membrane could be represented by a series combination of parallel conductance and capacitance elements. The deduced support layer thickness matched optical measurements and the effective polyamide skin layer thickness matched recently reported data using high resolution TEM. During the filtration process using silica and alginate as model foulants, the conductance of all the electrical elements initially decreased with time and then increased as fouling progressed. The increase was associated with a local increase in salinity due to the development of cake enhanced concentration polarization. The capacitance of the skin layer decreased as the fouling process progressed and this could be evidence of the built up of foulant layers on membrane surface. By flushing the fouled membrane with NaCl solution for about 17 h, the membrane returned back to its original state, with its capacitance and conductance values similar to the unfouled membrane.

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## 1. Introduction

Over the past decades, it has been shown that impedance spectroscopy can provide a non-invasive means of characterising the electrical properties of many systems from one of the earliest estimations of the thickness of the plasma membrane of a living cell [1] to a label-free detection tool for many different types of biosensors [2–3] and molecular films [4] and lipid bilayers [5–6]. Impedance spectroscopy provides valuable information to understanding the structural-functional

relationship of these systems. Electrical impedance spectroscopy (EIS) has also been applied to the engineering field, such as evaluation of corrosion rates by polymer coated surfaces [7–8] as well as characterisation of synthetic filtration membranes and fouling phenomena [9–18]. Membrane fouling is a costly problem in water and desalination industries and non-invasive observation of fouling by methods, such as EIS, can provide valuable insights and the basis for online monitoring.

In the field of membrane filtration, the focus of EIS has been on its novel application to various membrane fouling related problems [19–24]. Currently in water treatment plants, increases in transmembrane pressure, TMP (for Microfiltration MF/Ultrafiltration UF) and/or differential pressure across membrane elements (for RO) and the drop in normalised flux, is used as an indication for membrane fouling. However,

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by the time such changes occur, the membranes might have been severely fouled or clogged. On-line monitoring of membranes to provide early indicators of incipient fouling during separation processes would have the benefits of more efficient cleaning cycles and chemical dosing, prolonged membrane lifetime, reduced plant downtime, less power consumption and verification of the effectiveness of cleaning.

EIS has been demonstrated in several studies [19,21,23] to be a sensitive monitoring tool to detect early fouling in reverse osmosis (RO) systems. In our recent work [23], EIS was used to detect silica and BSA fouling on a lab-scale RO system. A typical RO crossflow cell was equipped with electrodes to allow four-terminal in-situ EIS measurements during the fouling process. EIS responded almost immediately to any changes that occurred on the membrane surface. In particular, during the early stage of silica fouling, the electrical impedance spectra changed significantly and well ahead of changes in the TMP. However, the study did not quantitatively describe the structural changes of the membrane and its surface during the membrane fouling process.

In this paper, we expand on our prior work to quantitatively describe the changes in the electrical properties of the membrane associated with the fouling process. A more detailed analysis in identifying the Maxwell Wagner elements of the distinct layers was performed in this study. For example, measurement of the porosity of individual membrane layers was performed in a more accurate manner using the evaporometry method. As a result, a more representative thickness of each layer could be obtained. The RO polysulfone and polyamide (PA) layers were physically removed from the non-woven fabric in order to identify corresponding characteristic frequencies. New insights were obtained into the effective PA layer thickness. For the fouling study, the model foulant in the feed solution was a combination of silica and alginic acid. The latter is a surrogate for the extracellular polymeric substances (EPS) produced by microorganisms which feature strongly in sea water desalination. Although EIS has been applied to detect RO fouling and scaling problems in several studies [19–21,25], none of them has used EIS to detect the removal of foulant layers from the membrane surface. In this paper, we also examine monitoring of cleaning operations using EIS.

## 2. Theory

Electrical impedance,  $Z$  is determined by measuring the amplitude of voltage  $v_0$  and phase difference  $\theta$  that develops across a system when a small alternating current (AC) with amplitude of  $i_0$  of known frequency ( $\omega$ ) is applied to it. The magnitude of the impedance is given by:

$$|Z| = \frac{v_0}{i_0}. \quad (1)$$

The impedance of a homogenous material can be expressed in terms of a conductance element,  $G$ , and capacitance  $C$ , which describe the ability of the material to conduct and to store electric charge respectively. They can be calculated from the magnitude of the impedance and the phase difference as follows.

$$G = \frac{\cos\theta}{|Z|} \text{ and } C = \frac{\sin\theta}{\omega|Z|} \quad (2)$$

where  $\omega$  is the angular frequency. For a simple homogeneous sample of material, the impedance is frequency dependent but the conductance,  $G$  and the capacitance,  $C$  are frequency independent.

In practice, however, the capacitance and conductance of a material sample are themselves frequency dependent. This can arise from the presence of sub-structural layers or from electrochemical diffusion effects. The latter will be considered below.

For a non-homogenous system such as a filtration membrane which is composed of distinct layers with different properties, the overall parallel combination of the conductance and capacitance themselves become dependent on the frequency of measurement. Thus for a system comprised of  $N$  substructural layers, each with a frequency independent conductance and capacitance, the total impedance is given by:

$$Z_N(\omega) = \sum_{n=1}^N \frac{1}{G_n + j\omega C_n} \quad (3)$$

where  $n$  is used to identify the frequency independent conductance and capacitance of each of the  $n^{\text{th}}$  layers. When the membrane has such substructural elements the overall conductance and capacitance also become frequency dependent.

$$G(\omega) = \frac{\cos\theta(\omega)}{|Z(\omega)|} \text{ and } C(\omega) = \frac{\sin\theta(\omega)}{|Z(\omega)|} \quad (4)$$

where  $|Z(\omega)|$  is given by Eq. (3).

The form of the dispersion of the impedance magnitude with frequency is often very insensitive to the number of substructural elements. However, the dispersions of the overall conductance,  $G(\omega)$ , and capacitance,  $C(\omega)$ , much more clearly reveal the presence of substructural layers in the system. The substructural resolution that can be obtained using EIS thus depends crucially on accurate measurement of the phase at each frequency in order to determine the conductance and capacitance at each frequency [11].

Both conductance and capacitance of an element are related to the layer's material and geometrical properties,

$$G_n = \frac{\sigma_n}{\delta_n} \quad (5)$$

$$C_n = \frac{\epsilon_0 \epsilon_n}{\delta_n} \quad (6)$$

where  $\sigma_n$  is the conductivity,  $\epsilon_n$  is the dielectric constant of the  $n^{\text{th}}$  layer,  $\delta_n$  is the thickness of the layer and  $\epsilon_0$  is the permittivity of free space ( $= 8.85418 \times 10^{-12}$  F/m).

As referred to earlier, frequency dependent capacitance and conductance can also arise from electrochemical diffusion effects. There are several types of such effects. At very low frequencies ( $< 10$  Hz), diffusion polarization can occur near the membrane-solution interface that, for instance, arises from differences, relative to the external solution, in the transport numbers for cations and anions in the membrane. The resulting (AC) concentration profiles can manifest as a capacitance and this makes a large contribution to the total membrane system capacitance. Such a "phenomenological" capacitance arises because the conductance in the diffusion polarization region is dependent on the ion concentrations which themselves build up and collapse during each half-cycle of the AC signal applied and lag behind (in time) the AC current flowing. This leads to a conductance element that has an AC component out of phase with the applied voltage at low frequencies. The total conductance of this region therefore is a (mathematically) complex quantity with real and imaginary components. The imaginary component of this conductance appears as a frequency dependent capacitance. This can be seen as follows. The capacitance can be written in terms of the imaginary part of the admittance as:

$$C = \frac{I_m\{Y(\omega)\}}{j\omega} \quad (7)$$

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