Abstract

Separation membranes typically have a thin skin layer supported on a more porous and thicker base layer. Next to the surface a diffusion polarization layer will develop during separation processes. Each of these layers has associated electrical conductance and charge storage properties (capacitance).

When an alternating current (AC) is passed through such a system, the presence of such substructural and diffusion polarization layers will manifest a dispersion with frequency of the overall capacitance and conductance of the system.

If the overall capacitance (C) and conductance (G) of the system is measured with sufficient precision over a wide range of frequencies of the AC current, it is possible to deconvolve the overall impedance dispersion to yield the separate electrical parameters (capacitance and conductance) for each of the individual layers present.

The capacitance of a layer can arise in two ways:

- For a layer of a dielectric material, capacitance is related to its charge storage properties which are determined by its electrical polarizability (dielectric constant) and its thickness.
- In diffusion polarization layers, transport effects give rise to phenomenological capacitances that arise from phase shifts induced by diffusion limited processes. This type of phenomenological capacitance manifests at very low frequencies.

Introduction

Fouling of membranes, resulting in increased power consumption and a reduction in membrane lifespan has been recognized as a major operational and economic issue for water treatment plants [1]. Membrane fouling has traditionally been measured by the decline in the permeate flux at constant operating conditions.

Early detection is important, because the onset of fouling occurs at a critical flux [2, 3, 4, 5] which is difficult to predict a priori. With high flux reverse osmosis systems, and particularly with low salinity feeds at high recovery rates, the measurement of fouling by flux decline poses problems in practice since the flux declines along the module [1].

Developments in the manufacture of membranes has reached the stage where improvements in the performance of membrane separation systems is now largely dependent on improving the operational strategies, rather than improvements in the membranes incorporated in the separation modules. The monitoring of fouling is an important parameter in developing such optimal strategies.

Electrical Impedance Spectroscopy of Membranes

Electrical Impedance Spectroscopy has proven to be a valuable tool in analyzing the structures of micro and ultrafiltration membranes [6]. The technique was also applied to the study of the fouling of ion exchange membranes [7]. Other studies [8, 9] of membranes, using Electrical Impedance Spectroscopy, showed that surface
Characterizing Fouling in Separation Membranes

During operation, particularly at high flux rates, a diffusion polarization layer also forms at the membrane surface. This ‘layer’ itself can be represented by a parallel combination of a conductance and capacitance. Further, when fouling occurs, this diffusion polarization layer is modified and leads to distinct changes in the impedance spectrum.

When membrane fouling occurs, the dielectric parameters are changed and these changes can be readily determined from in situ impedance spectroscopy measurements, during actual membrane operation. Some examples of the type of data that can be so obtained are described below for nanofiltration and reverse osmosis filtration membranes.

Reverse Osmosis Membrane Fouling

The fouling of a reverse osmosis membrane was also demonstrated [10] using the INPHAZE HiRes-EIS system over the frequency range of 0.1 Hz to 100 kHz [11]. Both the conductance and impedance showed dramatic changes when the reverse osmosis membrane was fouled by a small amount of precipitated divalent salts.

The impedance spectra for the fouled and unfouled membrane revealed the presence of three distinct elements: the skin layer; the membrane sub-layer; and a diffusion polarization/fouling layer.

Examples of the impedance data for the unfouled and fouled membrane are shown in Figures 3a and 3b. The
full lines plotted on these graphs were obtained by data reduction in terms of the three basic impedance elements. In the data reduction process the interfacial diffusion polarization layer is modelled using a series of frequency-independent impedance components (conductances and capacitances). The data reduction showed that the basic membrane elements [skin layer and sub layer] did not change on fouling. However, fouling produced a very distinct change in the parameters describing the interfacial polarization layer where fouling led to a conductance scaling factor of 0.56.

**Comment:** In a reverse osmosis membrane there are no distinct pores. The basic membrane elements (skin layer and sub layer) are therefore not likely to undergo major changes on fouling. However, the formation of a foulant (cake) layer on the surface would be expected to lead to significant changes in the membrane-solution interface where diffusion polarization occurs.

Membrane fouling can therefore be simply described in terms of the conductance scaling factor for the interfacial polarization layer. Reduction of data in terms of characteristic of the basic dielectric parameters yields direct insight into the state of fouling of the membrane.

**Nanofiltration Membrane Fouling**

Nanofiltration membranes, unlike reverse osmosis membranes, have well defined pores. The membrane has a skin layer containing nano-sized pores, a sub layer, and in operation an interfacial polarization layer. Fouling can therefore be expected to produce changes in the membrane parameters per se as well as changes in the interfacial polarization layer.

Examples of the impedance data [10] for the unfouled and fouled membrane are shown in Figures 5a and 5b. The full lines plotted on these graphs were obtained by data reduction in terms of the three basic impedance elements. In the data reduction process the interfacial diffusion polarization layer is modelled using a series of frequency-independent impedance components (conductance and capacitance). The data reduction showed that the basic membrane elements (skin layer and sub layer) changed on fouling. Fouling produced very significant changes in the parameters describing the skin layer (and to a lesser extent the membrane sublayer) as well as a large change in the interfacial polarization layer where fouling led to a conductance scaling factor of 0.4 (see Figures 6a and 6b).

**Comment:** In a nanofiltration membrane there are distinct pores. The basic membrane elements (skin and sublayer) are likely to undergo major changes on fouling in addition to the changes in the interfacial polarization layer that will also occur.

Membrane fouling can therefore be described in terms of the conductance scaling factor for the interfacial polarization layer as well as changes in the impedance elements for the skin layer. There are also additional indicators of fouling, such as changes in the capacitance of the skin or the conductance of the sublayer, which could be used to confirm the observations on fouling based on the changes in the interfacial polarization layer and skin.

**Summary**

High resolution impedance spectroscopy can readily determine the substructural elements of separation membranes and monitor changes that occur in these elements during membrane fouling processes. Using online data reduction methods a simple parameter such as the interfacial polarization conductance scaling factor or a skin conductance parameter can be deduced, that provides an immediate measure of membrane fouling.

**Acknowledgements**

The authors would like to thank Dr. J. Kavanagh and Mr. S. Hassein for discussions, intellectual and technical input to the study of membrane fouling using electrical impedance spectroscopy.

**References**


Figure 5a: Conductance and capacitance of a nanofiltration membrane system before and after fouling with CaCO₃ [10]. Components and values for modelling the skin, sub and diffusion polarization layers and the solution. Fouling can be principally attributed to decreases in the conductances of the skin, sublayer and diffusion polarization layers (F) and characterised by a shift in the diffusion polarization capacitance (G & H) to lower frequencies.
Figure 5b  Impedance magnitude and phase of a nanofiltration membrane system before and after fouling with CaCO$_3$. Note in particular the large increase in the impedance and the distinct change in the phase as a function of frequency that occurred on fouling.


