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# Mechanistic modelling of the dielectric impedance of layered membrane architectures

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## Abstract

Electrochemical impedance spectroscopy (EIS) is frequently used to investigate properties of layered membrane systems in aqueous electrolyte solutions. However, attributing features of the underlying plots to chemical or structural characteristics of the system is difficult, as it usually requires the assumption of structural elements of an analogous equivalent electrical circuit. Furthermore, some parameters are not easily accessible in experimental studies, making the analysis of EIS subject to interpretation rather than rigorous understanding. We address these difficulties by conducting direct numerical simulations of layered structures composed of electrolyte solutions, ion exchange membranes and membrane modification layers, using our  $E_nPE_n$  framework. By changing the parameters of the model, it is possible to isolate features of impedance spectra as they relate to chemical and structural properties of different layers and interfaces. For the first time, we show the general influence of the thickness and charge density of surface modification layers on the complex impedance of such layered membrane architectures in multi-ionic mixtures. The results can be used to characterize and understand impedance spectra gained by experiments. This is important in increasingly complex systems with multiple layers and complex multi-ionic electrolyte solutions. Understanding the influence of design parameters on the performance of a membrane is important for a wide range of applications, such as ion selective electrodes, desalination and deionization. The dynamic  $E_nPE_n$  framework closes the gap between experimental EIS and the uncertainty introduced by assuming equivalent circuits.

**Keywords:** impedance spectroscopy, ion exchange membranes, dynamic  $E_nPE_n$ , monovalent ion selectivity

## 1. Introduction

Thin film assemblies coated on top of surfaces are frequently employed in a variety of applications in order to optimize specific ion fluxes. The performance of layered materials is significantly influenced by the properties of the outermost layer, which can have thicknesses in the nanometer range. Surface modifications have been used to add protective layers to electrodes [1], control ion retention in membranes [2], or introduce catalytic properties in different substrates [3, 4, 5]. A notable technique receiving much attention recently is layer-by-layer assembly, which allows creating structures of alternating positively and negatively charged polyelectrolytes [6, 7].

The surface modification of ion exchange membranes and membrane electrode assemblies is one of the most promising applications of multilayer architectures. Properties, such as the monovalent ion selectivity of ion exchange membranes, can be enhanced by adding charged multilayers on their surface [8, 9]. Layered membranes are employed in different areas of electrochemistry. Monovalent ion selective membranes are used in electro dialysis for the fractionation of ions and in ion-selective

electrodes for analytical purposes [8, 10, 11, 12]. Surface modified membranes have also been applied to improve the current efficiency of fuel cells and batteries by reducing the fuel cross-over or the cross-mixing of electrode solutions [9, 13, 14]. The underlying mechanisms also play a role in nanofiltration and reverse osmosis [15].

Achieving a satisfying trade-off between selectivity and conductivity is the main goal of modifying the surface of ion-exchange membranes [16]. To this end, understanding the correlation of the characteristics of layered semipermeable membranes and the involved ion transport processes is fundamental. Ion transport in electrolyte and polyelectrolyte solutions can be described by the coupled Nernst-Planck and Poisson equations. Solving this non-linear set of equations is challenging due to wide range of time scales inherent to diffusion and charge relaxation. Often, the problem is simplified by several assumptions [17]. Of special difficulty is resolving the membrane/electrolyte and membrane/modification layer interfaces, since large gradients of concentration and electric potential occur in these regions [18]. The complexity of the problem is significantly increased if

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