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#### Review

# Study of proton exchange membrane fuel cells using electrochemical impedance spectroscopy technique – A review

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#### HIGHLIGHTS

#### G R A P H I C A L A B S T R A C T

- Electrochemical impedance spectroscopy in diagnosing PEM fuel cells is reviewed.
- The EIS technique and various equivalent circuit elements are discussed.
- Modifications to impedance measurements and interpretations are presented.
- Critical points misinterpreted in the EIS measurement models are discussed.

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#### 1. Introduction

High dependency of conventional power generators to fossil fuels which has resulted in global warming and environmental pollution in the recent decades have motivated the research and development toward new, clean, inexpensive and robust power generation devices. Among them, fuel cells have been accepted as



#### ABSTRACT

Electrochemical impedance spectroscopy (EIS) technique is well established in recent years to study and diagnose proton exchange membrane fuel cells (PEMFC). In this paper, the recent published research results in this field are reviewed. The EIS technique and equivalent circuit concept are introduced and recent applications of EIS in PEM fuel cell modeling and diagnosis are thoroughly reviewed. The paper also includes outstanding improvements conducted in EIS measurement and hardware. Finally the critical points that can be misinterpreted in the EIS measurement models in the field of PEM fuel cells are discussed.

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a prospective alternative for nowadays sources of energy. The fuel cell has considerably high power density with no emission or any moving parts. This increases the potential of being a reliable and robust system for long-term usage. Also, compared to batteries, the power (i.e., proportional to the fuel cell size) and capacitance (i.e., proportional to the reservoir size) characteristics of fuel cells are independent, and hence they can be employed in a wide range of applications [1]. Despite the general success made toward the advancement of fuel cells, their performance and applicability are limited due to several factors. In essence, the final cost of a fuel cell is the major barrier for commercialization.



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Also, the best fuel for the low temperature fuel cells is hydrogen that has low volumetric energy density and is hard to store. Hydrogen has to be completely pure (especially in proton exchange membrane fuel cells) and alternative fuels usually need reforming [1]. Moreover, robustness and durability under start—stop cycles are not reliable in the current form of the fuel cells as has been indicated in the strategic planning of US department of energy (DOE) which is directed toward the development of a portable power fuel cell system (<250 W) with an energy density of 900 Wh L<sup>-1</sup> by 2015, a transportation fuel cell system with 5000-h durability by 2017, and a micro-CHP (Combined Heat and Power) fuel cell system (5 kW) with 60,000-h durability and a medium-scale CHP fuel cell system (100 kW–3 MW) with 80,000-h durability by 2020 [2].

Among various kinds of fuel cells, a proton exchange membrane fuel cell (PEMFC) which performs at low operating temperature and relatively high power density has captured the most attention. In PEMFC, the hydrogen gas is fed into the anode; while usually the air (or pure oxygen) is fed to the cathode. The hydrogen is split into the electron and positive ions which move through a protonconducting membrane to the anode. The transported protons are combined with the oxygen in the cathode, producing water as byproduct. The balance in the water transport through and out of the cell is very crucial to avoid flooding and dehydration of the membrane.

There are different electrochemical methods developed to study and diagnose the PEM fuel cells. These methods have been thoroughly reviewed by Wu et al. [3]. Among different techniques used the polarization curve, current interruption, cyclic voltammetry and electrochemical impedance spectroscopy are the most wellknown electrochemical techniques in fuel cell studies. The polarization curve is the most common method of presenting and discussing the PEM fuel cells performance [1,3,4]. In this technique, the cell potential is plotted against the cell current (or current density) in various operating conditions. Usually, the plot is divided into three (or more) parts. It is believed that each part has a dominant performance loss. In the current interruption technique, the ohmic load is almost immediately released from the fuel cell and the voltage change is plotted vs. time. This method is often employed to investigate the ohmic losses in fuel cells [1,3]. In the cyclic voltammetry technique, the voltage increases linearly and sweeps back while the current is recording. The resultant voltagecurrent plot has peaks on potentials related to electrochemical reactions of the system [1,3]. Electrochemical impedance spectroscopy (EIS) is a well-established diagnostic and modeling method that has widely been used in electrochemistry [5-7] due to its flexibility (which enables covering a wide range of aspects of fuel cells) and accuracy.

In this paper, the studies conducted on PEM fuel cells using electrochemical impedance spectroscopy are reviewed. One of the most comprehensive reviews conducted in this field is the work published by Yuan et al. [4] in 2007. They discussed about various measurement and feeding modes in EIS as well as interpretation and EIS applications in optimization and contamination effects. This paper thoroughly reviews the recent efforts published in this field as well as aspects like Kramer-Kronig relations and process and measurement models, which were not covered in Ref. [4]. After introducing the EIS method and different aspects of the EIS technique (including the equivalent circuit and various defined elements based on electrochemistry reactions), the recent EIS applications are reviewed. The paper also highlights the novel studies performed and ideas developed based on the EIS method, and finally discusses common uncertainties and possible errors in the interpretation of the EIS measurements.

#### 2. Electrochemical impedance spectroscopy

As mentioned before, the EIS technique performs based on imposing a harmonic perturbation to the system and measuring the impedance of the system in a wide range of frequencies. To measure the impedance in different frequencies, a frequency response analyzer (FRA) and a load bank are required. A sinusoidal wave is produced by FRA and applied to fuel cell via the load bank. Then, the system response is captured by FRA and an impedance spectrum is calculated [4]. The measurements can be conducted in the galvanostatic or potentiostatic modes, perturbing the current or voltage by the harmonic wave, respectively. It has been reported that there is no considerable differences between the galvanostatic and potentiostatic modes results [4]. However, the slope of the polarization curve is not significant in most of the performance domain, and hence a small perturbation in the voltage can cause a considerable change in the current, overloading the cell and the potentiostat circuits. Hence, the galvanostatic mode is usually preferred during impedance measurements [4]. The impedance measurement can also be conducted in both in-situ and ex-situ manners [4]. In the in-situ configuration, which has been widely used, the AC impedance of the fuel cell is measured as a whole; whereas, in the ex-situ measurement different components of the fuel cell are studied separately.

#### 2.1. AC amplitude selection

One of the most important parameters in the AC impedance measurement is the AC amplitude. It is clear that the larger the AC amplitudes the easier the separation of the measured response from noises. However, the amplitude is limited due to the linearity assumption: in essence, the impedance is estimated by measuring the changes in the potential over the changes in the current. This calculation can be accepted if the potential changes linearly with respect to the current. Since this is not true for PEM fuel cells and other electrochemistry systems, the signal amplitude has to be small enough to satisfy the linearity assumption. Different studies [8–11] have indicated the AC signal amplitude equal to 5% of the DC performance current. For instance, the study conducted by Dale et al. [11] on the impedance spectra of a 2 kW stack for the selected AC amplitudes of 1%, 5%, 7.5%, 10% and 15% of the DC load shows that i) the spectrum is very scattered for low load currents, especially at a 1% AC signal amplitude for which it becomes difficult for FRA to distinguish between the response and noise; and ii) there is no considerable difference between the impedance plots in the amplitude range of 5%-10%.

#### 2.2. Impedance plots

The Nyquist plot is the most usual way of presenting the impedance spectrum. In this graph, the imaginary part of the impedance (mostly presented with the negative sign) is plotted against the real part. The plot normally contains of two or more semicircles in different frequency ranges. Although the Nyquist plot is the most common representation in the EIS measurement technique, Bode magnitude (magnitude vs. frequency) and phase (phase vs. frequency) plots have also been used in the past [11–13] to extract information that cannot be readily obtained from the Nyquist plot. For instance, the local maximum values in the Bode phase plot show the characteristic frequencies which are proportional to the inverse of the time constants of the processes [13].

#### 2.3. Wires impedance issue

The connecting wires can create significant magnetic fields affecting the impedance spectrum measurements [11]. This effect

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