



# A parametric model for solid oxide fuel cells based on measurements made on cell materials and components



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## HIGHLIGHTS

- Developed a parametric model for solid oxide fuel cells based on out of cell measurements.
- Calculated performance curves are in good agreement with experimental cell test data.
- Ohmic contribution can be substantial even in thin electrolyte film solid oxide fuel cells.

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## ABSTRACT

A parametric equation describing polarization in solid oxide fuel cells (SOFC) in terms of experimentally measurable parameters is presented. The equation explicitly describes activation and concentration polarizations at the two electrodes and the ohmic loss as functions of current density. Using known values of parameters measured on cell materials and components, various polarizations are estimated as functions of current density and the possible performance characteristics are assessed. The calculated performance curves using measurements made on cell materials and components are in good agreement with actual fuel cell tests. Using the model, prospects for ultra-high power density SOFC at intermediate temperatures (<800 °C) are examined. The results show that even in thin electrolyte film anode-supported cells, the ohmic contribution can be substantial, not all of it being attributable to electrolyte and electrode materials. The results also show that the electrode particle size has a substantial effect on the activation polarization.

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## 1. Introduction

Considerable work has been reported on solid oxide fuel cells (SOFC) over the past couple of decades [1–3]. Various materials for the three components, namely cathode, electrolyte and anode, have been investigated over the past 30+ years. The state-of-the-art materials for SOFC are: 8 mol.% Y<sub>2</sub>O<sub>3</sub>-stabilized zirconia (YSZ) for the electrolyte, as a constituent in porous Ni + YSZ composite anode and as a constituent in porous Sr-doped LaMnO<sub>3</sub> (LSM) + YSZ composite cathode. Much of the early work was based on using porous LSM as the cathode without YSZ dispersed in it. Over the last couple of decades many other highly active cathodes have emerged, the vast majority of them being mixed ionic electronic conducting (MIEC) perovskites containing transition metals capable of exhibiting multiple valence states. Much of the early work was also based

on using YSZ plates of 150–200 microns in thickness (or in some cases even thicker) for the electrolyte with screen-printed anode and cathode. Such cells are referred to as electrolyte-supported cells. In order to minimize the ohmic contribution to the total cell resistance such cells have to be operated at high temperatures, typically ~900–1000 °C, to ensure reasonable performance (~0.25 Wcm<sup>-2</sup>). The use of single phase LSM also leads to generally poor cathode performance at temperatures below 900 °C. High temperature operation poses significant challenges related to materials degradation especially when used in a planar geometry and with a metallic interconnect.

In subsequent work in this field processes were developed for fabricating cells comprising thin YSZ electrolyte film supported on a porous electrode (typically the anode). This allowed for a significant lowering of the total ohmic contribution thereby enhancing performance. Using state-of-the-art materials maximum power densities approaching 2 Wcm<sup>-2</sup> at a temperature as low as 800 °C were demonstrated in anode-supported button cells [2,3]. This

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represented not only an order of magnitude improvement in performance but that too at a temperature 200° lower. Many SOFC researchers have been involved in developing such high performance thin electrolyte, electrode-supported cells at lower operating temperatures over the past couple of decades. Further improvements in performance can be realized by using other perovskite cathodes, especially those containing Co and Fe as B-site constituents exhibiting mixed ionic electronic conducting (MIEC) properties. It is to be emphasized, however, that fine-grained two-phase LSM + YSZ (or LSM + rare earth oxide doped ceria) cathodes perform nearly as well as many single phase LSC and LSF-based cathodes assuming suitable microstructures have been developed. Also, some of the highest performance reported to date has been on cells made with composite cathodes. Finally, the lowering of the ohmic contribution requires the use of electrolyte materials with higher ionic conductivity than YSZ. These materials include scandia-stabilized zirconia (ScSZ), Sr-doped and Mg-doped LaGaO<sub>3</sub> (LSGM) and rare earth oxide doped ceria. Limited amount of work has been reported on SOFCs made with these other electrolyte materials in a thin film form.

While much progress has been made it also appears that there have not been significant further gains in performance, beyond those achieved about fifteen years ago. One of the difficulties has been in accurately measuring the various polarization losses which has made it challenging to target those areas requiring more effort. There are five sources of voltage loss: (a) Ohmic loss – voltage loss associated with the electrolyte, the electrodes and contact regions between the electrodes and the electrolyte; (b) Concentration polarization at the cathode – voltage loss associated with the transport of the oxidant through the porous cathode, (c) Concentration polarization at the anode – voltage loss associated with the transport of the fuel through the porous anode, (d) Activation polarization at the cathode – voltage loss associated with the electrochemical reduction of the active constituent in the oxidant (oxygen) and (e) Activation polarization at the anode – voltage loss associated with the electrochemical oxidation of the active constituent in the fuel (typically hydrogen). The activation polarization involves a number of series steps such as gas adsorption, dissociation, electron transfer and transfer of ionic species into (or out of) the electrolyte at the electrode catalyst (e.g. LSM)/electrolyte (e.g. YSZ) interface. In a typical cell it is usually difficult to separately measure anode and cathode polarization losses and also it is equally difficult to experimentally separate out concentration and activation polarization losses at either of the two electrodes. The most commonly used technique is electrochemical impedance spectroscopy (EIS). Almost always the EIS spectra overlap and it is often not possible to unequivocally determine the various contributions. And even if the EIS spectra can be described using a number of equivalent circuits it is generally difficult to assure uniqueness (multiple equivalent circuits can be used to describe a given experimental data set thus making physical interpretation difficult). It appears that to realize further improvements in cell performance may require accurate identification of the various polarization losses, their sources and their dependence on material and microstructural parameters, atmosphere and temperature. The objective of this work is to use a parametric model for SOFC [4], quantitatively estimate the various polarization losses based on measurements made on cell materials and components and determine what parameters may need to be optimized to improve the performance, beyond what has been achieved to date.

## 2. A parametric model for SOFC

Several models which take into account gas transport through

porous electrodes, electrochemical reactions at the electrodes (near electrode/electrolyte interfaces) including various reaction steps and the ohmic loss have been developed [5–11]. The vast majority of them, however, are numerical in nature due to the analytical complexities involved. Also, the vast majority of the models are based on many assumed parameters regarding the microstructure of the electrodes, assumed chemical reaction steps and parameters related to the reaction steps (e.g. an assumed rate determining step). Thus, quantitative validation of virtually all of these models is impossible for lack of experimentally verifiable/measurable parameters used in many of the models. The main difficulty lies in the fact that multiple *assumed* parameters are required for modeling, while the experimental voltage vs. current density traces are featureless. As a consequence, the *inverse* problem of determining the various parameters from the measured cell performance curves lacks uniqueness. Essentially the same difficulty arises in the use of EIS on actual cells since the inverse problem of determining various cell parameters from EIS spectra lacks uniqueness.

It appears that an approach of using a parametric model which is based on parameters that can be experimentally measured on cell materials and components, at least in principle, could be the first step towards developing a thorough understanding of the various voltage losses that occur in a typical SOFC [4]. Calculation of cell performance curves based on measured parameters on cell materials and cell components is a *forward* problem, and thus in principle it has a unique solution for a given set of parameters [12]. It is understood, however, that there usually will be multiple sets of different parameters which can give the same cell performance. Such an approach can estimate the dominant sources of voltage losses thus identifying areas which require further work in order to increase the performance. Detailed numerical modeling may then be the next step to allow for the inclusion of multi-dimensional features in addition to issues such as fuel utilization and non-isothermal operation.

A typical anode-supported SOFC consists of at least five distinct layers: (a) A porous anode-support characterized by high electronic conductivity to minimize the ohmic loss and high porosity for easy transport of gaseous fuel (to minimize anode concentration polarization); (b) A porous anode functional layer adjacent to the electrolyte characterized by fine microstructure for enhanced electrocatalysis (electrochemical oxidation of fuel; lowering of the anode activation polarization); (c) A dense, thin (to minimize the ohmic loss) film electrolyte; (d) A porous cathode functional layer adjacent to the electrolyte characterized by fine microstructure for enhanced electrocatalysis (electrochemical reduction of the oxidant; lowering of the cathode activation polarization); and (e) A porous cathode current collector layer characterized by high electronic conductivity to minimize the ohmic loss and high porosity for easy transport of the oxidant (to minimize cathode concentration polarization). In a parametric model voltage vs. current density polarization curves of an SOFC may be adequately described by a generic equation of the form [3,13].

$$V(i) = E_o - iR_i - \eta_{act}^a - \eta_{act}^c - \eta_{conc}^a - \eta_{conc}^c \quad (1)$$

In Equation (1)  $E_o$  is the open circuit voltage,  $i$  is the current density,  $R_i$  is the ohmic area specific resistance ( $\Omega \text{ cm}^2$ ),  $\eta_{act}^a$  is the activation polarization at the anode,  $\eta_{act}^c$  is the activation polarization at the cathode,  $\eta_{conc}^a$  is the concentration polarization at the anode and  $\eta_{conc}^c$  is the concentration polarization at the cathode. The voltage vs. current density polarization curves may further be described by Refs. [3,13].

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