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Model validation and performance analysis of regenerative solid oxide cells: Electrolytic operation

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ABSTRACT

The use of regenerative, high temperature solid oxide cells (SOCs) as energy storage devices has the potential for round-trip efficiencies that are competitive with other storage technologies. The focus of the current study is to investigate regenerative SOC operation (i.e., working in both fuel cell and electrolysis modes) through a combination of modeling and numerical simulation. As an intermediate step, this paper focuses on the electrolysis mode and presents a dynamic cell model that couples the reversible electrochemistry, reactant chemistry, and the thermo-fluidic phenomena inside a cell channel. The model is calibrated and validated using available experimental and numerical data for button cells, single cells, and multi-cell stacks supplied with either steam or syngas. Parametric studies are also performed to show how the investigated parameters affect model validity. The results show that the present model can accurately simulate the electrolytic cell behavior, especially in the low current range, which is a favored operating point in practical systems. It is observed that improvements in stack-level model precision require further investigation to better represent the contact resistance of the stack components and to improve the estimation of the activation polarization throughout the operating envelope. It is also concluded that the CO₂ electrochemical reaction can be neglected when the concentration of the steam supplied to the cell is high enough to support the water–gas shift reaction. Copyright © 2013, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

1. Introduction

Regenerative (or reversible) solid oxide cells (rSOCs) can operate in two modes: power-producing fuel cell mode and syngas generating electrolysis mode. There are many attractive applications for rSOCs that include, but are not limited to electrical energy storage (EES) [1]. Some recent studies show that rSOC systems, if properly designed, have the potential to store energy in electrolysis mode and release it later in fuel cell mode while achieving round-trip efficiencies competitive with other available storage technologies [2–4].

A regenerative SOC system necessarily requires pumps, tanks, and heat exchangers to make the device realizable and ultimately, an entire system must be designed and developed to perform with high round-trip efficiency. The round-trip efficiency, which is a key figure of merit for developing energy storage systems, is the fraction of the original electrical energy recovered (or released to the external circuit) after the storage and utilization cycle [2,4]. A critical requirement in developing system models to facilitate the design of energy storage systems is a high fidelity cell-stack level model that can be employed to predict both steady-state and dynamic

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stack performance so that considerations such as SOC operating conditions (e.g., T, P, composition, utilization), desired tank storage state points (T, P, and composition), thermal management of the EES system, and operating strategies for load change and mode-switching can be properly evaluated. Thus, the present work seeks to develop a high-fidelity, dynamic SOC stack simulation tool for the purposes outlined above.

Steady-state and dynamic behavior of solid oxide fuel cells (forward, power-producing) at both cell and system levels have been investigated in several prior works (a thorough review is presented in Ref. [5]), including that of the authors [6–8]. The effects of operating parameters on solid oxide fuel cell (SOFC) performance are well-predicted and can be employed for preliminary system design and specification; although more attention and experimental clarification are always needed to reduce model error in prototype systems.

In the reverse operational mode (i.e., electrolysis mode), both experimental and numerical prediction of solid oxide electrolysis cell (SOEC) performance has yet to be reliably established given its relative technological infancy. As the round-trip efficiency of the total system is related to both modes of operation, further understanding of the complex SOEC operating mechanisms in terms of heat, mass, and charge transfer phenomena that are closely coupled with heterogeneous and electrochemical reaction chemistry is valuable. Most of the extant literature involving SOECs focuses on hydrogen production under both steady-state and dynamic conditions [9–11]. Syngas and alternative fuels production using SOECs [12–14,39] and regenerative solid oxide fuel cells [15,16] are also receiving increased attention. In an energy storage application using rSOCs, operation at lower temperature (~650 °C) and higher pressure (~20 bar) are desirable. However, accurate and robust prediction of SOEC performance for the present application requires more supporting experimental and numerical research since the desired operating conditions are very different from that of typical SOFCs/SOECs. It is also worth noting that much of the previous work regarding SOEC modeling relies on minor changes to existing SOFC models with limited experimental validation. Therefore, in some cases the predicted voltage–current characteristic curves are far from the typical experimental results found in the literature for SOECs.

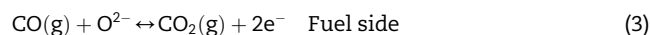
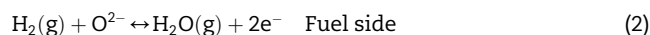
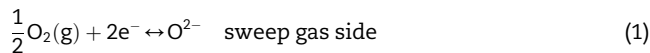
The model validation cases investigated in this paper have shown that there are several issues which need to be resolved in developing a reliable SOEC model capable of operating in both modes. Consequently, as an intermediate step toward developing a sophisticated dynamic rSOC model and identifying favorable electrochemical and thermal performance, this paper focuses on the development of a steady-state cell model for SOECs. A companion paper by the authors investigates fully reversible operation of regenerative SOCs [40]. The present work improves and extends existing models to enable exploration of cell geometry, operating conditions, and feed gas effects. In this paper, the modeling approach is first presented and then results from model calibration and validation with the available experimental and numerical data in the literature are discussed. Throughout the validation cases, the model is also employed to investigate the effect of several parameters on performance and to provide insight for future

research and development related to system design and optimization.

2. Theory and operation of SOECs

2.1. Principles of operation

Multiple reactions can occur simultaneously in the fuel channel of an SOEC depending on the inlet fuel composition, operating conditions, and electrode material set as:



In addition to the above electrochemical reactions, reactant gases containing CH₄, CO₂ and CO mixtures can participate in catalyzed heterogeneous reactions within an electrode/electrode support material set to either produce or consume water. For example, in the presence of Ni containing electrodes, the above gases can undergo steam reforming (SR) (or methanation), as well as the water–gas shift reaction (WGS) (or reverse water–gas shift (RWGS)). These reactions are discussed in greater detail in subsequent sections.

Laboratory data show that the kinetics of the CO₂ electrochemical reduction reaction at the SOEC cathode are generally slow compared with steam reduction, and CO₂ consumption mostly depends on the RWGS reaction when steam is present. It is also observed that there are almost no changes in the apparent area specific resistance (ASR) for SOEC operation when supplied with both steam and CO₂ [17].

2.2. SOEC materials and stack configuration

In either regenerative or stand-alone operation, SOECs employ similar material sets and geometric configurations as SOFCs [18]. Due to the lower power density (because of the increased current path), as well as the higher manufacturing cost of tubular SOCs, the considered geometry here is of planar type. Planar designs are also considered in several experimental studies from small- to large-scale systems for hydrogen and syngas production [12,14]. A schematic of the selected configuration is shown in Fig. 1.

2.3. Inlet compositions

For the proposed energy storage application, the purpose of SOEC operation is to convert an H₂O/CO₂-rich gas mixture into a fuel (CO, CH₄, H₂) for storage and later usage in discharge mode (i.e., fuel cell power generation). Although any combination of H₂, CO, CO₂, H₂O, CH₄ and N₂ is certainly possible as a feedstock gas mixture; a gas mixture rich in H₂O and CO₂ content is considered as the supply stream into the fuel channel (i.e., cathode side in electrolysis mode) in order to effectively generate enough fuel for storage. The presence of a certain amount of hydrogen in the fuel composition is also

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