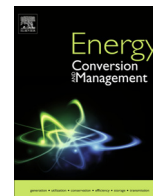




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# Performance comparison of solid oxide steam electrolysis cells with/without the addition of methane



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## ABSTRACT

Hydrogen is considered a clean energy carrier for the future. At present, the production of hydrogen via a solid oxide electrolysis cell is of interest because water is the only reactant used; however, hydrogen production through electrolysis technology is still costly due to high electrical energy consumption. To reduce this energy demand, an addition of methane to the anode side of the solid oxide electrolysis cell, where it behaves like the anode side of the solid oxide fuel cell and generates heat and electricity to accomplish the electrolysis process, is one interesting method. In this study, modeling of the solid oxide fuel-assisted electrolysis cell is performed based on an electrochemical model to analyze the performance of the electrolyzer with/without the addition of methane in terms of the power input and the energy efficiency. In addition, the effect on the electrolyzer cell by key operating parameters, such as current density, steam fraction, steam-to-carbon ratio, temperature, pressure, steam utilization and fuel utilization, is presented. The simulation analysis shows that the performance of the solid oxide fuel-assisted electrolysis cell is higher than that of conventional solid oxide electrolysis cell, as it requires a lower power input. Furthermore, it is possible to run the solid oxide fuel-assisted electrolysis cell without an external electrical energy input.

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## 1. Introduction

Currently, hydrogen is considered as a carrier of clean and sustainable energy. Hydrogen can be used in a combustion device without greenhouse gas emission, it is used as a reactant in many industries such as the hydrogenation process [1], and it can be used in a fuel cell for producing electrical energy and thermal energy [2]. To date, hydrogen is mainly produced via the thermochemical process from fossil fuels [3–5]; however, because of the depletion and the environmental problem of fossil fuels [6], the renewable and sustainable processes have received more attention. Water electrolysis via the electrochemical process is one of promising processes for hydrogen production because it uses only water as a reactant. This water electrolysis, which uses electricity to decompose water into hydrogen and oxygen, is the cleanest method when using electricity produced by nuclear energy or renewable

energy [7]. Among various types of electrolysis cells, the solid oxide electrolysis cell (SOEC) has received considerable attention because its high temperature operation (700–1000 °C) leads to lower electrical energy consumption and higher reaction kinetics compared with low temperature electrolysis cells (alkaline and proton exchange membrane). Furthermore, heat generation is a by-product in the SOEC because of voltage losses (activation overpotential, concentration overpotential and ohmic loss), and it can be used as a heat source for steam fed into the SOEC [8].

As mentioned above, the SOEC is an attractive process for hydrogen production; however, it is expensive for competition in the market due to two main reasons. Firstly, high temperature SOEC has materials and catalyst limitations that lead to a high fabrication cost. Secondly, although thermal energy demand can replace electrical energy demand with high temperature operation of SOEC, the SOEC still requires high electrical energy consumption; this in turn creates a high electricity cost. Although, the SOEC is not used commercially to produce hydrogen, improvement and development of the SOEC is still necessary and interesting [9]. Many of the recent efforts in SOEC development have been devoted

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### Nomenclature

$D_{AB}$	molecular binary diffusion coefficient ( $\text{m}^2 \text{s}^{-1}$ )	$R$	gas constant ( $\text{J mol}^{-1} \text{K}^{-1}$ )
$D_{Ak}$	Knudsen diffusion coefficient of species A ( $\text{m}^2 \text{s}^{-1}$ )	$R_{\text{ele}}$	electrochemical reaction rate ( $\text{mol m}^{-2} \text{s}^{-1}$ )
$D_{\text{an}}^{\text{eff}}$	effective diffusivity of the gas mixtures at the anode side ( $\text{m}^2 \text{s}^{-1}$ )	$T$	temperature (K)
$D_{\text{ca}}^{\text{eff}}$	effective diffusivity of the gas mixtures at the cathode side ( $\text{m}^2 \text{s}^{-1}$ )	$U_{\text{steam}}$	steam utilization
$E$	open-circuit voltage (V)	$U_{\text{fuel}}$	fuel utilization
$E^0$	potential at standard pressure (V)	$V_{\text{SOEC}}$	cell voltage of the SOEC (V)
$E_i$	activation energy of the electrode ( $\text{J mol}^{-1}$ )	$V_{\text{SOFC}}$	cell voltage of the SOFC (V)
$F$	Faraday's constant ( $\text{C mol}^{-1}$ )	$W$	cell width (m)
$J$	current density ( $\text{A m}^{-2}$ )	<i>Greek letters</i>	
$J_{0,i}$	electrode exchange current density ( $\text{A m}^{-2}$ )	$\alpha$	transfer coefficient
$k_i$	pre-exponential factor of the electrode ( $\Omega^{-1} \text{m}^{-2}$ )	$\varepsilon_{\text{SOFC}}$	efficiency of SOFC (%)
$K_{\text{eq,SR}}$	equilibrium constants of steam reforming reaction	$\varepsilon$	electrode porosity
$K_{\text{eq,WGS}}$	equilibrium constants of water–gas shift reaction	$\eta_{\text{act}}$	activation overpotential (V)
$L$	cell length (m)	$\eta_{\text{conc}}$	concentration overpotential (V)
$LHV_i$	lower heating value of component i at inlet	$\eta_{\text{ohm}}$	ohmic loss (V)
$n$	number of electrons participating in the electrochemical reaction	$\sigma_{\text{an}}$	electrical conductivity of the anode ( $\Omega^{-1} \text{m}^{-1}$ )
$\dot{N}_{i,\text{in}}$	inlet flow rate of gas component i ( $\text{mol s}^{-1}$ )	$\sigma_{\text{ca}}$	electrical conductivity of the cathode ( $\Omega^{-1} \text{m}^{-1}$ )
$P$	pressure (Pa)	$\sigma_{\text{ele}}$	ionic conductivity of the electrolyte ( $\Omega^{-1} \text{m}^{-1}$ )
$P_{\text{H}_2}$	partial pressure of hydrogen (Pa)	$\tau_{\text{an}}$	anode thickness (m)
$P_{\text{H}_2\text{O}}$	partial pressure of steam (Pa)	$\tau_{\text{ca}}$	cathode thickness (m)
$P_{\text{O}_2}$	partial pressure of oxygen (Pa)	$\tau_{\text{ele}}$	electrolyte thickness (m)
$P_{\text{H}_2}^{\text{TPB}}$	partial pressure of hydrogen at triple-phase boundary (Pa)	$\zeta$	electrode tortuosity
$P_{\text{H}_2\text{O}}^{\text{TPB}}$	partial pressure of steam at triple-phase boundary (Pa)	<i>Subscripts</i>	
$P_{\text{O}_2}^{\text{TPB}}$	partial pressure of oxygen at triple-phase boundary (Pa)	an	anode
$P_{\text{SOEC}}$	power density of SOEC ( $\text{W m}^{-2}$ )	ca	cathode
$P_{\text{SOFEC}}$	power density of SOFEC ( $\text{W m}^{-2}$ )	<i>Superscripts</i>	
		TPB	triple-phase boundary

to reduce its operating costs by a decrease in SOEC electricity consumption. Pham et al. [10] first came up with the idea to reduce electrical energy by adding a natural gas such as methane to react with oxygen at the anode side of the SOEC. In addition, Martinez-Frias et al. [11] proposed a novel and high-efficiency SOEC. In their experiment, natural gas is fed into the anode side of single SOEC at 700 °C. It was found that natural gas reacts with oxygen produced in the anode side through partial oxidation or total oxidation, which causes a voltage reduction as much as 1 V compared with conventional SOEC. However, they found that carbon deposition on the anode material can occur when the SOEC operates at high temperature (900 °C). Avoidance of carbon deposition can be achieved by feeding steam with natural gas at the anode.

Apart from this, the solid oxide fuel-assisted electrolysis cell (SOFEC) has been the focus of much research. The mixture of methane and steam is fed to the anode side of the SOEC, followed by the steam reforming of methane and water–gas shift reaction. Hydrogen produced is further reacted with oxygen; thus, the electrochemical reaction occurs and leads to electricity production. It can be implied that the anode side of SOEC works like the anode side of solid oxide fuel cell (SOFC). Tao and Virkar [12] developed the cathode materials for use in hybrid electrochemical devices (SOFC/SOEC/SOFEC) that comprise the SOFC and the SOFEC for co-production of hydrogen and electricity directly from distributed natural gas or alternative fuels. Wang et al. [13] also developed electrode materials, which are a Co–CeO<sub>2</sub>–YSZ cathode and a Pd–C–CeO<sub>2</sub>–YSZ anode, to replace Ni/YSZ electrodes and inhibit the carbon deposition that occurs from methane at high operating temperature. Moreover, Luo et al. [14] studied the performance of the SOFEC fueled by carbon monoxide and methane based on

experimentation and elementary reaction modeling. It was found that the performance of CH<sub>4</sub>-SOFEC is higher than that of CO-SOFEC. The mechanism shows that methane is not electrochemically oxidized directly but converted into carbon monoxide and hydrogen first through a steam reforming reaction and then followed by electrochemical oxidation and hydrogen production. As a consequence, the SOFEC can produce not only hydrogen at the cathode but also generate electricity at the anode.

From above literatures, it can be observed that many previous studies have focused on improving the materials of SOFEC. An investigation into SOFEC based on mathematical modeling, which is required to characterize the SOFEC electrical performance related to reaction and mass transport processes that occurs in the SOFEC, has been limited. Luo et al. [14] proposed an elementary reaction kinetic model of SOFEC coupling with heterogeneous elementary reactions and electrochemical reactions. The proposed model was calibrated and validated with the experimental data and the predicted polarization curves agreed well with the experimental data. Although, the SOFEC model is commonly governed by the conservation equations and electrochemical reaction, an electrochemical model of the SOFEC describing the conversion of electrical power to hydrogen plays an important role in characterizing the electrical performance of SOFEC. The inclusion of all voltage losses (i.e., activation, ohmic and concentration losses) in the electrochemical model results in a more accurate prediction of the SOFEC, and it could lead to a more optimal operating condition and design of SOFEC.

In this study, the performance of SOFEC for hydrogen production is investigated by considering its electrical characteristics obtained from a detailed an electrochemical model. The

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