



Dynamic electro-thermal modeling of co-electrolysis of steam and carbon dioxide in a tubular solid oxide electrolysis cell



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ABSTRACT

SOEC (Solid oxide electrolysis cell) has great potential to store unstable renewable energy in form of steady chemical energy. For this application, transient behavior in variable working condition is crucial. A 2D (two-dimensional) dynamic model was developed to predict the dynamic response of H₂O/CO₂ co-electrolysis in TSOEC (tubular solid oxide electrolysis cell). The dynamic model comprehensively considered the macroscopic fluid flow, microscopic diffusion, heat transfer and electrochemical/chemical reactions in the TSOEC. In this paper, the TNV (thermal neutral voltage) of TSOEC was specifically defined and calculated. The time constants of charge/mass/heat transport processes were evaluated. Further, the responses of current, gas concentrations, temperature, efficiency, conversions to electricity/gas flow/temperature step inputs were respectively studied. Through the study, appropriate transient operations were designed to improve efficiency and conversions.

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1. Introduction

With fossil fuels running out and environment deteriorating, new forms of energy are urgently demanded. RES (Renewable energy sources) like wind and solar energy can provide people sustaining power in an environmentally-friendly way [1–6]. RES is intermittent, unstable and seasonal-changing. The characteristics bring the difficulty in integrating RES into grid and propose a need for energy storage. Steam and carbon dioxide co-electrolysis in solid oxide electrolysis cell is a promising long-term energy storage way by converting H₂O/CO₂ to syngas. Syngas can be further synthesized into various hydrocarbon fuels by Fischer-Tropsch process [7]. Co-electrolysis in SOECs can not only store and transport renewable energy seasonally, but also effectively reduce greenhouse gas.

Many scholars and research institutes have carried out their researches about SOECs. Risø National Laboratory and Idaho National Laboratory as representatives have successfully demonstrated respective SOEC (Solid oxide electrolysis cell) stacks facilities [8,9]. Most researches focus on SOECs operated in a certain

working condition (steady state) [10–13]. Facing the application of RES storage, SOEC is chronically operated in variable working conditions. Therefore, the transient behaviors of SOEC are quite significant. Presently, a limited number of studies on transient operation of SOEC are reported. Petipas et al. [14] tested steam electrolysis in a single SOEC with square wave input for 140 cumulated hours. In transient operation, the degradation rate was found to stabilize at 5% (70 mV) per 1000 h, in accordance with steady-state operation. Besides dynamic experiments, dynamic modeling is needed to study the transients of transport processes and understand the effect of multi-factors to response time. To prevent the issues of cell component fracture, Cai and Udagawa et al. [15,16] built a 1D dynamic model of a planar SOEC stack for steam electrolysis to control stack temperature through variation of air flow rate. For the application of RES, time delay of SOEC in variable working conditions should be paid attention to, which is rarely discussed in present papers. Abundant dynamic models of SOFC, the reverse process of SOEC, provide good references [17–24]. Bhattacharyya et al. [17] reviewed existing dynamic models and compared the response time constants according to the corresponding plots in the studies. The review indicated the time constants differed from tens of microseconds to thousands of seconds, depending on physical and chemical characteristics, operating condition, cell configuration, control strategies, etc. In

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Nomenclature	
Abbreviation	
cc	Cathode channel
el/ca	Interface of electrolyte and cathode
LHV	Lower heating value (J mol^{-1})
LSM	Lanthanum strontium manganate
MEA	Membrane electrode assembly
MR	Methanation reaction
PEN	Positive-electrolyte-negative
RES	Renewable energy sources
ScSZ	Scandium stabilized zirconium
SOEC	Solid oxide electrolysis cell
SOFC	Solid oxide fuel cell
TNV	Thermal neutral voltage
TSOEC	Tubular solid oxide electrolysis cell
TPB	Triple phase boundary
WGSR	Water–gas shift reaction
YSZ	Yttrium stabilized zirconium
English letter	
C_{dl}	Specific interface double-layer capacitance between electronic and ionic conductor phases
c_i	Molar concentration of gas-phase species i
C_p	Heat capacity ($\text{J kg}^{-1} \text{K}^{-1}$)
D	Diffusion coefficient ($\text{m}^2 \text{s}^{-1}$)
E_{act}	Active Energy (J mol^{-1})
i_0	Exchange Current Density (A m^{-3})
k	Reaction rate constant
K_p	Reaction equilibrium constant
M	Molar mass (kg mol^{-1})
n_e	Number of electrons participating in the reaction
n_{el}/n_{io}	Fraction number of electronic/ionic conductor particles
N_{tot}	Total number of particles per unit volume
p	Pressure (Pa)
P	Whole range connection probabilities of same kind particles
q	Molar flow rate (mol s^{-1})
Q_M	Mass source term ($\text{kg m}^{-3} \text{s}^{-1}$)
Q_{heat}	Heat source term (W m^{-3})
r	Average pore/particle radius (m)
R	Universal gas constant ($8.314 \text{ J mol}^{-1} \text{K}^{-1}$)
Rt	Reaction production rate ($\text{mol s}^{-1} \text{m}^{-3}$)
S_i	Molar entropies of species i
S_{Ni}	Ni active surface area per unit volume ($\text{m}^2 \text{m}^{-3}$)
S_{TPB}/S_{act}	TPB active area per unit volume ($\text{m}^2 \text{m}^{-3}$)
T	Temperature (K)
v	Velocity (m s^{-1})
V	Voltage or potential (V)
x_i	Molar fraction of gas-phase species i
Z	Mean coordination number
Greek letters	
α	Transfer coefficient
β	Interface conductivity ($\Omega^{-1} \text{m}^{-2}$)
γ	Tuning parameter of exchange current density
ε	Porosity
ζ	Conversion
φ	Molar ratio
η	Efficiency
θ	Contact angle between the electronic and ionic conductors particles
κ	Permeability (m^2)
λ	Heat conductivity ($\text{W m}^{-1} \text{K}^{-1}$)
M	Dynamic viscosity ($\text{kg s}^{-1} \text{m}^{-1}$)
P	Density (kg m^{-3})
Σ	Electric conductivity (S m^{-1})
τ	Tortuosity
Subscript or superscript	
<i>an</i>	Anode layer
<i>ca</i>	Cathode layer
<i>f</i>	Forward reaction
<i>eff</i>	Effective
<i>el</i>	Electronic
<i>elec</i>	Electrolyte
<i>in</i>	Inlet
<i>ion</i>	Ionic
<i>irr</i>	Irreversible
<i>Kn</i>	Knudsen diffusion
<i>mole</i>	Molecular diffusion
<i>ohm</i>	Ohmic
<i>out</i>	Outlet
<i>r</i>	Reverse reaction
<i>ref</i>	Reference condition (at equilibrium state)
<i>Reform</i>	Direct Internal reforming reaction
<i>rev</i>	Reversible
<i>shift</i>	Water–gas shift reaction
<i>tot</i>	Total gas conversion ratio

addition, some models [17,19,21,22] were isothermal, neglecting the processes of heat transfer, some models simplified the diffusion of gases species in the electrodes [18,21–24]. The model simplification could greatly affect the model precision and predicted transient behaviors of SOFC. However, the heat effect of SOEC is quite different from that of SOEC. The heat effect of SOEC mainly determined by endothermic electrolysis and exothermic polarization and ohmic losses happening in PEN (positive-electrolyte-negative) component, could be endothermic mode, exothermic mode and thermoneutral mode. Therefore, the heat transfer process of SOEC is more complicated. The conclusions drawn from SOFC could not be absolutely available for SOEC. In this paper, a two-dimension dynamic model, considering the macroscopic fluid flow, microscopic diffusion, heat transfer and electrochemical/chemical reactions, was developed to predict the transient

behaviors of co-electrolysis in a TSOEC (tubular solid oxide electrolysis cell).

2. Model description

Previously, this TSOEC model has been developed for steady-state simulation of co-electrolysis [25]. Therefore, this section just provides a brief description of model. The governing equations and parameters have been provided in Ref. [25]. More detailed description of the model can be found in Ref. [25].

2.1. Model assumptions and calculation domain

Dynamic model of TSOEC coupled governing equations of charge, mass, momentum, energy transport equations. The model

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