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Chemically-induced mechanical instability of samaria-doped ceria electrolyte for solid oxide electrolysis cells

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ABSTRACT

Doped-ceria is an attractive electrolyte material for solid oxide electrolysis cells (SOECs) operated at intermediate temperatures. However, ceria is highly prone to break down under high applied voltages and low oxygen partial pressures at the fuel side. This phenomenon is analyzed for the typical $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9-\delta}$ electrolyte based on the chemically-induced stress, which is caused by the inhomogeneous distribution of oxygen non-stoichiometry throughout the thickness of electrolyte plate. The sensitivities of the maximum tensile stresses are explored under typical SOEC operating parameters such as temperature, applied voltage and oxygen partial pressure. Varying from short-circuit of solid oxide fuel cell (SOFC) mode to high voltage of SOEC conditions, the applied voltage sharpens the maximum tensile stress by seven times and raises the minimum permitted oxygen partial pressure at the cathode-electrolyte interface by a factor of $10^{4.5}$ at most. The analysis results indicate that a ceria-based electrolyte under SOEC conditions denotes a definite trend of collapse at 700 °C even 600 °C, suggesting the inapplicability of doped-ceria electrolyte in SOEC mode.

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Introduction

Solid oxide electrolysis cells (SOECs) have drawn increasing attention, as a valid system to store the surplus energy from the renewable resources. However, the high operating temperature may cause a series of problems. Similar with the availability in solid oxide fuel cells (SOFCs), it is desirable that

doped-ceria is a potential material served as the electrolyte in SOEC mode [1], due to its high oxygen-ion conductivity at intermediate temperatures. But, as being widely acknowledged, the ceria tends to be reduced by the conversion of Ce^{4+} to Ce^{3+} under a low-oxygen partial pressure. The reduction induces expansion in the lattice [2–4]. Moreover, the chemical expansion depends on the oxygen non-stoichiometry, which

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is corresponding to the extent of the reduction, and varies with the oxygen partial pressure. As for an electrolyte plate under both SOFC and SOEC conditions, there is a differential expansion distribution across its thickness when it's subject to a gradient of oxygen potential. It is the non-uniform that induces so-called chemically-induced stress. This stress plays a crucial role in material mechanical fracture.

Although there are abundant literature about the evaluation of the stress in doped-ceria electrolytes under SOFC conditions [5–7], no work is reported in SOECs. And it is experimentally recognized that SOEC using a doped-ceria electrolyte is prone to break down in the structure [8,9], especially under a high operational voltage. The yttrium stabilized zirconia (YSZ) without any chemically-induced stresses is applied as the SOEC electrolyte. There are literature [10,11] using the build-up of extreme oxygen pressures near the anode, which is caused by the electrode overpotential, as an argument for cracking and delamination of YSZ electrolyte at the SOEC anode. As for doped-ceria electrolytes, except the high oxygen partial pressure, the extra chemically-induced stress may sharpen the inapplicability in SOECs.

At the initial part, an irreversible SOEC electrochemical situation and its corresponding V – I relations are clarified [12,13]. On pointing out the electrode overpotential, which is actually the most dominant factor to the structural fracture, the mechanical stability is essentially determined by the applied voltage V_{ex} , oxygen partial pressure P and operating temperature T . By using electrochemical principles, point defect properties and chemically-combined mechanical laws, this work has carried out an evaluation of the stress across a plate electrolyte of typical samaria-doped ceria (SDC) material under SOEC operating conditions. According to the estimated mechanical stability, the fracture preference has found its theoretical basis.

Evaluation approach

SOEC process with SDC electrolyte

When an external voltage (V_{ex}) is applied to an SOEC with a typical doped-ceria electrolyte of $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9-\delta}$ (SDC20), Fig. 1a [12,13], oxygen ions are generated at the cathode, then transport through the SDC electrolyte to the anode. The cathode is exposed to the fuel environment with low oxygen partial pressure (P_{fuel}), while the anode is exposed to the air atmosphere with high oxygen partial pressure (P_{air}). So oxygen ions are delivered from the P_{fuel} side to the P_{air} side, driven by an applied cell voltage. Meanwhile, electrons transport in the same direction with the oxygen ions, due to the electronic conductivity nature of the doped-ceria electrolyte. The electrolysis process is associated with an oxygen partial pressure change, which is schematically shown in Fig. 1b. The positions of $x = 0$ and $x = L$ refer to the two ends of the SDC electrolyte exclusive of the electrode reaction, which will be discussed later on.

In the SOEC mode, the applied voltage needs to overcome the driving force caused by chemical potential difference ($\Delta\mu_{O_2}$) between the two ends of the electrolyte. This chemical

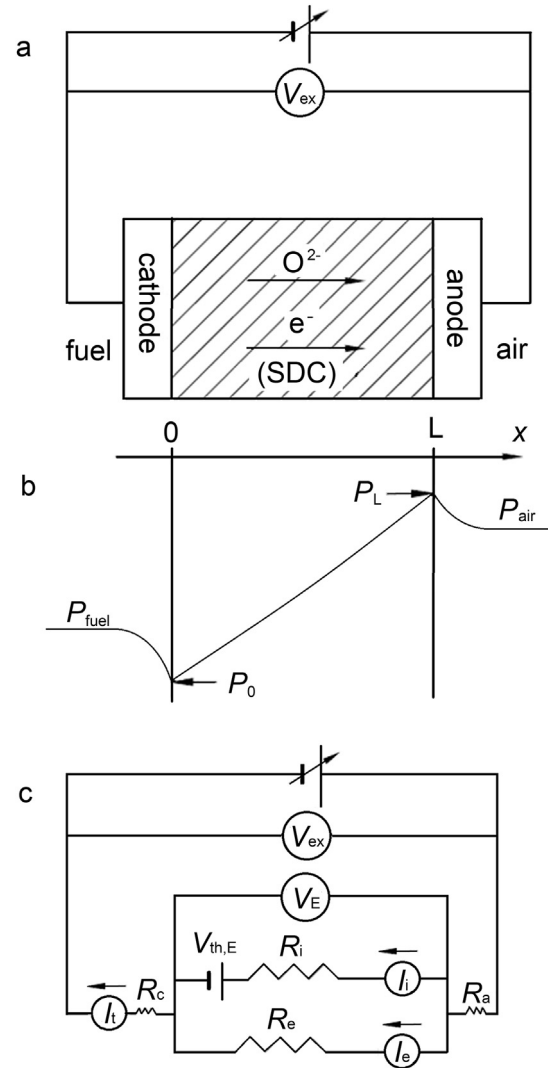


Fig. 1 – (a) Schematic representation, (b) the oxygen partial pressure variation, and (c) the equivalent circuit of an SDC-based SOEC.

potential is related to oxygen partial pressure P_{O_2} (which one abbreviate to P for convenience):

$$\mu_{O_2} = \mu_{O_2}^{\ominus}(T) + kT \ln P \quad (1)$$

and thus the equivalent potential caused by $\Delta\mu_{O_2}$ can be expressed by Nernst equation:

$$V_{th,E} = \frac{kT}{4q} \ln \frac{P_L}{P_0} \quad (2)$$

where $V_{th,E}$ is the Nernst voltage across the electrolyte; k is the Boltzmann constant; T is the temperature; q is the elementary charge; P_L and P_0 are the oxygen partial pressure at $x = L$ and $x = 0$, respectively. Transcending this counteracting potential, the applied cell voltage drives oxygen ions against its chemical potential. So [12,13],

$$V_E = I_i R_i + V_{th,E} \quad (3)$$

where V_E is the applied cell voltage across the interior electrolyte (from $x = 0$ to $x = L$). I_i is the ionic current through the

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