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Failure of solid oxide fuel cells by electrochemically induced pressure

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Abstract

It is well known that many electrochemical devices such as fuel cells, electrolyzers, batteries, etc. degrade under various operating conditions. Many devices are based on solid electrolytes. In many cases, degradation manifests as electrode delamination and/or cracking of the electrolyte. An apparently obvious conclusion often reached is that this must be due to weak interfaces and some approach to mechanical strengthening or toughening should alleviate the problem. It is the intent of this manuscript to show that such electrochemical degradation often occurs due to electrochemical-mechanical coupling effects which result in extremely large internal pressures. So large can be the pressures that no amount conventional strengthening or toughening can prevent degradation. In fact, even a perfect lattice can be disrupted by electrochemically induced pressure leading to failures. In order to prevent failures, it is necessary to fully understand the role of coupling and develop strategies of mitigating high pressure formation by optimizing ion and electron transporting properties of the membrane. This manuscript discusses a fundamental mechanism of electrochemical failure of solid oxide fuel cells, solid oxide electrolyzer cells, and oxygen separators based on oxygen ion conducting solid electrolytes. Experimental results on two types solid electrolyte cells are presented in support of the mechanism.

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Nomenclature

p_{O_2} oxygen (partial) pressure

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E_C	cell voltage
E	nernst voltage
E_A	applied voltage
T	temperature
R_i	ionic area specific resistance of the cell
R_e	electronic area specific resistance of the cell
r_i^a	anode area specific ionic charge transfer resistance
r_e^a	anode area specific electron transfer resistance
$I_{O^{2-}}$	oxygen ion current density
I_e	electronic current density
e	electronic charge
k_B	Boltzmann constant
K_I	mode I stress intensity factor
K_{Ic}	fracture toughness (critical stress intensity factor)
V_c	volume of a penny-shaped crack of radius c under internal pressure
Y	Young's modulus of elasticity
ν	Poisson's ratio

1. Introduction

Many active electrochemical devices such as fuel cells, batteries, electrolyzers involve the transport of predominantly ionic species through membranes under generated and/or applied voltages. Examples include solid oxide fuel cells (SOFC), solid oxide electrolyzer cells (SOEC), proton exchange membrane fuel cells (PEMFC), li-ion batteries (LIB), etc. The transporting ion(s) depend upon the type of the device. In a typical SOFC or an SOEC, high temperature solid electrolyte used is usually an O^{2-} conductor. In PEMFC, the membrane is a proton (H^+) conductor. In LIB, the electrolyte is a Li^+ conductor. During both charging and discharging, electrochemical (half-cell) reactions occur at the two electrodes. The sum of the two half-cell reactions gives a complete chemical reaction. For example, in a fuel cell the half-cell reactions may be: (1) $\frac{1}{2}O_2 + 2e^- \rightarrow O^{2-}$ and (2) $O^{2-} + H_2 \rightarrow H_2O + 2e^-$ with the net reaction being $H_2 + \frac{1}{2}O_2 \rightarrow H_2O$. In the SOEC in which the objective is to split H_2O to form H_2 and $\frac{1}{2}O_2$, the half-cell reactions are: (1) $H_2O + 2e^- \rightarrow H_2 + O^{2-}$ and (2) $O^{2-} \rightarrow \frac{1}{2}O_2 + 2e^-$ with the net reaction being $H_2O \rightarrow H_2 + \frac{1}{2}O_2$. In many cases, the same cells are used for SOFC and SOEC. However, the rate of degradation has been observed to be much faster in the SOEC mode as compared to the SOFC mode [1, 2]. Specifically, it has been found that the oxygen electrode in SOEC (under which O_2 evolves) often completely delaminates. The nature of delamination may at a first glance be interpreted that the interface must be

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