



Durability evaluation of reversible solid oxide cells



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HIGHLIGHTS

- Durability evaluation of solid oxide cells in the reversible mode.
- SOFC is not suitable for electrolysis operation directly, while SOEC has an acceptable performance in fuel cell operation.
- One Ceramtec SOEC showed performance improvement during long-term electrolysis operation.

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ABSTRACT

An experimental investigation on the performance and durability of single solid oxide cells (SOCs) is under way at the Idaho National Laboratory. Reversible operation of SOCs includes electricity generation in the fuel cell mode and hydrogen generation in the electrolysis mode. Degradation is a more significant issue when operating SOCs in the electrolysis mode. In order to understand and mitigate the degradation issues in high temperature electrolysis, single SOCs with different configurations from several manufacturers have been evaluated for initial performance and long-term durability. Cells were obtained from four industrial partners. Cells from Ceramtec Inc. and Materials and Systems Research Inc. (MSRI) showed improved durability in electrolysis mode compared to previous stack tests. Cells from Saint Gobain Advanced Materials Inc. (St. Gobain) and SOFCPower Inc. demonstrated stable performance in the fuel cell mode, but rapid degradation in the electrolysis mode, especially at high current density. Electrolyte–electrode delamination was found to have a significant impact on degradation in some cases. Enhanced bonding between electrolyte and electrode and modification of the electrode microstructure helped to mitigate degradation. Polarization scans and AC impedance measurements were performed during the tests to characterize cell performance and degradation.

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1. Introduction

Large-scale non-fossil hydrogen production is an important technology that impacts the hydrogen economy. Approximately 53 million metric tons of hydrogen were produced in 2010 globally. An annual growth rate of 5.6% was forecast for 2011–2016 [1]. However, most hydrogen production is based on fossil fuels including natural gas, oil, and coal. Non-fossil large-scale hydrogen production methods are gaining increasing interest all over the world. High temperature electrolysis (HTE) is one of the most efficient technologies for the production of carbon-free hydrogen at large scale [2,3]. INL has demonstrated HTE at the multi-kW scale with a hydrogen production rate in excess of 5000 NL h⁻¹ [3,4]. However,

technical barriers need to be resolved before commercialization of HTE technology. The major issue for HTE is long-term performance degradation of the solid oxide electrolysis cells (SOECs) [5–8].

Besides the demand for large-scale hydrogen production, there is growing interest in reversible solid oxide cells (RSOC) [9–15]. By integrating an RSOC system into a power plant, hydrogen can be produced through high temperature electrolysis at times of low power demand and stored as an energy source. During periods of high power demand, additional electricity can be generated from the stored hydrogen by RSOC operation in the fuel cell mode [16,17]. However, although solid oxide fuel cells (SOFCs) can be directly operated in the electrolysis mode, practically they exhibit much higher degradation rates in the electrolysis mode than in fuel cell mode [18,19]. Therefore, degradation in the electrolysis mode becomes a significant problem for RSOC application.

In our previous SOEC stack tests, air electrode delamination, Cr vapor poisoning, microstructure degradation, and seal leakage

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Nomenclature

ASR	area specific resistance ($\Omega \text{ cm}^2$)
HTE	high temperature electrolysis
i	current density (A cm^{-2})
INL	Idaho National Laboratory
MSRI	Materials and Systems Research Inc.
P&ID	Piping and instrument diagram
RSOC	reversible solid oxide cell
sccm	standard cubic centimeters per minute
SOC	solid oxide cell
SOEC	solid oxide electrolysis cell
SOFC	solid oxide fuel cell
St. Gobain	Saint Gobain Advanced Materials Inc.
V	cell voltage (V)

were found to significantly affect the durability of the stack [20,21]. The investigation on degradation issues was focused on single cells in this paper. A new experimental apparatus was developed for single cell tests. SOCs obtained from Ceramtec, MSRI, St. Gobain, and SOFCPower were investigated experimentally. Initial performance and long-term durability tests were conducted in both the fuel cell and electrolysis modes of operation.

2. Materials and experimental apparatus

The state-of-the-art SOCs used for this study were provided by Ceramtec Inc., Materials and Systems Research Inc. (MSRI), Saint Gobain Advanced Materials (St. Gobain), and SOFCPower Inc.. The cells provided from Ceramtec were electrolyte-supported button cells with 2.25 cm^2 active area, while the others use electrode-supported square cells, $5 \text{ cm} \times 5 \text{ cm}$ in dimension. Materials used in the Ceramtec cells are ScSZ electrolyte, Ni–Ceria steam/hydrogen electrode, and La–Co–Fe oxide-based perovskite air electrode. MSRI, St. Gobain, and SOFCPower used Ni/YSZ-supported cells with 8YSZ electrolyte. LSCF is used as the air electrode material in MSRI and SOFCPower cells, while in St. Gobain cells both modified LSM and LSCF are used. Ceramtec and MSRI cells were fabricated as SOECs, with the microstructure of the cells specifically optimized for operation in the electrolysis mode. St. Gobain and SOFCPower cells were fabricated as SOFCs, and were optimized for long-term operation in the fuel cell mode.

A newly developed single-cell test fixture was used to test the SOCs from MSRI, St. Gobain, and SOFCPower. An exploded view of the test fixture is shown in Fig. 1. The apparatus is designed for single cell and short stack tests in the reversible mode. In addition, Cr poisoning is minimized during long-term tests. Referring to Fig. 1, a hydrogen/steam mixture is fed from the bottom through a 6.35 mm OD coiled Inconel tube into the inlet hole in the bottom of the Hastelloy-X (HastX) base plate. The flow then passes through a slot at the bottom of the Alumina cell holder. A mica/glass cell gasket is placed between the cell holder and the nickel plate for sealing. The nickel plate works as the current collector for the steam electrode. A corrugated nickel flow field and a nickel felt are used for managing the hydrogen/steam flow and for electric conduction. After passing along the bottom of the cell, the flow exits through another slot and vents out via a 9.53 mm OD inconel tube. The outlet tube is sized larger than the inlet tube to minimize the back pressure on the cell seal.

On the air side of the cell, a gold-plated perforated inconel plate is used as the current collector and air flow distributor. Air is introduced through a tube that is welded to the inconel plate. Air flow is distributed along the air side of the cell through an array of

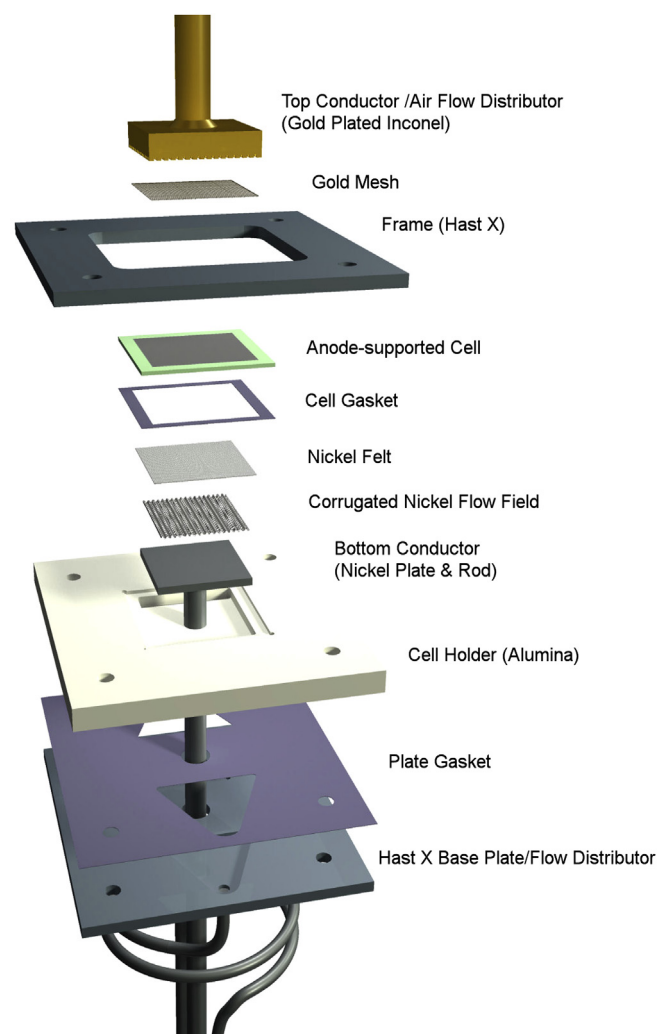


Fig. 1. Exploded view of the cell fixture used for testing MSRI, St. Gobain, and SOFCPower SOCs.

flow channels milled into the bottom of the inconel plate. Air exhaust gas vents out to the furnace. A gold mesh is placed between the air electrode and the plate to minimize ohmic loss and to further improve air flow distribution.

A fixed compression load is applied to the solid oxide cell by means of weights, as shown in the test stand overview, Fig. 2. The load is transferred via an alumina tube from the dead weights to the top cell contact plate. This load simultaneously compresses the cell against the nickel felt, the flow field and the current collector on the bottom steam/hydrogen side of the cell and against the gold mesh on the air side. It also compresses the cell against the seal around the outer edge of the cell which rests on the shelf milled into the alumina cell holder. The HastX weight plates are held in alignment outside of the furnace by the upper portion of the threaded rods which extend upward for this purpose.

The compressive load can also be applied by means of springs. Spring loading is more compact and easier to implement than adding weights. Spring loading was used for the MSRI SOC tests, due to the requirement of incremental loading during the gasket curing process. Weights were used in the St. Gobain and SOFCPower SOC tests.

A photograph of the test stand installed in the furnace base for testing St. Gobain SOCs is shown in Fig. 3. Note that the upper part of the alumina load transfer tube is located outside of the furnace.

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