Introduction to Solid Oxide Fuel Cell Button Cell Testing

US Fuel Cell Council’s Solid Oxide Fuel Cell Focus Group

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INTRODUCTION

Solid oxide fuel cells are being tested by many organizations on different scales. At the research and development stage, so-called button cells are used to optimize materials and fabrication processes. Typically, these are circular cells of one to three centimeter diameter held in tubular fixtures. At the next level, single cells representing the anticipated stack geometry are evaluated. In planar designs, single cells will have sizes of 10 to 500 square centimeters. Here the purpose is to evaluate design alternatives. Next, stacks of cells are tested to address manifolding and contact issues and eventually the whole system is run.

Different organizations follow different practices in testing cells and reporting data. To help communication, the Fuel Cell Council is establishing a series of recommendations for conducting tests at the various levels. This document defines recommended practices for button cell testing.

PREAMBLE

Testing of fuel cells involves the use of hydrogen and other flammable or toxic gases. Elevated temperatures and electrical hazards are present. Following proper safety rules is important.

Similarly, to assure accuracy of the measurements, standard quality assurance procedures need to be followed.

PURPOSE

The primary purpose of button cell testing is to provide a method of evaluating the effects of changes in cell component materials (anode, cathode, electrolyte, interconnect, contact paste, interconnect coatings, etc) and fabrication processes on cell performance. Button cells are the test instrument for improving materials or processes in solid oxide fuel cells. Novel compositions or processing/microstructures may, or may not, lead to improvements in cell performance and need to be characterized and compared to the state of the art components. Either symmetric configurations of anode/electrolyte/anode or cathode/electrolyte/cathode or actual cells can be used.

Evaluation and characterization is done by determining (1) polarization curves and (2) impedance spectra. Both are measures of the material properties, and by reporting the polarization curves and/or impedance spectra for a given type of cell, conclusions are reached about the quality of the material or about the methods used to process it. However, the polarization curves and impedance spectra are also influenced by sometimes hard to recognize experimental variables, as for example, unsuitable current collection materials, minor leaks in seals or electrolytes, pressure differentials between the two gas streams, and mechanical forces on the electrodes. While it is always desirable to obtain accurate results, many experimenters will use button cell testing as a relative rather than absolute test method. By defining a reference polarization curve or impedance spectrum for well defined materials and operating conditions, the changes relative to the reference condition are determined and interpreted.

APPARATUS

Most button cell tests are done in a tube furnace, where the anode/electrolyte/cathode discs is held concentrically between two tubes of either glass or ceramic. The tubes must be sealed hermetically against the disc. Seals are made with gold foil or rings. To maintain a good seal, one of the tubes is stationary, and the other is spring loaded to provide a moderate amount of pressure. Reducing and oxidizing gases are supplied to either side of the cell by much thinner inner tubes that end a few millimeters above the respective electrodes. A typical set-up is shown in Fig.1
Button cells are normally tested at a constant temperature. Care must be taken to use a furnace with uniform temperature profile long enough for the reactive gases to reach the prescribed operating temperature. Since button cells do not have a defined flow-field, fuel and oxidant utilizations must be less than 5% by default, and gas flow-rates need to be relatively high.

The fuel and air streams are typically humidified to about 3% water. This is done by passing the fuel and air streams through a water bubbler at 25 C, making sure that the gas lines between bubbler and test apparatus are hot enough to prevent condensation.

One of the greatest challenges in button cell testing is to eliminate leaks, either in the electrolyte or seals. Leaks manifest themselves in decreased cell potentials. To determine whether a cell is well enough sealed for characterization, the open circuit potential is measured and compared to the calculated Nernst potential for the respective gas compositions and temperatures. Values slightly below theoretical (20-30 mV) will not invalidate the test but should be identified.
Note that the choice of current collector material can impact measured cell performance. Platinum is commonly used in the form of a wire mesh, which is welded to a platinum lead wire. However, it has been reported that Pt volatilization and subsequent deposition at the cathode/electrolyte interface can significantly affect the cell performance. Au has lower volatility than Pt and may therefore be a better choice for an “inert” current collector. Ag is a poor choice as an inert current collector due to its very high volatility at typical SOFC operating temperatures, so it should only be used if it is being considered as a candidate material in the cell/stack system under investigation. The wire mesh current collectors are either pressed by mechanical forces or cemented with platinum or gold ink to the electrodes. Care must be taken that the ohmic resistance of the wire mesh between the lead wire connection and the farthest point of the electrode is small; no more than about 5% of the impedance of the cell as measured by the left intercept of the semicircle.

Note also, that the lead wires need to be as heavy and short as possible, when impedance spectra are being obtained, because they will contribute to the measured ohmic resistance and sometimes to inductive effects. 0.5-1 mm wire diameter is recommended. To mitigate these effects, 4-wire set ups are preferred. Further, at least the current collector wires and ideally the cell assembly need to be shielded when impedance spectra are obtained.

To summarize, the following set-up is recommended:

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cell dimension</td>
<td>2-4 cm²</td>
</tr>
<tr>
<td>Tube material</td>
<td>alumina, zirconia or quartz</td>
</tr>
<tr>
<td>Seal material</td>
<td>gold</td>
</tr>
<tr>
<td>Current collectors</td>
<td>gold mesh and gold or platinum leads</td>
</tr>
<tr>
<td>Contact</td>
<td>mechanical load on current collector (300-500 g/cm²) or a contact paste</td>
</tr>
<tr>
<td>Fuel utilization</td>
<td>less than 5%</td>
</tr>
<tr>
<td>Air utilization</td>
<td>less than 5%</td>
</tr>
<tr>
<td>Humidification</td>
<td>3% H₂O</td>
</tr>
</tbody>
</table>

Anode, cathode and electrolyte compositions and thicknesses will depend on the nature of the experiment. As a default, the following compositions and dimensions can be used for the active components of an anode supported cell:

- **Anode**: Ni/8YSZ, typically 60wt% NiO, 40 wt% ZrO₂, 0.5-1.0mm thick, 30% porosity.
- **Cathode**: 50%LSM/50%YSZ near the electrolyte, 100% LSM in the bulk, or LSCF with a ceria interlayer, 0.05mm thick, 35% porosity.
- **Electrolyte**: 8 YSZ, 0.05 mm thick

Thinner or thicker components can off-course be used, but the thickness of the cathode, electrolyte or anode will affect the electrochemical properties and comparisons cannot be made unless the dimensions are constant.
TEST PROCEDURES

Conditioning

Newly fabricated button cells are usually not in a fully equilibrated state and need thermal and electrochemical conditioning before electrochemical measurements can be taken. Typically, cells are heated up under flowing air at a rate of one degree per minute to burn off any organic binders. When the operating temperature is reached, the air on the anode side is changed to an inert gas and then to a reducing gas that will convert the nickel oxide to nickel metal. Nitrogen or argon with 2-4% hydrogen is a good choice.

Cells with lanthanum manganite cathodes need to be operated under electrical load at a current density that is close to the design point for a few hours at the designated temperature and operating conditions. Since the electrochemical performance of the lanthanum manganite improves when current is passed through it, stable performance needs to be established. A cell potential that is constant to within 10 millivolts for an hour is considered stable.

Polarization Curves

The single most important measurement with button cells is to determine the voltage/current density characteristic or polarization curve. To determine the polarizations curve, a potentiostat is the preferred instrument. Either the current can be the independent variable and the cell potential is measured or vice versa. Multiple identical tests with steady state operation between tests are recommended to assure reproducibility. When reporting a polarization curve for a new material, it is useful to report also a polarization curve for a well established material under identical test conditions.

To obtain the actual polarization curve, the cell current is changed from about zero to an upper current limit at a rate of 20 mA/sec with 5 data points per second, and recording the cell potential. The upper limit depends on the cell. Typically, it is the current where the cell potential is at 0.3-0.5 volts. Similarly, in a potentiodynamic mode, the current is held at a given value, and the potential is changed at about 10mV/sec.

Most polarization curves are slightly curved at low and high current densities. The “area specific resistance”, or ASR, is the inverse slope of the straight portion of the curve, which can be calculated by regression analysis.

Impedance Spectra

Impedance spectroscopy is a useful tool to differentiate between various sources of ohmic or non-ohmic resistances.

Impedance spectra can be obtained with or without a reference electrode, but in either case, the impedance spectrometer must be integrated into the electric circuit in such a way that the spectrum can be obtained in an operating fuel cell without interrupting the current flow. In symmetric cells, the impedance spectrum is obtained at open circuit potential or at a small bias potential.

When reference electrodes are used, positioning them is of critical importance to avoid contributions from the electric field on the impedances. If the reference is to close to either the working or counter electrode, the measured impedances will be affected by gradients in the field. A general rule is to have a spacing of at least three times the thickness of the cell between the reference and working electrode. There is no universally accepted optimum location for the reference electrode, but two good options are shown below:
Fig. 2  Placement of Reference Electrodes

Option 1 shows the reference electrode, RE, as a half moon shape on the side of the anode or cathode. In this case it is important that the counter electrode is shaped in the same way as the working electrode and the spacing between the reference electrode and the working electrode, WE, must be at least three times larger than the thickness of the cell. The working and counter electrode must be strictly orthogonal.

Option 2 shows the reference electrode in the center of an annular working electrode. This position gives very good results but is harder to make, especially with thin electrolytes. Again, the spacing between the reference and working electrode must be at least three times the thickness of the electrolyte.

Either of these two options can be used for symmetric cells where the working and counter electrode are made of the same material. In this case, the measured quantities are divided by two.
INTERPRETATION AND REPORTING OF RESULTS

Polarization curves are usually reported as plots of cell potential versus current density in milliamps per square centimeter, where the voltage scale is on the left and the power density can be shown on the right. The linear part in the middle of the curve is the area specific resistance. An ASR of 0.5 ohm cm or lower represents good cell performance.

When reporting polarization curves, the nature and thickness of the electrolyte needs to be identified together with temperature and gas compositions. As mentioned, it would also be useful but not always feasible to show the polarization curve for a standard nickel anode or LSM cathode together with the new material or process. Also, reporting the open circuit potential is very advisable.

Impedance spectroscopy can differentiate between charge transfer resistances at the electrodes, any diffusion related impedances, and ohmic resistances in the electrolyte or electrodes. When no reference electrode is used, the left intercept of the semicircle with the real impedance axis is usually interpreted as ohmic resistance of the cell, and the diameter of the semicircle as the polarization resistance as shown in Fig. 3. With reference electrodes, the latter can be further differentiated as described in pertinent textbooks.

Fig. 3 Typical Impedance Spectrum of a SOFC without reference Electrode