SINGLE SOLID OXIDE FUEL CELL TESTING USING SILVER PASTE FOR SEALING AND CURRENT COLLECTION

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ABSTRACT

A new method for sealing and joining solid oxide fuel cells (SOFCs) for single-cell testing is described in this paper. This technique uses silver paste for sealing and joining. YSZ electrolyte SOFCs assembled by this technique showed OCVs close to theoretical values, indicating the sealant is very effective. Experiments have shown SOFCs assembled by this technique operated stably at 800°C for over 10 hours and at 700°C for over 90 hours.

INTRODUCTION

Solid oxide fuel cells (SOFCs) have become more attractive because of their promising application in different areas. When researching and selecting SOFC component materials, it is important to test them in actual SOFCs, typically single cells. However, even to get the simplest experimental SOFC working, several key aspects must be considered and done. These aspects include making a core cell (electrolyte with cathode and anode), sealing the cell, applying lead wires for current collection, feeding fuel gas and oxidant at appropriate flow rates, and heating the cell. There have been many discussions about how to make a core cell (1), but much less work has been reported on the testing techniques.

Sealing is very important for any fuel cell because the cell voltage is obtained through the separation of the fuel gas and oxidant. It is more challenging in SOFCs because they are operated at high temperatures, 600° to 1000°C, where the choice of sealant materials is limited. Additionally, sealant failure at high temperatures can lead to burning or even explosion of the resulting fuel-oxidant mixture. There are a few sealing techniques commonly used for SOFC single-cell tests. The earliest developed and most commonly used sealing technique might be glass sealing (2,3). It is a technique that can yield strong gas-tight seals and that places no strict requirement on the shape of the SOFC. However, the glass sealing material generally has low thermal expansion coefficients compared with SOFC components, leading to substantial stresses on the SOFC during temperature changes, potentially leading to fracture. Also, many glasses contain
components that may diffuse into the nearby SOFC active components and deleteriously affect performance. Finally, processing the glass seals is time-consuming.

Another commonly used sealing technique is mechanical compression using mica as the sealant material (4). This technique has the advantage of providing good sealing at temperatures as high as 800°C that can survive several thermal cycles. It is also very convenient to use, with no annealing required. But the disadvantage is that the more complex cell test fixture requires applying a compressive force, and the SOFC must have sufficient strength and flatness to avoid fracture during testing.

Generally, when anode or cathode material is used as film other than bulk, the conduction along the film (sheet conductivity) is limited. In many cases it is not enough to get good performance from the SOFC cell. Therefore, a platinum or silver mesh was pressed onto the electrode to act as current collector. The mesh is very conductive but relatively rigid, and it would detach from the electrode during drying or annealing. Silver paste could be used to paint the silver grids on the electrode for current collection.

In this paper, we describe a SOFC assembly technique using silver as sealant and current collector. As thin electrolyte SOFCs have developed, SOFCs operated at temperatures lower than 800°C with reasonable power density are available (5,6). The melting point of silver is 961.78°C (7), so it can be used as sealant for these SOFCs. The lower melting point of silver results in a lower sintering temperature, so it is easy to sinter silver to full density. An additional advantage of silver is its good electrical conductivity, allowing anode and cathode currents to be collected through the silver sealing material. Some details, including the assembly design, lead wire connection, fuel gas feeding, and testing of this technique are now discussed.

**SOFC ASSEMBLY AND EXPERIMENT**

**Core SOFC Preparation**

We took an electrolyte-supported SOFC as an example. The core of the SOFC is a flat electrolyte-supported single-cell pellet. The electrolyte was made by pressing purchased 8 mol% yttrium stabilized zirconia (YSZ) (Beijing Institute of Construction Materials) into pellets with diameter of 13 mm and thickness of about 1 mm and sintering them at 1400°C for 2 hr. Ni-YSZ (7:3 wt) was applied on one side of the electrolyte as anode and LaSrMnO-YSZ (7:3 wt) on the other side as cathode. The pellet was cosintered at 1200°C for 1 hr. In this case, both cathode and anode materials were used as films. Silver grids were painted or screen printed on the surface of both electrodes.

**Assembling the SOFC**

The assembling of the SOFC is illustrated in Figure 1. We took an alumina tube (1) with the outside diameter close to the diameter of the SOFC pellet. We fixed the tube straight up and applied some silver paste onto the ring area of the upper open end of the tube (2). Then we carefully put the SOFC pellet with the anode side (3) down right on top of the tube and pressed the pellet down gently. The pellet was attached to the tube while some silver paste was pushed to the edge of the joint, forming the initial seal. We
then moved the set to an oven and dried the silver paste at 150°C for 2 hr. Note that the alumina tube needs to be kept straight up or the pellet on top of it will slip off during drying in the oven. After drying, the pellet was bonded firmly onto the tube. It was removed from the oven and cooled to room temperature, then more silver paste was applied to the joint edge of the pellet and the tube. It is good to apply silver paste to cover all the edges of the pellet (4). This is particularly necessary for anode-supported SOFCs. Then, the set was put back into the oven to dry at 150°C for 1 hr. After the set cooled down, we bundled two silver wires (5) (6) on the alumina tube close to the joint (7). The wires were insulated by thin ceramic tubes (not shown in the figure). Together with the anode lead wires in thin ceramic tubes, the other two silver wires (8) (9) in thin ceramic tubes (10) were bounded and fixed to the alumina tubes as cathode leading wires by at least two bundles of silver wire, as shown in Figure 2 (1). The thin insulator ceramic tubes (10) for cathode lead wires exceeded the top of the pellet a little to avoid a short with the anode. Some silver paste was applied to connect the anode leading wire with the sealant (Figure 1) (7). Silver paste was also used to make the contact for cathode lead wires on the surface of the cathode (11). The set was put back into the oven to dry the silver paste for another hour.

![Figure 1. Illustration of SOFC assembled by silver sealant. 1 - alumina tube; 2 - Ag paste for attaching; 3 - anode; 4 - Ag paste for sealing; 5 - lead wire A1; 6 - lead wire A2; 7 - Ag paste for lead wire contact; 8 - leading wire C1; 9 - leading wire C2; 10 - insulated ceramic tube; 11 - Ag paste for lead wire contact.](image1)

![Figure 2. Practical setup of SOFC for single-cell testing; 1 - SOFC pellet; 2 - anode lead wire; 3 - alumina tube; 4 - insulate tube; 5 - cathode lead wire; 6 - bounding wire; 7 - fuel gas feeding tube.](image2)
In many cases, it is desirable to control the composition and the flow rate of oxidant gas to the cell. In this case, a second tube is sealed to the cathode side of the cell and the cathode connection can also be made through the silver seal.

**SOFC Single-Cell Testing**

A practical setup of SOFCs for single-cell testing based on the SOFC assembly shown in Figure 1 is shown in Figure 2. A thinner ceramic tube for fuel gas feeding (2) is put into the alumina tube with the open end almost touching the anode of the SOFC pellet. When the SOFC is operated, fuel gas is fed into the cell by the thin tube and the reactant gas and the residential fuel gas are pushed out through the space between the alumina tube and the gas feeding tube. This set is put into a tubular heating furnace with the cell pellet part in the middle position. A voltmeter, an amp meter, and a changeable load resistance are connected. To start the SOFC test, the fuel flow begins and the ramp rate of the furnace is set.

**DISCUSSION**

**Feasibility of the Technique**

It only takes about 3 hr to complete the assembly for the cell test. No high temperature annealing is needed. Experiments showed no cracks were caused by the stress applied by the silver sealant for YSZ electrolyte SOFC by this technique. (An advantage is that silver is quite malleable at high temperatures and can accommodate some mismatch between the SOFC and the tube.) Gold has been used as a solder to seal SOFCs, but we used silver sealant in the form of paste. Generally, silver paste is a mixture of fine silver powder, binder, and solvent. The paste/slurry form makes it convenient to apply to the required places. After it is dried at a lower temperature, the binder can hold all the silver particles together tightly and join different parts (i.e., the cell pellet and alumina tube) together. When the temperature increases, as when a SOFC is tested, the binder evaporates while the silver is sintered to full density. Besides this feasibility, silver sealant at the joint (Figure 1) (7) can also lead the anode connecting wires from outside easily because silver itself is conductive. This is significant for glass sealing because the current produced inside the tube must be led out from inside. To make a good contact from inside and lead a wire out is very difficult.

From Figure 2, it can be seen that in this SOFC assembly technique fuel gas was let in through a ceramic tube without any additional sealing. It is well known that a possible mixing of fuel gas and oxidant in an enclosed space is very dangerous. Feeding fuel gas without a seal lets the exhausted gas be extruded out instantly without any danger of mixing with oxygen within the SOFC.

**Reliability**

Figure 3 shows the OCV for an electrolyte-supported SOFC by this technique. It is close to theoretical values, indicating the gas leakage is very small and silver paste can be used as a sealant for SOFC. This has also been demonstrated by many experiments and the rate of success is almost 100%.
Figure 3. OCV obtained from a SOFC assembled by silver sealant. The theoretical OCV for a H₂-O₂ fuel cell is also shown for comparison.

Experiments showed the silver sealant can stand temperatures as high as 800°C for more than 10 hours. At lower temperatures, it can last even longer. Liu and Barnett (8) applied this technique in their Ni-YSZ anode-supported SOFC and obtained an OCV 30 mV less than the theoretical expectation. In one case, the SOFC was operated at 700°C for about 90 hr and voltage did not decrease significantly. As mentioned before, silver can be sintered to dense at lower temperatures, so there is no annealing before the cell test. As the temperature increases during the test, the silver is sintered to dense in situ. And it becomes even denser at higher temperatures. There might be some evaporation of silver at high temperatures that would limit its operation life; however, most SOFC experiments do not operate for that long. This technique works for most cases.

CONCLUSIONS

Silver paste is a very convenient material for sealing and joining SOFCs for testing. The sealing effect of silver is very good. Combined with the seal-free way of feeding fuel gas, the presented SOFC assembling technique is very practical in SOFC research.

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