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Short communication

Silver wire seal design for planar solid oxide fuel cell stack

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Abstract

A method for sealing solid oxide fuel cells with silver wire gaskets was developed and tested. The 1.6 mm diameter gaskets were fitted into machined channels, 1.5 mm deep in the interconnect plates. The channels were machined into 430 stainless steel plate along the edge of both surfaces and around alternate gas inlets. The interconnect plates were 6.35 mm thick, 152.4 mm long and 76.2 mm wide. A one-cell-stack was assembled for pressure testing with a stainless steel sheet in place of the ceramic membrane. The gas connections were brazed to the stack with a nickel–chromium brazing alloy. The apparatus was bolted together and tested for gas leakage at 137.8 kPa between room temperature and 500 °C. At room temperature, the measured leak rate was 5.14 kPa min^{-1}. With the stack heated to 500 °C, the leak rate decreased to 75.8 Pa min^{-1}.

Keywords: Solid oxide fuel cell; Compression sealing; Pressure test; Interconnect design; Silver gasket

1. Introduction

The solid oxide fuel cell (SOFC) is an alternative to traditional electric power generating devices such as gas turbines, diesel generators and thermoelectric converters [8]. One of the major drawbacks of the SOFC is the need for gas-tight sealing around the edge of the cell. In case of a significant gas leak, the resulting combustion would create a hot spot leading to failure by local reaction/melting. Sealing the stack remains an unsolved problem with SOFCs. Several sealing techniques have been developed over the years, although standards have yet to be set. From a materials standpoint, the seal must satisfy several criteria. First, the seal must have long-term stability. Second, the seal must not cause degradation of the materials with which it is in contact (e.g. stabilized zirconia, interconnect and electrodes) at the elevated temperatures and harsh environments typical of SOFCs during operation. Third, the seal has to survive many thermal cycles during routine operation. The issue of thermal cycling is critical for materials which have different coefficients of thermal expansion, especially for SOFCs with metallic interconnects. This is one of the most challenging components of the seal development. Finally, the seal should be cost effective.

Several types of materials have been used as sealants in SOFCs. The three most popular types are glass, high temperature cements and compression metal seals. Most SOFC development has focused on glass or glass–ceramic seals. The disadvantage of glass is its susceptibility to thermal shock. High temperature cements are porous and lack adequate wetting of the metal interconnect and ceramic membrane surfaces.

Currently, the most popular type of sealants used are compression metal seals. These types of seals are radically different from those mentioned above since bonding is not required. Sufficient compressive load is applied to deform the metal gaskets and therefore prevent gas leakage. The combination of elastic and plastic deformation of the metal seals leads to an increased tolerance of the SOFC stack of thermal expansion mismatch as well as thermal cycling and vibrations [2]. Since the ceramic cell and the metallic interconnect are not rigidly bonded to each other (as in the case of glass–ceramic seals), residual stresses do not develop or are substantially reduced. Research in the area of compressive metal seals for SOFCs is still relatively new and very little data is available.

It is possible to purchase metallic O-ring seals commercially. These vary in shape, material and cost and certain types are already being used by fuel cell manufacturers. In

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the present paper, a new compression sealing technique using silver wire gaskets is investigated. The test apparatus used for examining the integrity of the gaskets is described, and measured leak rates are reported.

2. Experimental

2.1. Design of apparatus

In the course of developing this SOFC stack, several design and fabrication processes were investigated. Following a thorough materials analysis, 430 stainless steel was identified as the optimum material for the interconnect on the basis of strength, stiffness, conductivity, oxidation resistance and cost. The interconnect plates were cut to the design size of 76.2 mm × 152.4 mm × 6.35 mm. Fig. 1 shows top and bottom views of the interconnect plate.

Gas flow channels and gasket channels (1 and 1.5 mm deep, respectively) were machined from the 430 plates. Tube fittings of 316 stainless steel were brazed to the stack gas inlets and outlets using a chromium–nickel brazing alloy. The gaskets were pure silver wire (1.6 mm diameter). An attempt was made to use sterling silver (7.5 wt.% Cu) wire gaskets, but due to the solid solution strengthening, the alloy was not sufficiently deformable to form a seal. The stack was assembled and bolted together with sufficient torque to ensure adequate sealing by the silver gaskets.

The design requires the fuel cell membrane to be fabricated with six holes, in line with the interconnect gas inlets. Sealing gaskets are located between the fuel cell membrane and the interconnect plate. Both gases enter from one end of the stack in a co-flow fashion, the oxidant from the outer holes and the fuel from the inner hole, as shown in Fig. 2. The oxidant gas flows through the inlet tube, across the stack between the interconnect and cathode and then exits through a tube at the opposite end. The fuel gas follows a similar path across the stack on the anode side. To better visualize the gas path inside of the stack, slices at different locations are shown in Figs. 3–5. The square inset in Figs. 4 and 5 are enlargements of the stack gas inlets. These views show the internal manifolding arrangement as well as the location

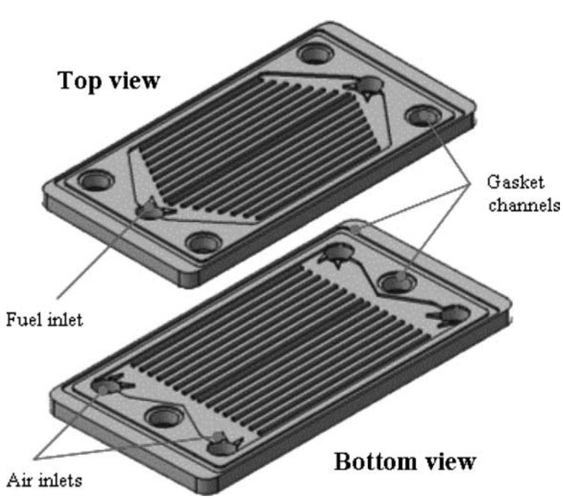

Fig. 1. Interconnect plates.

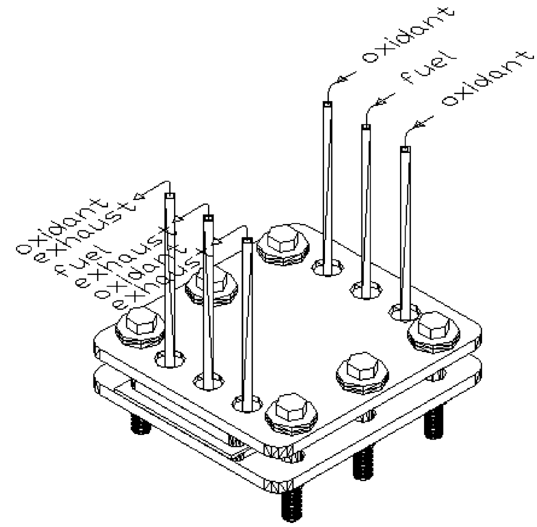

Fig. 2. Picture of SOFC stack showing gas inlets and outlets.

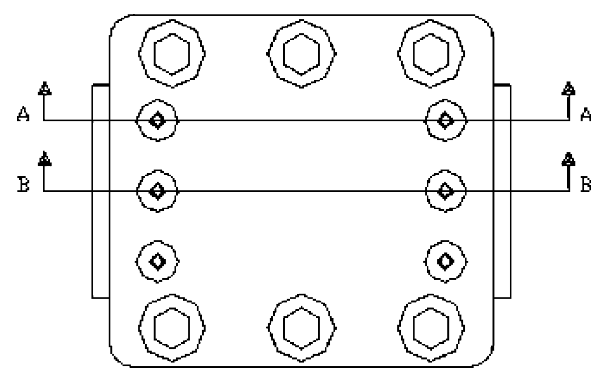

Fig. 3. Top view of stack showing slice direction.

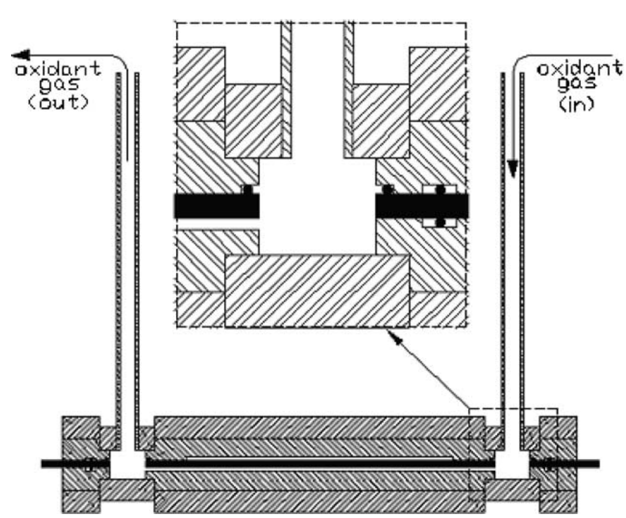

Fig. 4. Slice A–A.

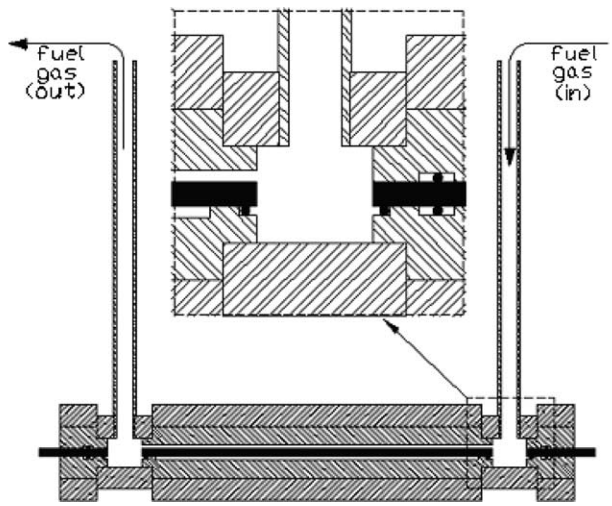

Fig. 5. Slice B–B.

of the silver gasket O-rings (shown as small black circles), and the fuel cell membrane (shown as a black layer). These figures illustrate the design of the seals and the distribution and flow of gas in the cell.

In addition, a test system was constructed (Fig. 6) which had the ability to measure the pressure loss with time inside the stack at various temperatures.

To evaluate the integrity of the seals, the system was pressurized and monitored for leakage. Pressure tests were conducted with air, and cycling was performed from room temperature to 500 °C. The leak rate tests were done with a steel plate in place of the ceramic fuel cell membrane.

2.2. Testing procedure

The procedure for all the pressure tests can be described as follows:

(1) The stack was heated by flowing externally heated air through it.
(2) When the stack temperature reached the desired test temperature, the exit gate valve was closed. This corresponds to the start time (point A, Fig. 7) for the internal pressure of the system.
(3) When the pressure reached 137.8 kPa, the entrance gate valve was closed, isolating the system. At this point

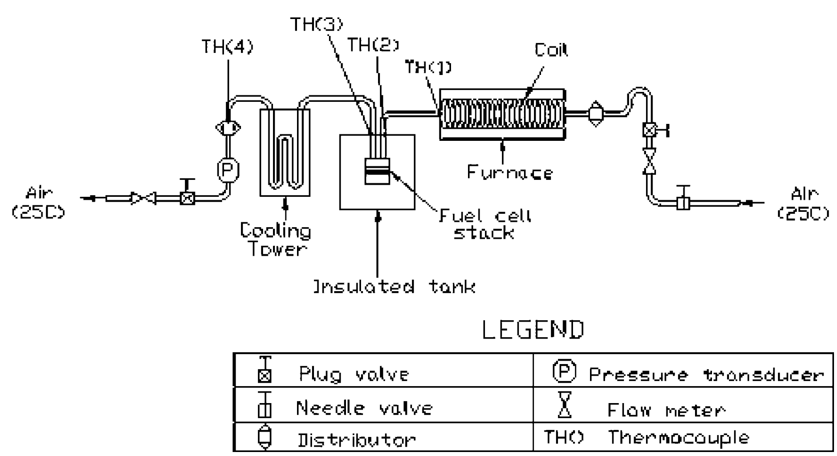

Fig. 6. Testing apparatus to measure leakage in SOFC stack (side view).

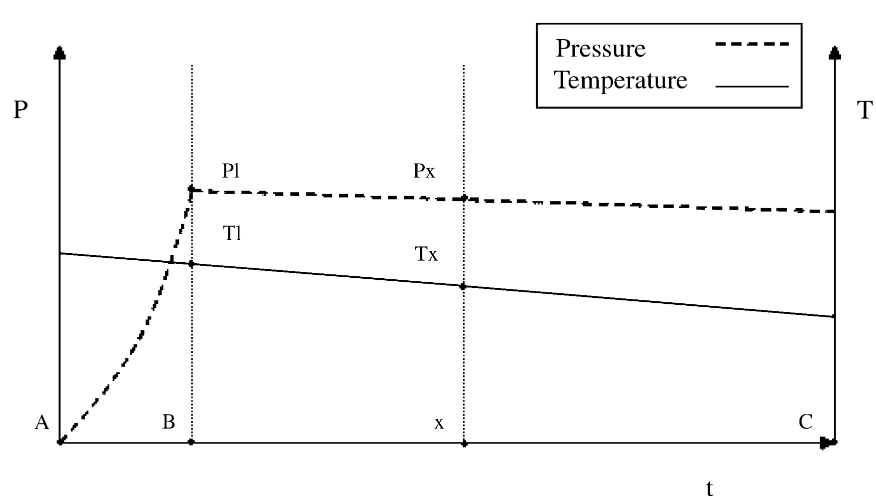

Fig. 7. Schematic representation of pressure and temperature change vs. time in the stack.

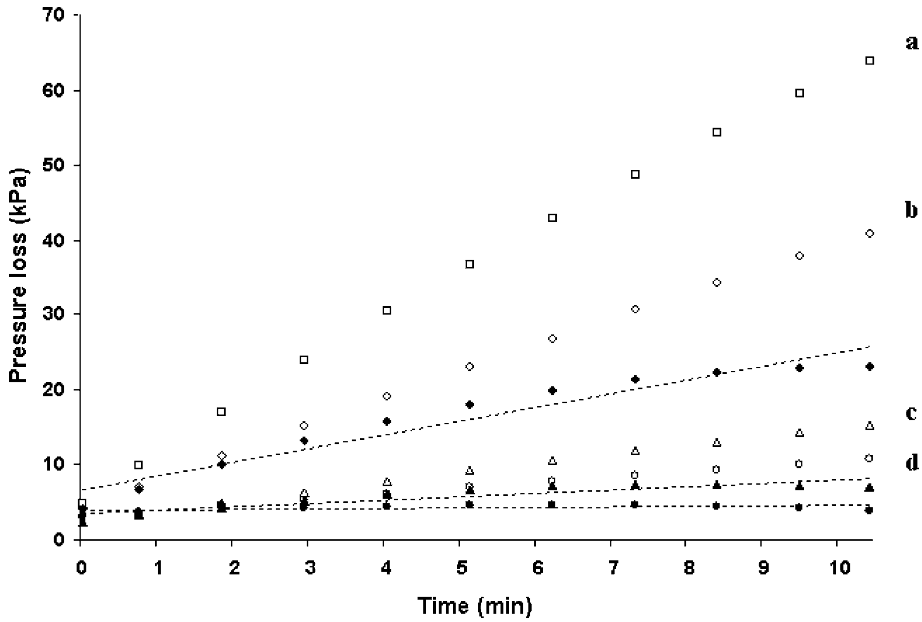

Fig. 8. Lines a, b, c and d represent the overall pressure loss (as measured by the pressure transducer) at 25, 200, 400 and 500 °C, respectively. The shaded data points characterize the corresponding pressure loss after the temperature has been compensated. Lines were drawn for each of these curves to give a linear approximation of the pressure loss with time.

(point B, Fig. 7), the pressure was recorded in the system at 5 s intervals for 10 min (point C, Fig. 7).

3. Results and discussion

Because no auxiliary heat was provided to maintain constant temperature at the stack location, there was a significant drop in temperature during testing with an associated drop in the pressure in the system. Thus, there are two separate causes for pressure loss in the system, the first is gas leakage and the second is temperature drop. In order to plot the leak rate, the effect of temperature change on pressure was compensated. Fig. 7 represents the output reading of a calibration pressure test. Both temperature and pressure decrease approximately linearly with time.

The pressure loss curves for the stack at different temperatures are shown in Fig. 8. The slope of the lines in Fig. 8 provides the average rate of pressure loss. At 25 °C, when the temperature effect is eliminated, the rate of pressure loss is 5.14 kPa min^{-1}. At 500 °C, the rate of pressure loss is 75.8 Pa min^{-1}. The obvious trend demonstrated is that the leakage rate decreases with increasing temperature. This effect is likely due to the larger CTE of silver and its decreasing strength and modulus with temperature.

4. Conclusions

A single-cell planar SOFC stack was fabricated and used to test the new compression sealing concept utilizing pure silver wire gaskets. The leak rates were tested with a steel plate in place of the ceramic fuel cell membrane at 25, 200, 400 and 500 °C. The results clearly imply a trend toward better sealing as temperature increased. SOFCs normally operate at elevated temperatures (800 °C), and therefore, the most relevant results were at 500 °C. The approximate leak rate calculated at this temperature was 75.8 Pa min^{-1}. Only one similar study was found in the literature [2]. The lowest leak rate measured for metallic seals at room temperature was 3.59 kPa min^{-1}.

5. Recommendations

The results of this experiment show that significantly low leak rates were obtained using the silver wire design. However, it is important to note that several problems were encountered through the course of this study. These problems are listed below.

5.1. Manufacturing of flat interconnect plates

Flat plates are required to ensure that the ceramic membrane does not crack upon loading. Brazing the gas fittings after the plates were machined caused slight warpage to the interconnect.

5.2. Sealing of gas fittings

One of the greatest difficulties encountered in assembling the stack components was to join the tube fittings to the interconnect plate. Difficulties occurred while attempting to make an airtight seal. Initially, threads were tapped into the interconnect plates to accept Swagelok tube fittings which was known to be a source of leakage. The idea was to fill the volume between the fitting and the thread with a high temperature sealant, effectively preventing gases from escaping. The filler materials utilized were silver paste and glass. However, the silver paste was overly porous and the glass cracked during thermal cycling causing some leakage at the joint.

Brazing the joint was considered as an option, however, because the braze material is exposed to severe high temperature oxidation conditions, only a limited range of metals were suitable for the application. Several commercial brazes were investigated and it was found that silver braze was ideal in terms of cost and oxidation resistance. Silver braze is also recommended to be used to bond stainless steel. However, the majority of commercial brazing alloys containing silver have a lower maximum operating temperature than what is required for the fuel cell stack. The only suitable commercially available brazing alloys capable of exceeding the maximum stack operating temperature contain either gold or platinum. These materials were not considered on account of cost.

Another alternative was to weld the fittings to the interconnect plates. The method was attempted but proved to be unsuccessful due to excessive grain coarsening which caused microcracks to form in the weld (a problem commonly encountered when welding ferritic stainless steel). Therefore, even after welding, the joint region remained a major source of gas leakage. This problem was eventually solved by applying a fine particle chromium–nickel braze to the stack and heating it in a reducing furnace. The attempt was successful, with the alloy effectively filling all the cracks and creating a gas-tight seal.

5.3. Uniform load distribution

The bolts which provided compression to the stack were not symmetrically positioned. This caused the silver gasket to deform non-uniformly, and higher torques were required to ensure a gas-tight seal.

In order to prevent or reduce these problems in future stack design and fabrication, a few recommendations are proposed below:

- Change the cell to a circular shape instead of rectangular. The advantages of this are:
  - Ease of manufacturing (the interconnect plates can then be machined on a lathe rather than a milling machine, which is more time consuming and costly).
  - Better sealing (more bolts can be added around the perimeter of the plates to supply a more symmetrical load distribution).
  - Less risk of warpage of both interconnect and ceramic.
- Machine the interconnect plates once all the metal joining operations have taken place to ensure flatness of the contact surfaces.
- Ensure uniform cross-section of the wire gaskets. The present method of welding the ends of a wire together leaves a ridge at the joint which acts as a source of leakage.

Acknowledgements

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