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ScienceDirect
Journal of Power Sources 171 (2007) 96–100
JOURNAL OF POWER SOURCES
www.elsevier.com/locate/jpowsour

Short communication

Cycling of three solid oxide fuel cell types

Waldemar Bujalski, Chinnan M. Dikwal, Kevin Kendall *

Department of Chemical Engineering, University of Birmingham B15 2TT, UK

Received 23 October 2006; received in revised form 9 January 2007; accepted 13 January 2007
Available online 20 January 2007

Abstract

One of the key problems of SOFCs is their slow start-up and cycling performance which is due to the thermal shock problems of zirconia electrolyte and its associated electrode and interconnect materials. Typical start-up times range from 2 to 6 h. Faster cycles can cause degradation in performance and in material integrity. The purpose of this paper is to study the transient performance of SOFCs under various (e.g. thermal and/or current load) cycling conditions, typifying start-up and shut-down as well as variable working conditions of the systems, in order to understand the degradation mechanisms. Three types of SOFC have been compared; the planar stack with metal interconnects represented by the Forschungszentrum Jülich (FZJ) configuration; the Rolls Royce Fuel Cell Systems Ltd. (RRFCS) integrated planar tube; and the Adelan pure tube. The objective was to cycle in temperature from ambient to the operating condition several times to check if degradation was occurring. To obtain thermally shock resistant systems, cell dimensions had to be reduced to the millimeter scale.

Keywords: Cycling SOFC; Thermal cycles; Planar; FZJ; Integrated planar; RRFCS; Tubular; Adelan

1. Introduction

Cycling of solid oxide fuel cells (SOFCs) is a major issue because degradation can increase rapidly with the number of temperature cycles and especially with redox cycles where the nickel anode is repeatedly oxidised and reduced. These phenomena are strongly dependent on the cell and stack architecture and so it is important to find design criteria for minimizing cycling degradation effects in different geometries. This study compares three types of SOFC design: planar, integrated planar (also sometimes referred to as plane tubular) and pure tubular in order to illustrate differences in cycling performance. The damaging effect of cycling on SOFCs has been described before to some extent but requires much further elucidation [1–7].

The main objective was to set up three SOFC geometries in thermal (and/or current load) cycling tests so that comparisons could be drawn between them. The performance of each geometry could then be measured over a number of cycles and compared to a theory based on the temperature gradients in the zirconia elements which could cause fracture or delamination according to a critical size criterion. Thus, the main factor dictating cycling failure was shown to be the dimension of the thermally stressed ceramic cell.

The work reported here was largely carried out in the real-SOFC project, a European Integrated Project aimed at solving the persisting generic problems of ageing with planar solid oxide fuel cells in a concerted action of the European fuel cell industry and research institutions. This includes gaining full understanding of degradation processes, then finding solutions to these problems to reduce ageing effects.

2. Experimental

A two-cell planar SOFC stack containing a metal interconnect plate was donated by FZJ (Fig. 1(a)) [8–11]. The two anode supported cells are placed inside metal frames. The metal frames, interconnect and end plates are sealed with a glass-ceramic. A mechanical load of 50 kg provides pressure for sealing. Hydrogen or purge gas was fed to the fuel side and air to the oxidant inlet at 2.88 L min^{-1} (3% humidity) and 6.84 L min^{-1}, respectively. The assembly was heated to 800 °C at 2 °C min^{-1} and also cooled at a similar rate as described in the test protocol below.

Single integrated planar tube arrays of cells were contributed by RRFCS Ltd. [12,13]. Each tube had 10 cells on each side and

* Corresponding author. Tel.: +44 1214142739; fax: +44 1214145377.
E-mail address: k.kendall@bham.ac.uk (K. Kendall).

0378-7753/$ – see front matter
doi:10.1016/j.jpowsour.2007.01.029

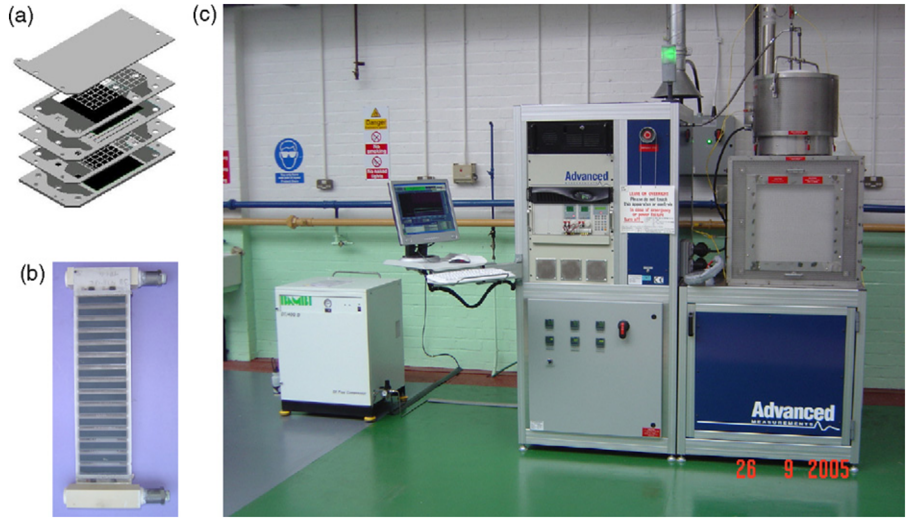

Fig. 1. (a) FZJ planar two cells stack; (b) RRFCS integrated planar tube (15 dual-cells version); (c) Advanced Measurement Inc. test station.

comprised a support structure onto which the anode, electrolyte, cathode, interconnect and sealing layers had been printed by colloidal methods (Fig. 1(b)). The tube was fed with fuel on the inside by attaching manifolds and pipes to each end. Air was flowed across the outside of the tube by feeding air into a box that surrounded the tube, to imitate the flow across a tube stack. The box was located in the furnace of an Advanced Measurement Inc. test station shown in Fig. 1(c). The temperature of the furnace was ramped up at 1 °C min^{-1} to 900 °C and preheated air and fuel were then supplied to the tube to carry out the cycling tests. Air flow was 5 L min^{-1} and fuel was 1.5 L min^{-1} (3% humidity) in a typical run.

Each test was carried out according to the specific protocol defined by the Real-SOFC project. Reduction for the RRFCS plane tube was achieved by increasing the hydrogen (containing about 3% of water vapour) flow to the anodes while gradually dropping the nitrogen flow to zero (the Jülich stack was reduced at source and provided as such for our tests). After reduction was attained, the open circuit voltage of the cells was measured. Then current was drawn from the cells to perform current loading cycle. 0.1 A was drawn for 40 s, then another 0.1 A was drawn and the process repeated until the maximum current of 2.7 A was achieved. This current was held for 5 min and then the current was lowered in 0.1 A steps until open circuit. After recording the baseline *I*–*V* curve at this temperature the tube was subjected to pre-planned thermal cycles consisting of ramping the temperature at 2 °C min^{-1} to 950 °C (the maximum value used) and then going down to the next temperature level in the same 50 °C steps till reaching the minimum of 800 °C used in the test. At each temperature the *I*–*V* curves were recorded.

A similar procedure was applied to the Jülich stack. However, due to this stack's different design, the nominal test temperature was set at 800 °C and the current steps applied were much larger comparing to the RRFCS tube case described above (i.e. 4 A per step) and because of the equipment limitations at the time, the maximum attainable value was restricted to 16 A. For both systems, after dwelling at the operating temperature for a certain time, the furnace was then cooled down to about 300 °C using controlled cooling rates of 1–2 °C min^{-1} still using hydrogen (which was than replaced with a "safe gas" i.e. mixture of nitrogen and about 5% of hydrogen for further cooling to room temperature), and another heating cycle then commenced. The change in tube/stack performance after a number of current load and/or thermal cycles was observed.

Both the above systems were susceptible to thermal shock damage and so were restricted to low heating/cooling rates of 1–2 °C min^{-1}. However, the Adelan tubular cell is known to withstand temperature ramps of 4000 °C min^{-1} (conservative estimate on the basis of the system reaching the operating temperature of about 800 °C in less than 10 s) and so this was tested in the rapid heating system of Fig. 2. Butane was fed through a valve and a venturi to draw in some air which heated a catalyst mesh before entering the tubular cell to produce heat and power. The electrical output was observed to drive a fan and could be measured by attaching a voltmeter and ammeter to the cell. The tubular cells were also tested on pure hydrogen fuel (using 20 mL min^{-1} flow rate) at up to 200 °C min^{-1} temper-

Fig. 2. Adelan tubular SOFC and hand held demonstration device that powers a fan.