



Performance evaluation of a tubular direct carbon fuel cell operating in a packed bed of carbon



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ABSTRACT

The DCFC (direct carbon fuel cell) technology, based on the direct electrochemical oxidation of carbon, has the potential to double the electric efficiency and half the CO₂ emissions compared to conventional coal fired power plants. In order to assess the scalability of the technology in terms of fabrication and fuel feed system, and to elucidate the possible causes of the cell degradation, a tubular DCFC has been fabricated and operated in a pulverised carbon packed bed at around 800 °C. The cell was operated for a total period of 11 days with many thermal cycles. The electrochemical impedance spectroscopy was used to elucidate the possible causes of the cell degradation. Post-mortem analysis of the cell with SEM (scanning electron microscopy) and XRD (X-ray diffraction) confirmed structural stability of both air and fuel electrodes. A peak power density of 30 mW cm⁻² was obtained by direct contact of carbon to the fuel electrode with high purity He as the purge gas. The cell, at the end of operation was still found to produce 60% of the power relative to the power at the beginning of operation, and this study demonstrates the feasibility of continuous operation of the tubular fuel cell in a packed bed of carbon.

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1. Introduction

Today's coal fired power plants are inefficient (~30%) and produce around 1–1.2 kg CO₂ emissions per kW-h of electricity produced along with significant other pollutants such as particulate matter and NO_x/SO_x gas emissions. This leads to coal fired power stations being a major contributor to the pollutants in the atmosphere and greenhouse gas emissions. A range of alternative coal combustion technologies are currently being trailed including IGCC (Integrated Gasification Combined Cycle), ultra supercritical coal combustion, and oxy-fuel combustion. These technologies are capable of delivering a reduction in CO₂ emissions via efficiency gains, however, the efficiency gains are only incremental. Although there are worldwide efforts to find technologies for CO₂ capture and sequestration, this introduces even more inefficiency as the energy associated with separation, capture and storage of CO₂ from conventional power stations is high, leading to an increase in fuel use of up to 25%. Coal has accounted for 45% of the total energy related CO₂ emissions in 2011 [1]. There are around 860 billion tonnes of coal reserves worldwide and are estimated to last more than a century at current rate of consumption [2]. Therefore, more

efficient technologies need to be developed to reduce pollution and CO₂ emissions, and prolong the life of the existing coal reserves.

Fuel cells are well known to be one of the most efficient technologies to convert chemical energy to electricity via electrochemical oxidation. To date the majority of fuel cells use gaseous fuels such as hydrogen or CO. Neither of these fuels occurs naturally at levels that could be used for power generation, and both H₂ and CO have to be produced, typically through the gasification of coal or reforming of natural gas. This leads to a substantial reduction in their total 'well-to-wheel' efficiency. In contrast there are a wide range of solid high carbon fuels that occur naturally (such as coal) or are a waste product of an industrial or agricultural process. In a DCFC (direct carbon fuel cell), the chemical energy in solid carbon fuel is transformed into electricity through direct electrochemical oxidation. In a DCFC, the carbon fuel could be converted into electricity at efficiencies higher than any other technology with stand alone electric efficiency's potentially over 70%, and with combined heat and power this could increase to around 90%. These electrical efficiencies are around twice that of modern coal fired power plants leading to the amount of CO₂ for storage/sequestration being at least halved if the DCFC were fuelled on coal or near zero if fuelled on sustainable biomass. Moreover, the by-product of fuel oxidation in a DCFC is pure CO₂, as opposed to normal flue gas which often contains less than 20% CO₂, and hence the flue exhaust only requires compression before transportation and storage

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leading to further energy savings even when compared to proposed SOFC (solid oxide fuel cell) hybrid systems [3–7].

In general DCFC development has been restricted to small cells (button cells of around 1 cm² active area) tested over short time periods with the investigations focussed on fundamental aspects of the technology such as materials for fuel electrode, fuel oxidation mechanisms and materials issues due to high temperature and corrosive environments [6–12]. The latter is more critical in the case of molten salt based DCFCs. As with gas fed molten carbonate fuel cells, these systems offer good performance but are technically challenging to develop to a reasonable scale or operate for long time periods due to the highly reactive and mobile nature of the cell components [8–10,13]. Recent studies by Kulkarni et al. have shown that it is possible to eliminate molten components from the cell structure through the use of mixed ionic electronic conducting electrodes which allow for the entire surface of the electrode to be utilised for electrochemical oxidation reactions of the solid fuel [14].

This solid state DCFC with solid carbon as the fuel feed offers simplicity in operation and non-corrosive environment compared to molten carbonate electrolyte based DCFC as well as fuel in molten carbonate fed DCFC, however, the delivery of solid carbon to anode/electrolyte interface becomes more challenging. For commercialisation of DCFC technology, there needs to be a parallel effort in regards to scale-up of the technology, and the issues associated with fuelling, fabrication and long-term operation of the device. Tubular fuel cell designs have a number of advantages over conventional designs as discussed elsewhere [15,16], however, the tubular design was chosen in this case as it allows the fuel to be easily fed to the anode. In the present study, a solid oxide electrolyte (8 mol% yttria stabilised zirconia) supported tubular cell has been fabricated and operated in a pulverised carbon packed bed for a period of 11 days, to assess the scalability of the technology in terms of fabrication, fuel feed and operation. This paper presents the results on the operation and diagnostic analysis to elucidate the possible causes of the cell degradation.

2. Experimental

2.1. Cell fabrication and assembly

The closed-end electrolyte tubes were fabricated from 8 mol% Y₂O₃–ZrO₂ powder (8YSZ) (Tosoh Corporation, Japan) by isostatic pressing and sintering at 1500 °C for 2 h. The sintered tube dimensions were 9.9 mm OD, 8.7 mm ID, wall thickness 0.6 mm and a length of ~155 mm. The density was >99% of the theoretical value. Lanthanum strontium cobalt ferrite (LSCF-HP of composition La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-δ}) powder obtained from Fuel Cell Materials Inc., OH, USA was used as the anode and cathode for the tubular cells. The LSCF (lanthanum strontium cobalt ferrite) catalyst ink was prepared by milling 50 wt% of powder with 50 wt% of ink vehicle (Fuel Cell Materials Inc, OH, USA). Both LSCF anode (outside of tube) and cathode (inside of tube) were prepared by brush coating the LSCF ink on the outside and inside of the tube on a 40 mm long portion of the tube (5 mm before the closed-end of the tube) producing a cell active area of 10.9 cm².

In order to determine the polarisation losses due to the cathode (on inside of the tube), a 10 mm long portion of the electrolyte tube was brush coated with LSCF on outside of the tube, 5 mm away from the fuel electrode, to act as a reference electrode. This was followed by heat treatment at 800 °C in air for 2 h. Silver wires (0.5–1 mm diameter) bonded to the electrodes by Pt paste were used as current collectors for all the three electrodes. The three-electrode arrangement was only used to determine polarisation losses due to the air electrode by operating the cell with air supply

both to inside and outside of the tube. These investigations were performed at 750 and 800 °C with all three electrodes exposed to air before evaluating tubular cells with the carbon fuel.

For operation of the tubular cell on carbon fuel, the reactor design, shown schematically in Fig. 1, was used. It consists of a 35 mm ID SS (stainless steel) crucible with a flange. A brass flange (internally water cooled) with a number of ports for gases and electrical connections serves as a cover of the reactor as shown in Fig. 1. The tubular cell was first assembled on the reactor brass head. The SS crucible was filled with Vulcan XC-72 carbon (Cabot Corporation, USA), and the tube cell assembly inserted into the carbon bed slowly. After the assembly of the cell in the S.S. crucible it was inserted into a heated vertical chamber of a furnace up to its flange.

2.2. Cathode evaluation procedure

In order to determine cathode contribution to overall cell losses, electrochemical impedance spectroscopy was performed in a 3-electrode arrangement (air electrode as working and reference electrodes, fuel electrode as counter electrode). The EIS (electrochemical impedance spectroscopy) were obtained with IM6e Impedance analyser (Zahner, Germany) with scans performed from 100 mHz to 200 kHz frequency at an excitation signal of 20 mV. The EIS were obtained at 750 °C under OCV (open circuit voltage) and at 800 °C under OCV as well as under various values of current loads. The impedance spectra were modelled using ZView™ software (Scribner Associates Inc., USA).

2.3. Cell operation with carbon fuel

The complete reactor set-up resides inside a test station that allows the operation of the tubular cell up to 1000 °C, and has provision to supply gases such as H₂, CO, N₂, He to the outer and air to the inner chamber of the electrolyte tube. The station has multiple levels of safety redundancy for the operator and the cell, and functions in a failsafe mode for continuous operation over a period of number of days. The cell is heated to the operating temperature with helium gas (Ultra high purity with ~1 ppm oxygen, BOC) flowing in the outer chamber (through the carbon bed) and air in the inner chamber of the electrolyte tube. Once the temperature, as

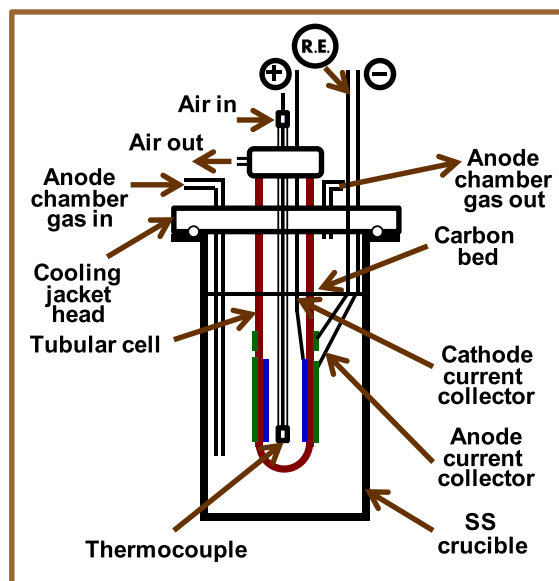


Fig. 1. A schematic design of the direct carbon fuel cell reactor employed for the evaluation of the tubular cells in a packed bed of solid carbon.

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