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Application of infrared thermal imaging to the study of pellet solid oxide fuel cells

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Abstract

The application of infrared thermal imaging to the study of solid oxide fuel cells is demonstrated. The temperature increase accompanying polarisation of gadolinium doped ceria pellet cells is measured and the effect of temperature increase on polarisation characteristics is modelled. Temperature increases of the order of 2.5 °C were measured for heavily loaded pellet cells. Measurement accuracy of 0.1 °C and spatial resolution of 0.5 mm allow temperature distribution heterogeneity to be clearly discerned. A total heat transfer coefficient is derived from experimental results that allow the development of a model that predicts the extent of self-heating. For pellet fuel cells, self-heating is not expected to have a large effect on the polarisation characteristics; however, for thin electrolytes and high current density the effect becomes appreciable. © 2007 Elsevier B.V. All rights reserved.

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1. Introduction

Solid oxide fuel cells (SOFCs) are high temperature fuel cell variants that employ oxide ion conducting ceramic electrolytes operating in the temperature range of 500–1000 °C [1]. These devices show excellent potential for a broad range of power generation applications and particularly for large scale and residential combined heat and power (CHP). However, elevated temperature operation imparts certain challenges with regard to factors such as ensuring sealing integrity and minimising mechanical stresses due to thermal expansion coefficient mismatch. Cells and stacks may suffer from significant temperature variation during operation that will lead to performance limitation. The ability to accurately measure the temperature distribution of SOFC systems and components is thus a powerful research and design tool for the advancement of this technology.

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As a non-contact method, infrared (IR) thermal imaging does not disturb the behaviour of the fuel cell, it allows measurements on operational fuel cells, provides high spatial resolution (<1 mm) that generates detailed data sets suitable for model input and has fast acquisition time that enables dynamic phenomena to be studied.

This paper reports work performed to assess areas in which infrared thermometry can be applied to SOFCs, and discusses how the resulting thermal images can be used to develop models that predict the effect of temperature on electrochemical performance. Application of the technique to the thermo-mechanical analysis of SOFCs will be the subject of a future publication.

1.1. Thermal imaging of fuel cells

Incorporation of temperature measuring devices (e.g. thermocouples and thermistors) is necessary for proper control, test and operation of fuel cells. This ranges from the crude placement of a single sensor to highly instrumented arrangements that allow 2D mapping of a single cell or plate-by-plate profiling of stacks [2].

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In order to get an accurate measure of the temperature of a cell and detect temperature changes as they happen at the electrode, it is necessary to place the sensor as close to the electrodes (or electrolyte) as possible. However, incorporation of thermocouples close to the interface of current collector and electrode may lead to membrane penetration in a polymer electrolyte fuel cell (PEFC) or cracking of an SOFC during cell assembly or heat-up. For PEFCs, thin film temperature sensors have been developed that allow *in situ* measurement within the membrane electrolyte [3]; however, this is not practical for SOFCs.

Adzic et al. [4] have used a 'flat type' thermocouple to probe the temperature distribution at the cathode of a planar SOFC. The thermocouple was placed in close proximity (as close as 5μ m), but not in contact with the electrode surface and a spatial resolution of 0.5 mm was achieved. Localised hot spots were measured that were as much as $16 \,^{\circ}$ C above the mean surface temperature.

An alternative to discreet temperature sensors is the use of infrared measurements. This technique has the advantage of not requiring sensor placement or connecting leads, and high spatial resolution and frame collection rates can be achieved. The technique is ideal for measuring the outer surface temperature of a fuel cell or stack; however, in order to investigate the temperature within a fuel cell, the construction needs to be modified to allow optical access to the surface of interest (i.e. electrode surfaces).

Infrared thermal imaging has been performed on PEFCs using IR transparent windows made from zinc selenide (>75%) transmission over $8-12 \,\mu\text{m}$) [5] and barium fluoride (>85%) transmission over 8-12 µm, although a correction was still performed) [6]. These studies showed that significant temperature variation results across the surface of a PEFC due to variation in current density, humidification and reactant composition. The work of Wang et al. [6] demonstrates how electrode surface temperature increases with current density. For a current density of $1.38 \,\mathrm{A}\,\mathrm{cm}^{-2}$ a maximum spatial distribution of ca. 6 °C was observed over the $2.9 \text{ cm} \times 3.9 \text{ cm}$ electrode surface, with over 10 °C temperature rise recorded in some parts of the electrode. These results show that it is important to have a measure of the temperature at the electrode of the fuel cell since the bulk fuel cell temperature and electrode temperature can vary significantly (over 8 °C in this study). In addition, it was also reported that a significant time was necessary for the electrode to reach a stable temperature after a current step change. Over 1800s was required in this instance in order for the heat generated to radiate through the test cell to the environment [6].

1.2. Pellet fuel cells

For materials development and investigation of different fuels, so-called 'pellet' or 'button' SOFCs are routinely employed; see Ref. [7] for an example of their use. These are normally electrolyte supported devices (typ. 250–1500 μ m in thickness) with screen-printed or painted anode and cathode and metallic gauze current collectors. The active surface area is typically of the order of 1 cm². A reference electrode is sometimes incorporated in order to explicitly determine the performance of the anode and cathode [8]. Pellet cells are not intended for technological applications, but rather for screening purposes. The relatively thick electrolyte support and ambient-air 'breathing' cathode usually precludes high performance. However, their small size and homogeneous reactant supply to the electrodes allows the useful assumption that current density and temperature are spatially invariant. It is also generally assumed when analysing pellet cell results that the temperature does not change when the cell is polarised. This is important since even a small change in temperature can lead to an appreciable change in electrolyte conductivity or electrochemical activity. In this work, the temperature distribution characteristics of operating pellet cells are examined and the assumptions about their behaviour considered.

Pellet fuel cells operating in the 'intermediate' temperature (IT) range of 500–650 °C are considered in this study [9]. This temperature regime of operation has the advantage of allowing more rapid start-up and shut-down; reduced rates of corrosion; simplified system requirements and lower component cost compared with high temperature SOFCs. To achieve the requisite performance at lower temperatures, materials are required that have superior conductivity and electrochemical activity than conventionally employed SOFC materials. In this case, gadolinium doped ceria (CGO) is used as the electrolyte [10], Ni/CGO cermet as the anode, and La_{1-x}Sr_xCo_yFe_{1-y}O_{3- δ} (LSCF), a perovskite based on LaCoO₃ and specially developed for IT-SOFC operation, as the cathode [8].

2. Experimental

2.1. Materials preparation

Cylindrical electrolyte pellets were produced by uniaxially pressing $Ce_{0.9}Gd_{0.1}O_{1.95}$ powder (Rhodia) at 1 tonne (30 s), followed by sintering at 1450 °C in air for 2 h. Pellets with a thickness of 0.35 mm and a diameter of 16 mm were prepared.

The anode cermet slurry was prepared using a 40 wt.% $Ce_{0.9}Gd_{0.1}O_{1.95}$ (CGO-10) and 60 eq.wt.% of Ni in the form of green NiO (Alpha Aesar) mixed with various additives including binders, dispersant, plasticizer and solvent. The doctor blade technique was used to coat the electrode onto the electrolyte which, after being sintered and reduced at 1300 °C in air for 2 h, had a thickness of 60 μ m. Each pellet was initially reduced in 10% H₂/N₂ (+2.3% H₂O) as the electrodes were raised from ambient to working (550–650 °C) temperature.

The cathode was $(La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta})/30$ wt.% CGO10 paint coated to give a final thickness of ca. 30 μ m. The area specific resistance (ASR) of the cathode was calculated from symmetrical cell ac impedance measurements to be 0.84 Ω cm² at 600 °C. Current collectors were affixed to the electrodes using some additional electrode material and sintering. Pt gauze was used as the current collector on the anode side and Au gauze on the cathode, both 0.06 mm wire diameter and 0.25 mm aperture. Pt must be used at the anode due to the requirement of a higher sintering temperature.

Both the anode and the cathode were deposited with a diameter of 1 cm, giving a geometric surface area of 0.785 cm^2 .

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