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Journal of Power Sources 86 (2000) 401–403

JOURNAL OF POWER SOURCES

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Cannock landfill gas powering a small tubular solid oxide fuel cell — a case study

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Accepted 8 October 1999

Abstract

Cannock landfill gas — mainly a mixture of methane and carbon dioxide — can successfully power a small tubular solid oxide fuel cell. Initial experiments showed a relatively rapid falling off in power due to poisoning with hydrogen sulphide. A simple de-sulphurisation system alleviated this problem. Even greater performance was achieved by the pre-addition of air to help in the reforming of the gas, giving little loss of power over the lifetime of the experiment. © 2000 Elsevier Science S.A. All rights reserved.

Keywords: Methane; Carbon dioxide; Landfill gas

1. Introduction

Small tubular solid oxide fuel cells made from 8% yttria-doped zirconia with a wall thickness of 200 μm have previously been shown to have fast start up and excellent heat shock resistance capability [1,2]. A perfect test fuel for such a system would be one that is transient in nature and, preferably, cheap and readily available. One such possible fuel is biogas which is transient, variable in composition, cheap and presently under utilised. The available methane in biogas is such that a solid oxide fuel cell is well able to utilise it as a fuel source [3]. Here samples of biogas have been obtained from Cannock landfill site and are used to generate electrical power in a small tubular solid oxide fuel cell. The initial results are quite encouraging as, with a minimum of pretreatment, continuous electrical power was produced for the duration of the experiment.

2. Experimental

Small diameter solid oxide fuel cells were prepared with an internal nickel cermet anode, produced from a mixture of 0.9 g ZrO_2, 9.5 g NiO and 0.5 g CeO_2, sintered at 1300°C for 1 h. An LSM: lanthanum–strontium manganite cathode 3 cm long, giving a total cathode surface area of 2.4 cm^2 was painted onto the outside of the tube. Nickel wire was used to collect the current at the anode and silver wire to deliver current to the cathode [4]. Individual cells were used to keep the system as simple as possible. Cannock landfill gas was delivered from 10l bags through a mass controller into the fuel cell. The fuel cell was kept inside a furnace held at 850°C. The electrical power produced by the fuel cell was measured by potentiostat (Fig. 1).

Analysis by gas chromatography showed Cannock landfill gas to be a mixture of the following gases:

Nitrogen	18%
Methane	56%
Carbon dioxide	26%
Hydrogen sulphide	< 1%

This gas was metered into the fuel cell at a rate of 6 ml/min and electrical power readings taken at regular

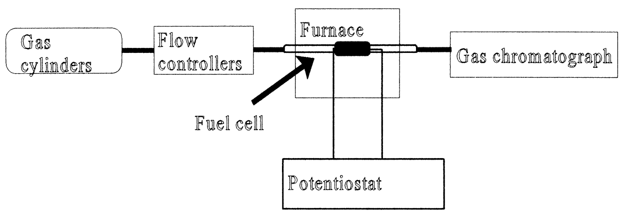

Fig. 1. Experimental system for testing a small tubular SOFC on Cannock landfill gas.

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PII: S0378-7753(99)00417-6

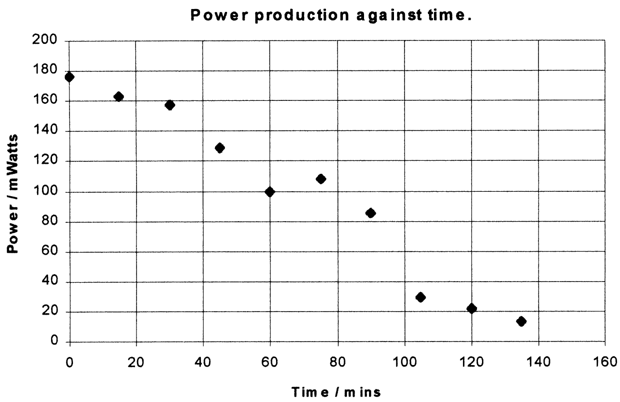

Fig. 2. Power output against time of Cannock landfill gas.

intervals. Between readings the cell was kept at a constant voltage of 0.7 V.

3. Initial results

With no pretreatment the power output of the cell quickly declined due to poisoning by the small quantity of hydrogen sulphide within the gas. Hydrogen sulphide is a well known poison for nickel reforming catalyst [5] and the effect on the power output of Cannock landfill gas is shown in Fig. 2.

In order to improve on this a de-sulphurisation stage was introduced between the mass flow controller and the fuel cell. This consisted of bubbling the gas through a 5% solution of sodium carbonate. De-sulphurisation greatly reduced the cells power loss as can be seen in Fig. 3.

As can be seen from the above graph de-sulphurisation has greatly improved the longevity of a cell powered by Cannock landfill gas. After some 6 h running, the cell is still producing around 70% of its maximum power output.

It was considered that this could be improved upon by augmenting the natural reforming going on within the fuel cell. In a methane–carbon dioxide mixture the carbon dioxide present will reform the methane into a mixture of

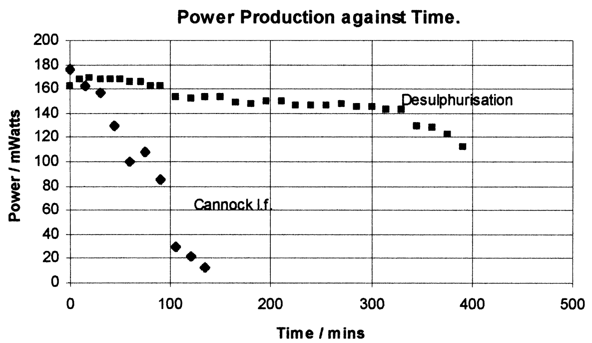

Fig. 3. The effect of de-sulphurisation on Cannock landfill gas.

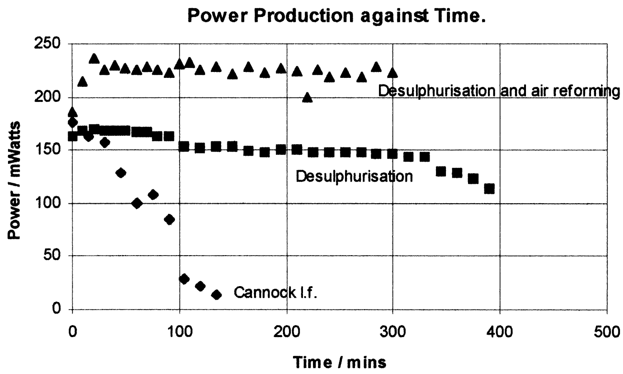

Fig. 4. Desulphurisation and air reforming of Cannock landfill gas.

hydrogen and carbon monoxide [6] which are more available fuels for the cell:

$$CH_4 + CO_2 = 2H_2 + 2CO. \quad (1)$$

This helps prevent the fast build up of carbon which occurs in cells powered by the direct electrochemical oxidation of methane [3]. Unfortunately, there is not enough carbon dioxide available in Cannock landfill gas to complete a stoichiometric reforming of methane, so to enable complete reforming this must be augmented in some way. One of the easier ways of doing this is by air reforming, as the oxygen in air will reform methane to carbon monoxide and hydrogen:

$$CH_4 + 1/2O_2 = 2H_2 + CO. \quad (2)$$

To achieve this air was premixed with a sample of Cannock landfill gas giving a final mixture of, 44% air, 40% methane and 16% carbon dioxide. This mixture was then desulphurised as before and used to power a fuel cell.

As can be seen from Fig. 4 the effect is to increase both the longevity and the overall power production. After 120 min the power production had risen from start up to 120% and was still at 119% of the start up after 300 min. Even taking into account the maximum power production which came about after 20 min running time, the final power production was still 94% of this value. This improvement in power production comes about from the greater availability of hydrogen and carbon monoxide for the air-reformed biogas. The greater longevity is probably a result of the direct oxidation of the available hydrogen sulphide to sulphur dioxide further limiting the poisoning of the nickel catalyst.

4. Conclusion

It is possible to produce power from a small tubular solid oxide fuel cell using biogas. Cannock landfill gas showed problems in longevity. These problems were overcome by a simple desulphurisation system and by augmenting the inherent carbon dioxide reforming with partial oxidation. The final experiment produced a cell running at 18.5% efficiency for 5 h at a current density of 524 mA

cm^{-2} centimetre squared, with little overall loss of power after an initial gain of approx. 20% shortly after start up.

Greater longevity tests are required to find the life expectancy of a small tubular solid oxide fuel cell running under these conditions as well as cycle tests to show the effect of shutdowns and start ups on the cell performance. Other landfill gases need to be studied as well as other sources of biogas to see if they are comparable in performance to that of Cannock landfill gas.

Acknowledgements

I would like to thank David Alcock and Poplars for the supply of Cannock landfill gas, a vital necessity for this study.

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