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Biogas powering a small tubular solid oxide fuel cell

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Abstract

Biogas has been used to power a small tubular solid oxide fuel cell (SOFC). It was demonstrated that biogas could provide power equivalent to hydrogen, even when the methane content was reduced below the value at which normal combustion could occur. The carbon dioxide content of biogas was especially beneficial because it aided the internal reforming process. But carbon deposition was a problem unless air was added to the biogas before it entered the cell. When air was premixed, the biogas was comparable with hydrogen in the power produced. However, a problem was the variability of biogas samples. Of the three types tested, only one produced a consistent power output. © 1998 Elsevier Science S.A.

Keywords: Biogas; SOFC; Power output

1. Introduction

It would be advantageous for the commercialisation of fuel cells [1,2] if a power source not fully utilised by conventional power systems could be employed, thus providing a niche for the new technology. Biogas, the material given off by anaerobic bacterial digestion of organic matter [3], offers one such opportunity. Although such fuel, derived from sewage, was first used to power street lights in Exeter in 1896, most biogas is currently vented to atmosphere, except for selected land-fill and sewage plant sites.

The main problem of biogas is that its composition depends greatly on the source, and also varies with time. After a few years, for example in landfill production, the methane content of biogas diminishes and ordinary combustion engines do not ignite satisfactorily. This is a substantial difficulty for internal combustion generators but should not have much effect on solid oxide fuel cell (SOFC) generation. Indeed, this paper shows that SOFC generators thrive on higher carbon dioxide contents, and provide equal power across a wide range of biogas compositions.

First, the experimental tubular SOFC is described. Then, preliminary experiments show how simulated biogas compares to hydrogen in power production. It is demonstrated that adding air to the biogas allows improved internal reforming and prevents carbon formation. With this addition, three types of biogas are compared. One, from Risley landfill, compared well with hydrogen in terms of power output.

2. Biogas

Under air-free (anaerobic) conditions certain bacteria will metabolise organic material producing methane and carbon dioxide as by-products. The reactions of methanogenesis are complex and involve the concerted efforts of different types of bacteria. Because of the many different kinds and populations of bacteria, the final mixture of gases released by anaerobic digestion can vary dramatically. This usually varies between a 2:1 and a 1:1 mixture of methane to carbon dioxide with varying amounts of other volatile ‘impurities’.

3. Experimental

Small tubular SOFCs were made by mixing 8-Y zirconia (Viking Chemicals, Denmark) into a plastic paste with water and polymer, then extruding the mixture through a 2-mm diameter tube die with 200 μm wall thickness. After drying, the tubes were fired to 1450°C to give electrolyte with excellent strength and ionic conductivity. Nickel cermet anodes were painted on the inside and lanthanum strontium manganite on the outside. Metal wires were wrapped inside and outside the tubes to provide current collection. The resulting cell was so robust that it could be heated to

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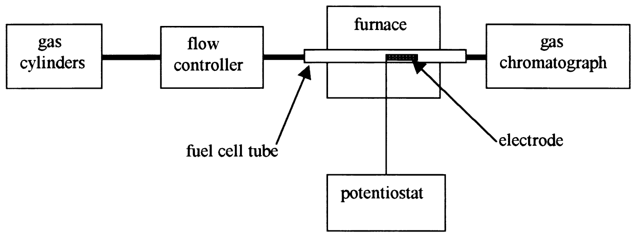

Fig. 1. Experimental system for testing SOFC on biogas.

800°C operating temperature in a few seconds. Also, it could be passed through the hot wall of the furnace to provide a polymer seal in the cold zone for the gas inlet [4,5].

The test system is shown in Fig. 1. It comprised a fuel feed from gas cylinders, via a flow controller, to the fuel cell. The power output from the cell was measured using a potentiostat, while the exit gas composition was studied by gas chromatography. Simulated biogas was made by mixing methane and carbon dioxide in 2:1 ratio. Ordinary natural gas was supplied from the laboratory pipeline. Biogas samples were fed in from 10-litre plastic bags.

4. Results

In the first experiments, hydrogen fuel was compared with natural gas and methane/carbon dioxide mixtures to simulate biogas. The results (Fig. 2) showed that the natural gas coked up immediately on the hot anode, whereas the simulated biogas ran about half as well as hydrogen. When the simulated biogas was saturated with water by passing through a bubbler, the performance improved further. However, coking was still a major problem, leading to the early demise of the cell by blockage.

The best way to remove this carbon was to inject air into the biogas before it entered the cell anode. 30 ml/min of natural gas, 15 ml/min of carbon dioxide and 75 ml/min of air were used. The voltage then rose as shown in Fig. 3. No carbon was observed after several hours running under this condition.

Several air flow rates were tested including 50, 40 and 30 ml/min. The power varied with air addition, but under certain conditions was higher than that obtained with pure

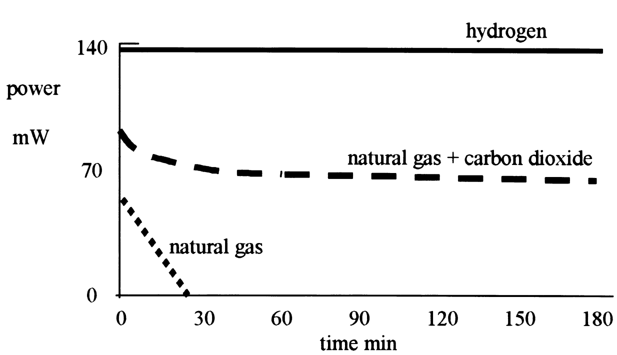

Fig. 2. Power production from SOFC comparing different gases.

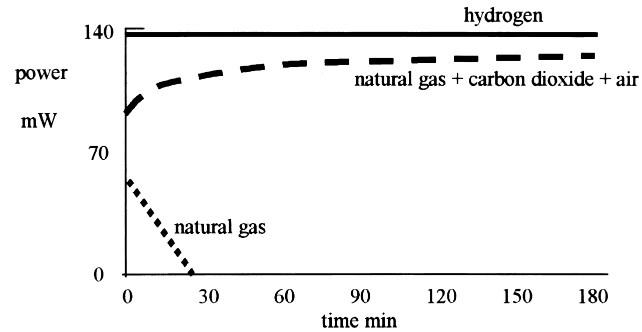

Fig. 3. Improvement in performance with air added to simulated biogas.

hydrogen fuel. The optimum amount of air for power and longevity has not yet been determined. Further tests were carried out to find how low a level of methane could be tolerated in the biogas. Significant power was still generated at 20% methane to carbon dioxide. This experiment proves that SOFCs are better than combustion engines for power generation from biogas at low methane levels. Combustion engines cannot ignite under these conditions.

Three kinds of real biogas were then tested as shown by the results in Fig. 4. The bottom line was given by the laboratory-prepared biogas, which coked the cell after about 30 min. Biogas from Llanypwll landfill site was somewhat better, but this deposited carbon after an hour. The best sample was from Risley landfill site. This continued performing well after 1 h. Adding air to these samples is likely to produce substantial improvements.

5. Applications

Biogas is freely available from both biological and man-made sources. Marshes and mizzens have long been known to produce a combustible gas, but these are now limited to unpopulated areas and are thus difficult to utilise. Man-made sources of biogas, both accidental and deliberate, are much more readily available for utilisation as a power source. Landfill sites and sewage works produce copious quantities of biogas as a by-product. Conventional power sources have been used to produce heat and power from these sources but suffer from ignition problems at low

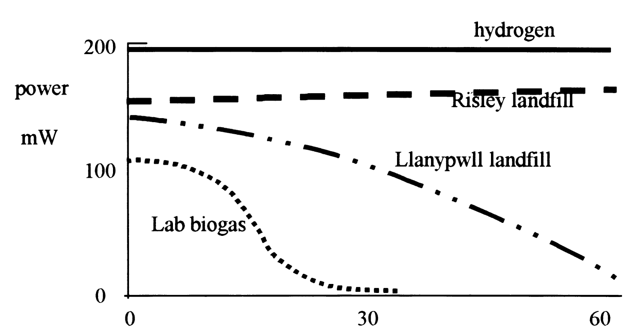

Fig. 4. Power production of biogas samples at 30 ml/min flow rate.

methane ratios. Thus, these sources are available for exploitation by fuel cell technology [6–8].

Another man-made source of biogas is deliberate anaerobic digestion to produce methane as a heating fuel. This is mainly carried on in third-world countries where mains supplies of gas and electricity are limited to the cities. Using this gas in a fuel cell system would enable the production of electricity in areas where it is currently unavailable. Biogas produced in a digester has its own unique problems. If the gas is produced in a batch process the methane content varies with time, and the early gas consists mainly of carbon dioxide and some hydrogen which is useless for power production. Thus there is a need to monitor the gas for methane content. This is not such a problem in a continuous system but due to the small quantities involved in the batch process, the methane content can vary considerably over small intervals of time.

6. Problems

A fuel cell can still run reasonably well at the extremes of low methane concentration but there is the problem of trace impurities. The main problem impurity is sulfur, which in high concentration will poison the catalytic nickel surface of the cell anode. This sulfur comes in two forms, hydrogen sulfide and volatile organic sulfur. This has to be dealt with in a two-stage process; first all the sulfur present is reduced to hydrogen sulfide with hydrogen over a catalyst of mixed transition metals, followed by removal by passing the resultant gas over iron filings. This does not have to be 100% effective as SOFCs are resistant to low levels of hydrogen sulfide.

7. Conclusions

This study has demonstrated that biogas can produce significant power from SOFCs running at 850°C. Risley landfill gas performed almost as well as hydrogen, and simulated biogas (30 ml/min methane, 15 ml/min carbon dioxide) performed better than hydrogen if air (30 ml/min) was added as a premix to the gas to prevent coking. This suggests that SOFCs could find a niche market in the biogas utilisation area, especially with gases which cannot be ignited because of their low methane content, making the gases unsuitable for combustion engine generators. Future work will concentrate on the impurity effects in biogas, especially on the lifetime of the electrodes.

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