Internal Reforming in Intermediate Temperature SOFCs running on Natural Gas

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There is increasing interest in the development of intermediate temperature solid oxide fuel cells (IT-SOFCs), utilising either gadolinia-doped ceria as the electrolyte material or supported SOFCs with ultra-thin yttria-stabilised zirconia electrolyte layers, which show excellent current densities at lower operating temperatures than those used for conventional zirconia based SOFCs. Such intermediate temperature SOFCs will lessen the demand on other components within the SOFC, in particular the interconnect and manifolding materials, and hence reduce manufacturing costs, whilst still potentially maintaining the advantages of SOFCs over other fuel cell systems in terms of fuel processing, efficiency and reduced system complexity.

Although in recent years there have been various studies of internal reforming in zirconia-based SOFCs [1-3], there has been a comparative lack of studies on ceria-gadolinia based SOFCs using natural gas, where the lower operating temperature makes internal reforming a potential issue. We have undertaken a detailed study of methane steam reforming over a range of different nickel/ceria-gadolinia anodes, including anodes derived from intermetallic $CeNi_5$ alloys prepared by inert gas atomisation, over the operating temperature range 500-700°C, for use in internally reforming intermediate temperature ceria-gadolinia based SOFCs. The influence of anode formulation, preparation route, structure and pre-treatment on the reforming characteristics and on carbon deposition has been investigated for each anode as a function of operating temperature and steam/methane ratio, with particular emphasis on the behaviour at low steam/methane ratios. All the nickel/ceria anodes studied show good activity towards methane steam reforming at 600°C and 650°C, with lower but still significant, activities observed at lower reaction temperatures, together with a corresponding decrease in the CO selectivity. At 650°C high selectivity to CO is observed for all methane-rich or equimolar methane/steam mixtures. The amount of carbon deposited on the anodes during reforming over this temperature range is very low when a steam/methane ratio of ≥ 1 is employed. The conversions observed at 650°C are consistent with steam reforming rather than methane pyrolysis being the predominant reaction pathway under reaction conditions.

The nickel/ceria anodes derived from gas atomised intermetallic precursors have been characterised structurally and evaluated for their methane reforming characteristics and their resistance to carbon deposition. Their catalytic behaviour has been compared with that of anodes prepared by the conventional mixed oxide ceramic processing route and anodes generated from intermetallic cast alloys, with very favourable results.

The effect of doping nickel/ceria-gadolinia anodes with small quantities of ruthenium has been investigated. Incorporating small amounts of ruthenium results in an anode with increased activity for methane reforming, that is resistant to carbon deposition, in the relevant operating temperature range of ceria-gadolinia based SOFCs.

[1] C.M. Finnerty, N.J. Coe, R.H. Cunningham and R.M. Ormerod, Catalysis Today, 46 (1998) 137.
[2] R.M. Ormerod, Stud. Surf. Sci. Catal., 122 (1999) 35
[3] C.M. Finnerty and R.M. Ormerod, J. Power Sources, 86 (2000) 390.