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Characterisation of Rare Earth Doped Neodymium Cuprates for Solid Oxide Fuel Cell Applications

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Abstract – Rare earth doped neodymium cuprates have been characterised for SOFC applications. Results of conductivity and AC impedance show that the materials may be suitable for both cathode and cathode current collector applications. Furthermore significant sinter activity at stack conditioning temperatures suggest that the current collection function may also be combined into an in-situ fired contact which may be useful both for stack fabrication and improving degradation.

Cathode materials have long been an area of significant development effort within the SOFC research community. As a result of this effort a number of materials systems have become well established and some of these are being used in devices approaching commercial reality. However, there is still a need to explore interesting new materials systems which may offer advantages in terms of performance, durability and robustness over the existing cathode materials, thereby extending the capabilities of future generations of SOFC. Ceramics which exhibit longer range crystallographic features, such as layering, may offer many potentially interesting properties due to the defect structures which can be introduced through doping and which could result in considerable levels of ionic and electrical conductivity along these layers. One such family of materials are rare earth doped neodymium cuprates ($Nd_{2-x}Ln_xCuO_{4+\delta}$) where Ln in this study was either Ce or Pr (NCCO and NPCO respectively). What separates these materials from other commonly used cathode materials is the fact that they exhibit n type semiconductivity as opposed to the p type conductivity exhibited by most other materials used in this application. It is hoped that the n-type conductivity may confer operational advantages when operating at high oxygen utilizations or high cell polarisations where the local cathode oxygen partial pressure is reduced. Also, as it contains neither Mn nor Sr (both of which have been implicated in nucleating Cr deposition [1]) it may also have benefits in terms of Cr tolerance.

Samples were synthesised with dopant levels of between x = 0.15 and 0.35 and test specimens prepared by either uniaxial pressing to form pellets or by screen printing of cuprate inks on to electrolyte pellets of YSZ of GDC form porous electrode films. Powders were characterised by XRD, 4 point DC conductivity and 2 electrode AC impedance. In general the structure can be indexed to a tetragonal T' structure similar to the K_2NiF_4 T type structure, however at values of X approaching 0.2 secondary phases can often be observed. These were attributed to dopant rich precipitates appearing close to or above the dopant solubility limit. This limit is also where maximum conductivities were observed (x=0.2-0.25, depending of the dopant), with maximum values of $60Scm^{-1}$ being observed at 800°C. Significant microstructural development at typical stack conditioning temperatures (around 900°C) was also observed and there is scope to create an in-situ fired combined current collector and cell contact. So far results obtained using these in-situ structures have shown Rp values of $0.066\Omega cm^2$ to $0.115\ \Omega cm^2$ at around 750°C. These are good enough for mid temperature SOFC operation in the range of 700 to 850°C. SEM examination of has also revealed that in certain circumstances interesting microstructural segregation occurs, which may provide a route to allow some level of functional grading in these structures. Cathode performance will be discussed with reference to dopant level, type and electrode microstructure.

The general performance observed for the insitu-fired structures suggests possible applications in the temperature range of 850-700°C and may give a method for the incorporation of current collection and cell to interconnect contact in a single layer. This may be advantageous for stack assembly and reduced degradation, especially if the unfired layer can be engineered to allow some degree of compliance during stack build and so provide a reliable and consistent cathode contact to the interconnect. Now that the basic cathode behaviour envelope has been investigated and shown to be promising, further testing both at cell and half cell level are now required to investigate whether the promised advantages of this material, such as improved performance in reduced oxygen partial pressure and a tolerance to Cr poisoning, can be realized.

References

[1] S.P Jiang, Z.P. Zhang, X.G. Zheng, *J. Euro. Ceram. Soc.*, **22**, 361, (2002).