Electrodes for Oxidation of Methane

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Introduction: Converting hydrocarbons in a solid oxide fuel cell (SOFC) have proved to be tricky (1). In general high R_p values are obtained and a low stability of the electrodes is observed (2). Attempts have been made to improve the performance and stability of the anodes by using a composite of copper and ceria (a known carbon oxidation catalyst (3)). In has been shown possible to fabricate composite anodes, which gives a fair performance and are stable for prolonged periods of time in i.e. methane (4). This work presents some preliminary results obtained using different types of oxide-based electrodes for the conversion of methane. It is shown that the electrodes containing ceria show good ability with respect to ageing, but that the R_p values (typically around 1 Ωcm^2 at 1000°C for the best electrodes) obtained are much to high for practical applications.

Experimental: The electrodes were fabricated using standard ceramic processing and slurry spraying. The electrodes where sprayed onto Risø three electrode pellets. The measurements were done using a three-atmosphere setup. As a reference gas air was used and nitrogen was used as the surrounding atmosphere. The electrochemical characterisation is performed using a combination of a Solartron 1250 frequency response analyser and a Solartron 1287 electrochemical interface. The measurements are performed within a frequency range 65500 Hz to 1 mHz with an amplitude of approximately 28 mV.

Results: Selected impedance spectres are shown in figure 1-3. In general 1-3 distinct arcs are observed in the spectres.

Discussion: In general the ASR values obtained in this study are much to high for practical applications. The best electrodes show an ASR of 1.2 Ωcm^2 at 1000°C. The stability of the new alloy/YSZ and the new alloy/CGO are remarkably good. Only the ceria based electrodes showed long time stability under a flow of methane at OCV.

Conclusion: Electrodes with good stability in flowing methane at OCV has been fabricated. The best electrodes show an R_p of 1.2 Ωcm^2 and consist of a 50/50 % (w/w) mixture of CGO10 and a new alloy. The ceria-based electrodes are stable over a prolonged period of time in methane at OCV.

References:
1. C.M. Finnerty, N.J. Coe, R.H. Cunningham, R-M. Ormerod, *Catal. Today* **46,** 137 (1998)
2. G. Wang, W.L. Worrell, S. Park, J.M. Vohs and R.J. Gorte, *J. Electrochem. Soc.,* **148(8),** A864 (2001).
3. A.Trovarelli, *Catal. Rev.-Sci. Eng.,* **38,** 439 (1996).
4. H. Kim, C. Lu, W.L. Worrell, J.M. Vohs and R.J. Gorte, *J. Electrochem. Soc.,* **149(3),** A247 (2002).

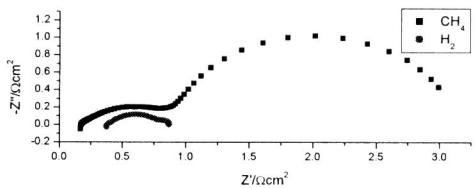

Figure 1. EIS of an Au/CGO40 composite anode in wet H_2 and wet CH_4 respectively at 950°C.

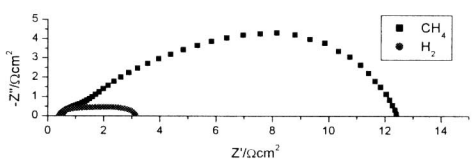

Figure 2. EIS of a Cu/CGO40 composite anode at 950°C in wet H_2 and wet CH_4 respectively.

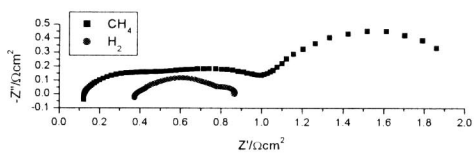

Figure 3. EIS of a Ni/CGO40 composite anode at 950°C in wet H_2 and wet CH_4 respectively.

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