

Introduction

Polymer electrolyte fuel cells (PEFCs) are one of the most interesting alternatives for a pollution-free energy production. One of the major challenges in the development of PEFCs is to exploit the whole capacity that inherits a given membrane electrode assembly (MEA). Mass transport effects lead to an inhomogeneous electrochemical activity over the electrode area. In order to investigate in detail the influence of gas composition on the cell performance and to elucidate mass transport effects in the cell current-voltage curves, measurements of current density distribution and electrochemical impedance spectra (EIS) were recorded.

Experimental

At DLR a flexible dry powder spraying method for the manufacturing of membrane electrode assemblies (MEA) has been developed which was used to prepare membrane electrode assemblies (MEAs). In addition commercially available catalysts coated membranes (CCMs) from Ion Power Inc., (N111 IP) and gas diffusion layers (GDLs) from SGL SIGRACET 35 DC have been used.

Results and Discussion

The measured EIS can be simulated using an equivalent circuit taking into account the uncompensated membrane resistance (electrolyte resistance), kinetics of the oxygen reduction reaction, the kinetics of hydrogen oxidation reaction, mass transport limitations and the porous structure of anode and cathode.

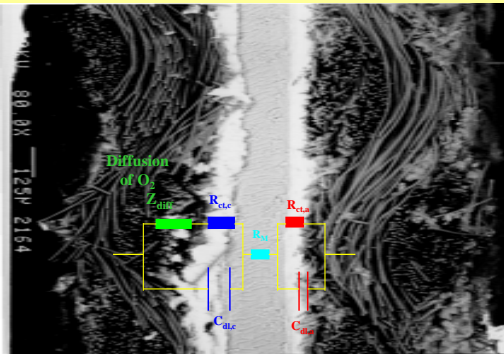


Fig.1: Equivalent circuit (EC) used for the simulation of EIS inserted into SEM picture of a MEA cross section

Results

The used electrode model (Fig.2) is the cylindrical-pore model proposed by Göhr, where the impedance elements of the pore's wall surface (Z_q) are the double layer capacity (C_{dl}), charge transfer resistance (R_{ct}) and impedance elements of oxygen diffusion (Z_{diff}).

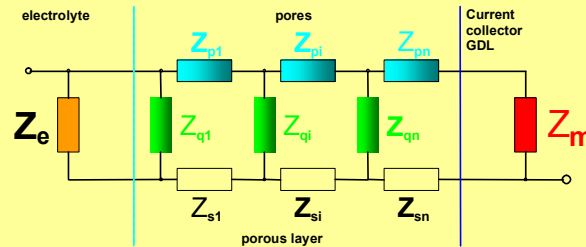


Fig.2: Cylindrical homogeneous porous electrode model

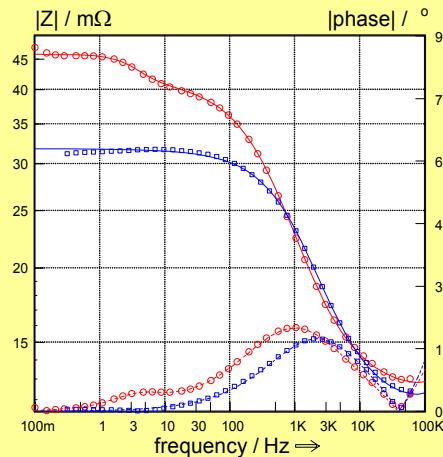


Fig.3: Comparison of EIS (Bode plot) of the same in house made MEA produced with carbon supported (60 wt% Pt) catalyst and 20 wt% Nafion, operated with H_2/O_2 (\square) and H_2/air (\circ) at 500 mAcm^{-2}

Results

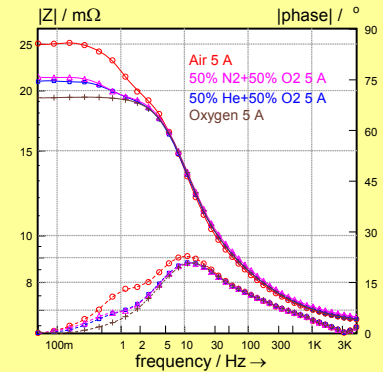


Fig.4: Comparison of EIS (Bode plot) measured at 5 A, 80°C , $\lambda=1.5$, N111 IP CCM, cathode fed with different gas composition

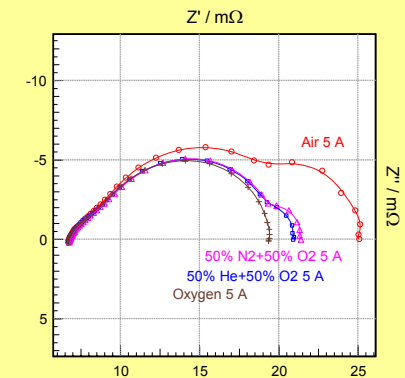


Fig.5: Comparison of EIS (Nyquist plot) measured at 5 A, 80°C , $\lambda=1.5$, N111 IP CCM, cathode fed with different gas composition

Conclusions

Major source of performance loss during operation of fuel cells is the slow kinetic of the oxygen reduction reaction (ORR). During operation of the cell with air instead with pure oxygen one can find two sources of performance loss at the cathode:

- decreasing of charge transfer reaction rate with decreasing partial pressure of oxygen corresponding to an increase of the charge transfer resistance ($R_{ct,C}$).
- appearance of an additional diffusion impedance term in the low frequency range of the impedance spectra, for example at 3 Hz in Fig.3.