EIS Investigations on Novel Electrodes and Membranes for Low Temperature Fuel Cells

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The information of current voltage characteristics in investigating fuel cells is limited. A more powerful tool to investigate polymer electrolyte fuel cells (PEFC) [1] and other low temperature fuel cells is electrochemical impedance spectroscopy (EIS). Under different operating conditions limiting processes inside the fuel cell e.g. diffusion, charge transfer or ohmic resistance can be determined.

Proton conducting membranes

Microporous PTFE and PVDF matrices are filled with sulfuric acid and triflic acid to obtain proton conducting membranes. These membranes are tested in respect of the applicability in H_2/O_2 and methanol fuel cells.
The ionic conductivity of these membranes is measured using AC impedance spectroscopy (5 to 50 kHz).
Different inorganic oxides (TiO_2, ZrO_2, SiO_2 Al_2O_3) are used as fillers in these composite membranes. The influence of the particle size and content of these fillers on the proton conductivity as well as on the extent of the methanol crossover [2] is examined.

In addition to the characterization of the pure membranes membrane electrode assemblies (MEAs) were prepared from different proton conducting membranes and characterized with the electrochemical impedance spectroscopy (EIS) technique. The amplitude of the AC signal was 5 to 10 mV rms and the frequency was typically varied from 10 mHz to 100 kHz.

Methanol oxidation in sulfuric acid

According to Conway and Harrington [3] the impedance characteristic of methanol anodes can be explained by a multi-stage reaction with an adsorbed intermediate (without diffusion limiting).

$$H_3COH \xrightarrow[v_1]{[Pt]} (CO)_{ads} + 4e^- + 4H^+ \xrightarrow[v_2]{+H_2O\ [Ru]} CO_2 + 2e^- + 2H^+$$

The electrode can be simulated by the circuit in figure 1.

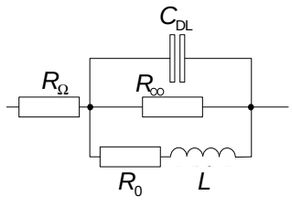

Figure 1: equivalent circuit to fit experimental data

For these measurements gas diffusion electrodes were prepared by a rolling method. They consist of a diffusion layer, a catalyst layer and metal grid for current collection. The anodes active layer is composed of PTFE (Hostaflon), Graphite (Timcal), Shawinigan Black (Chevron) and Oppanol B100 (BASF) as binder. The catalyst loading was 3,5 mg/cm^2 Pt/Ru on Vulcan XC72 (Johnson Matthey) [4].

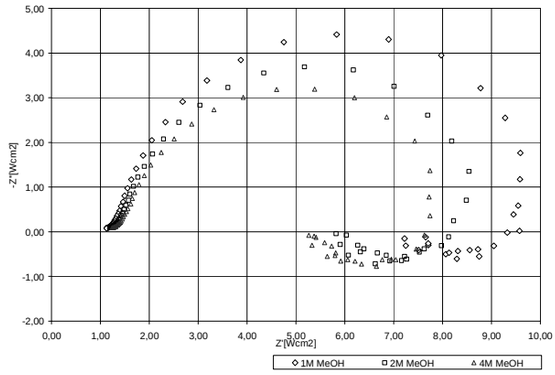

Figure 2: influence of the methanol concentration
E vs. sat. $Hg/Hg_2SO_4/SO_4^{2-}$ = -250mV = 400mV vs. NHE, T=60°C, c(H_2SO_4)=1M, U_{Mod}=10mV
3mHz <= f_{Mod} <= 1kHz, 10pts./dec.

The results for fitting the curve with the circuit described in figure 1 above are summarized in the table below.

	RΩ [Ω]	R∞ [Ω]	R0 [Ω]	L [H]	τ
1M	1,24	8,45	26,33	135	5,13
2M	1,32	7,07	15,83	63	3,95
4M	1,39	6,33	11,95	42	3,52

With increasing methanol concentration the ohmic resistance increases while the time constant for the methanol oxidation slightly decreases.

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