TIME RESOLVED ELECTROCHEMICAL IMPEDANCE SPECTRA (EIS) DURING CARBON MONOXIDE-POISONING OF Pt- AND PtRu-ANODES IN MEMBRANE FUEL CELLS

N. Wagner, M. Schulze
Deutsches Zentrum für Luft- und Raumfahrt (DLR),
Institut für Technische Thermodynamik
Pfaffenwaldring 38-40, D-70569 Stuttgart, Germany
E-mail: norbert.wagner@dlr.de

Polymer Electrolyte Fuel Cells (PEFC) have been receiving more and more attention due to their high energy conversion efficiency and emission free-operation for powering electric vehicles. The highest performance is achieved with pure hydrogen (H_2) which is the preferred fuel for low-temperature fuel cells. However, pure H_2 has several limitations. The storage systems for liquid or compressed H_2 are heavy and bulky. Furthermore, H_2 refueling is costly and takes time. A solution is to generate hydrogen on board of the electric vehicle by reforming either hydrocarbons (e.g. natural gas) or alcohol (e.g. methanol). The reformate feed gas may contain up to 2.5% carbon monoxide (CO) by volume, which can be reduced to about 50 ppm CO using a selective oxidizer. Although Pt has proven to be the most effective catalyst for hydrogen oxidation in PEFCs [1], even small traces of carbon monoxide can cause significant reductions in fuel cell performance. In order to improve the carbon monoxide tolerance of the anode in fuel cells new catalysts like Pt/Ru are investigated.

The progressive poisoning with carbon monoxide of a fuel cell was monitored using Time Resolved Electrochemical Impedance Spectroscopy (TREIS). For this, measurements at distinct time intervals during the experiment were performed. The poisoning causes a change of the state of the fuel cell which is reflected in the recorded impedance spectra (Fig.1 and Fig.2).

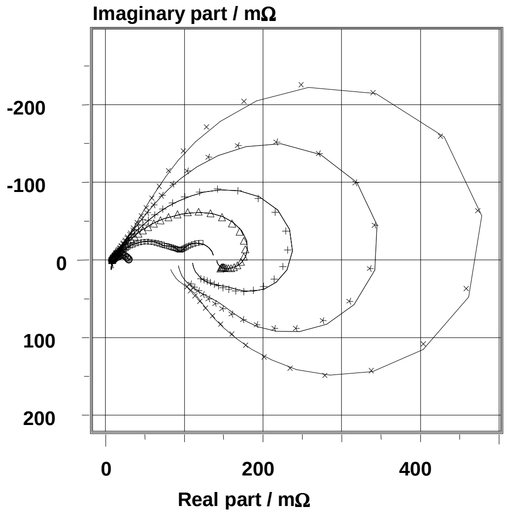

Fig.1: Nyquist plot of EIS measured at different times during poisoning of the Pt-anode with CO

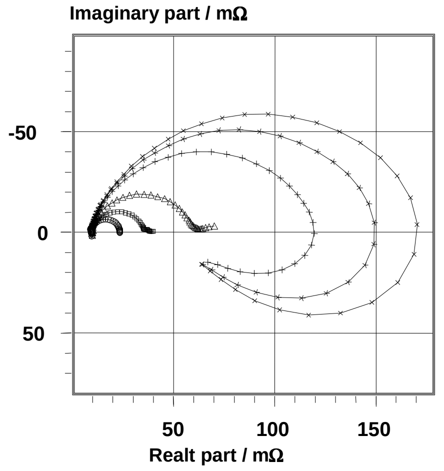

Fig.2: Nyquist plot of EIS measured at different times during poisoning of the PtRu-anode with CO

Besides an increase of the total impedance of the fuel cell, the occurrence and the increase of a pseudo-inductive behaviour is observed. For the evaluation of the series measurements, enhanced mathematical procedures, like the real-time drift compensation, the time course interpolation and an additional refinement were applied to reduce the influence of the changing state of the fuel cell in the obtained spectra [2]. Due to the experimental conditions, the evaluation of the spectra can be considerably simplified.

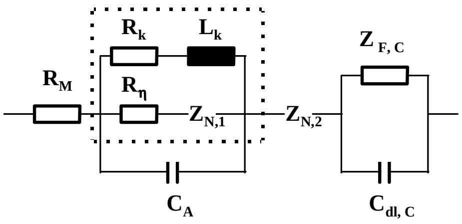

Fig. 3: Equivalent circuit for evaluation of the impedance spectra measured during poisoning of the anode with CO

The transient change of the charge transfer resistances shows that the degradation of the fuel cell performance during the poisoning is dominated by an increase of the anodic resistance whereas the cathodic one can be considered to be nearly constant during the whole experiment. The increasing pseudo-inductive behaviour can be explained by means of a surface relaxation process due to the competitive oxidation of hydrogen and carbon monoxide at the anode.

[1] N. Wagner, *J. Appl. Electrochem.*, **32** (2002) 859

[2] C.A. Schiller, F. Richter, E. Gülzow, N. Wagner, *Phys. Chem. Chem. Phys.*, **3** (2001) 374