On the Effects of Irradiation during X-Ray Imaging of PEFC

J. Eller[1)], J. Roth[1)], F. Marone[2)],
M. Stampanoni[2)], F.N. Büchi[1)]
[1)] Electrochemistry Laboratory, [2)] Swiss Light Source
Paul Scherrer Institut
CH-5232 Villigen PSI, Switzerland

X-Ray imaging, either with radiography for 2D- or tomography for 3D-information has become a quite popular tool for characterizing the liquid water phase in the gas diffusion layer of polymer electrolyte fuel cells (PEFC) [1-5]. Mostly synchrotron radiation (SR) with high brilliance is used to achieve adequate temporal resolution. The photon energies used are in the range between about 10 and 30 keV.

In this range of photon energies, the materials of interest, carbonaceous materials and water are weakly absorbers while the noble metal catalyst, usually platinum, is a strong absorber. As photoelectric ionization is the main mode of interaction in the used photon energy range, effects on the materials in the PEFC should be considered. The ejected photoelectrons ionize further atoms, creating a cascade of secondary electrons and enabling radical formation.
In particular the polymers, viz. the ionomer in membrane and the catalyst layer as well as PTFE (hydrophobic agent in GDL and binder in catalyst layer) are expected to be susceptible to modification by radiation [6].

Even though data on the effects of X-ray radiation on PEFC during imaging [7, 8] and spectroscopic analysis [9] exist, still the mechanism of damage is unclear and a number of questions prevail, such as the electrochemical mechanism of cell performance degradation, the nature of recovery phenomena, the role of strongly absorbing platinum, or the importance of the beam intensity.

In-situ experiments were performed at the TOMCAT beamline of the Swiss Light Source (SLS) at an energy of 13.5 keV. To judge on the influence of X-ray intensity the flux density was varied between 6×10^{9} and 6×10^{11} ph/(s mm^2). Experiments were performed with cells where the entire active area was in the beam and the electrochemical performance was closely followed during irradiation. The release of fluoride and sulfate in the product water and from ex-situ exposed catalyst coated membrane (CCM) samples was analyzed by ion chromatography. This experimental design allows for assessment of the effects of radiation on the electrochemical properties.

During SR exposure of running PEFCs a clear loss of cell performance was observed. The performance degradation was depending the cell's operation conditions (current density, cell temperature, feed gas humidification). The degradation was stronger for MEA orientation perpendicular than parallel to SR orientation, but independent of beam intensity up to an accumulated X-ray energy of 1.5 J/cm^2 in the CCM. Electrochemical impedance spectroscopy measurements revealed a pronounced increase of cathode charge transfer resistance and a stable or slightly decreasing bulk membrane proton conductivity. At specific circumstances (high current densities and accumulated irradiation energy in excess of 1.5 J/cm^2) SR exposure led to temporary HFR increase and/or temporary increase of cell voltage.

Overall, PEFC setups with MEA orientation always parallel to SR (through-plane radiography conditions) are preferable for XTM investigation in terms of SR resistivity due to self-shielding of the catalyst layer in such a configuration. Nevertheless, careful consideration of radiation dose must be considered to prevent material degradation and fuel cell performance in case of application any X-ray source as diagnostic tool, is it radiography or tomography [7].

1. C. Hartnig, I. Manke, R. Kuhn, N. Kardjilov, J. Banhart and W. Lehnert, *Applied Physics Letters*, 2008, **92**, 134106.
2. P. Krüger, H. Markötter, J. Haussmann, M. Klages, T. Arlt, J. Banhart, C. Hartnig, I. Manke and J. Scholta, *J. Power Sources*, 2011, **196**, 5250-5255.
3. J. Eller, T. Rosen, F. Marone, M. Stampanoni, A. Wokaun and F.N. Büchi, *J. Electrochem. Soc.*, 2011, **158**, B963-B970.
4. J. Lee, J. Hinebaugh and A. Bazylak, *J. Power Sources*, 2013, **227**, 123-130.
5. T. Rosén, J. Eller, J. Kang, N. I. Prasianakis, J. Mantzaras and F.N. Büchi, *J. Electrochem. Soc.*, 2012, **159**, F536-F544.
6. J. H. O'Donnell, in *The Effects of Radiation on High-Technology Polymers*, American Chemical Society, 1989, vol. 381, ch. 1, pp. 1-13.
7. A. Schneider, C. Wieser, J. Roth and L. Helfen, *J. Power Sources*, 2010, **195**, 6349-6355.
8. J. Roth, J. Eller and F.N. Büchi, *J. Electrochem. Soc.*, 2012, **159**, F449-F455.
9. M. Schulze, M. Lorenz, N. Wagner and E. Gülzow, *Fresenius J. Anal. Chem.*, 1999, **365**, 106-113.