

A Comprehensive Corrosion Study on Metallic Materials for HT-PEFC Application

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High-temperature polymer electrolyte fuel cells (HT-PEFCs) operate in the range of 120-180 °C and thus embrace high electrode kinetics, a facile controllable cooling system and a greater CO tolerance compared to LT-PEFCs. However, they imply the need to employ proton conductive electrolytes like phosphoric acid doped polybenzimidazole membranes. This in turn represents a quite aggressive environment for the components of a fuel cell [1, 2]. For this reason bipolar plates are currently made of graphitic composite materials. Indeed, they underlie moderate degradation due to corrosion phenomena, but otherwise they exhibit low power densities and a high effort of manufacturing. Therefore metallic materials come into play as promising candidates for bipolar plates. They provide some crucial advantages, like enhanced volumetric and gravimetric power density, increased ductility, high mechanical stability, high electronic and thermic conductivity and the capability of low-cost mass production [3, 4]. Though, this challenge would be too easy without a huge “but”! We still need further understandings of the electrochemical corrosion at the interface metal (bipolar plate) and electrolyte (phosphoric acid) at elevated temperatures.

To avoid looking for the Holy Grail in the dark, we primarily focus in the present work on investigation of the fundamental corrosion mechanism in phosphoric acid that depends strongly on passive layer formation of the respective metallic material. This affects the generation of mixed potentials (corrosion potentials), provided that there is no thermodynamically stable oxide layer. Using an electrochemical cell, which was specially designed for these experimental requirements, we analyzed significant parameters affecting corrosion of various metals and stainless steels by means of potentiodynamic measurements coupled with impedance spectroscopy. It concerns, amongst others, the effect of pH, temperature and degree of aeration of phosphoric acid as well as composition, topography and impact of polarization. Additionally, post-mortem characterizations using SEM/EDX, XPS and ICP-OES were carried out in order to investigate the metallic surface and the corresponding dissolved ions in the electrolyte.

Figure 1 exemplarily shows the formation of the protective passivation layer of steel 1.4301, which is attributed to the current decline even beyond 1.0 V in the anodic branch (red and black curve). Also a significant shift of the corrosion potential E_{cor} from -100 mV to 250 mV and a mitigation of the corrosion current I_{cor} by two orders of magnitude is remarkable when avoiding negative polarization (dotted curve). This derives from the absence of the passive layer due to the reduction of the oxide layer as a side reaction at negative polarization. Enhancing the temperature to 130 °C leads to an increase of the

corrosion current I_{cor} from about $4 \cdot 10^{-6}$ to $2 \cdot 10^{-4}$ A·cm⁻² (red curve). For instance, the corrosion rates decrease for alloys containing niobium, copper or molybdenum, while nickel reveals an adverse effect.

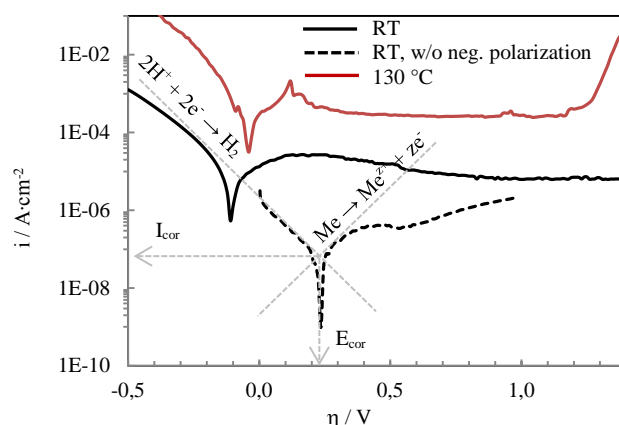


Fig. 1: Tafel plots of stainless steel 1.4301 in phosphoric acid (85 wt.-%) measured against RHE at standard conditions.

The correlation between impedance of the electrochemical interface and polarization at 130 °C is shown as Nyquist diagrams in figure 2. The charge transfer resistance R_D is clearly higher in the potential region from 0.35 V to 1.0 V when a solid oxide layer is built up (black and grey line). By this reason the dissolution of the metal and accordingly the corrosion are inhibited. Notable are the disappeared passive layer and the low R_D at open circuit voltage, which corresponds to E_{cor} (OCV, red semi arc). Negative voltage and an excess of 1.2 V, which are quite destructive for the oxide layer, induce small values for R_D and rising corrosion rates. Fortunately, such relative potentials do not appear in a running fuel cell.

Furthermore, several promising ceramic and graphitic coatings applied by PVD and CVD technique and an in-house sprayed carbon-based protective film are going to be investigated in an analogous way.

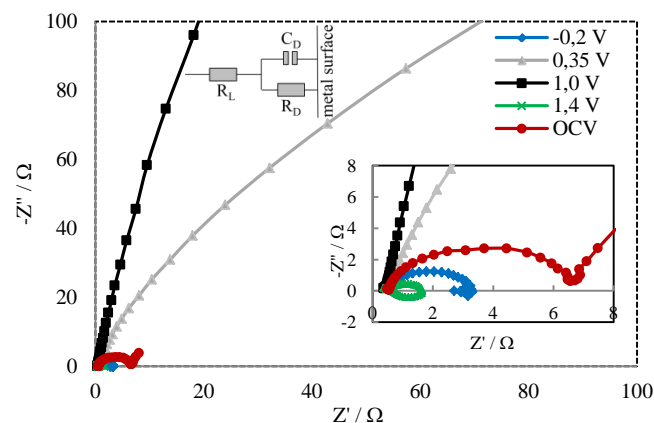


Fig. 2: Nyquist plots of material 1.4301 at 130 °C as a function of applied potential in the range of 100 mHz–100 kHz. Insets show magnification of high frequency region (lower right corner) and electric circuit (above).

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