CO Poisoning in a Polymer Electrolyte Fuel Cell and CO Mitigation by Using Dilute H_2O_2

D.P.J. Barz, A. Frey, V.M. Schmidt

Mannheim University of Applied Sciences
Institute of Environmental Engineering,
D-68163 Mannheim, Germany

Trace CO concentrations in the fuel gas of a polymer electrolyte fuel cell (PEFC) inhibits the electrochemical H_2 oxidation at the anode leading to significant lower cell performances. The H_2 electrooxidation activity in the presence of CO can be expressed as ratio of the current density for pure hydrogen oxidation ($j_{(H2)}$) and that measured in H_2/CO as fuel gas ($j_{(H2/CO)}$)[1]:

$$j_{(H2/CO)} = j_{(H2)}\,(1-\theta_{CO})^2 \quad \text{(at T, p and } U_{cell} = \text{const)} \qquad (1)$$

with the CO coverage θ_{CO} at the fuel cell anode. The other cell parameters should be kept constant.

Current density-voltage curves of a PEFC single cell with Pt as anode and cathode catalysts (with a loading of 0.3 mg cm^{-2} for each electrode) were recorded in H_2 and H_2/CO with 20, 50, 100, 250, 500 and 1000 ppm CO. The cell temperature was 80 °C and the pressure of the fuel gas and the oxidant (pure O_2) was 0.20. Both gas flows were kept constant at 350 ml min^{-1}.

The CO coverages at constant cell voltages were calculated according to (1). The coverage follows a Langmuir isotherm attaining a constant θ_{CO} of 0.8-0.85 at 1000 ppm CO in H_2 (see Fig. 1).

The cell performance in H_2/CO fuel gases can be enhanced by oxidizing adsorbed CO using H_2O_2 as precursor for active oxygen[2]. Dilute H_2O_2 solutions can be passed through a flow tube reactor in order to decompose H_2O_2 and to produce O_2 [3]. By using defined H_2O_2 concentrations and constant flow rates, the stochiometry λ_{CO} can be adjusted for complete CO oxidation according to the overall reaction:

$$H_2O_2 + CO \rightarrow CO_2 + H_2O \qquad (2)$$

Fig. 2 shows the ratio $j_{(H2/CO)} / j_{(H2)}$ as a function of λ_{CO} for 1000 ppm CO in H_2. At this high CO concentration the CO removal is 40 % at U_{cell}=0.70 V and λ_{CO}=95. However, higher λ_{CO}-values should exceed the flammability limit for O_2/H_2 mixture (ca. 5 % O_2).
For lower CO concentrations it was found that at λ_{CO}=140-160 the CO adlayer is completely removed from the fuel cell anode. Under these conditions the same cell performance in H_2/CO is obtained as in pure hydrogen up to CO concentrations of 500 ppm.

References
1. R.J. Bellows, E.P. Marucchi-Sous, D.T. Buckley, *Ind. Eng. Chem. Res.*, 35 (1996) 1235.
2. J. Divisek, H.-F. Oetjen, V.M. Schmidt, US Patent 6,068,943 (May 30, 2000).
3. D.P.J. Barz, V.M. Schmidt, *Phys.Chem. Chem. Phys.*, 3 (2001) 330.

Figure 1: CO coverages at the fuel cell anode as function of CO concentration at two cell voltages; Symbols: experimentals values obtained following (1); solid lines: fit according Langmuir Isotherm.

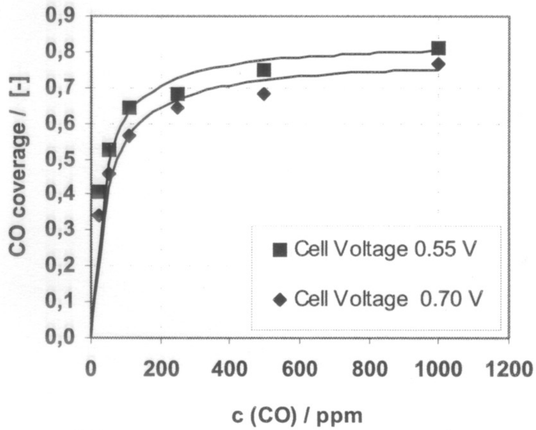

Figure 2: $j_{(H2/CO)} / j_{(H2)}$ as function of the CO stochiometry (λ_{CO}-value); cell voltage: 0.7 and 0.8 V; fuel gas: H_2/1000 ppm CO; 5 % wt H_2O_2 in H_2O for CO oxidation.