Experimental Analysis of the Effects of CO and CO2 on High Temperature PEM Fuel Cell Performance using Electrochemical Impedance Spectroscopy

Andreasen, Søren Juhl; Vang, Jakob Rabjerg

Published in:
Progress in MEA Materials for Medium and High Temperature Polymer Electrolyte Fuel Cells - Book of abstracts

Publication date:
2010

Document Version
Accepted author manuscript, peer reviewed version

Citation for published version (APA):
Andreasen, S. J., & Vang, J. R. (2010). Experimental Analysis of the Effects of CO and CO2 on High Temperature PEM Fuel Cell Performance using Electrochemical Impedance Spectroscopy. In D. Jones (Ed.), *Progress in MEA Materials for Medium and High Temperature Polymer Electrolyte Fuel Cells - Book of abstracts* University Montpellier II.

AALBORG UNIVERSITY

Experimental analysis of the effects of CO and CO_2 on High Temperature PEM Fuel Cell Performance using Electrochemical Impedance Spectroscopy

Søren Juhl Andreasen, Jakob Rabjerg Vang and Søren Knudsen Kær*

Department of Energy Technology, Aalborg University, Pontoppidanstræde 101, 9220 Aalborg East, Denmark

Introduction

This work presents the results of using electrochemical impedance spectroscopy to analyse the behaviour of a BASF Celtec P2100 MEA operated under varying operating conditions with different temperatures and gas concentrations. Figure 1 shows the experimental setup used for these measurements.

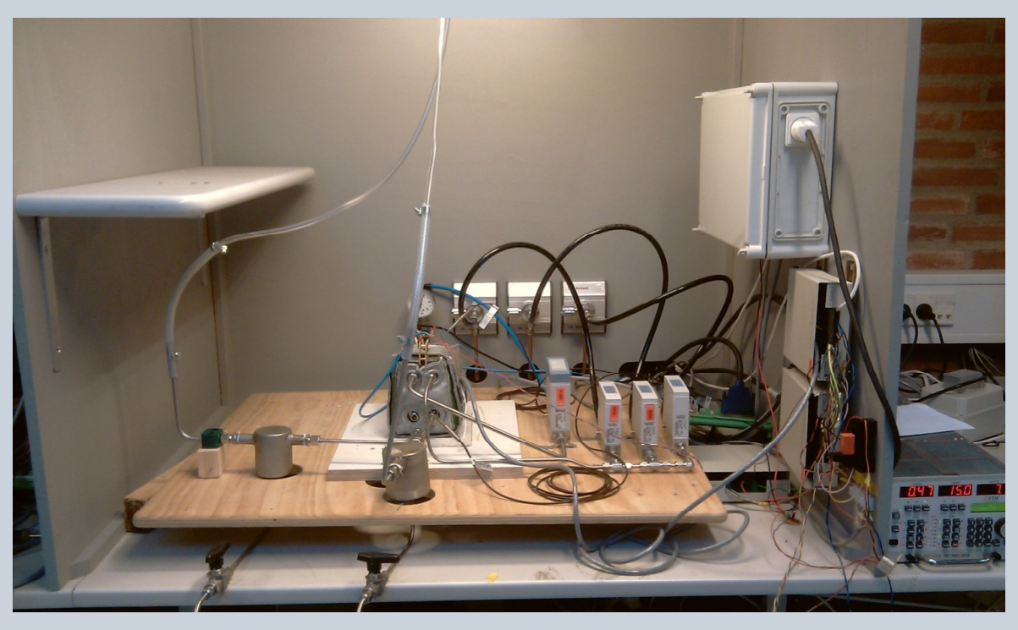

Figure 1: Single cell setup capable of mixing different concentrations of H_2, CO and CO_2 to simulate reformate gas performance .

The setup uses two separate Labview Real Time systems, one for controlling the fuel cell mass flows and temperature, and one for conditioning the current and interpreting the fuel cell voltage response to the galvanostatic impedance measurements. The impedance measurement results are interpreted and converted into Nyquist plots, as shown in figure 2.

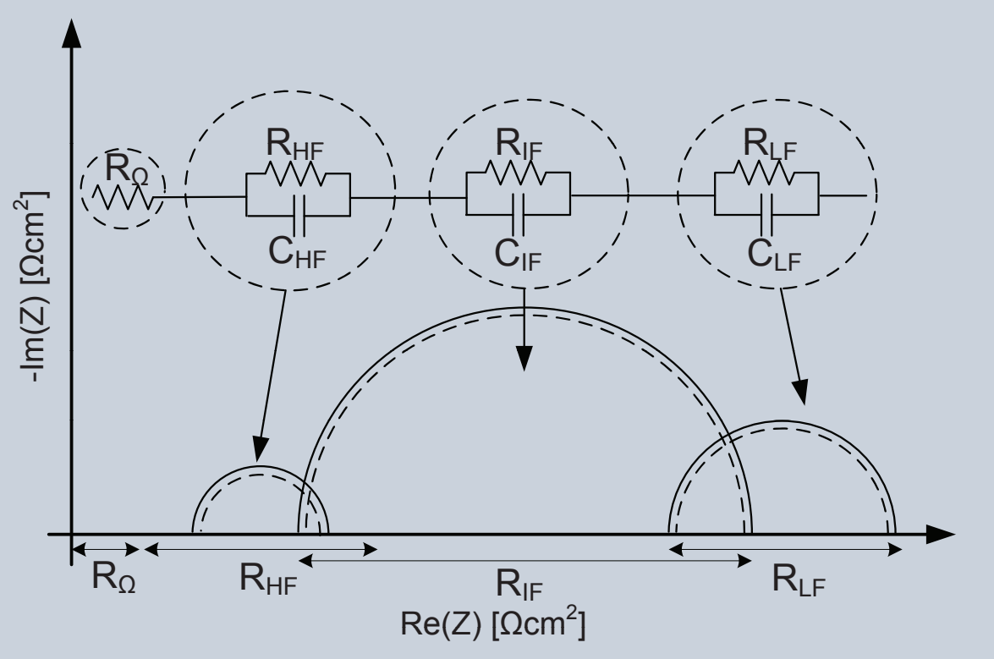

Figure 2: Nyquist plot of the shown electrical circuit. Subscripts referring to high, intermediate or low frequency (HF,IF and LF), and Ω to the ohmic losses of the fuel cell.

The results can be used to construct simple equivalent circuit models or used to validate more complex physical models and give insight in the dynamic processes occurring in the different layers of the fuel cell. Figure 3 presents the different operating conditions under which the automated measurement system has tested the BASF MEA.

M. No.	T_{FC} [°C]	I_{FC} [A]	y_{CO} [%]	y_{CO_2} [%]
0	100	5	0	0
1	100	10	0	0
2	100	15	0	0
3	110	5	0	0
4	110	10	0	0
5	110	15	0	0
6	120	5	0	0
7	120	10	0	0
8	120	15	0	0
9	130	5	0	0
10	130	10	0	0
11	130	15	0	0
12	140	5	0	0
13	140	10	0	0
14	140	15	0	0
15	150	5	0	0
16	150	10	0	0
17	150	15	0	0
18	160	5	0	0
19	160	10	0	0
20	160	15	0	0
21	170	5	0	0
22	170	10	0	0
23	170	15	0	0
24	180	5	0	0
25	180	10	0	0
26	180	15	0	0
27	180	10	0.15	0
28	180	10	0.15	25
29	180	15	0.15	0
30	180	15	0.15	25
31	160	5	0	0
32	160	10	0.15	0
33	160	10	0.15	25
34	160	15	0.15	0
35	160	15	0.15	25
36	140	5	0	0
37	140	10	0.15	0
38	140	10	0.15	25
39	140	15	0.15	0
40	140	15	0.15	25
41	120	5	0	0
42	120	10	0.15	0
43	120	10	0.15	25
44	120	15	0.15	0
45	120	15	0.15	25
46	100	5	0	0
47	180	5	0	0
48	180	10	0.25	0
49	180	10	0.25	25
50	180	15	0.25	0
51	180	15	0.25	25
52	160	5	0	0
53	160	10	0.25	0
54	160	10	0.25	25
55	160	15	0.25	0
56	160	15	0.25	25
57	140	5	0	0
58	140	10	0.25	0
59	140	10	0.25	25
60	140	15	0.25	0
61	140	15	0.25	25
62	180	5	0	0
63	180	10	0.5	0
64	180	10	0.5	25
65	180	15	0.5	0
66	180	15	0.5	25
67	160	5	0	0
68	160	10	0.5	0
69	160	10	0.5	25
70	160	15	0.5	0
71	160	15	0.5	25
72	140	5	0	0
73	140	10	0.5	0
74	140	10	0.5	25
75	140	15	0.5	0
76	140	15	0.5	25
77	180	5	0	0
78	180	10	1	0
79	180	10	1	25
80	180	15	1	0
81	180	15	1	25
82	160	5	0	0
83	160	10	1	0
84	160	10	1	25
85	160	15	1	0
86	160	15	1	25
87	140	5	0	0
88	140	10	1	0
89	140	10	1	25
90	140	15	1	0
91	140	15	1	25
92	180	10	1	0
93	140	10	1	25

Figure 3: Table of the different operating points examined in this work, using different combinations of DC currents, gas concentrations and temperatures.

HTPEM Fuel Cell Electrochemical Impedance Spectroscopy

Automated electrochemical impedance spectroscopy measurements have resulted in a full map of the impedance of a BASF Celtec P2100 HTPEM fuel cell at different temperature (se figure 4 and 6). The polarisation curves created before each of the EIS measurements is seen in figure 5.

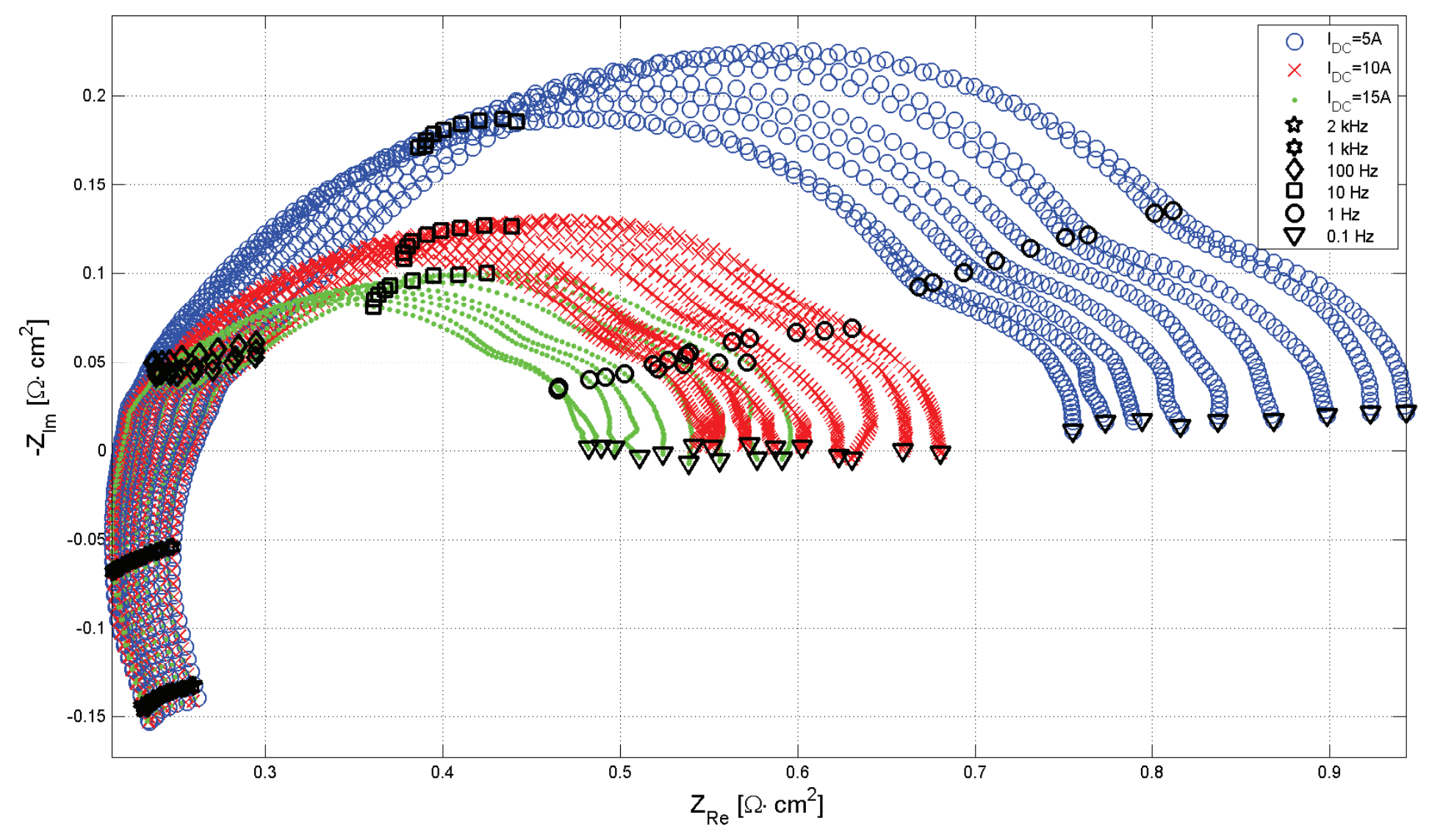

Figure 4: Nyquist plot of fuel cell impedance at different temperatures and DC currents.

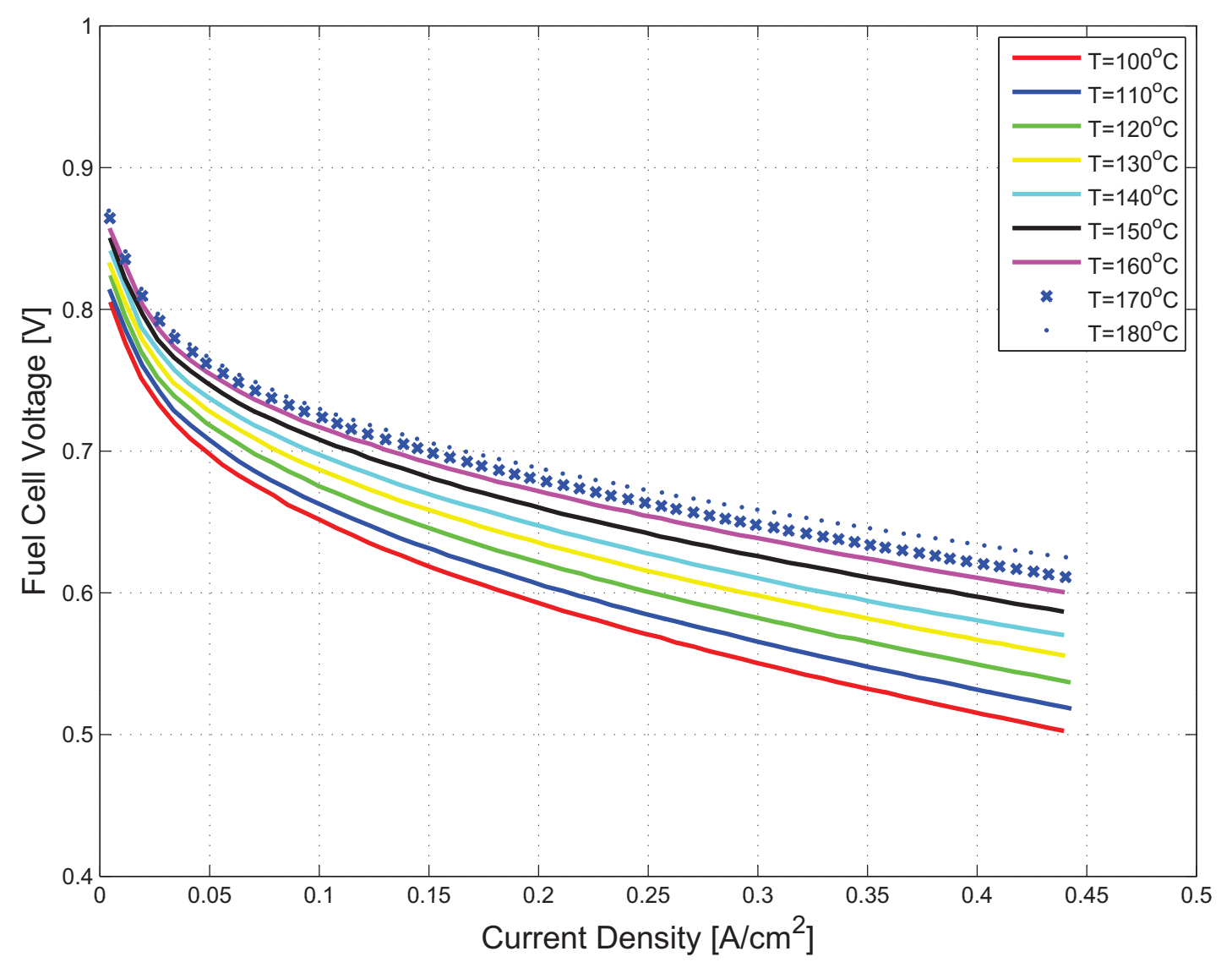

Figure 5: Polarisation curve using pure hydrogen at different temperatures.

Varying temperatures increase the conductivity of the MEA, clearly depicted in the impedance at high frequencies, but also the intermediate and low frequency performance is changed due to expected effects occurring in the catalyst and diffusion layers. The effect of adding CO to the anode gas immediately increases the ohmic losses of the fuel cell, but due higher local temperatures because of higher expected activation losses, increasing the CO concentration losses lower these losses (figure 7).

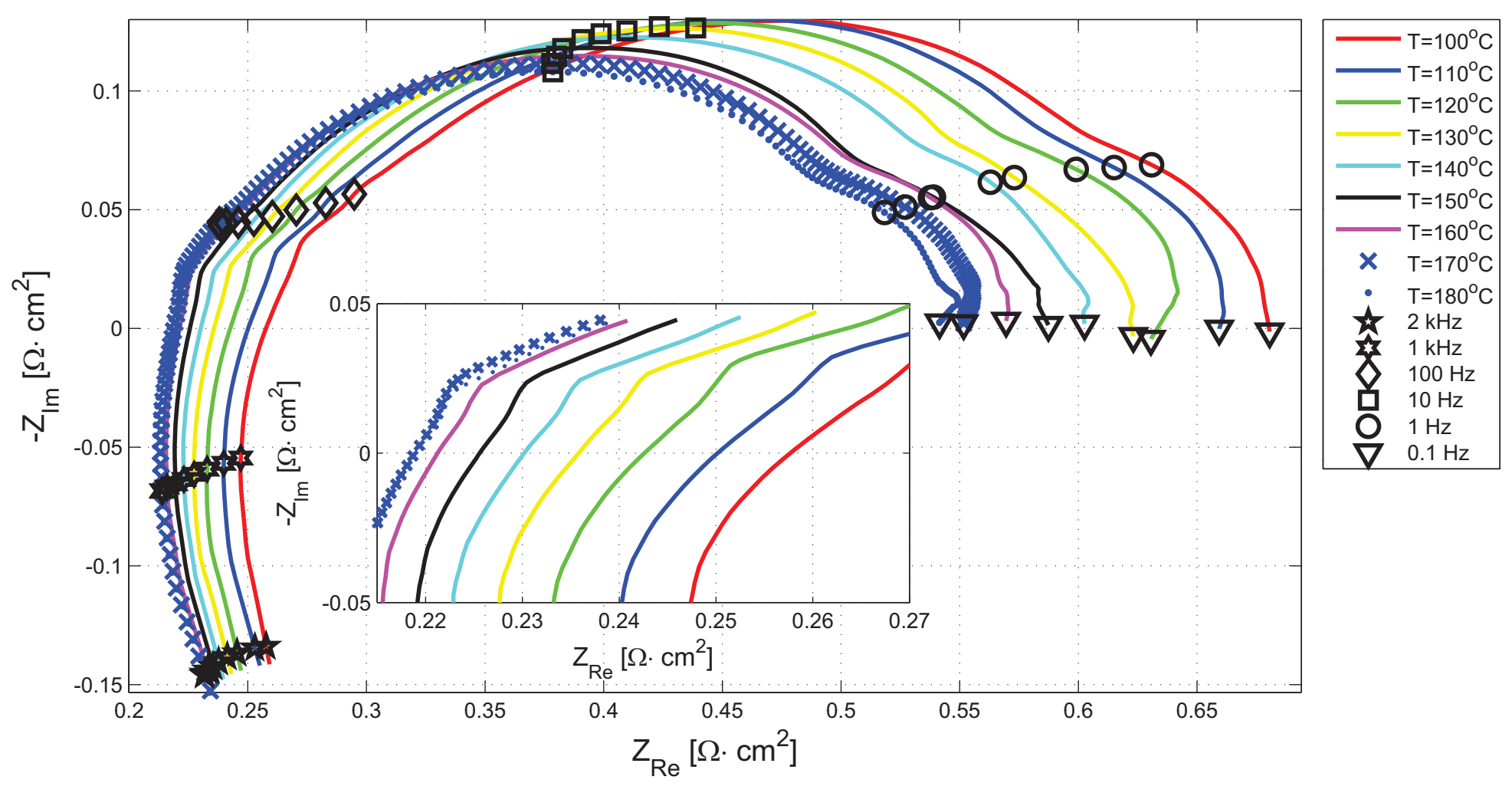

Figure 6: Nyquist plot of fuel cell impedance at 10A DC current and different temperatures.

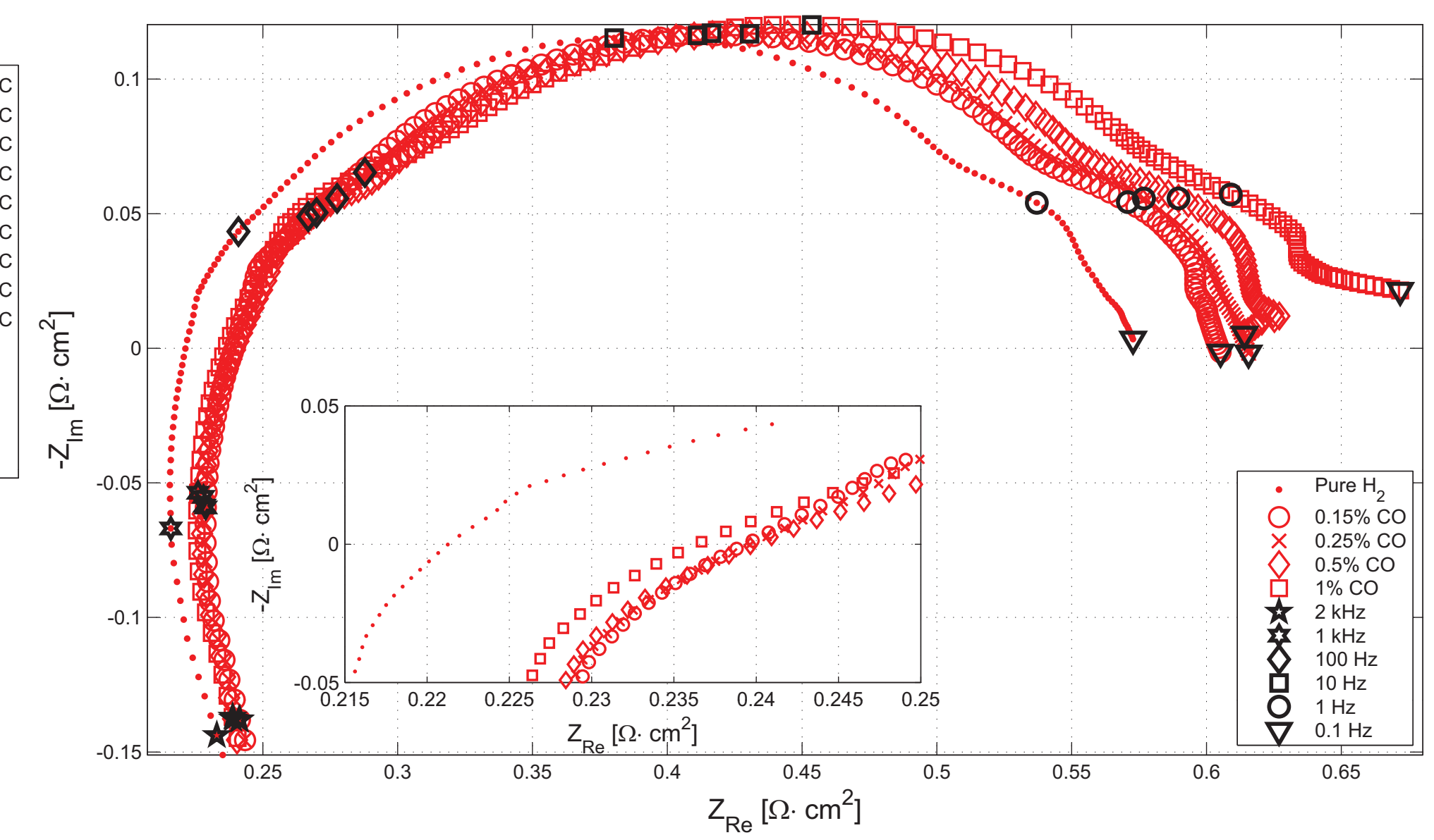

Figure 7: Nyquist plot of fuel cell impedance at 160 °C and different CO concentrations.

Nyquist plots comparing the impedance at different temperatures and adding CO and CO_2 to the anode gas reveals combined effects of increasing losses and additional cooling due to higher gas flows. The additional activation losses are suggested to increase local temperatures in the cell which in turn shift the Nyquist plot to the left. The additional CO_2 increases the cell cooling and greatly affects the fuel cell at temperatures (see figure 9). The affect of adding CO_2 on the polarisation curve of the fuel cell is shown in figure 8.

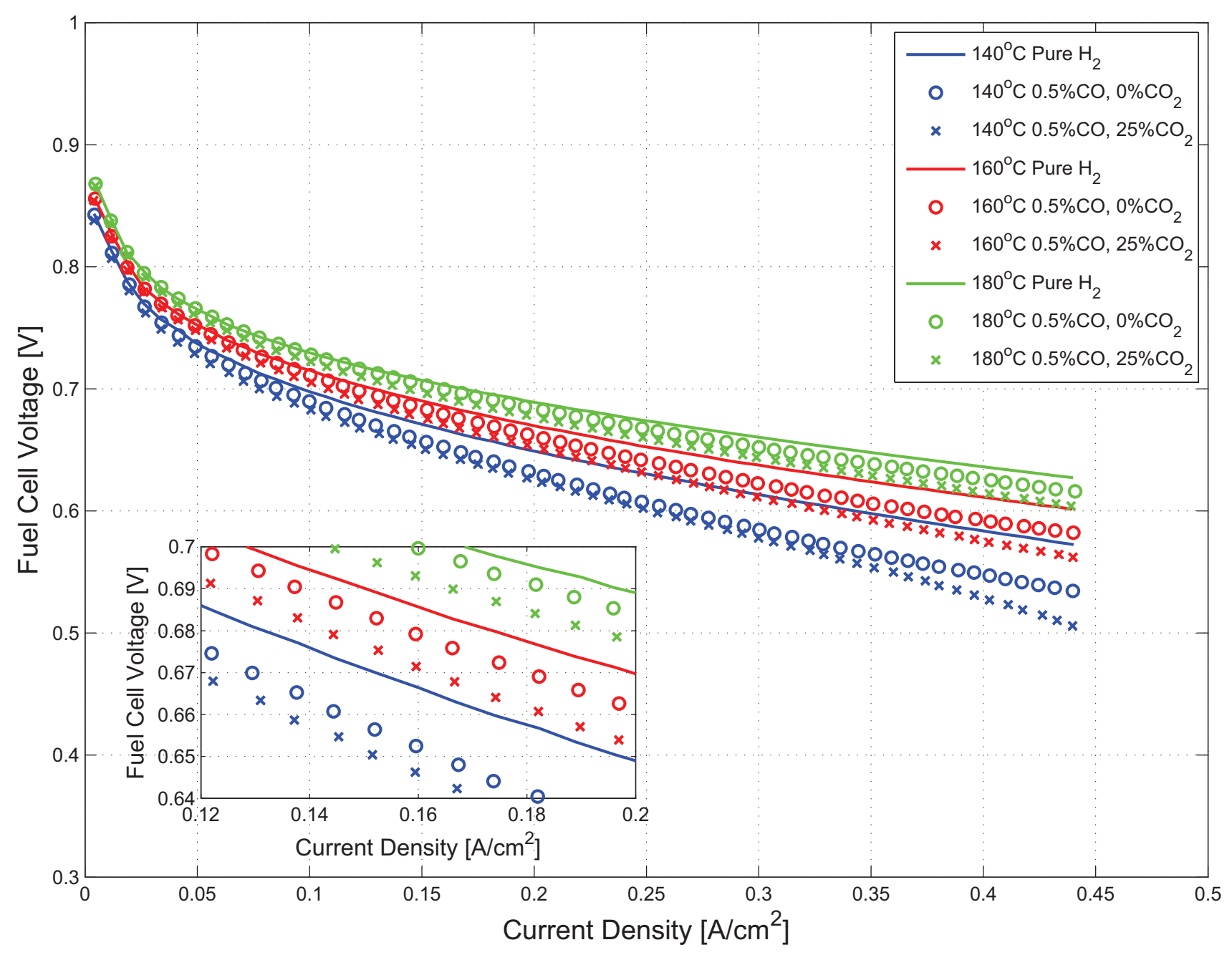

Figure 8: Polarisation curve using 0.5% CO at different temperatures with and without CO2.

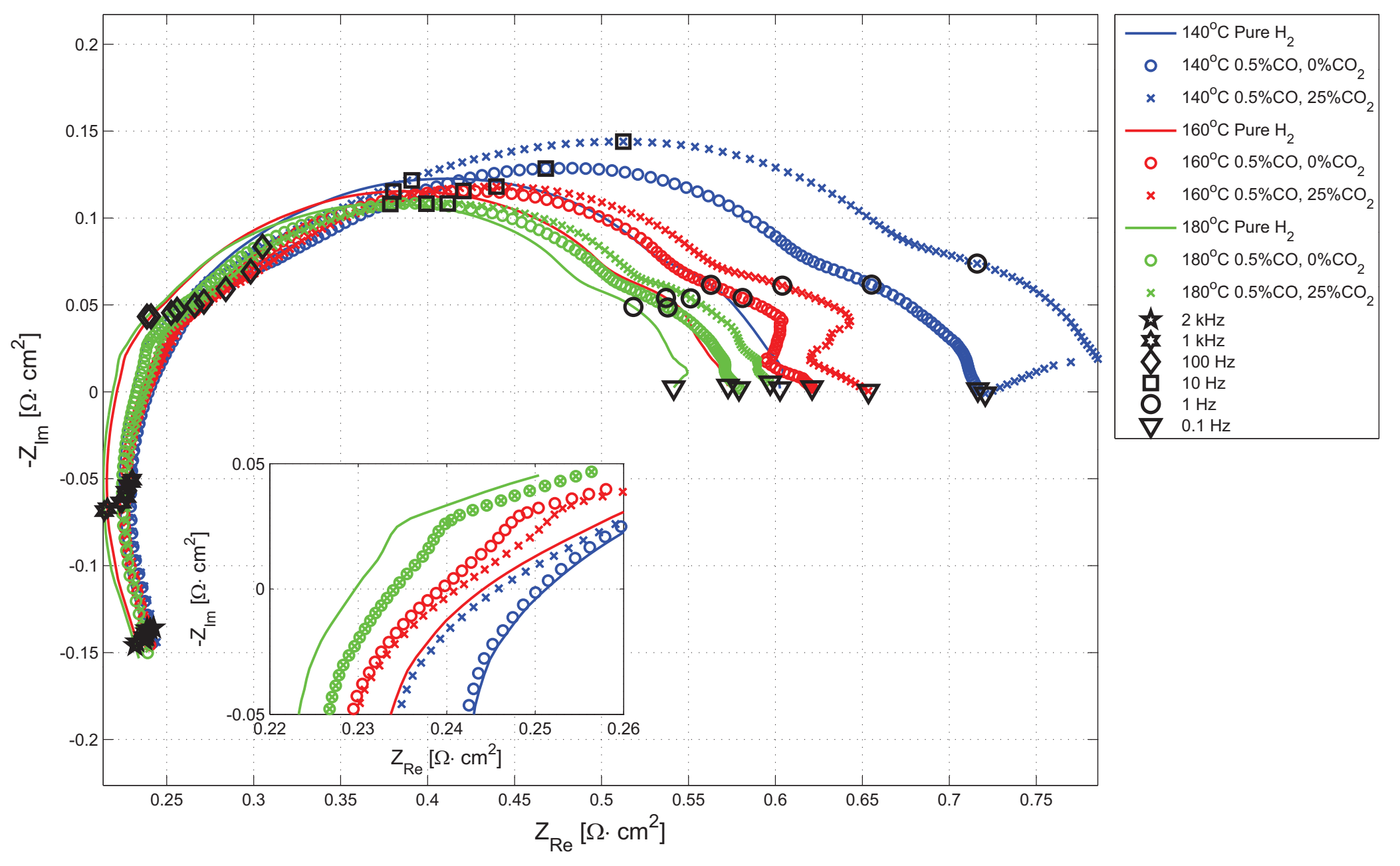

Figure 9: Nyquist plot of fuel cell impedance at 10A DC current, 0.5% CO, different temperatures with and without CO_2.

The different operating conditions and their effect on the electrical fuel cell performance can be modelled by using simple equivalent electrical circuit models. Figure 10 shows the fitted parameters of the layout in figure 2 to the measurement conducted at different temperatures.

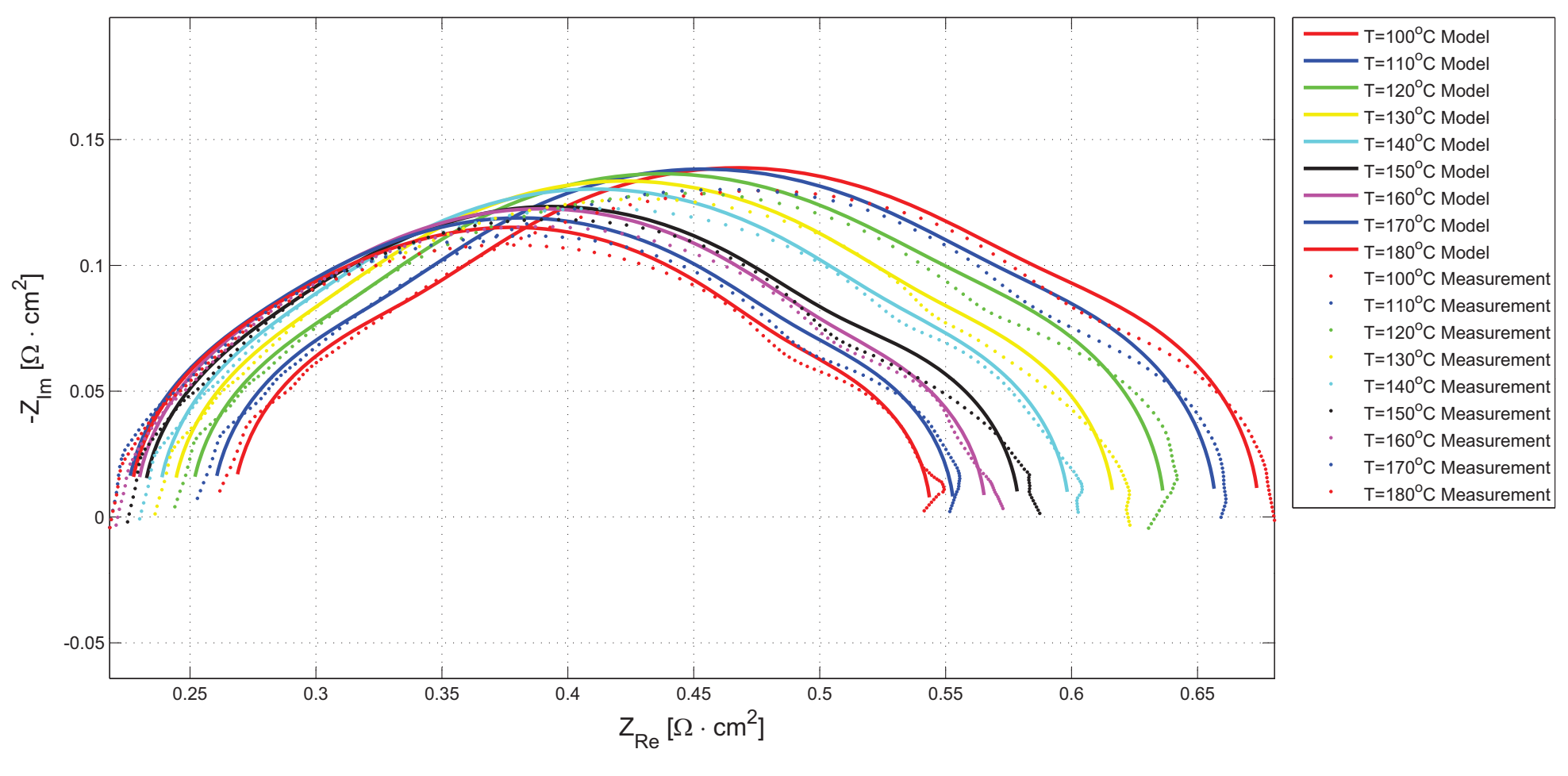

Figure 10: Nyquist plot of measurements and fitted equivalent circuit model corresponding to the model shown in figure 2 at varying temperatures.

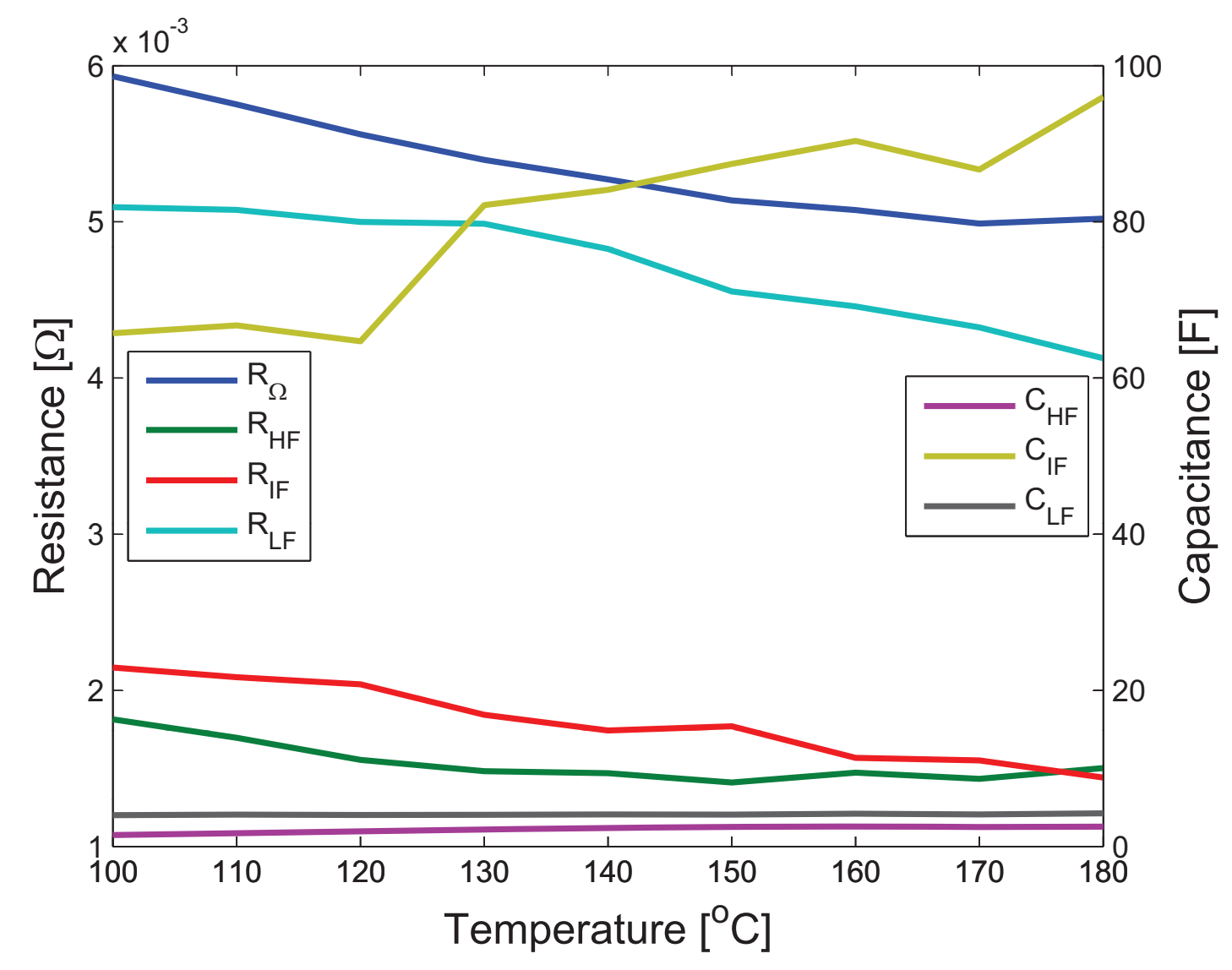

Figure 11: Plot of how the fitted equivalent circuit parameters vary with changing temperatures.

In figure 11 the variation in resistances and capacitances of the equivalent circuit is presented as a function of operating temperature.

Conclusions

Using electrochemical impedance spectroscopy, a BASF P2100 has been characterized in different operating points with varying temperature and gas concentrations. The developed experimental setup is designed such that measurements are consistent and reproducible. Increasing the number of measurements in each operating point increases the reliability of the conclusions drawn from these measurements.

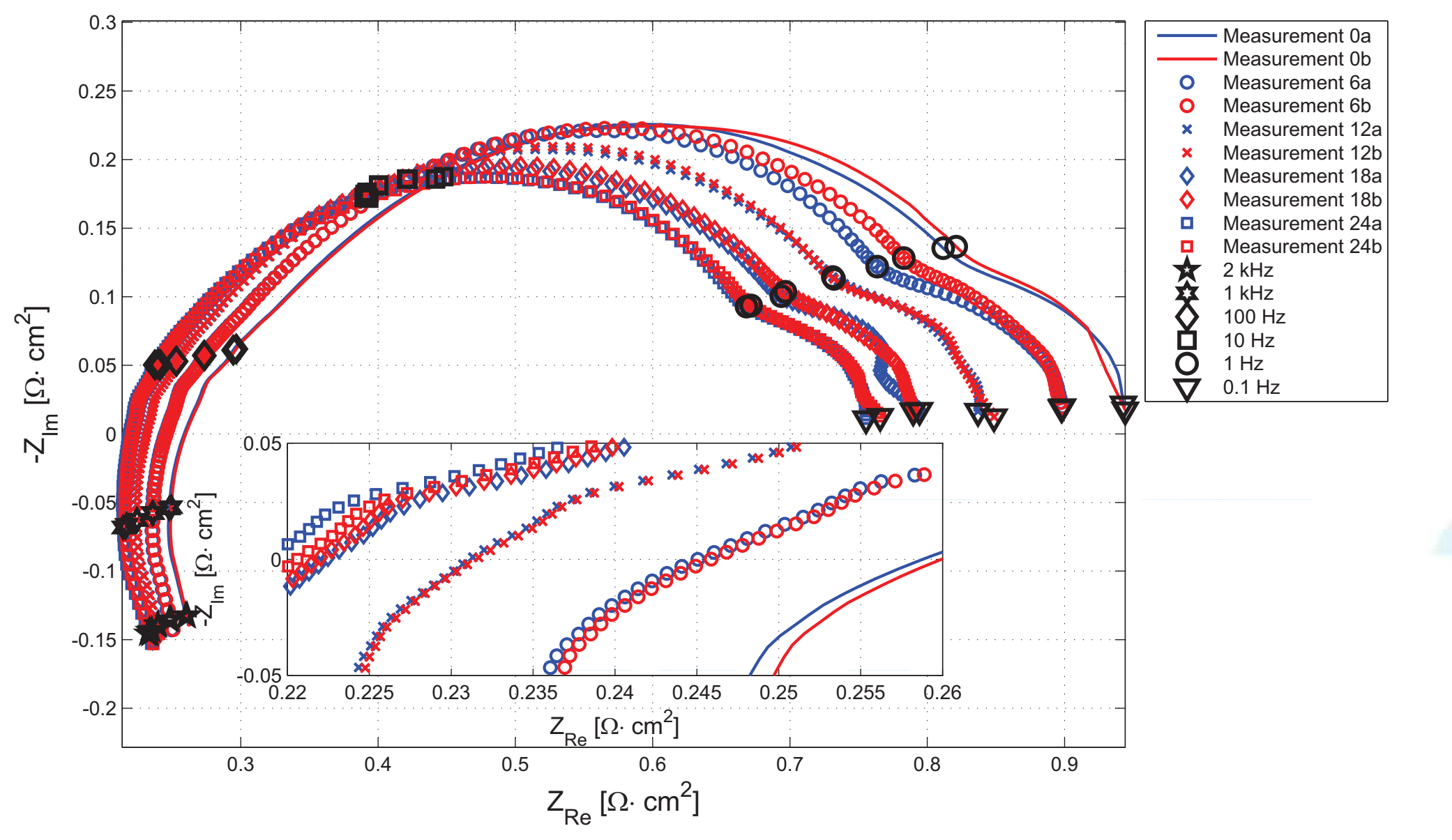

Figure 12: Nyquist plot of different measurements, with 2 consecutive measurements in each operating point.

The measurements conducted on pure hydrogen with varying temperatures, show decreasing high frequency resistances with increasing temperatures, which was also expected, due to the better proton conductive abilities of the membrane at higher temperatures.

When introducing CO to the anode gas, a higher high frequency resistance is visible, this is expected to be due to the higher activation losses because of the additional CO. When increasing the CO content this high frequency resistance surprisingly decreases, which is expected to be due to increasing local temperatures because of the additional activation losses involved with the CO adsorption/desorption processes.

When adding CO_2 to the anode gas at the concentrations found in the output stream of a steam reformer, the DC resistance is also effected. Careful analysis of the Nyquist plot of these measurements suggest that multiple phenomena affect the impedance of the fuel cell. At 180℃ and 160℃ the high frequency impedance is lower when CO_2 is absent. At 140 ℃ and 120℃ the additional CO_2 decreases this resistance. This effect is expected to be a combination of losses and locally higher temperatures due to CO and increased cooling due to the high flow of CO_2.

The experimental results gained from the electrochemical impedance spectroscopy measurements, can be used to model the nonlinear behaviour of the fuel cell as a function of temperature, CO and CO_2 content.

Future Work

Future work with this fuel cell setup will include scheduled testing during the break-in process, identifying the parameters changing during break-in, and also evaluating new break-in methods, that could speed up the sometimes time consuming break-in process. Future work also includes more detailed physical models that describe the impedance behaviour in more detail than equivalent circuit models. Developments of implementing this method on operating fuel cells for state-of-health diagnosis could provide an efficient way of giving system control inputs for optimal system performance.

www.et.aau.dk

***sja@et.aau.dk**