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Introduction

Degradation is an important topic as it is one of the main issues hindering the commercial break though of high temperature proton exchange membrane fuel cells (HT-PEMFC). In this work, a literature survey on the HT-PEMFC degradation mechanisms was done and a lifetime prognosis model that can help predict degradation patterns was prepared based on literature data. Literature data on Pt particle agglomeration and Pt loss was used to model the degradation due to activation loss and data on membrane conductivity loss due to phosphoric acid loss was used to simulate the ohmic losses. The concept of modeling build map is shown in Fig 1.

Model Specification

In this work we only focus on the activation loss and ohmic losses. The resulting cell voltage is obtained by subtracting the losses from the Nernst potential:

$$E_{cell} = E - \eta_{act}(t) - \eta_{ohm}(t) = E - A\ln\left(\frac{i}{i_0\, a(t)}\right) - \frac{\delta_m}{\sigma_m(t)}\, i \qquad (1)$$

$$a(t) = \frac{m_{active}(t)}{A_{geo}} \frac{3}{\rho_{Pt} \frac{d(t)}{2}} \qquad (2)$$

where α, δ_m, σ_m, m_{active}, A_{geo}, ρ_{Pt} and d are the degradation factor, membrane thickness [cm], membrane conductivity [S/cm], active mass of Pt [g], geometric area [cm^2], density of Pt particle [g/cm^3], and the average diameter of Pt particle cm, respectively.

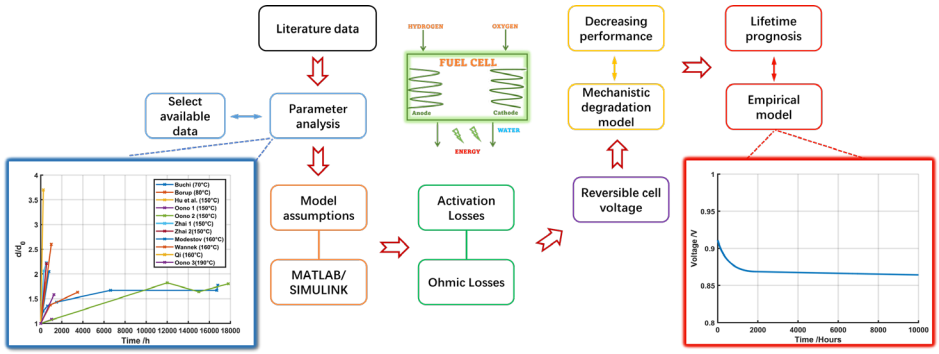

Fig 1. Schematic representation of degradation model

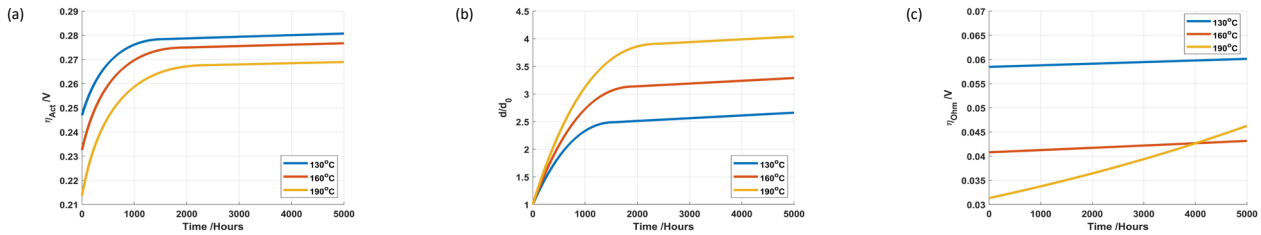

Fig 2. Simulation results of temperature dependency of activation and ohmic losses over time (a) activation overpotential over time, (b) diameter relative to the initial diameter over time and (c) ohmic overpotential over time

Results

- From the Fig 2a, the initial voltage loss decreases with increasing temperature from almost 0.25 V at 130 °C to little more than 0.21 V at 190 °C. Although the difference decreases with time, the higher temperature is always favorable. The reasons are most likely the enhanced reaction kinetics at higher temperatures.
- On the other hand, the Pt particle diameter that describes Pt agglomeration increases with increase in operating temperature (Fig 2b). Therefore, higher operating temperature increases fuel cell performance by increasing the fuel cell reaction kinetics, but at the same time exacerbates Pt agglomeration.
- The evolution of the ohmic overpotential over time for different temperatures can be seen in Fig 2c and shows higher losses at lower temperature at the beginning of life due to better membrane conductivity at higher temperatures.

Conclusion

- The major underlying chemical mechanisms contributing to degradation are identified as loss of electrochemical surface area (ECSA) and loss of conductivity. They are both found to be affected by at least temperature and operation mode.
- Based on analysis on voltage decay rate and lifetime prognosis, the operating temperature range between 160 °C and 170 °C can be said optimal.

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