Electrochemical Impedance Spectroscopy-based Screening of Membrane Effects via Gas Diffusion Electrode Half-Cells for PEMFC Performance Optimization

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Supporting Information contents:

SI 1. Definition and calculation of the ionomer-to-carbon (I/C) ratio

In this work, the ionomer-to-carbon (I/C) ratio is defined volumetrically as the ratio between the volume of PFSA ionomer (V_I) and the volume of the carbon support (V_C) see Equation S1:

$$\frac{I}{C} = \frac{V_I}{V_C} \qquad (1)$$

The volumetric I/C ratio was selected as a formulation parameter because it more directly reflects the spatial ionomer fraction within the catalyst layer, which governs proton transport pathways and ionomer–catalyst contact.

For practical ink preparation, the volumetric I/C ratio was converted into a gravimetric ionomer-to-carbon ratio (m_I/m_C) using the densities of the respective components:

$$\frac{m_I}{m_C} = \frac{V_I}{V_C} \cdot \frac{\rho_I}{\rho_C}$$

where

- m_I and m_C are the masses of ionomer and carbon (g), respectively,
- ρ_I is the density of the dry PFSA ionomer (g/cm^3), and
- ρ_C is the density of the carbon support (g/cm^3).

For the present study, a volumetric I/C value of 0.9 was used for all inks. Using representative density values for PFSA ionomer and carbon black (as commonly reported in the literature), the corresponding gravimetric I/C ratio was calculated and used to determine the required ionomer and catalyst masses. Once the solid masses were fixed, the dispersion composition was adjusted to achieve a total solid content of 10 wt%, with a solvent mixture consisting of 30 wt% water and 70 wt% 1-propanol.

The publicly accessible, peer-reviewed characterization of the same catalyst material, including TEM-based particle size distributions and surface chemical analysis XPS, provided in the Supporting Information of Ref. [35].
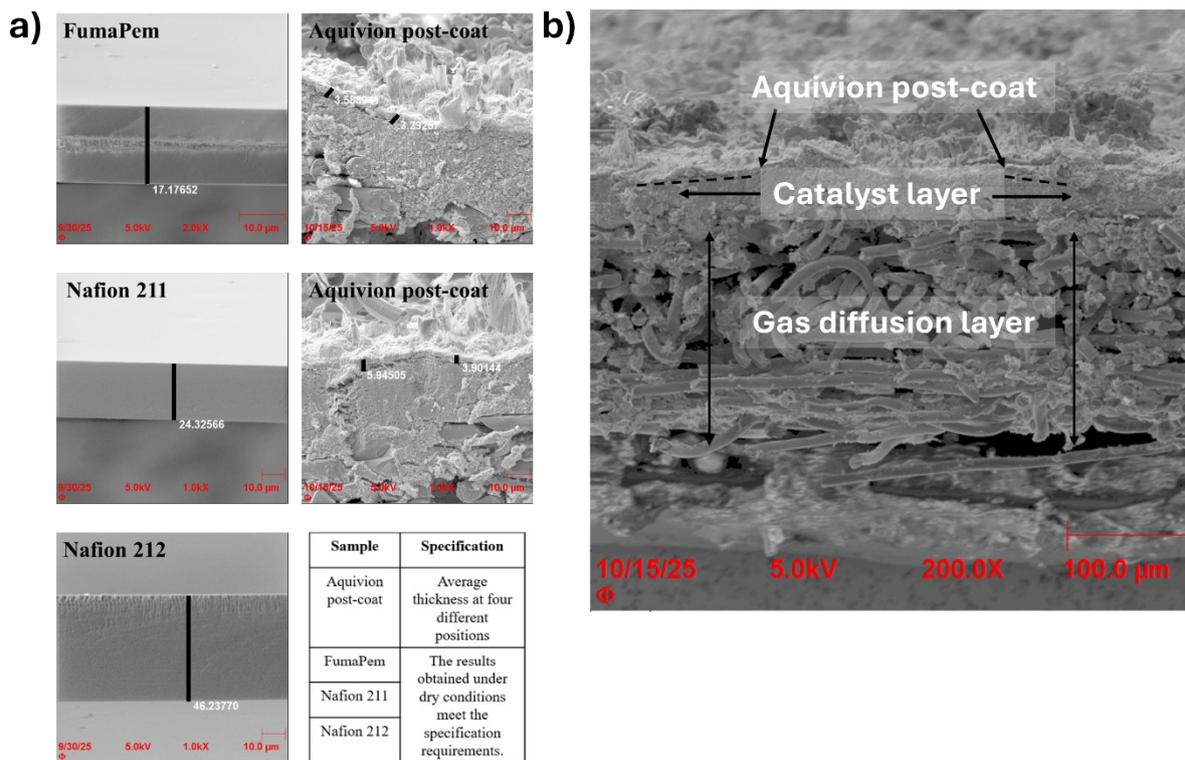
Figure S1: a) FIB-SEM of Aquivion post-coat, FumaPem, Nafion 211 and Nafion 212. b) FIB-SEM of whole dCCS sample with Aquivion post-coat.
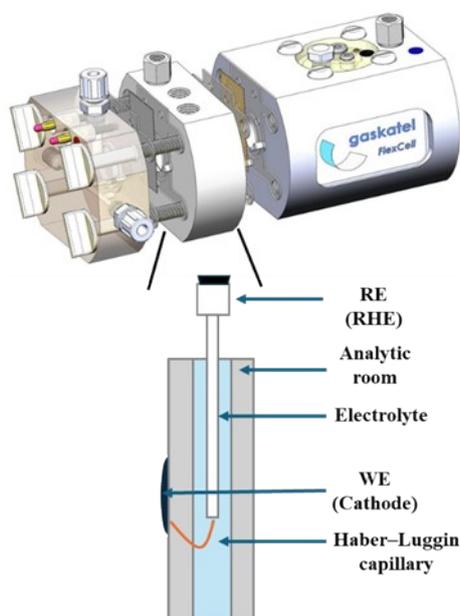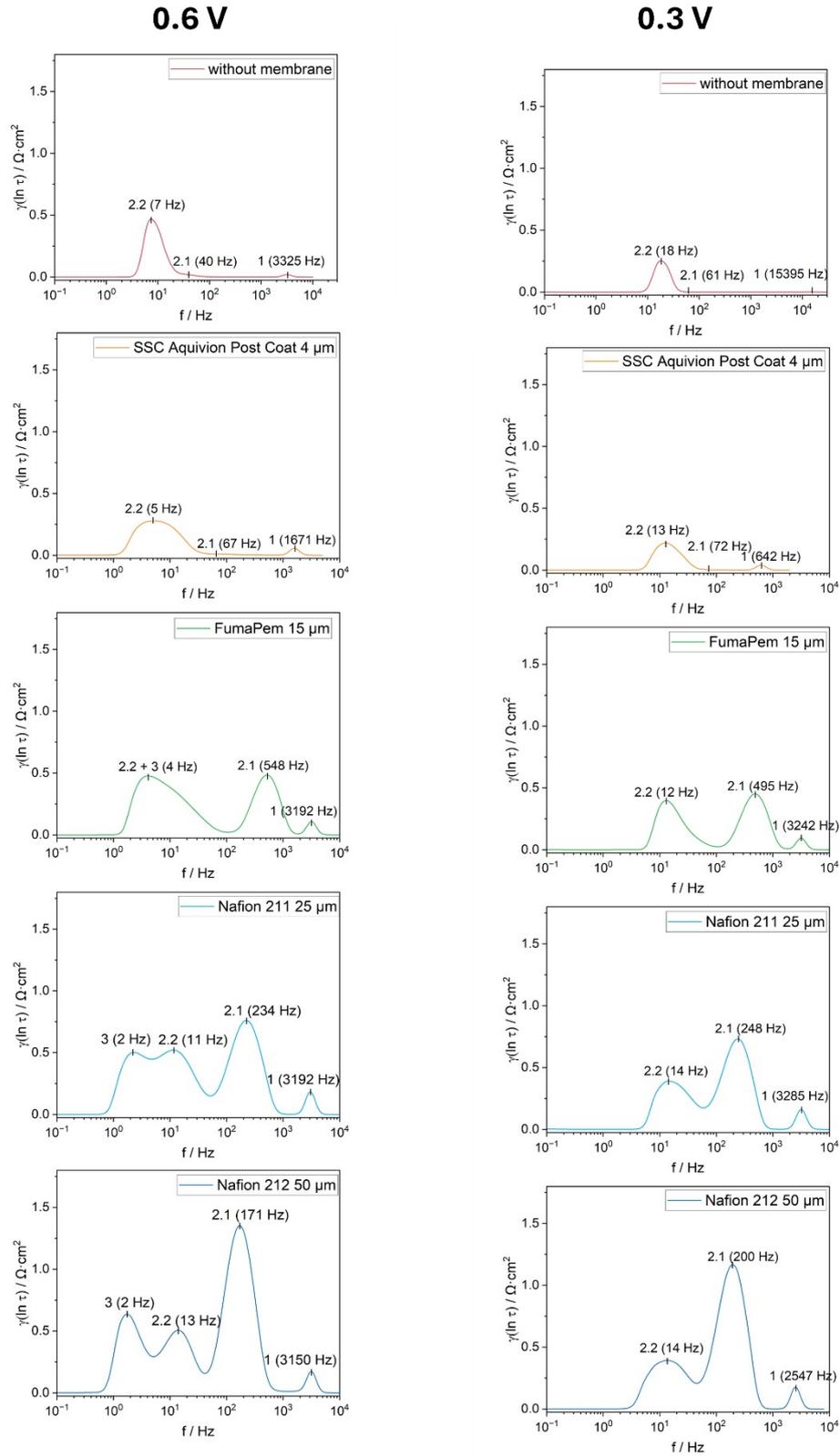

Figure S2: Cross-sectional schematic of the GDE half-cell setup.

Figure S3: DRT peak positions for all samples at 0.6V and 0.3V.