Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supplementary Material

Interfacial Analysis of PEM Electrolyzer Using X-ray Computed Tomography

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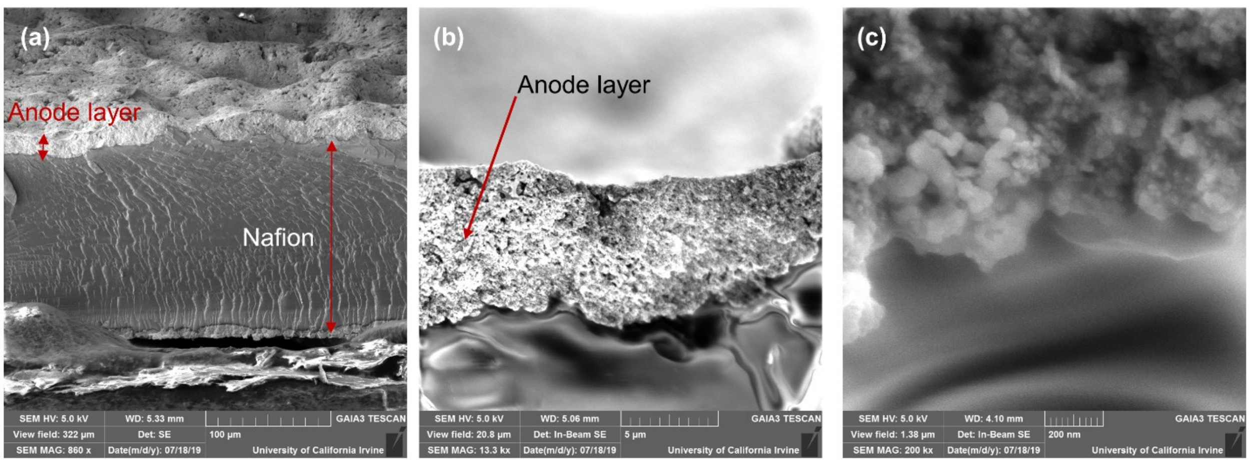

Figure S1: Cross-section SEM images of the anode portion of the CCM at the beginning of life. a) Low-resolution overview, b) zoom-in and c) high-resolution cross-section image.

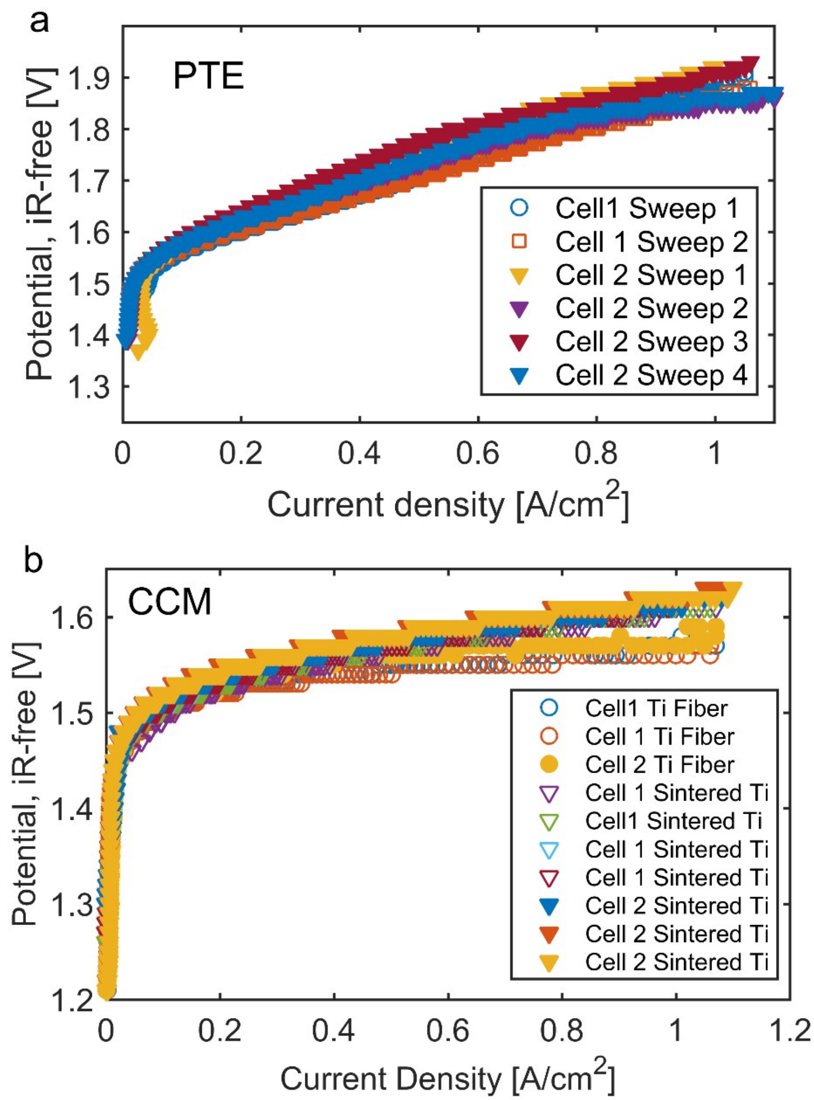

Figure S2: Polarization sweeps for a) PTE and b) CCM configurations for the electrolyzers. The plots show sweeps from 1.4 V to a potential when 1 A/cm^2 is reached at 5 mV/s. Plot b also shows the comparison between Sintered Ti and Ti fiber PTLs.

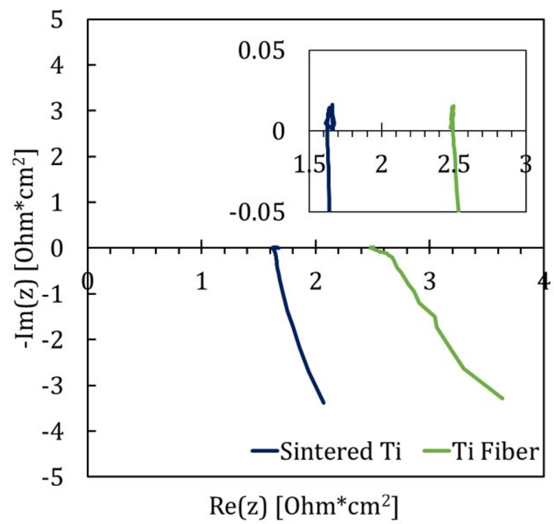

Figure S3: Representative high frequency resistance plotted for the sintered titanium and titanium fiber PTL samples from the EIS tests at the beamtime. (S2, inset) The real axis intersection points for the sintered titanium and titanium fiber samples, respectively.

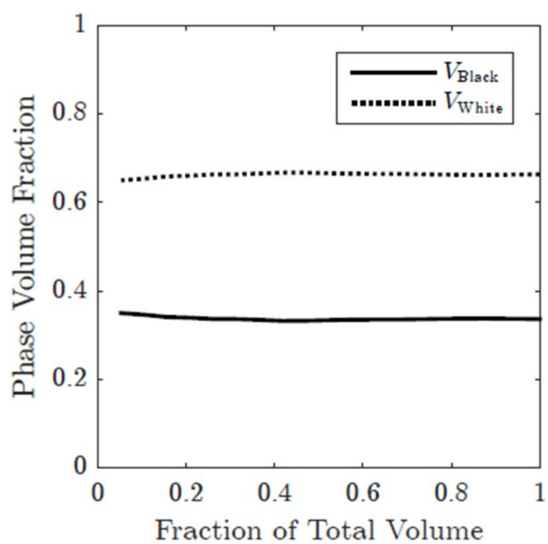

Figure S4: TauFactor representative volume analysis (RVA) with L=constant for the sintered titanium sample. Fraction of total volume equal to 1 is the full sample used for this work. V_{black} is the void volume fraction, V_{white} is the titanium volume fraction. Volume fractions remain constant across the volume sweep with the thru-plane length constant.

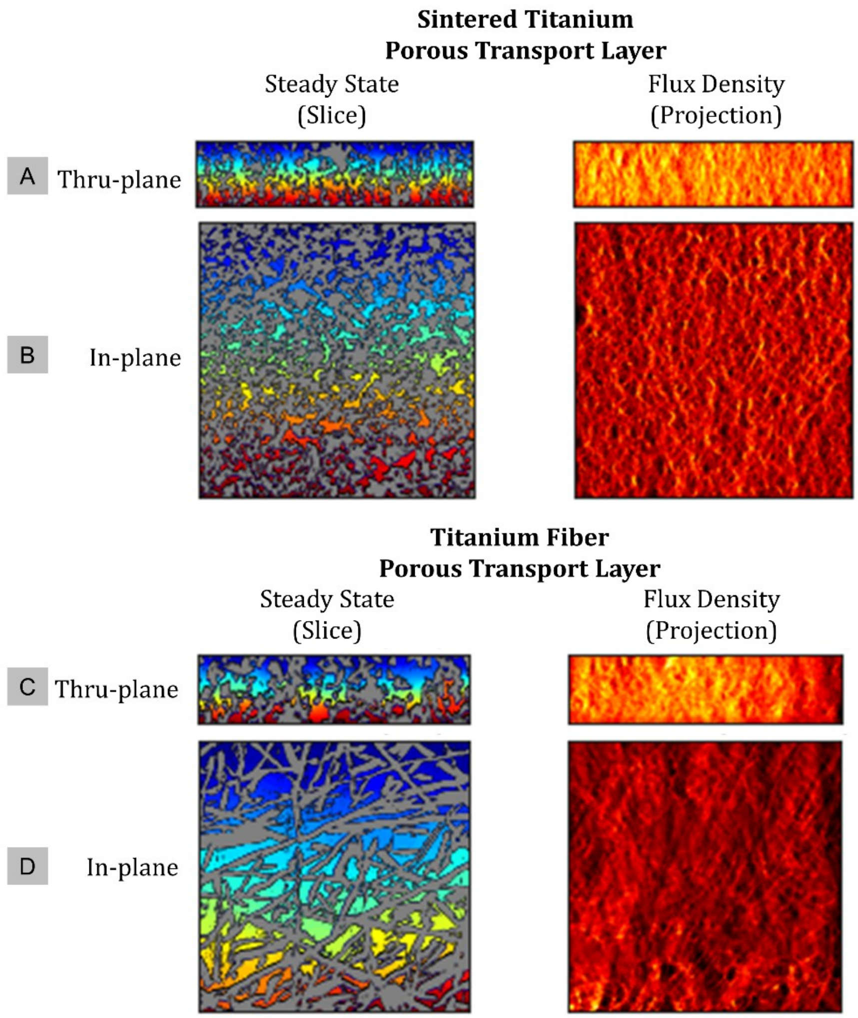

Figure S5: TauFactor results showing the cross section and flux density in the thru-plane for (a) sintered titanium and (c) titanium fiber and in-plane for (b) sintered titanium and (d) titanium fiber. The grey areas in the steady state slice is titanium metal. The flux density is a projection of all the analyzed slices in the two directions. These results come directly from the TauFactor application.

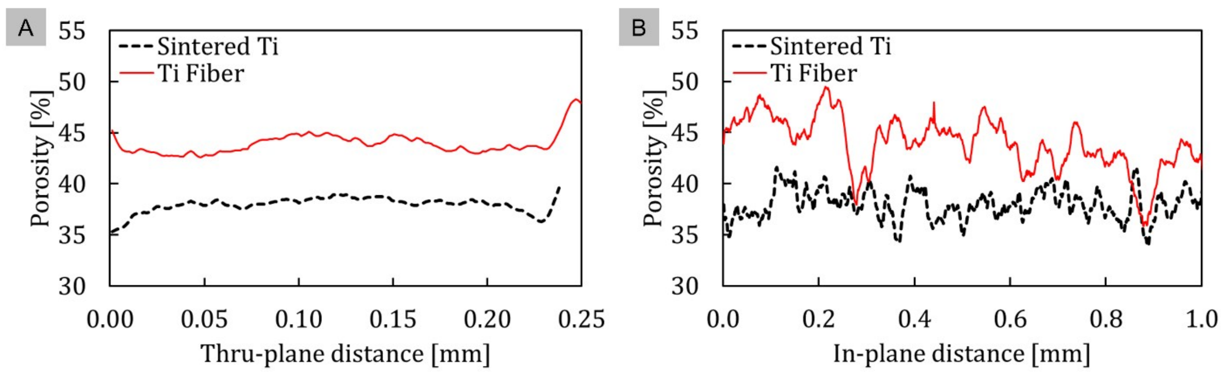

Figure S6: Porosity results in the (a) thru-plane direction and (b) in-plane direction for both the sintered titanium and titanium fiber.

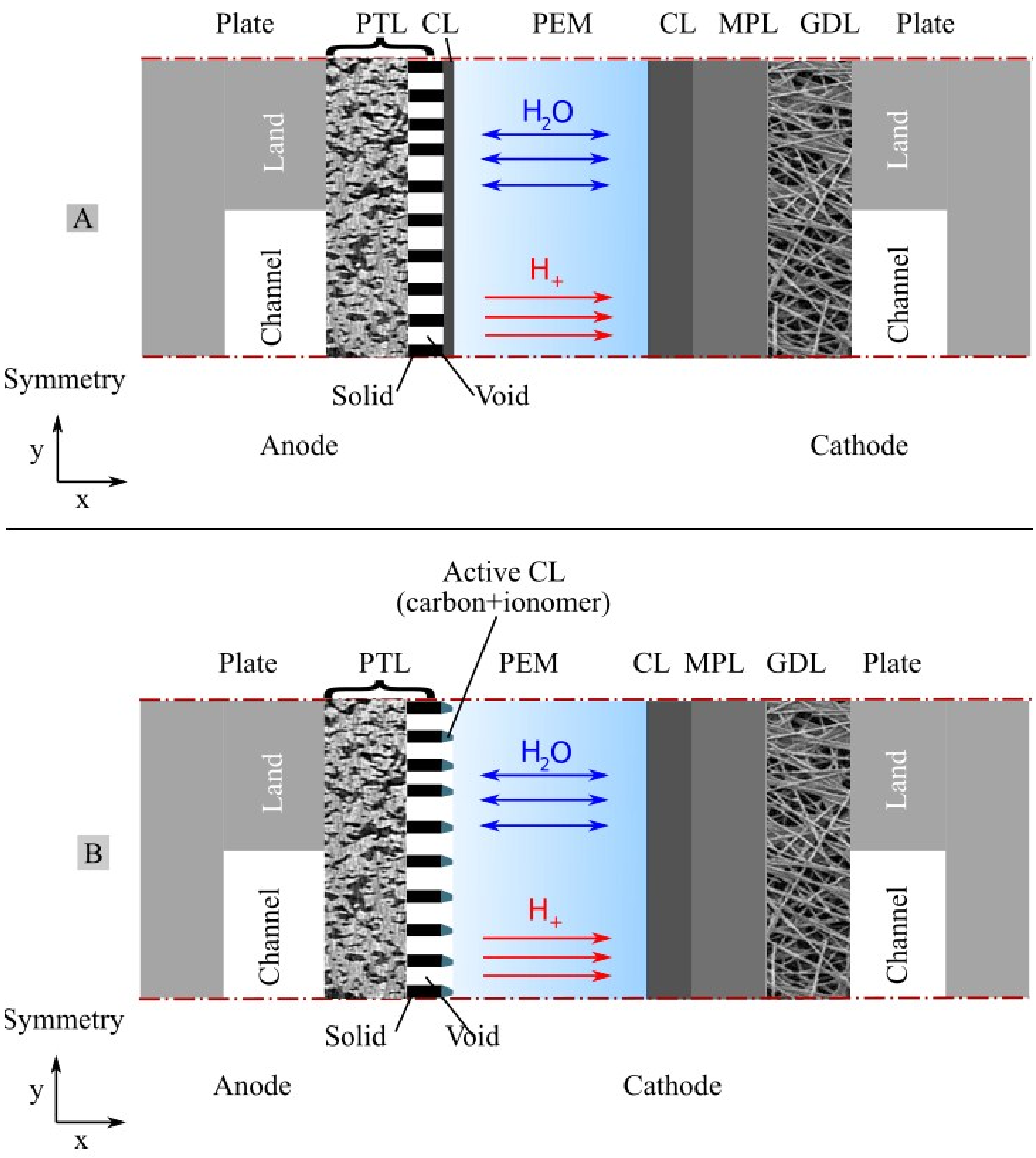

Figure S7: Simulation domain for different electrolyzer architectures. (a) Catalyst coated membrane (CCM) configuration, (b) Porous Transport Electrode (PTE) configuration

Table S1: List of parameters used for electrolyzer simulation

Parameter	**Value**
Operating conditions	Same as in experiment
PTL thickness	254 μm
GDL thickness	190 μm
MPL thickness	45 μm
Membrane thickness	150 μm
Cathode CL thickness	5 μm
Anode CL zone thickness	5 μm
Anode roughness PTL void pore height	30 μm
Anode roughness PTL solid part height	15 μm
A_v (anode specific catalyst area)	2.5×10^6 1/m
A_v (cathode specific catalyst area)	3.0×10^6 1/m
$i_{0,OER}$ (OER exchange current density)	$5 \times 10^{-2} A/m^2$
$i_{0,HER}$ (HER exchange current density)	$215[A/m^2]\exp\left[\frac{17000[J/mol]}{R}\left(\frac{1}{T_t}-\frac{1}{T}\right)\right]$
$\alpha_{OER,a}$ (OER anodic transfer coefficient)	0.6
$\alpha_{OER,c}$ (OER cathodic transfer coefficient)	0.6
$\alpha_{HER,a}$ (HER anodic transfer coefficient)	0.5
$\alpha_{HER,c}$ (HER cathodic transfer coefficient)	0.5
p_{ref} (Reference pressure)	$1 \times 10^5\ Pa$
E_{OER} (OER reference potential)	$1.23\ [V] - 8.45 \times 10^{-4}(T - T_t)$
$k_{T,Ti}$ (Titanium thermal conductivity)	$6[W/m \cdot K]$
PTL water retention curve	Shown in Fig. S8
Ti and Ir bulk electric conductivity	12,000 S/m [a]

a) See equation S1 that correlates bulk and effective electric conductivity

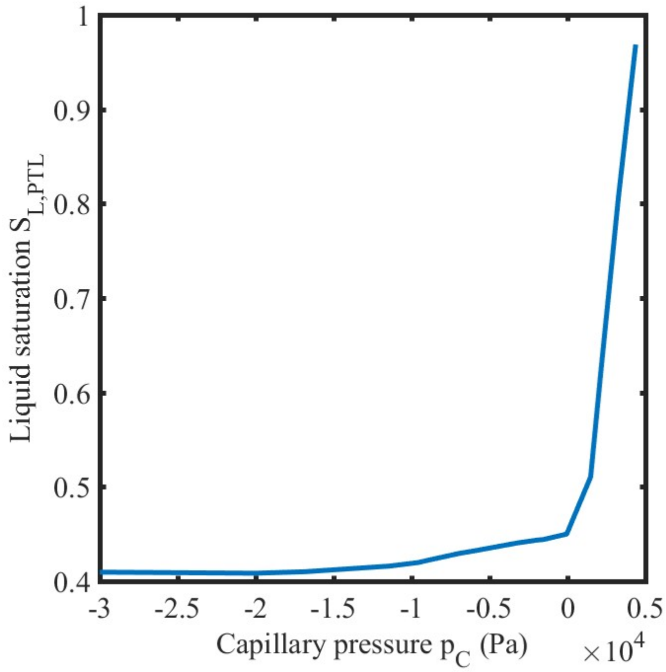

Figure S8: Water retention curve of porous transport layer

$$\sigma_{eff} = \sigma_{bulk} \times \epsilon_{solid}^{1.5} \quad [S1]$$

Where σ_{eff} is effective electric conductivity, σ_{bulk} is bulk value reported in Table S1, ϵ_{solid} is the volume fraction of solid material and 1.5 is Bruggeman constant.

Permeability of gas and liquid phases is saturation dependent[1]:

$k_L = k_{rL} \times k_0 = S_L^3 * k_0$

Where k_0 is absolute permeability. S_L is liquid water saturation.

References

1. I. V. Zenyuk, P. K. Das and A. Z. Weber, *Journal of The Electrochemical Society*, 2016, **163**, F691-F703.