Aging Iridium Oxide Catalyst Inks: A Formulation Strategy to Enhance Ink Processability for Polymer Electrolyte Membrane Water Electrolyzers

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Supporting Information

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1. Effect of resting procedure: steady-shear rheology

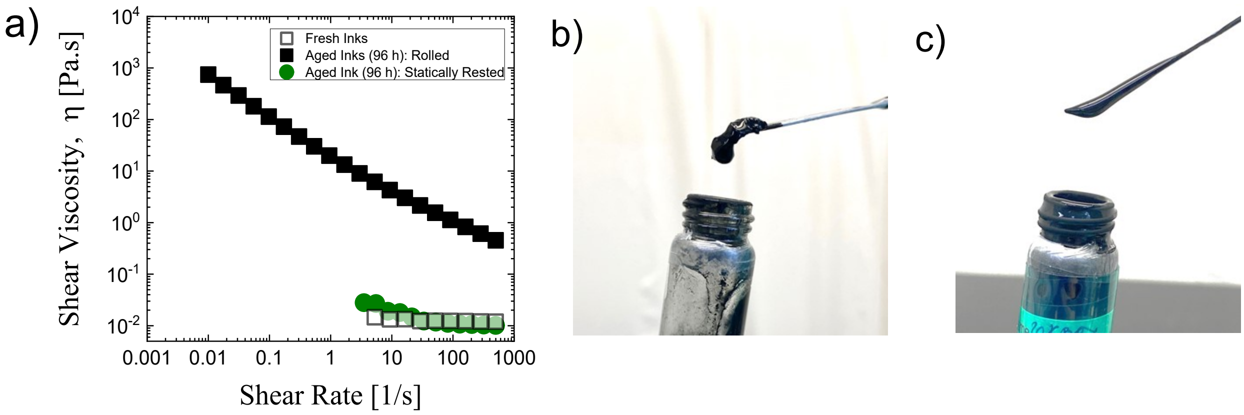

Figure SI1. Steady rheology comparison of aged inks ((20% IrO2 – 90% H2O) – 0.2 I/C) with different resting procedures. The data includes rolled inks and statically rested inks that were sonicated (1 h) prior to making measurements. The data of fresh inks is also included. Images of b) statistically rested and c) rolled inks after 96 h aging time examining the ink sample at the bottom of the vial.

2. Effect of IrO_2 concentration on ink aging: steady-shear rheology

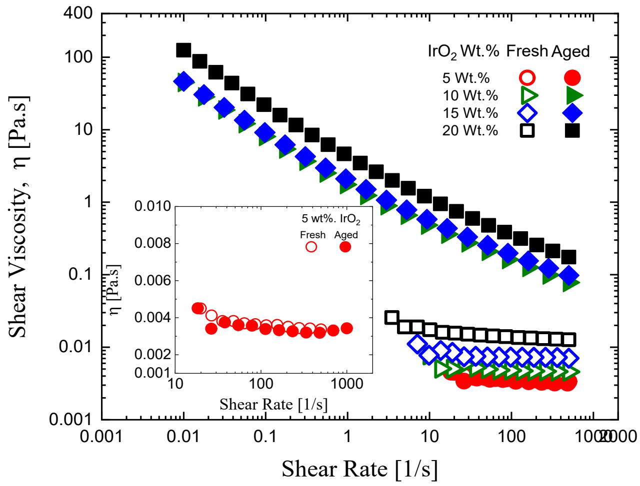

Figure SI2. Steady-shear rheology of water-rich (90 wt.% H_2O) inks: Fresh and Aged (t_a ~ 96 h) at a series of IrO_2 concentrations. The ionomer concentration was fixed at 0.2 I/C. The inset shows the data of inks at 5 wt% IrO_2 concentration.

3. Effect of ionomer concentration on ink aging: Frequency sweep data

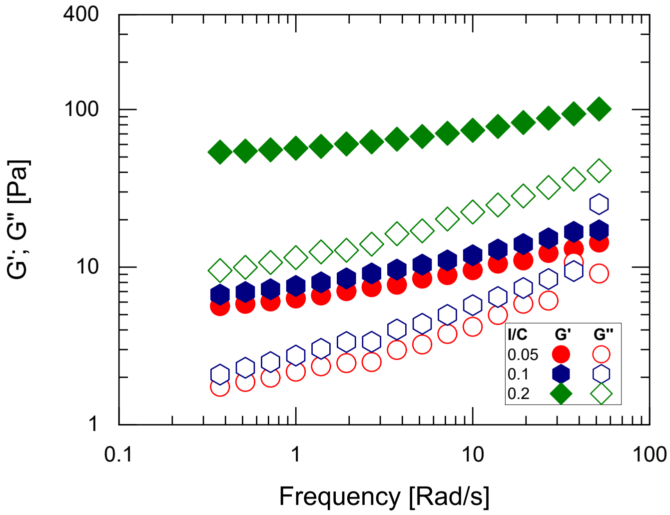

Figure SI3. Frequency sweep measurements of aged inks (20% IrO_2 – 90% H_2O) at different I/Cs tested after 96 h aging time.

4. USAXS: Volume size distribution plots of fresh water-rich inks

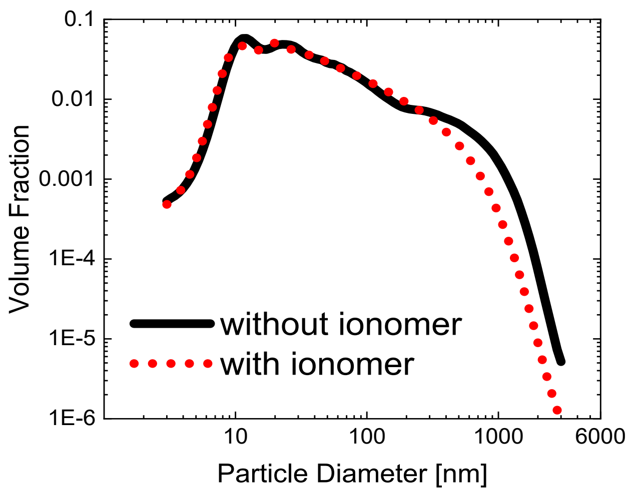

Figure SI4. Volume size distribution comparison of water-rich inks (30 wt% IrO2 – 0.1 I/C – 90%H2O/10%nPA) with and without ionomer (0.1 I/C) extracted from USAXS scattering curves shown in Figure 4a.

5. Electrophoretic Mobility and Dynamic Light Scattering Measurements

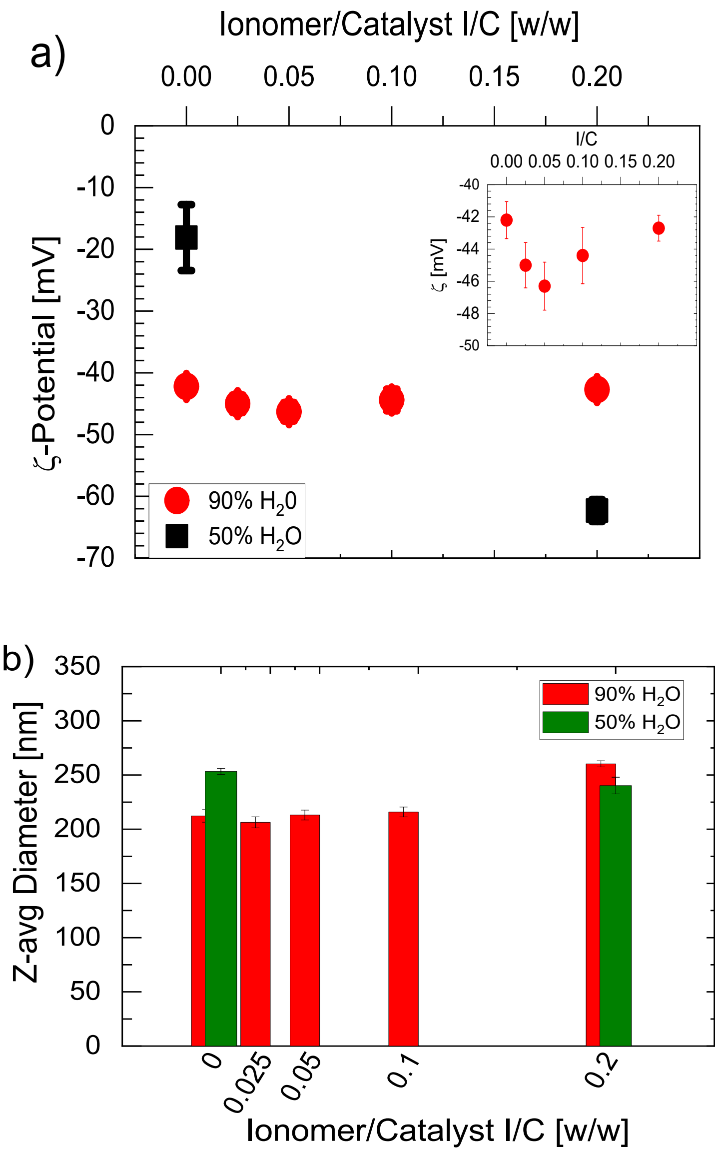

Figure SI5. a) Zeta potential and Z_{avg} size measurements of 0.1% IrO_2 inks at different I/C (D2020). The data include both water-rich (90% H_2O) and alcohol-rich (50% H_2O) inks. The inset in Figure a shows zoomed-in zeta potential data of water-rich inks.

6. Steady-shear rheology of water-rich neat ionomer dispersions at different aging times

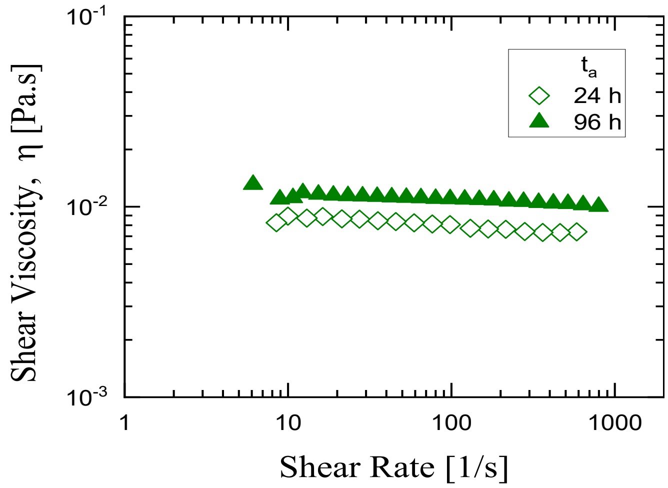

Figure SI6. Steady-shear rheology of water-rich ionomer dispersions (4.75% ionomer – 90% H_2O) at different aging times.

7. Transient rheology of aged water-rich inks

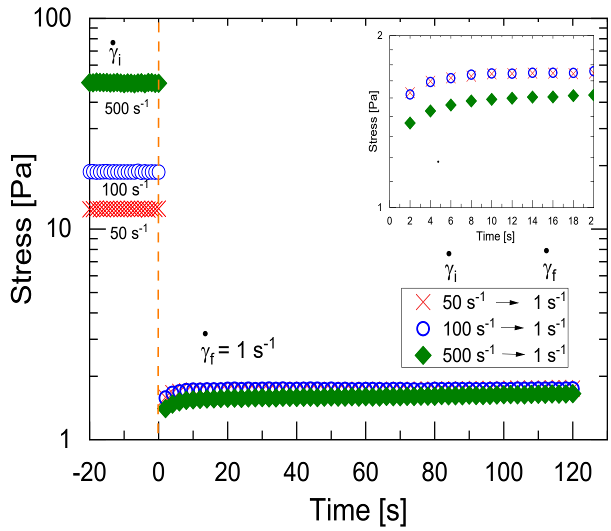

Figure SI7. Stress evolution in shear rate step-down jump experiments. Shear rate is dropped from different initial shear rates (50, 100, and 500 s^{-1}) to 1 s^{-1}. The data corresponds to aged (96 h) water-rich inks. 20% IrO_2 – 0.2 I/C – 90% H_2O.

8. Steady-shear rheology data of water-rich inks containing Nafion D2020 and D2021

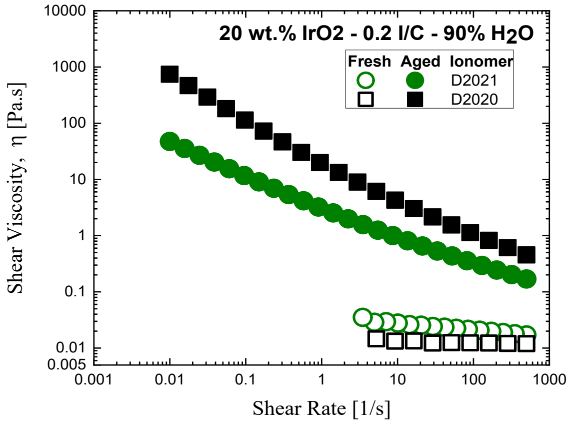

Figure SI8. Steady-shear rheology of comparison of fresh and aged (96 h) inks consisting of Nafion D2020 and D2021 dispersions.

9. EIS Polarization Curves

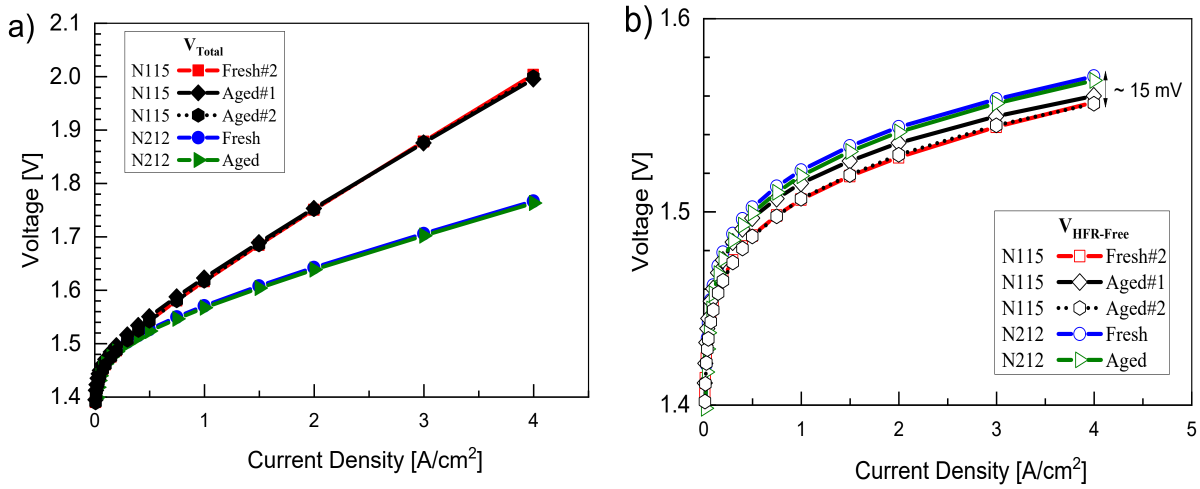

Figure SI9. Polarization curves measured from EIS of electrodes made from fresh and aged water-rich inks: a) V_{Total} and b) HFR-corrected ($V_{HFR\text{-}free}$) polarization curves. The measurements collected using N212 membrane is also included. The Ir loadings of Fresh and Aged cases tested using N212 membrane were 0.502 ± 0.03 and 0.598 ± 0.015 mgIr/cm^2, respectively.

10. Cyclic Voltammetry Data

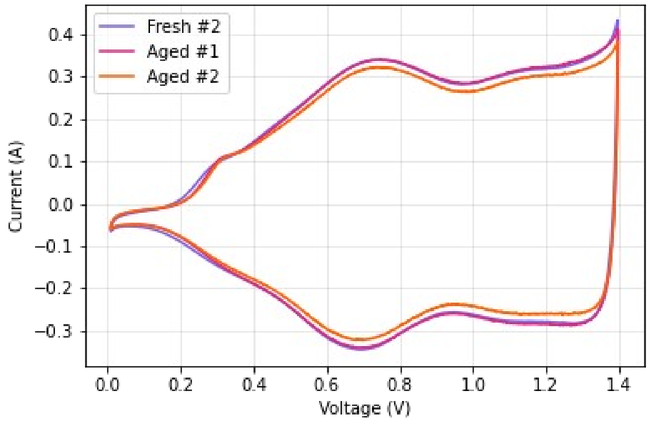

Figure SI10: Comparison of cyclic voltammetry (CV) following conditioning and EIS measurement for samples where a CV following conditioning was measured.