Electronic Supplementary Material (ESI) for RSC Advances

Nafion®-Stabilised Pt/C Electrocatalysts with Efficient Catalyst Layer Ionomer Distribution for Proton Exchange Membrane Fuel Cells

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SUPPORTING INFORMATION

Ionomer content measured by TGA

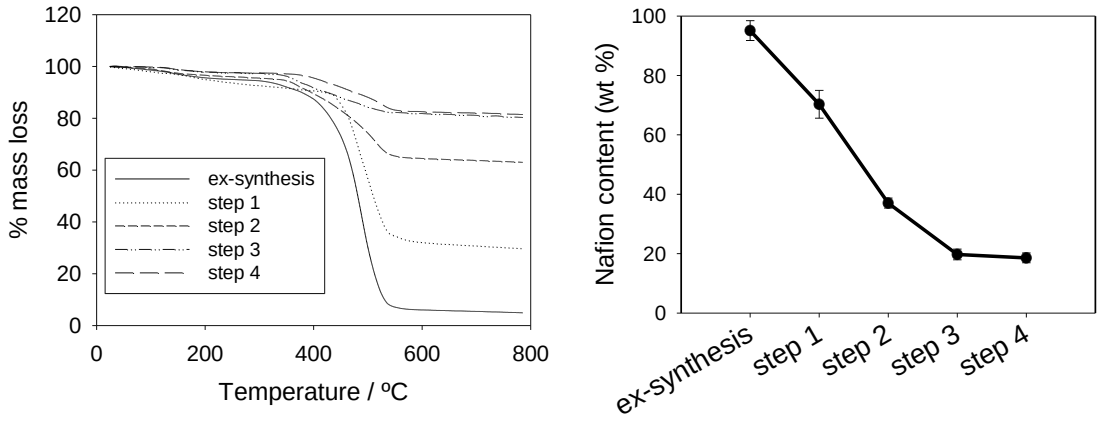

Figure S1 - a) Thermogravimetric mass loss profiles obtained in air with a 50 K.min^{-1} thermal ramp and b) Nafion content of the colloidal Pt as a function of the number of centrifugation steps. The ionomer content of the colloidal Pt decreased with successive centrifugation cycles, but a residual 20 wt % Nafion remained after 3 cycles, and could not be removed by further treatment, indicating that this residual Nafion is strongly bound within the product.

Effect of catalyst loading on utilisation

To examine the effect of working electrode catalyst loading L_{Pt} on the measured ECSA, a series of electrodes were prepared at catalyst loadings of 20, 40 and 80 $\mu g_{Pt}\,cm^{-2}$ for the Nafion-Pt/C A and E-Tek catalysts. The Nafion content was 20 % NFP for both catalysts, representing an optimal ionomer content for the Nafion-Pt/C catalyst, but a sub-optimal ionomer content for the E-Tek catalyst (previous work has found the optimal ionomer content for the E-Tek catalyst to be ca. 33 % NFP).

The CVs in Figure S2 (a) and (b) are normalised by Pt loading and show clear differences in the relationship between L_{Pt} and mass-specific current in the H_{upd} region between the catalyst with optimal ionomer content (Nafion-Pt/C A) and that with sub-optimal ionomer content (E-Tek). Utilisation values based on ECSAs calculated from the H_{upd} charges are shown as a function of L_{Pt} in Figure S2 (c). The Nafion-Pt/C electrodes prepared at 20 % NFP gave around 100 % utilisation at all loadings, whilst the utilisation for the E-Tek electrodes fell from a maximum 76 % at low loading (20 $\mu g_{Pt}.cm^{-2}$) to just 50 % at high loading (80 $\mu g_{Pt}\,cm^{-2}$).

These results demonstrate that at high Pt loadings (L_{Pt} > 40 $\mu g_{Pt}\,cm^{-2}$), thin-film catalyst layers with sub-optimal ionomer loadings exhibit poor utilisation, whilst those prepared at optimal ionomer loading show 100 % utilisation. This demonstrates the viability of using the RDE method for screening catalyst ink compositions to investigate optimal ionomer content, prior to fabricating full MEAs.

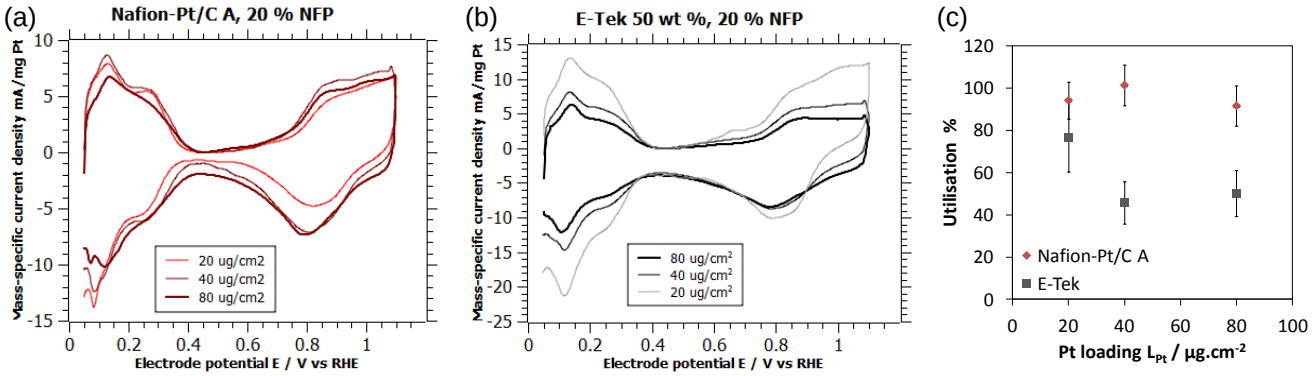

Figure S2 - Cyclic voltammograms recorded for working electrodes prepared at various Pt loadings (20, 40, 80 ug cm^{-2}) in N_2-purged 0.1M $HClO_4$ at 25 mV s^{-1} for (a) Nafion-Pt/C A catalyst with optimal Nafion content (20% NFP); (b) E-Tek catalyst with sub-optimal Nafion content (20 % NFP). (c) catalyst utilisation as a function of Pt loading on the working electrode for Nafion-Pt/C A and E-Tek catalysts prepared at 20 % NFP.

Oxygen Reduction Reaction – supplementary data

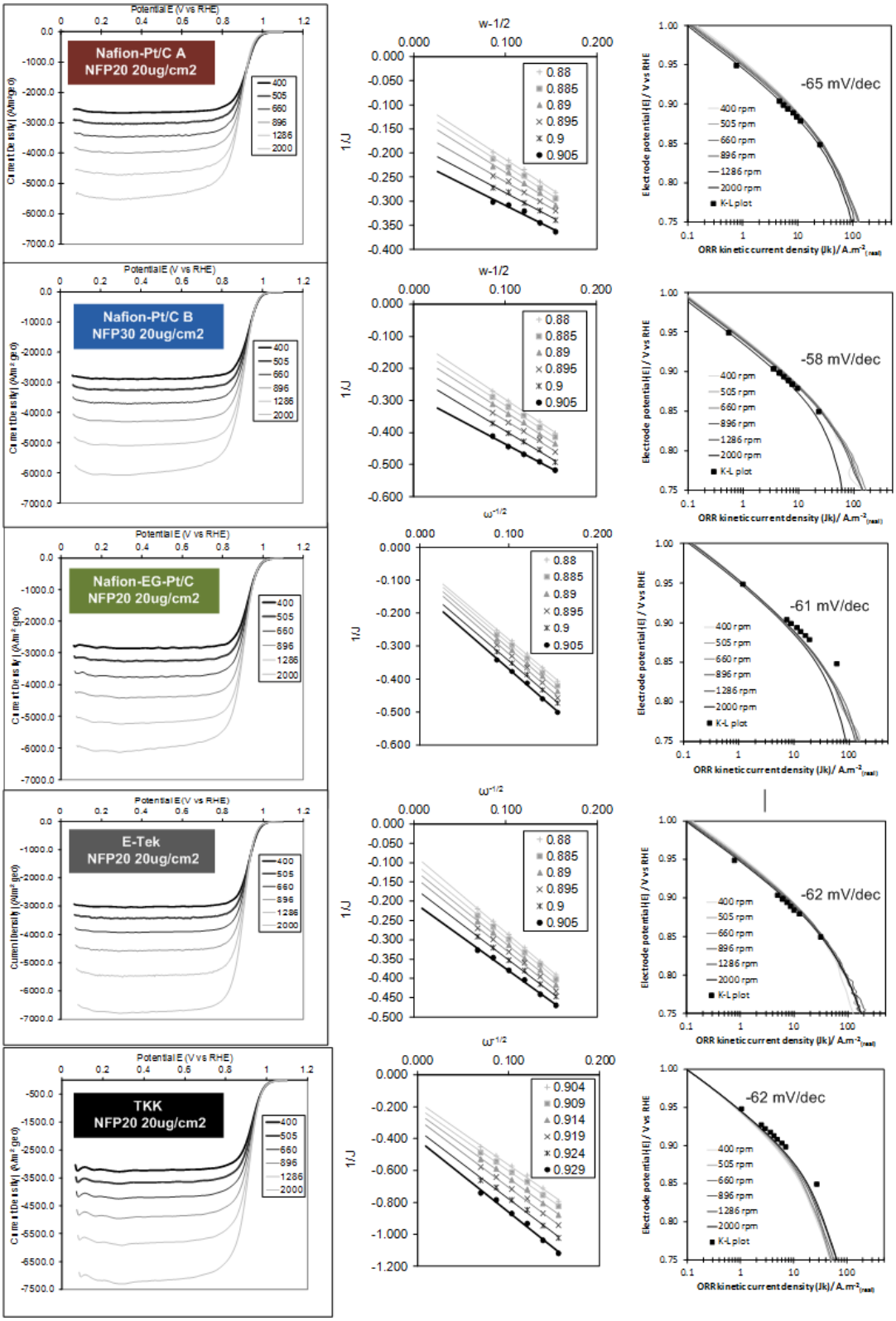

Figure S3 - (left) iR-corrected, background-subtracted RDE data at various rotation rates; (centre) Koutecky-Levich plots; (right) mass-transport corrected Tafel plots for each electrocatalyst under test in O_2-saturated 0.1 M $HClO_4$ at 25ºC.