Supporting Information to:

Ultra-thin Proton Conducting Carrier Layers for Scalable Integration of Atomically Thin 2D Materials with Proton Exchange Polymers for next-generation PEMs

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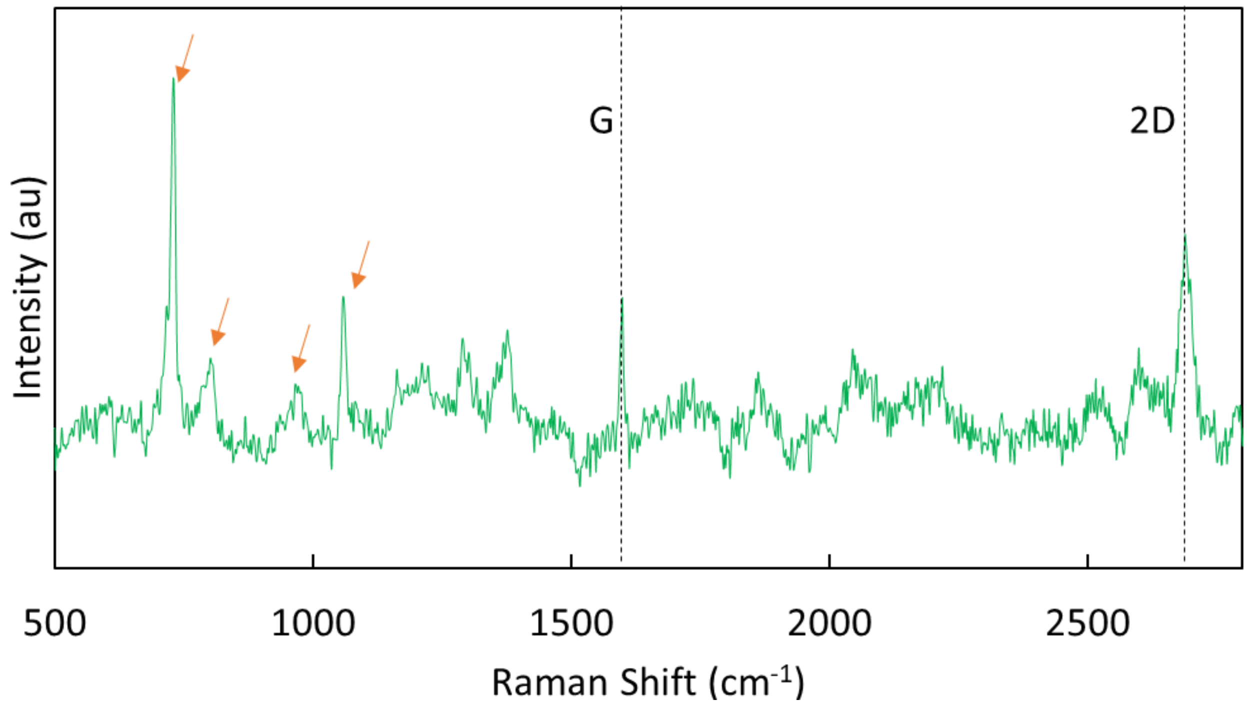

Figure S1. Raman spectra of graphene transferred to N211 via hot press method with an extended scan range.

Additional characteristic Nafion peaks are observed at ~730 cm^{-1}, ~810 cm^{-1}, ~980 cm^{-1} and 1060 cm^{-1} (orange arrows). Peaks at ~1580 cm^{-1} and ~2690 cm^{-1} are attributed to the G and 2D peaks of graphene, respectively. Dotted lines serve as a guide to the eye.

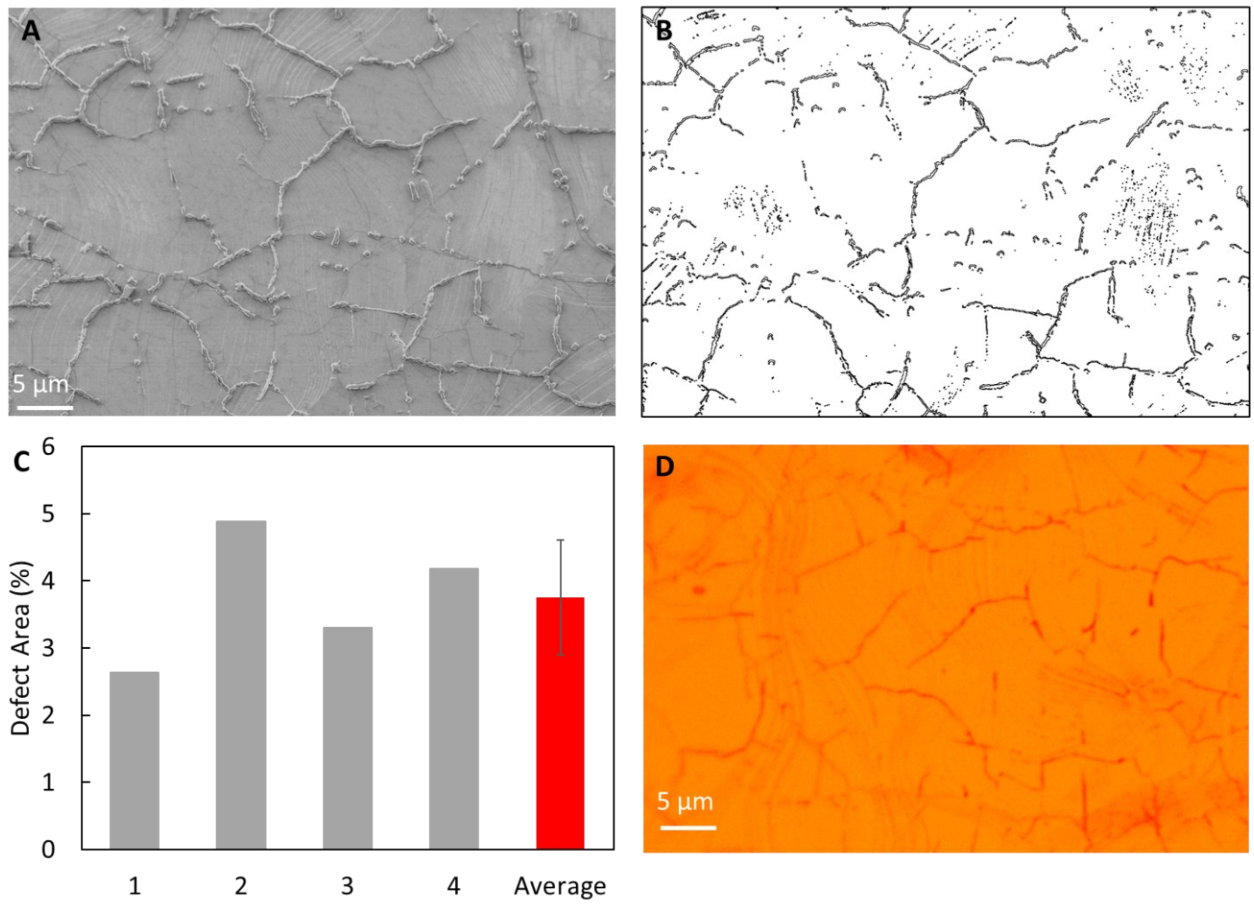

Figure S2. Quantifying defects from hot press transfer method.

A) SEM image of graphene transferred to Nafion 211 via 140ºC hot press transfer, showing defects along wrinkles. B) The same image as in A after thresholding in ImageJ is used to estimate the average defective area. C) Plot of the calculated percent defect area as determined from 4 different SEM images and the average (Red Bar). The average defect area of ~3.5% is comparable to other reports in literature.[1,2] D) Image of graphene on Cu foil after oxidization in air at 220ºC for 10 minutes. The dark orange lines indicate oxidized area underneath defects in graphene on Cu foil. The features observed here are similar to graphene transferred to Nafion using hot press transfer.

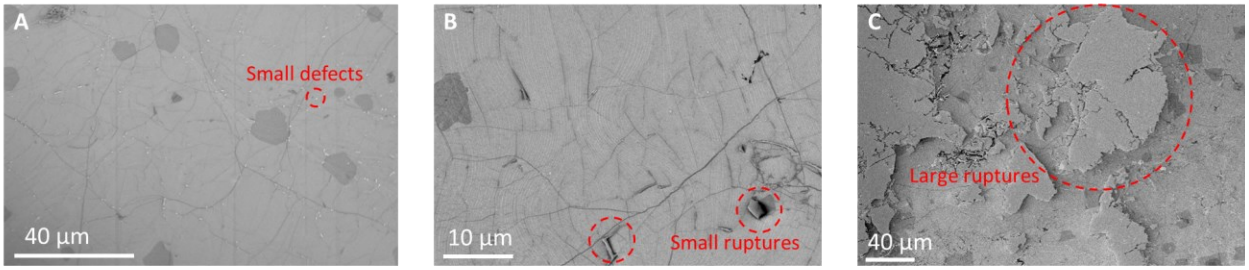

Figure S3. Defects observed for cold press transfers.

Graphene transferred to N211 via casting a thin layer of Nafion on G+Cu, then pressing at room temperature to N211. SEM of the transferred graphene shows areas with A) small, isolated defects, B) small ruptures, and C) large ruptures. The ruptures occur due to poor contact between the N211 and the graphene. The regions of large ruptures are sparsely occurring, with most of the defects observed being small defects and ruptures.

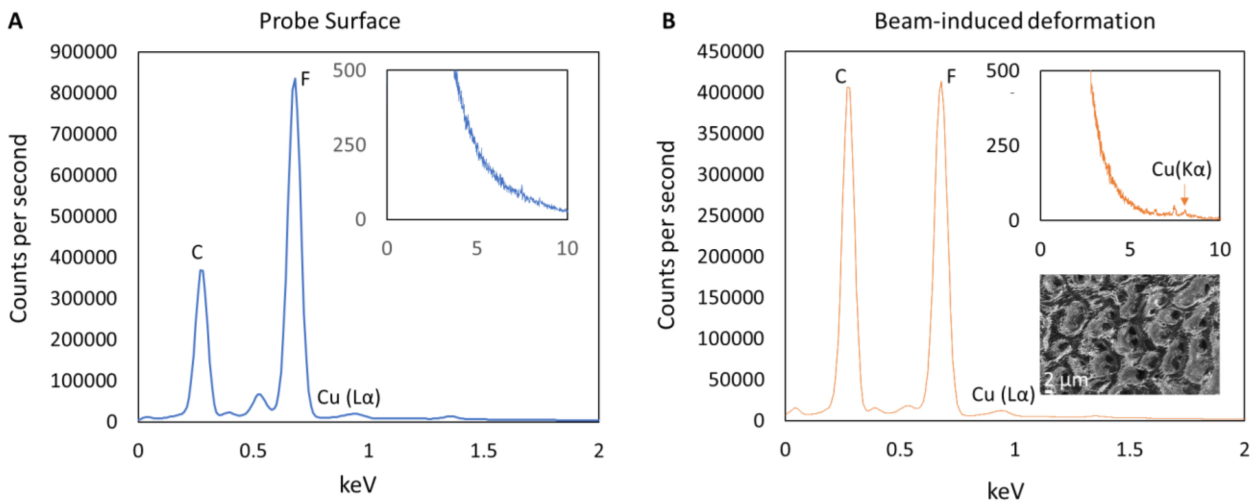

Figure S4. EDS of N211|G before HCl soaking.

Graphene was transferred to N211 via hot pressing and EDS collected of the A) surface and B) after inducing deformations to the surface. For the surface probe measurements, the magnification was kept low (<800×) to reduce charging/damage to the sample. Upon increasing the magnification (>800×), deformation of the sample was observed (inset of B). For both measurements, the Lα peak for Cu at ~0.93 keV is visible but only when the sample is undergoing beam-induced damage is the Kα peak at ~8.04 keV visible suggesting the presence of Cu within the Nafion and not being limited to only the surface.

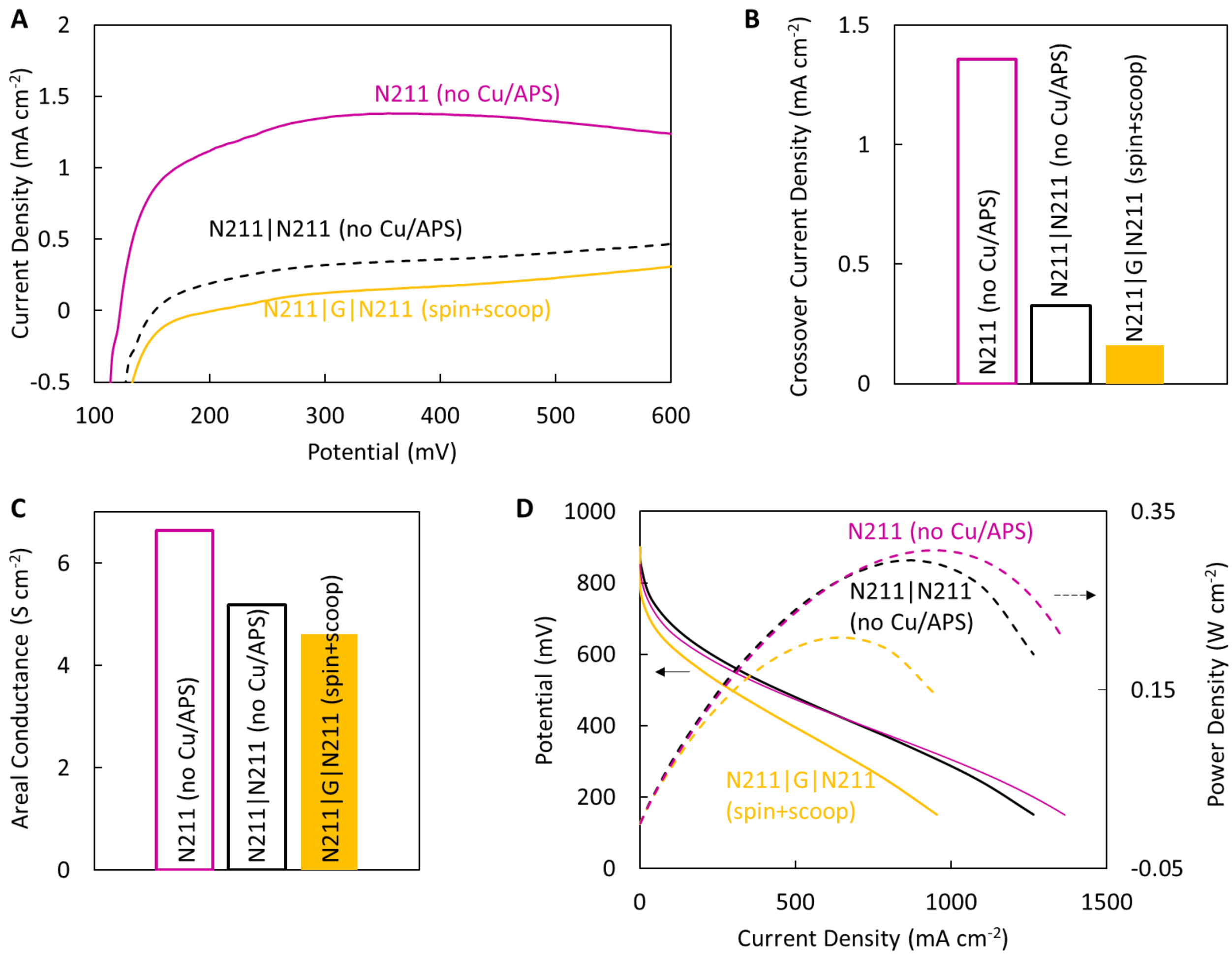

Figure S5: Comparison of sandwich membranes to single layer N211.

A) H_2 crossover current density as a function of potential for N211, N211|N211, and N211|G|N211 (spin+scoop) membranes. B) Crossover current density extracted from (A) for each membrane at 0.4V. C) Areal proton conductance measured by supplying H_2 to both sides of the membrane. D) I-V curves (left axis, solid lines) and power density curves (right axis, dotted lines) for each membrane in the custom-built H_2/Air fuel cell at atmospheric pressure and room temperature.

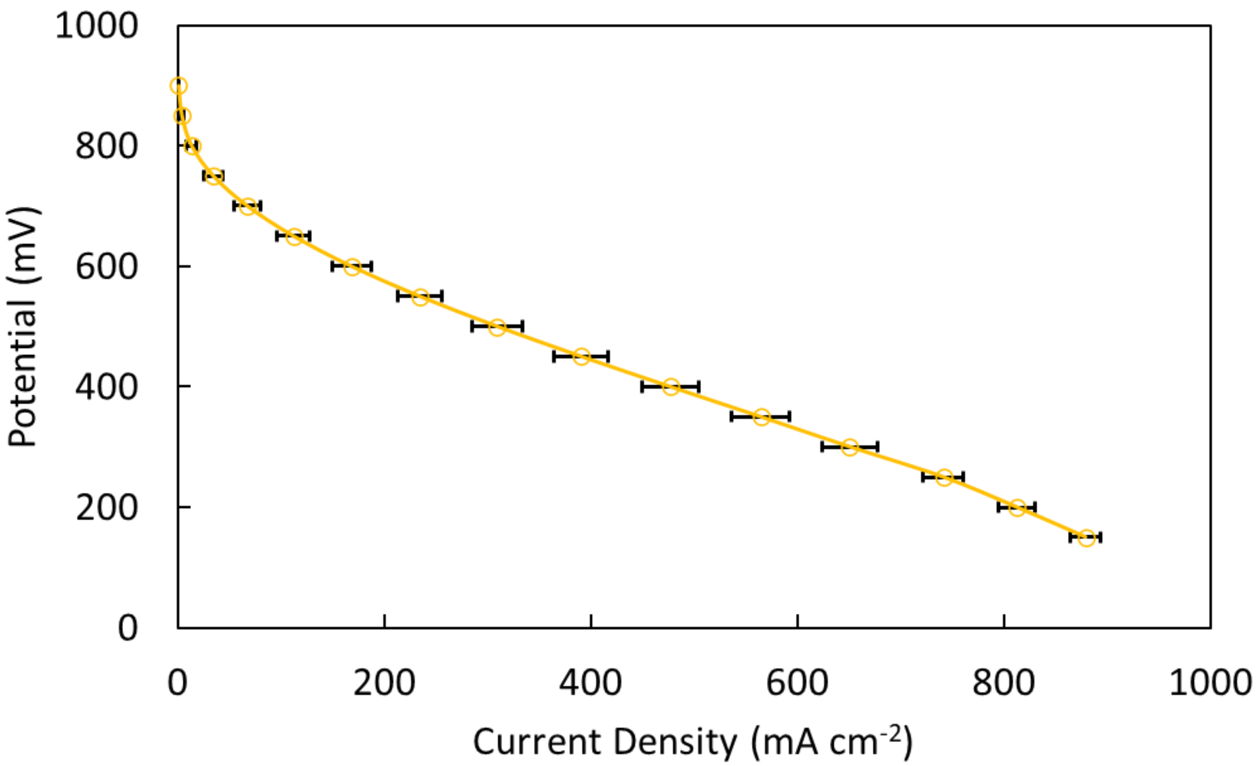

Figure S6. Stability of N211|G|N211 (spin+scoop) membranes when operated in the custom-built H_2/Air fuel cell at beginning of life.

Three polarization curves were collected 10 minutes apart for a N211|G|N211 (spin+scoop) membrane. The average at each point is calculated and plotted here along with respective standard deviation in current density, showing the stability of the membrane over the course of the measurements. All measurements were completed at room temperature and atmospheric pressure.

Supporting Information References

1 N. K. Moehring, P. Chaturvedi, P. Cheng, W. Ko, A.-P. Li, M. S. H. H. Boutilier and P. R. Kidambi, *ACS Nano*, 2022, **16**, 16003–16018.

2 P. R. Kidambi, R. A. Terry, L. Wang, M. S. H. Boutilier, D. Jang, J. Kong, R. Karnik, M. S. H Boutilier, D. Jang, J. Kong and R. Karnik, *Nanoscale*, 2017, **9**, 8496–8507.