Supporting Information

Graphene coating of Nafion® membranes for enhanced fuel cell performance

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Figure S1. Measurement setup schematics. (a) Measurement of proton conductivity in fuel cell test rig with electrochemical impedance spectroscopy (EIS) and recording of polarization curves by galvanostatic method (b) Measurement of methanol permeation with liquid methanol sensor. (c) Hydrogen permeation measurement through membrane in dedicated cell. A mass spectrometer (MS) is used to analyze hydrogen concentration at cathode outlet.
Figure S2. (a) ADF-STEM cross-sectional micrograph (same as Fig. 2c). (b-d) Respective multiple linear least-squares (MLLS) fits of the spectral graphene, Nafion, and epoxy references (shown in Fig. 2e) to the EELS mapping of the same area as in a).
Figure S3. Supporting data of a previous coating attempt using commercial graphene nano-platelet material (a) SEM top-view image observed under 29 ° tilting angle. Individual flakes can be distinguished and do not form a smooth and closed film. (b) Smoothed EELS reference spectra for graphene, the epoxy resin and Nafion created from the locally integrated STEM-EELS signal after background subtraction. (c-e) Respective MLLS fits of the internal references shown in (b) to the recorded STEM-EELS spectrum image. (f-h) Elemental distributions of C, O and F based on their respective K edges in the according STEM-EELS spectrum image.
**Figure S4.** Raman spectrum of e-G thin film, normalized to G-peak.
Figure S5. Hydrogen concentration over time, measured by mass spectrometer. The hydrogen permeation through graphene-coated and reference membrane in dry and hydrated state is determined.
Table S6. Methanol Permeabilities of graphene-coated membranes, measured at 1M and 5M MeOH concentration.

| Membrane                  | MeOH Permeability [cm² s⁻¹] x 10⁻⁷ |
|---------------------------|-------------------------------------|
|                           | 1M MeOH               | 5M MeOH               |
| Nafion® N115 reference    | 32.4                  | 243.5                 |
| N115 + 32.5 µg cm⁻² e-G   | 14.3                  | 129.4                 |
| N115 + 65 µg cm⁻² e-G     | 12.9                  | 108.2                 |
| N115 + 130 µg cm⁻² e-G    | 12.3                  | 12.5                  |