## **Electronic supplementary information:**

## Effect of Membrane Mechanics on AEM Fuel Cell Performance

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## 1.1 Current Density vs pH relationship

Figure S1: Current density vs pH of mTPN1-TMA membranes operated at pH (a) 9 and (b) 10

In order to estimate the effect of current on the pH of the water exiting the cell, the pH was measured at the output as a function of the current density. We first set the pH, pH9 and pH10, by varying the gas flow at zero current as described in section 2.4. The current density was then increased and the station was run at constant current for an hour before collecting the water exiting the cell measuring the pH. The pH as a function of current density is plotted above for mTPN1-TMA membranes. From the figures we see that indeed the pH is a function of current density, but at the density where the experiments described here were performed the actual corresponding to pH=9 was pH=8.6 and 8.8 for the anode and cathode respectively. Similarly pH=10 corresponded to pH=9.7 and 9.8

at the anode and cathode respectively. In both cases, therefore, the pH within the cell was not expected to vary significantly from that at the anode and cathode.

## 1.2 Electrochemical Measurements

The cyclic voltammograms and EIS measurements before and after the durability test for both mTPN1-TMA and Sustainion membranes at pH 9 and 10 are shown in Fig. S2 and S3 respectively. The electrochemically active surface area (ECSA) of the Pt catalyst was calculated from the peak current densities for hydrogen adsorption reaction.

The ECSA values are mentioned in Table 1. From the table, we can see that the total decrease in area after the durability test for mTPN1-TMA membranes at pH 9 was 72.5% and at pH 10 was 84.31%. The total decrease in area for sustainion membranes at pH 9 was 48.7% and at pH 10 was 54.6%.





Figure S2: Cyclic voltammograms of the cathode electrode in inert gas atmosphere before and after durability test for (1) mTPN1-TMA membranes at (a) pH-9 (b) pH-10 (2) Sustainion membranes at (c) pH-9 (d) pH-10

	Initial ECSA of Pt (cm²/g)	Final ECSA of Pt (cm²/g)	Change (%)
mTPN1-TMA_pH9	65.29	17.91	72.56
mTPN1-TMA_pH10	51.12	8.02	84.31
Sustainion_pH9	61.52	31.58	48.67
Sustainion_pH10	63.78	28.95	54.60

Table 1. The active Pt surface area change

The EIS measurements for both mPN1 and sustainion membranes at pH 9 and 10 in Fig. S3 suggest that the ohmic resistance of the cell is  $0.17\Omega$  at pH 10 for both membranes while the resistance is higher i.e.,  $0.21\Omega$  for mTPN1-TMA and  $0.19\Omega$  for sustainion membranes at pH 9. Both membranes experience higher resistance at pH9. From the table, we can see that the total increase in the ohmic resistance after the durability test for mTPN1-TMA membranes at pH 9 was 42.86% and at pH 10 it was 58.8%, while for sustainion membranes it was 47.4% at pH 9 and 70.6% at pH 10.



Figure S3: EIS spectra of AEMFC in inert gas atmosphere before and after durability test for (1) mTPN1-TMA membranes at (a) pH-9 (b) pH-10 (2) Sustainion membranes at (c) pH-9 (d) pH-10

	Initial Ohmic resistance (Ohm)	Final Ohmic resistance (Ohm)	Change (%)
mTPN1-TMA_pH9	0.21	0.3	42.86
mTPN1-TMA_pH10	0.17	0.27	58.8
Sustainion_pH9	0.19	0.28	47.4

Sustainion_pH10	0.17	0.29	70.6

Table 2. Change in Ohmic resistance