Supplementary Information

## Hierarchically Porous, Biaxially Woven Carbon Nanotube Sheet Arrays for Next-Generation Anion-Exchange Membrane Water Electrolyzers

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| Sample name              | Ohmic resistance | Charge-transfer<br>resistance | Mass transport resistance |  |
|--------------------------|------------------|-------------------------------|---------------------------|--|
| NiFeO <sub>x</sub> _CNTS | 65               | 177                           | 0.112                     |  |
| NiFeO <sub>x</sub> _NP   | 126              | 495                           | 0.228                     |  |
| Pt_CNTS                  | 75               | 12.0                          | 0.06                      |  |
| Pt_NP                    | 99               | 12.5                          | 0.2                       |  |

Table S1. Comparison of ohmic, charge-transfer, and mass transport resistances of four

AEMWEs obtained using the Nyquist plot.

| Reference<br>number<br>This<br>work | Cathode<br>material<br><b>NiFeO</b> x   | Cathode<br>loading<br>[mg cm <sup>-</sup><br><sup>2</sup> ]<br><b>2</b> | Membrane<br>FAA-3-50 | Anode material<br>NiFe  | Anode<br>loading<br>[mg cm <sup>-</sup><br><sup>2</sup> ]<br>1 | Temperatur<br>e<br>[°C]<br>70 | Cell<br>voltage<br>[V]<br>2.05 | Current<br>density<br>[A cm <sup>-2</sup> ]<br>1480 |
|-------------------------------------|---|---|----------------------|---|--|-------------------------------|--------------------------------|---|
| 44                                  | Ni <sub>12</sub> P <sub>5</sub> Ni <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> | 3   | YAB                  | Ni <sub>12</sub> P <sub>5</sub> Ni <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> | 3  | 50                            | 2.05                           | 650   |
| 46                                  | Ni  | 0.18  | FAA-3-50             | CuCoO   | 0.4 40   |                               | 2.05                           | 75  |
| 47                                  | Ni  | 1.45  | FAA-3-50             | Ni  | 1.45   | 40                            | 2.05                           | 60  |
| 45                                  | NiCoS   | -   | Nafion               | IrO2  | -  | 50                            | 2.05                           | 610   |
| 48                                  | NiCoO-NiCo/C  | 1.5   | X37-50               | CuCoO   | 30   | 50                            | 1.85                           | 504   |
| 49                                  | NiCu  | 5   | Fumapem-<br>3-pe-30  | Ir  | 3  | 50                            | 1.9                            | 1350  |
| 50                                  | Ni  | 0.38  | FAA-3-50             | CuCoO   | 0.4  | _                             | 2.05                           | 110   |
| 51                                  | NiFeCo-P  | -   | X37-50               | NiFeO   | -  | 50                            | 1.72                           | 500   |
| 52                                  | NiMnO   | 1   | Fumapem-<br>3-pe-30  | NiCoFe  | 3  | 50                            | 2.0                            | 1150  |
| 53                                  | Ni/(CeO <sub>2</sub> -<br>La <sub>2</sub> O <sub>3</sub> )/C                    | 2.7   | A201                 | ACTA 3030   | -  | _                             | 1.9                            | 300   |
| 54                                  | Ni/(CeO <sub>2</sub> -<br>La <sub>2</sub> O <sub>3</sub> )/C                    | 7.4   | A201                 | ACTA 3030   | 30   | 60                            | 1.9                            | 350   |

Table S2. Comparison of AEMWE performances using the non-noble metal-based HER

catalysts reported in the literature<sup>44-54</sup> and in this work.

| Reference<br>number | Cathode<br>material | Cathode<br>loading<br>[mg cm <sup>-2</sup> ] | Membrane | Anode<br>material | Anode<br>loading<br>[mg cm <sup>-2</sup> ] | Temperatur<br>e<br>[°C] | Cell<br>voltage<br>[V] | Current<br>density<br>[A cm <sup>-2</sup> ] |
|---------------------|---------------------|--|----------|-------------------|--|-------------------------|------------------------|---|
| This<br>work        | Pt                  | 2  | FAA-3-50 | NiFe              | 1  | 70                      | 2.05                   | 4000  |
| 55                  | PtNi/CP             | 0.001856                                     | A201     | Ni/CP             | 0.00851                                    | 50                      | 1.9                    | 250   |
| 57                  | RuSe <sub>2</sub>   | 2  | FAA-3-50 | IrO <sub>2</sub>  | 2.5  | 80                      | 1.8                    | 730   |
| 56                  | TNTA+Pd             | 1.7  | A201     | IrO <sub>2</sub>  | 5  | 60                      | 2.05                   | 1000  |
| 58                  | Pt/C                | -  | A201     | IrO <sub>2</sub>  | -  | 50                      | 1.8                    | 1030  |
| 59                  | Pt/C                | 0.4  | FAA-3-50 | IrO <sub>2</sub>  | 2  | 70                      | 2.05                   | 2400  |
| 60                  | Pt/C                | 1  | FAA-3-50 | IrO <sub>2</sub>  | 0.5  | 80                      | 1.73                   | 1000  |
| 61                  | Pt/C                | 1  |          | CuCoO             |  | 40                      | 1.9                    | 2250  |
| 62                  | Pt/C                | 1  |          | CuCoO             | 0.2  | 45                      | 1.9                    | 1900  |
| 63                  | Pt/C                | 1  | X37-50   | COO-NP            | 3  | 50                      | 2.0                    | 1150  |
| 7                   | Pt/C                | 1  | X37-50   | NiFeOx            |  | -                       | 1.9                    | 2000  |
| 64                  | Pt/C                | -  | X37-50   | NiFeV             | 3.95                                       | 50                      | 1.9                    | 2850  |

Table S3. Comparison of AEMWE performances using the noble metal-based HER catalysts

reported in the literature<sup>7,55-64</sup> and in this work.



**Figure S1**. Photograph of process: (a) drawing carbon nanotube sheet (CNTS) from the vertically aligned carbon nanotube (VACNT) forest and (b) resulting single CNTS array.



Figure S2. X-ray diffraction (XRD) patterns of SUS paper.



Figure S3. Contact angles of four electrodes: (a) NiFeO<sub>x</sub>\_CNTS, (b) NiFeO<sub>x</sub>\_NP, (c) Pt\_CNTS, and (d) Pt\_NP.



Figure S4. Cyclic voltammograms of NiFeO<sub>x</sub>\_CNTS and NiFeO<sub>x</sub>\_NP.



Figure S5. Cyclic voltammograms of Pt\_CNTSs and Pt\_NPs.



**Figure S6**. Polarization curves for AEMWE using Pt\_CNTS, Pt\_NP (Pt black), and 40 wt.% Pt/C.



**Figure S7**. Comparison of polarization curves of two AEMWEs ((a) NiFeO<sub>x</sub>\_CNTS and (b) Pt\_CNTS) before and after the durability test.