

# **Supporting Information**

## **Regenerable Nickel-functionalized Activated Carbon Cathodes Enhanced by Metal Adsorption to Improve Hydrogen Production in Microbial Electrolysis Cells**

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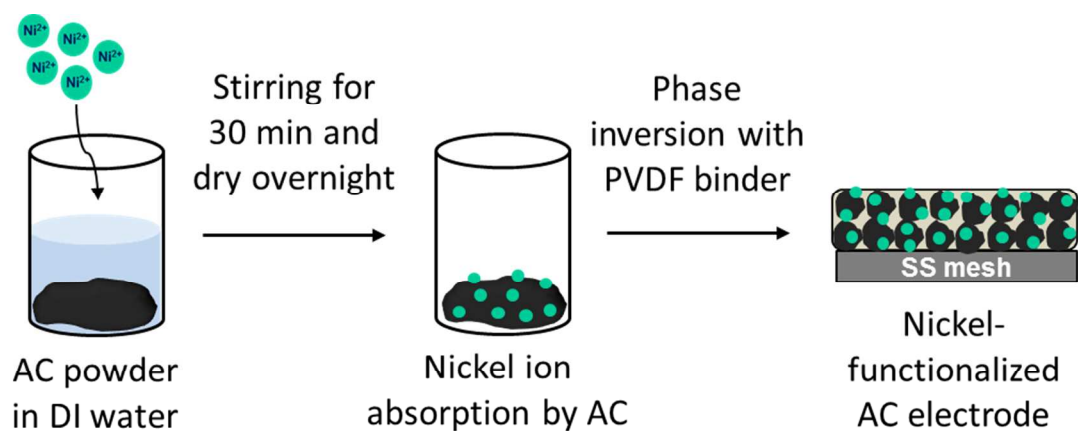
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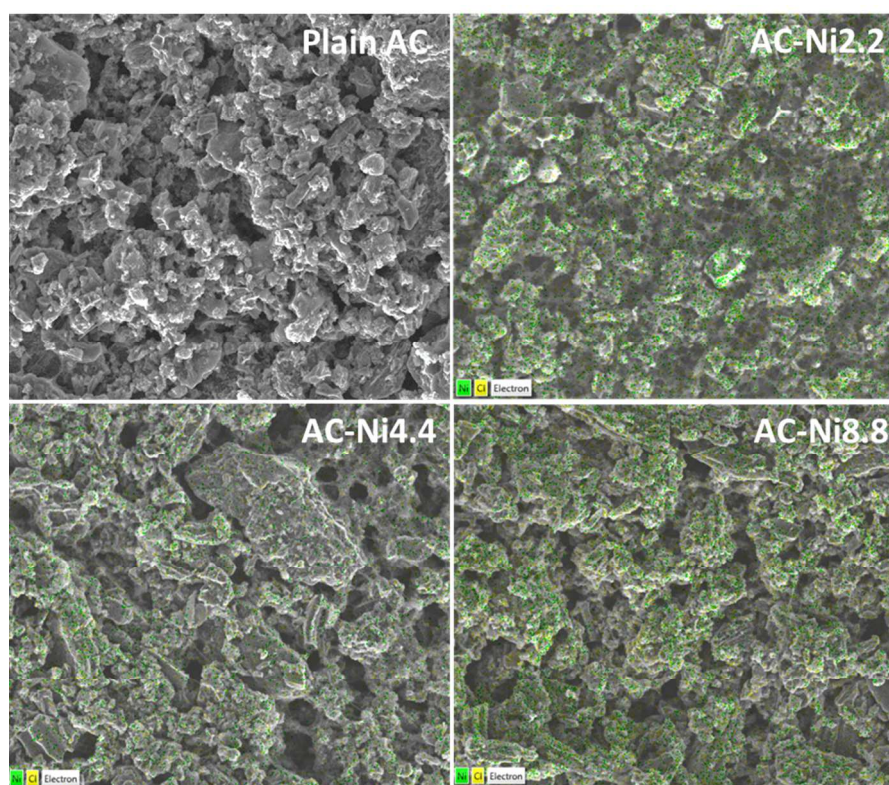
Number of Pages: 9

Number of Figures: 10

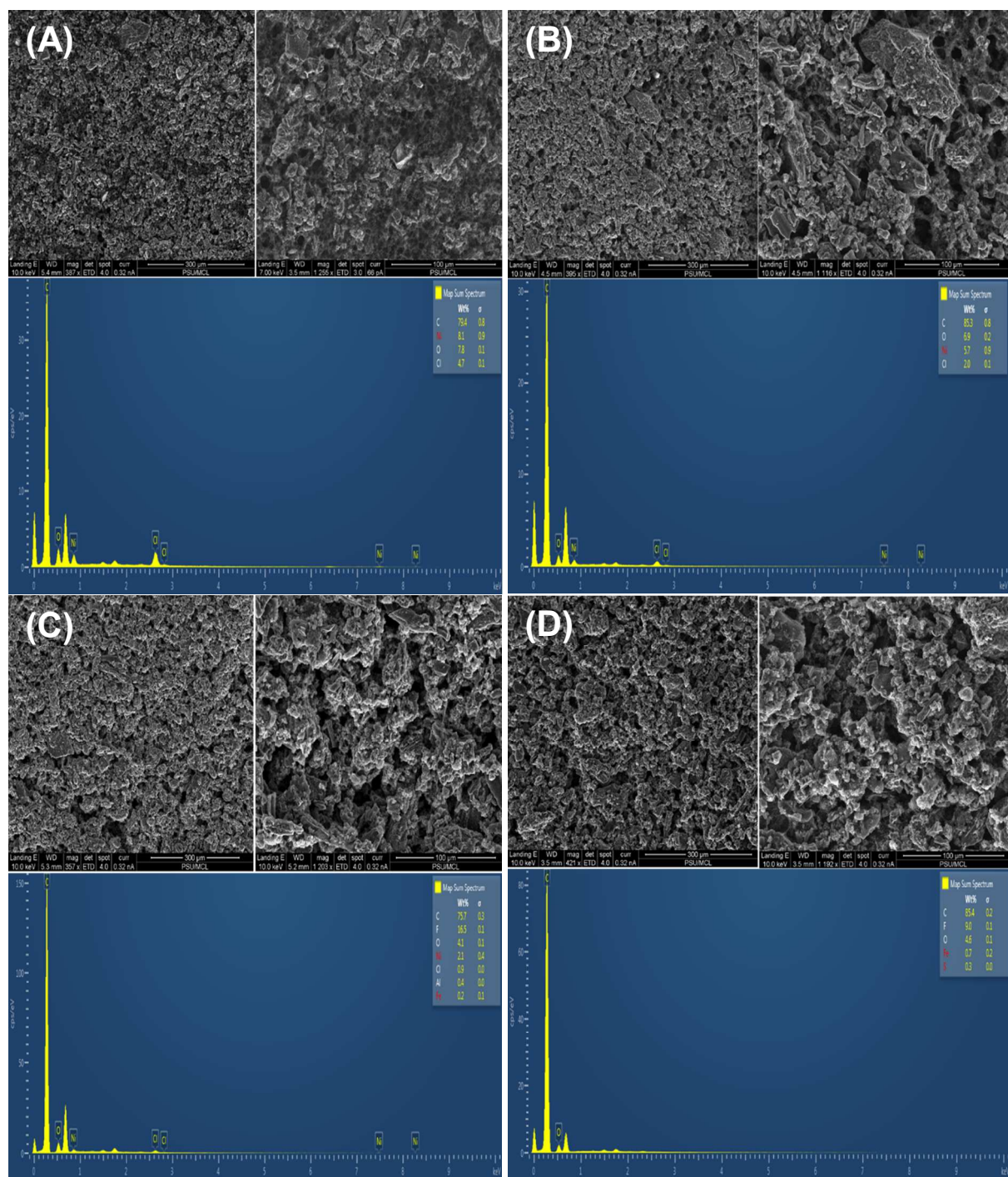
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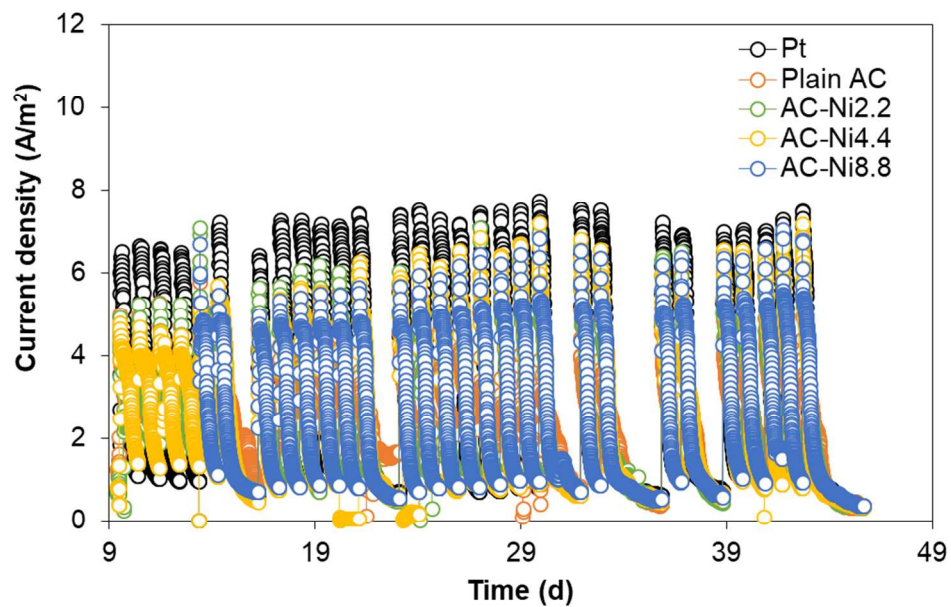
**Figure S1.** Illustration of procedures to fabricate AC-Ni electrodes.



**Figure S2.** EDS images using AC-Ni2.2, AC-Ni4.4, AC-Ni8.8, and plain AC. The colors indicate nickel (green) and chloride (yellow).

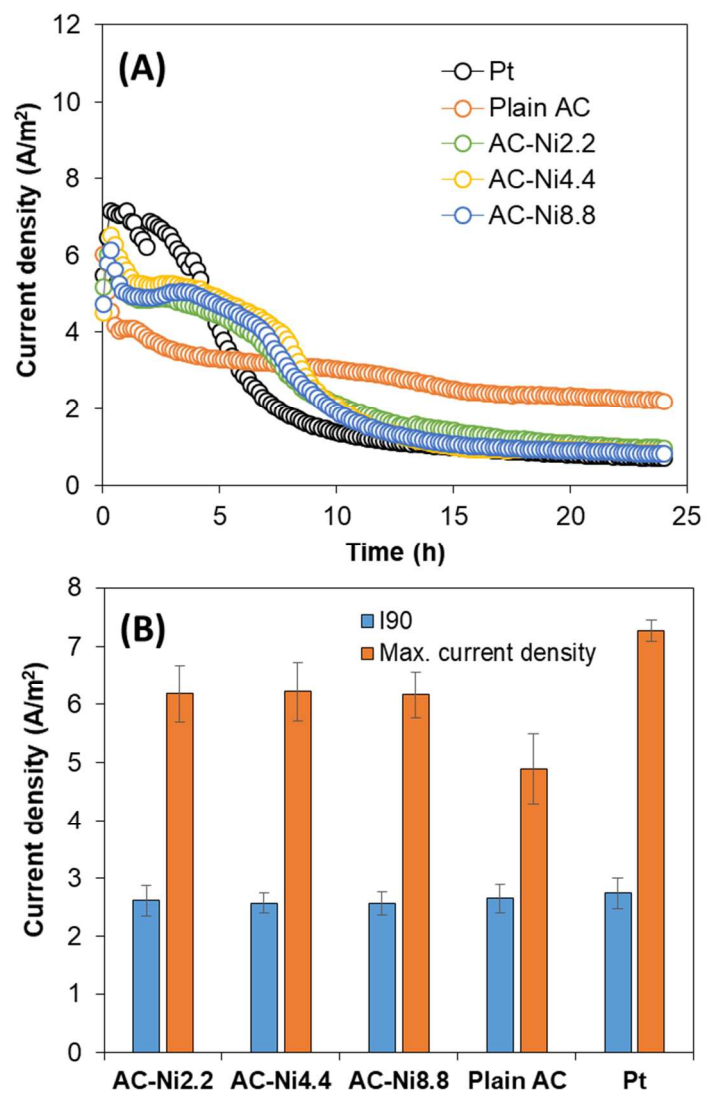


**Figure S3.** SEM images and EDS spectrums of (A) AC-Ni<sub>2.2</sub>, (B) AC-Ni<sub>4.4</sub>, (C) AC-Ni<sub>8.8</sub> and (D) plain AC electrodes.

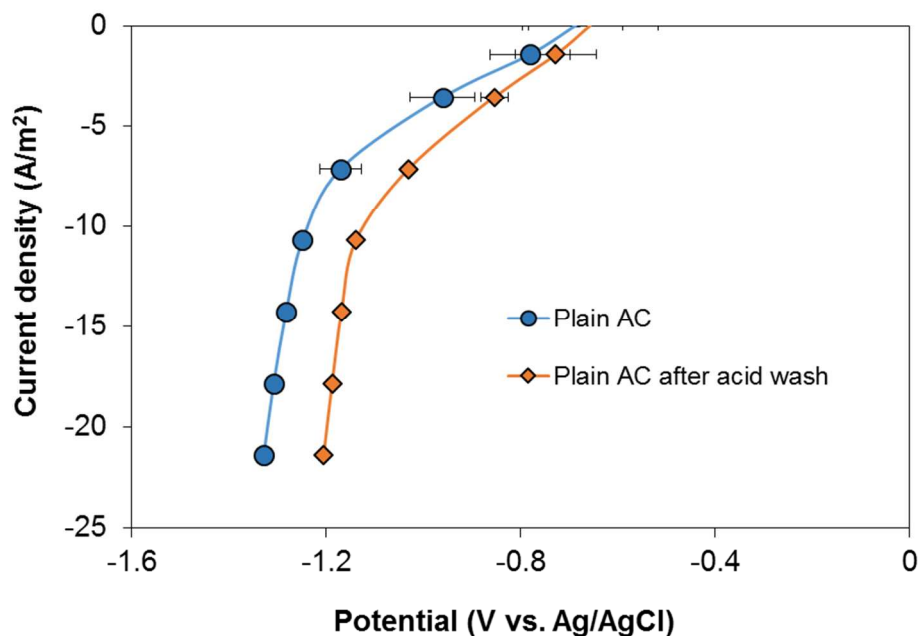


**Figure S4.** Current generations of cubic MECs with AC-Ni, plain AC and Pt cathodes over 30 days. Data was not shown for acclimation period (before day 9).

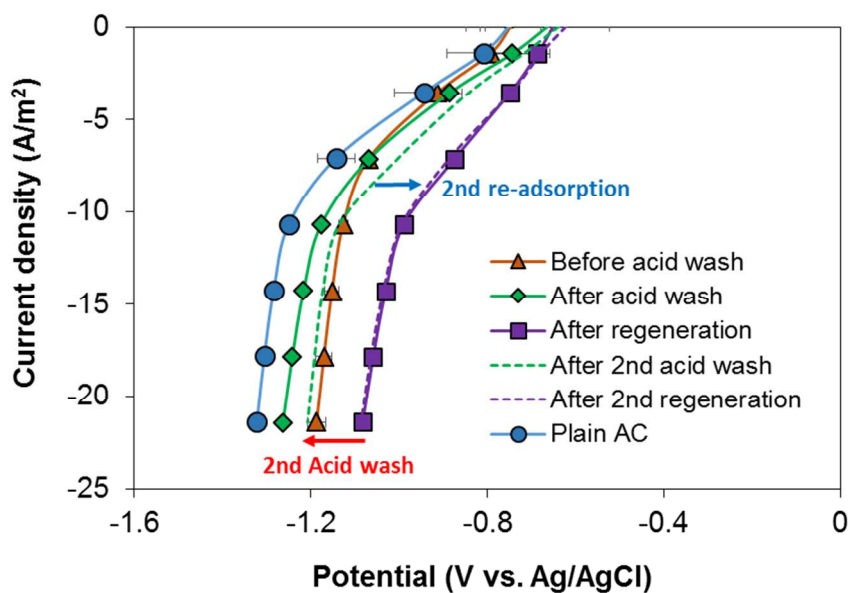




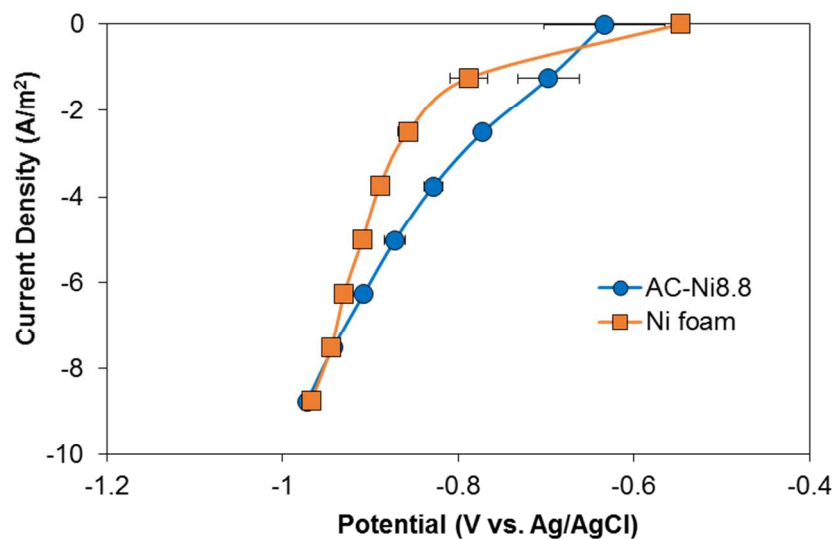
**Figure S5.** (A) Current generations of cubic MECs with AC-Ni, plain AC and Pt cathodes for a single cycle (from day 26), (B) Comparison of maximum and  $I_{90}$  current densities. Error bars indicate standard deviation ( $n \geq 15$ ).



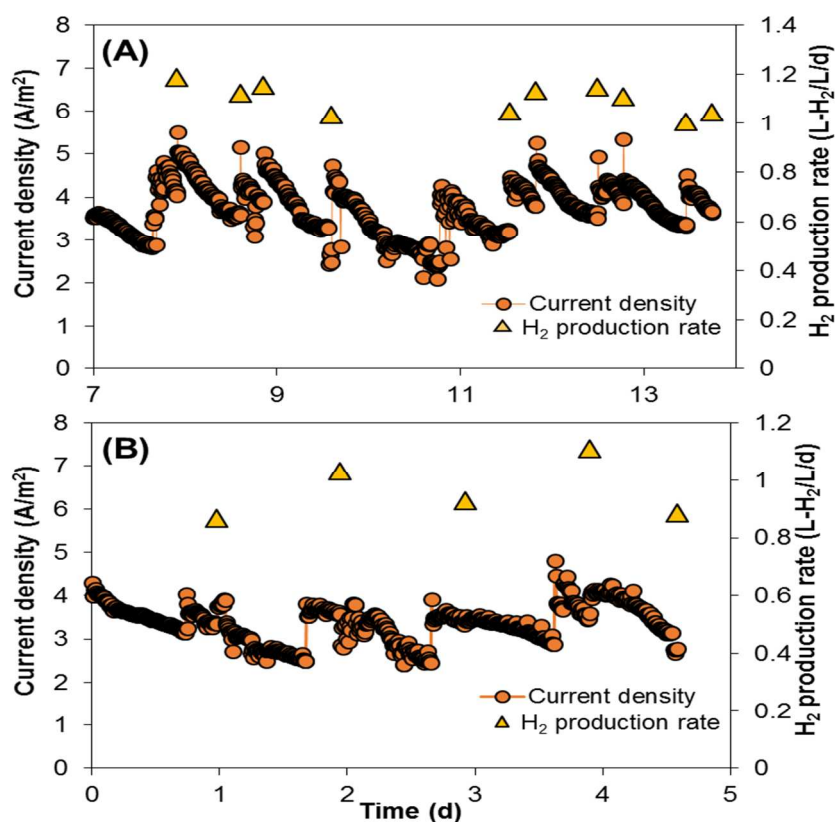
**Figure S6.** Chronopotentiometry (CP) test results for plain AC and acid-washed plain AC electrode. Error bars indicate standard deviation ( $n=3$ ).



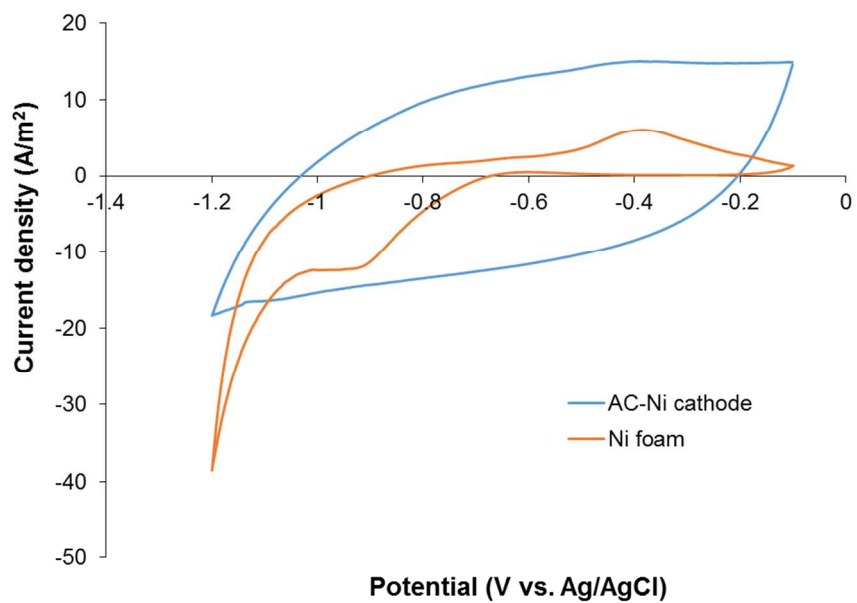
**Figure S7.** Repeated regeneration test results for AC-Ni cathodes. Dashed lines indicate results after a 2nd (repeated) acid wash and regeneration tests. Error bars indicate standard deviation ( $n \geq 2$ ).



**Figure S8.** Chronopotentiometry (CP) test results for the AC-Ni8.8 and Ni foam with catholyte recirculation. Error bars indicate standard deviation (n=3).



**Figure S9.** Continuous current generation and hydrogen production rates of MECs with (A) AC-Ni8.8 cathode and (B) Ni foam in a larger scale MEC (total volume: 168 mL) with catholyte recirculation.



**Figure S10.** Cyclic voltammetry (CV) results for AC-Ni<sub>4.4</sub> cathode (blue) and Ni foam (orange). Voltage was swept from -1.2 V to -0.1 V (vs. Ag/AgCl) at scan rate of 1 mV/sec.



**Table S1.** Estimated price of AC-Ni and Pt/C electrodes for the MECs and commercialized price of Ni foam.

	Catalyst			Binder		Current collector		
	Activated carbon (\$/m <sup>2</sup> )	NiCl <sub>2</sub> · 6H <sub>2</sub> O (\$/m <sup>2</sup> )	Pt/C powder (\$/m <sup>2</sup> )	PVDF (\$/m <sup>2</sup> )	Nafion binder (\$/m <sup>2</sup> )	Nickel foam (\$/m <sup>2</sup> )	SS mesh (\$/m <sup>2</sup> )	Total price (\$/m <sup>2</sup> )
AC-Ni	0.4	4.4	n.a.	1	n.a.	n.a.	12	18
Nickel foam	n.a.	n.a.	n.a.	n.a.	n.a.	20	n.a.	20
Pt/C	n.a.	n.a.	1630	n.a.	210	n.a.	12	1852

\*n.a: Not available

\*\*All price sources can be found at <http://www.alibaba.com>

**Table S2.** Final catholyte pHs of cubic MECs with AC-Ni, plain AC and Pt cathodes after 24 h cycle and the time periods (h) that required for 90% of the coulombs to be generated (I<sub>90</sub> time). Standard deviations indicate n≥15 for final catholyte pHs and n=5 for I<sub>90</sub> time.

	AC-Ni2.2	AC-Ni4.4	AC-Ni8.8	Plain AC	Pt
Final catholyte pH	9.8±0.3	10.1±0.3	10.5±0.2	8.4±0.1	10.2±0.3
I <sub>90</sub> time (h)	18.2±0.3	16.8±0.6	17.3±0.5	20.2±0.7	16.7±0.5