

Supporting information

Vapor-fed cathode microbial electrolysis cells enables greatly improved performance

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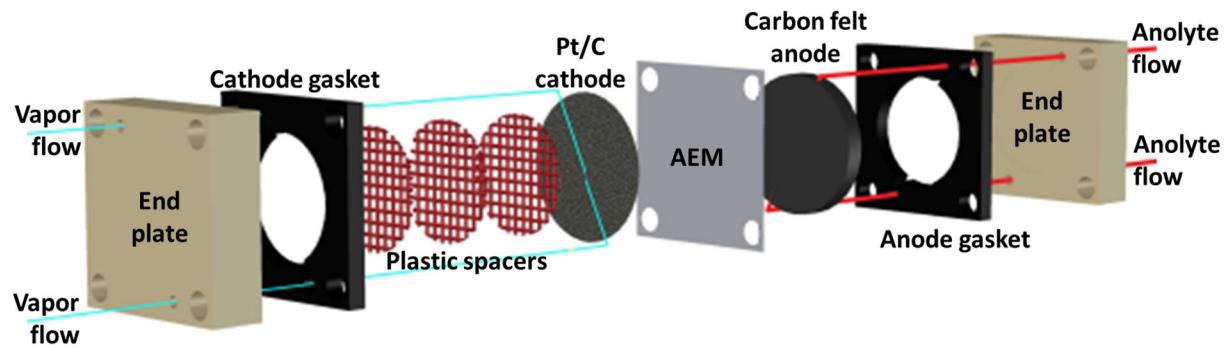


Figure S1. Exploded view of the vapor-fed MEC.

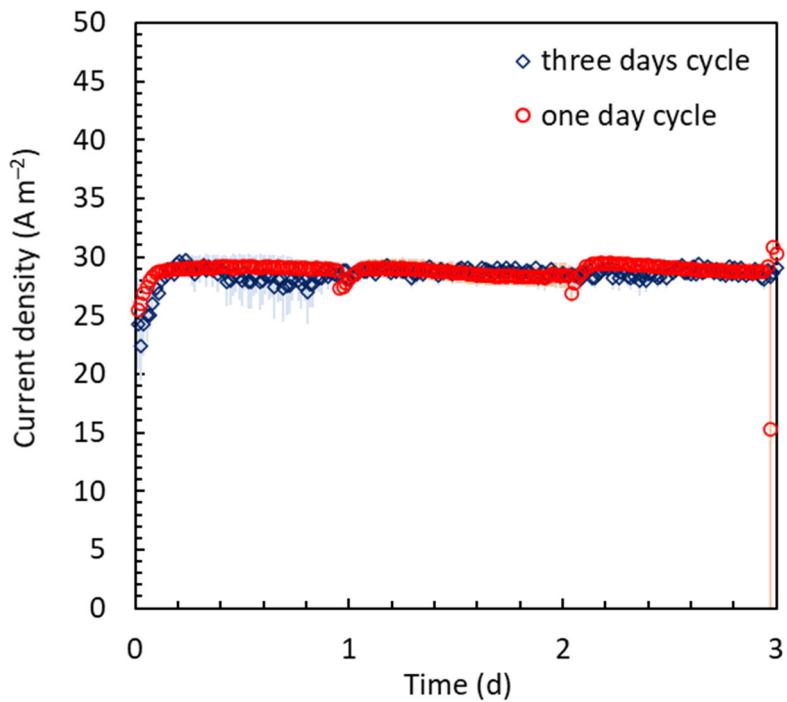


Figure S2. Comparison of the current density delivered by the MEC at $V_{\text{app}} = 0.90$ V over three days by feeding the MEC daily or only once. COD removal ranged from 14 ± 1 % at $V_{\text{app}} = 0.80$ V to 28 ± 2 % at $V_{\text{app}} = 1.10$ V. The COD removal during the three days cycle at $V_{\text{app}} = 0.80$ V was 53 ± 2 %.

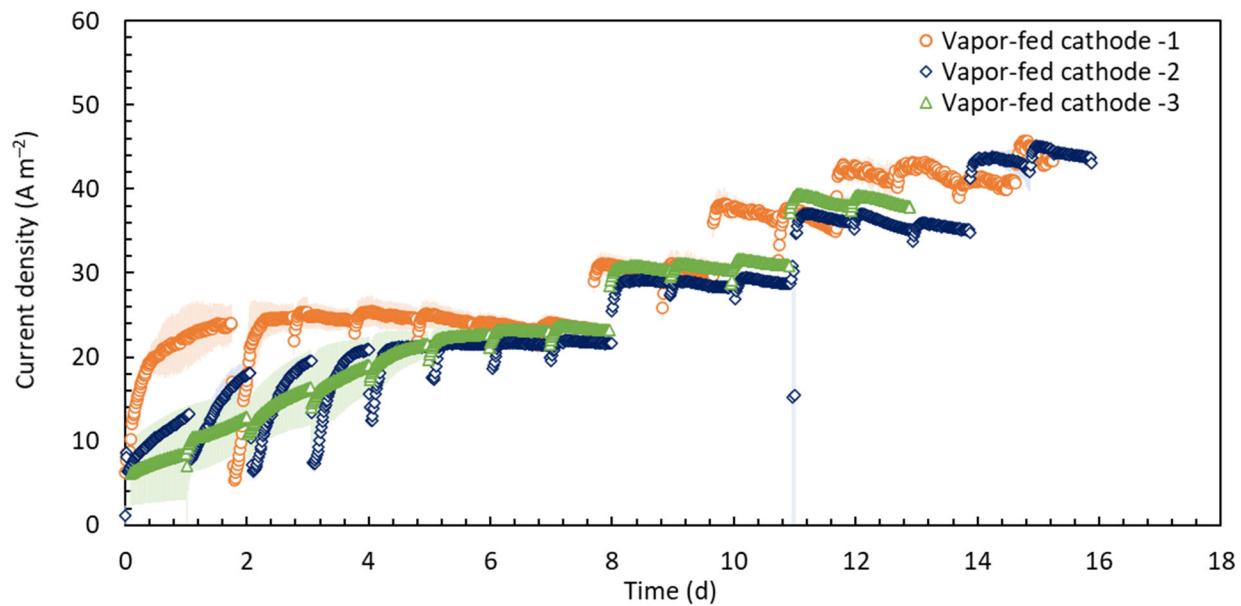


Figure S3. Current density profile over time of the MEC at different applied voltages and operating conditions. The applied voltages were increased from 0.8 V to 1.1 V at different times.

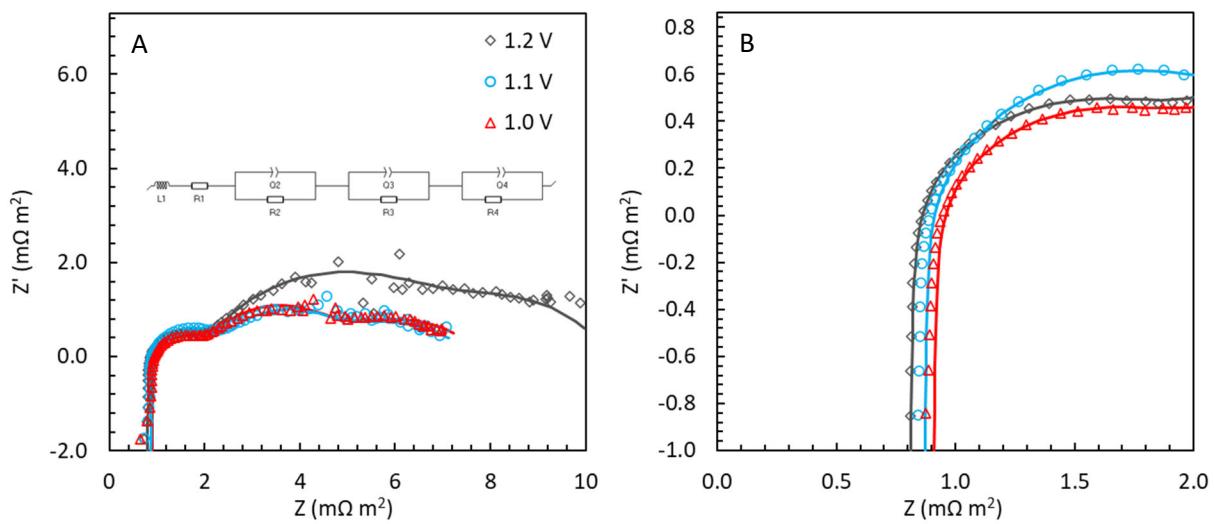


Figure S4. (A) EIS spectra at different applied voltages and corresponding equivalent circuit used for fitting the spectra. (B) Enlargement of the ohmic resistance region.

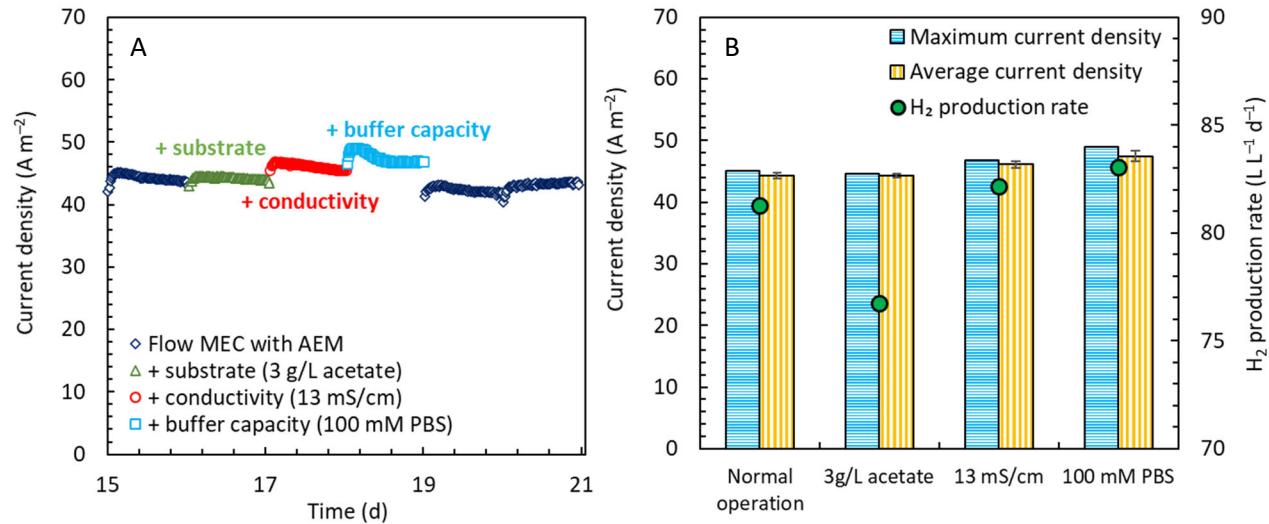


Figure S5. Impact of the additional substrate, solution conductivity and buffer capacity on the (A) current density and (B) H_2 production rate at $V_{\text{app}} = 1.1 \text{ V}$ in the vapor-fed cathode MEC.

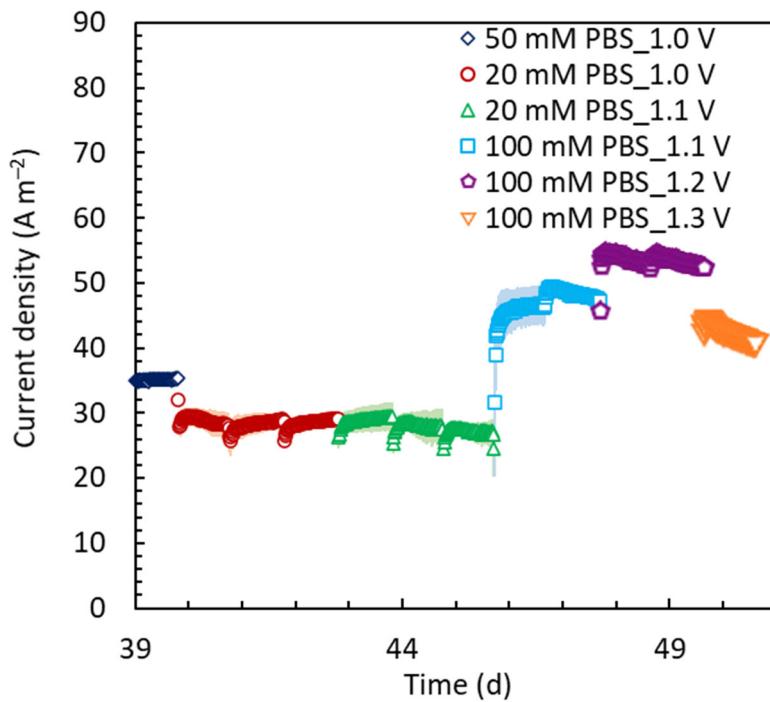


Figure S6. Impact of the buffer capacity and applied voltage on the vapor-fed cathode MEC current density.

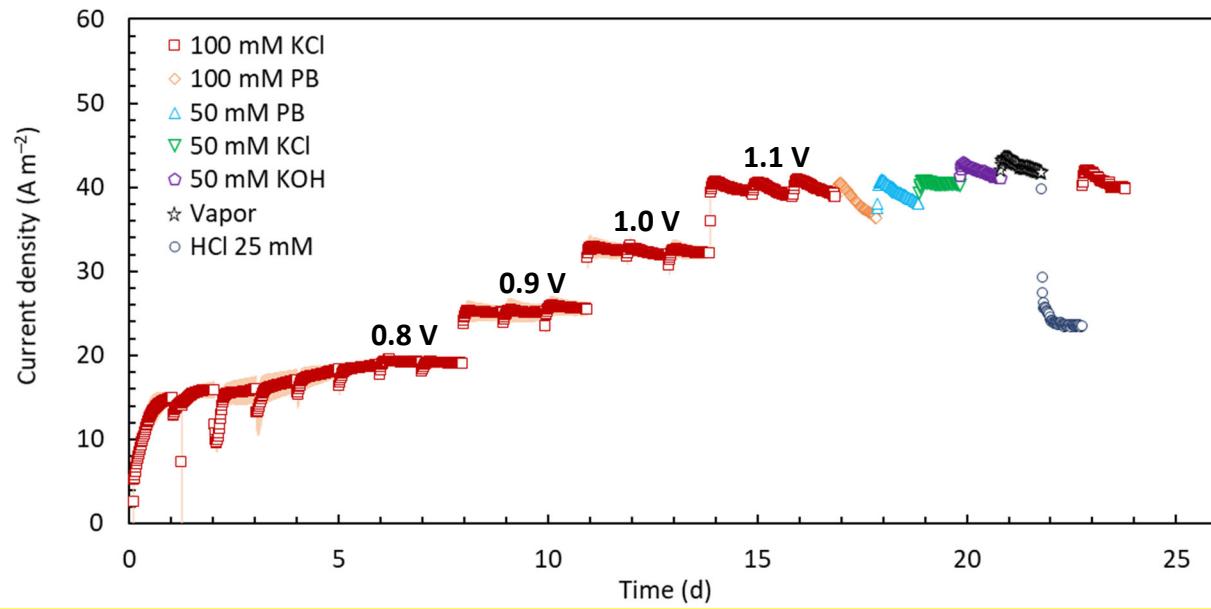


Figure S7. Performance of the liquid catholyte MEC fed different catholyte over time.

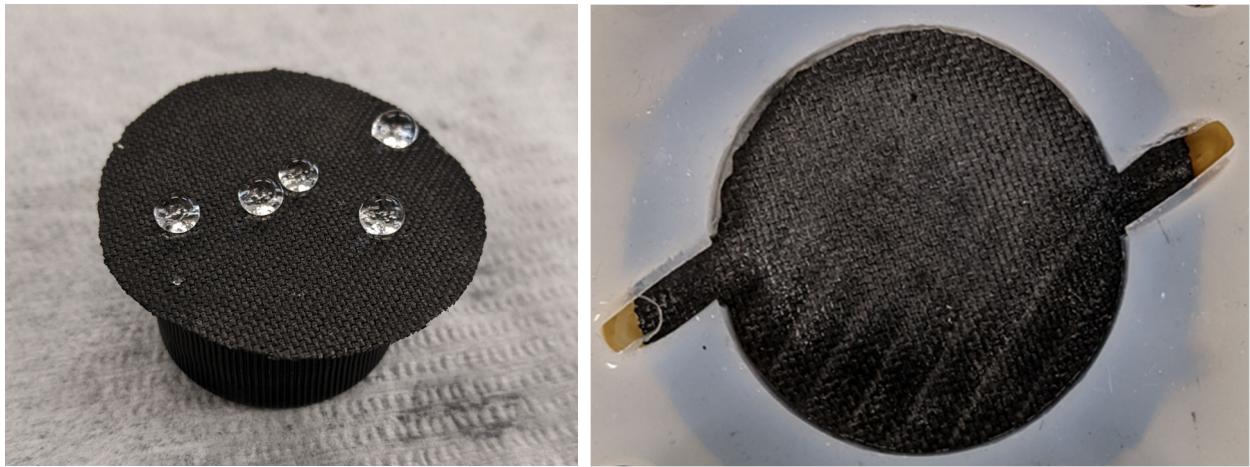


Figure S8. Photos of the electrolyte side of the cathode. The hydrophobic wet-proofed carbon cloth used to support the catalyst layer likely limited the amount of solution reaching the cathode catalyst.

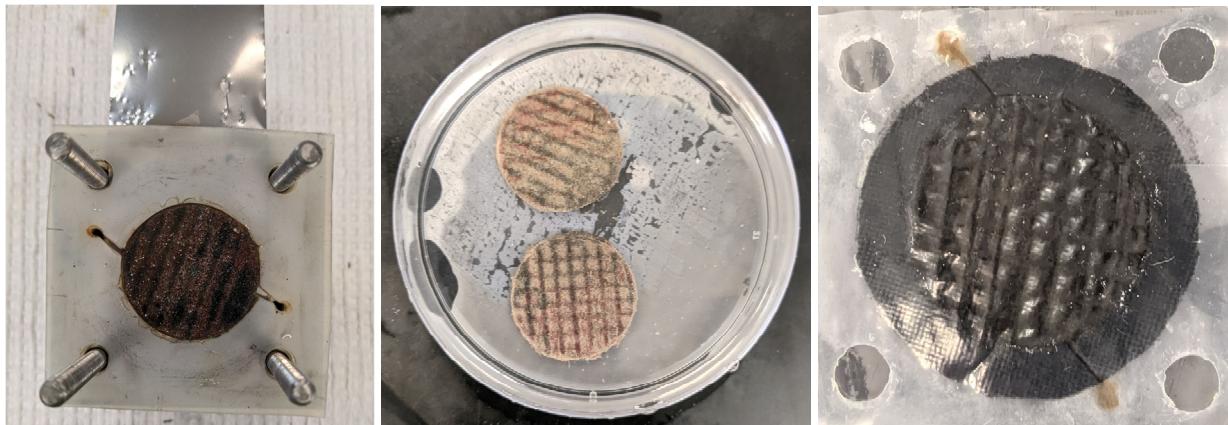


Figure S9. Photos of the anode felts and cathode MEAs used in the vapor-fed MEC.

Table S1: Comparison of the performance of gas-cathode MEC.

Membrane type	Applied voltage (V)	Current density (A/m ²)	H ₂ production rate (L/L-d)	Electrode Area (cm ²)	Reference
AEM	1.1 V	43.1 ± 0.6	72 ± 2	7	This study
CEM	1.2	10.0	0.98	15	²⁵
CEM	1.0	2.25 ± 0.05	0.3	256	¹⁸
AEM	1.0	2.37 ± 0.04	0.3	256	¹⁸
CEM	0 V anode potential vs SHE	17.9 ± 1.6	20 ± 2	11	²⁶
CEM	0 V anode potential vs SHE	5.2 ± 0.2	< 6	11	²⁷

Literature cited:

- (1) Tartakovsky, B.; Manuel, M. F.; Neburchilov, V.; Wang, H.; Guiot, S. R. Biocatalyzed Hydrogen Production in a Continuous Flow Microbial Fuel Cell with a Gas Phase Cathode. *J. Power Sources* **2008**, *182* (1), 291–297. <https://doi.org/10.1016/j.jpowsour.2008.03.062>.
- (2) Rozendal, R. A.; Hamelers, H. V. M.; Molenkamp, R. J.; Buisman, J. N. Performance of Single Chamber Biocatalyzed Electrolysis with Different Types of Ion Exchange Membranes. *Water Res.* **2007**, *41* (9), 1984–1994. <https://doi.org/10.1016/j.watres.2007.01.019>.
- (3) Satinover, S. J.; Schell, D.; Borole, A. P. Achieving High Hydrogen Productivities of 20 L/L-Day via Microbial Electrolysis of Corn Stover Fermentation Products. *Appl. Energy* **2020**, *259* (November 2019), 114126. <https://doi.org/10.1016/j.apenergy.2019.114126>.
- (4) Satinover, S. J.; Rodriguez, M.; Borole, A. P. Microbial Electrolysis Cell Recovery after Inducing Operational Failure Conditions. *Biochem. Eng. J.* **2020**, *164* (September), 107800. <https://doi.org/10.1016/j.bej.2020.107800>.