Supplementary Data:

Maximizing Coulombic recovery and solids reduction from primary sludge by controlling retention time and pH in a flat-plate microbial electrolysis cell

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S1. Calculation of the anode's projected area

We calculated the required projected area of the anode based on the following values:

- Anode volume: 500 mL = 0.5 L
- HRT: 9-day based on a daily feed rate of 56 mL/day
- Expected current density: 1.5 A/m^2
- PS-TCOD$_{in}$: Of the measured TCOD of 8 gCOD/L, we assume that 50% is bioavailable, or 4 gBOD/L[1]

The anode's projected area should be

$$Anode\ projected\ area = \frac{4\ gBOD}{L} \times 0.5L \times \frac{1}{9\ day} \times \frac{1\ e^-}{8\ gBOD} \times \frac{96485\ C}{1\ e^-} \times \frac{1\ day}{24 \times 60 \times 60 sec} \times \frac{m^2}{1.5\ A} = 0.02\ m^2 = 200\ cm^2$$

S2. Analysis of the open-circuit potential in semi-continuous operation

The open-circuit potential (OCP) was measured with no current in an electrochemical cell.[2] OCP values indicate the thermodynamic equilibrium of the electrochemical cell. The theoretical reaction potential and, correspondingly, the OCP changed as the concentration of reactants and products vary. When acetate medium (5 mM of acetate and 5 mM of bicarbonate at pH 7 and 30°C) are used in the anode, the theoretical potential is -0.304 V vs SHE, which corresponds to -0.574 V vs Ag/AgCl. Changing the acetate concentration to 125 mM to match with a similar PS COD concentration (~8 g COD of PS L^{-1}),

$$\frac{8\ gCOD}{L} \times \frac{1\ mol\ of\ acetate}{64\ g\ COD} \times \frac{1000\ mmol}{1\ mol} = 125\ mM$$

the theoretical potential slightly changes to -0.584 V vs Ag/AgCl. The pH has a bigger impact on the potential, a Nernstian voltage drop of ~60 mV per one pH unit.[3]

PS is comprised of complex organic solids that must be hydrolyzed and fermented into simple organic molecules, such as volatile fatty acids. Thus, it is difficult to calculate the exact theoretical potential for PS organics. In our study, we measured the OCP at the anode for each operating condition with PS; the results are summarized in Table S1.

Table S1. Maximum current densities, open-circuit potential, and pH in the batch and semi-continuous experiments

HRTs	Maximum current density (A m^{-2})[1]	Open circuit potential at anode (V)[2]	pH[3]
Batch	1.5 (± 0.71)	-0.113 (± 0.090)	6.5 ~ 8.5
15 day	0.35 (± 0.06)	-0.090 (± 0.026)	7.32 (± 0.24)
12 day	1.08 (± 0.22)	-0.133 (± 0.017)	7.24 (± 0.30)
9 day	2.21 (± 0.28)	-0.228 (± 0.009)	7.49 (± 0.33)
6 day	2.08 (± 0.14)	-0.245 (± 0.013)	8.13 (± 0.29)
12 day neutral pH	1.34 (± 0.11)	-0.197 (± 0.015)	7.30 (± 0.17)
12 day high pH	1.40 (± 0.04)	-0.253 (± 0.008)	8.08 (± 0.11)

[1]Average value on the steady-state condition at each HRT. In the case of batch operations, the value was averaged with two consecutive runs

[2]Open circuit potentials at anode are all versus SHE.

[3]pHs in the semi-continuous operation were all measured at the end of everyday feeding cycle

S3. Long-term operation of the PS-fed MEC

Figures S1 and S2 show the anodes and membranes after long-term operation (~300 days with batch and semi-continuous periods) without changing the materials.

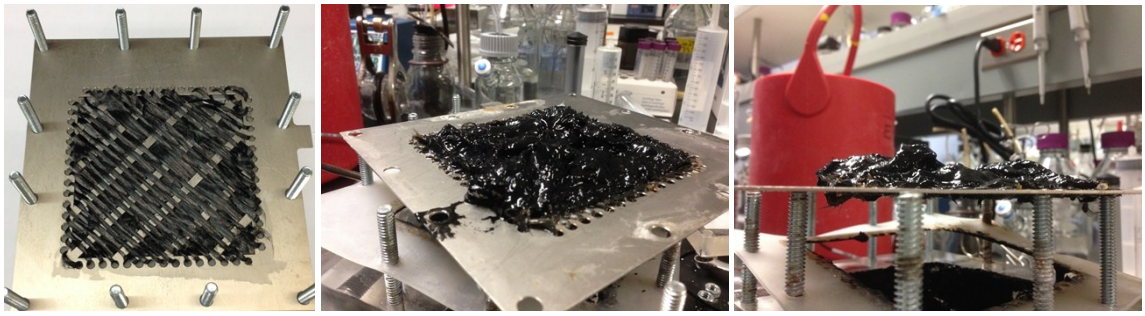

Figure S1. Photos of anode fibers: i) initial (left) and ii) after 12-day HRT at high pH (right).

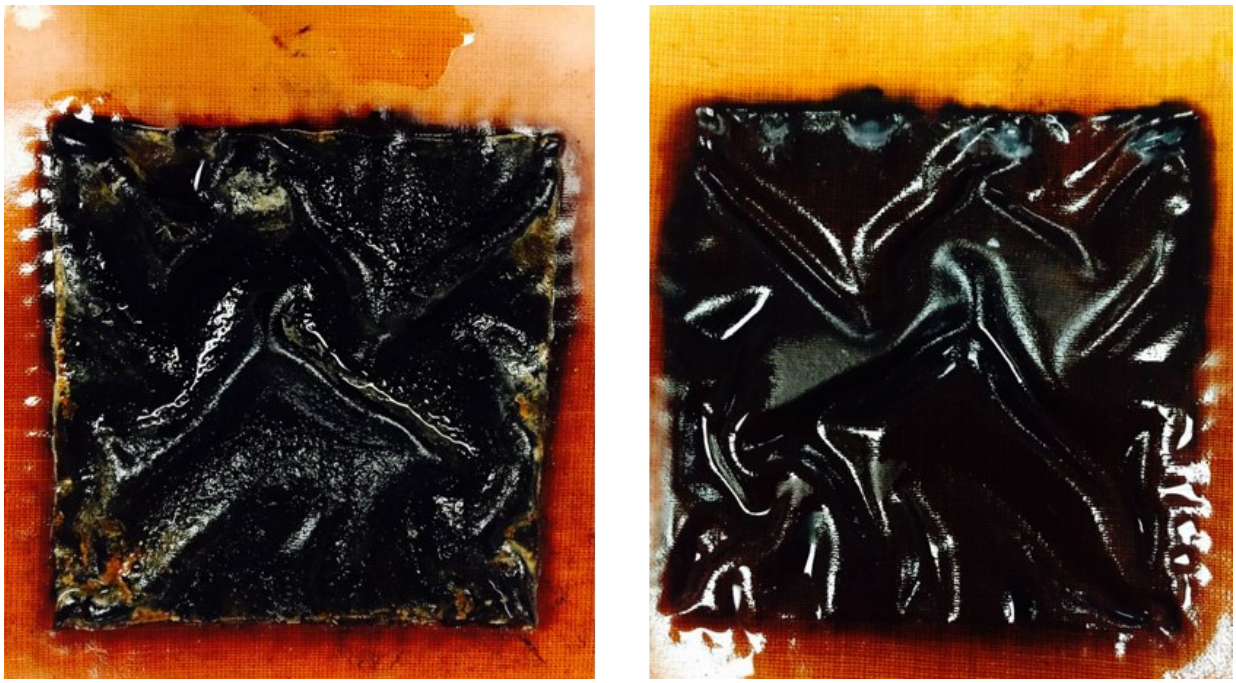

Figure S2. Photos of the membrane at the end of the long-term operation for ~300 days: anode side (left) and cathode side (right).

S4. Sludge dewaterability

Dewaterability of the MEC digested sludge was evaluated using the time-to-filter (TTF) test.[4] In brief, we placed 10 mL of sludge sample onto the filtration device with Whatman No. 1 (47-mm diameter) connected to the graduated Falcon Tubes inside the vacuum flask. After the sample was added to the filter paper and waited 10 min, I turned on the pump with a vacuum pressure 52~56 cm Hg. We recorded the time to collect each mL of filtrate until 5 mL. TTF tests were performed in triplicate at room temperature.

We performed time-to-filter (TTF) tests for PS influent and effluents for both of the final runs with 12-day HRTs. Figure S5 shows that the TTF value of the influent PS, 3.6 min, was reduced substantially by MEC treatment to 1.7 min for the neutral pH and to 0.5 min for high pH. The improved dewaterability with high pH likely was consistent with a previous study observed with AD sludge at high pH.[5]

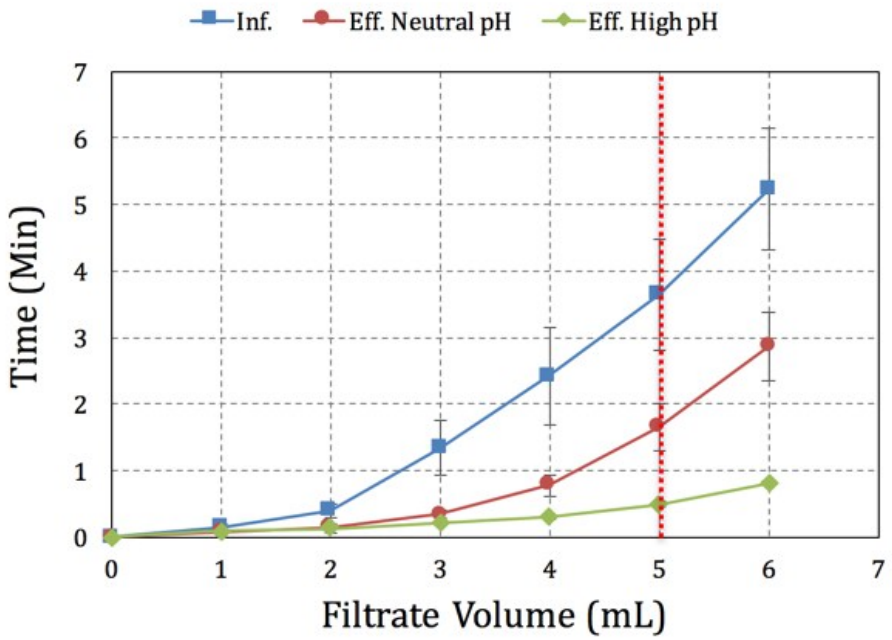

Figure S3. Results of time-to-filter (TTF) tests of PS influent and effluent at 12-day HRT. A vertical dotted red line indicates when 50% of initial loaded sample volume was filtered.

References used in the supplementary data

1. D. Ki, P. Parameswaran, S. C. Popat, B. E. Rittmann and C. I. Torres, *Bioresour. Technol.*, 2015, **195**, 83-88.
2. B. E. Logan, B. Hamelers, R. A. Rozendal, R.A., U. Schrorder, J. Keller, S. Freguia, P. Aelterman, W. Verstraete and K. Rabaey, *Environ. Sci. Technol.*, 2006, **40**, 5181-5192.
3. S. C. Popat and C. I. Torres, *Bioresour. Technol.,* 2015, **215**, 265-273.
4. American Public Health Association, *Standard Methods for the Examination of Water and Wastewater*, Washington D.C., 20th edn., 2012.
5. O. G. Apul, I. Atalar, G. T. Zorba and F. D. Sanin, *Drying Technol.*, 2010, , **28**, 901-909.