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4	Energy self-sustained treatment of swine wastewater in a microbial							
5	electrochemical technology-centered hybrid system							
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The continuous stirred microbial electrochemical system (CSMES) was developed 39 by integrating microbial electrochemical system (MES) and continuous stirred tank 40 reactor (CSTR) into a single design. A three-phase separator was kept to maintain the 41 advantage of CSTR, and simultaneously separated the CSMES into two zones, 42 including the microbial electrochemical zone (MEZ) (Fig. S1b) and complete mixing 43 zone (CMZ)(Fig.S1c). In the CMZ, continuous mixing of the activated sludge and 44 pollutants was achieved by a micro-motor, where high rate of hydrolysis and 45 acidification occurred and volatile fatty acids (VFAs) were harvested. A portion of 46 the VFAs could be transferred to the MEZ, where they were further degraded for 47 electricity generation in the anode. In addition, the removal of VFAs from the CMZ 48 also released their inhibition on methanogens, which might enhance the 49 methanogenesis process in CMZ. 50

The tide-type biocathode microbial electrochemical system (TBMES) was 51 characterized by its unique way of oxygen supply. By using a U-shaped glass siphon 52 outlet for effluent drainage, a continuous feeding and periodical draining process 53 (feeding-draining process) could be accomplished in TBMES. During the feeding-54 draining process, the packed CAC biocathode could contact with air intermittently, 55 which enabled it to entrap air bubbles with the rising of liquid level in the cathode 56 chamber. Therefore, an intermittent aerobic/anoxic condition could be achieved in 57 58 cathode chamber, which was suitable for the occurrence of simultaneous nitrification and denitrification process. Since the liquid level of cathode rose and fell regularly 59



during the feeding-draining process, which was similar to the process of ocean tide,

61 the biocathode was named as "tide-type biocathode".

60

79 Fig. S1 (a) Schematic diagram of the CSMES-TBMES system (b) Microbial
80 electrochemical zone, MEZ (c) Complete mixing zone, CMZ
81





86 Fig. S2 Effects of PAC and PFS addition on removal of (a) COD and (b) SS at various

87	concentrations (PAC:	polvalı	uminium	chloride:	PFS:	poly	ferric	sulphate	e)
•••	• • • • • • • • • • • • • • • • • • • •				••••••••	~-	P ~ - 1		o en price e	-





+0

Time (d)

The ammonia toxicity experiment for CSMES was operated at a fixed COD concentration of 6000 mg L⁻¹ (using sucrose as the carbon source) in continuous flow mode (HRT = 12 h), with the TAN (total ammonia nitrogen) concentration varied from 300 to 1200 mg L⁻¹ and increased in a stepwise manner. The system was operated for at least 15 days at each TAN concentration. Current generation of CSMES at different TAN concentrations was shown in Fig. S4.

114



115

Fig.S4 Current generation of CSMES at different TAN concentrations (TAN: total





Fig. S5 Variation of total ammonium nitrogen concentrations and ammonium nitrogen

removal of CSMES treating swine wastewater in the three-operation phase







149 as NO_3^2 -N removal of TBMES treating the effluent of CSMES in Phase III

173 Mass balance calculation

- 174 (1) COD in the influent of CSMES
- $COD_{CSMES-Inf} = COD_{CSMES} \times Q_{CSMES} \times t = 6745 \text{ mg } L^{-1} \times 0.17 \text{ L } h^{-1} \times 24 \text{ h} = 27591.6 \text{ mg} \cdot d^{-1}$
- 176 (2) COD removal in the CSMES
- $COD_{CSMES-Re} = (COD_{in}-COD_{out}) \times Q_{CSMES} \times t = (6745-1207) mgL^{-1} \times 0.17 Lh^{-1}$
- 178 ¹×24h=22595.1mg·d⁻¹
- 179 (3) COD consumed for electricity generation in the CSMES
- 180 COD_{CSMES-Ele} = COD_{CSMES-Re}×CE=22595.1 mg·d⁻¹×0.6% = 135.6 mg·d⁻¹
- 181 (4) COD in the influent of TBMES
- 182 COD_{TBMES-Inf}=COD_{TBMES}×Q_{TBMES}×t=1207 mg L⁻¹×0.0084 L h⁻¹×24 h=243.3 mg·d⁻¹
- 183 (5) COD removal in the TBMES
- 184 COD_{TBMES-Re}=(COD_{in}-COD_{out})×Q_{TBMES}×t=(1207-128) mgL⁻¹×0.0084 L h⁻¹×24 h=217.5
- $mg \cdot d^{-1}$
- 186 (6) COD consumed for electricity generation in the TBMES
- $COD_{TBMES-Ele} = COD_{TBMES-Re} \times CE = 217.5 \text{ mg} \cdot d^{-1} \times 34.2\% = 74.3 \text{ mg} \cdot d^{-1}$

201 Estimation of energy production and consumption

1. Electrical energy production of CSMES and TBMES 202

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- $EP_{\text{Electric}} = \frac{4I^2 RT}{1000V}$ 204 * MERGEFORMAT (1) $EP_{CH_4} = \frac{Y_{CH_4}q_{CH_4}T\eta_{CH_4}}{24 \times 3600V}$ 205 * MERGEFORMAT (2) 206 $_{D}$ CD

$$EP_{\text{CSMES}} = EP_{\text{Electric}} + EP_{\text{CH}_4} \qquad \land \text{* MERGEFORMAT (3)}$$
$$I^2 R T$$

$$EP_{\text{TBMES}} = \frac{1 - RT}{1000V}$$
207 * MERGEFORMAT (4)
208

- 209 *EP*_{Clectric}—— Electrical energy production by the CSMES based on the average current output, kWh m⁻³; 210
- 211 EP_{CH4}—Electrical energy converted from the energy produced by methane 212 combustion;
- 213 EP_{TBMES} —Electrical energy production by the TBMES based on the average current

214 output, kWh
$$m^{-3}$$
;

215 *I*—— Average current output in stable operation period of Phase III, 0.015 ± 0.001 A

for the CSMES,
$$0.022 \pm 0.018$$
 A for the TBMES;

- 217 *R*—External resistance, 10 Ω for the CSMES, 5 Ω for the TBMES;
- 218 T—Hydraulic retention time, 24 h for the CSMES, 48 h for the TBMES;
- 219 V—Working volume, 4×10^{-3} m³ for the CSMES, 3.9×10^{-4} m³ for the TBMES;
- 220 Y_{CH4} —Methane combustion, 35.8 kJ L⁻¹ CH₄;
- 221 η_{CH4} —Conversion efficiency of methane to electricity, 33%.

222 2. Energy consumption of TBMES

$$EC_{\text{TBMES}} = \frac{gQE}{\eta_{\text{pump}}} \cdot \frac{T}{V}$$

* MERGEFORMAT (5)

224 EC_{TBMES} —Energy consumption of TBMES treating 1 m³ of pretreated swine 225 wastewater, kWh m⁻³;

- 227 Q flow rate of the feeding-draining process, 2.6 $\times 10^{-7}$ m³ s⁻¹;
- 228 E hydraulic pressure head, 0.07 m;
- 229 η_{pump} —energy conversation efficiency of the pump, 64 %¹

Table S1 Detail costs of the components composed the CSMES-TBMES system

Cost analyzia	Manufactures Infa	Unite price	Dosage ^e		Cost (\$)	
Cost analysis	Manufacturer Info		CSMES	TBMES	CSMES	TBMES
Carbon fiber brush	Toray, 12K carbon fiber	1.5 \$ cm ⁻¹	120 cm	15 cm	180	22.5
ACAC ^a	Nankai University	$60 \ \text{m}^{-2 \ \text{d}}$	$0.03 \ m^2$		1.8	_
CAC ^b	Beijing Sanye Carbon Co. Ltd.	0.5 \$ kg ⁻¹	—	0.35 kg	_	0.175
Stainless steel mesh	Hebei Enfu Stainless Steel Mesh Co.	2.5 \$ m ⁻²	_	0.01 m ²	_	0.025
PTFE	Shanghai Hesen Electric Co. Ltd.	67 \$ L ⁻¹	_	0.098 L	_	6.5
Glass fiber separator		8 \$ m ⁻²	—	0.18 m ²	_	1.4
Reactor	Harbin Hongda Perspex Factory	230/30 \$	1	1	230	30
Others ^c		50/7.5\$			50	7.5
Total					461.8	66.7

b: CAC: columnar activated carbon

c: The item "other" mainly includes the cost of DI water, screws, glue, necessary tools etc.

d: The cost of ACAC was calculated to be $30 \sim 60 \ \text{sm}^{-2}$, $60 \ \text{sm}^{-2}$ was chosen in this calculation

e: The dosage is based on the use for CSMES or TBMES

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240 **References**

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