**Electronic Supplementary Information** 

## Paper-based Microfluidic Aluminum-Air Batteries: Toward Next-Generation Miniaturized Power Supply

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Fig. S1 (a) Dimensions of the anode and cathode electrodes. (b) Display of the prototypical paper-based  $\mu$ Al-air battery from top and bottom view. (c) A prototypical paper-based  $\mu$ Al-air battery during discharge.



**Fig. S2** (a) Flow rate measurement of the electrolyte (1.5 M KOH solution) on paper channel (VWR grade 417). (b) The volume of the electrolyte accumulated on absorbent pads was plotted against elapsed time. The flow rate calculated by linear regression is marked.



**Fig. S3** A paper-based  $\mu$ Al-air pouch battery before (a-1) and during discharging (a-2). Two paper-based  $\mu$ Al-air pouch batteries connected in series before (b-1) and during discharging (b-2). (c) Polarization (*V-I*) and power density curve of a single paper-based  $\mu$ Al-air battery in pouch configuration.



**Fig. S4** Polarization (*V-I*) and power density curves of paper-based  $\mu$ Al-air battery with 1.5 M (a), 2.0 M (b) and 2.5 M (c) KOH solution by continuous staircase linear scan voltammetry.



**Fig. S5** Display of the prototypical paper-based non-fluidic Al-air battery from top (a) and bottom (b) view. Reaction area for both anode and cathode electrodes are  $8 \times 5 \text{ mm}^2$ . The size of paper is  $10 \times 7 \text{ mm}^2$ .



Fig. S6 Discharge curves of paper-based  $\mu$ Al-air battery (a) and non-fluidic Al-air battery (b) under different current density (5, 10, 20 mA/cm<sup>2</sup>). The insets are the optical photos of the anode aluminum foil after discharge.



Fig. S7 Aluminum foil (size 8×5 mm<sup>2</sup>) immersed in 1.5 M KOH solution.



**Fig. S8** Performance of non-fluidic Al-air battery using gas diffusion layer air electrode at the cathode. (a) Polarization (*V-I*) and power density curves recorded at different scan numbers. (b) Discharge curves of

paper-based and non-fluidic Al-air battery using gas diffusion layer air electrode under different current density (5, 10, 20 mA/cm<sup>2</sup>). The insets are the optical photos of the anode aluminum foil after discharge.



**Fig. S9** Polarization (*V-I*) and power density curve (a) and discharge curve (b) of the paper-based µAl-air battery by using 2 M NaCl as electrolyte. The insets are the optical photos of the anode aluminum foil after discharge.

Anode	Cathode	Electrolyte	OCV <sup>a</sup> (V)	Maximum Power density (mW/cm <sup>2</sup> )	Discharge V (V) @ I (mA/cm <sup>2</sup> ) <sup>b</sup>	Specific capacity (Ah/g)	Energy density (Wh/kg)	Features	Ref <sup>i</sup>
Al foil	MnO <sub>2</sub> in carbon	EMIm(HF) <sub>2.3</sub> F <sup>c</sup>			1.1@1.5	2.1 <sup>f</sup>	2300 <sup>f</sup>	Primary Al-air battery with non- aqueous electrolyte	1
Al	N, S doped porous carbon	6 M KOH	1.64	46	0.9@20	0.288	260	Primary Al-air battery	2
Al	La <sub>0.7</sub> (Sr <sub>0.15</sub> Pd <sub>0.15</sub> )MnO <sub>3</sub>	4 М КОН	1.8	265.6	-	-	-	Primary Al-air battery	3
Al	carbon fiber paper supported Ag	4 M NaOH	1.85	109.5	1.56@30	2.78 <sup>f</sup>	4340 <sup>f</sup>	Primary Al-air battery	4
Carbon treated Al	MnO <sub>2</sub> pasted on Ni mesh	2 M NaCl	-	-	0.4@10	1.2	480	Primary Al-air battery	5
Al mesh	La <sub>2</sub> O <sub>3</sub> , SrO, MnO <sub>2</sub> and carbon on Ni foam	KOH gel	-	91.13	1.2@18	1.166 <sup>f</sup>	1230 <sup>f</sup>	All-solid-state battery	6
Ultrafine- grained Al	Ag powder on Ni mesh	2 M NaCl	-	-	0.388@10	2.72 <sup>f</sup>	960 <sup>f</sup>	Primary Al-air battery	7
		4 M KOH	-	-	1.45@10	2.48 f	3600 <sup>f</sup>		
Al alloy	silver manganate nanoplates loaded air electrode	6 М КОН	1.4	105	0.91@100	$2.84^{\rm f}$	2550 <sup>f</sup>	Primary Al-air flow battery (electrolyte flow rate 100 mL/min) controlled by a pump	8
6061					1.10@50	2.64 <sup>f</sup>	2540 <sup>f</sup>		
Al wire	MWCNT coated paper	BPS <sup>c</sup>	0.7	0.38 <sup>d</sup>	-	-	-	Cable-shaped flexible battery	9
Al spring	Ag coated CNT <sup>c</sup> sheet	Hydrogel PVA <sup>c</sup> KOH	1.7	1.33	1.25@0.5	0.935 <sup>f</sup>	1168 <sup>f</sup>	All-solid-state, fiber-shape, stretchable battery	10
Al foil	Carbon on steel mesh	1.5 M KOH	1.27	0.6				Disposable, small-size, paper- based battery	11
Al foil	Carbon black	12 wt% NaCl	0.7	0.015	0.54@0.005	0.496 <sup>f</sup>	270 <sup>f</sup>	- Shape-reconfigurable battery	12
						0.128 <sup>g</sup>	69 <sup>g</sup>		
Al foil	Mn <sub>3</sub> O <sub>4</sub> /C on carbon paper	SiO <sub>2</sub> -modified chitosan hydrogel	1.43	3.8	1.05@1	0.289 <sup>f</sup>	303 <sup>f</sup>	Al-air coin battery with SiO <sub>2</sub> - modified chitosan hydrogel membrane	13

Table S1. Comparisons of the metal-air batteries

Al foil	Carbon paper	5 M NaOH	1.6	21	0.9@10	1.273 <sup>g</sup>	1150 <sup>g</sup>	Paper-based Al-air battery with suppressed Al corrosion	14
Al foil	MnO <sub>2</sub> /CNT <sup>c</sup> on carbon paper	4 M NaCl	1.5	-	0.6@1	2.462 <sup>g</sup>	1480 <sup>g</sup>	Paper-based flexible battery	15
Al ink	MnO <sub>2</sub> /CNT <sup>c</sup> on paper	4 M NaCl	0.8	-	0.6-0.2@1	0.814 <sup>g</sup>	-	Paper-based paintable battery	
Zn film	LaNiO3/NCNT <sup>c</sup>	PVA <sup>c</sup> gelled KOH	1.3	28	1.2@1	0.45 <sup>g</sup>	581 <sup>g</sup>	Flexible, polymer-electrolyte, rechargeable battery	16
Spiral Zn plate	Fe/N/C on air electrode	alkline gel polymer	1.14	-	0.8@0.1	4.5 Ah/L <sup>h</sup>	3.6 Wh/L <sup>h</sup>	All-solid-state cable-type flexible battery	17
Zn spring	RuO <sub>2</sub> on CNT <sup>c</sup>	Hydrogel polymer	1.29	-	1@1°	6 Ah/L <sup>h</sup>	5.7 Wh/L <sup>h</sup>	Rechargeable, flexible, stretchable, fiber-shape battery	18
Zn foil	NCNF <sup>a</sup> film cathode	Alkaline PVA <sup>c</sup> gel	-	-	1.0@2	0.378 <sup>f</sup>	378 <sup>f</sup>	Flexible battery with good mechanical and cycling stability	19
Zn foil	Co/N/O tri-doped graphene mesh	Alkaline PVA <sup>c</sup> gel	1.45	28	1.19@1.0	-	-	Rechargeable, bendable, flexible, solid battery	20
Zn film	Ultrathin Co <sub>3</sub> O <sub>4</sub> on carbon cloth	PVA <sup>c</sup> gel KOH	1.3	17	1.05@2	0.542 <sup>f</sup>	546 <sup>f</sup>	Flexible thin Zn-air battery	21
Zn plate	CuCo <sub>2</sub> S <sub>4</sub> nanosheets	PVA <sup>c</sup> gel KOH	1.2	-	1.28@1.0	0.331 <sup>f</sup>	424 <sup>f</sup>	Flexible all solid Zn-air battery	22
EGILM <sup>a</sup>	Carbon fiber@Pt	PAA <sup>c</sup> gel KOH	1.85	1.25	1.2@0.5	0.21 <sup>f</sup>	250 <sup>f</sup>	Cable-shaped, soft, elastic, flexible liquid metal-air battery	23
Al foil	Pd/C on graphite foil	1.5 M KOH	1.55	22.5	1.05@20	4.1 Ah/L <sup>h</sup>	4.3 Ah/L <sup>h</sup>	Small, thin, low-cost paper-based µAl-air battery with high power and energy density (electrolyte flow rate 24 µL/min)	
					1.05@20	2.75 <sup>g</sup>	2900 <sup>g</sup>		This
					1.17@10	2.11 <sup>g</sup>	2470 <sup>g</sup>		work
					1.30@5	1.47 <sup>g</sup>	1910 <sup>g</sup>		
		2 M NaCl	0.70	11.3	0.46@20	2.20 <sup>g</sup>	1010 <sup>g</sup>		

<sup>a</sup>Open circuit voltage

<sup>b</sup>Discharge voltage (V) at discharge current (mA/cm<sup>2</sup>)

<sup>c</sup>EMIm(HF)<sub>2.3</sub>F: 1-ethyl-3-methylimidazolium oligo-fluoro-hydrogenate; BPS: phosphate-buffered saline; CNT: carbon nanotubes; NCNT: nitrogen-doped carbon nanotubes; PAA: poly acrylic acid; NCNF: nanoporous carbon nanofiber films; PVA:

polyvinyl alcohol; EGILM: Eutectic gallium-indium liquid metal.

<sup>d</sup>0.38 mW/cm.

 $^{\rm e}1$  V at 1 A/g.

<sup>f</sup>The capacity and energy density was calculated based on the consumed Al mass.

<sup>g</sup>The capacity and energy density was calculated based on the whole Al mass.

<sup>h</sup>The capacity and energy density was calculated based on the whole volume of the device.

<sup>i</sup>The primary batteries are shown in black text (Ref 1-8). The miniaturized batteries are shown in blue text (Ref 9-23).



Fig. S10 Mechanically recharging tests. Discharge curves of three paper-based  $\mu$ Al-air battery by using the same cathode electrode.



**Fig. S11** (a) Discharge curves of three paper-based non-fluidic Al-air battery by using the same anode electrode, but new cathode electrode as well as paper and electrolyte. (b, c) Discharge curves of three paper-based non-fluidic Al-air battery by using the same cathode electrode, but new anode electrode as well as paper and electrolyte; (b) the cathode was used without drying; (c) the cathode was dried in vacuum for 2 h after each discharge, and then reassembled into a non-fluidic Al-air battery for the next discharge.



**Fig. S12** Polarization (*V-I*) and power density curves of paper-based  $\mu$ Al-air batteries with different electrode sizes.



**Fig. S13** Running time measurement of a mini-fan (power rating 10 mW) powered by a paper-based  $\mu$ Alair battery (geometric electrode active area: 8×15 mm<sup>2</sup>). The inset shows that after 70 min discharge the Al anode is almost fully consumed.

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