Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Integrated-milliampere-level hydroelectric generator utilizing chemical-doped P-type and N-type graphite

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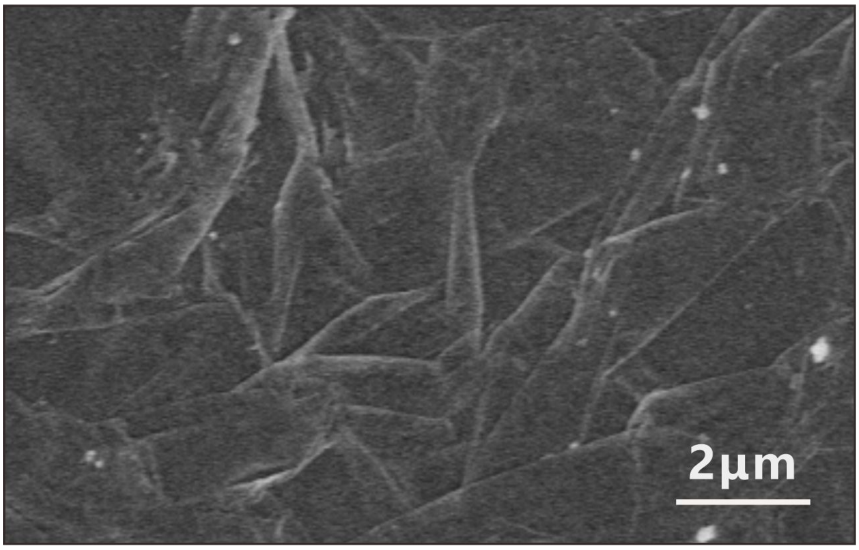

Figure S1. SEM image for the surface morphology of a typical graphite foil prepared.

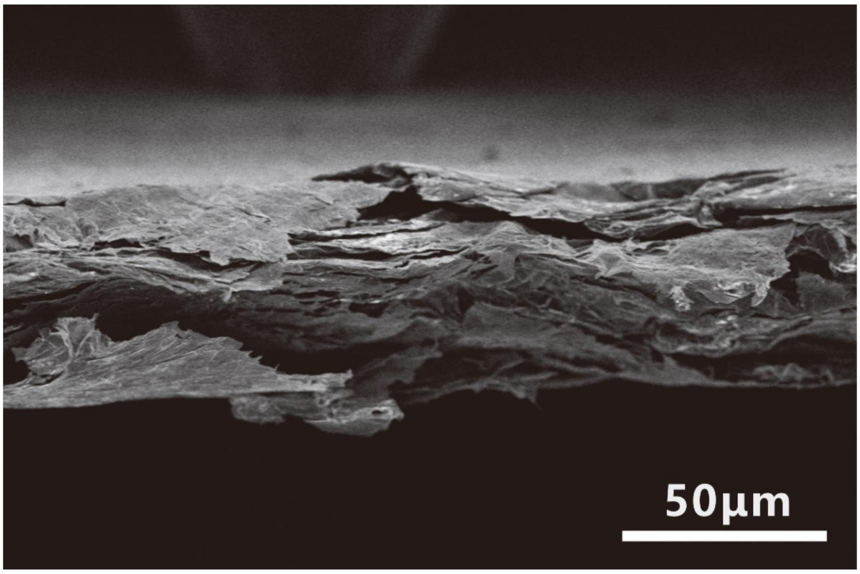

Figure S2. SEM image for the cross-section morphology of a typical graphite foil prepared.

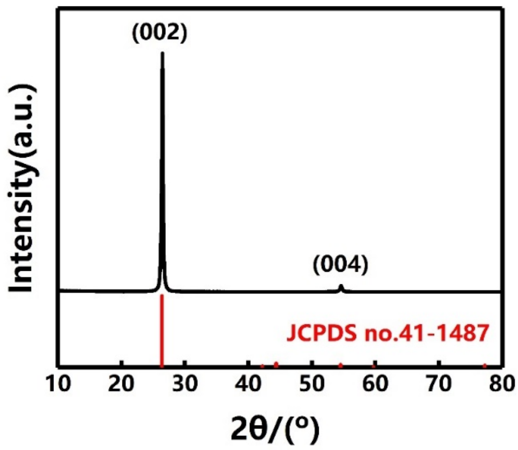

Figure S3. The XRD patterns of graphite foil and the standard card of graphite (JCPDS no. 411487).

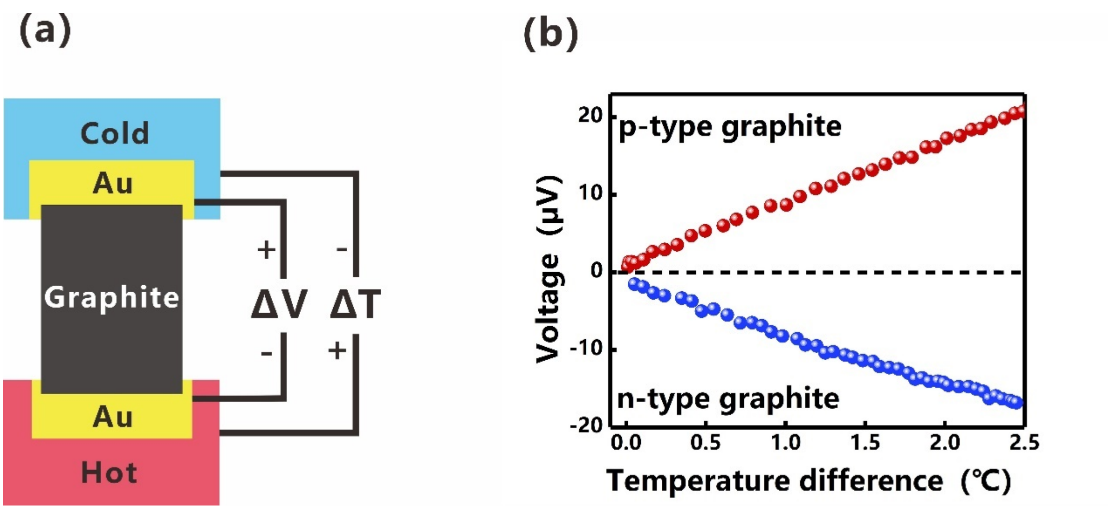

Figure S4. (a) Schematic illustration of the apparatus for measuring Seebeck coefficient at room temperature in in nitrogen atmospheres. (b) Measurement results of the P-type graphite and the N-type graphite.

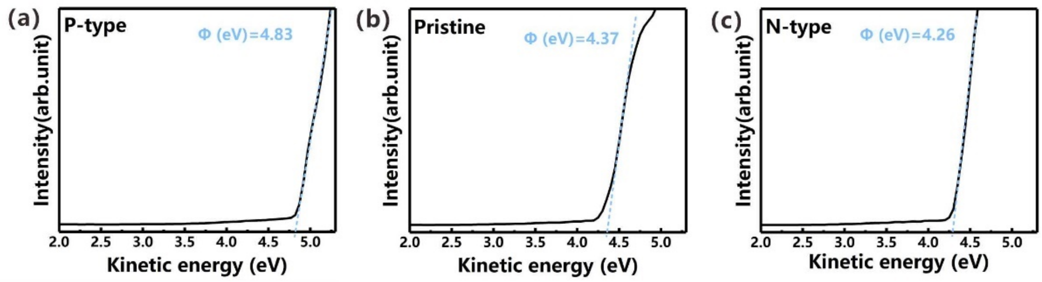

Figure S5. UPS spectra around the secondary-electron cutoff edge for (a) P-type, (b) Pristine and (c) N-type graphite.

The value of work function, defined as the difference between the vacuum level and the Fermi level, is calculated by the following equation:

$$\varphi = h\nu - (E_f - E_{cutoff})$$

Where φ is the work function, hν is the stimulating photonic energy (21.22 eV in this test), E_f is the Fermi edge energy, and E_{cutoff} is the cutoff edge energy. Shift the UPS data to align the Fermi edge energy with the stimulating photonic energy, ensuring that the value of the work function matches the cut-off edge energy.

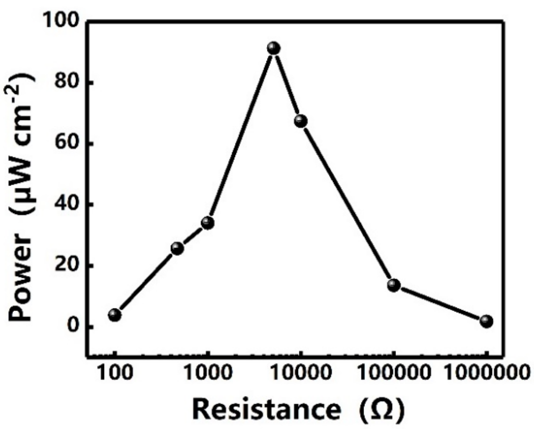

Figure S6. Power density of the WG-HEG as a function of external resistance ranging from 100 Ω to 10^6 Ω.

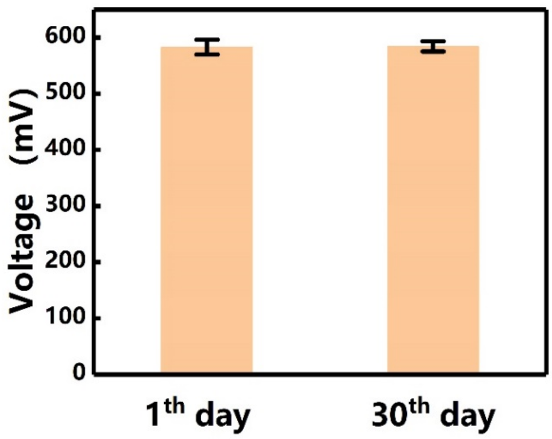

Figure S7. Open-circuit voltage from samples after being exposed to the ambient environment for 30 days and relatively fresh samples. The error bars in this supporting information represent the standard deviation of the statistical means from multiple measurements (n is 5 or more).

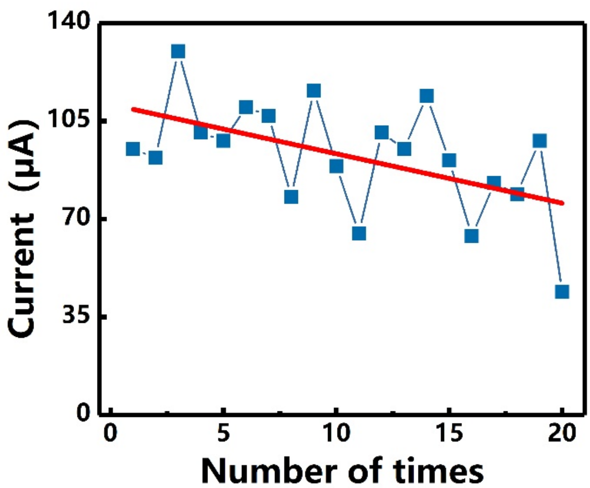

Figure S8. The current output of the WG-HEG was measured in response to multiple immersions of graphene in and out of tap water.

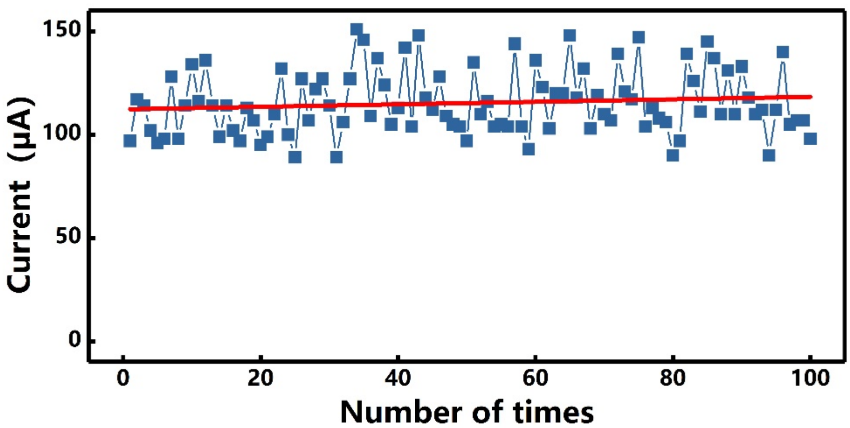

Figure S9. The current output of the WG-HEG was measured in response to multiple immersions of graphene in and out of tap water. The sample was subjected to simple impregnation after every 10 discharges.

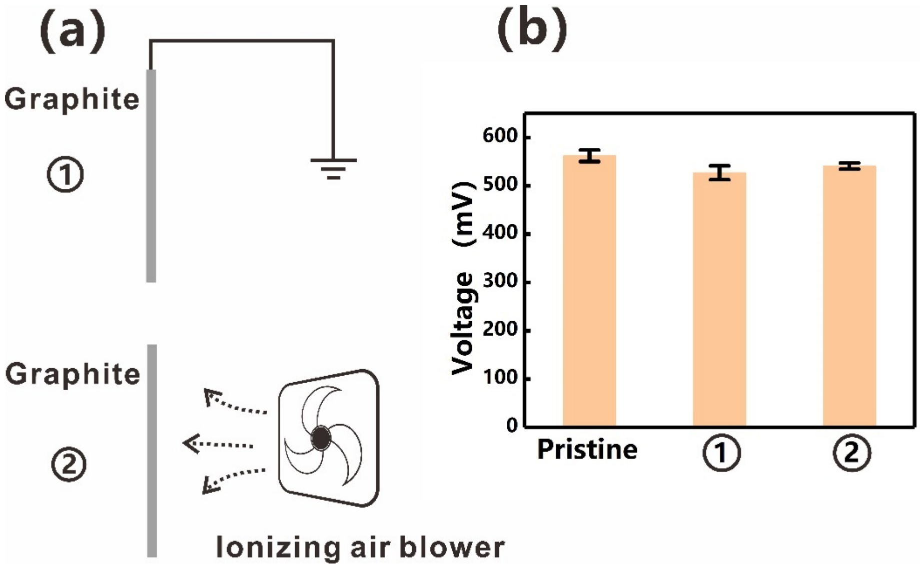

Figure S10.(a) ①Grounding and ②using an ionizing air blower for 1 hour. (b) Measurement results after removing possible pre-existing charges on surfaces.

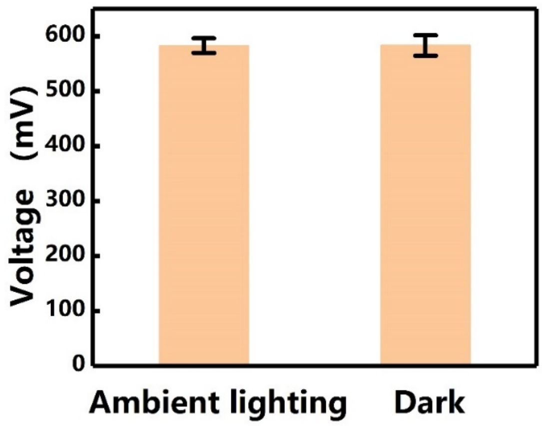

Figure S11. Open-circuit voltage from the WG-HEG devices with ambient lighting and in the dark.

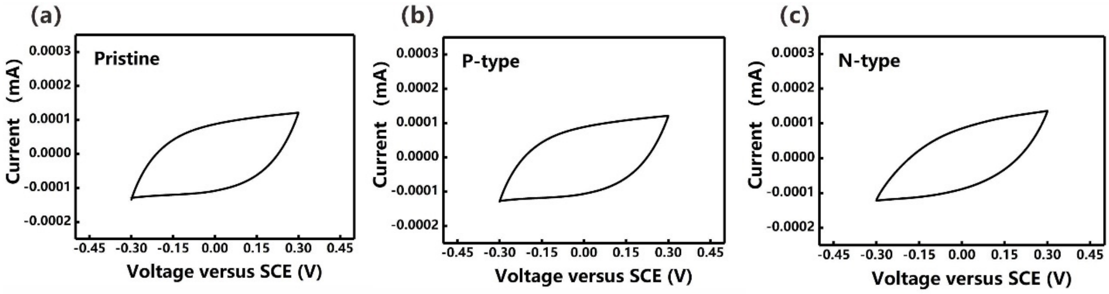

Figure S12. Cyclic voltammetry of the (a) Pristine, (b) P-type and (c) N-type graphite. The Cyclic voltammetry of graphite was measured in a Tap water with a Pt wire as the counter electrode, a saturated calomel reference electrode (filled with saturated KCl with a potential of 0.241 V versus SHE) and graphene as the working electrode. The CVs were taken at a scan rate of 10 mV s-1 for each electrode.

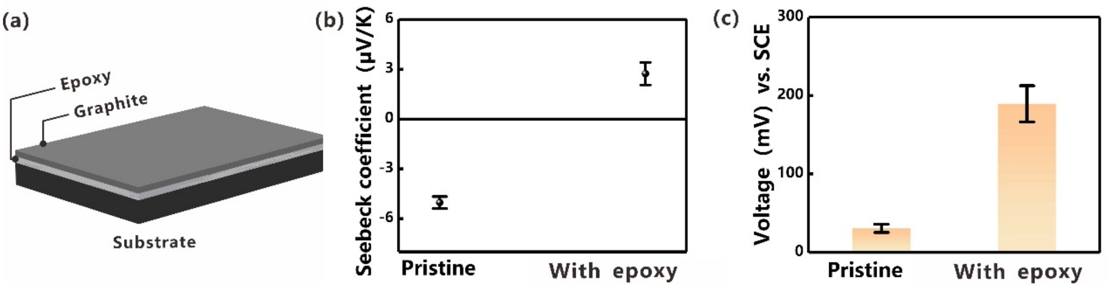

Figure S13. (a) Schematic drawing showing the graphite with epoxy (b) The average room temperature (290 K) Seebeck coefficient of the pristine graphite and graphite with epoxy. (c) The open-circuit voltage between graphite foils (pristine and with epoxy) and a saturated calomel electrode reference electrode at room temperature (290 K).

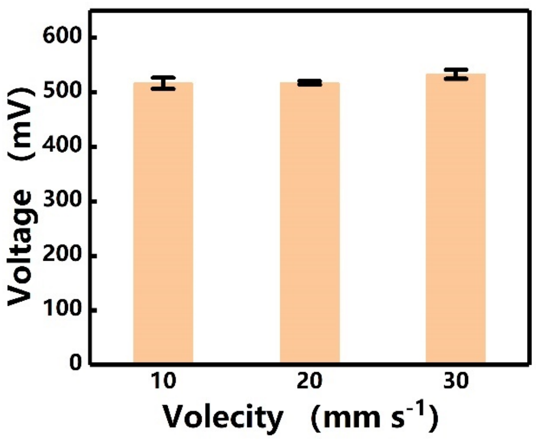

Figure S14. Open-circuit voltage from the WG-HEG devices with different immersing speed.

Table S1. The performance comparison of reported hydroelectric generators

Hydroelectricity	Materials (Electrode)	Series number	Parallel number	Voltage (V)	Current (mA)	Refs
Moisture-induced						
	Protein wires (Au)	17	5	10	0.0011	1[1]
	Polyelectrolyte (carbon tape)	1600	500	1000	0.016	2[2]
	Ionic liquid (Ag/AgCl)	4	4	1	0.05	3[3]
	Nafion and poly (n-isopropylacrylamide) (carbon paper)	2	2	4	0.04	4[4]
	Carbon-coated cotton fabric (Fe)	4	4	1.2	0.08	5[5]
	Kraft paper And reassembled graphite (--)	30	1	12	0.000025	6[6]
	Poly(4-styrensulfonic acid) (Au)	3	1.5cm^2	2.3	0.2	7[7]
	Graphene oxide and sodium polyacrylate (Au\Ag)	68	-	26.2	-	8[8]
	Graphene oxide (Au)	60	-	11.2	-	9[9]
	Graphene oxide (silver paste)	5	-	2	-	10[10]
	Graphene oxide (Al)	5	5	1.32	10.5	11[11]
	Ionic hydrogel and functionalized carbon (-)	5	5	3	0.03	12[12]
	Poly(vinyl alcohol), phytic acid and glycerol-water binary solvent (Ag/Cu)	260	300	210	7	13[13]
	Graphene oxide (Au)	8	8	1.21	0.00397	14[14]
	Titanium dioxide (indium tin oxide/Al)	3	3	1.3	0.025	15[15]
	Biological nanofibrils (Pt)	4	4	0.4	0.0001	16[16]
	Protein nanofibrils (ITO/Ti)	5	5	2.44	0.00961	17[17]
	Polyvinyl alcohol and Poly (4-styrensulfonic acid)	10	10	9.2	0.0102	18[18]

	(carbon tape)					
	Carbon black (Cu)	60	-	33	-	19[19]
	Fluorinated oxidized graphene (-)	15	4	10	0.005	20[20]
	Geobacter sulfurreducens (ITO)	4	4	1.07	0.001	21[21]
	Cellulose nanofiber (Cu)	4	-	3	-	22[22]
	Molybdenum disulfide (stainless-steel)	12	1	1.7	0.00233	23[23]
	Biological nanofibrils (Pt)	3	3	250	0.00012	24[24]
	High-valent metal cation (Au)	4	4	0.35	0.00175	25[25]
	poly(4-styrensulfonic acid) and poly(vinyl alcohol) (Ag nanowires)	2	2	1.2	0.004	26[26]
	HCl/polyvinyl alcohol (carbon nanotube)	6	-	1.5	-	27[27]
	Graphene oxide (Ag wire)	10	10	3	0.042	28[28]
	MXene/cellulose/ polystyrene sulfonic acid (Ag nanowires)	4	4	1.1	0.0058	29[29]
	Cationic silk nanofibrils (Pt)	3	3	0.36	0.00035	30[30]
	LiCl-loaded cellulon paper and carbon-black-loaded cellulon paper (carbon tape)	3	2	2	0.01	31[31]
	Nanochannel anodic aluminum oxide (nanoporous carbon nanotubes/In-Ga)	20	1	18	0.0012	32[32]
	Sodium alginate , silicon dioxide nanofiber , and reduced graphene oxide (Au)	21	16	11	1.3	33[33]
	Poly([2-(methacryloyloxy)ethyl] Dimethyl-(3-sulfopropyl) ammonium hydroxide-co-acrylicacid) (Au)	5	-	2	-	34[34]
	Graphene oxide (Ag/AgCl ink)	6	1	1.9	0.0334	35[35]
	Graphene oxide (Au/Ag)	15	1	18	0.00012	36[36]

	Electroactive bacteria (ITO)	30	30	10	0.052	37[37]
	TiO2 nanowires/cotton fibers and an asymmetrically distributed polypyrrole (Fe)	3	3	1.45	0.018	38[38]
	Poly (4-styrensulfonic acid) (Al)	5	5	5.5	0.0047	39[39]
	Polyvinyl alcohol (Al /Au-coated stainless-steel)	5	5	3.2	0.00014	40[40]
	Print paper (Stainless steel metal plate)	5	5	1	0.00012	41[41]
	Graphene oxide/ reduced graphene oxide (Ag)	175	1	192	0.17	42[42]
	Polyacrylonitrile and sodium dodecyl benzene sulfonate (ITO/Zn/Al)	5	5	3.6	0.012	43[43]
	TiO_2/ZrO_2 composite nanofiber membrane (ITO/ Au-coated Al)	4	4	2.98	0.00001582	1[44]
	3-Acrylamidopropyl[trimethylammo-nium] chloride and poly (sodium4-styrene sulfonate) (Ag)	5	5	0.045	0.0000008	1[45]
	Graphene oxide (carbon)	5	5	11.9	0.0827	1[46]
Liquid-induced						
	cellulose/carbon nanotubes (Cu)	1	6	0.16	0.0007	18[47]
	CuO nanowires (silver paste)	4	4	0.75	0.0012	45[48]
	Single-layer MoS_2 (silver paste)	3	3[49]	14	0.000015	1[49]
	Silicon nanowires (graphite)	3	4	3.7	0.0012	1[50]
	Graphene and polytetrafluoroethylene (-)	3	3	1.1	0.008	1[51]
	Carbon nanotube/transition metal oxide (-)	8	1	1.8	0.04	1[52]
(Tribovoltaic effect)	Water (Pt/ indium gallium zinc oxide/Al)	6	3	2.5	0.014	2[53]
(Tribovoltaic effect)	Water (silicon/Al)	3	1	2.7	0.0007	1[54]
Evaporation-induced						
	carbon black	4	4	1.25	0.0001	1[55]

	(multiwalled carbon nanotube)					
	Al_2O_3 and carbon black (conductive carbon paste)	6	6	16	0.00845	47[56]
	MoS_2/SiO_2 (silver paste)	6	6	4.2	0.0024	48[57]
	Carbon black/PVA (Fe)	100	101.07	10	2.2	49[58]
	poly(styrene sulfonicacid) and polyacrylicacid cellulose (conductive carbon ink)	2	1	0.8	0.00045	50[59]
	Functionalized conductive carbon black (Cu)	6	6	3.7	0.34	51[60]
	Nanostructured silicon nanowire arrays (graphite/PEDOT:PSS/ cellulose fabric/)	4	-	2.4	-	52[61]
	Silicon nanowire arrays (graphite/Ag)	4	-	1.1	-	53[62]
	Carbon nanospheres and TiO_2 nanowires (carbon ink)	4	4	6.2	0.00055	54[63]
	Reduced graphene oxide (super-aligned carbon nanotube buckypapers)	6	1	2.34	0.1	55[64]
	A piece of wood (carbon paste)	4	4	1.2	0.04	56[65]
	Al_2O_3 (carbon paste)	6	6	14.8	0.0022	57[66]
	PEDOT:PSS (gold-coated electrode)	4	4	3.1	0.1834	58[67]
	SiO_2 nanofiber (Al)	5	5	2.1	0.0017	1[68]

Only one of the works with similar performance is selected to be displayed in the main text, and the work displayed in the main text is in brackets.

6/49(49)
41/55(55)
14/26/29/51(51)
31/17(17)
50/57(57)
12/28(28)
37/46(46)

Additionally, no work utilizing active electrode materials was presented in the main text. In the form, active electrode materials are denoted in red font, while acidic materials are indicated in blue font. Both types of materials are marked with asterisks. It is essential to emphasize that meticulous consideration should be given to eliminating any potential contributions resulting from the interaction between the liquid and metal electrodes. If the electrodes or functional channels comprise active materials, they may come into contact with water, leading to the formation of a metal-air battery under ambient conditions. This process would cause electrochemical energy to be irreversibly converted into electricity. Thus, contributions resulting from the metal-air battery should be avoided, as they may lead to inaccurate or misleading results[69].

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