Experimental Section

Chemical reagent

PVA (17-99) was purchased from Linyi Yiebiao Chemical Industry, Ketjen Black (ECP-600JD), stainless steel mesh, zinc flake (100 μm) was purchased from Guangdong Canrd New Energy Technology Co. , Ltd.Zinc sulfate heptahydrate (analytically pure) was purchased from Sinopharm Chemical Reagents Co. Diaphragms (GF/D) were purchased from Chongqing olegeeino Technology Co.

Electrodes and electrolytes

Fabrication of Kb-P60 electrode

0.6 g of PVA was placed in 400 ml of water and stirred at 90° C for 4 h. 0.4 g of KB was added and stirred at 90° C until evaporation. The sample was processed in an oven at 80° for 12 h. The resulting product was KB-P60, which was mixed with KB (conductive agent) and PTFE in a mass ratio of 8:1:1, and then pressed onto a stainless-steel mesh.

Pure KB Partial Electrode Fabrication

The KB electrodes were prepared by mixing KB and PTFE binder at a mass ratio of 9:1 and then pressing the mixture onto a stainless-steel mesh. The mass loadings were all controlled to be around 20 mg cm^{-2}. Two electrolytes were used, which can be classified as cathode electrolyte and anode electrolyte. The cathode electrolyte was prepared by dissolving 0.2 M ZnI_2 and 3 M $ZnSO_4$ in H_2O, and the anode electrolyte was an aqueous solution containing 3 M $ZnSO_4$.

Electrochemical measurements

The electrochemical performance of Zn-I_2 battery was tested using CR-2023 button cells assembled at room temperature. In Zn-I_2 battery, KB-P60 electrodes or KB electrodes were matched with Zn foils in which glass fibers were used as diaphragms. In the assembly process, 30 μL of cathode electrolyte was first dropped into the KB-P60 electrode or KB electrode. Then an appropriate volume of anode electrolyte was dripped in to wet the glass fibers. Constant current discharge/charge measurements were performed at room temperature using a battery test system (CT-4008T, Neware, Shenzhen, China) in the potential range of 0.6-1.6 V. The battery was then charged with the electrolyte at a potential of 0.6-1.6 V. The electrode was then charged with the electrolyte at a potential of 0.6-1.6 V. Redox pairs of I^-/I_3^- were investigated using cyclic voltammetry (CV) technique in the voltage range of 0.6-1.6 V at a scan rate of 0.2 mVs^{-1}. All current densities were calculated from the mass of iodine in the cathode solution.

Materials characterization

Powder X-ray diffraction (XRD) analyses were carried out on a Rigaku smart lab se using a Cu-Kα radiation source. The samples were scanned over a range of 5 to 80° with a scanning speed of 10° min^{-1}. The morphology and elemental distribution were observed with a field emission scanning electron microscope HITACHI SU 8010) equipped with energy dispersive X-ray spectroscopy (EDS). X-ray photoelectron spectroscopy (XPS) was collected by an Escalab 250Xi instrument (Thermo Fisher Scientific, USA) and Al Kα monochromatic radiation was used as the X-ray source. Sputtering was carried out using an embedded argon ion gun, and the sputtering rate was standardized by SiO_2 (25 nm min^{-1}) with a sputtering area of 1 mm × 1 mm. Raman spectra were performed using a LabRAM HR Evolution confocal Raman microscope (Horiba Jobin Yvon) with a 532 nm laser. electrochemical measurements were performed on an electrochemical workstation (CHI760E, Chenhua, Shanghai) for constant-current charge/discharge testing of Zn-I_2 battery.

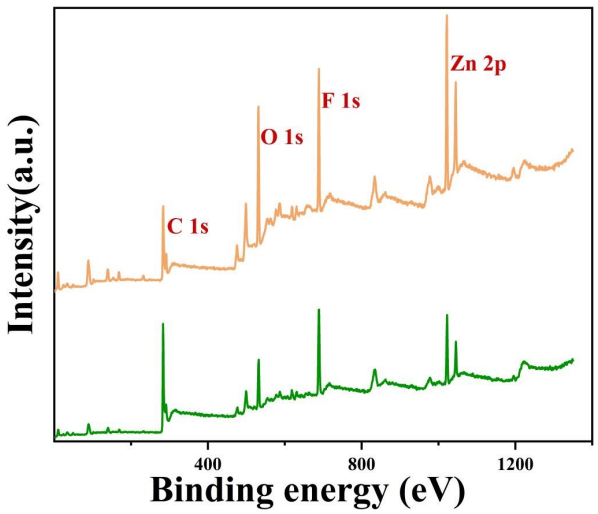


Fig. S1 Full spectrum of XPS for positive tabs.

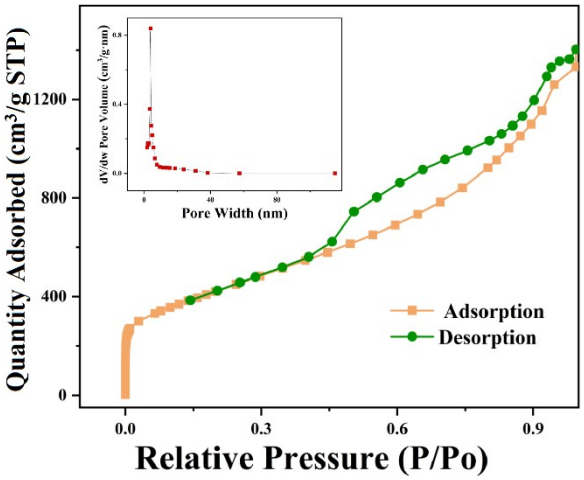
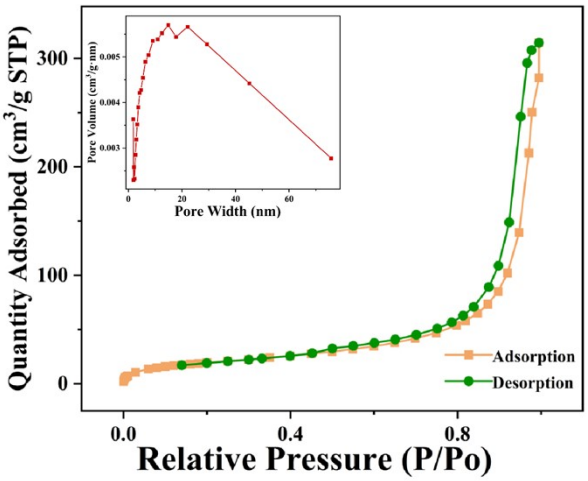


Fig. S2 N_2 adsorption/desorption isotherm curve and pore-size distribution of KB.

Fig. S3 N_2 adsorption/desorption isotherm curve and pore-size distribution of KB-P60.

Scheme S1 Elemental C and I content on charged and discharged pole pieces as shown by EDS test

Elements	Line	Intensity(c/s)	Conc	Units	Error 2-sig	MDL 3-sig



C (charge)	Ka	26.01	17.545	wt.%	1.017	.842
I (charge)	La	13.35	6.121	wt.%	.712	.884
C (discharge)	Ka	73.81	47.901	wt.%	1.916	.858
I (discharge)	La	15.14	17.093	wt.%	2.108	2.351

Fig. S4 Anode photos of KB battery (Left) and KB-P60 battery (Right) after charge and discharge cycle at 20 A g^{-1} current density.

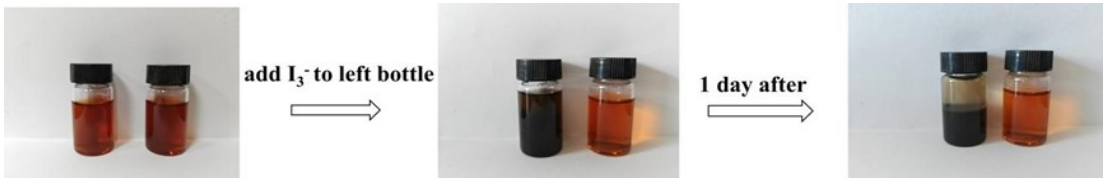


Fig. S5 Before and after the polyiodide solution is captured by PVA