Electronic Supplementary Information (ESI)

Binder free hierarchical mixed capacitive deionization electrode based on polyoxometalate and polypyrrole for brackish water desalination

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Materials and characterization

All chemicals were of analytical grade, commercially available from Sinopharm Chemical Reagent Co. Ltd (Shanghai, China) and used as received without further purification. Raman spectroscopy was conducted with an excitation wavelength of 633 nm (LabRAMHR-800, HORIBA). The morphology was observed on an ultra plus field emission scanning electron microscope (SEM, ultra plus, ZEISS). Infrared spectra were recorded as KBr pellets on a Thermo Scientific Nicolet 6700 Fourier Transform Infrared (FTIR) spectrometer in the 400-4000 cm^{-1} range. XPS was performed with Mg Kα radiation (1253.6 eV) as an excitation source (ESCALab MKII, Thermo Scientific, Waltham, MA). Electrochemical experiments were conducted on CHI-660E electrochemical workstation.

Electrochemical property study

PPy/EGM or **P_2Mo_{18}@PPy/EGM** can be employed as working electrode directly. In charge-discharge property study, three-electrode system was employed. Carbon rod and Ag/AgCl electrodes served as counter and reference electrodes. 1 M NaCl was chosen as electrolyte. The charge-discharge property was conducted in scope of -0.6 to 0.6 V (vs. Ag/AgCl). For CV tests, the voltage window is in -0.2 to 0.6 V (vs. Ag/AgCl) with scanning rate at 10 $mV \cdot s^{-1}$. The specific capacitance ($F \cdot g^{-1}$) was calculated based on galvanostatic charge-discharge test according to Eq(1). In this equation, I and t are the charge-discharge current and time. E and m represent the charge-discharge potential windowand mass loading of the active material. The HER, OER and overall water splitting of **P_2Mo_{18}@PPy/EGM(B)** were studied in NaCl (600 $mg \cdot L^{-1}$).

$$C = I \cdot t / E \cdot m \quad (1)$$

CDI property study

CDI tests were conducted on a home-made device equipped with a continuous recycling system. The effective area of electrode is 2 × 2 cm^2. In experiment, the concentration of the NaCl solution was continuously recorded and measured by the ion conductivity meter (DDS-307, Leici, Shanghai, China). The volume was set at 50 mL, the flow rate was fixed at 20 $mL \cdot min^{-1}$ and the operating voltage is maintained at 1.2 V. During experiment, the temperature was maintained at 25 ˚C. Salt adsorption capacity (SAC, $mg \cdot g^{-1}$) and salt adsorption capacity (SAR, $mg \cdot g^{-1} \cdot min^{-1}$) were calculated according to Eq(2) and Eq(3). In these equations, V is the volume of NaCl solution and m is the total mass of the electrode material, c_0 and c are the initial and final concentrations of NaCl solution, t represents the time of adsorption experiment

$$SAC = (c_0 - c) \times V/m \quad (2); \quad SAR = SAC/t \quad (3)$$

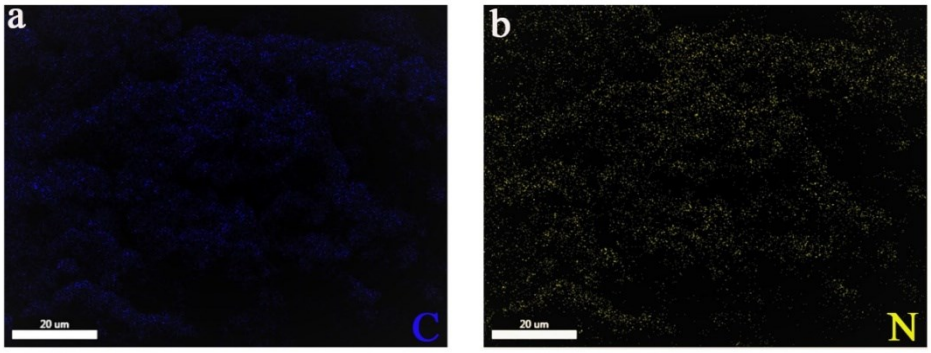

Fig. S1 EDX spectrum of **PPy/EGM**: (a) C, (b) N

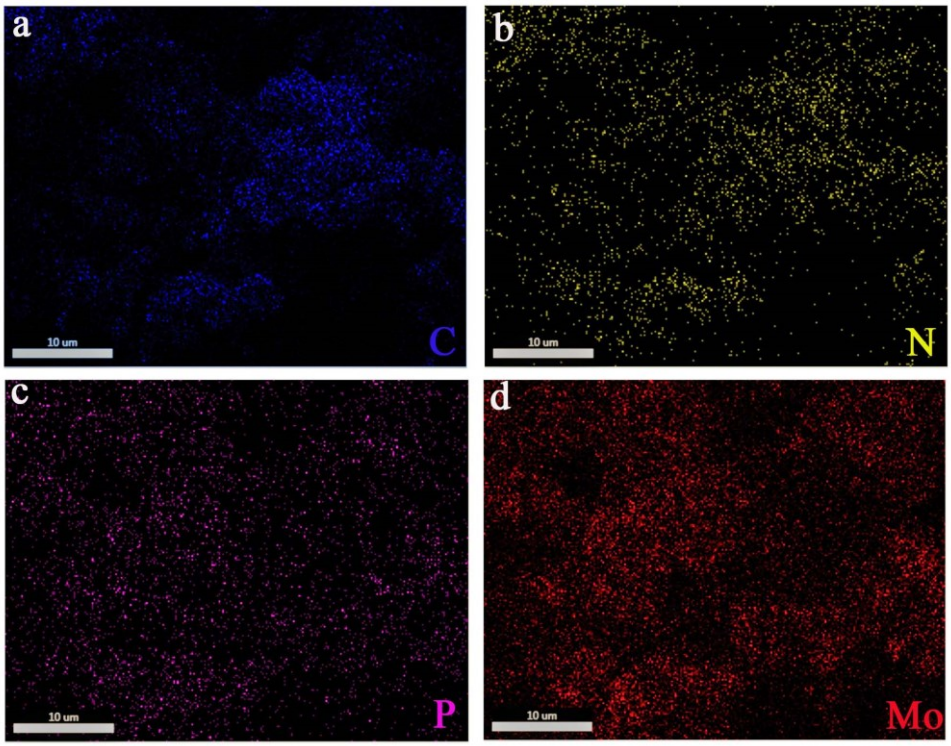

Fig. S2 EDX spectrum of **P_2Mo_{18}@PPy/EGM(B)**: (a) C, (b) N, (c) P, (d) Mo

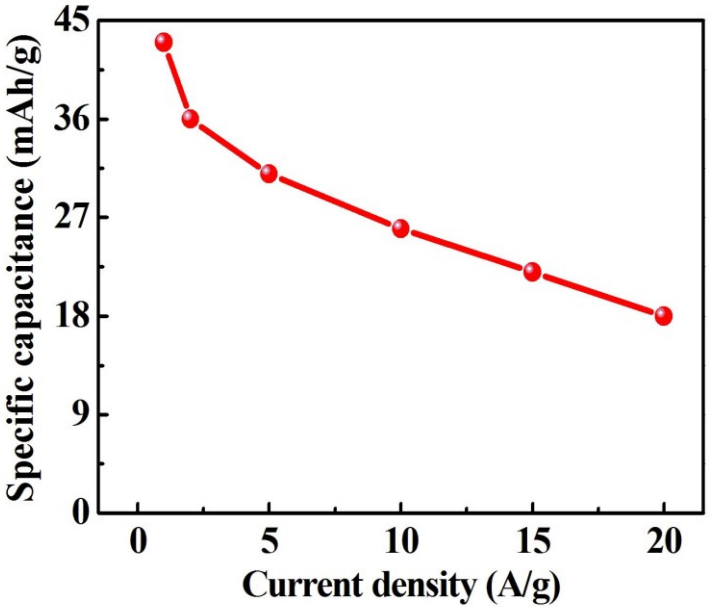

Fig. S3 the specific capacitance of **EGM**

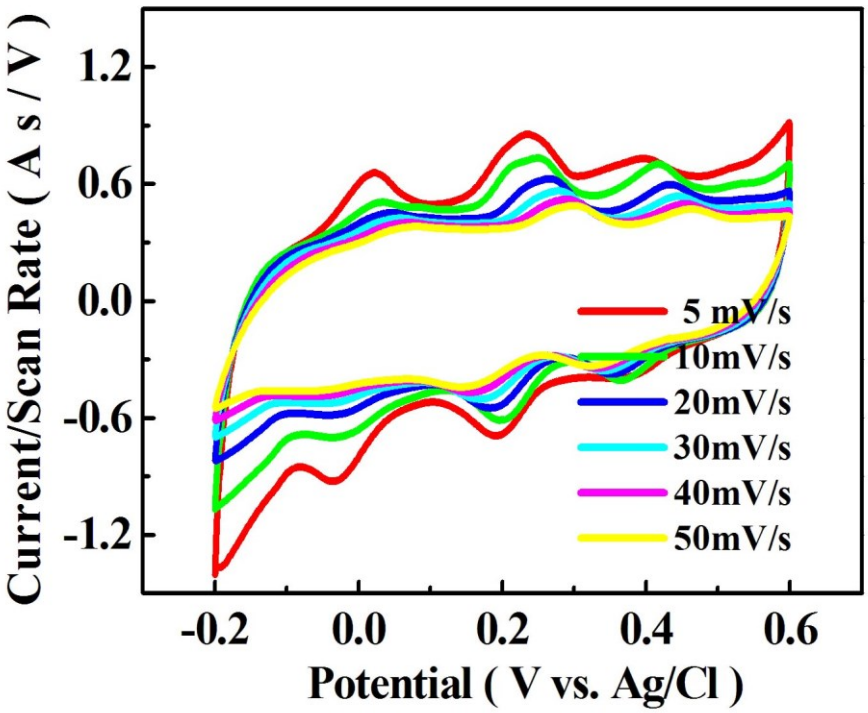

Fig. S4 the connection between current density/scan rate and potential

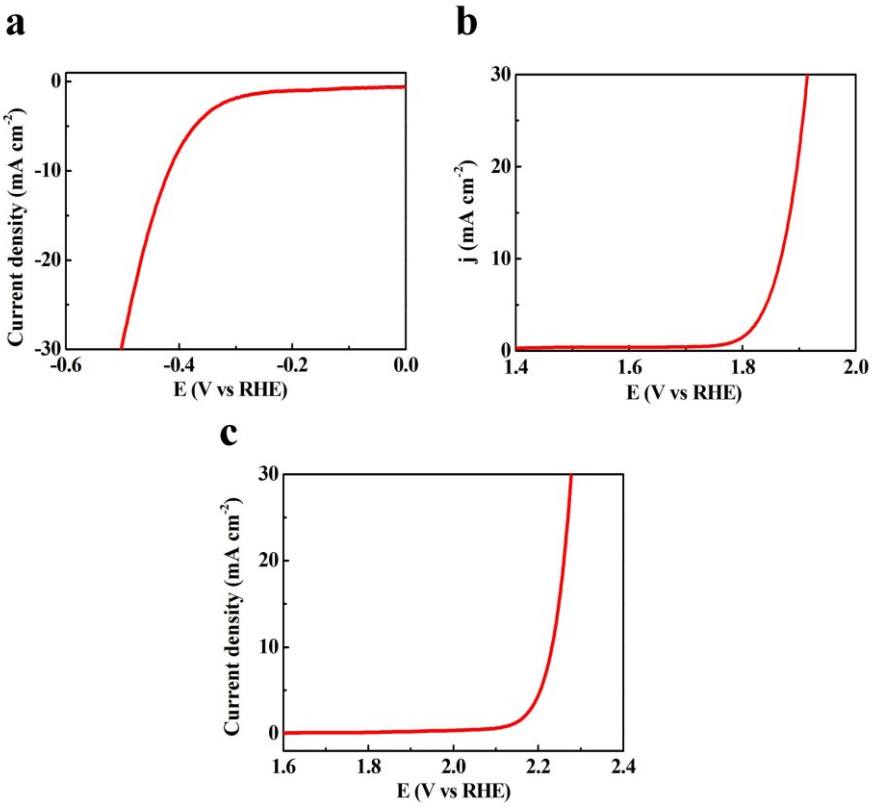

Fig. S5 (a) LSV of **P_2Mo_{18}@PPy/EGM(B)** in HER; (b) LSV of **P_2Mo_{18}@PPy/EGM(B)** in HER; (c) LSV of **P_2Mo_{18}@PPy/EGM(B)** in overall water splitting