Electronic Supplementary Material (ESI) for Nanoscale.

Dual-functional Mediators of High-Entropy Prussian Blue Analogues for Lithiophilicity and Sulfihilicity in Li–S Batteries

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1. Raw materials and instruments

All raw materials, including manganese nitrate tetrahydrate ($Mn(NO_3)_2·4H_2O$), cobalt nitrate hexahydrate ($Co(NO_3)_2·6H_2O$), nickel nitrate hexahydrate ($Ni(NO_3)_2·6H_2O$), copper nitrate trihydrate ($Cu(NO_3)_2·3H_2O$), zinc nitrate hexahydrate ($Zn(NO_3)_2·6H_2O$), trisodium citrate dihydrate ($Na_3C_6H_5O_7·2H_2O$), and potassium hexacyanoferrate(III) ($K_3[Fe(CN)_6]$), high-purity de-ionized water, sublimed sulfur (99.98%, Sigma-Aldrich), poly(vinylidene fluoride) (PVDF), and N-methylpyrrolidone (NMP), lithium (Li) metal foils (200 μm in thickness, 14.0 mm in diameter), aluminum (Al) foil and Celgard 2400 polypropylene (PP) membranes, and lithium bis (trifluromethanesulfonyl) imide (1.0 M LiTFSI in DME : DOL =1 : 1 vol.% with 1.0 wt.% $LiNO_3$), were purchased from commercial sources. All reagents used in this experiment were of analytical grade and were used without further purification.

The phase and purity of the material were characterized by X-ray diffraction (XRD) on Smart Lab/3kW X-ray Diffractometer. The morphology of samples was observed by scanning electron microscope (SEM, Zeiss Gemini 300) under the acceleration voltage of 5.0 kV. After disassembling the coin cell and cleaning it with 1,2-dimethoxyethane (DME) in the glovebox. Transmission electron microscopy (TEM) investigations were performed by a JEM-2100 instrument. The chemical states were measured using the X-ray photoelectron spectroscope (XPS) on an ESCALAB250Xi (Thermo Scientific, U.K.) associated with a standard mono-chromatied Al Kα source (energy: 1486.68 eV).

2. Supplementary Figures

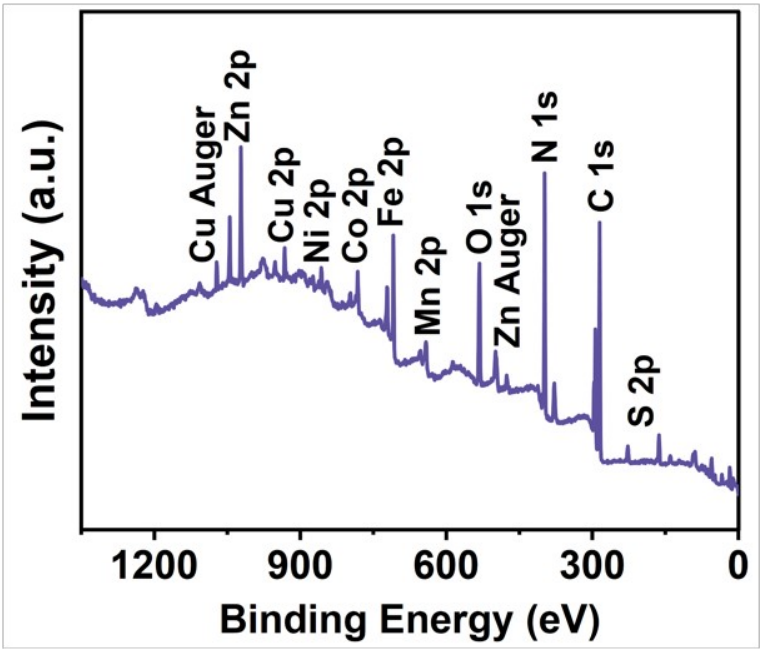

Fig. S1 XPS survey spectrum for HE-PBA.

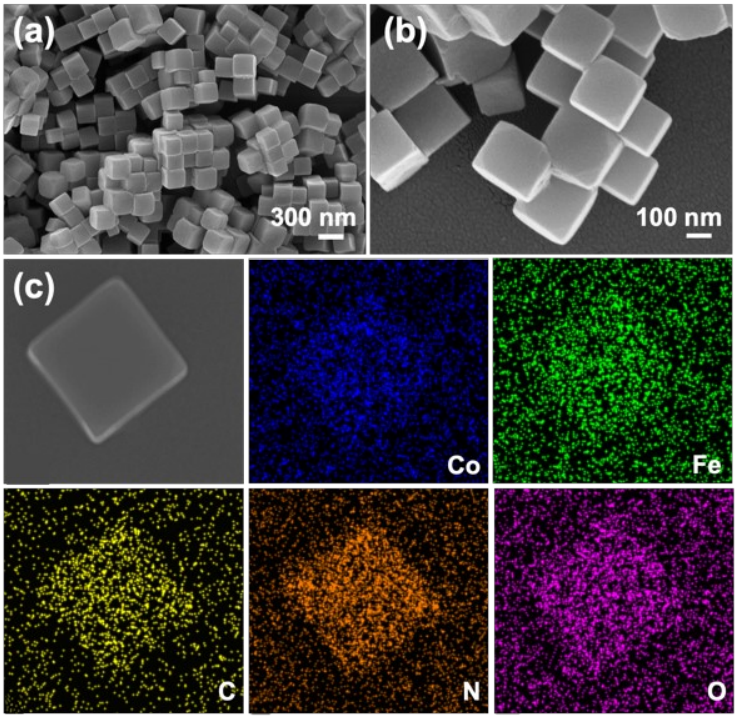

Fig. S2 (a-b) SEM images of CoFe-PBA. (c) The corresponding EDS mapping for elements of Co, Fe, C, N, and O on CoFe-PBA.

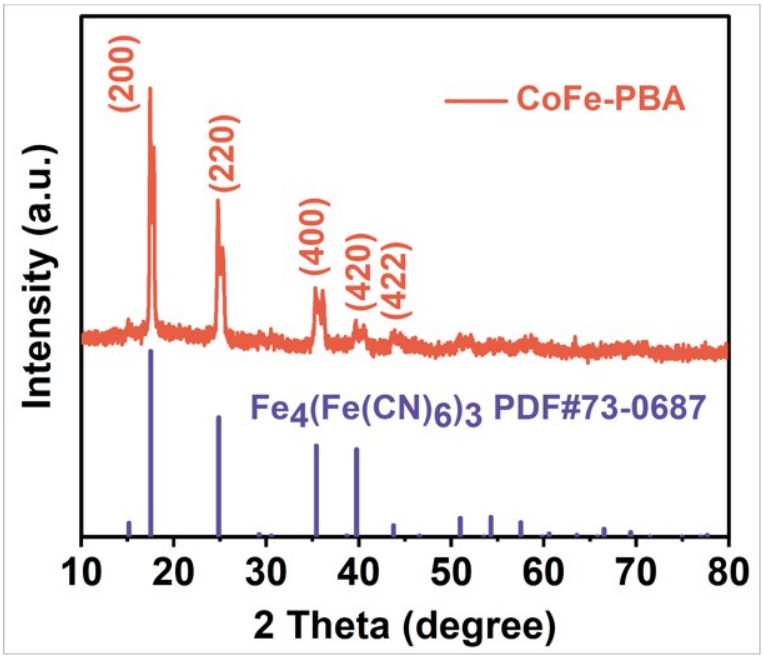

Figure S3. XRD patterns of CoFe-PBA.

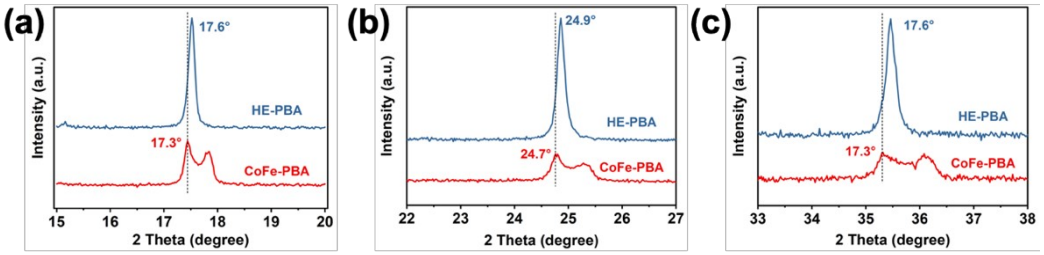

Fig. S4 The enlarged view of XRD patterns of HE-PBA and CoFe-PBA.

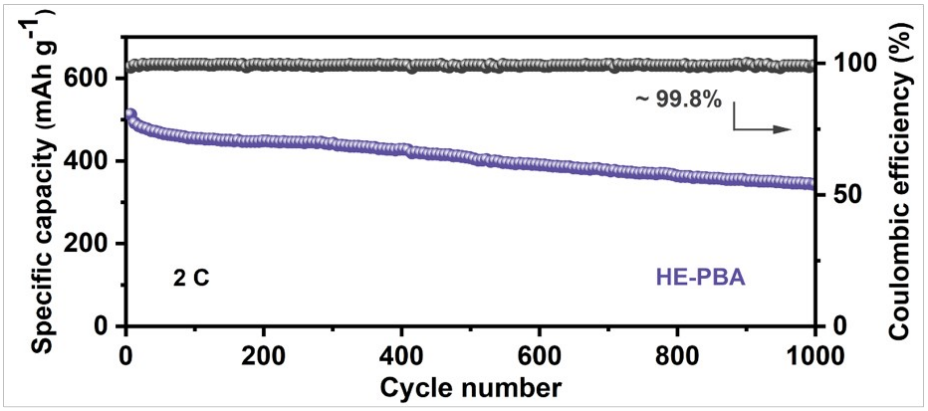

Fig. S5 Cycle performance of HE-PBA/S at 2 C over 1000 cycles.

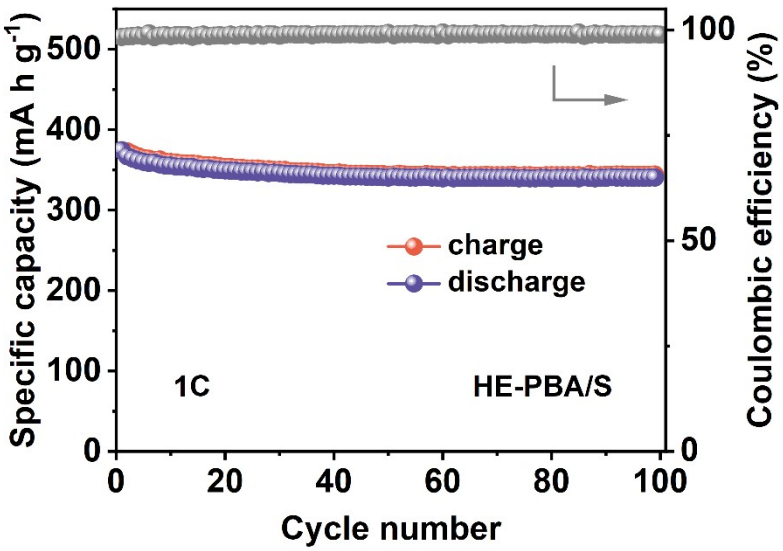

Fig. S6 Cycle performance at 1.0 C over 100 cycles with sulfur loading of 2.1 mg cm^{-2}.

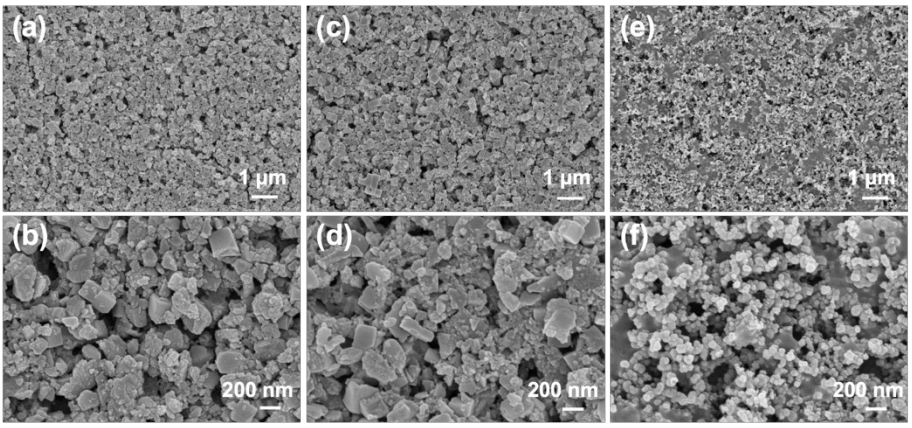

Fig. S7 SEM images of (a-b) HE-PBA@Cu, (c-d) CoFe-PBA@Cu and (e-f) bare Cu surface.

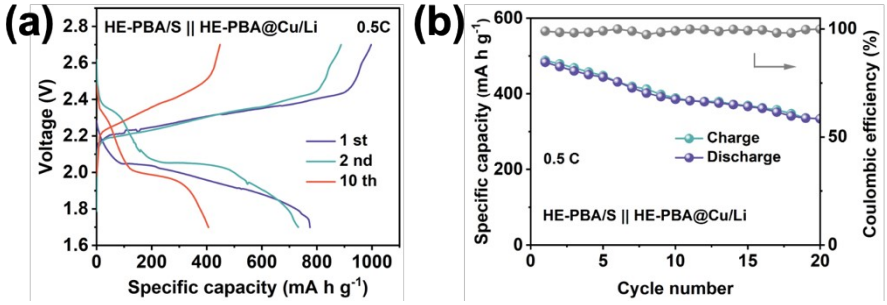

Fig. S8 (a) The galvanostatic charge/discharge profiles at 0.5 C for HE-PBA/S || HE-PBA@Cu/Li. (b) Cycle performance at 0.5 C over 20 cycles for HE-PBA/S || HE-PBA@Cu/Li.