Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Supporting Information for

An aqueous dissolved polysulfide cathode for lithium-sulfur batteries

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Experimental Section

Preparation of aqueous lithium-polysulfide batteries. The preparation and assembly of the anode side were conducted in an Ar-filled glove box (<1 p.p.m. of H_2O and O_2). The lithium foil with a thickness of 0.2 mm was cut to a rectangle of 8×40 mm, and then pressed onto a Cu mesh (Nilaco Cor., 100 mesh) current collector. An organic electrolyte of 1 M $LiClO_4$ in ethylene carbonate (EC)/dimethyl carbonate (DMC) with a volume ratio 1:1 was added into this glass cylinder. LATP glass ceramic separator was supplied by Ohara Inc., Japan. The thickness and the electrical conductivity of the LATP plate at 25℃ was ca. 150 μm and 1.05×10^{-4} S cm^{-1}. The LATP was fixed on top of the glass cylinder thus sealing in the anode. The film of CoS deposited on brass mesh was used as a current collector of aqueous cathode. First, by using reactive RF magnetron sputtering equipment a Co metal film (150 nm in thickness) was sputtered onto two sides of a brass mesh (100 mesh). The resultant Co-sputtered brass was immersed in 0.005 M polysulfide solution and scanned anodically from 0.1-3 V vs. a Pt electrode, then a black, strongly adhered CoS layer was evenly formed. The current collector was stuck on the other cylindrical shape of the glass shell, in which the aqueous cathode containing Li_2S_4 and LiOH was added with a total volume of 1 ml, which was then sealed with the backside of the LATP employed in the anode assembly. The Li_2S_4 solution was prepared as follows: first, 1 M Li_2S (Aldrich) was dissolved in aqueous solution containing 1 M LiOH (Wako Chemicals), then 3 M sulfur (Wako Chemicals) were slowly added into the solution. The different concentration catholytes were prepared using above obtained 1 M Li_2S_4 solution. The resulting mixture was stirred and bubbled with purified nitrogen. The completed battery had a configuration of (-) Cu mesh/Li metal/organic electrolyte/LATP/aqueous cathode/CoS/brass mesh (+).

Electrochemical measurements. The electrochemical reaction was investigated using CV and galvanostatic methods at room temperature in 1 atm using a Hokuto charging/discharging machine and Solartron instruments.

Characterizations. The current collector of aqueous cathode before and after the first cycle (discharge and charge) were detected by XPS and SEM measurements. The XPS measurements were performed using a VG theta probe. SEM observation was performed on a Hitachi S4800.

Supplementary Figures

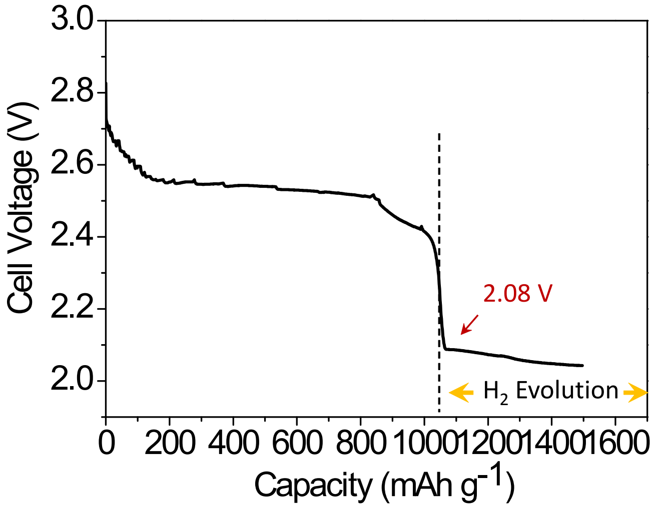

Fig. S1 The discharge profile of an aqueous lithium-polysulfide battery with a 0.01M solution of Li_2S_4 as catholyte at 0.2 mA cm^{-2}. The H_2 evolution potential was adjusted to be 2.08 V.

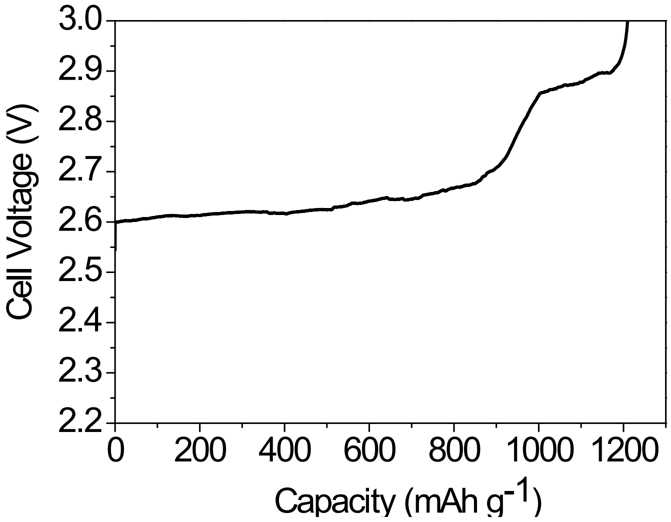

Fig. S2 The recharge profile of an aqueous lithium-polysulfide battery with a 0.1 M solution of Li_2S_4 as catholyte at 0.2 mA cm^{-2}. The lower plateau involves oxidation reaction between soluble Li_2S and soluble Li_2S_4; the upper plateau involves oxidation reaction between soluble Li_2S_4 and insoluble sulfur.

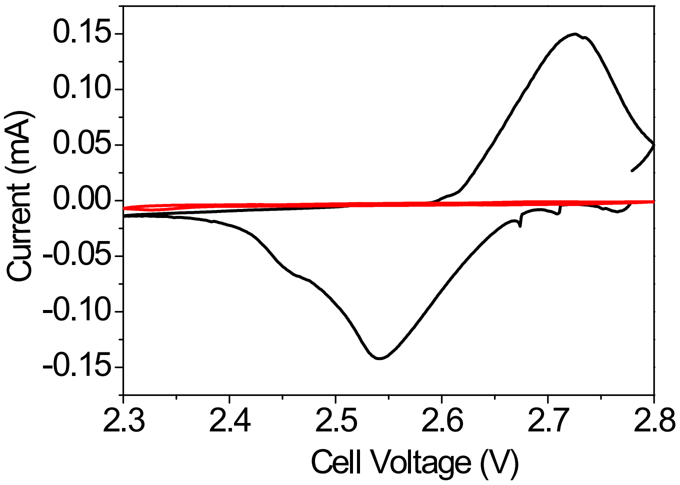

Fig. S3 CV of an aqueous lithium-polysulfide battery with a 0.01 M solution of Li_2S_4 (black) and 0.2 M LiOH aqueous solution without Li_2S_4 (red) as catholytes between 2.30 and 2.80 V versus Li^+/Li at a rate of 0.005 mV S^{-1}.

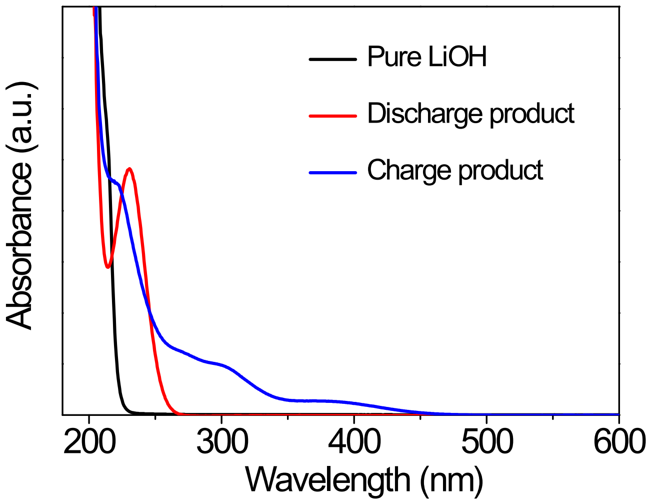

Fig. S4 UV-visible absorption spectra of (red line) discharge and (blue line) charge products. Solutions have the same concentration, 0.5 mM.

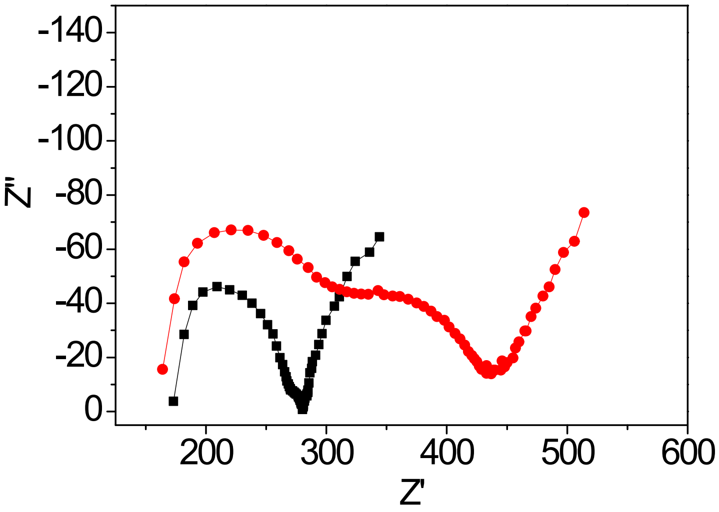

Fig. S5 Impedance spectra of the battery with a (black line) initial LATP plate and (red line) LATP plate after using 50 days.

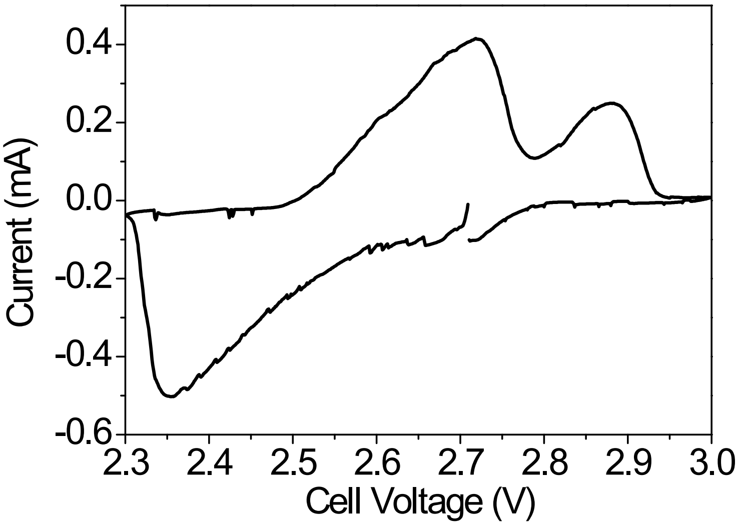

Fig. S6 CV of an aqueous lithium-sulfur battery with 3.2 mg solid sulfur and 14.2 mg of 0.1 M Li_2S_4 as cathode between 2.30 and 3.00 V versus Li^+/Li at a rate of 0.001 mV S^{-1}.