## **Supporting Information**

## First Implementation of Alkaline Polymer Electrolyte Water Electrolysis Working Only with Pure Water

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## **Supplementary figures**

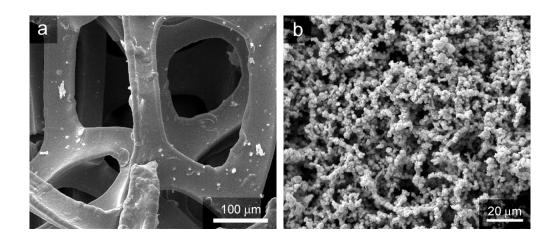


Figure S1 SEM images for the Ni foam skeleton (a), which is filled with Ni powder to be used as the anode electrode substrate (b).

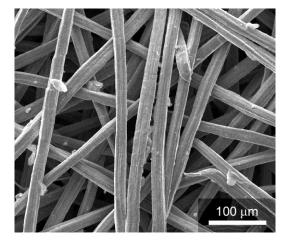


Figure S2 SEM image for the stainless steel fiber felt which is used as the cathode electrode skeleton.

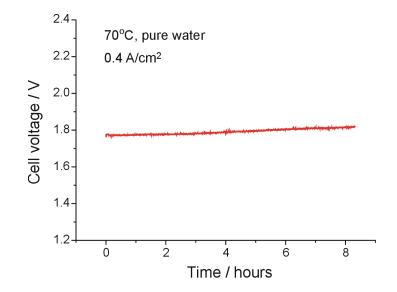


Figure S3 A stability test of the APE water electrolyzer running at 0.4 A/cm<sup>2</sup>, 70°C.

## Methods

**Fabrication of Ni-Fe anode.** An ethanol solution containing 0.5 M Ni(NO<sub>3</sub>)<sub>2</sub> and 0.5 M  $Fe(NO_3)_3$  was sprayed onto the porous Ni electrode substrate (a Ni-foam skeleton filled with Ni powder, Figure S1) on a hot plate, whose temperature was set to 60°C. The thus-prepared Ni-Fe precursor electrode was electrochemically reduced at room temperature with a current density of 40mA/cm<sup>2</sup> in 1M KOH solution. Finally, the Ni-Fe electrode was washed with deionized water repeatedly and dried for later use.

**Fabrication of Ni-Mo cathode.** 1 M Ni(NO<sub>3</sub>)<sub>2</sub> ethanol solution was mixed with 1 M  $(NH_4)_2MoO_4$  ammonia solution to produce a light green precipitate. The atomic ratio of Ni/Mo was controlled to be 1:2. The thus-obtained precipitate was washed and filtered, and dispersed in ethanol, and then sprayed onto the stainless steel fiber felt (SSFF) skeleton (Figure S2). The Ni-Mo precursor electrode was reduced under H<sub>2</sub> atmosphere at 500°C for 30 min.

Electrochemical test. The catalytic activity of the above-obtained Ni-Fe anode and Ni-Mo cathode were tested in 1M KOH solution in a thermostatic cell. The counter electrode was a Ni foam, and the reference electrode was Hg/HgO. All solutions were prepared using deionized water (18 M $\Omega$ ·cm). The potentiostat was a CHI-660 electrochemical station.

Fabrication of membrane-electrode assembly (MEA). The above-prepared Ni-Fe anode and Ni-Mo cathode were impregnated with certain amount of self-crosslinking quaternary ammonia polysulfone (xQAPS) solution, and dried at 60°C. An xQAPS membrane was sandwiched with the ionomer-impregnated Ni-Fe anode and Ni-Mo cathode under a pressure of 2 MPa at 80°C for 2 min.

**Water electrolysis test.** The prototype of APE water electrolysis was run under constant current mode. Deionized water was circularly pumped into the cell, and the cell voltage was recorded after 30 min electrolysis under each current density.