Electronic Supplementary Information

Efficient Improvement of Lithium Ionic Conductivity for Polymer Electrolyte via Introducing porous Metal–Organic Frameworks

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Contents

Experimental section

Materials and Characterizations

Scanning electron microscopy (SEM) images were acquired by a HITACHI S4800 scanning electron microscope. X-ray diffraction (XRD) spectra were obtained through an Empyrean Panal Ytical instrument with Cu-Kα radiation under the range of 5–50º (λ = 1.5406 Å). Thermo gravimetric analysis (TGA) was performed by a Q500 thermal analyzer on heated from 23 to 800°C under N_2 atmosphere at a heating rate of 10 °C/min. Mechanical properties were determined by a CMT2533 universal tensile machine with electronic data evaluation at speed of 5 mm/min. The contact angles were tested by a Biolin Theta Flex optical instrument. The electrochemical impedance spectroscopy (EIS), cyclic voltammetry (CV), liner sweep voltammetry (LSV) and chronoamperometry (CA) curves were performed by a CHI660E electrochemical workstation. The cycle performances were recorded with a LANHE LAND battery cycler.

Synthesis of compound MOF-5

MOF-5 crystals were synthesized according to the reported in the literature.[1] $Zn(NO_3)_2 \cdot 6H_2O$ (900 mg), H_2BDC (166 mg), DMF (98 mL) and deionized water (2 mL) were loaded into 250 mL round bottom flask, vigorously stirred for 30 minutes at 75°C, then the solution were transferred to the Teflon-lined steel autoclave and heated at 110°C for 10 h. After cooled to room temperature, the white powder was collected by centrifugation and washed with DMF and CH_2Cl_2 and dried in a vacuum oven for 20 h at 115°C.

Synthesis of PVDF-HFP/MOF-5 composites electrolyte membranes

PVDF-HFP power was dissolved in DMA (5.8 mL) using magnetic stirring for 10 hours at 65°C. Different mass ratios of MOF-5 were added and stir for 1 hour. After cooling to room temperature, the degassed casting solution was dumped on a spotless glass and scraped to 200 μm with a doctor blade. The loading of MOF-5 in the composites varies from 1 to 5 wt%, which were marked as PVDF-HFP/MOF-5-X (X= I, II, III, IV and V). The thin films were produced by the immersion-precipitation phase inversion means. The thin films were dried before using at 100ºC

in a vacuum for 24 h. The composites electrolyte membranes were immersed in 1M $LiPF_6$ liquid electrolyte for 24 h to achieve the purpose of activation.

Fabrication of MOF-5 electrolyte membrane

MOF-5 was dispersed into isopropyl alcohol together with 60% PTFE emulsion at a mass ratio of 9:1 by milling and the paste was rolled into thin film about 200 μm and cut into pieces.[2] PVDF-HFP/MOF-5 composite membranes were cut into pieces directly. After drying 12 h in vacuum, MOF-5 electrolyte membrane was immersed in 1M $LiPF_6$ liquid electrolyte for 24 h to activate activation.

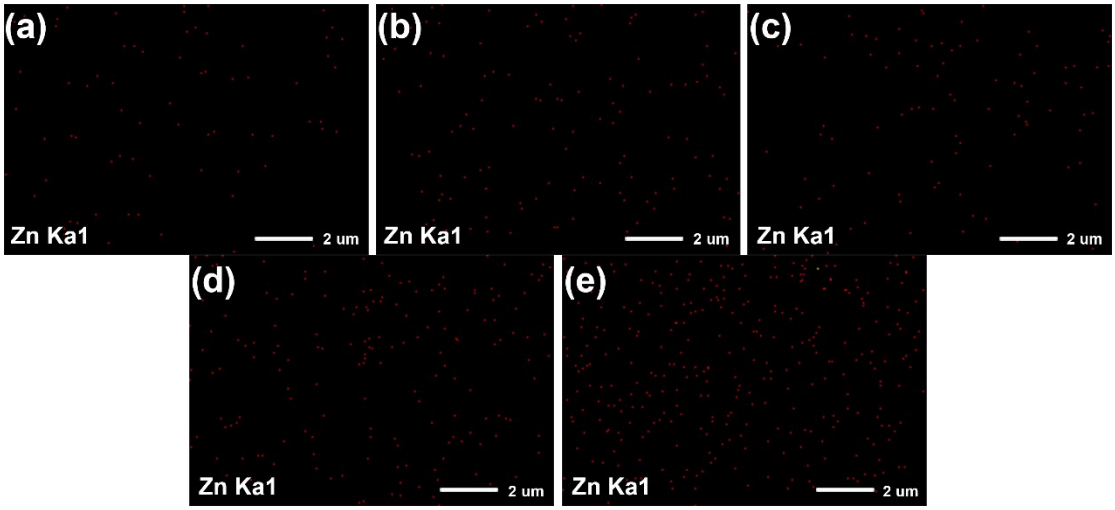

Fig. S1 Corresponding EDS mappings of pure PVDF-HFP and PVDF-HFP/MOF-5 composite membranes. (a) PVDF-HFP; (b) PVDF-HFP/MOF-5-I; (c) PVDF-HFP/MOF-5-II; (d) PVDF-HFP/MOF-5-III; (e) PVDF-HFP/MOF-5-IV and (f) PVDF-HFP/MOF-5-V.

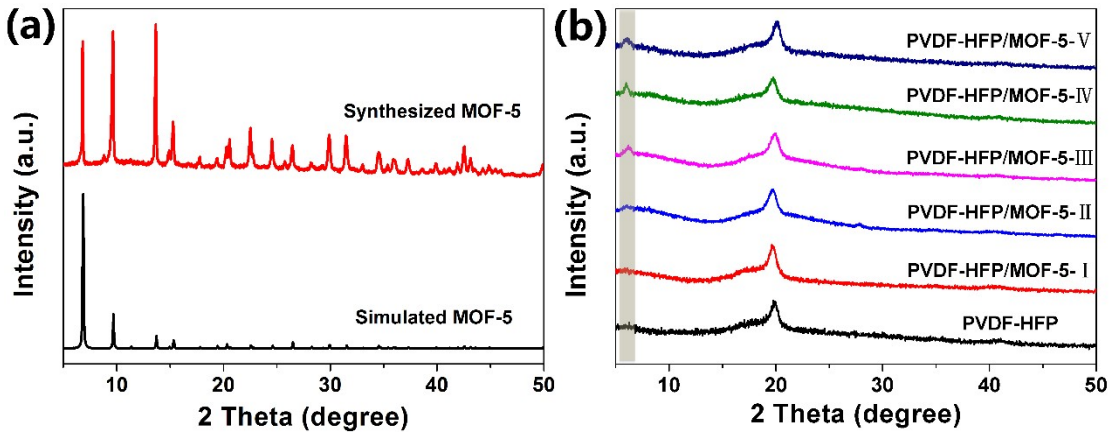

Fig. S2 (a) The experimental XRD patterns of MOF-5 agree well with the simulated ones; (b) The experimental XRD patterns of pure PVDF-HFP and various PVDF-HFP/MOF-5 composites.

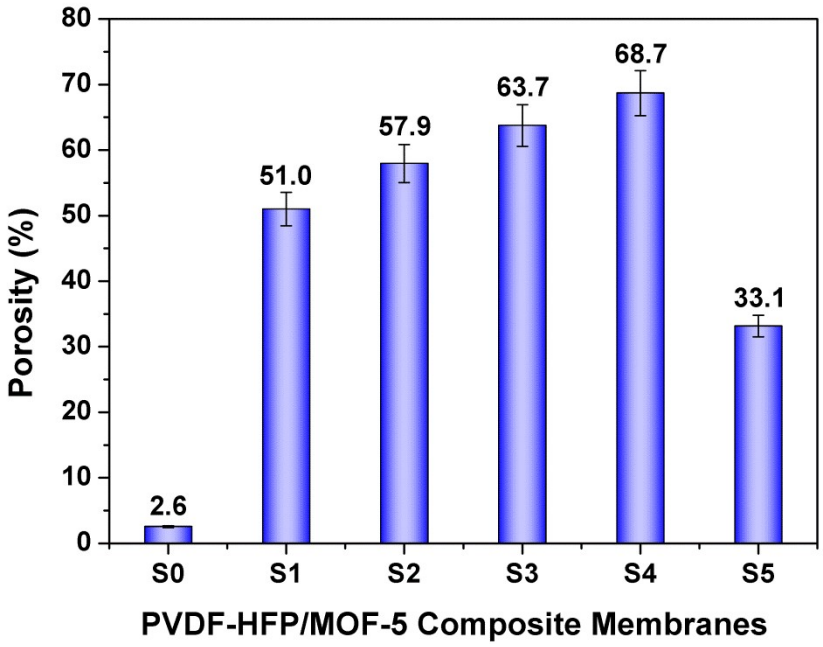

Fig. S3 Porosity of the PVDF-HFP/MOF-5 composites (PVDF-HFP/MOF-5-X (X = I, II, III, IV and V)).

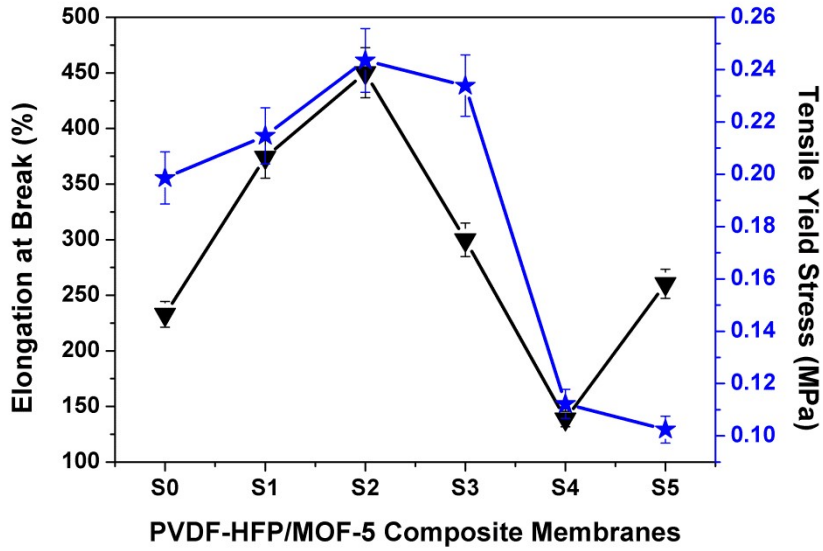

Fig. S4 Elongation at break and tensile yield stress of various PVDF-HFP/MOF-5 composite membranes.

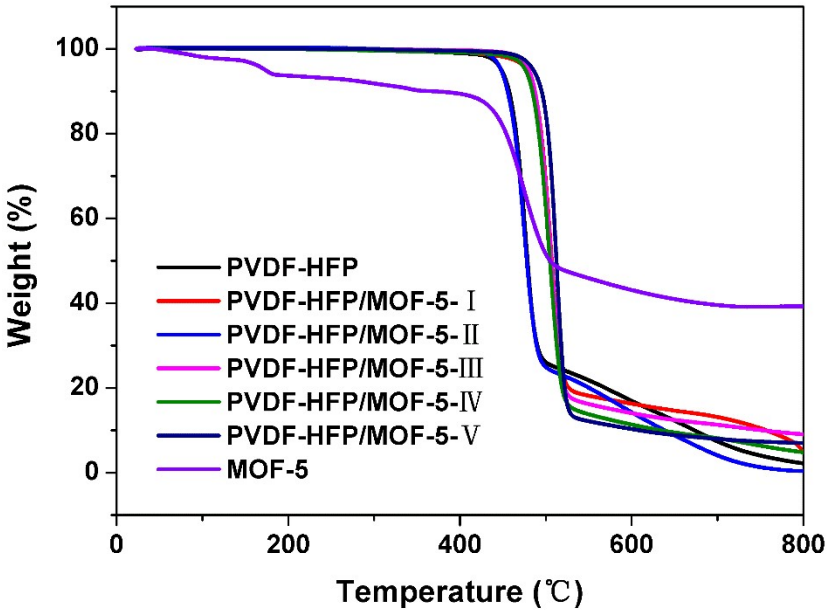

Fig. S5 TGA curves of pure PVDF-HFP, PVDF-HFP/MOF-5 composites and compound MOF-5 upon N_2 atmosphere at the range of 23–800°C.

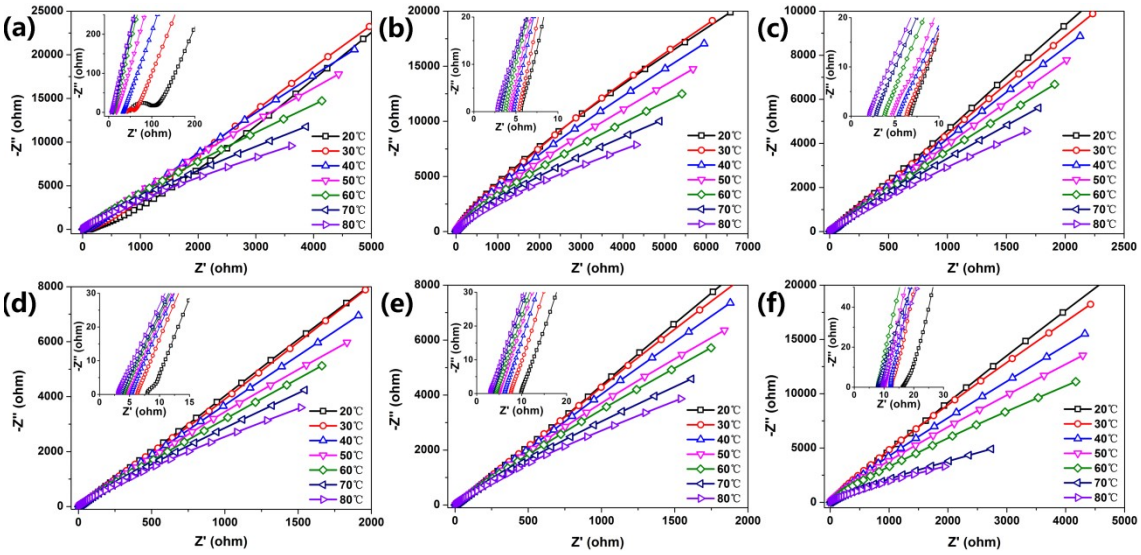

Fig. S6 EIS curves of PVDF-HFP/MOF-5 composites at temperature from 20 to 80ºC: (a) pure PVDF-HFP; (b) PVDF-HFP/MOF-5-I; (c) PVDF-HFP/MOF-5-II; (d) PVDF-HFP/MOF-5-III; (e) PVDF-HFP/MOF-5-IV and (f) PVDF-HFP/MOF-5-V.

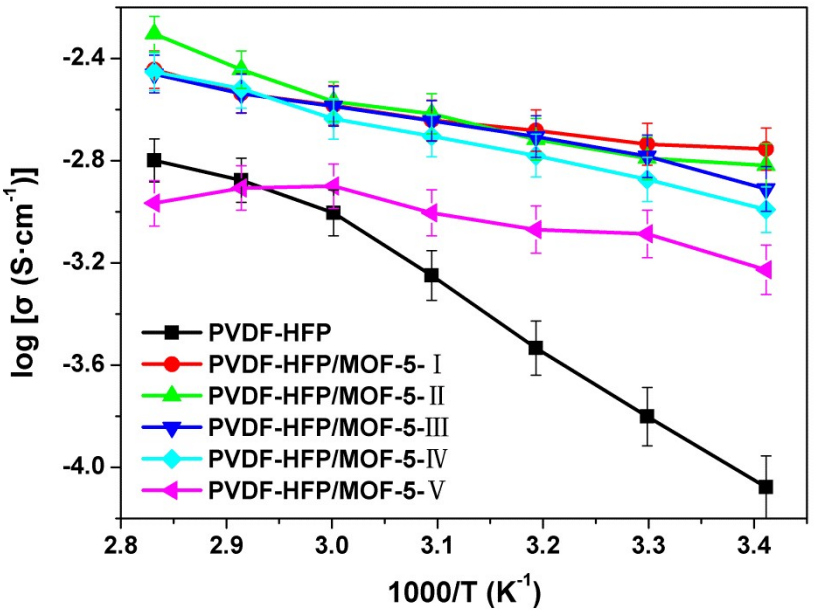

Fig. S7 Arrhenius plots of various PVDF-HFP/MOF-5 composites.

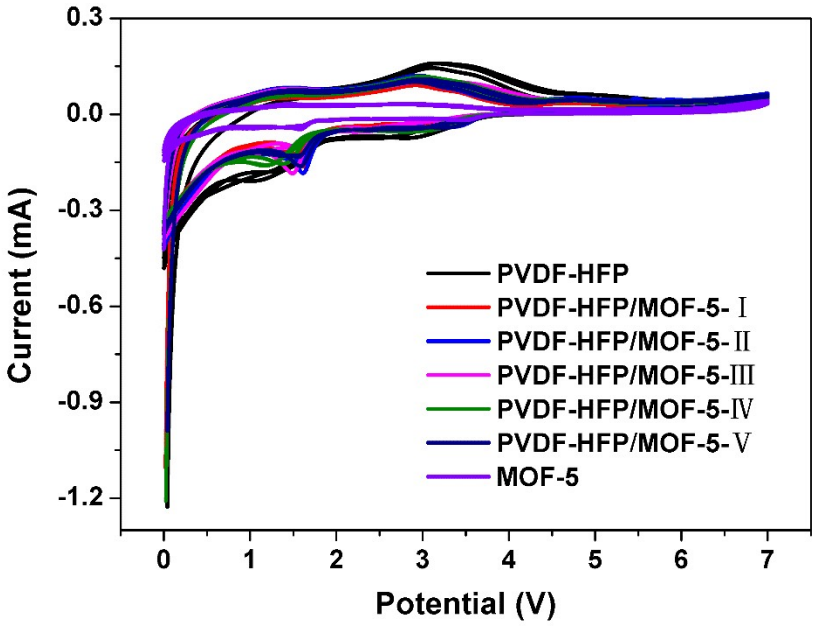

Fig. S8 CV curves of ss/electrolyte membrane/Li cell.

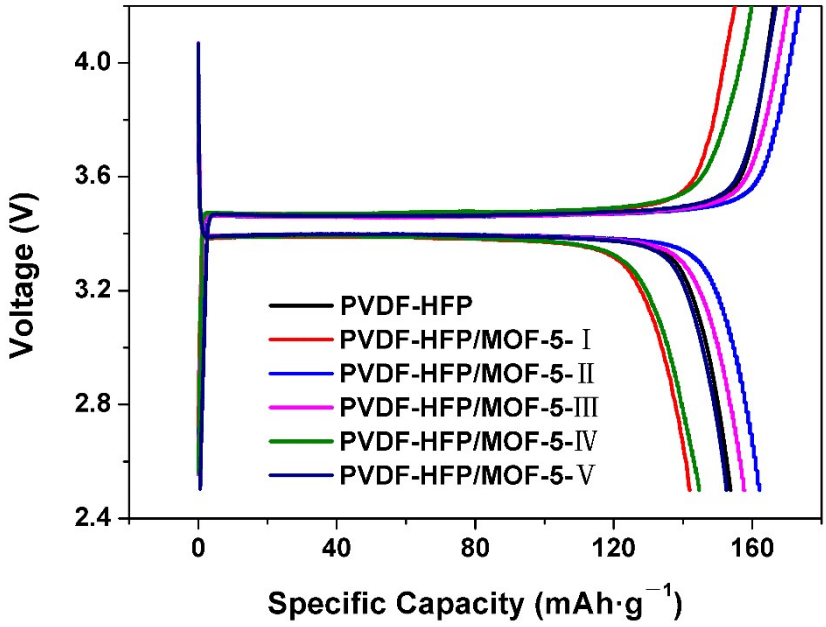

Fig. S9 Charge/discharge profiles of PVDF-HFP/MOF-5 composites at 0.1 C.

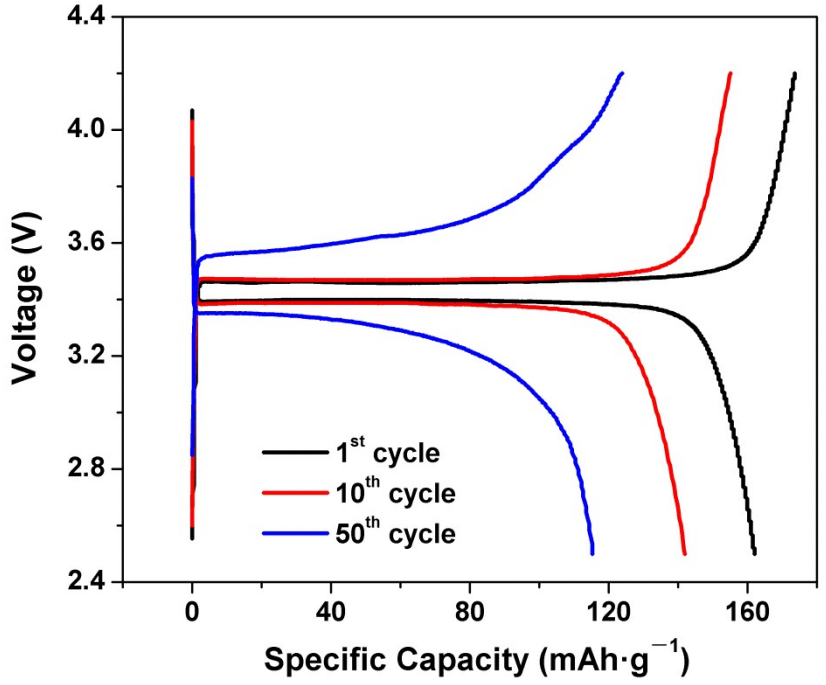

Fig. S10 Charge/discharge profiles of PVDF-HFP/MOF-5-II at 0.1 C (at 1st, 10th, 50th cycle).

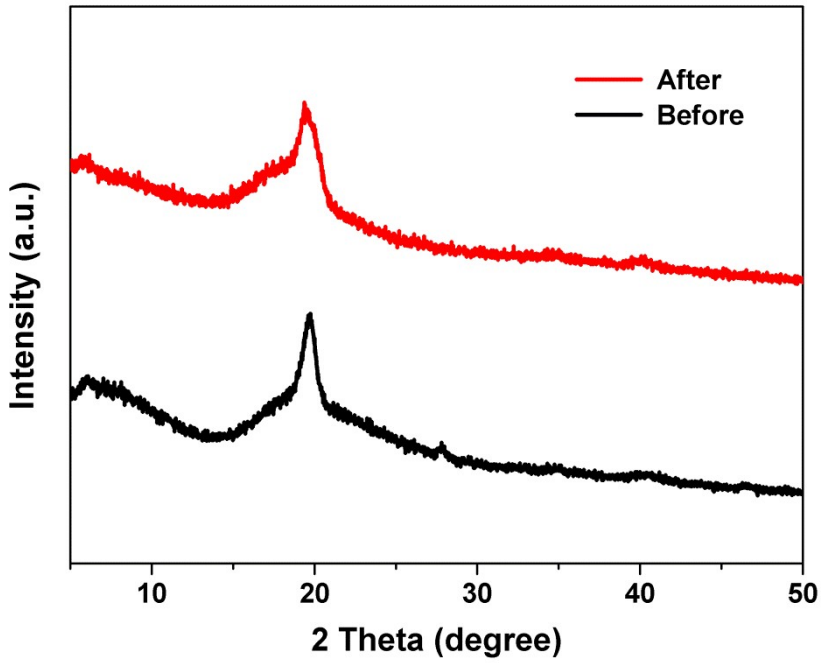

Fig.S11 The XRD patterns of PVDF-HFP/MOF-5-II composites before and after cycle.

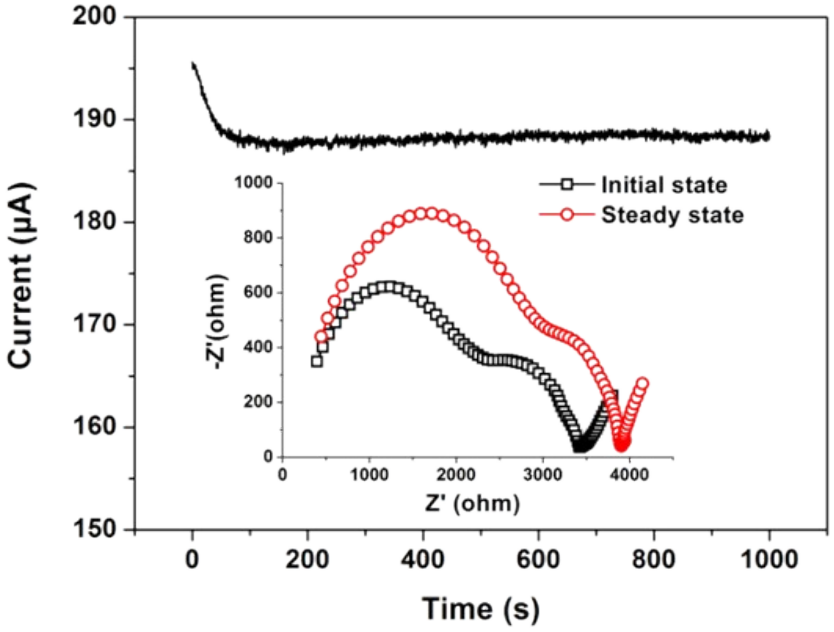

Fig. S12 CA curves of the Li/ MOF-5 electrolyte membrane/Li cell.

References

1. M. Arjmandi, A. Altaee, A. Arjmandi, M. Pourafshari Chenar, M. Peyravi and M. Jahanshahi, *Solid State Sci.*, 2020, **106**, 106292.
2. Z. Wang, R. Tan, H. Wang, L. Yang, J. Hu, H. Chen and F. Pan, *Adv. Mater.*, 2018, **30**, 1704436.