Supplementary Material

Pressure-Induced Enhancement of Proton Conduction across MOF Grain Boundaries

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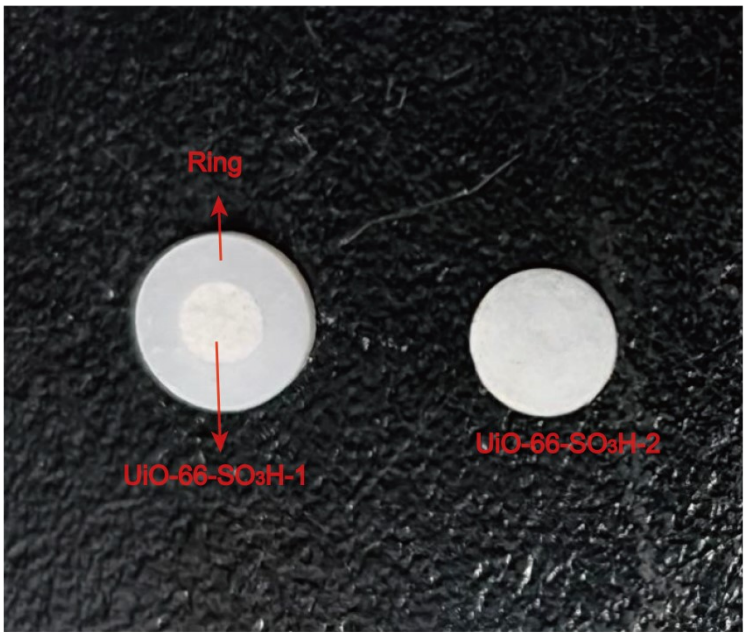

Fig. S1. Left: UiO-66-SO_3H-1 sample; Right: UiO-66-SO_3H-2 sample.

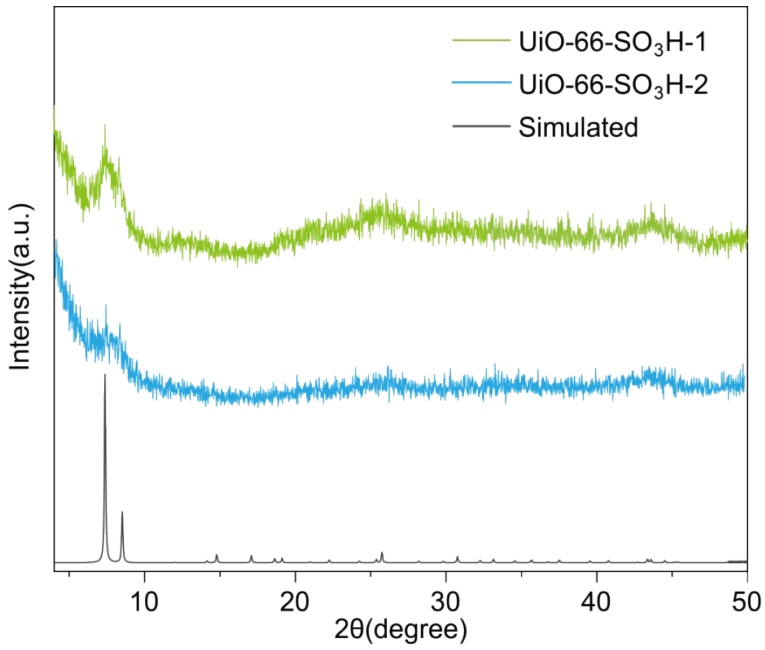

Fig. S2. PXRD of UiO-66-SO_3H-1 and UiO-66-SO_3H-2.

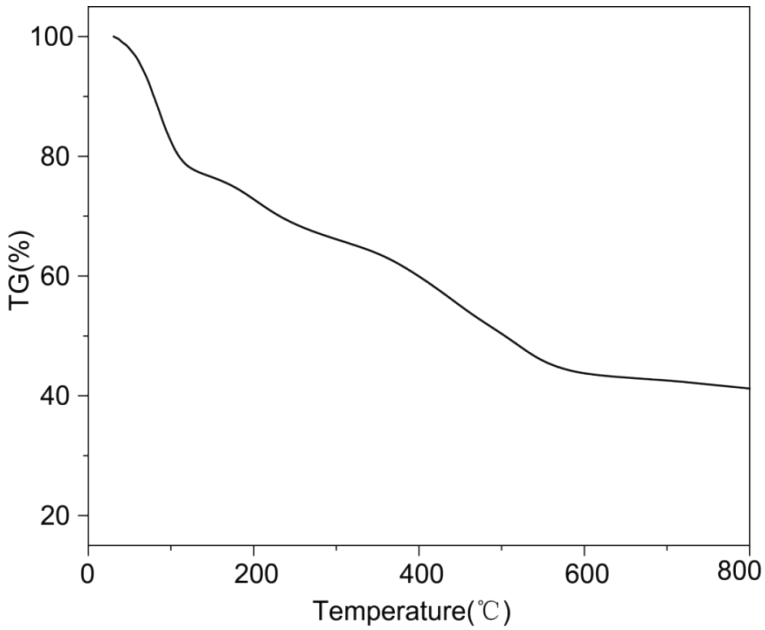

Fig. S3. TG of UiO-66-SO_3H.

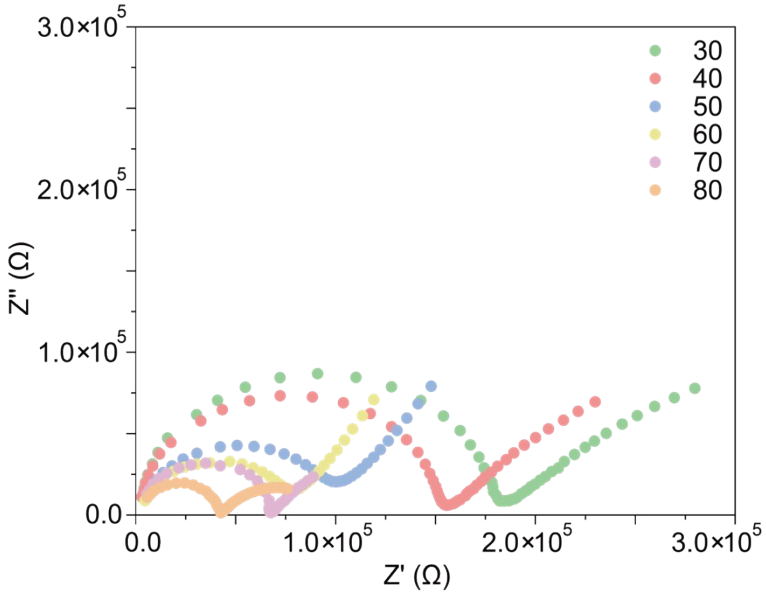

Fig. S4. Nyquist plot for UiO-66-SO_3H-1 at 100% RH versus different temperatures.

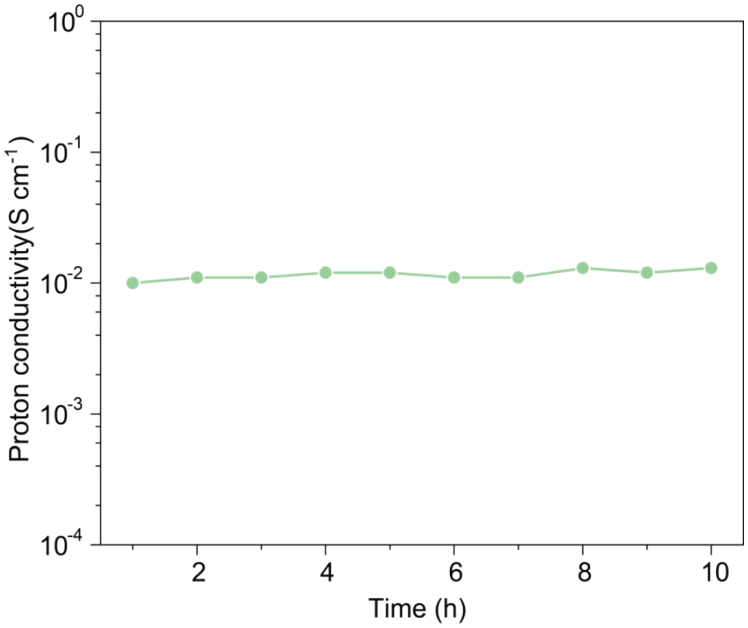

Fig. S5. Proton conductivity of UiO-66-SO_3H-2 versus time at 80 ℃ and 100% RH.

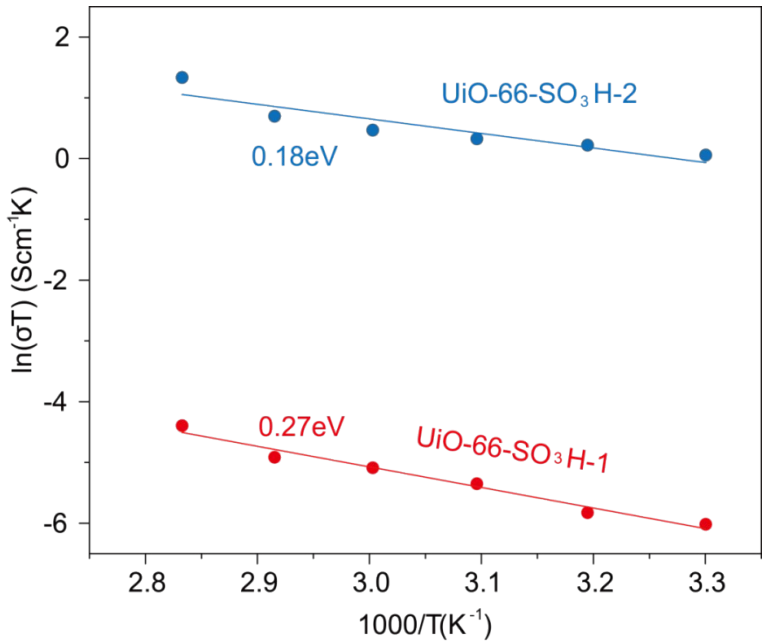

Fig. S6. Arrhenius plots of proton conductivity for UiO-66-SO_3H-1 and UiO-66-SO_3H-2.

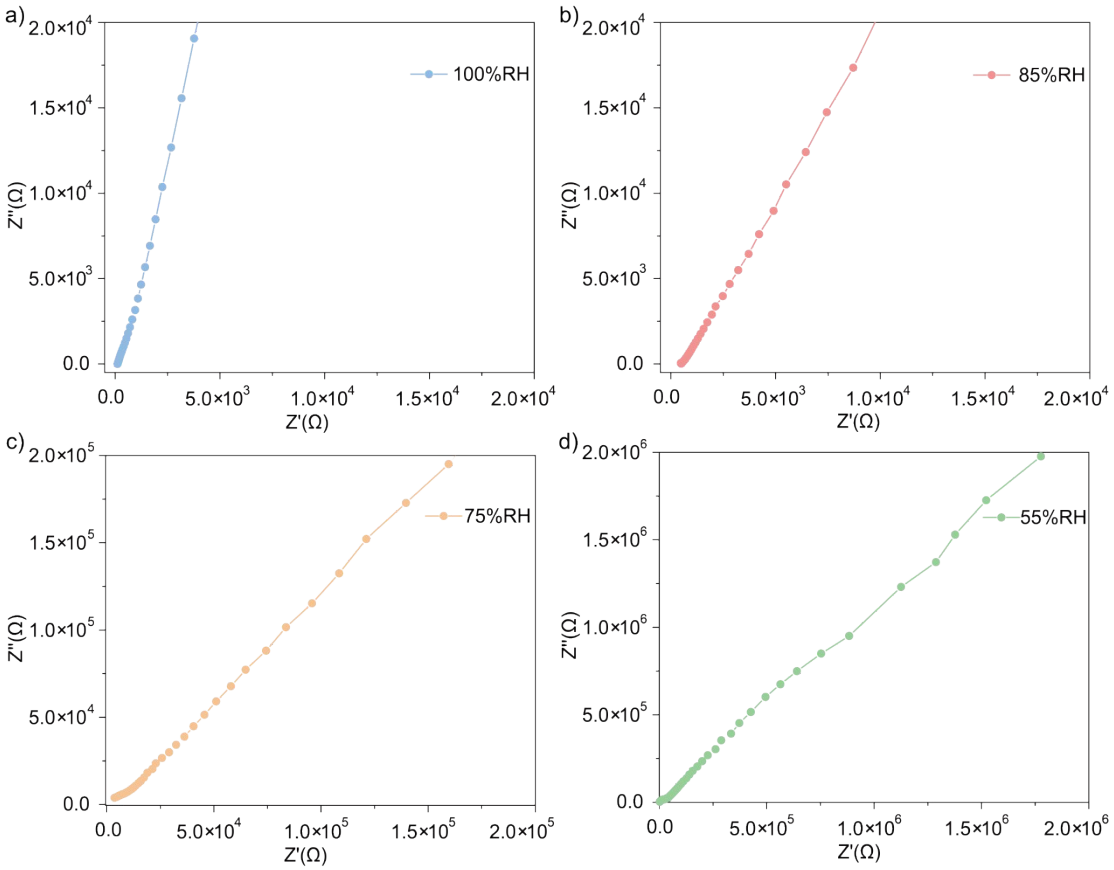

Fig. S7. Nyquist plot for UiO-66-SO_3H-2 at 30℃ versus different RHs.

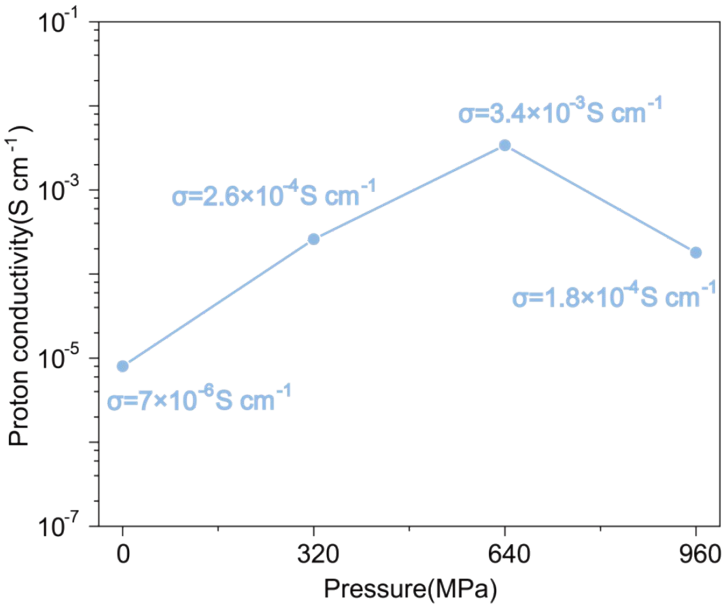

Fig. S8. Proton conductivity of UiO-66-SO_3H pellets as a function of applied pressure.

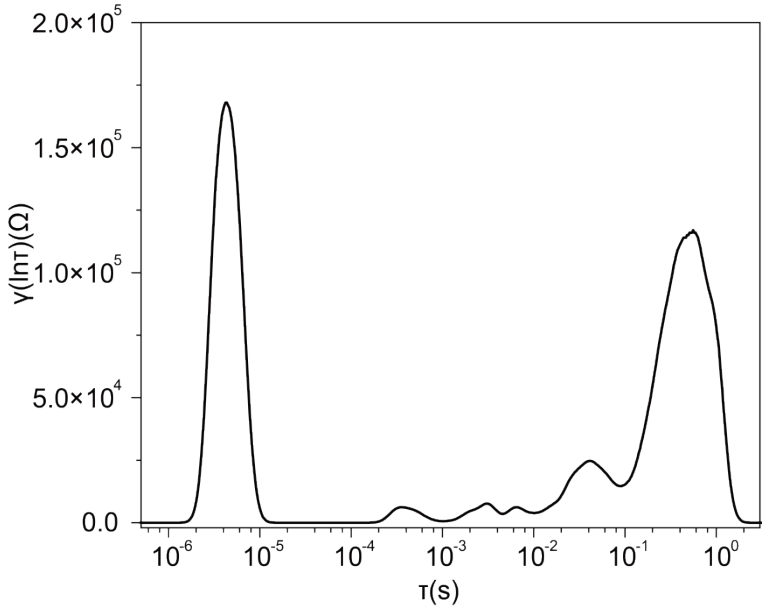

Fig. S9. DRT diagram of UiO-66-SO_3H-1 at 40℃.

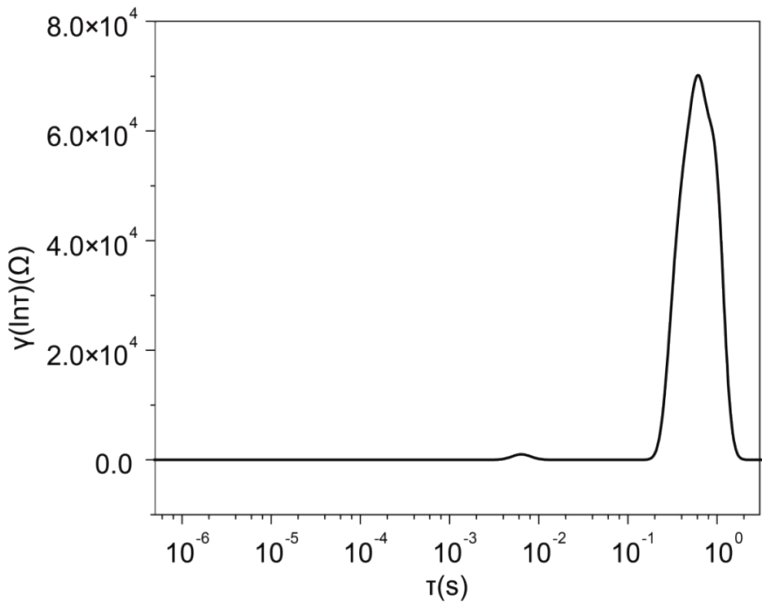

Fig. S10. DRT diagram of UiO-66-SO_3H-2 at 40℃.

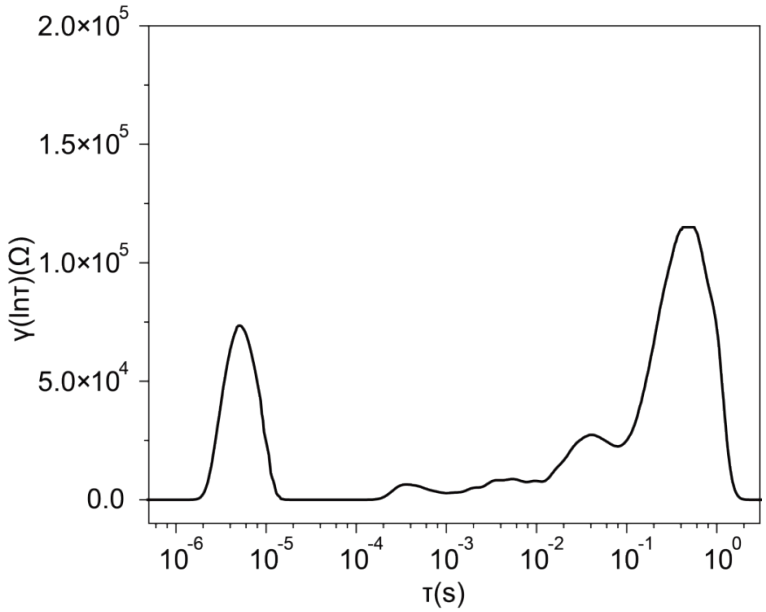

Fig. S11. DRT diagram of UiO-66-SO_3H-1 at 50℃.

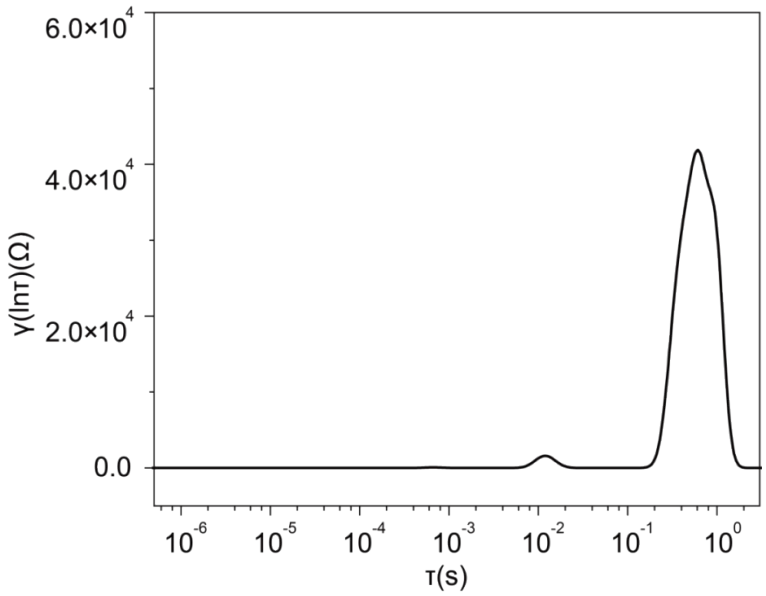

Fig. S12. DRT diagram of UiO-66-SO_3H-2 at 50℃.

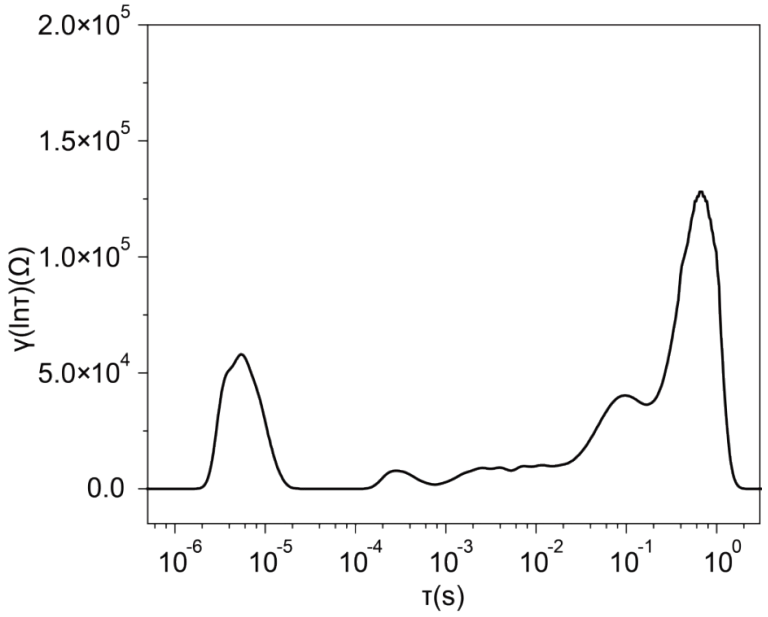

Fig. S13. DRT diagram of UiO-66-SO_3H-1 at 60℃.

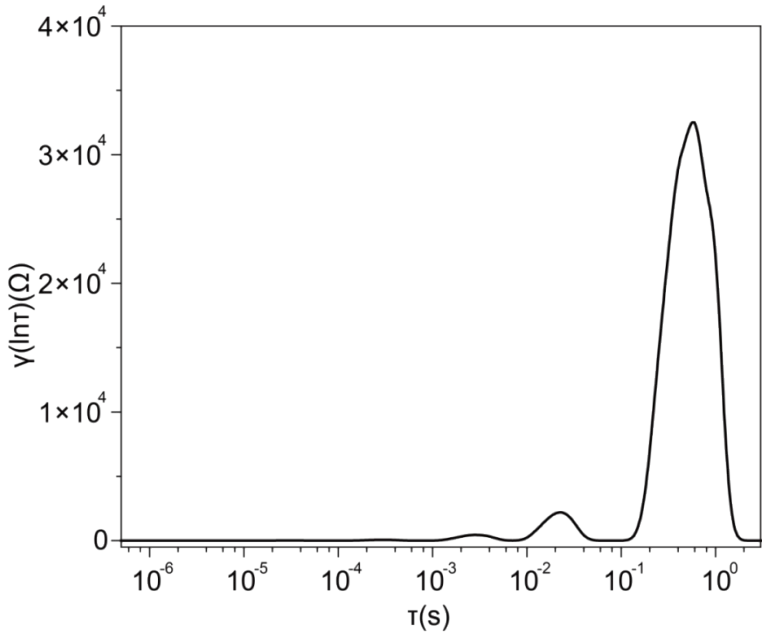

Fig. S14. DRT diagram of UiO-66-SO_3H-2 at 60℃.

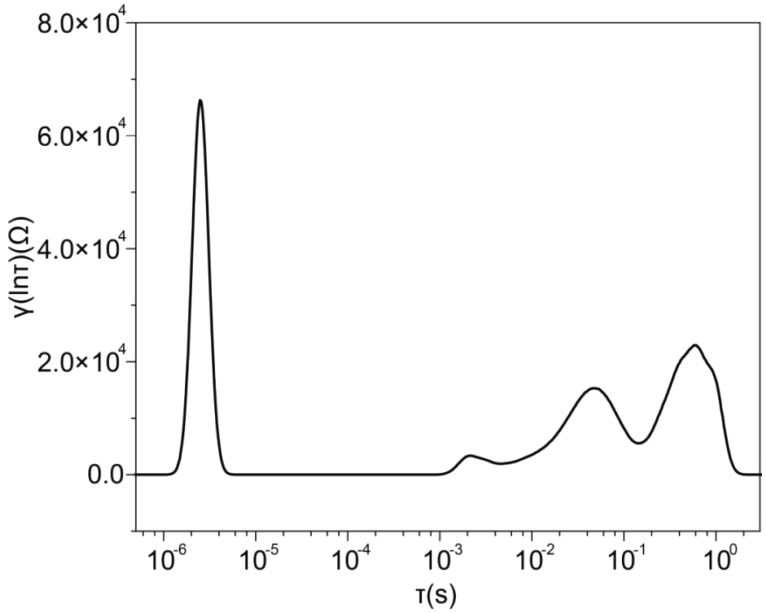

Fig. S15. DRT diagram of UiO-66-SO_3H-1 at 70℃.

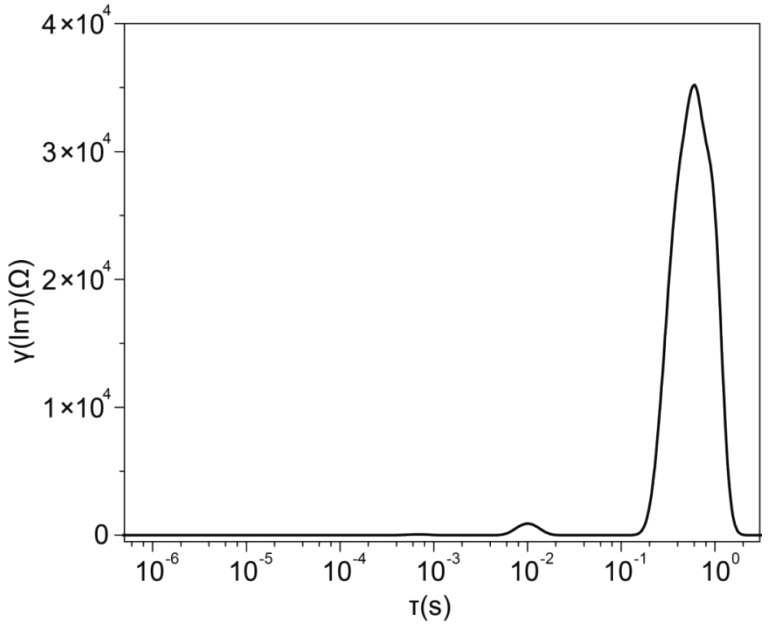

Fig. S16. DRT diagram of UiO-66-SO_3H-2 at 70℃.

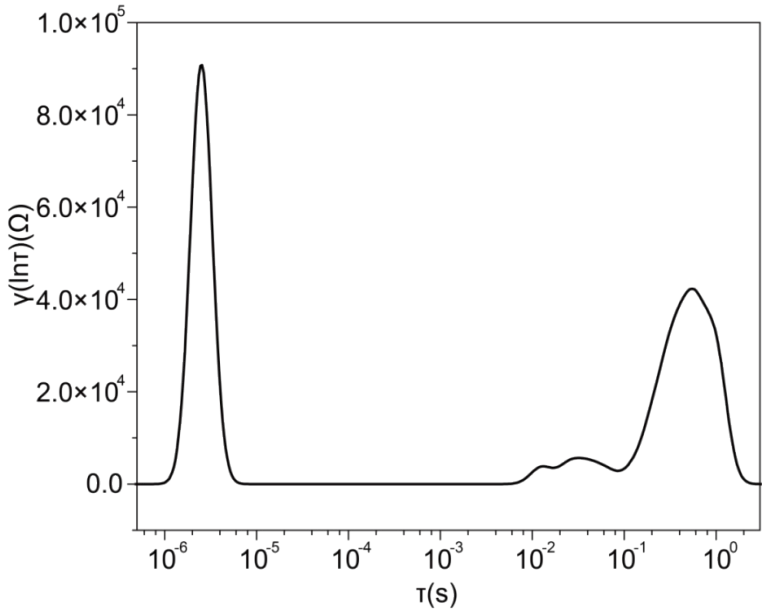

Fig. S17. DRT diagram of UiO-66-SO_3H-1 at 80℃.

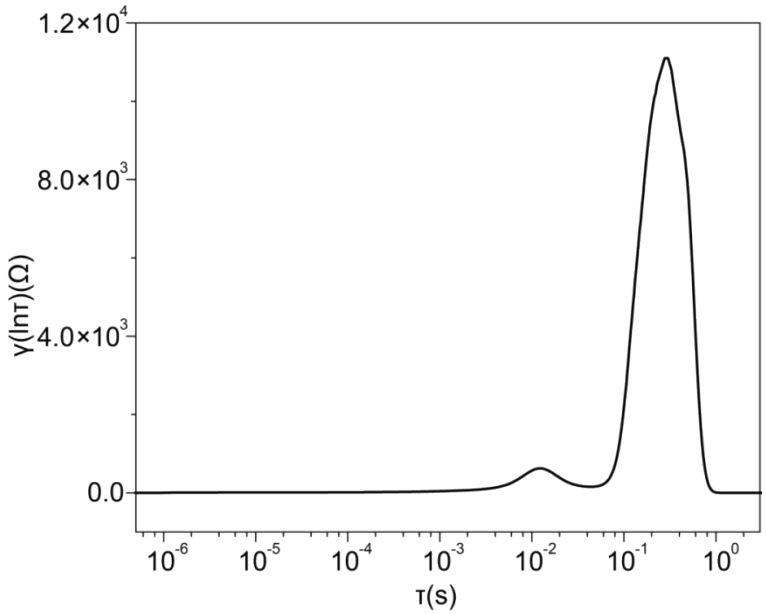

Fig. S18. DRT diagram of UiO-66-SO_3H-2 at 80℃.

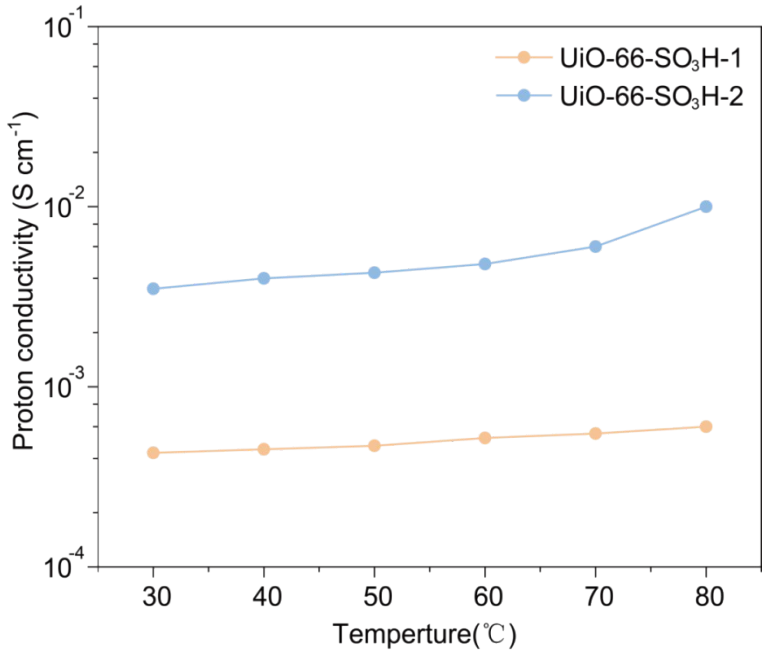

Fig. S19. Comparison of bulk proton conductivity among UiO-66-SO_3H-1 and UiO-66-SO_3H-2.

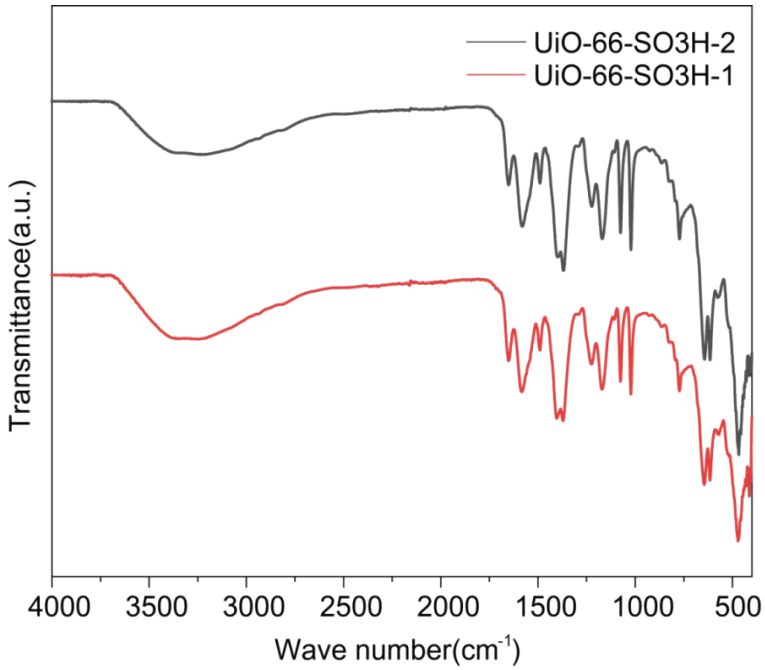

Fig. S20. FT-IR spectra of UiO-66-SO_3H-1 and UiO-66-SO_3H-2.

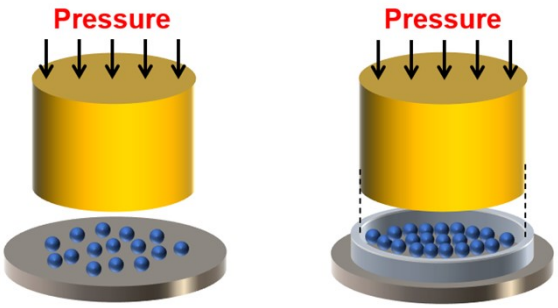

Fig. S21. Schematic comparison of the preparation protocols for UiO-66-SO_3H-3 (left) and UiO-66-SO_3H-2 (right).

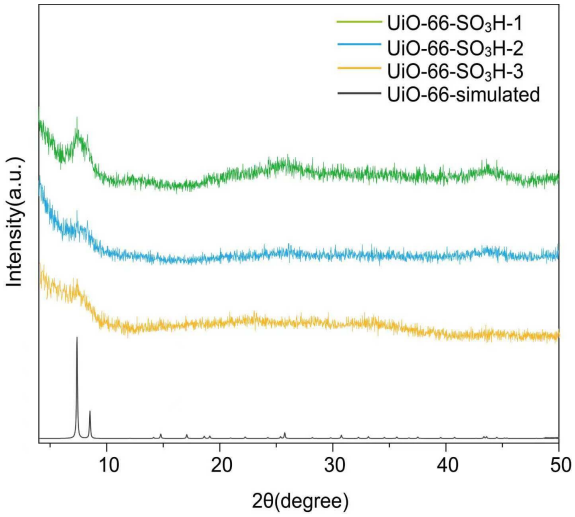

Fig. S22. PXRD of the three samples.

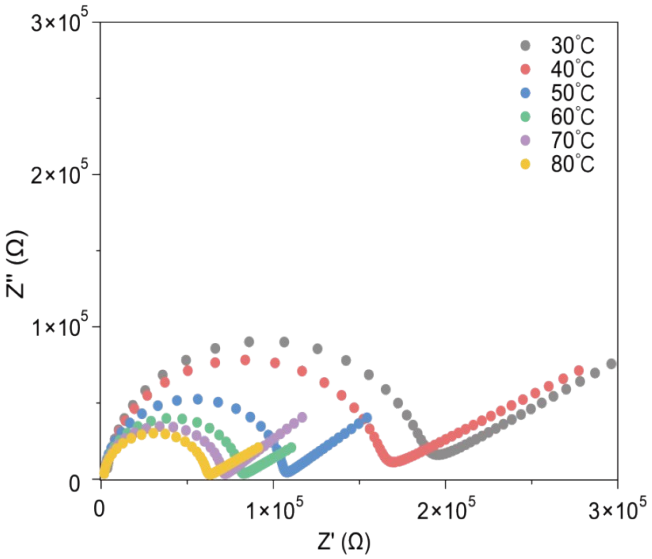

Fig. S23. Nyquist plot for UiO-66-SO_3H-3 at 100% RH versus different temperatures.

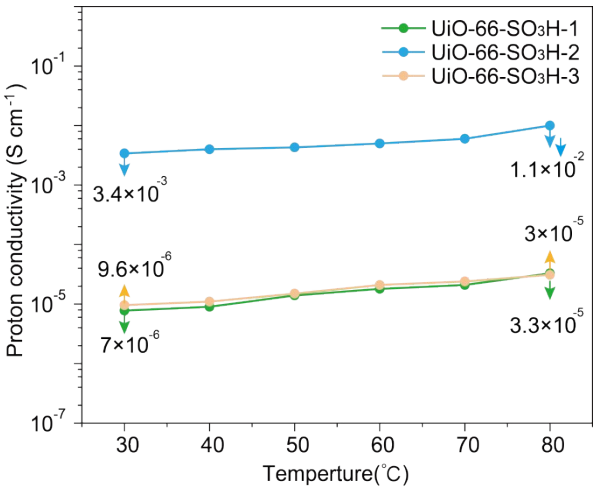

Fig. S24. Proton conductivity of the three samples.

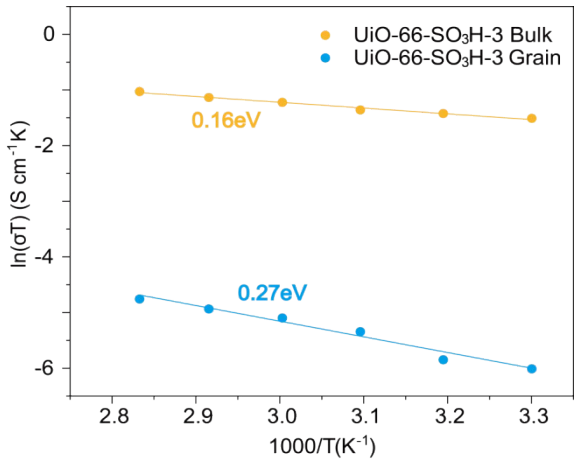

Fig. S25. Arrhenius plots of proton conductivity for UiO-66-SO_3H-$_3$ bulk and grain boundary part.

Table 1. Proton conductivity of other MOFs.

Compounds	Conductivity (S cm^{-1})	Conditions
UiO-66-SO_3H-2	1.13×10^{-2}	T = 80°C, 100%RH
MIL-101-SO_3H	1.16×10^{-2}	T = 80°C, 100%RH
CUST-861	2.96×10^{-2}	T =90°C, 98%RH
FDM-151	1.1×10^{-2}	T =95°C, 100%RH
MOF-bpe	1.95×10^{-2}	T=95 °C, 100% RH
MOF-bpy	2.6×10^{-2}	T=95 °C, 100% RH
MOP-1@DUT-68	7.58×10^{-3}	T=80 °C, 98% RH
MIP-202(Zr)	1.1×10^{-2}	T=90 °C, 95% RH
Im@MOF-808	3.45×10^{-2}	T=65°C, 98% RH
DUT-67(Zr)	2.98×10^{-3}	T=100°C, 98% RH
60-UiO-66-1.8	3×10^{-2}	T=100°C, 98% RH
ZrP-3	1.91×10^{-2}	T=90 °C, 95% RH
MOF-808-IMDC	1.11×10^{-2}	T=80°C, 98% RH
YCu161	1.84×10^{-3}	T=90°C, 98% RH
Cu-DSOA	1.9×10^{-3}	T=85°C, 98% RH
MOG-UiO-66	1.23×10^{-2}	T=80°C, 75% RH
MOG-MOF-808	3.06×10^{-2}	T=80°C, 75% RH
Nafion	8.0×10^{-2}	T = 80°C, 50%RH
$Zr_6O_4(OH)_8L_{4.2}\cdot xH_2O$	1.93×10^{-3}	T=65°C, 95% RH

Table S2. The fitting parameters of the Nyquist plots of UiO-66-SO_3H-1.

Tem(℃)	R1	R2	CPE1-T	CPE1-P	CPE2-T	CPE2-P
30	3295	178200	2.540×10^{-11}	1.013	3.522×10^{-6}	0.42821
40	3001	147370	2.606×10^{-11}	1.011	5.495×10^{-6}	0.56792
50	2807	93319	9.468×10^{-11}	0.93674	2.769×10^{-6}	0.87364
60	2682	73826	1.159×10^{-10}	0.92414	8.634×10^{-6}	0.33697
70	2600	63587	2.499×10^{-11}	1.015	3.896×10^{-6}	0.46375
80	2374	38168	3.462×10^{-11}	0.99999	4.137×10^{-6}	0.45731

Table S3. The fitting parameters of the Nyquist plots of UiO-66-SO_3H-2.

Tem(℃)	R	CPE-T	CPE-P
30	114.3	7.641×10^{-6}	0.83685
40	99.21	1.735×10^{-5}	0.71624
50	93.15	1.649×10^{-5}	0.78415
60	83.21	1.002×10^{-5}	0.82544
70	68.82	1.842×10^{-5}	0.81035
80	35.53	8.434×10^{-5}	0.67573

Table S4. The fitting parameters of the Nyquist plots of UiO-66-SO_3H-3.

Tem(℃)	R1	R2	CPE1-T	CPE1-P	CPE2-T	CPE2-P
30	1945	145280	5.047×10^{-11}	0.99933	2.6227×10^{-6}	0.3603
40	1840	123830	2.772×10^{-11}	0.99349	3.529×10^{-6}	0.33709
50	1783	96147	5.105×10^{-11}	1.012	7.043×10^{-6}	0.42861
60	1602	67228	8.126×10^{-10}	0.89714	9.121×10^{-6}	0.29087
70	1512	57664	7.020×10^{-11}	1.009	6.999×10^{-6}	0.44958
80	1400	45221	9.483×10^{-11}	1.014	1.338×10^{-5}	0.36942