Supplementary Information

Fabrication of Mixed Matrix Membranes with Regulated MOF Fillers via Incorporating Guest Molecules for Optimizing Light Hydrocarbons Separation Performance

Yang Feng[b, ‡], LiTing Yu[a, ‡], Kai Zhang[b], Weidong Fan[a], Lili Fan[a, *], Zixi Kang[a, *], Daofeng Sun[a]

[a]*School of Materials Science and Engineering, China University of Petroleum (East China), Qingdao, Shandong, 266580, PR China.*
[b]*College of Chemistry and Chemical Engineering, China University of Petroleum (East China), Qingdao, Shandong, 266580, PR China.*

Contents

1. Characterizations of PIM-1

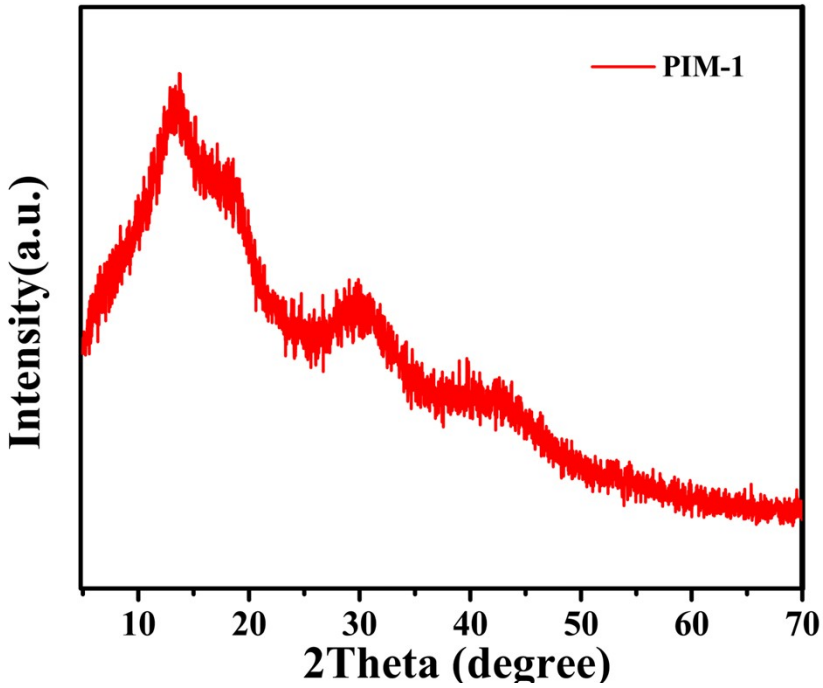

Fig. S1. PXRD patterns of **PIM-1**.

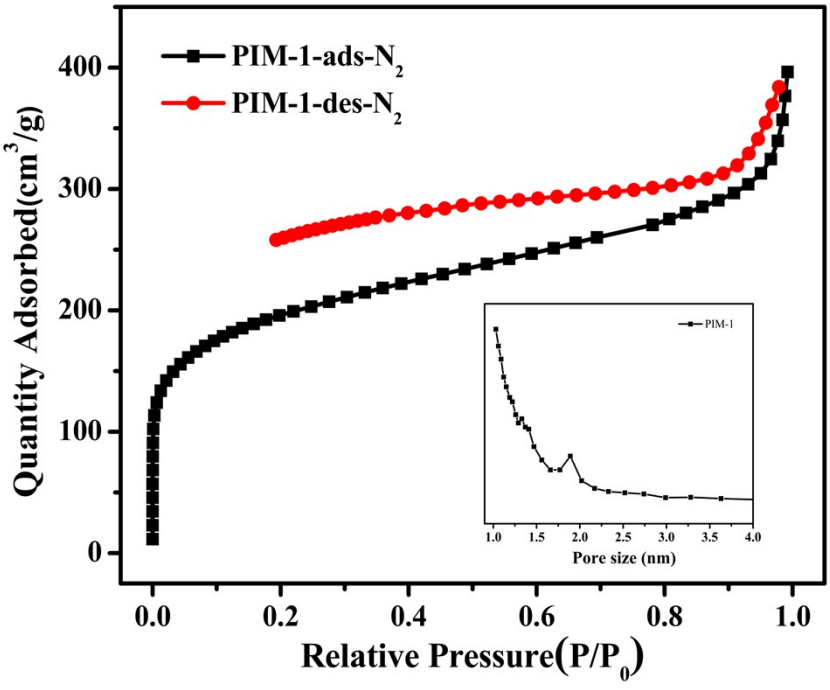

Fig. S2. N_2 adsorption/desorption isotherms of **PIM-1** at 77 K.

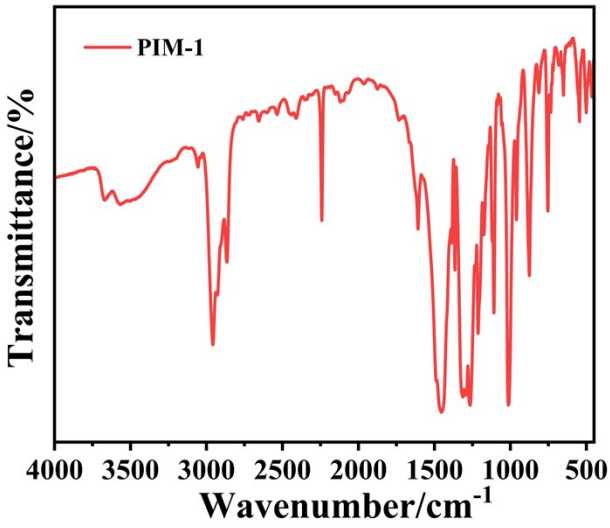

Fig. S3. FTIR spectrum of **PIM-1**.

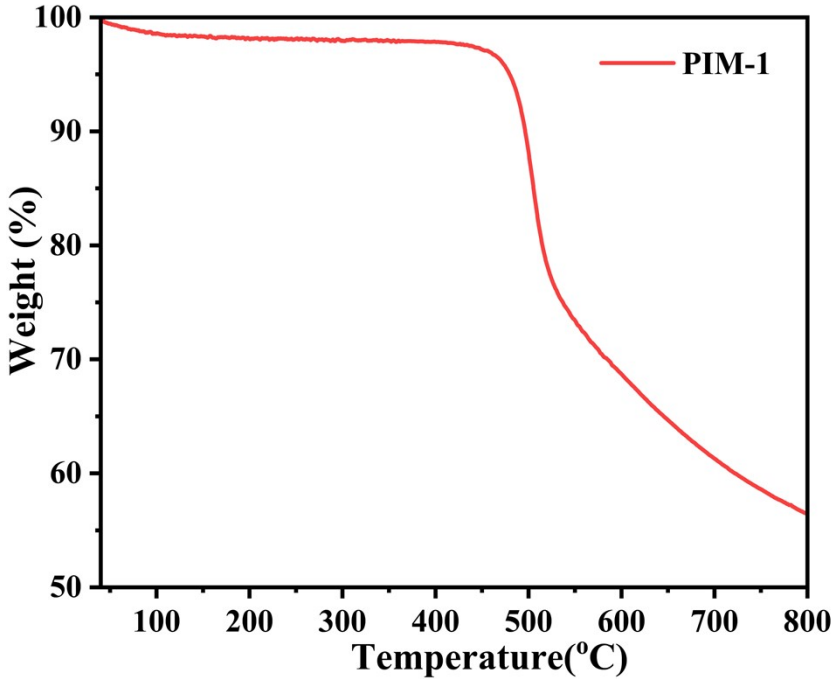

Fig. S4. TGA of **PIM-1** under a nitrogen atmosphere.

2. Characterizations of fillers

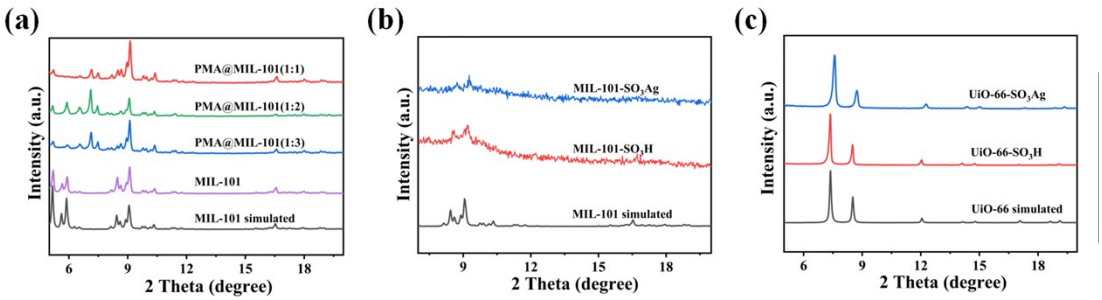

Fig. S5. The partial enlarged view of PXRD patterns for pristine and optimized (a) **MIL-101**, (b) **MIL-101-SO_3H** and (c) **UiO-66-SO_3H** series powder.

Table S1. The surface area calculated based on the N_2 (77 K) adsorption-desorption experiment for **MIL-101** and **UiO-66** series

Filler	BET(m^2/g)
MIL-101	2458
PMA@MIL-101(1:1)	1263
PMA@MIL-101(1:2)	1480
PMA@MIL-101(1:3)	1911
MIL-101-SO_3H	593
MIL-101-SO_3Ag	374
UiO-66-SO_3H	482
UiO-66-SO_3Ag	397

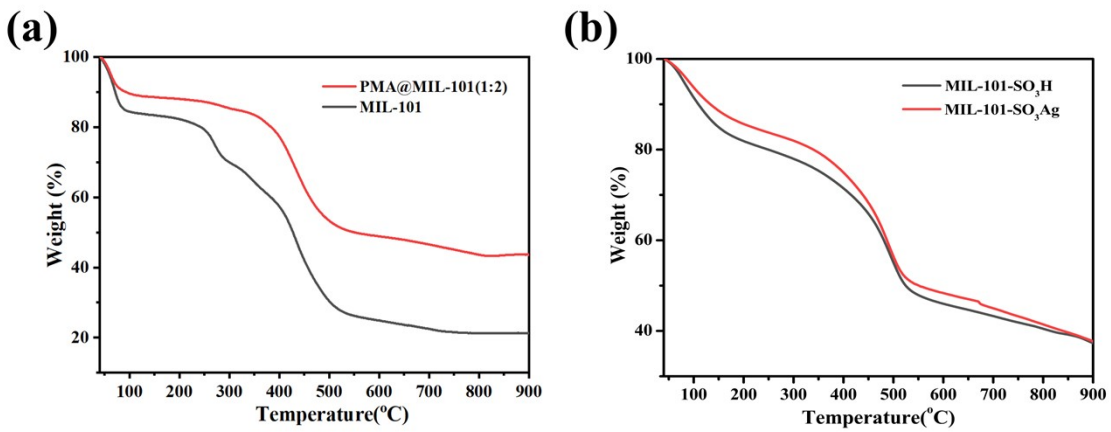

Fig. S6. TGA curves of pristine and optimized (a) **MIL-101** and (b) **MIL-101-SO_3H** series powder.

Table S2. The content of Mo in **PMA@MIL-101** series and Ag(I) in **MIL-10was 1-SO_3Ag /UiO-66-SO_3Ag** calculated by ICP results.

Filler	Mo (wt%)	Ag(I) (wt%)
PMA@MIL-101(1:3)	10.06	-
PMA@MIL-101(1:2)	16.40	-
PMA@MIL-101(1:1)	18.23	-
MIL-101-SO_3Ag	-	0.63
UiO-66-SO_3Ag	-	1.13

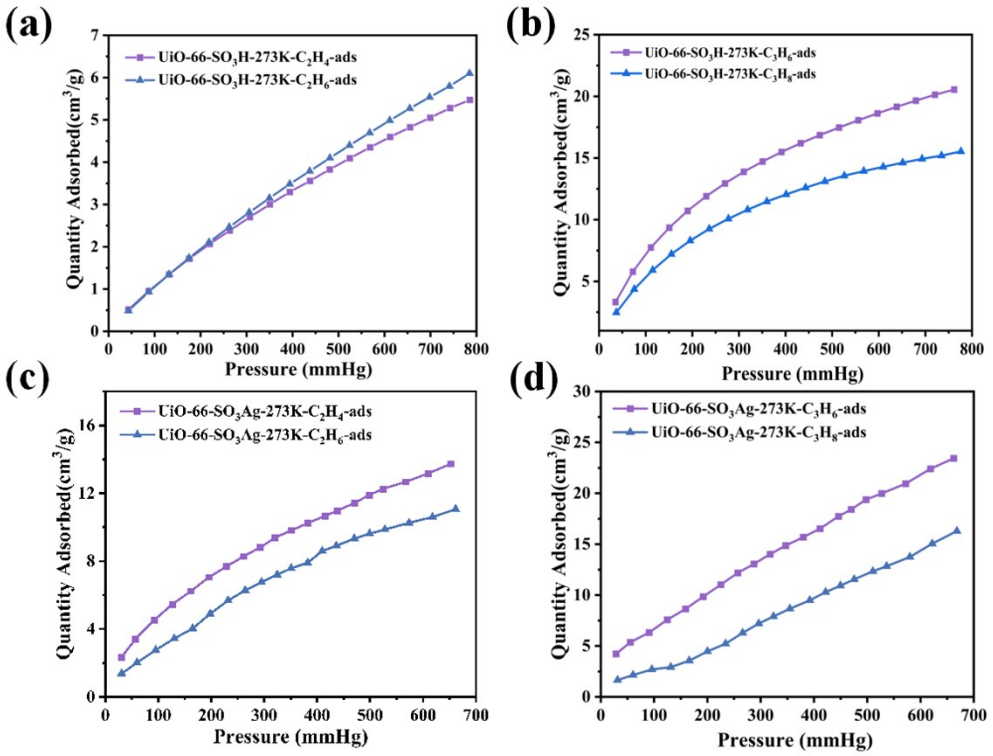

Fig. S7. **(a)** Comparison of the gas uptake of C_2H_4 and C_2H_6 for **UiO-66-SO$_3$H** at 273 K**; (b)** Comparison of the gas uptake of C_3H_6 and C_3H_8 for **UiO-66-SO$_3$H** at 273 K**; (c)** Comparison of the gas uptake of C_2H_4 and C_2H_6 for **UiO-66-SO$_3$Ag** at 273 K**; (d)** Comparison of the gas uptake of C_3H_6 and C_3H_8 for **UiO-66-SO$_3$Ag** at 273 K**.**

3. Characterizations and performance of membranes

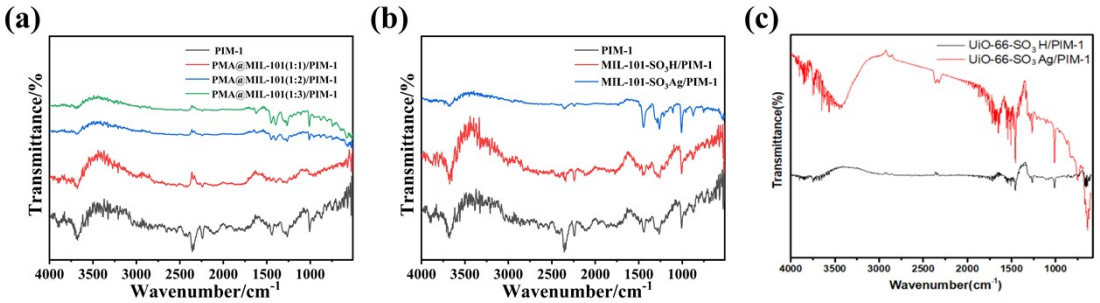

Fig. S8. Comparison of the FTIR for pristine and optimized (a) **MIL-101** and (b) **MIL-101-SO_3H** and (c) **UiO-66-SO_3H** membranes.

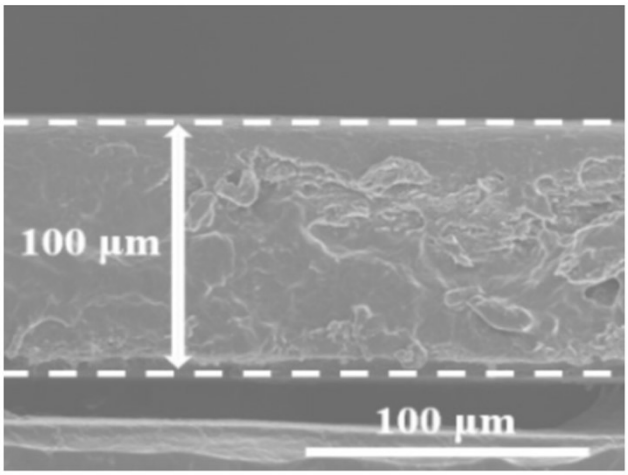

Fig. S9. The cross-sectional SEM images of **PIM-1.**

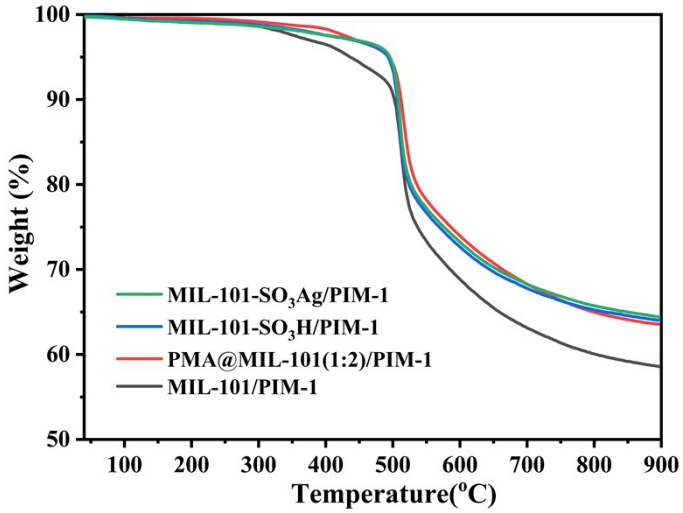

Fig. S10. TGA of **MIL-101 and MIL-101-SO_3H** series MMMs under a N_2 atmosphere**.**

Table S3 The thicknesses of the membranes measured based on the SEM image or with the digital micrometer

membrane	T_{SEM} (μm)	$T_{micrometer}$ (μm)
PIM-1	100±5	102±4
MIL-101/PIM-1	78±3	75±2
7.5% PMA@MIL-101(1:2)/PIM-1	62±1	65±1
MIL-101-SO_3H/PIM-1	74±2	75±3
MIL-101-SO_3Ag/PIM-1	85±3	82±2
UiO-66-SO_3H/PIM-1	76±2	73±2
UiO-66-SO_3Ag/PIM-1	81±2	85±4

Table S4 The permeability and the separation factor for all the membranes

Membrane	Permeability (Barrer)		C_2H_4/C_2H_6 selectivity	Permeability (Barrer)		C_3H_6/C_3H_8 selectivity
	C_2H_4	C_2H_6		C_3H_6	C_3H_8	
PIM-1	**1289±20**	**876±6**	**1.47±0.01**	**1562±18**	**698±7**	**2.23±0.03**
7.5%MIL-101/PIM-1	**1833±4**	**742±9**	**2.47±0.03**	**1421±35**	**348±13**	**4.08±0.06**
7.5%PMA@MIL-101(1:2)/PIM-1	**1632±9**	**567±9**	**2.88±0.06**	**1480±93**	**248±19**	**5.96±0.14**
10%PMA@MIL-101(1:2)/PIM-1	**3166±78**	**1702±36**	**1.86±0.07**	**1640±11**	**418±5**	**3.92±0.02**
MIL-101-SO_3H/PIM-1	**2037±134**	**1329±143**	**1.53±0.06**	**1828±56**	**769±59**	**2.38±0.11**
MIL-101-SO_3Ag/PIM-1	**1456±28**	**420±13**	**3.47±0.16**	**1663±6**	**428±7**	**3.89±0.07**
UiO-66-SO_3H/PIM-1	**2239±96**	**1566±66**	**1.43±0.07**	**3761±161**	**2149±200**	**1.75±0.10**
UiO-66-SO_3Ag/PIM-1	**869±5**	**349±2**	**2.49±0.03**	**1233±50**	**281±17**	**4.39±0.08**