Supplementary Information

One-Pot Synthesis of Necklace-Like MOF@CNTs: A Universal Strategy for Enhancing Molecular Separation Performance in Mixed-Matrix Membranes

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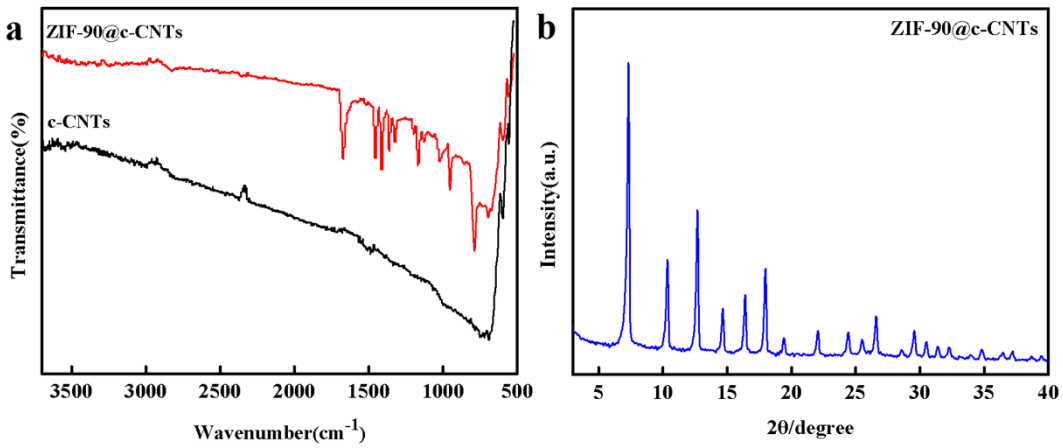

Fig. S1 (a) FTIR spectra of CNTs, and ZIF-90@c-CNTs, and (b) XRD patterns of ZIF-90@c-CNTs.

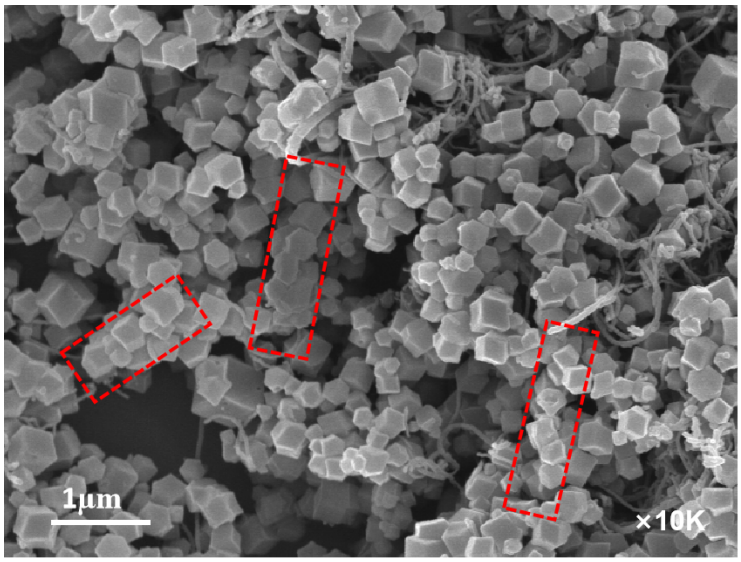

Fig. S2 SEM images of ZIF-90@c-CNTs.

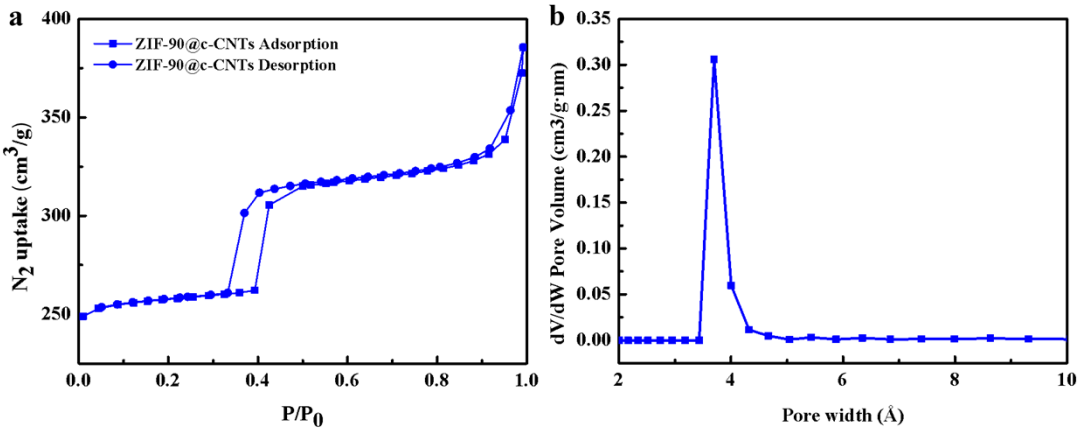

Fig. S3 (a) N_2 adsorption–desorption isotherms at 77 K and (b) pore size distribution data of as-synthesized ZIF-90@c-CNTs.

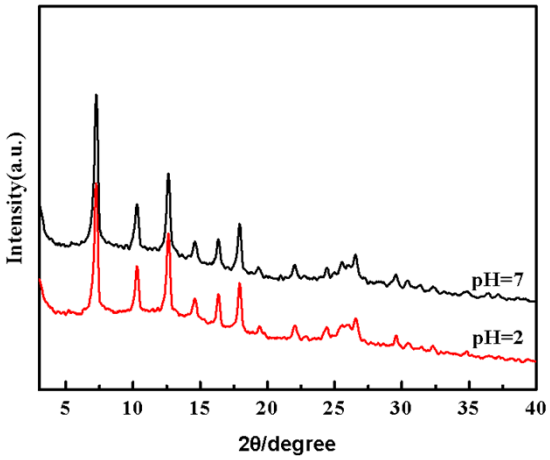

Fig. S4 XRD patterns of ZIF-90@c-CNTs before and after being immersed in pH=2 water.

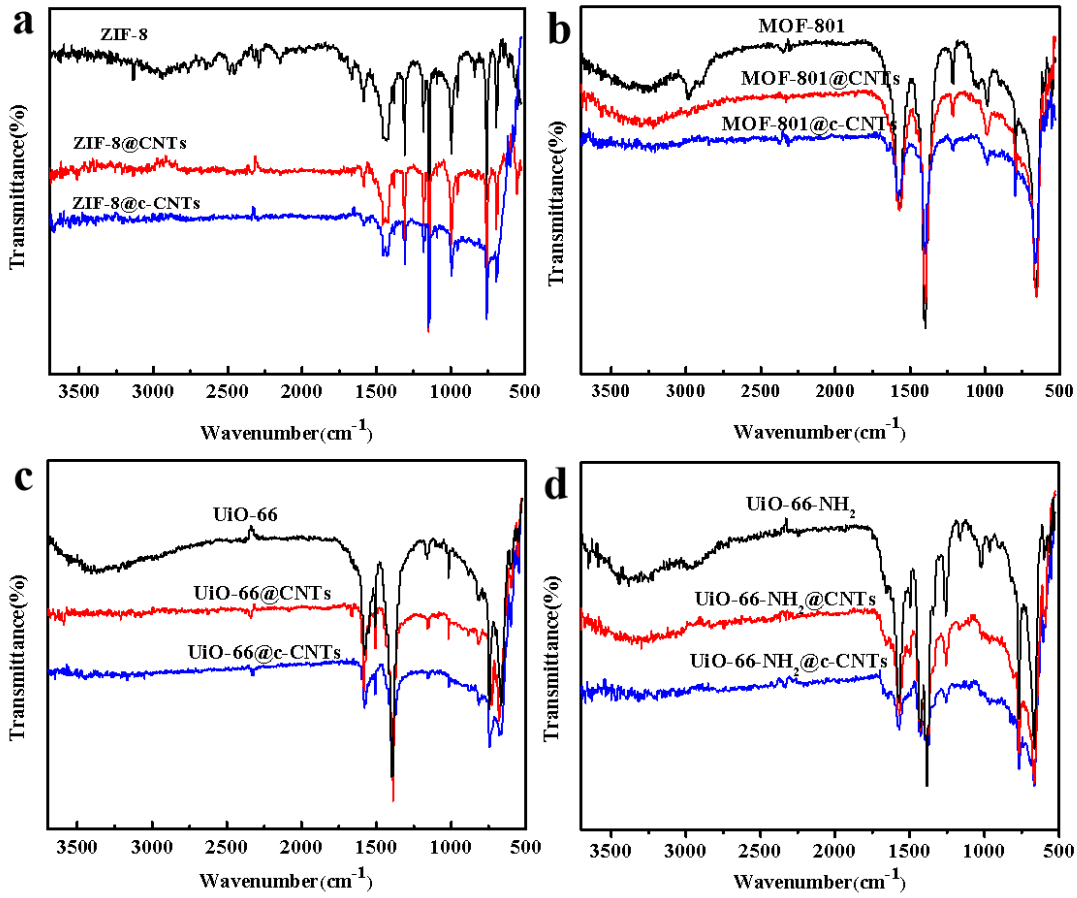

Fig. S5. FT-IR spectra of (a) ZIF-8, ZIF-8@CNTs and ZIF-8@c-CNTs, (b) MOF-801, MOF-801@CNTs and MOF-801@c-CNTs, (c) UiO-66, UiO-66@CNTs and UiO-66@c-CNTs, and (d) UiO-66-NH_2, UiO-66-NH_2@CNTs and UiO-66-NH_2@c-CNTs.

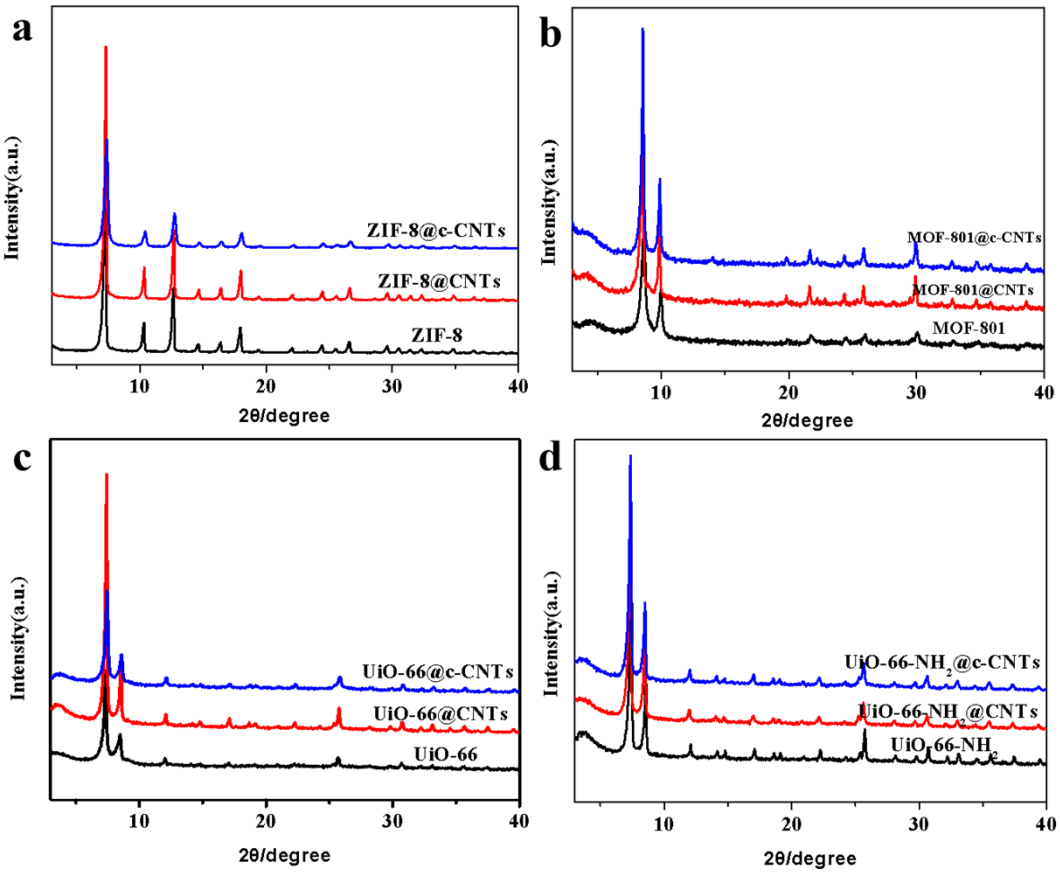

Fig. S6. PXRD patterns of (a) ZIF-8, ZIF-8@CNTs and ZIF-8@c-CNTs, (b) MOF-801, MOF-801@CNTs and MOF-801@c-CNTs, (c) UiO-66, UiO-66@CNTs and UiO-66@c-CNTs, and (d) UiO-66-NH_2, UiO-66-NH_2@CNTs and UiO-66-NH_2@c-CNTs.

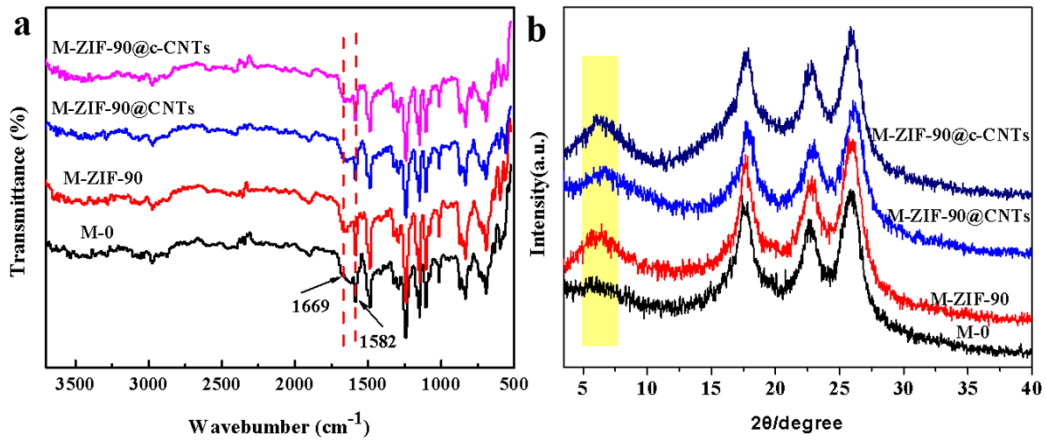

Fig. S7. (a) ATR-FTIR spectra and (b) XRD patterns of M-0 and modified nanofiltration membranes.

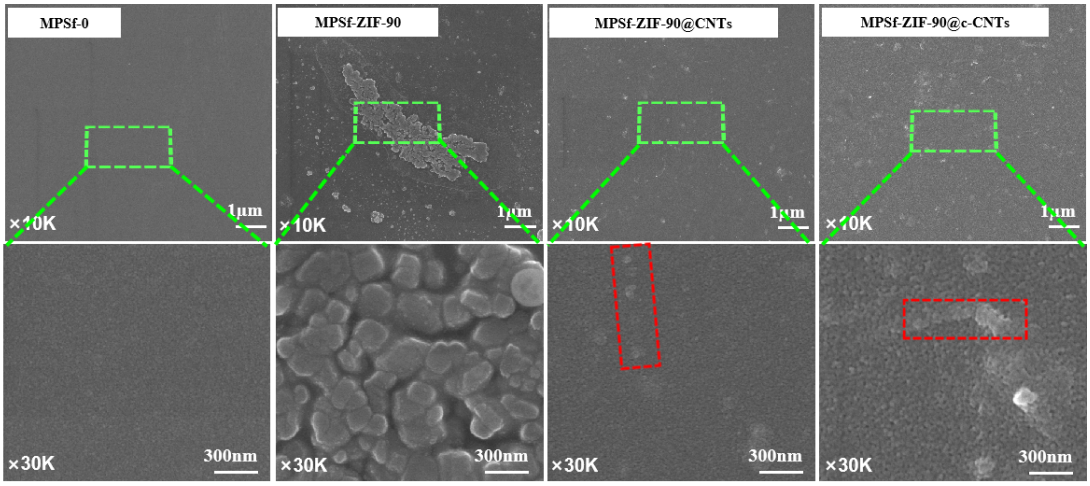

Fig. S8. The surface SEM of MPSf-0, MPSf-ZIF-90, MPSf-ZIF-90@CNTs and MPSf-ZIF-90@c-CNTs.

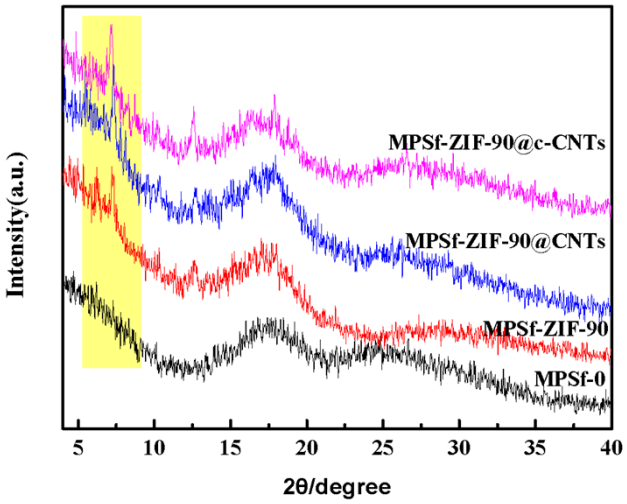

Fig. S9. XRD patterns of MPSf-0 and modified MMMs.

Table S1. Salt/dye separating ability and water permeability of nanofiltration membranes in our work and reported recently.

Membranes	Permeability (L·m-2·h-1·bar-1)	Dye rejection(%)		Salt rejection(%)		Ref.
MIL-53(Fe)/TMC	39.83	CR	99	NaCl	17.22	[S1]
Fe3O4@ZIF-8(0.3)	40.5	CR	99.6	NaCl	7.2	[S2]
PMIA/PVA/AGOs	17	CR	99.1	NaCl	4.2	[S3]
PA/ZIF-8-PSS	9.6	RhB	94.5	NaCl	~38	[S4]
ZrT-1-NH2-Lys/TMC	25.8	Chrome black T	99.2	NaCl	10.3	[S5]
CuTz-1/GO	40.2	CR	99.4	NaCl	0.3	[S6]
Cu-FeTCPP/TMC	80.7	CR	99.6	NaCl	3.8	[S7]
TA-Cu-TCPP/TMC	26.9	RhB	99.8	NaCl	6.6	[S8]
PAI/PRI-AlgFe3+	13.5	Methylene Blue	94.8	NaCl	19.9	[S9]
ZIF-90@CNT/PA	27.15	CR	99.55	NaCl	9.12	This work
ZIF-90@c-CNT/PA	27.80	CR	99.05	NaCl	8.08	This work

Table S2. CO_2/N_2 separation performances of theMPSf-ZIF-90@CNTs, MPSf-ZIF-90@c-CNTs and other MMMs.

Filler	Polymer matrix	Permeability of CO_2 (Barrer)	Selectivity of CO_2/N_2	Testing conditions	Ref.
ZIF-90	6FPA-DAM	720	27	25℃, 2 bar	[S10]
ZIF-90	Matrimid	6	/	25℃, 0.2 bar	[S11]
ZIF-90	Ultem	2.4	/	35℃, 4.6 bar	[S12]
ZIF-90	Polyetherimide	~120	~7.5	35℃, 1500 Torr	[S13]
ZIF-90@C_3N_4	Pebax	110.5	84.4	25℃, 2 bar	[S14]
ZIF-8-90	Pebax	306.1	51	35℃	[S15]
[BMIM][Br]@MIL-101(Cr)	Matrimid	7.9	19.7	30℃, 0.7 bar	[S16]
SSMMP/Im(nBu)I	PSf	9.62	24.3	25℃, 4 bar	[S17]
ZIF-67	PSf	12.6	36.9	25℃–35℃, 10 bar	[S18]
ZIF-67/[APTMS][Ac]	PSf	11.1	45.0	25℃–35℃, 10 bar	[S18]
ZIF-90@CNT	PSf	9.5	41.1	25℃, 2 bar	Thiswork
ZIF-90@c-CNT	PSf	9.98	42.27	25℃, 2 bar	This work

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