

Supplementary Figures and Tables

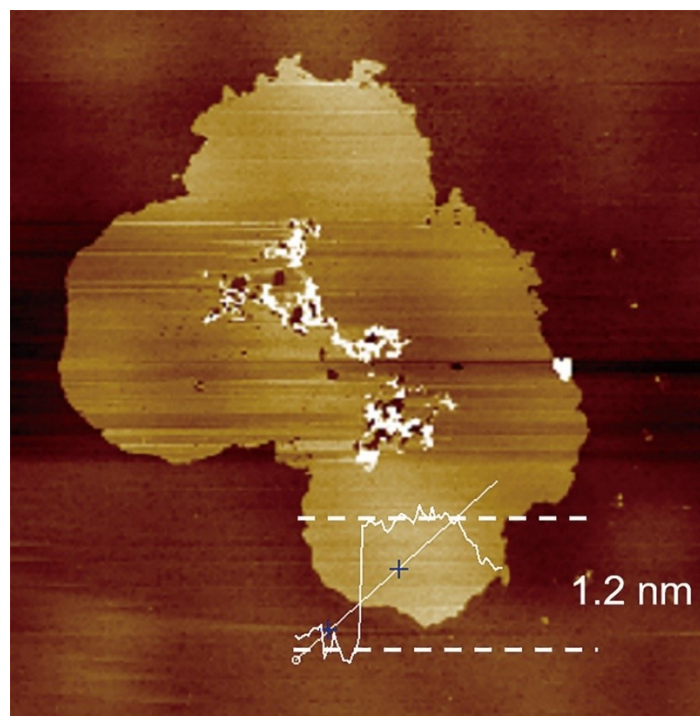


Figure S1 The AFM image of MoS₂ single layer.

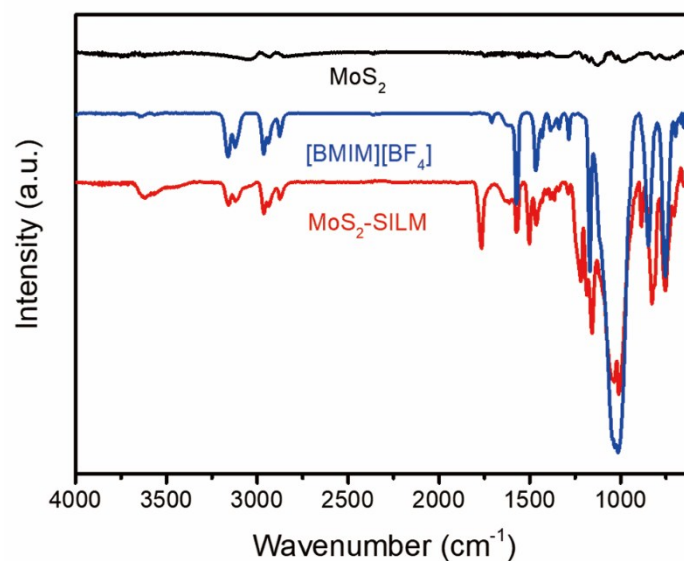


Figure S2 The FTIR spectra of MoS₂, [BMIM][BF₄] and MoS₂-SILM. Detail data are shown in Table S1.

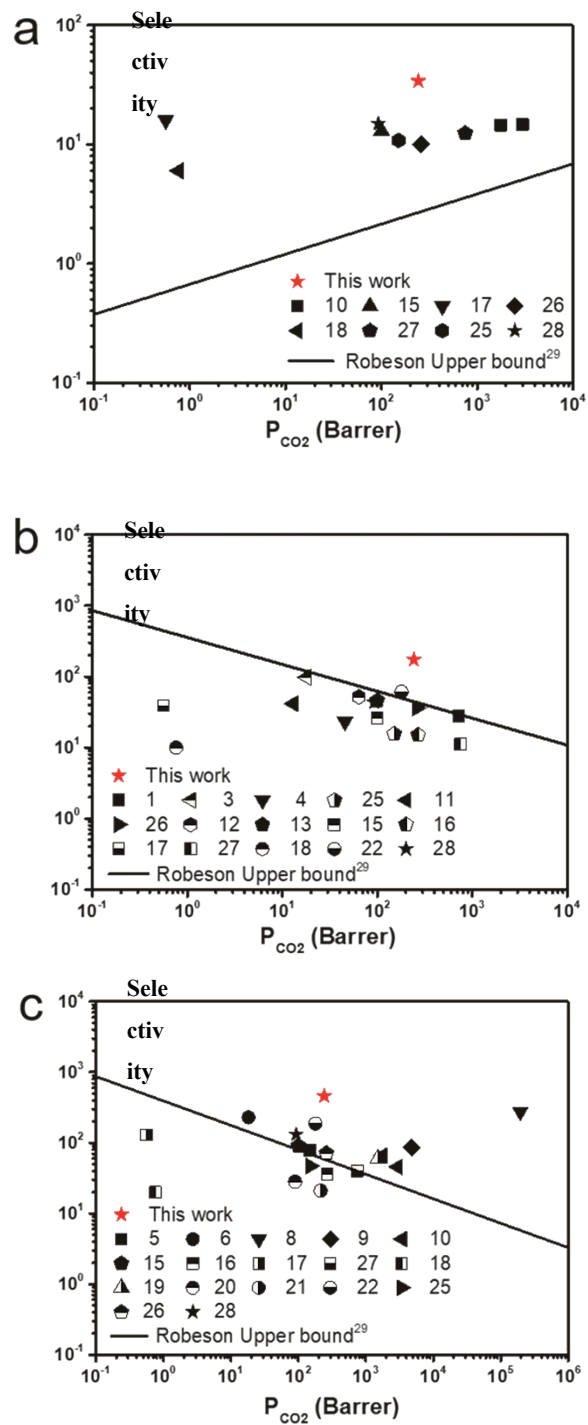


Figure S3 The separation performance (selectivity vs. permeability) of MoS₂-SILM under EEF compared with other membranes. (a) CO₂/H₂, (b) CO₂/CH₄ and (c) CO₂/N₂. The detail data is shown in Table S2 and from the Supplementary references.

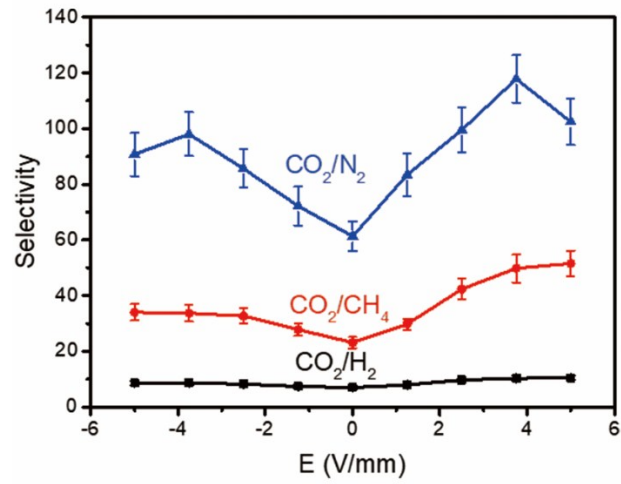


Figure S4 The mixed gas selectivity of MoS₂-SILM under EEF.

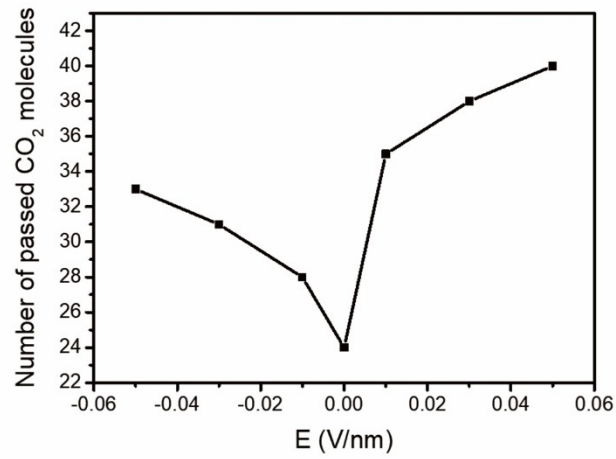


Figure S5 Molecular simulated CO₂ permeance under EEF.

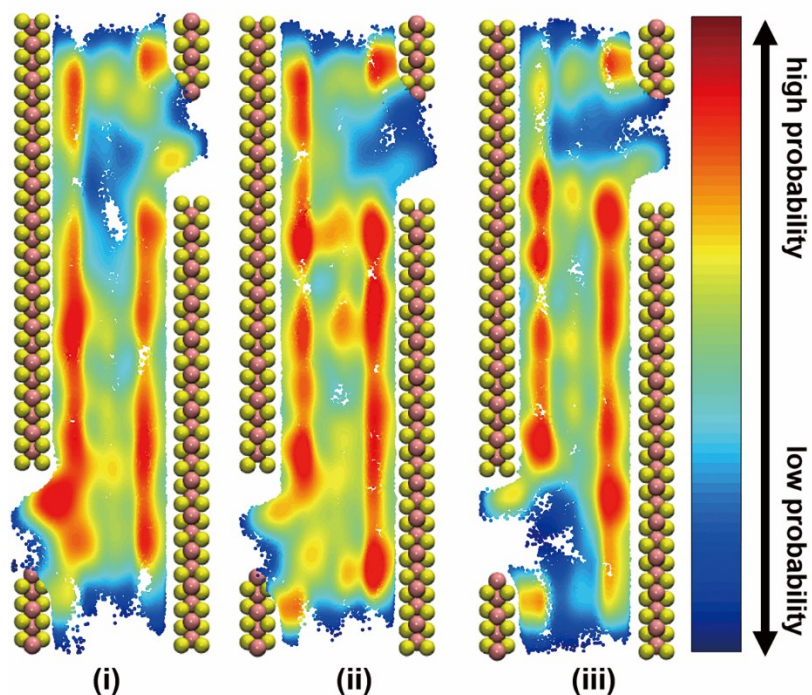


Figure S6 The density map of [BMIM]⁺ inside the nanochannel under EEF of (i) $E = -0.03$ V/nm, (ii) $E = 0$ V/nm, (iii) $E = 0.03$ V/nm. Blue indicates the region that low probability to find [BMIM]⁺, while red region indicates the high probability of finding [BMIM]⁺.

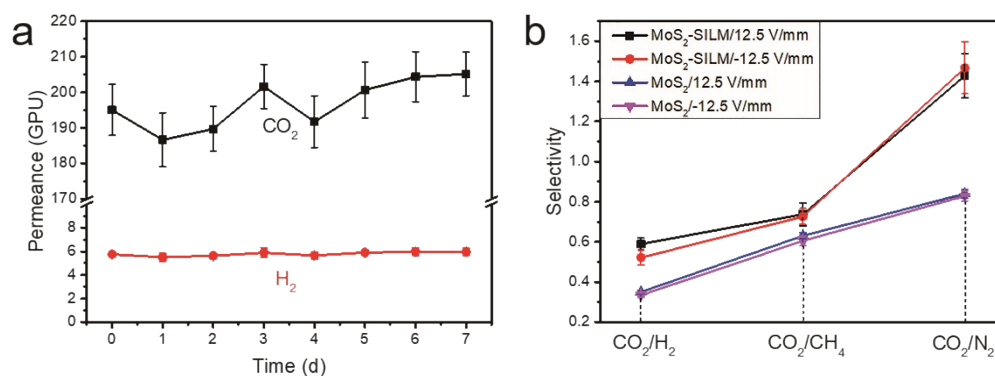


Figure S7 The durability of MoS₂-SILM. (a) is the permeance of CO₂ and H₂ tested under 3.75 V/mm for a week; (b) is the gas selectivity of MoS₂-SILM and MoS₂ under 12.5 V/mm and -12.5 V/mm after 1 day.

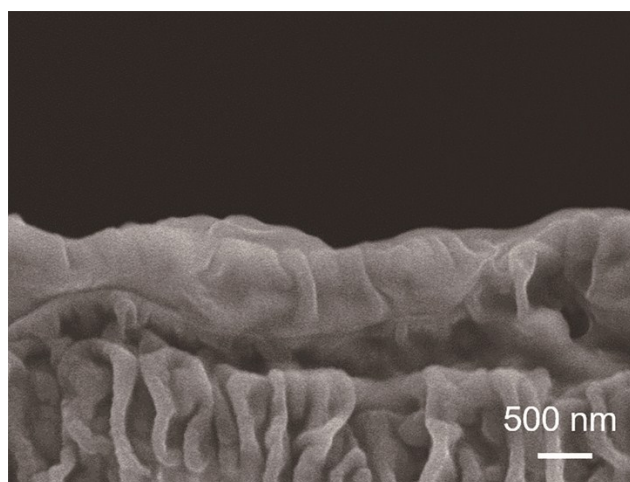


Figure S8 Cross-section SEM image of MoS₂-SILM after testing under 12.5 and -12.5 V/mm.

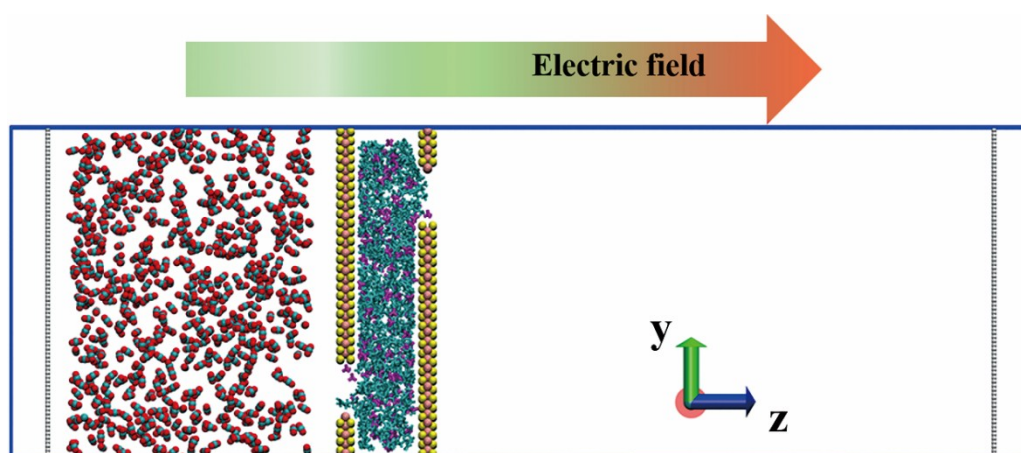


Fig. S9 A bichamber system for simulations of CO₂ permeance through the MoS₂-SILM. White, cyan, and red balls denote helium, carbon, and oxygen, respectively. Green-cyan and magenta stick represent [BMIM]⁺ and [BF₄]⁻ ions.

Table S1 The data of FTIR peaks in Fig. S2.

| [BMIM][BF₄] (cm⁻¹) | MoS₂-SILM (cm⁻¹) | Shifts (cm⁻¹) | Assignment |
|---|---|---------------------------------|---------------------------|
| 3160.81 | 3158.88 | -1.93 | vC4-H, vC5-H |
| 3122.24 | 3118.38 | -3.86 | vC2-H |
| 2939.03 | 2937.10 | -1.93 | vCH ₂ |
| 2877.32 | 2875.39 | -1.93 | vCH ₂ |
| 1708.65 | 1764.57 | 55.92 | |
| 1621.86 | 1614.15 | -7.71 | |
| 1465.66 | 1463.73 | -1.93 | vCH ₃ |
| 1384.66 | 1386.59 | 1.93 | wCH ₂ |
| 1373.09 | 1363.45 | -9.64 | vC2N1C5, wCH ₂ |
| 1338.38 | 1340.31 | 1.93 | vN-Bu, N-Me |
| 1284.38 | 1288.24 | 3.86 | vBF ₄ |
| 1253.52 | | | tCH ₂ , rC2-H |
| | 1218.81 | | vBF ₄ |
| 1168.67 | 1157.10 | -11.57 | vN-Bu, N-Me |
| 1031.74 | 1035.60 | 3.86 | vBF |
| 1014.39 | 1010.53 | -3.86 | vBF ₄ |
| 908.32 | 919.89 | 11.57 | |
| 846.61 | 827.32 | -19.29 | rC4-H, rC5-H |
| 808.04 | 813.82 | 5.78 | vC-C-C |
| 696.19 | 705.83 | 9.64 | vN-Bu, N-Me |

Table S2 The data of gas separation performance of different membranes in Figure 3.

| Number | Membrane | P_{CO_2} (Barrer) | P_{CO_2} (GPU) | CO_2/H_2 | CO_2/CH_4 | CO_2/N_2 | Ref |
|--------|---|------------------------|---------------------|------------|-------------|------------|--------------|
| 1 | ZIF-90/6FDADAM | 720 | 12 | | 28 | | 1 |
| 2 | PSF | - | 19.86 | | 23.12 | | 2 |
| 3 | DDR | 17.9 | 35.8 | | 98 | | 3 |
| 4 | MIL-53-NH2/organosilica | 45.6 | 430 | | 23.2 | | 4 |
| 5 | MMP-3/mPSf | 150 | 3000 | | | 78 | 5 |
| 6 | PAMAM dendrimer | 18.3 | 61 | | | 230 | 6 |
| 7 | DNMDAm–DGBAmE– TMC/PDMS/PS | - | 1601 | | | 138 | 7 |
| 8 | PVAm–PIP/PS | 195000 | 6500 | | | 277 | 8 |
| 9 | DAmBS–DGBAmE– TMC/PDMS/PS | 4781 | 5831 | | | 86 | 9 |
| 10 | SIPN:PEGMEA:PEGDA:PE GDME | 2980 | 11.9-14.9 | 14.7 | | 45.7 | 10 |
| 10 | SIPN:PEGDA:PEGDME | 1767 | 7.1-8.8 | 14.5 | | 65.9 | 10 |
| 11 | Matrimid® ZIF-8 | 12.96 | 180 | | 41.5 | | 11 |
| 12 | 6FDA-DAT/Ni2(dodbc) | 63.9 | 0.91-1.6 | | 52 | | 12 |
| 13 | 6FDA-ODA UiO-66 | 100 | 1.26-2.52 | | 46.1 | | 13 |
| 14 | Pebax/[emim][BF4]/GO | - | 981 | | | 44 | 14 |
| 15 | Pebax/GO | 100 | 20 | 13 | 26 | 91 | 15 |
| 16 | Pebax/[emim][BF4] | 269 | 306 | | 15 | 36 | 16 |
| 17 | PAN/[emim][Ac]GO/PTMSP | 0.56 | 37 | 16 | 39 | 130 | 17 |
| 18 | GO | 0.76 | 110 | 6 | 10 | 20 | 18 |
| 19 | 0.1water-[BMIM][BF ₄]-PES | 1518 | 13.8 | | | 60 | 19 |
| 20 | [BMIM][BF ₄]/AgO | 89.7 | 14.1 | | - | 28.2 | 20 |
| 21 | [BMIM][BF ₄]/CuO | 214.84 | 52.4 | | - | 21 | 21 |
| 22 | Pebax/0.5%Ag/50%[BMIM][BF ₄] | 180 | 3.2 | | 61 | 187.5 | 22 |
| 23 | [BMIM][BF ₄]/LiBF ₄ | - | 13.36 | | 8.25 | 8.4 | 23 |
| 24 | [BMIM][BF ₄]/cyanuric chloride | - | 19.2 | | 10.7 | 11 | 24 |
| 25 | Pebax®/PEG | 151 | 2.52 | 10.8 | 15.5 | 47 | 25 |
| 26 | IL-Pebax 165/ZIF-8 | 261 | 20 | 10 | 36 | 71 | 26 |
| 27 | PEO-PBT/PEG-DBE (PAN- PDMS) | 750 | 15 | 12.4 | 11.2 | 40 | 27 |
| 28 | MoS2-[BMIM][BF ₄] (1940 nm) | 92.9 | 47.88 | 14.95 | 43.52 | 131.42 | 28 |
| 29 | MoS2-[BMIM][BF ₄] (1120 nm) under positive EEF | 243 | 195 | 34 | 174 | 462 | This work |

Table S3 Lennard-Jones parameters and atom charge of MoS₂ used in this work.^{30,31}

| | CO ₂ | | MoS ₂ | |
|-----------------------|-----------------|---------|------------------|---------|
| | C | O | Mo | S |
| ϵ (Kcal/mol) | 0.0559 | 0.1600 | 0.0135 | 0.4612 |
| σ (Å) | 2.7570 | 2.5650 | 4.2000 | 3.1300 |
| q (e) | 0.6512 | -0.3256 | 0.7600 | -0.3800 |

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