Electronic supplementary information for:

Effect of pore density on gas permeation through nanoporous graphene membranes

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1. The 4N4H pore structure

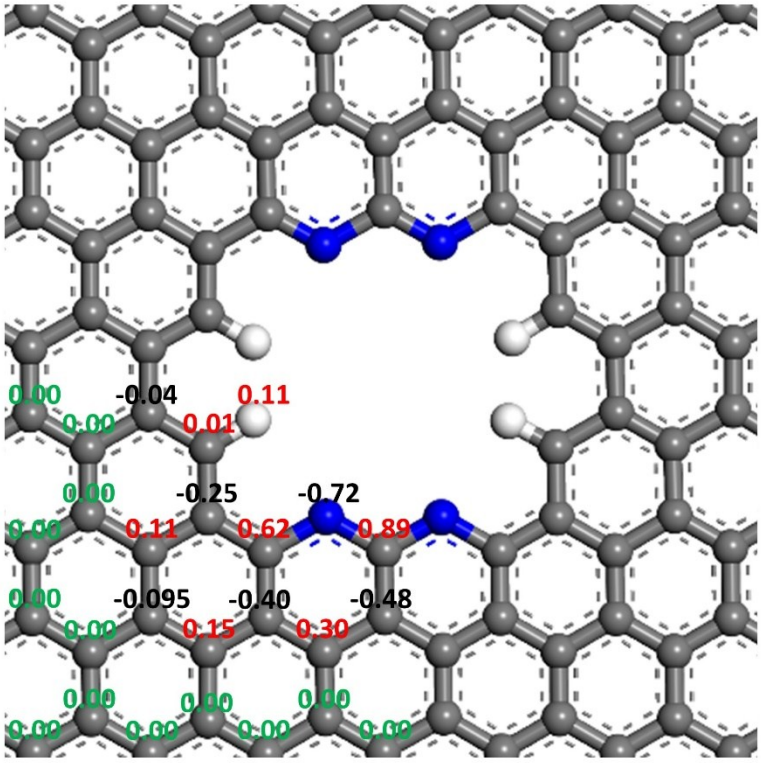

Fig. S1. The 4N4H pore structure (carbon, grey; nitrogen, blue; hydrogen, white). The partial atomic charges used in our simulations are provided next to each atom. Due to the D_{2h} symmetry of the pore structure, only the atoms in the lower left part are labelled with their atomic charges.

2. Force field parameters

Table S1. Lennard-Jones parameters for the porous graphene (atomic charges are in Fig. S1).

Porous graphene		
	ε (K)	σ (Å)
C	28.0	3.40
N	85.6	3.25
H	15.1	2.42
bonds	length (Å)	
C-C	1.42	
C-N	1.42	
C-H	1.10	

Table S2. Force field parameters for gas molecules.

CO_2			
	ε (K)	σ (Å)	q (e)
C	28.129	2.757	0.6512
O	80.507	3.033	-0.3256
bonds	length (Å)		
C-O	1.149		
He			
	ε (K)	σ (Å)	q (e)
He	10.956	2.641	0

3. Permeate side pressure, feed side pressure, and their difference at different pore densities

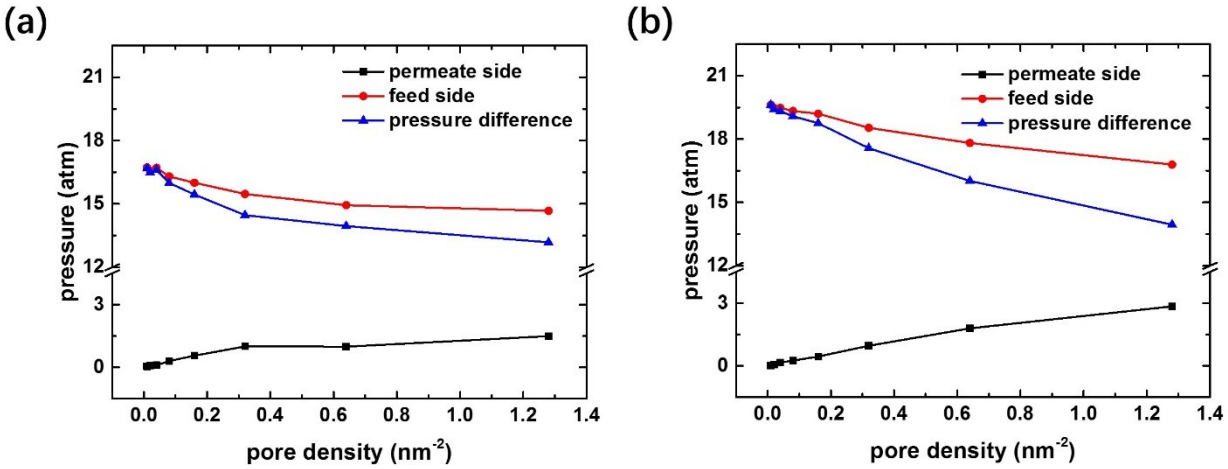

Fig. S2. Permeate side pressure, feed side pressure, and pressure difference vs. pore density for (a) CO_2 and (b) He. At 1 ns.

4. Exponential fitting

Instead of the linear fitting of the initial portion of the permeation number vs. time curves (Fig. 2 in the text), here we use an exponential decay model $y = a * (1 - e^{-\lambda t})$ to fit the whole curves (Fig. S3a,b). For each curve, the fitting yields a λ value; the half-time, $t_{1/2}$, is simply $ln(2)/\lambda$ and the permeation rate is $a/(2t_{1/2})$. Then total flux and flux per pore can be obtained (Fig. S3c,d). Compared with the linear fitting results (Fig. 3 in the text), one can see that the two methods give very similar results.

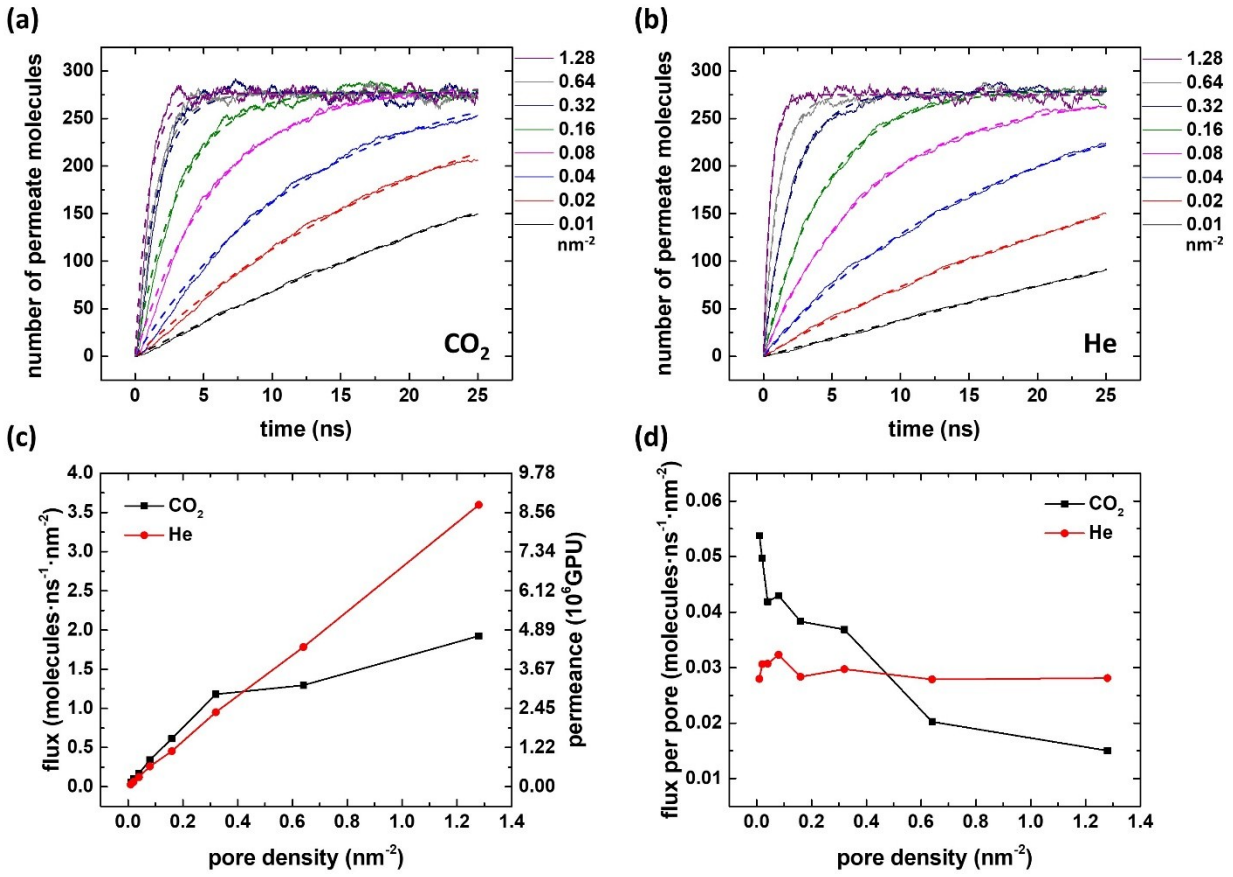

Fig. S3. The number of gas molecules permeating through the nanoporous graphene membrane with time for different pore densities (from 0.01 to 1.28 nm^{-2}): (a) CO_2; (b) He; Dashed lines represent the exponential decay model fitting. Flux vs pore density of graphene membranes for CO_2 and He permeation: (c) flux and permeance per unit membrane area; (d) flux per pore.

5. Flux for He molecule

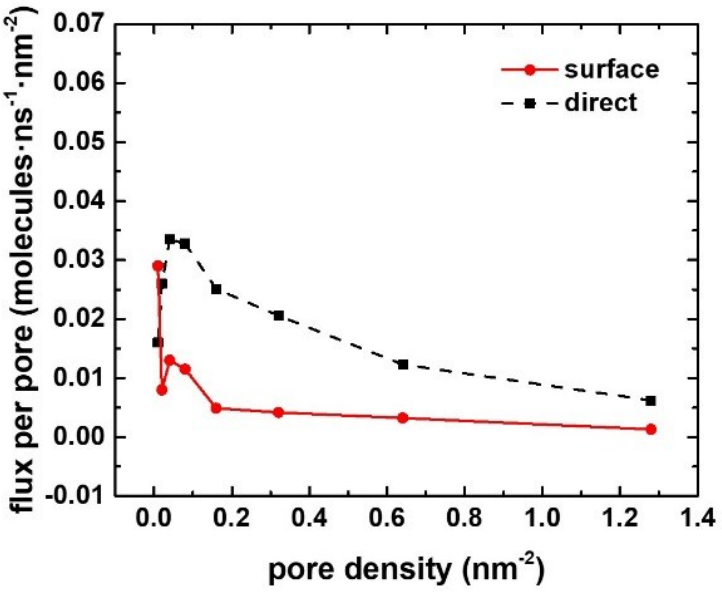

Fig. S4. The surface flux and the direct flux of He, across graphene membranes of different pore densities. The values are from the trajectory analysis of all gas molecules during the initial 1 ns.