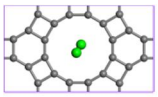
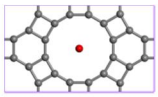
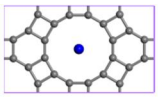
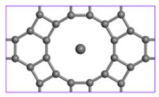
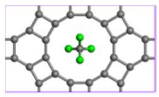
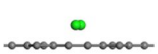
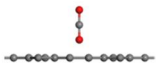
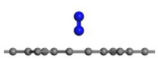
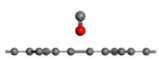
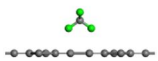
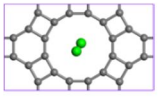
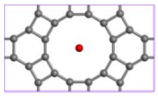
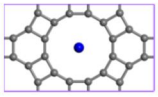
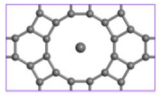
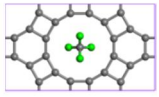
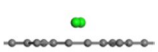
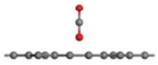
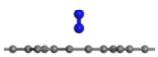
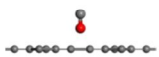
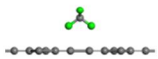
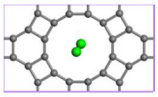
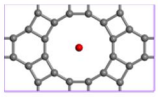
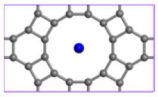
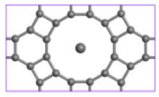
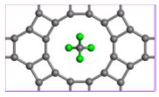
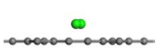
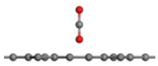
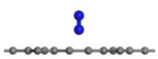
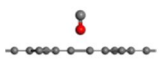
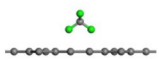
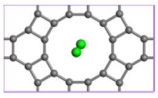
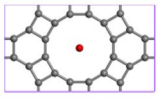
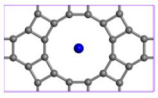
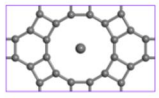
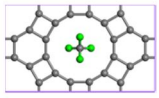
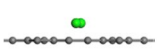
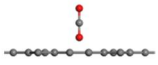
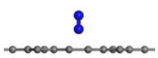
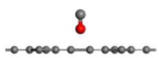
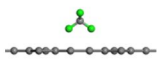
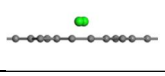
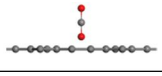
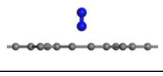
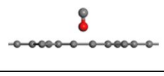
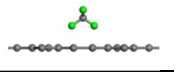
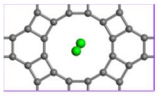
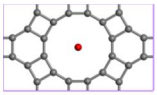
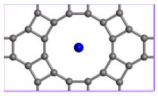
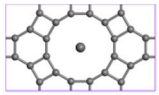
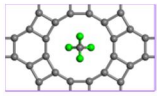
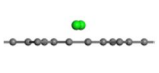
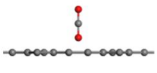
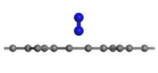
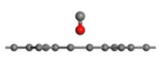
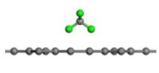
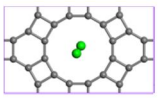
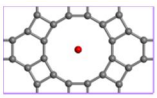
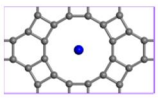
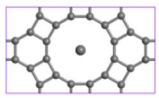
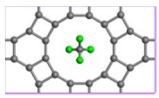
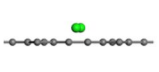
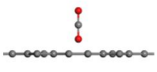
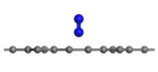
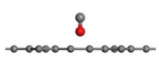
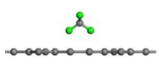
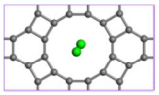
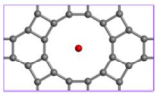
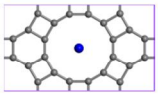
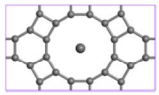
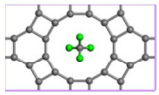
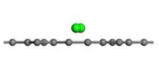
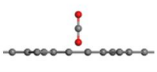
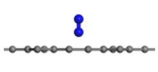
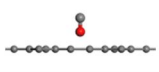
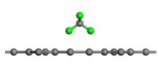
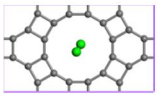
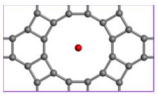
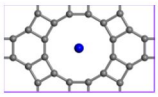
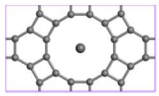
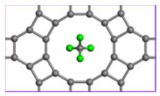
Table S1 The SS configurations with the adsorption heights (H_{ad}) and adsorption energies (E_{ad}) of gas molecules absorbed on the surface of strained graphenylene membrane. The gray, green, blue, and red spheres represent C, H, N, and O atom, respectively.

Strain Level	Property	H_2	CO_2	N_2	CO	CH_4
1%	Top View					
	Side View					
	H_{ad} (Å)	1.6	2.6	2.2	2.2	2.6
	E_{ad} (eV)	-0.20	-0.22	-0.25	-0.29	-0.25
2%	Top View					
	Side View					
	H_{ad} (Å)	1.6	2.6	2.2	2.2	2.6
	E_{ad} (eV)	-0.19	-0.22	-0.25	-0.29	-0.24
3%	Top View					
	Side View					
	H_{ad} (Å)	1.6	2.6	2.2	2.2	2.4
	E_{ad} (eV)	-0.19	-0.22	-0.25	-0.29	-0.24
4%	Top View					
	Side View					
	H_{ad} (Å)	1.4	2.4	2.2	2.2	2.4
	E_{ad} (eV)	-0.20	-0.23	-0.25	-0.29	-0.24
5%	Top View					
	Side View					
	H_{ad} (Å)	1.4	2.4	2.0	2.2	2.4
	E_{ad} (eV)	-0.20	-0.23	-0.25	-0.29	-0.24
6%	Top View					

	Side View					
	H_{ad} (Å)	1.4	2.2	2.0	2.0	2.4
	E_{ad} (eV)	-0.20	-0.24	-0.25	-0.29	-0.24
7%	Top View					
	Side View					
	H_{ad} (Å)	1.4	2.2	2.0	2.0	2.4
	E_{ad} (eV)	-0.19	-0.24	-0.24	-0.29	-0.24
8%	Top View					
	Side View					
	H_{ad} (Å)	1.2	2.2	2.0	2.0	2.4
	E_{ad} (eV)	-0.19	-0.24	-0.24	-0.28	-0.24
9%	Top View					
	Side View					
	H_{ad} (Å)	1.2	2.0	2.0	2.0	2.4
	E_{ad} (eV)	-0.19	-0.25	-0.24	-0.28	-0.23
10%	Top View					
	Side View					
	H_{ad} (Å)	1.2	2.0	2.0	2.0	2.2
	E_{ad} (eV)	-0.19	-0.25	-0.24	-0.28	-0.23
11%	Top View					
	Side View					
	H_{ad} (Å)	1.0	1.8	2.0	2.0	2.2
	E_{ad} (eV)	-0.19	-0.26	-0.24	-0.27	-0.23
12%	Top View					

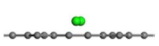
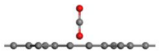
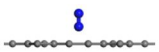
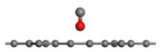
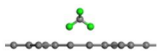
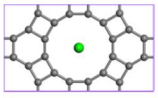
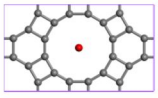
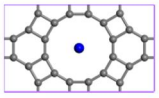
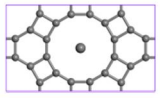
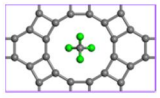
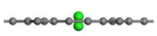
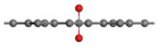
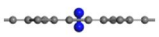
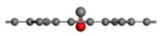
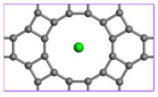
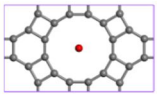
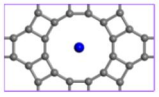
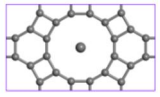
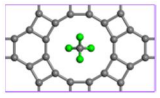
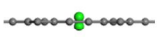
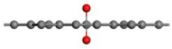
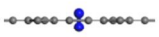
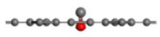
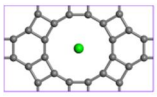
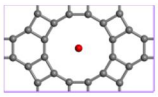
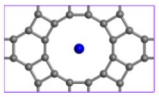
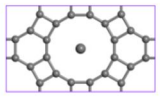
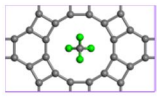
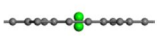
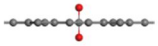
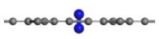
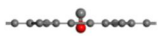
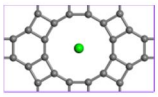
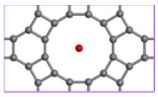
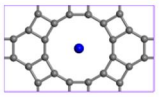
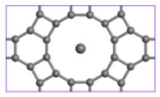
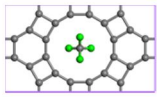
	Side View					
	H_{ad} (Å)	1.0	1.8	2.0	2.0	2.2
	E_{ad} (eV)	-0.19	-0.26	-0.23	-0.27	-0.23

Table S2 The TS configurations of gas molecules penetrating through strained graphenylene membrane. The gray, green, blue, and red spheres represent C, H, N, and O atom, respectively.

Strain level	**Property**	**H_2**	**CO_2**	**N_2**	**CO**	**CH_4**
1%	Top View					
	Side View					
2%	Top View					
	Side View					
3%	Top View					
	Side View					
4%	Top View					
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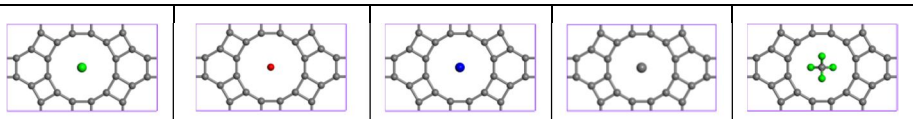
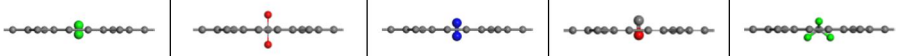
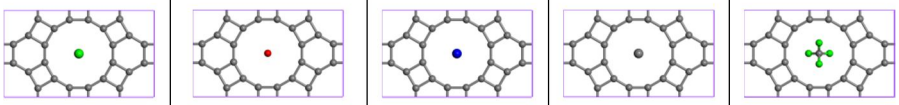
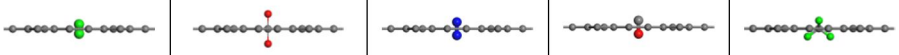
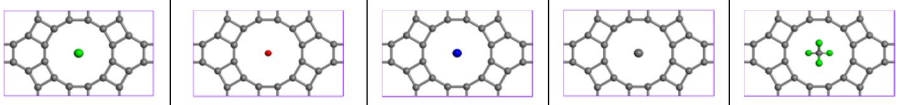
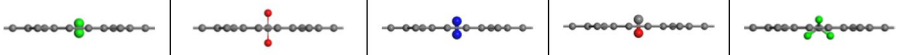
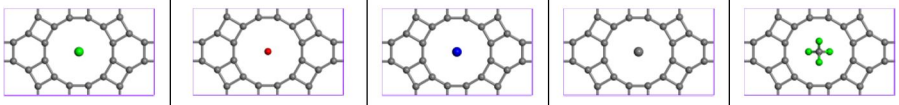
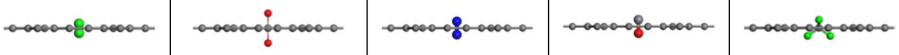
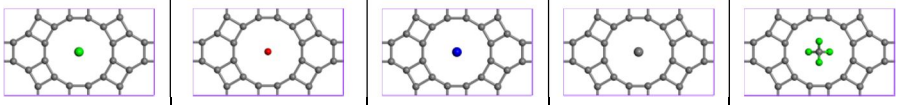
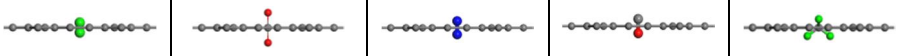
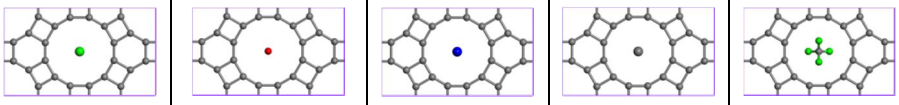
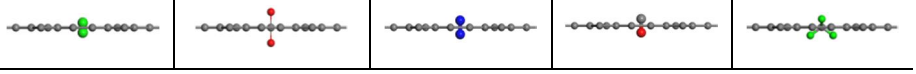
7%	Top View					
	Side View					
8%	Top View					
	Side View					
9%	Top View					
	Side View					
10%	Top View					
	Side View					
11%	Top View					
	Side View					
12%	Top View					
	Side View					

Classical Molecular Dynamics (MD) Simulation

The H_2 permeance of 3.04% strained graphenylene monolayer is investigated by MD simulation. The simulation model is shown as **Fig. S1**. An approximately square sheet of 3.04% strained graphenylene (97.72 Å×96.72 Å) serves as a mono-atomistic membrane and the membrane separates a gaseous mix of H_2, CO, CO_2 and CH_4 (consisting of 400 H_2 molecules, 200 CO molecules, 200 CO_2 molecules and 200 CH_4 molecules, a molar ratio of 2 : 1: 1 : 1). At the bottom of the permeate phase, a wall composed of 1904 helium atoms is used to impede gas molecules passing to the vacuum phase due to the periodicity along the Z axis.[1] A condensed-phase optimized

molecular potential for atomistic simulation studies (COMPASS) is used for describing the interatomic interactions.[2] The simulation is subject to an NVT ensemble, carried out at 300 K. The temperature of the system is controlled by the Andersen thermostat method, with a fixed time step of 1 fs. Data was collected every 5 ps, and full-precision trajectory was then recorded. Van der Waals interaction and Ewald electrostatic interaction are applied with a cutoff distance of 9.5 Å. Periodic boundary conditions are applied in the *x* and *y* directions. During the simulation, the 3.04% strained graphenylene and the wall composed of 1904 helium atoms are fixed. The MD simulation is carried out using Discover codes embedded in Materials Studio software.

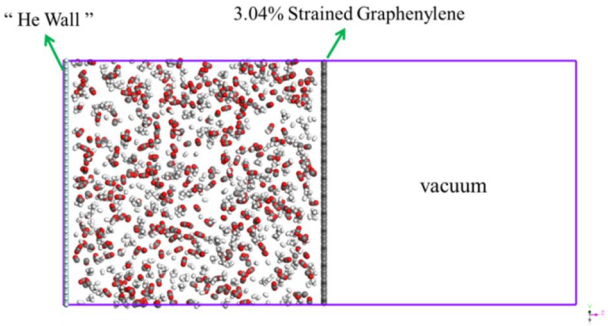


Fig. S1 Simulation model in our study. The gray, white, red, and turquoise blue spheres represent C, H, N, and He atom, respectively.

A molecule moving from the permeate phase to the vacuum phase is termed a permeation event.[3-4] Through analysis of molecules trajectories, the number of permeated H_2 molecules versus time can be obtained, as shown in **Fig. S2**. Based on the definition of flux and permeance, a theoretical model of the time-dependent number of the permeated H_2 molecules is deduced.[3-4] The relationship between flux J (mol s^{-1}) and permeance S (mol s^{-1} m^{-2} Pa^{-1}) is defined as

$$J = \frac{1}{N_A}\frac{\mathrm{dN}}{\mathrm{d}\tau} = A_g \cdot \Delta P \cdot S \tag{1}$$

Where A_g is the area of 3.04% strained graphenylene used in our simulations (A_g=9.45 $\times 10^{-17}$ m^2), $\triangle P$ is the pressure difference between the permeate phase and vacuum phase, N is the number of permeated H_2 molecules, τ is the time and N_A is the Avogadro constant. The pressure difference $\triangle P$ depends on the permeated H_2 molecules N. The initial partial pressure of H_2 is 1.78 MPa, and we assume the wall composed of helium atoms do not adsorb H_2 molecules, the $\triangle P$ can be defined as

$$\Delta P = \frac{400 - N_a - 2\mathrm{N}}{400} \times 1.78 \times 10^6 \text{ Pa} \tag{2}$$

where N_a is the average number of the H_2 molecules adsorbed on the adsorption layer.[3-4] By integrating eqn (1), it can obtain

$$N = (200 - \frac{N_a}{2}) \times (1 - e^{-5.06\times10^{11} S\tau}) \quad (3)$$

in which B=5.06×10^{11} S is the exponent of time decay. Then it applies eqn (3) to fit the curves given by the number of permeated H_2 molecules versus time and obtain the H_2 permeance. As shown in **Fig. S2**, the fitted parameter B=5.06×10^{11} S=8.89×10^{8}, and so the H_2 permeance of 3.04% strained graphenylene is 1.76×10^{-3} mol s^{-1} m^{-2} Pa^{-1}.

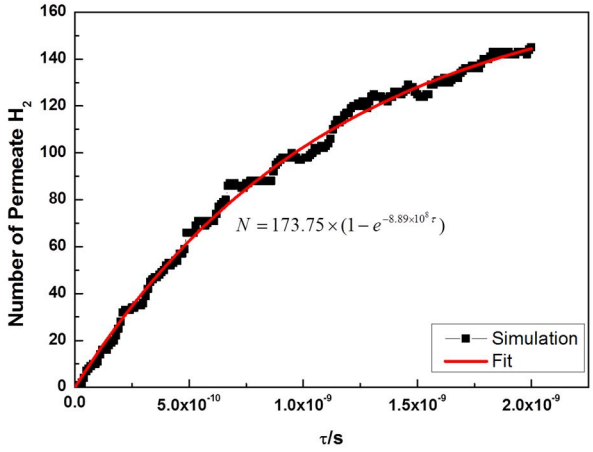


Fig. S2 the number of permeated H_2 molecules versus time.

Gas permeance calculated by kinetic theory represents the permeance of effective (permeable) area of a membrane while gas permeance obtained by MD simulation reflects the permeance of the whole membrane. In order to compare with the gas permeance obtained by kinetic theory, the gas permeance calculated by MD simulation must multiply a geometrical factor (η).[5] The η is defined as $\eta = A_m/A_p$, which A_m represents the area of a membrane, and A_p represents the effective (permeable) area of a membrane, and the estimated η value in this work is about 10. After considering the η, the H_2 permeance calculated by MD simulation is 1.76×10^{-2} mol s^{-1} m^{-2} Pa^{-1}, which is comparable with the H_2 permeance obtained by kinetic theory.

Reference

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