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Electronic supplementary information (ESI)

Theoretically Designed Two–Dimensional γ-C_4O as Effective Gas Separation Membrane for Hydrogen Purification

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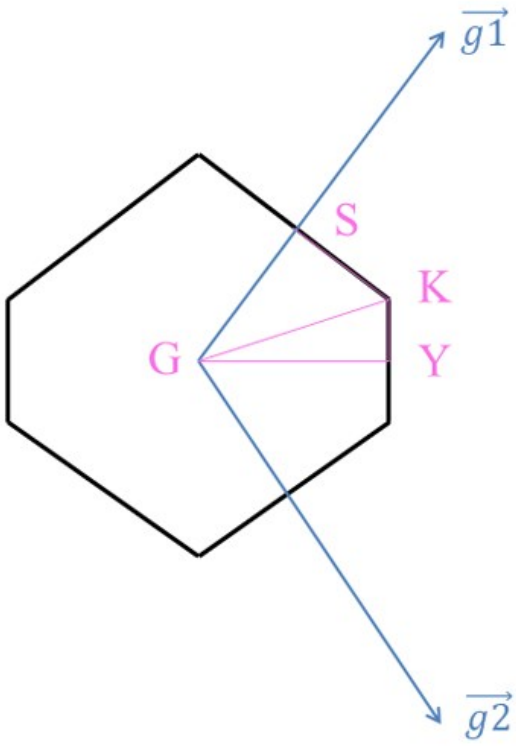

Fig. S1 Schematic illustration of the first Brillouin zone of γ-C_4X

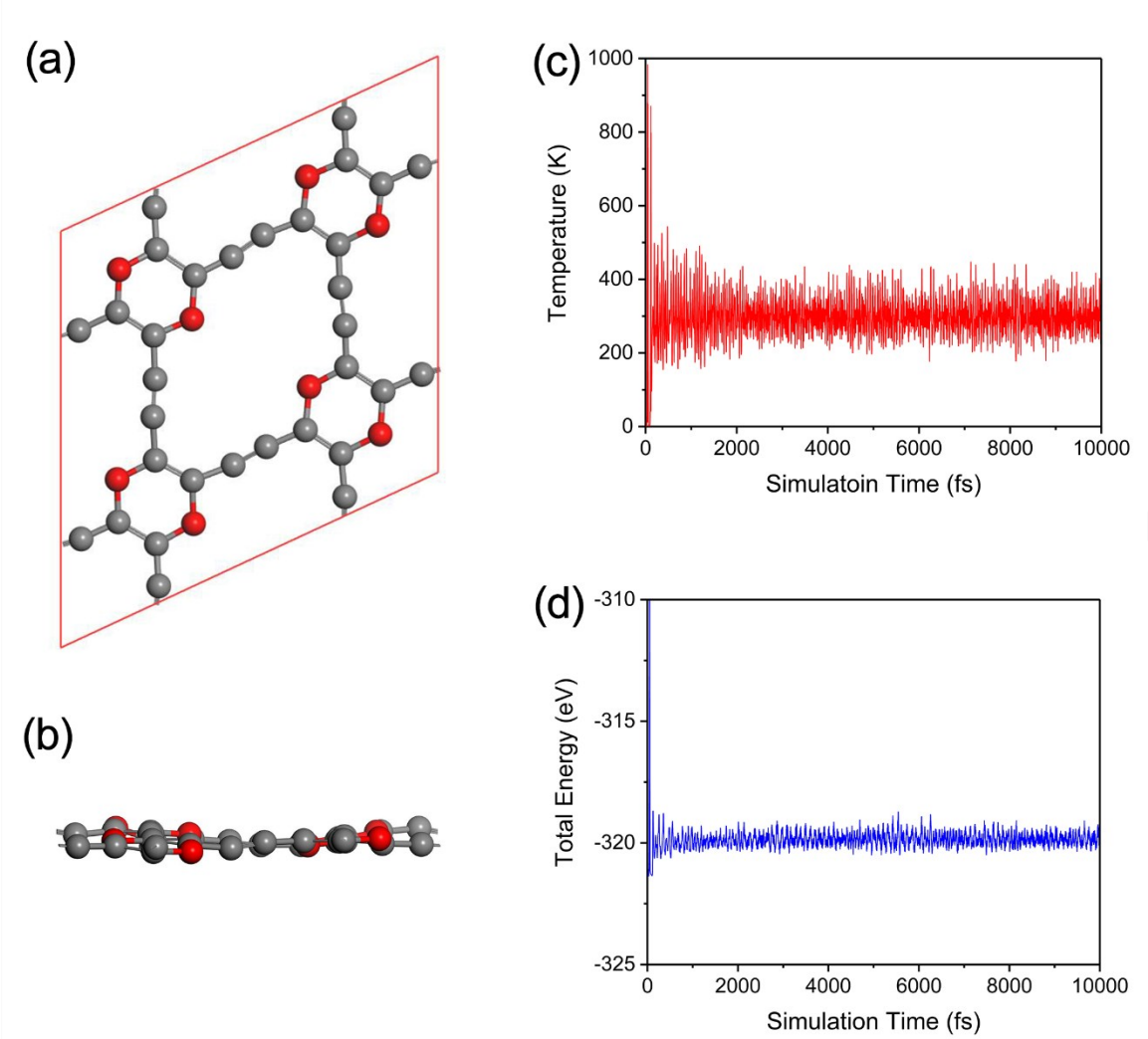

Fig. S2 (a) Top view and (b) side view of γ-C_4O after 10 ps FPMD simulation at 300K, and (c) temperature and (d) total energy changes during the simulation. Grey and red balls stand for C and O atoms.

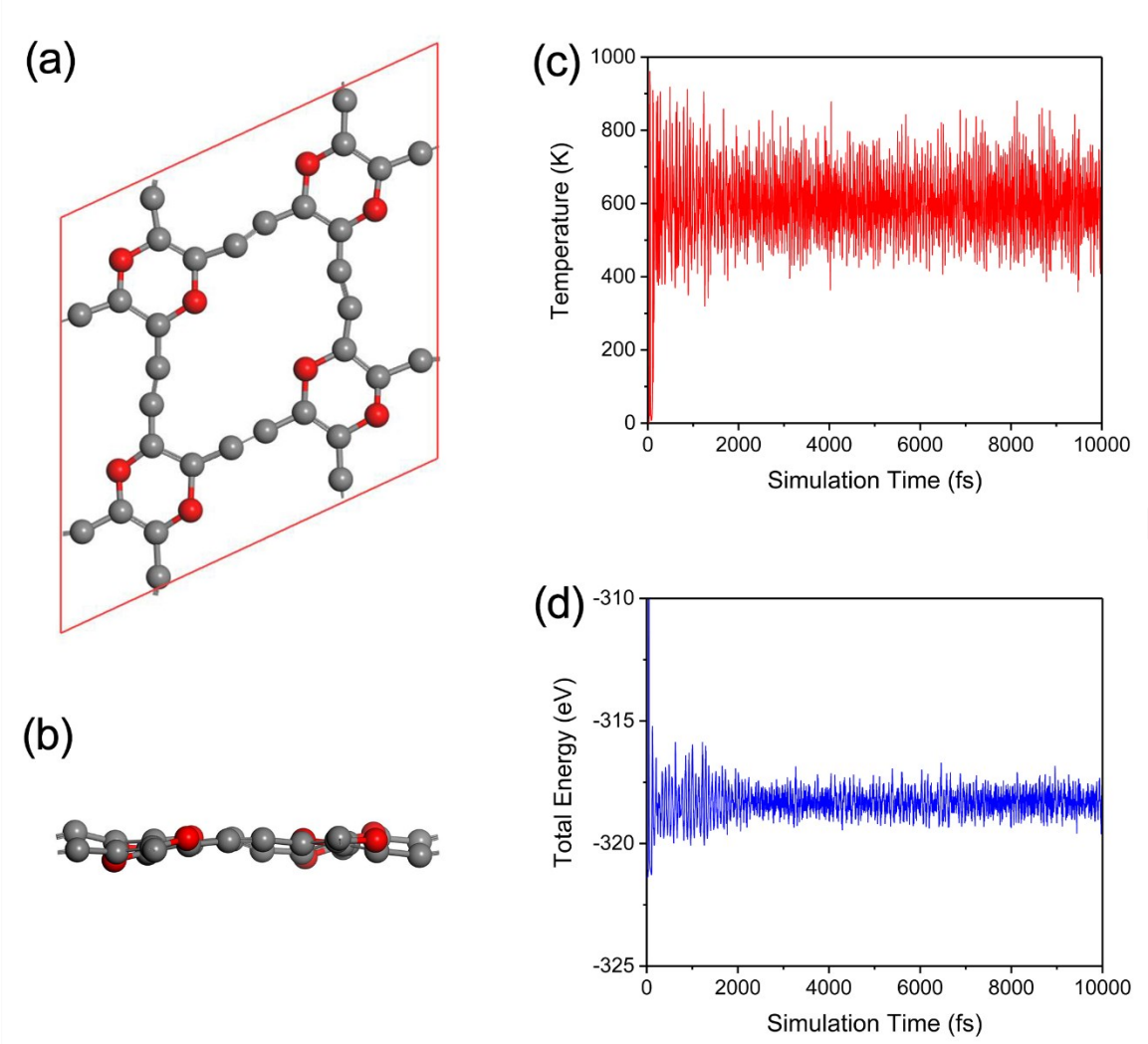

Fig. S3 (a) Top view and (b) side view of γ-C_4O after 10 ps FPMD simulation at 600K, and (c) temperature and (d) energy changes during the simulation. Grey and red balls stand for C and O atoms.

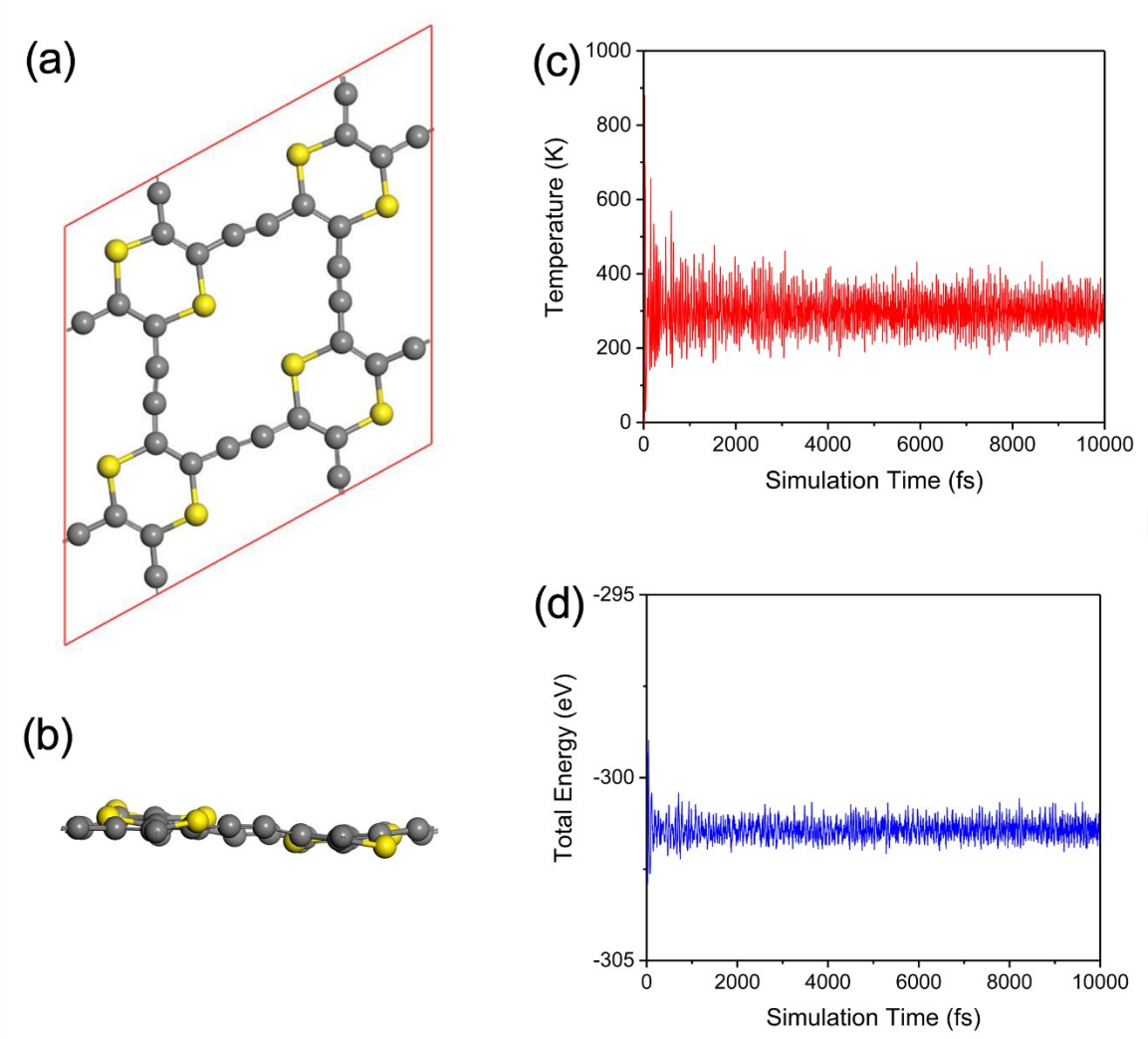

Fig. S4 (a) Top view and (b) side view of γ-C_4S after 10 ps FPMD simulation at 300K, and (c) temperature and (d) energy changes during the simulation. Grey and yellow balls stand for C and S atoms.

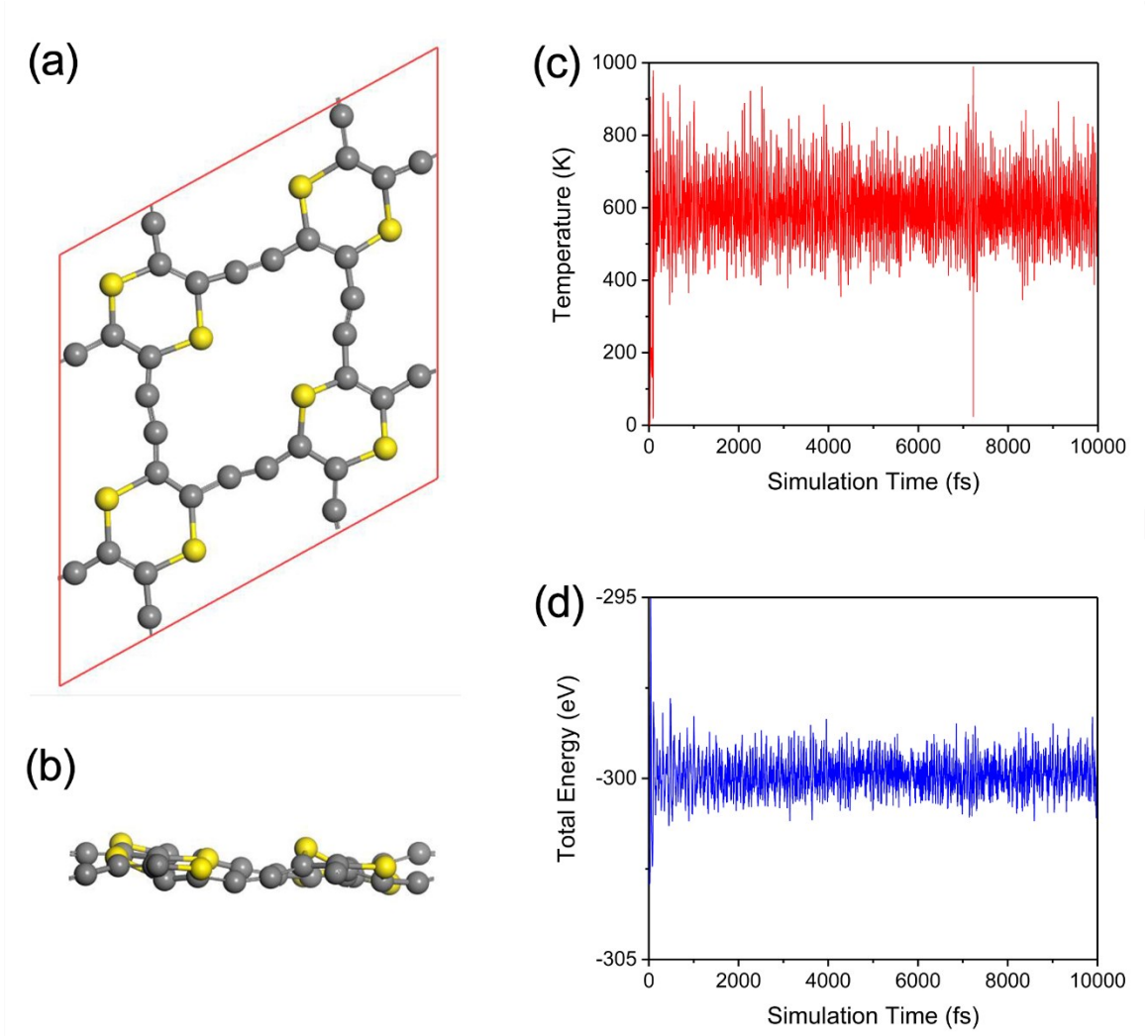

Fig. S5 (a) Top view and (b) side view of γ-C_4S after 10 ps FPMD simulation at 600K, and (c) temperature and (d) energy changes during the simulation. Grey and yellow balls stand for C and S atoms.

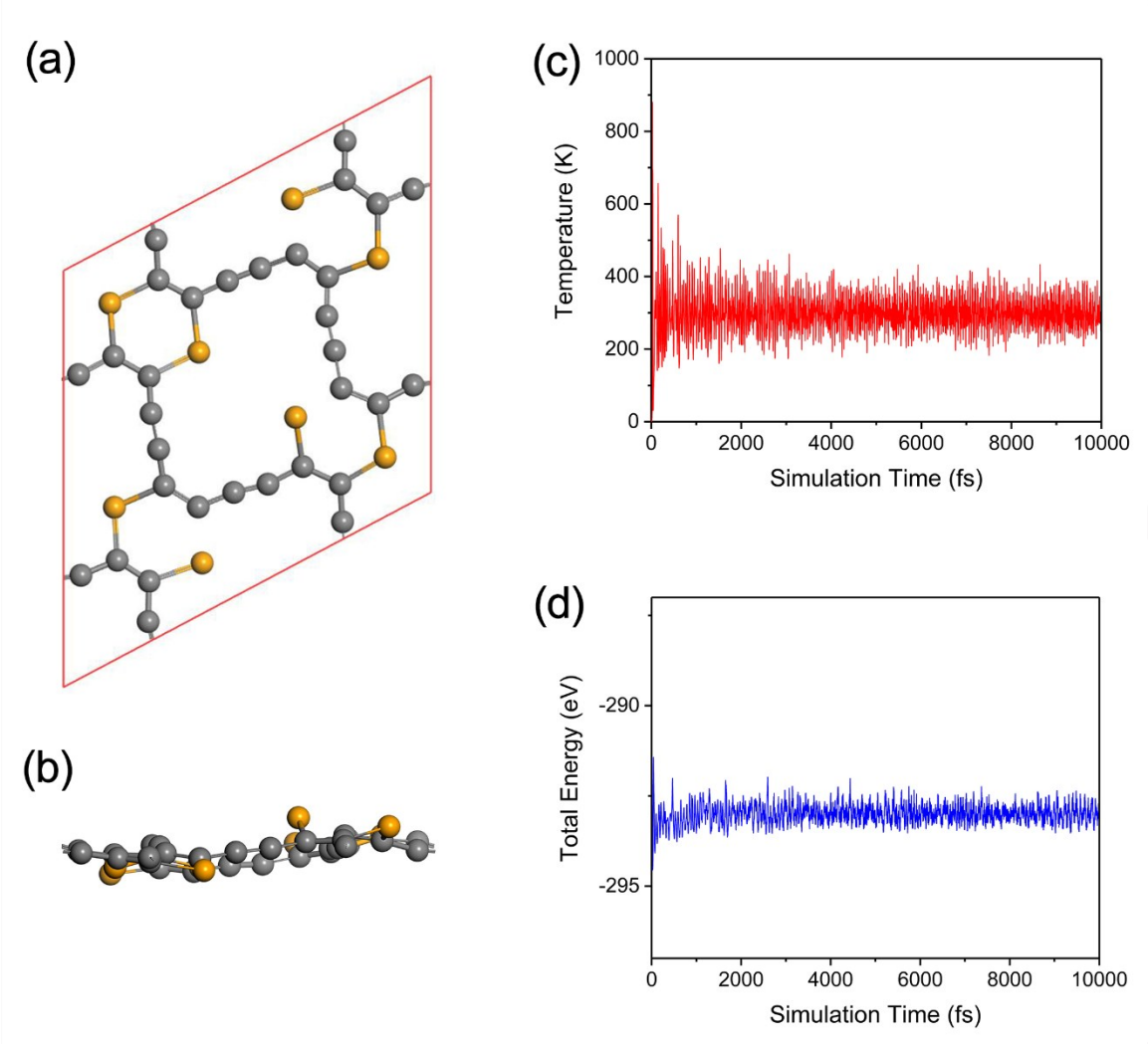

Fig. S6 (a) Top view and (b) side view of γ-C_4Se after 10 ps FPMD simulation at 300K, and (c) temperature and (d) energy changes during the simulation. Grey and brown balls stand for C and Se atoms.

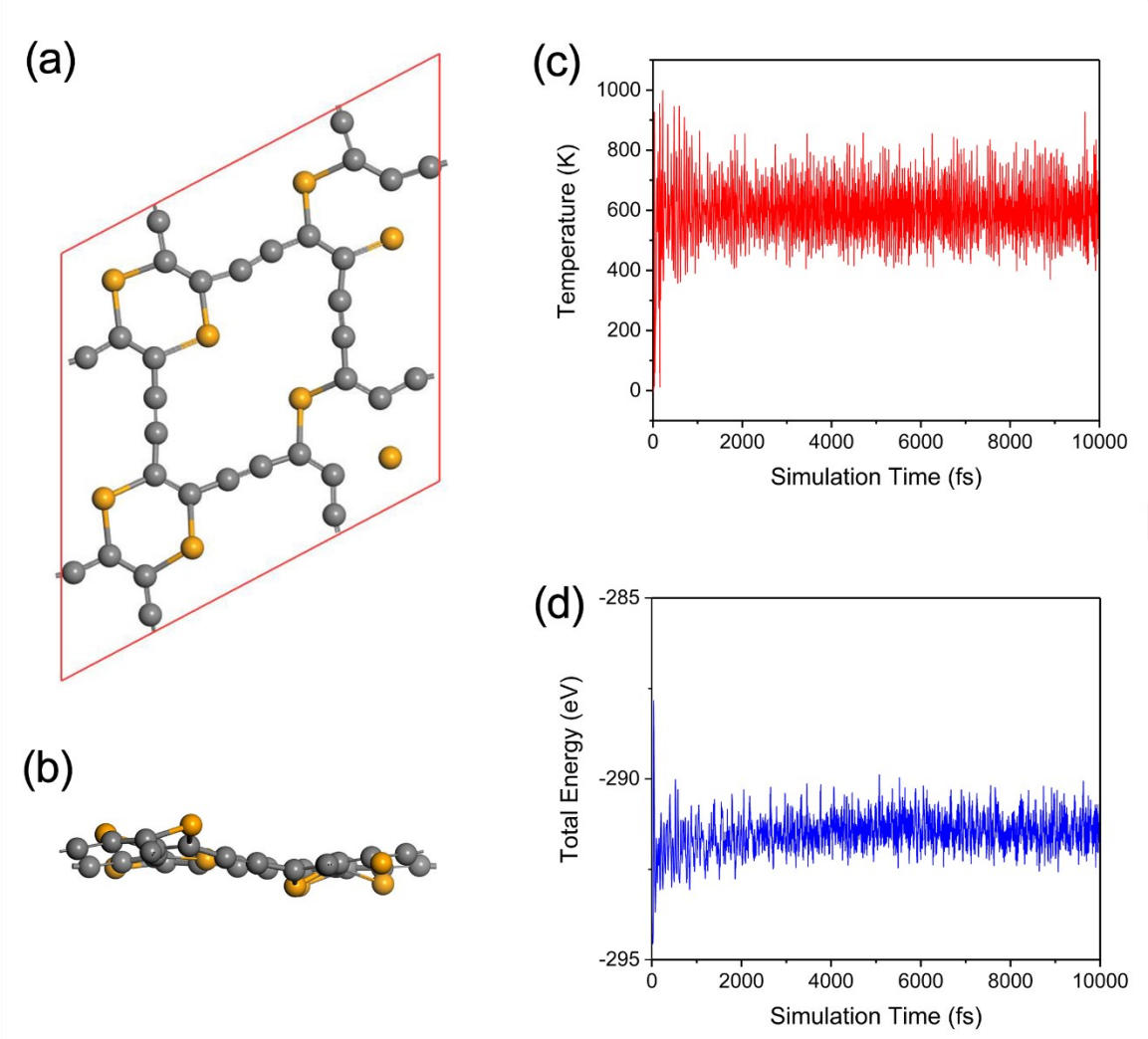

Fig. S7 (a) Top view and (b) side view of γ-C_4Se after 10 ps FPMD simulation at 600K, and (c) temperature and (d) energy changes during the simulation. Grey and brown balls stand for C and Se atoms.

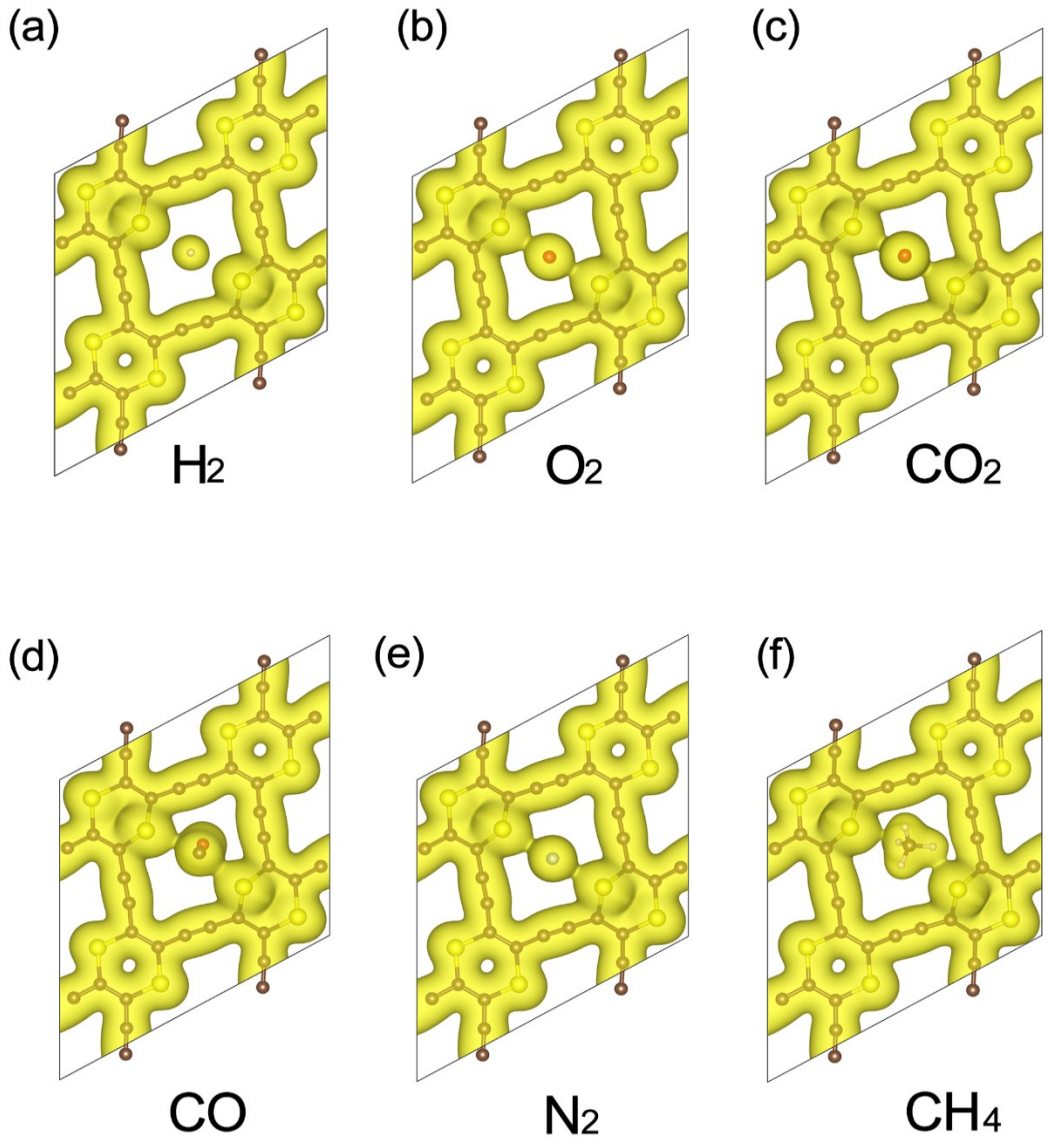

Fig. S8 Electron densities of the TS structures for H_2, O_2, CO_2, CO, N_2 and CH_4 penetrating γ-C_4S. The isovalue is 0.021 e $Å^{-3}$.

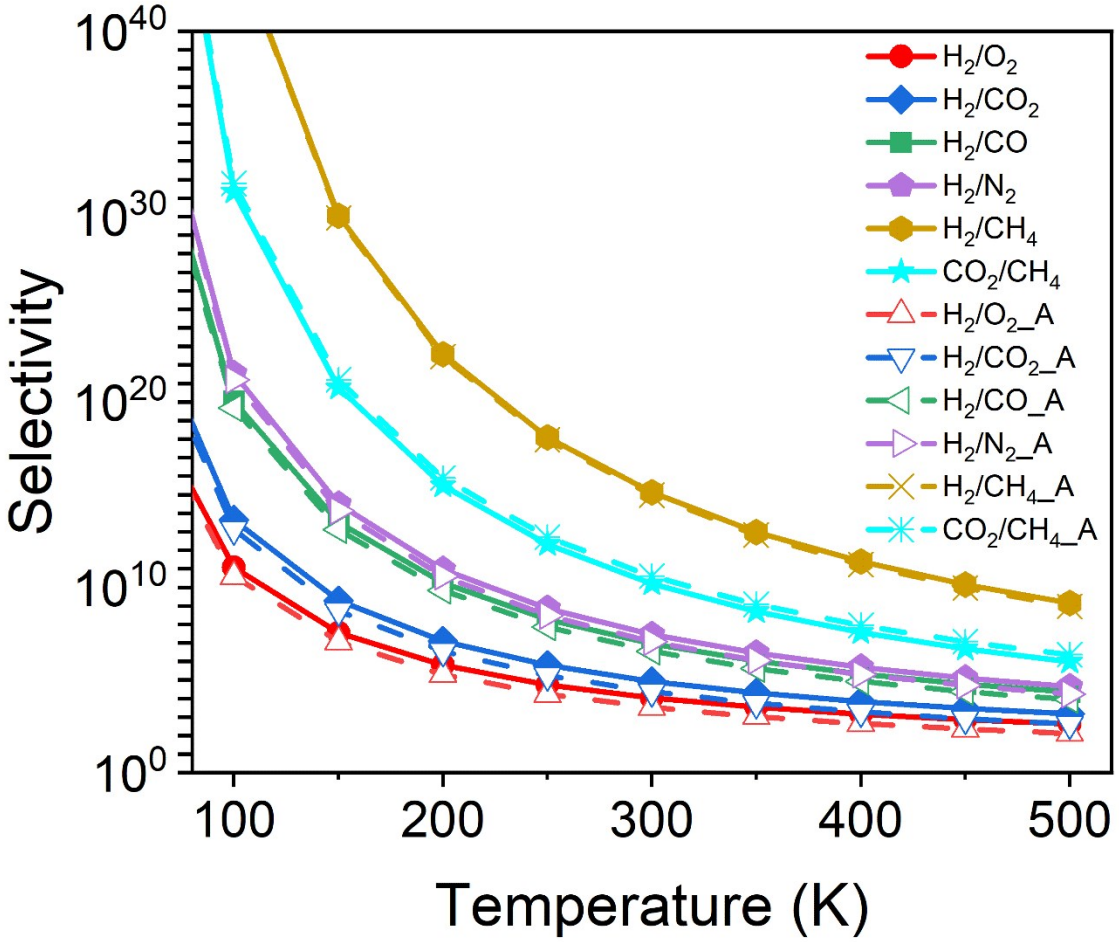

Fig. S9 Selectivity *versus* temperature for H_2/O_2, H_2/CO_2, H_2/CO, H_2/N_2, H_2/CH_4 and CO_2/CH_4 separation by γ-C_4O. The solid and dash lines stand for selectivity derived from the permeance and from the diffusion rate, respectively.

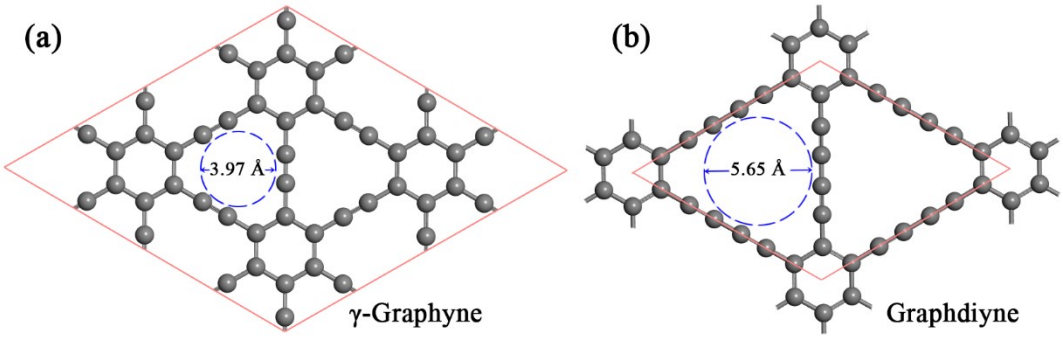

Fig. S10 The geometric structure of (a) γ-graphyne and (b) graphdiyne, with the size of intrinsic pore being marked.

Table S1. Top view and side view of the structures of Initial State (IS), Transition State (TS) and Final State (FS) for H_2, O_2, CO_2, CO, N_2 and CH_4 passing through the intrinsic pore of γ-C_4O.

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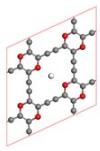
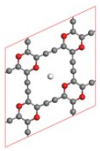
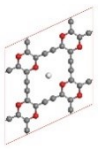
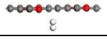
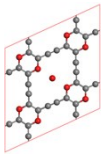
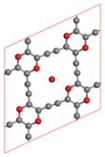
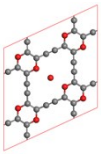
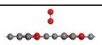
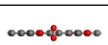
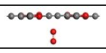
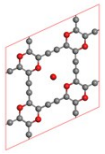
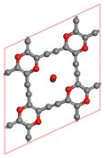
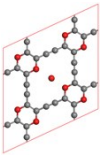
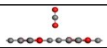
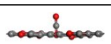
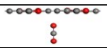
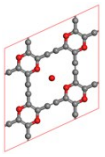
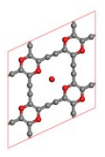
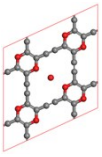
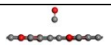
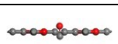
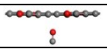
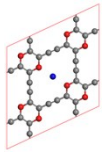
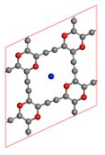
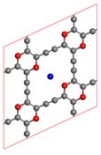
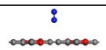
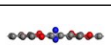
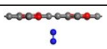
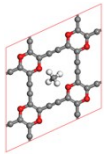
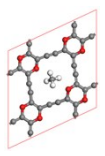
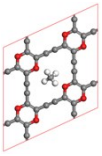
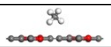
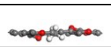
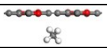
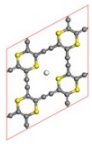
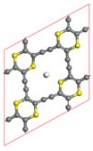
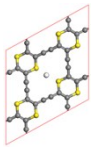
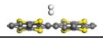
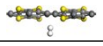
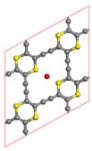
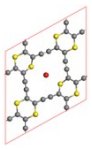
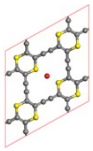
H_2	Top View			
	Side View			
O_2	Top View			
	Side View			
CO_2	Top View			
	Side View			
CO	Top View			
	Side View			
N_2	Top View			
	Side View			
CH_4	Top View			
	Side View			

Table S2. Top view and side view of the structures of IS, TS and FS for H_2, O_2, CO_2, CO, N_2 and CH_4 passing through the intrinsic pore of γ-C_4S.

		IS	TS	FS
H_2	Top View			
	Side View			
O_2	Top View			
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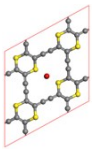
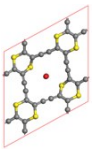
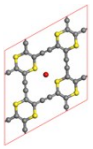
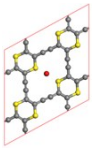
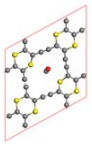
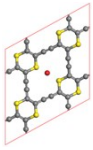
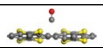
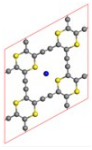
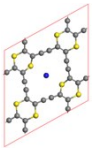
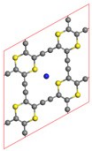
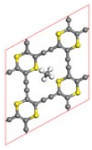
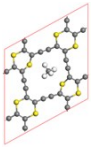
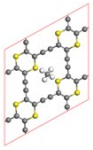
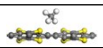
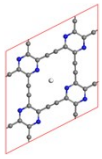
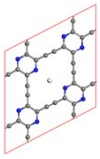
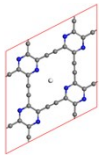
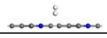
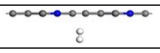
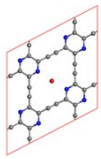
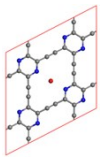
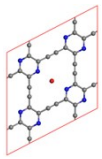
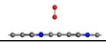
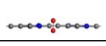
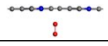
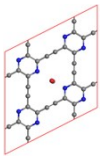
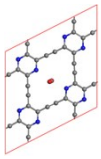
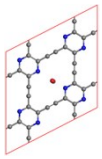
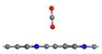
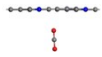
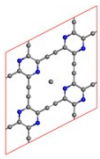
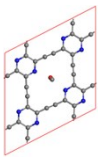
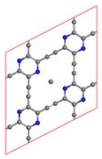
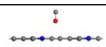
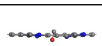
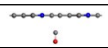
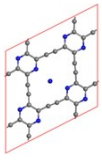
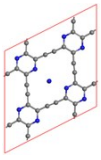
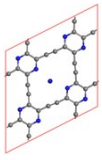
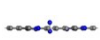
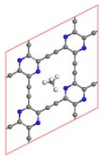
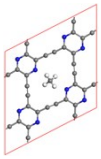
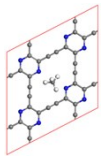
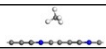
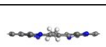
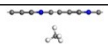
CO_2	Top View			
	Side View			
CO	Top View			
	Side View			
N_2	Top View			
	Side View			
CH_4	Top View			
	Side View			

Table S3. Top view and side view of the structures of IS, TS and FS for H_2, O_2, CO_2, CO, N_2 and CH_4 passing through the intrinsic pore of γ-C_4N.

		IS	TS	FS
H_2	Top View			
	Side View			
O_2	Top View			
	Side View			
CO_2	Top View			
	Side View			
CO	Top View			
	Side View			
N_2	Top View			
	Side View			
CH_4	Top View			
	Side View