Hidden Porous Boron Nitride as High-Efficiency Membrane for Hydrogen Purification

Shuyi Lin[1], Meiling Xu[*,1], Ziyang Qu[1], Yiwei Liang[1], Yan Li[2], Wenwen Cui[1], Jingming Shi[1], Qingxin Zeng[1], Jian Hao[1] and Yinwei Li[*,1]

[1] Laboratory of Quantum Materials Design and Application, School of Physics and Electronic Engineering, Jiangsu Normal University, Xuzhou 221116, China

[2] State Key Laboratory of Optoelectronic Materials and Technologies, School of Materials Science and Engineering, Sun Yat-Sen (Zhongshan) University, Guangzhou 510275, China

Corresponding Authors: xml@calypso.cn and yinwei_li@jsnu.edu.cn

Details on structure prediction: Unit cells containing 1-4 formula units were considered. In the first generation, a population of structures belonging to certain space group symmetries are randomly constructed. Starting from the second generation, 60% structures in the previous generation with the lower enthalpies are selected to produce the structures of next generation by the Particle Swarm Optimization (PSO) operators. The 40% structures in the new generation are randomly generated. A structure finger printing technique of bond characterization matrix is applied to the generated structures, so that identical structures are strictly forbidden. These procedures significantly enhance the diversity of the structures, which is crucial for structural global search efficiency. The structure search was considered converged when ~1000 successive structures were generated after a lowest energy structure was found.

Table S1. The optimized structural parameters and energy with respect to *h*-BN monolayer of *m*-BN, *t*-BN, *h*'-BN and *h*''-BN monolayers.

Space group	Compound	*a*,*b*,*c* (Å, deg)	Atomic position	Δ*E* (eV/atom)
*P*6*m*	*h'*-BN	*a*=*b*=6.8345	B(6*k*) (0.33868, 0.45753, 0.5)	0.414
		c=39.4365	N(6*k*) (0.44566, 0.31830, 0.5)	
		α=*β*=90		
		γ=120		
*P*4/*mbm*	*t*-BN	*a*=*b*=4.9430	B(4*h*) (0.828, 0.358, 0.5)	0.313
		c=10.2852	N(4*h*) (0.84318, 0.65682, 0.5)	
		α=*β*=*γ*=90		
$P6_2/m$	*h''*-BN	a=b=7.0122	B1(3*g*) (0.40584, 0.40584, 0.5)	0.530
		c=31.1736	B2(3*g*) (0.78705, 0.78705, 0.5)	
		α=*β*=90	N1(3*g*) (0.20627, 0.20627, 0.5)	
		γ=120	N2(3*g*) (1.0, 0.41238, 0.5)	
*P*2/*c*	*m*-BN	a=30.4617	B1(4*g*) (0.5 0.6436, 1.0)	0.305
		b=4.5553	B2(2*f*) (0.5, 0.8577, 0.75)	
		c=7.6572	N1(4*g*) (0.5, 0.68378, 0.59346)	
		α=90	N2(2*f*) (0.5, 0.17943, 0.75)	
		β=91.5912		
		γ=90		

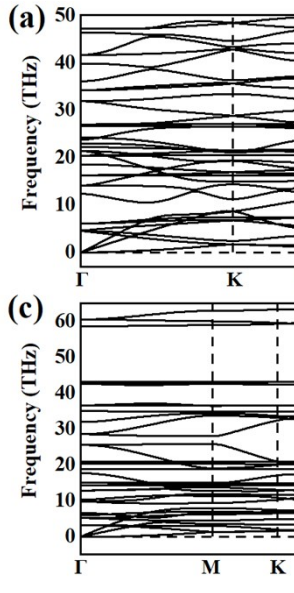

Figure S1. Phonon dispersion curves of (a) *h′*-BN, (b) *t*-BN, (c) *h″*-BN and (d) *m*-BN.

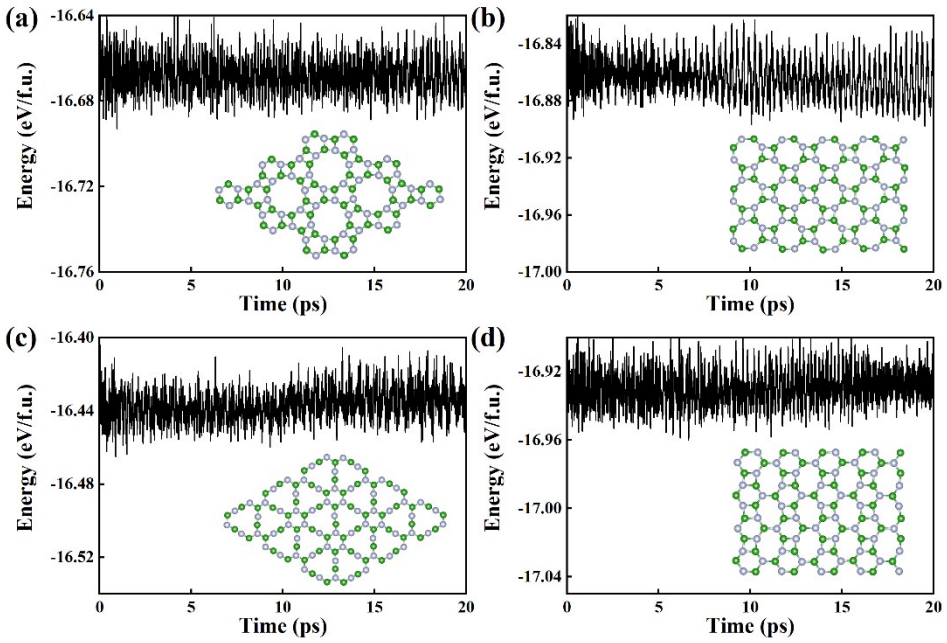

Figure S2. Total energy with respect to molecular dynamics (MD) simulation step at 300K of (a) *h′*-BN, (b) *t*-BN, (c) *h″*-BN and (d) *m*-BN. The insets are the structures after 20 ps MD simulation.

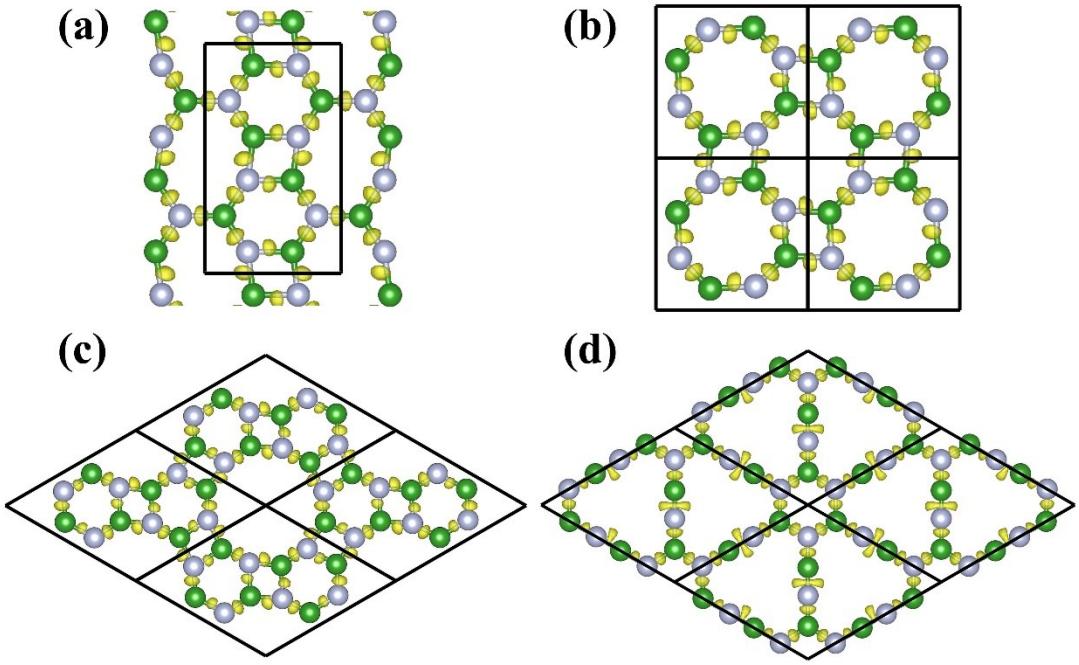

Figure S3. 3D Electronic local function (isovalue=0.85) for (a) *m*-BN, (b) *t*-BN, (c) *h′*-BN and (d) *h″*-BN

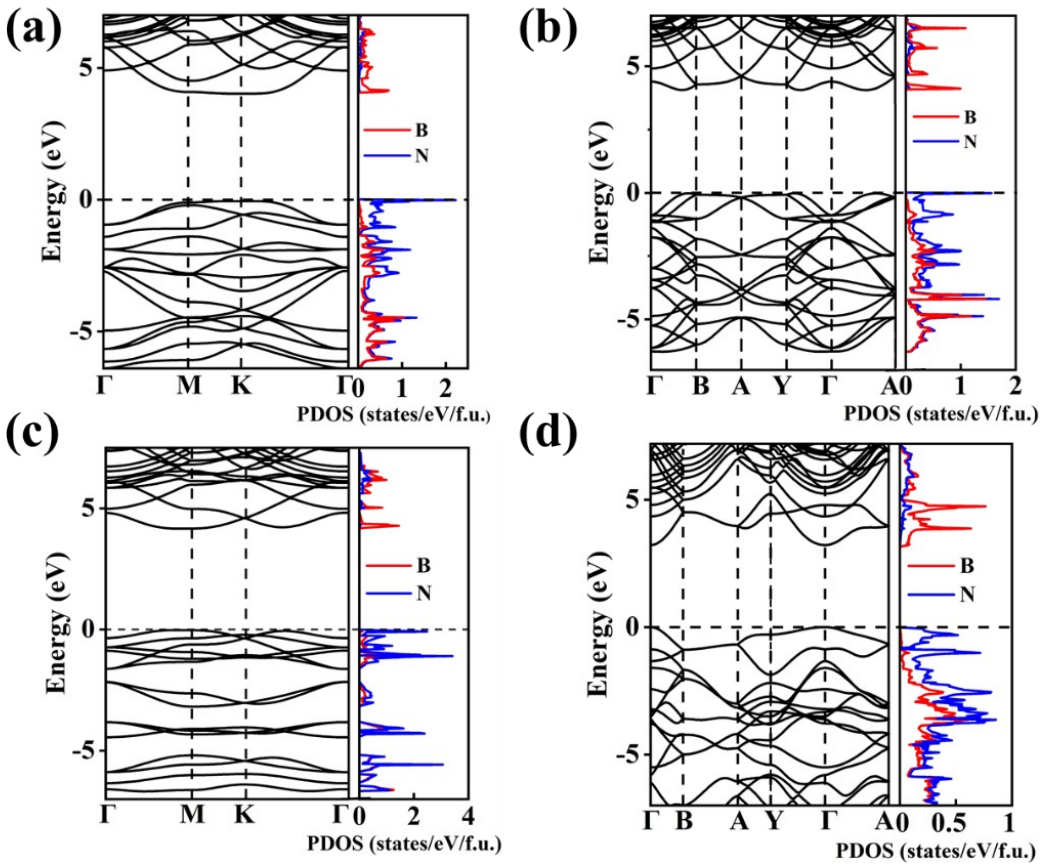

Figure S4. Calculated band structures and PDOS for (a) *h′*-BN, (b) *t*-BN, (c) *h″*-BN and (d) *m*-BN.

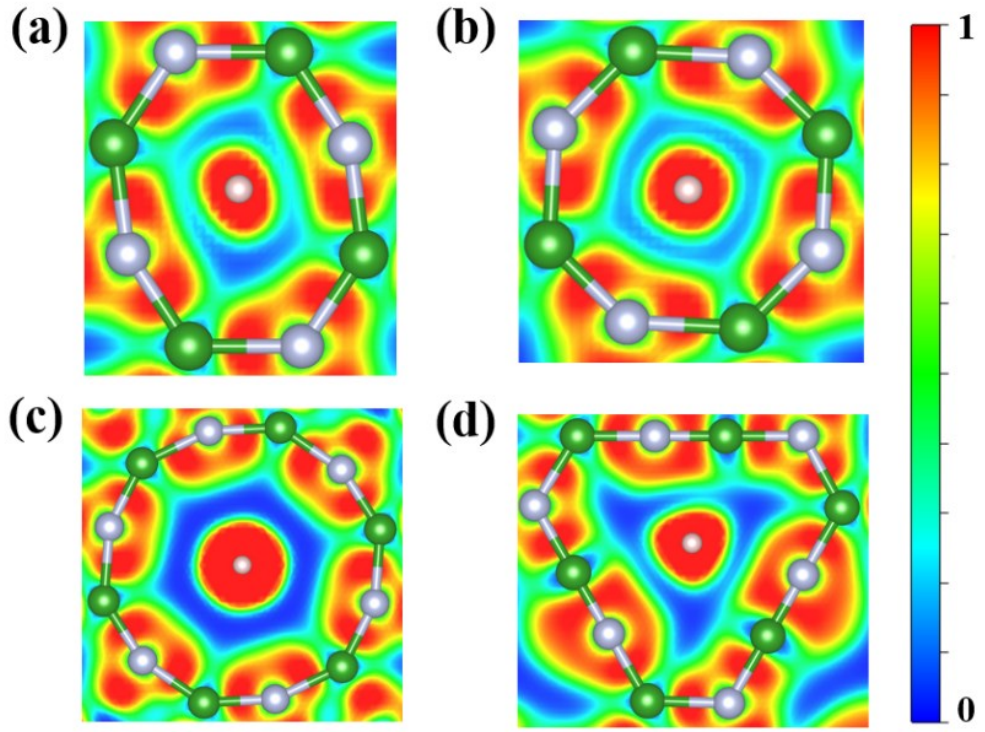

Figure S5. 2D plots of ELF on *z* axes when H_2 is in the center of pores for (a) *m*-BN, (b) *t*-BN, (c) *h*'-BN and (d) *h*"-BN.

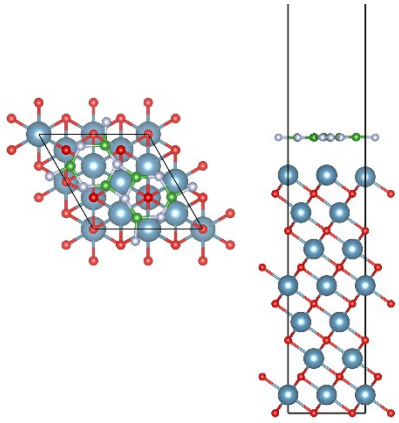

Figure S6. Top and side views of the optimized atomic structures for *h´*-BN/ CaO(111).