

Supporting Information

Details of CCSD(T)/CBS calculations:

In the MP2 and CCSD(T) calculation of ethyne-gas and benzene-gas complexes, the basis sets Aug-cc-pVDZ and Aug-cc-pVTZ were used. The MP2 correlation energies were extrapolated to the CBS limit by applying the two-point Helgaker extrapolation scheme:

$$E_{\text{MP2}}^{\text{CBS}} = \frac{E_X^{\text{MP2}} X^3 - E_Y^{\text{MP2}} Y^3}{X^3 - Y^3}$$

where X and Y are the cardinal numbers of the two basis sets used. In the present work, Aug-cc-pVDZ and Aug-cc-pVTZ were used for the MP2 extrapolation to the CBS limit. Then, CCSD(T) calculations using the Aug-cc-pVDZ basis set were performed. These CCSD(T) correlation energies were extrapolated to the complete basis limit by applying the following additivity scheme:

$$E_{\text{CCSD(T)}}^{\text{CBS}} = E_{\text{MP2}}^{\text{CBS}} + (E_{\text{CCSD(T)}} - E_{\text{MP2}})^{\text{avdz}}$$

According to this scheme, the CCSD(T) energies are computed with a small basis set (such as aug-cc-pVDZ). Then, the difference between the CCSD(T) and MP2 energies in the small basis set is added to the MP2/CBS result.

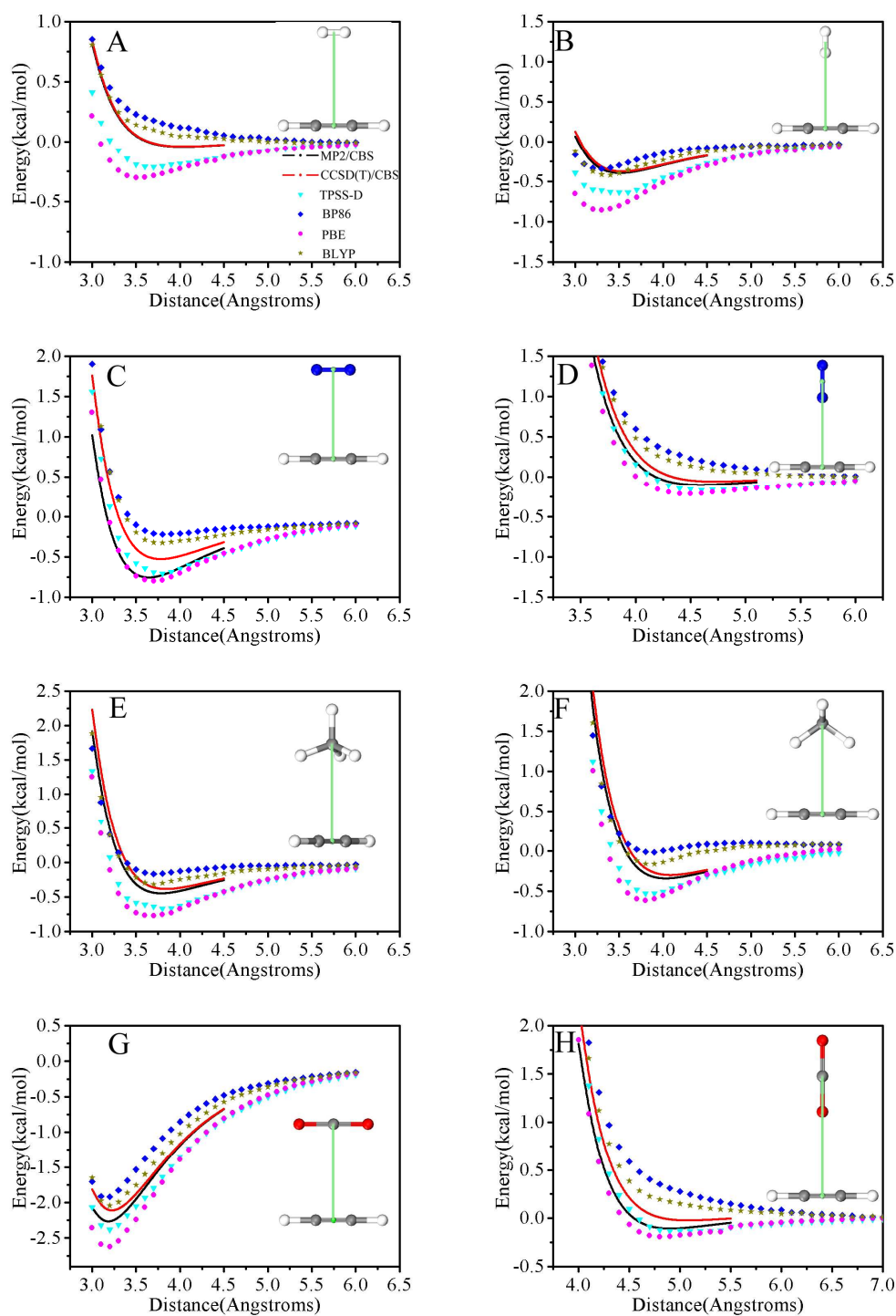


Figure S1. Comparison of calculated IEs of ethyne-gas complexes at the DFT-D/Aug-cc-pVTZ levels with the ones estimated at the MP2/CBS and CCSD(T)/CBS levels. The green lines between the two molecules denote the direction of scanning, and the x-axis donates the distance between center of mass of each molecule.

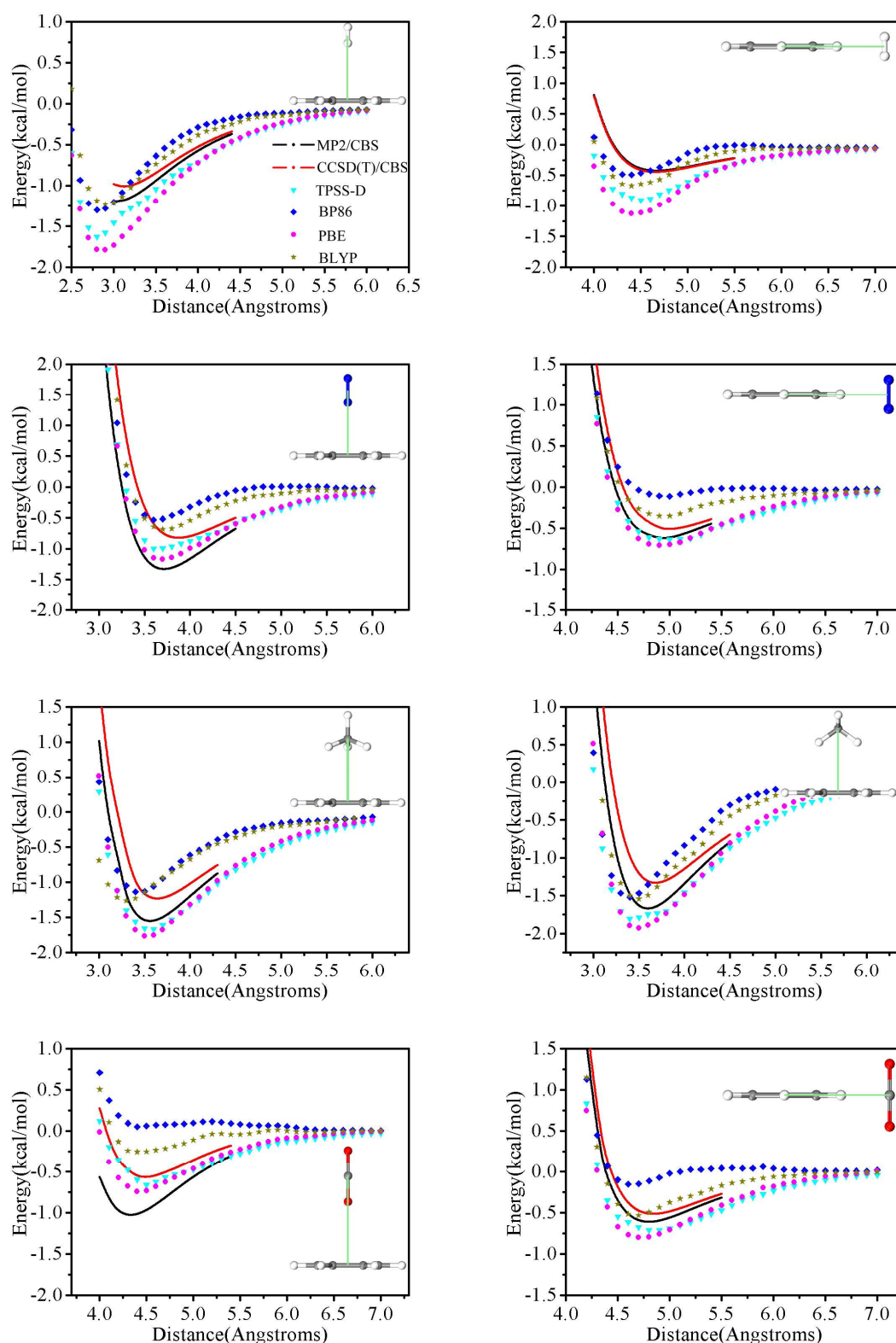


Figure S2. Comparison of calculated IEs of benzene-gas complexes at the DFT-D/Aug-cc-pVTZ levels with the ones estimated at the MP2/CBS and CCSD(T)/CBS levels. The green lines between the two molecules denote the direction of scanning, and the x-axis donates the distance between center of mass of each molecule.

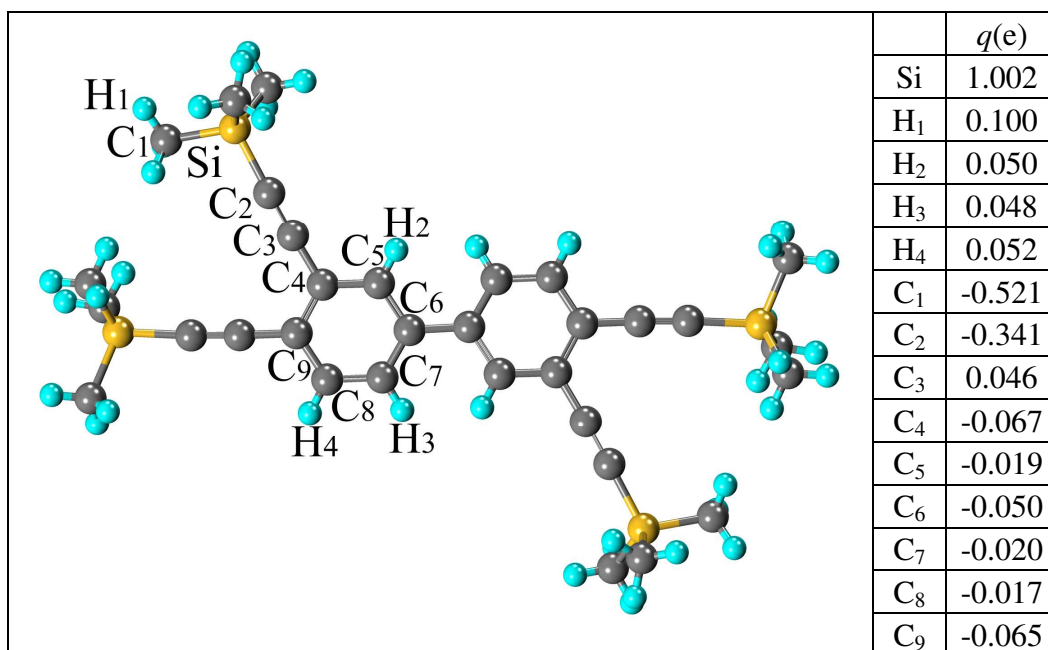


Figure S3. Atomic Charge on the TTB Molecule.

Table S1. Parameters for Gases and Frameworks in This Work

		σ (Å)	$\varepsilon/k_B(K)$	q (e)
TTB	C	3.90	23.93	
	H	3.20	3.82	
	Si	4.27	78.00	
Gases	H ₂	2.958	36.7	0
	CH ₄	3.73	148.0	0
	N_N ₂	3.31	36.0	-0.482
	Ps_N ₂	0	0	0.964
	C_CO ₂	2.80	27.0	0.700
	O_CO ₂	3.05	79.0	-0.350

Table S2. The fitting parameters of the LF equation to GCMC simulation results for the adsorption of H₂ and N₂ in TTB

	Value	Standard Error	Value	Standard Error	Value	Standard Error
	H ₂		N ₂ -Low pressure		N ₂ -High pressure	
M	43.7654	37.3361	4.9992	0.3739	1415.1974	2.4745E6
k	0.05778	0.05237	11.5088	10.4847	0.00334	5.86178
β	0.27913	0.01903	0.1598	0.09597	0.1308	0.3063

Table S3. The fitting parameters of the LF equation to GCMC simulation results for the adsorption of CO₂ and CH₄ in TTB

	Value	Standard Error	Value	Standard Error
	CO ₂		CH ₄	
M	19.1188	14.3227	2.3705	0.08342
k	0.00661	0.00451	0.01054	0.0009196
β	0.4100	0.02357	0.74873	0.02556