Benchmarking Adsorption Energetics and Reaction Mechanisms: Insights in Water–Carbon and H–Si Interfaces

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Abstract

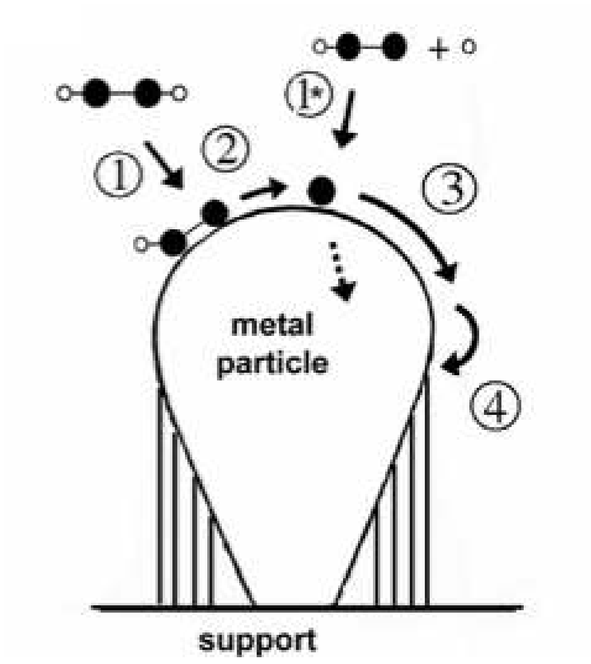
- Gas-phase molecules interacting with surfaces drive key physical and chemical processes, with reaction energetics shaped by bond rearrangement, charge transfer, and intermolecular forces.
- High-level ab initio electronic structure theories enhance our understanding of reaction barriers and molecule–surface interactions, providing deeper insight into catalytic and growth processes.
- In CVD growth of CNTs, precursor dissociation on metal catalysts governs growth kinetics and activation energies, enabling controlled synthesis.

Presented Work

Adsorption Energies on BN and 2d C: Adsorption of water on BN and graphene substrates, with a comparative analysis of computational methods to evaluate interaction strengths, binding geometries, and long-range correlation effects.

H_2 Adsorption on Si(100): Compare different adsorption/desorption mechanisms, benchmarking high-level periodic wavefunction methods against low-level electronic structure theory and experimental data.

Future Work

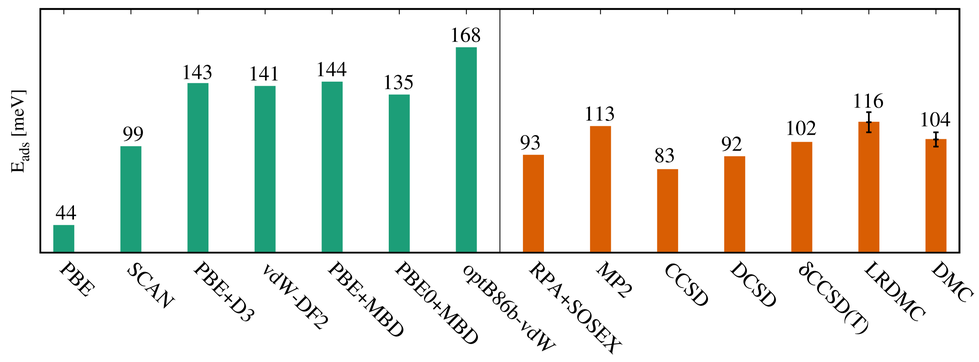


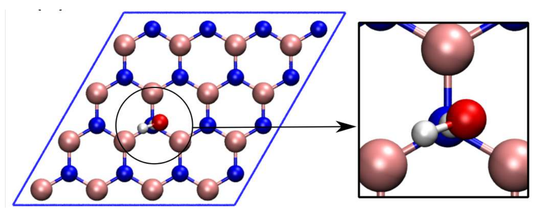
Study fundamental processes during CNT growth:

1. Adsorption of precursors molecule on the catalyst surface
2. Dissociation of the precursor molecule
3. Diffusion of the growth species in or on the catalyst particle
4. Nucleation and incorporation of carbon into the growing structure

S.Hofmann *et. al.*, PRL 95, 036101 (2005)

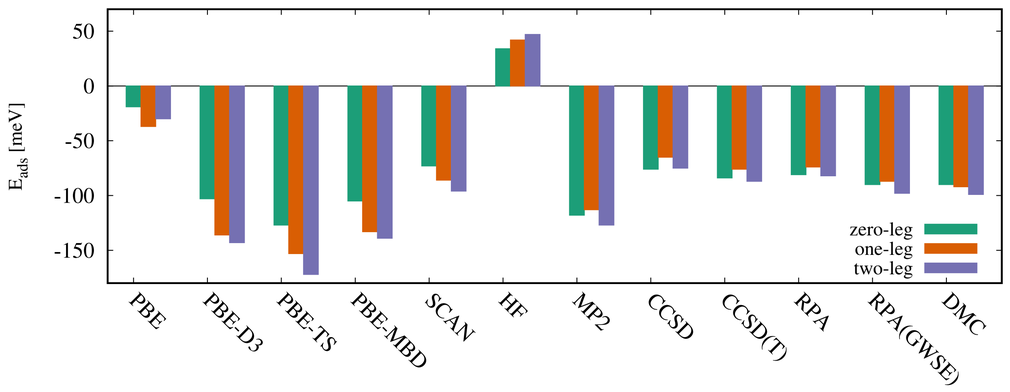
Water adsorption on h-BN

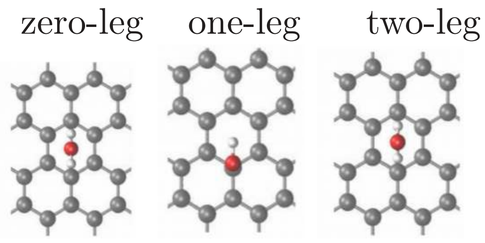




Y.S.Al-Hamdani *et. al.*, JCP 147, 044710 (2017)
T.Gruber *et. al.*, PRX 8, 021043 (2018)

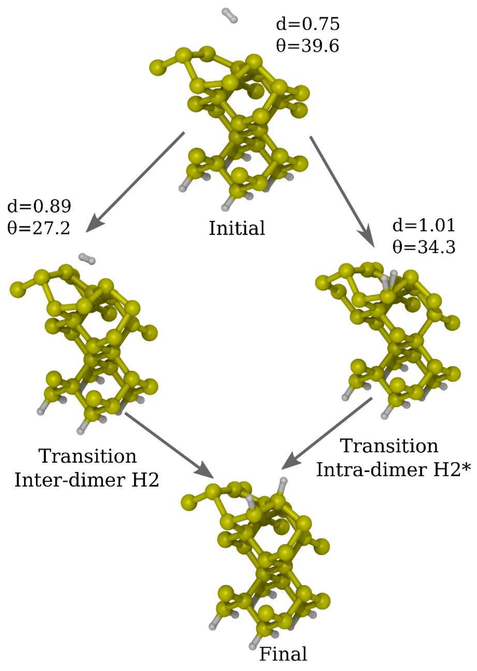
Water adsorption on Graphene

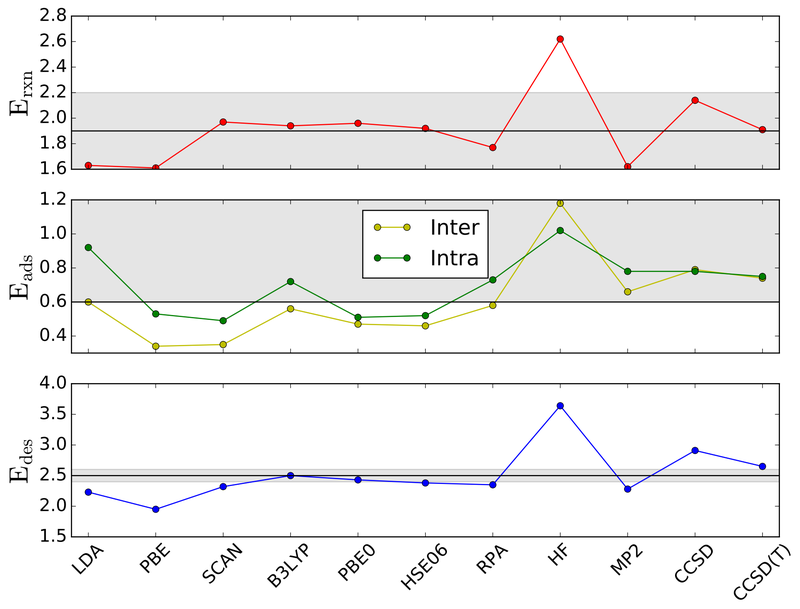




J.G.Brandenburg *et. al.*, J. Phys. Chem. Lett. 2019,10,3,358-368
A.O.Ajala *et. al.*, J. Chem. Theory Comput. 2019,15,4,2359-2374

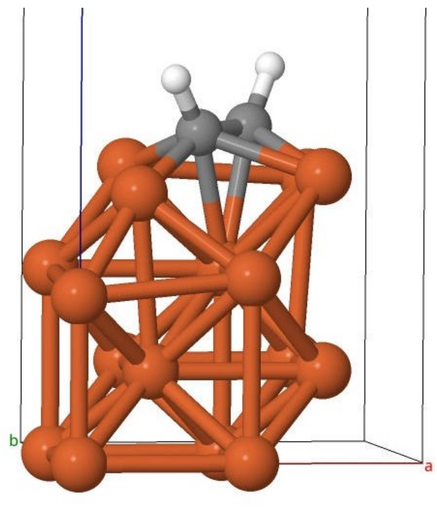
H_2 dissociative adsorption on Si (001)





T.Tsatsoulis *et. al.*, JCP 149, 244105 (2018)
C.Filippi *et. al.*, PRL 89, 166102 (2002)

Towards Predictive Modeling of CNT Growth



Reaction Intermediates & Growth Pathways: Future work should focus on clarifying the nature of reaction intermediates during CNT growth, distinguishing between adatoms, dimers, chains, and undissociated precursors.

Benchmarking Electronic Structure & Atomistic Models: To ensure reliability, benchmarking of electronic structure theories and atomistic models—including against high-level ab initio methods can improve predictive accuracy.

Work in progress