Electronic Supplementary Material (ESI) for Nanoscale Advances.

Supporting Information

A Comparative Study of Mechanisms of the Adsorption of CO_2 Confined within Graphene-MoS_2 Nanosheets: A DFT Trend Study

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CO_2 Adsorption Energies on Different Interlayer Spacing

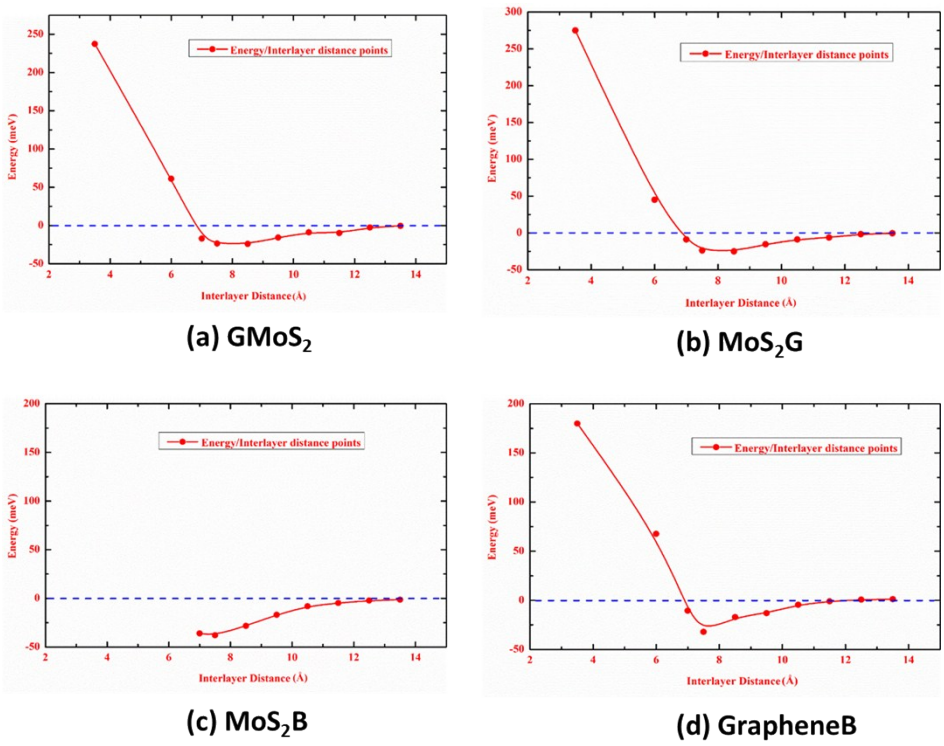

Fig. S1: Individual plots of CO_2 binding energy as a function of interlayer distance. (a) Graphene/MoS_2 hybrid, (b) MoS_2/Graphene hybrid, (c) MoS_2 bilayer, and (d) Graphene bilayer.

Adsorption energies of possible initial configurations of CO_2 adsorption on perfect multilayer and monolayer structures using PBE [31]

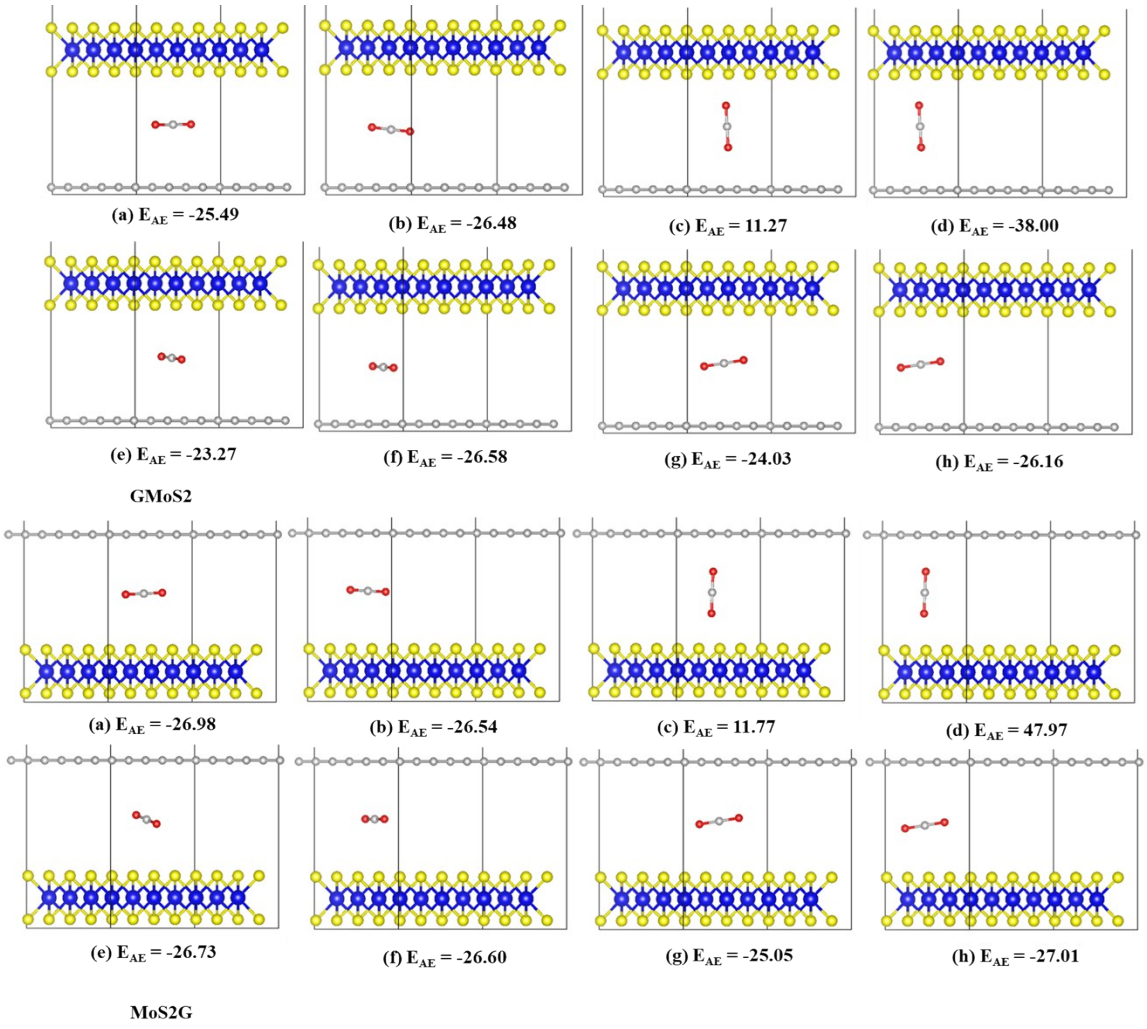

Fig. S2 Relaxed geometric structures of CO_2 adsorption on possible initial graphene/MoS_2 ($GMoS_2$) and MoS_2/graphene (MoS_2G) hybrid configurations. Position and orientation of CO_2 with respect to the surface are: (a) Centre/parallel, (b) Edge/parallel, (c) Centre/perpendicular, (d) Edge/perpendicular, (e) Centre/parallel (rotated 30°), (f) Edge/parallel (rotated 30°), (g) Centre/parallel (rotated 45°), and (h) Edge/parallel (rotated 45°). Energies are given in meV. Negative means heat release and blue, yellow, red and grey spheres depict Mo, S, O and C atoms, respectively.

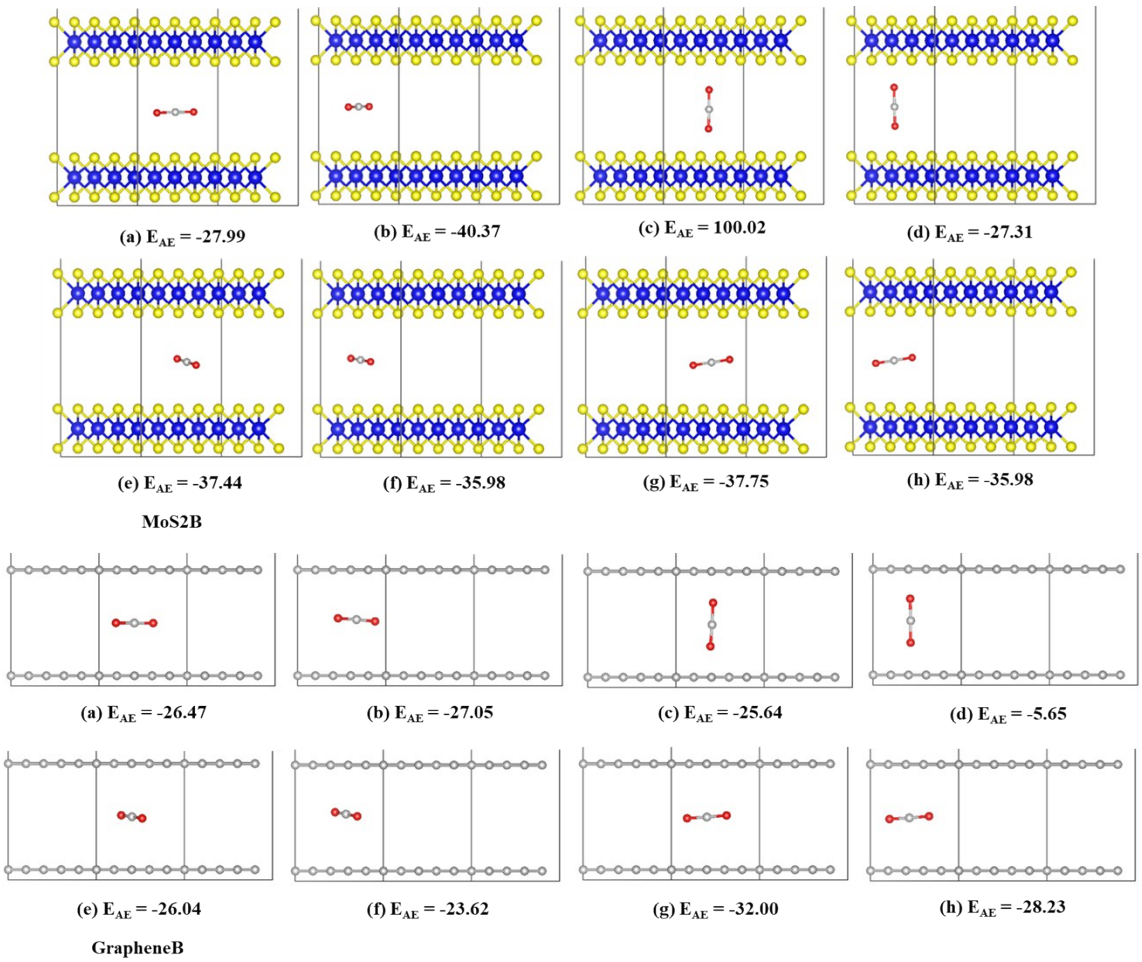

Fig. S3 Relaxed geometric structures of CO_2 adsorption on possible initial bilayers of MoS_2 (MoS_2B) and graphene (GrapheneB) configurations. Position and orientation of CO_2 with respect to the surface are: (a) Centre/parallel, (b) Edge/parallel, (c) Centre/perpendicular, (d) Edge/perpendicular, (e) Centre/parallel (rotated 30^o), (f) Edge/parallel (rotated 30^o), (g) Centre/parallel (rotated 45^o), and (h) Edge/parallel (rotated 45^o). Energies are given in meV. Negative means heat release and blue, yellow, red and grey spheres depict Mo, S, O and C atoms, respectively.

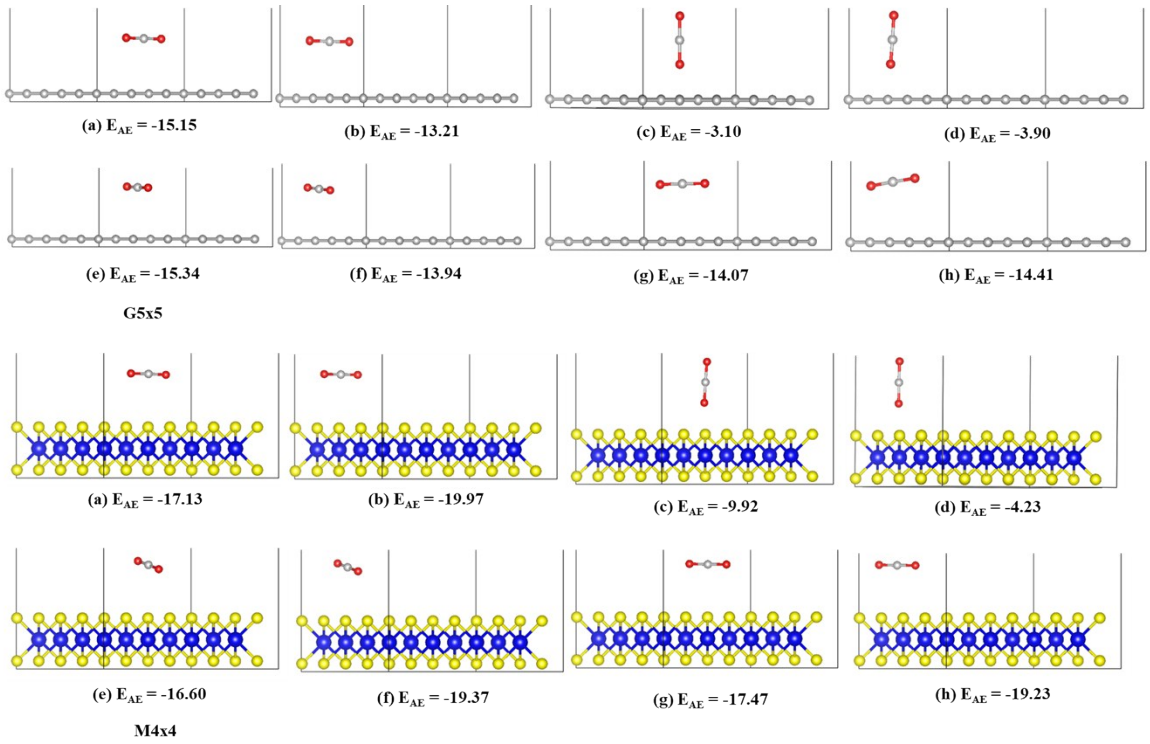

Fig. S4 Relaxed geometric structures of CO_2 adsorption on possible initial monolayers of graphene (G5x5) and MoS_2 (M4x4) configurations. Position and orientation of CO_2 with respect to the surface are: (a) Centre/parallel, (b) Edge/parallel, (c) Centre/perpendicular, (d) Edge/perpendicular, (e) Centre/parallel (rotated 30°), (f) Edge/parallel (rotated 30°), (g) Centre/parallel (rotated 45°), and (h) Edge/parallel (rotated 45°). Energies are given in meV. Negative means heat release and blue, yellow, red and grey spheres depict Mo, S, O and C atoms, respectively.

Test for Cutoff Energy and KPOINT Grid Required for Convergence

Table S1: Adsorption energies of CO_2 on bilayer, hybrid and monolayer structures of most stable structures at EDDIF=10^-4 and EDDIG=-0.03, NSW=300, ENCUT=500, 5×5×1 KPOINT

Structure	Adsorption Energies E_{AE} (meV)
Edge/Parallel (rotated 30°) (6GMoS_2)	-26.58
Edge/Parallel (rotated 45°) 7MoS_2G	-27.01
Edge/Parallel (3MoS_2)	-40.37
Centre/Parallel (rotated 45°) (8Graphene)	-32.00
Centre/Parallel (rotated 30°) (5G5x5)	-15.34
Edge/Parallel (3M4x4)	-19.97

Table S2: Adsorption energies of CO_2 on bilayer, hybrid and monolayer structures of most stable structures at EDDIF=10^-6 and EDDIG=-0.01, NSW=500, ENCUT=500, 5×5×1 KPOINT

Structure	Adsorption Energies E_{AE} (meV)
Edge/Parallel (rotated 30^o) (6GMoS_2)	-26.44
Edge/Parallel (rotated 45^o) 7MoS_2G	-26.84
Edge/Parallel (3MoS_2)	-39.85
Centre/Parallel (rotated 45^o) (8Graphene)	-32.01
Centre/Parallel (rotated 30^o) (5G5x5)	-15.35
Edge/Parallel (3M4x4)	-19.65

Table S3: Adsorption energies of CO_2 on bilayer, hybrid and monolayer structures of most stable structures at EDDIF=10^-6 and EDDIG=-0.03, NSW=500, ENCUT=600, 6x6x1

Structure	Adsorption Energies E_{AE} (meV)
Edge/Parallel (rotated 30^o) (6GMoS_2)	-27.16
Edge/Parallel (rotated 45^o) 7MoS_2G	-27.07
Edge/Parallel (3MoS_2)	-39.59
Centre/Parallel (rotated 45^o) (8Graphene)	-32.28
Centre/Parallel (rotated 30^o) (5G5x5)	-15.53
Edge/Parallel (3M4x4)	-19.37

Table S4: Adsorption energies of CO_2 on bilayer, hybrid and monolayer structures of most stable structures at EDDIF=10^-6 and EDDIG=-0.03, NSW=500, ENCUT=700, 7x7x1

Structure	Adsorption Energies E_{AE} (meV)
Edge/Parallel (rotated 30^o) (6GMoS_2)	-27.14
Edge/Parallel (rotated 45^o) 7MoS_2G	-27.61
Edge/Parallel (3MoS_2)	-39.63
Centre/Parallel (rotated 45^o) (8Graphene)	-32.37
Centre/Parallel (rotated 30^o) (5G5x5)	-16.38
Edge/Parallel (3M4x4)	-20.33

Density of State (DOS) Analysis

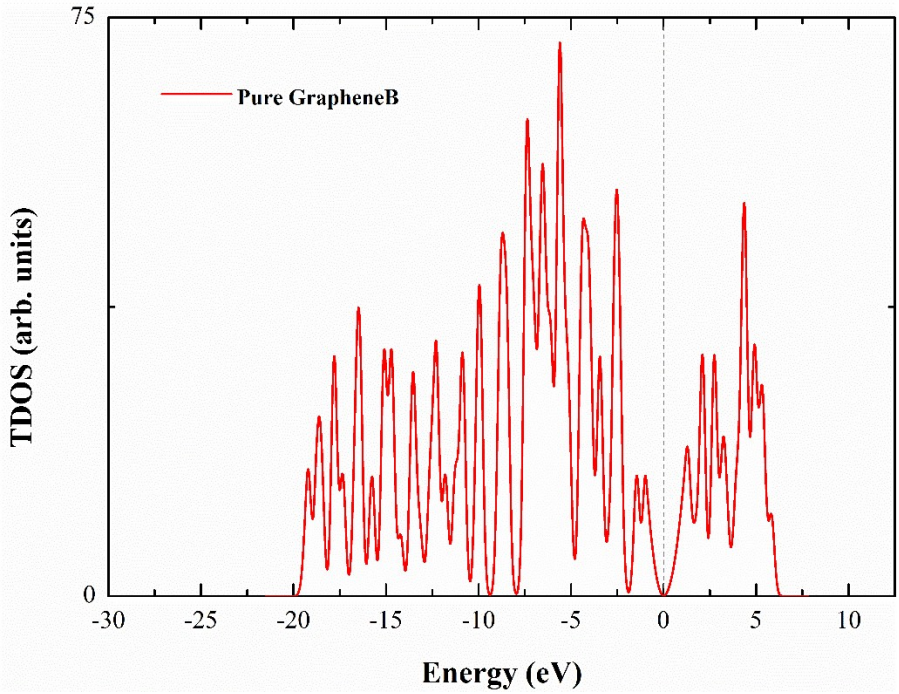

Fig. S5: Total density of state (TDOS) of pure graphene bilayer.

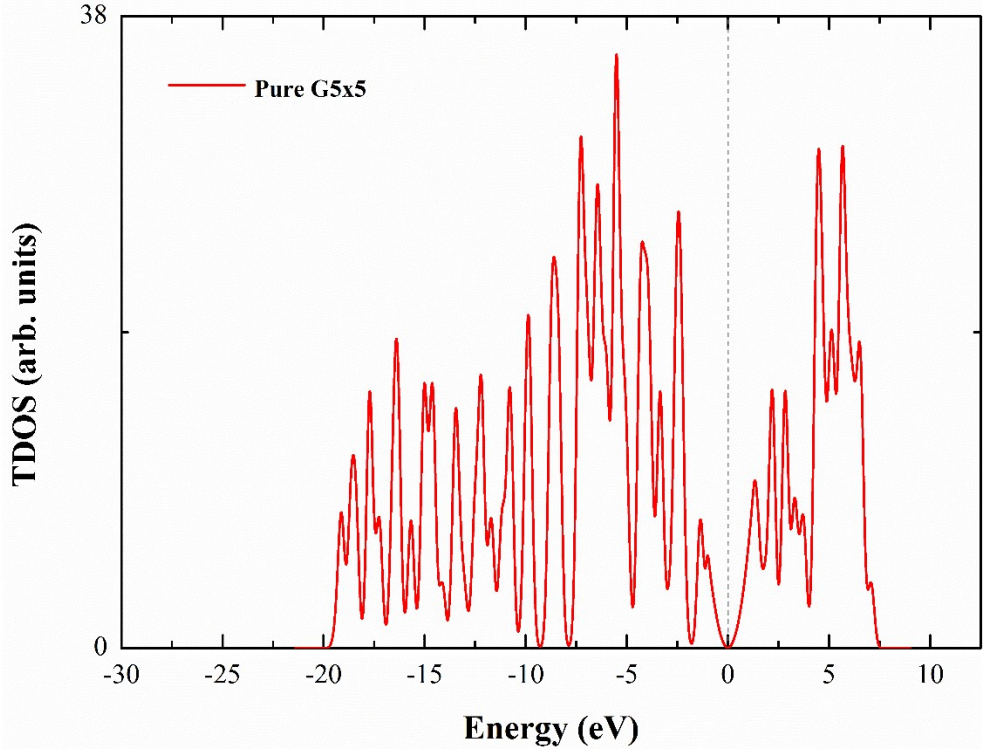

Fig. S6: Total density of state (TDOS) of pure 5x5 supercell graphene monolayer.

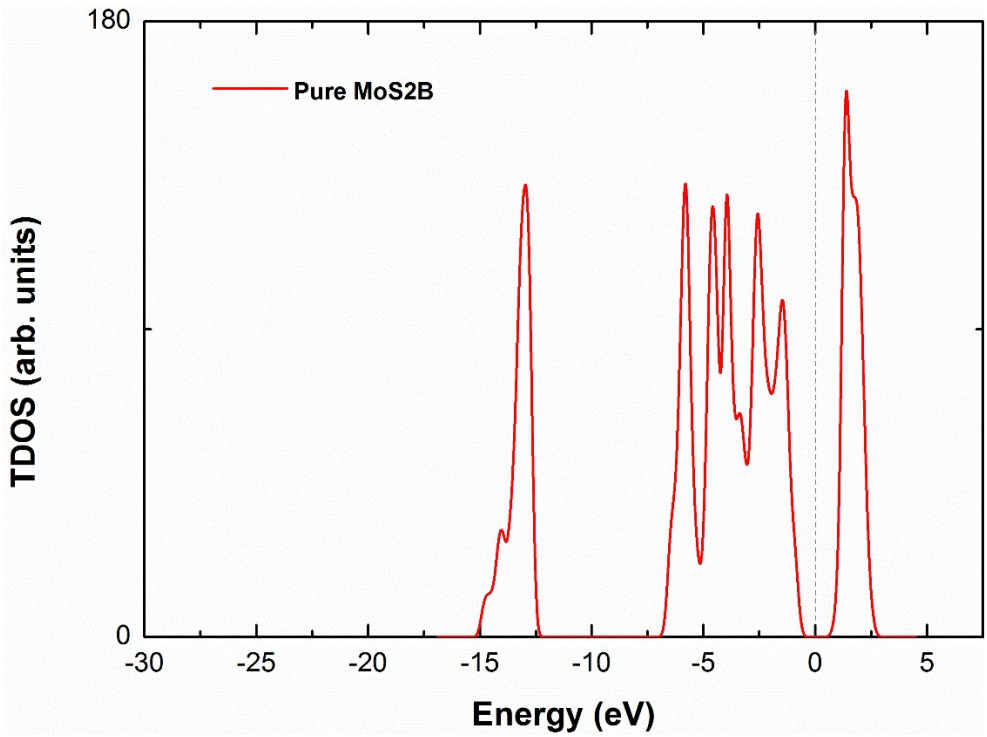

Fig. S7: Total density of state (TDOS) of pure MoS_2 bilayer.

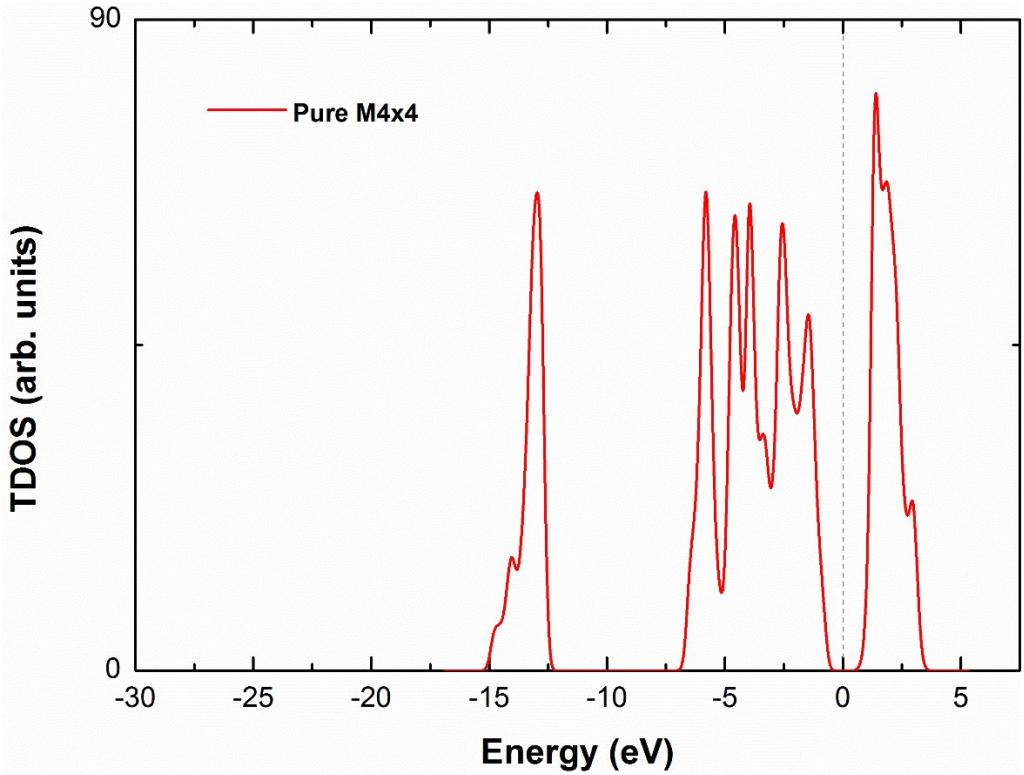

Fig. S8: Total density of state (TDOS) of pure 4x4 supercell MoS_2 monolayer.

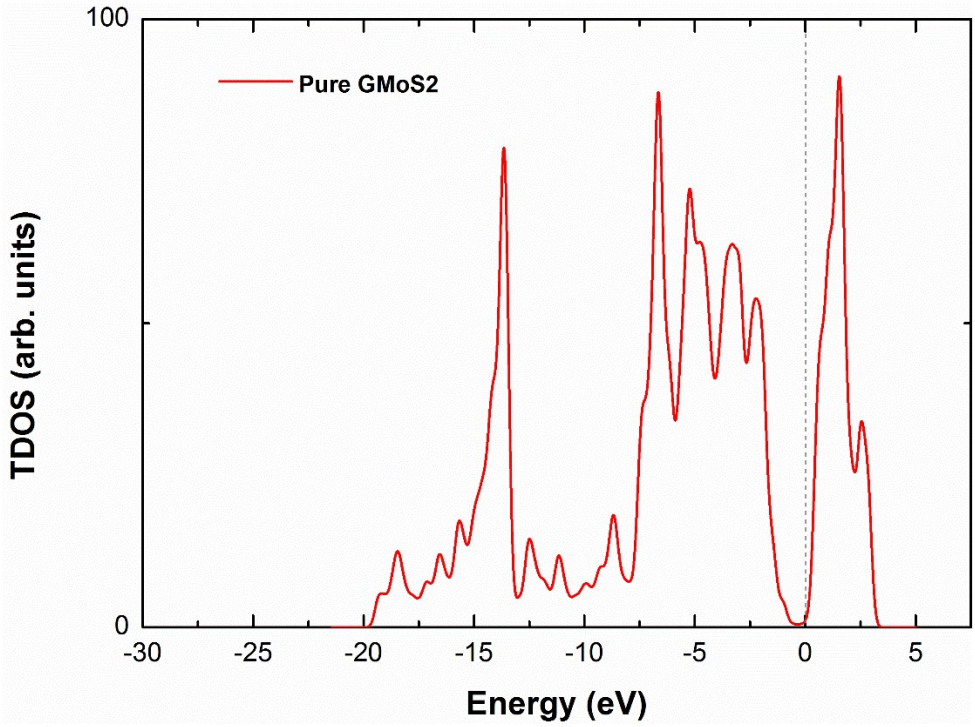

Fig. S9: Total density of state of pure Graphene/MoS_2 bilayer heterostructure.

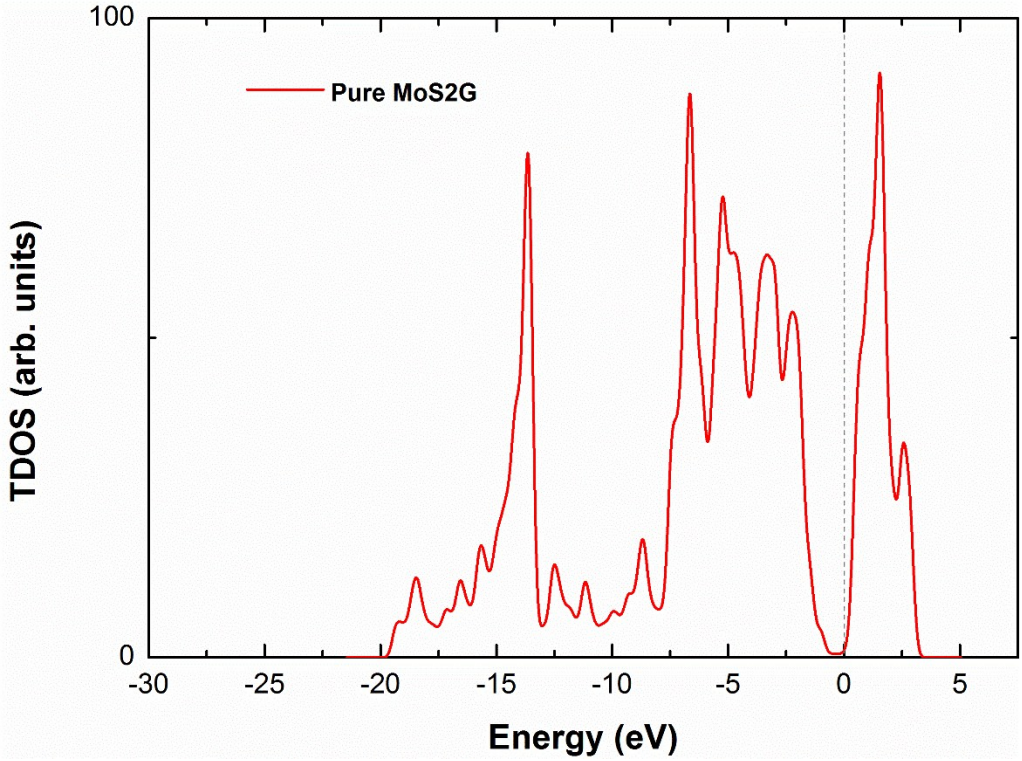

Fig. S10: Total density of state of pure MoS_2/Graphene bilayer heterostructure.

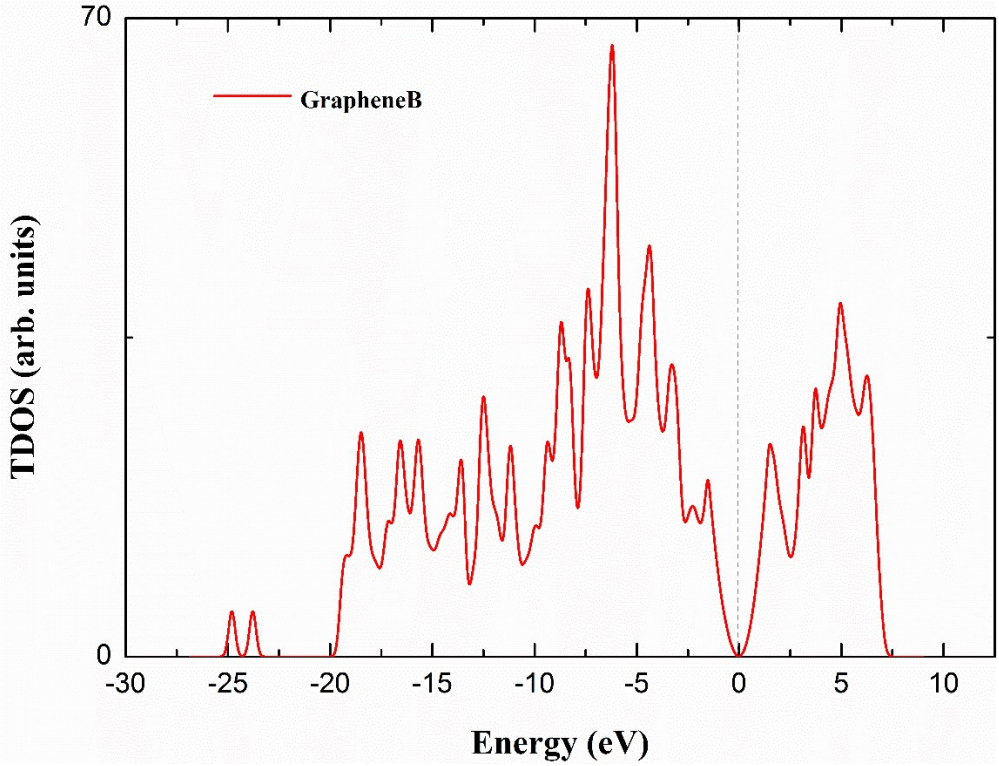

Fig. S11: Total density of state of graphene bilayer with adsorbed CO_2 molecule.

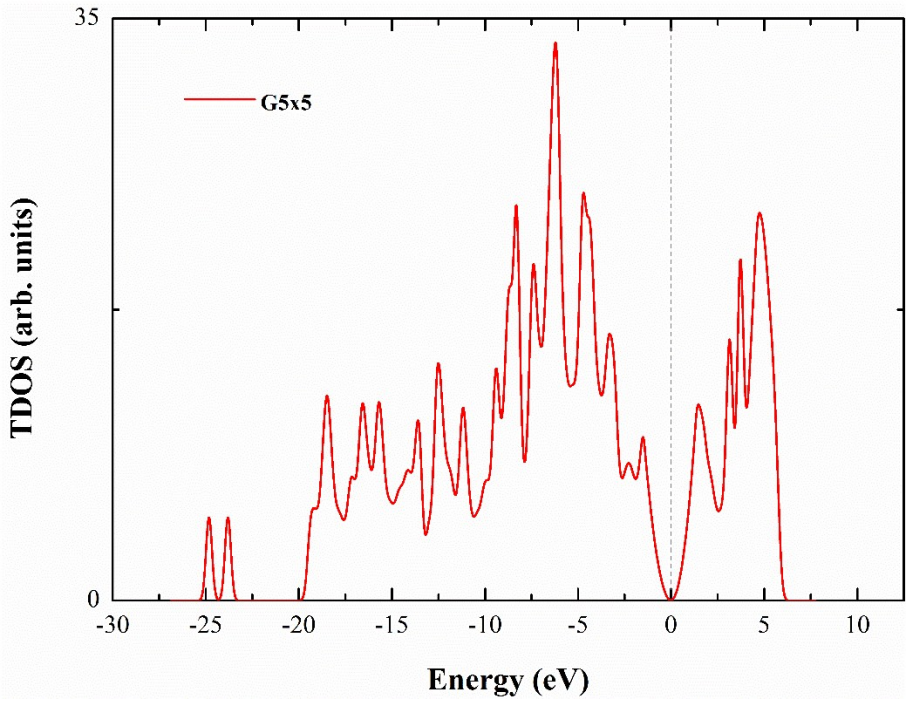

Fig. S12: Total density of state (TDOS) of 5x5 supercell graphene monolayer with adsorbed CO_2 molecule.

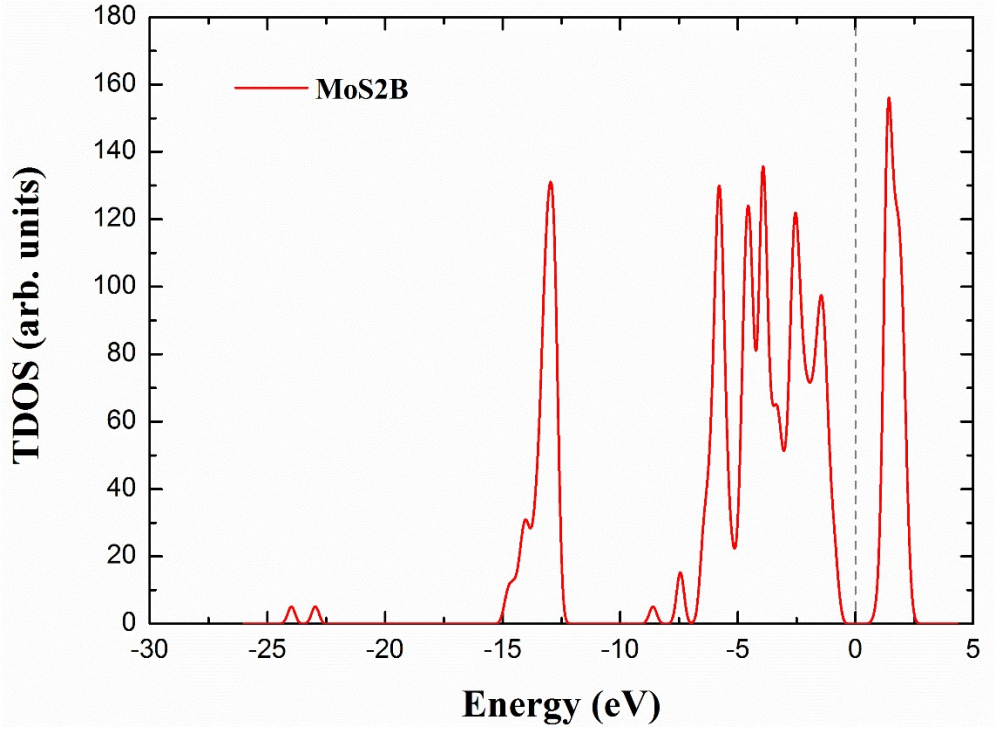

Fig. S13: Total density of state of MoS_2 bilayer with adsorbed CO_2 molecule.

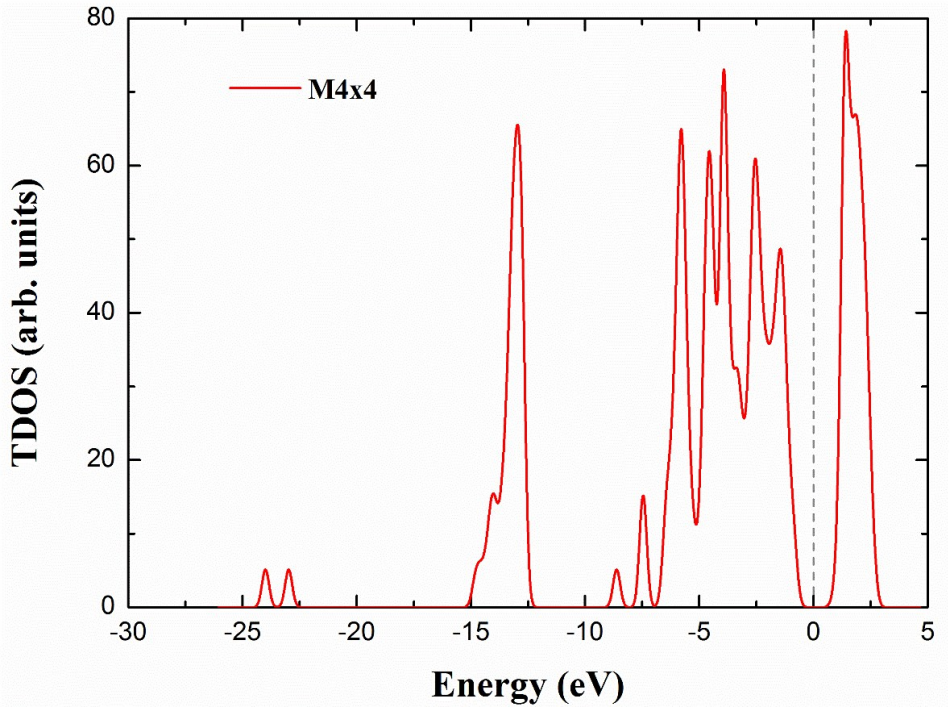

Fig. S14: Total density of state of MoS_2 bilayer with adsorbed CO_2 molecule.

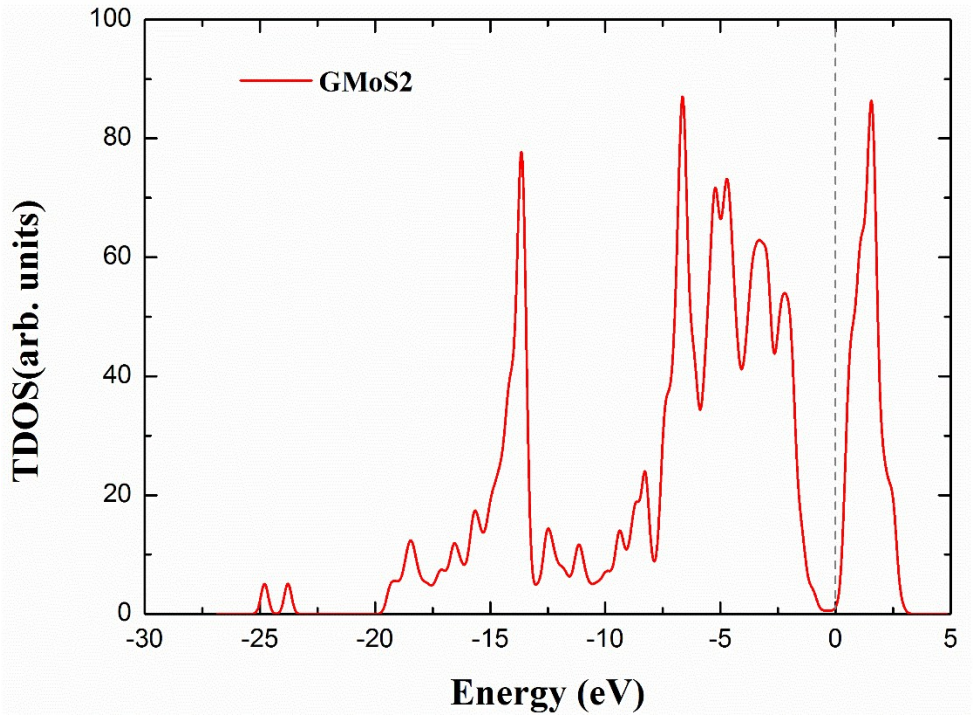

Fig. S15: Total density of state of MoS_2/Graphene bilayer heterostructure with adsorbed CO_2 molecule.

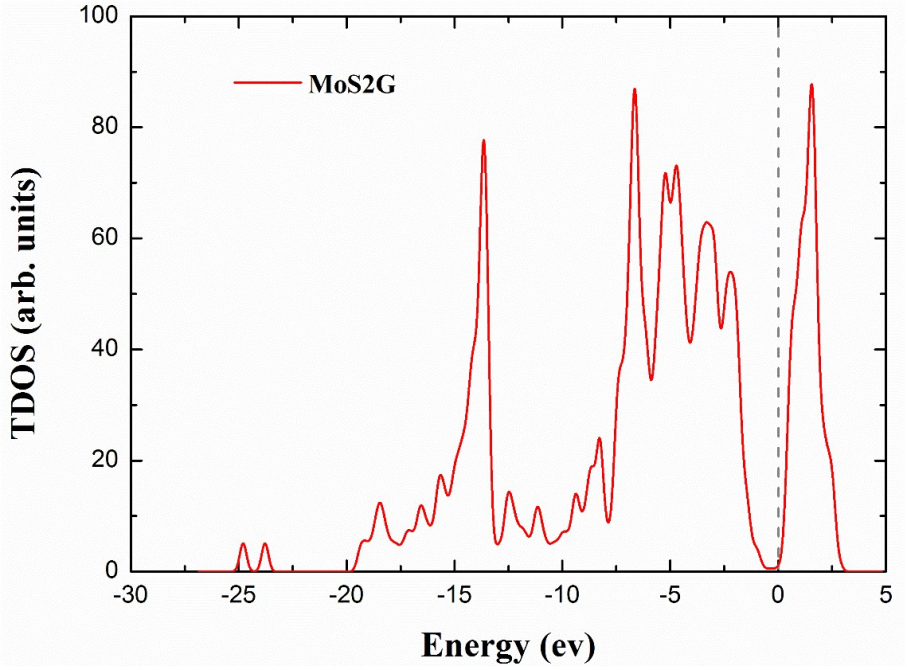

Fig. S16: Total density of state of MoS_2/Graphene bilayer heterostructure with adsorbed CO_2 molecule.

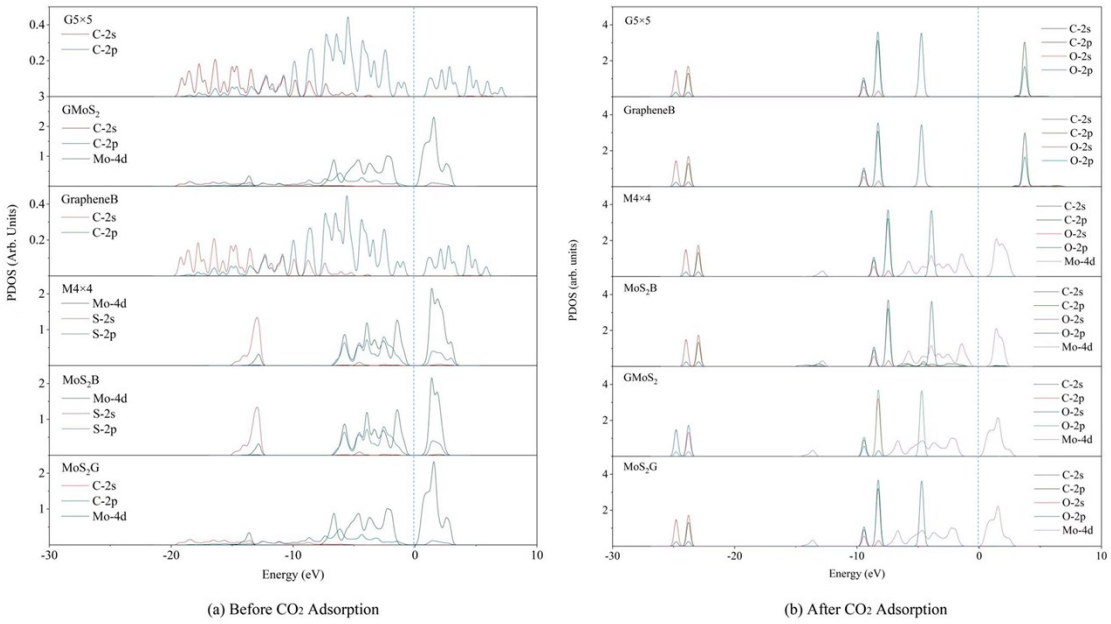

Figure S17: Projected DOS spectra before (a) and after (b) CO_2 adsorption. (G5×5) 5×5 supercell graphene monolayer, (M4×5) 4×4 supercell MoS_2 monolayer, (GrapheneB) graphene bilayer, (MoS_2B) MoS_2 bilayer, ($GMoS_2$) graphene/MoS_2 hybrid, and (MoS_2G) MoS_2/graphene hybrid. The dashed line represents the Fermi level.

Crystallographic Information Files (*.cif) of Graphene/MoS_2 Hybrids


GMoS2.cif


MoS2G.cif

Reference

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2. Liu, Y.; Wilcox, J., CO2 Adsorption on Carbon Models of Organic Constituents of Gas Shale and Coal. *Environ Sci Technol* **2011,** *Vol. 45* (No. 2), 809-814.
3. Li, J.; Hou, M.; Chen, Y.; Cen, W.; Chu, Y.; Yin, S., Enhanced CO 2 capture on graphene via N, S dual-doping. *Applied Surface Science* **2017,** *399*, 420-425.
4. Shijun Zhao; Jianming Xue; Kang, W., Gas adsorption on MoS2 monolayer from first-principles calculations. *Chemical Physics Letters* **2014,** *Vol. 595-596*, pp 35-42.