Electronic Supplementary Information

for

Graphene Thickness-controlled Photocatalysis and Surface-enhanced Raman Scattering

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Figure S-1: The XRD patterns of photoactive semiconductor panels of TiO_2 (a) and ZnO (b), corresponding to anatase TiO_2 (JCPDS 01-0562) and ZnO (JCPDS 89-0510) phases, respectively. No impure phase was observed.

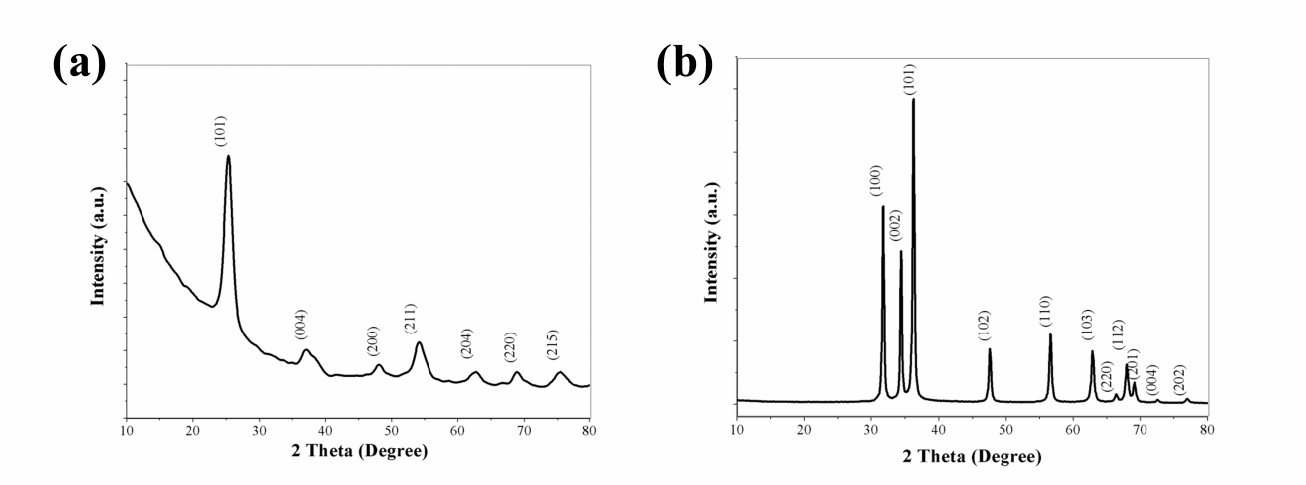


Figure S-2: The layer-by-layer assembly of graphene stacks on 300 nm SiO_2/Si substrates. The reason of using 300 nm SiO_2/Si substrates is for the clear visibility of different layers of graphene stacking. (a) The SLG deposited on the substrates clearly shows the light contrast with a specific shape of square. In (b), three layers of square-shaped graphene sheets stack on each other and generate the regions covered by one-layer (1L), two-layer (2L), and three-layer (3L) graphene indicated by arrows. No broken or discontinuous area is observed. These results clearly demonstrate that graphene stacking with well controlled coverage areas, sizes, shapes, and thicknesses can be precisely achieved by our layer-by-layer assembly.

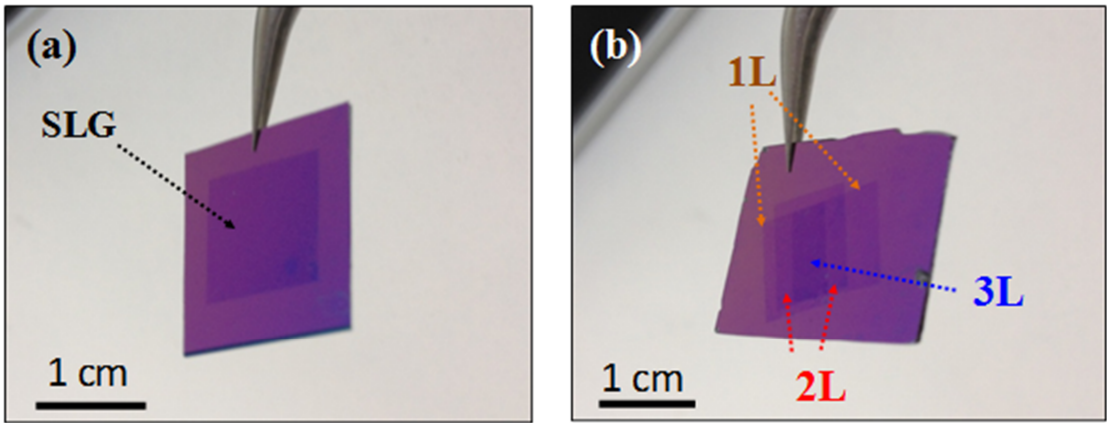


Figure S-3. AFM and TEM characterization of single-layer graphene produced by the CVD method. (a) AFM images of the graphene film with a thickness of 0.8 nm. (b) TEM images of transferred CVD graphene at the folded region showed the cross section of single-layer graphene. (c) The corresponding SAED pattern obtained from the graphene film in (b) showed a single set of diffraction spots with hexagonal array.

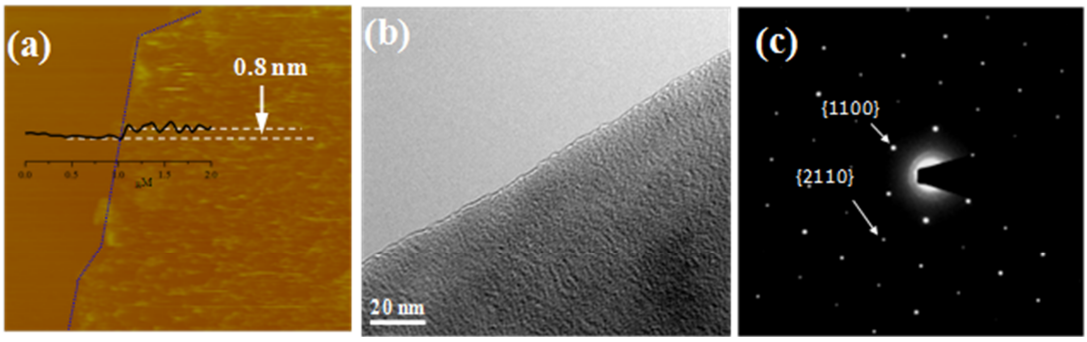


Figure S-4. The experimental setup for photodegradation tests in our photoreactor. The UV light emitted from the lamps can be fully reflected by the cylindrical mirror and focused to the center point, which is the location of GHPs with dye molecule solutions for photodegradation tests.

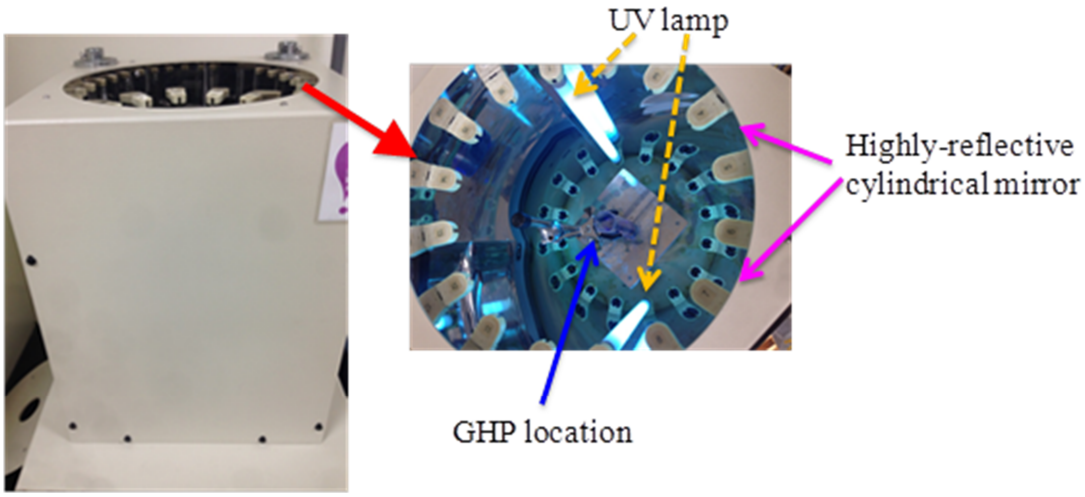
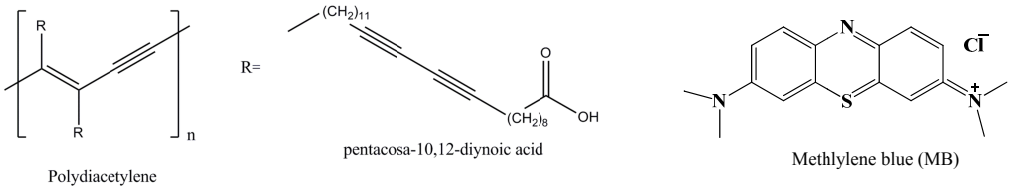
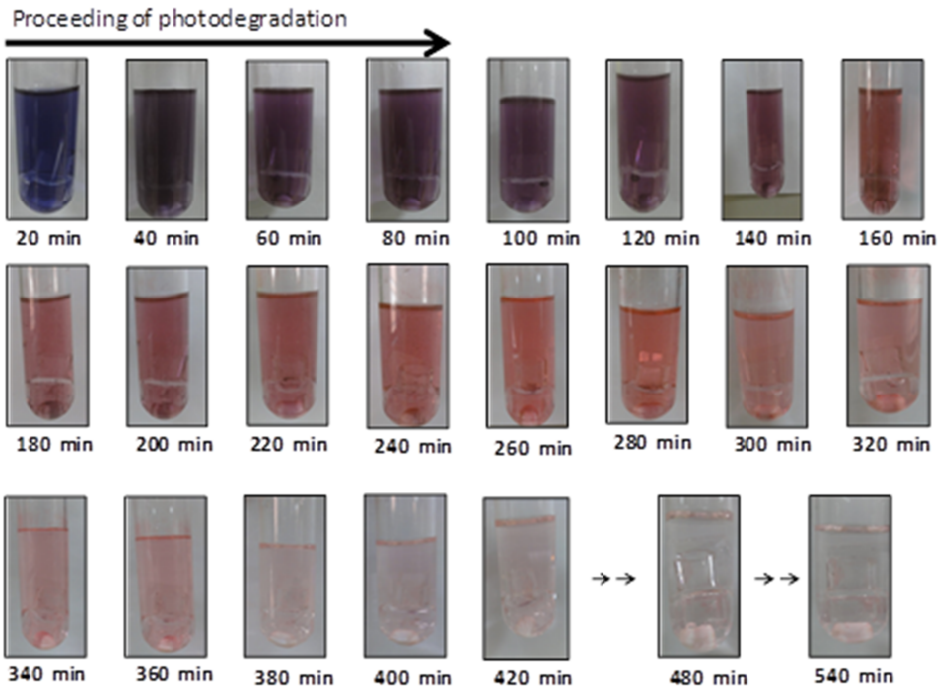


Figure S-5. The comparison of photodegradation efficiency of the polymer dye in the presence of various GHPs. The chemical structure of the polymer dye, polydiacetylene (PDA), and MB are shown in (a). The time profile of PDA photodegradation with bare TiO_2 panels is shown in (b). The total decomposition of PDA takes around 540 min with a three-stage color change (from deep blue to purple to pink). In (c), the results of PDA photodecomposition with different GHPs for 120 min were shown. By referencing the results in (b), the corresponding decomposition rate order is: **3L-GHP** > **5L-GHP** > **1L-GHP** > **7L-GHP** > TiO_2.

(a)



(b)



(c)

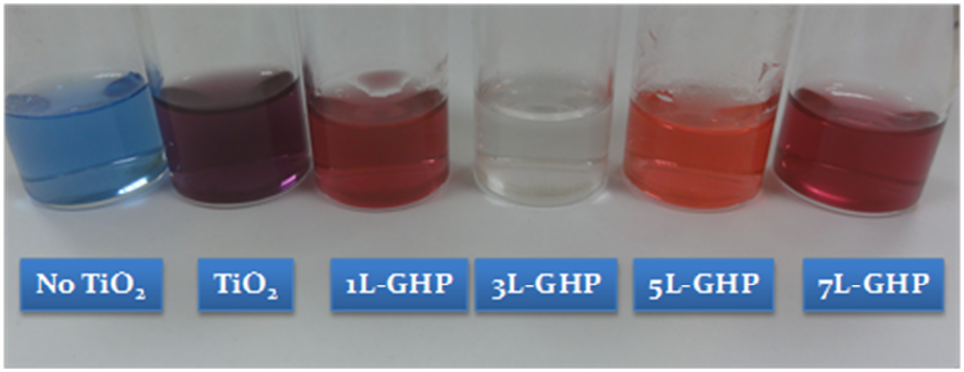


Figure S-6. (a) The MB photodegradation in the presence of GHPs (**1L-GHP** and **3L-GHP**) and their oxygen plasma treated products (**O-1L-GHP** and **O-3L-GHP**). (b) The corresponding rate constant plot for **1L-GHP** and **O-1L-GHP.**

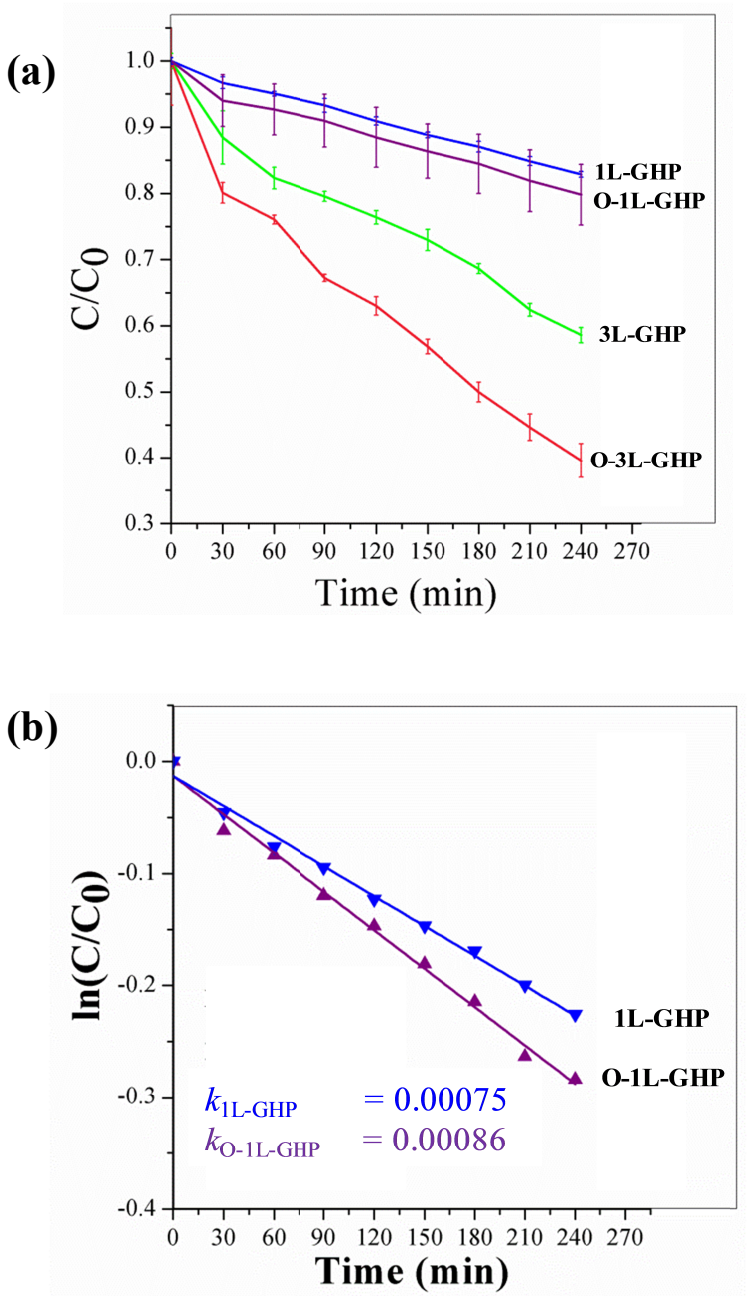


Figure S-7. The optical absorption spectra of GHPs. From (a) to (f) are results for **3L-GHP-4L**, **7L-GHP**, **5L-GHP**, **3L-GHP**, **1L-GHP,** bare TiO_2, and quartz, respectively.

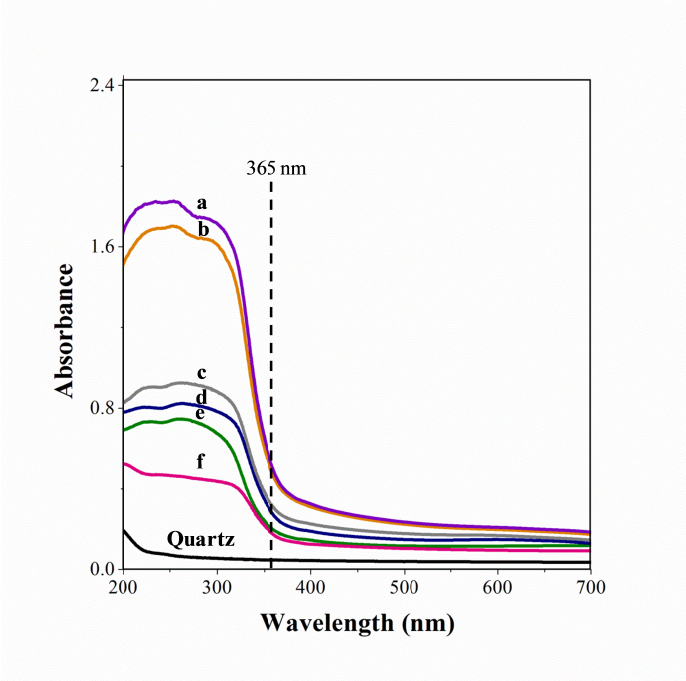


Figure S-8. The schematic presentation of photocatalytic and photoelectron scavenger (Au^{3+}) reduction mechanism of TiO_2 under UV light irradiation.[1]

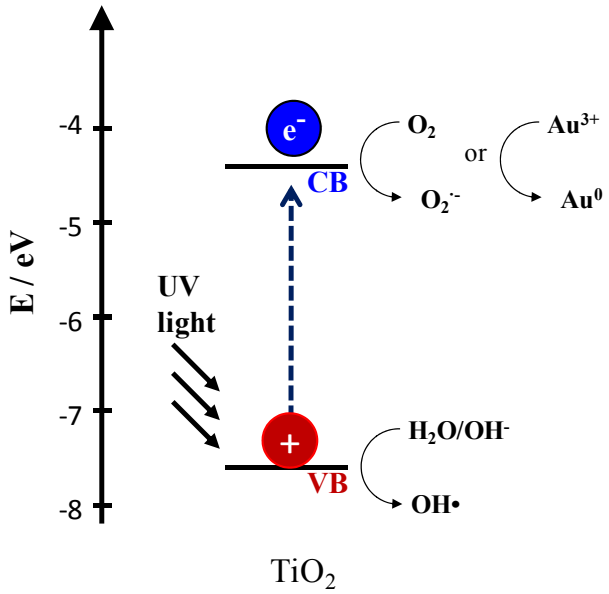
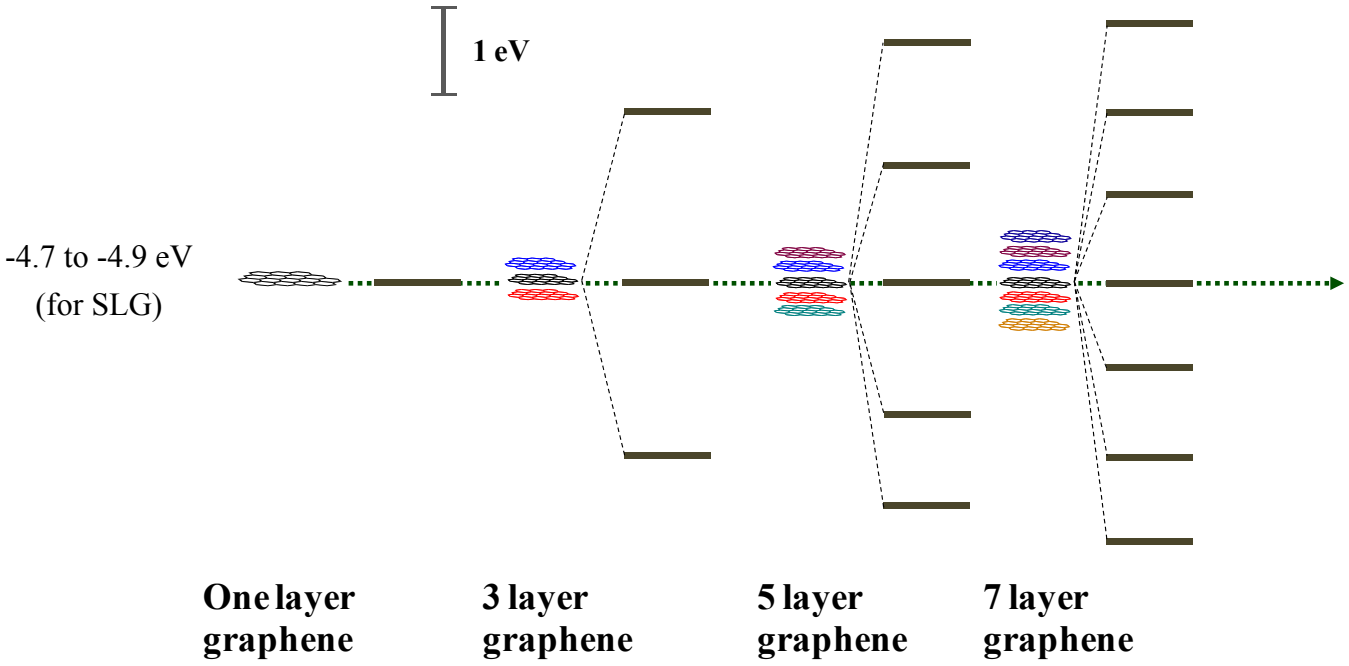


Figure S-9. The schematics of quantized energy levels of graphene sheet stacks with wave vectors perpendicular to graphene surface. The gaps of each energy levels in 1L, 3L, and 7L graphene were derived from the experimental results, reported by Hibino *et al.*,[2] of the reflectivity of low-energy electrons in 1 to 7 layer(s) of graphene films by low-energy electron microscopy (LEEM). All the upper energy levels above the CB of TiO_2 are not shown in scheme 1.



1. A. L. Linsebigler, G. Lu and J. T. Yates, *Chem. Rev.*, 1995, **95**, 735-758.
2. H. Hibino, H. Kageshima, F. Maeda, M. Nagase, Y. Kobayashi and H. Yamaguchi, *Phys. Rev. B*, 2008, **77**, 075413-075411.