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Electronic Supplementary Information (ESI)
For

Graphene Oxide Sheets as a Template for Molecular Assembly: Graphene Oxide Induce H-aggregates of Pyronin (Y) Dye

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Experimental Section

Synthesis of Graphene Oxide (GO):

In a typical synthesis of graphite oxide, 1.0 g of natural graphite flakes was dispersed in 50.0 mL of H_2SO_4 in a 500 mL erlenmeyer flask. Next, 1.0 g of $NaNO_3$ was added into the mixture under continuous stirring at 600 rpm. The erlenmeyer flask was placed into the ice-bath to decrease the reaction temperature in the range of 0-3^{o}C since an exothermic reaction occurs upon addition of $KMnO_4$ in the following step. Once everything has settled down, 6.0 g of $KMnO_4$ was added partially by controlling the temperature of the reaction via a thermometer placed into the reaction mixture. Then, the resulting mixture was stirred at 600 rpm for 30 min in the ice-bath. After the removal of ice-bath, the solution was heated to 35^{o}C and stirred for 3 h. A thick paste was formed after 3 h stirring at 35^{o}C. Then, the solution was placed into ice-bath again and 50.0 mL of water was added dropwise (causion, a severe exothermic reaction occurs upon addition of water into the reaction mixture and the reaction temperature may increase up to ~80 ^{o}C). A darkbrown slurry was formed after stirring of the solution for 30 min at room temperature. Finally, 100 mL of water and 8.0 ml of 30% H_2O_2 were added dropwise into the darkbrown slurry and the addition of 2.0 mL of excess H_2O_2 was followed until observation of a permanent yellow color indicating the complete oxidation of graphite. The resultant solution was filtered by suction filtration. Filtered product was washed with deionized water many times and dried under vacuum at 70 ^{o}C. The obtained graphite oxide was dispersed in water with a certain concentration and subsequently sonicated to give GO.

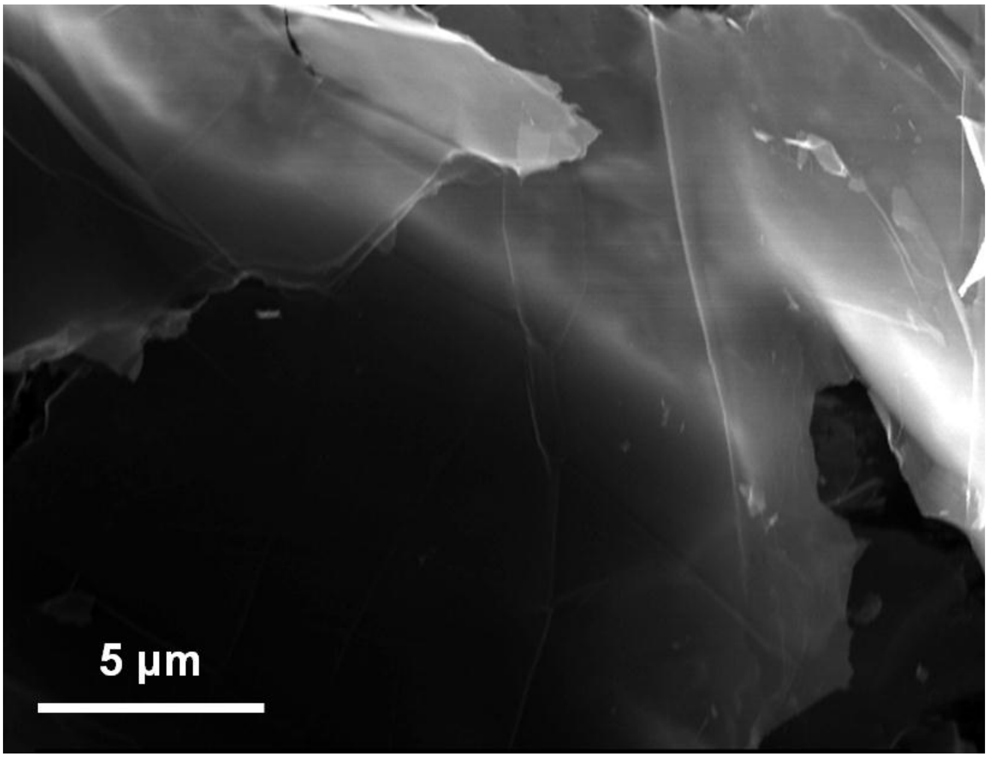
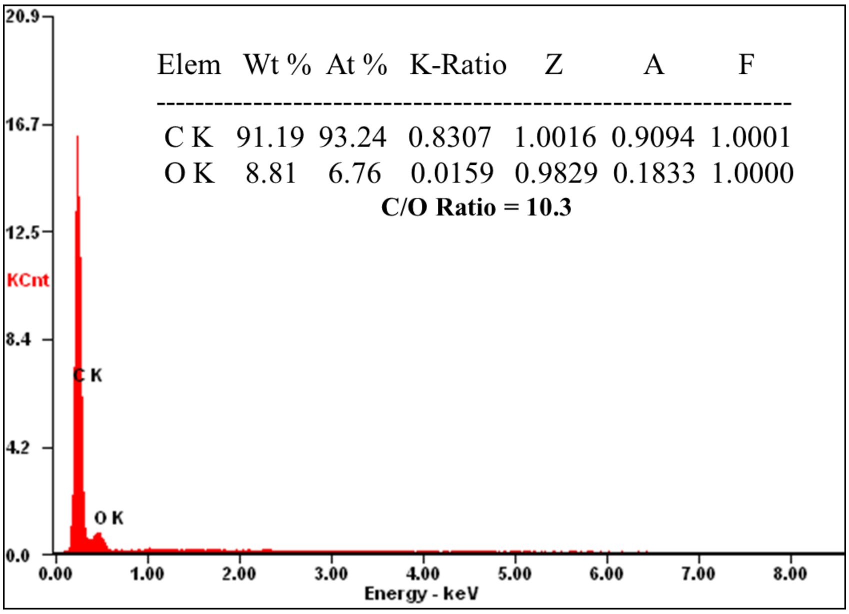


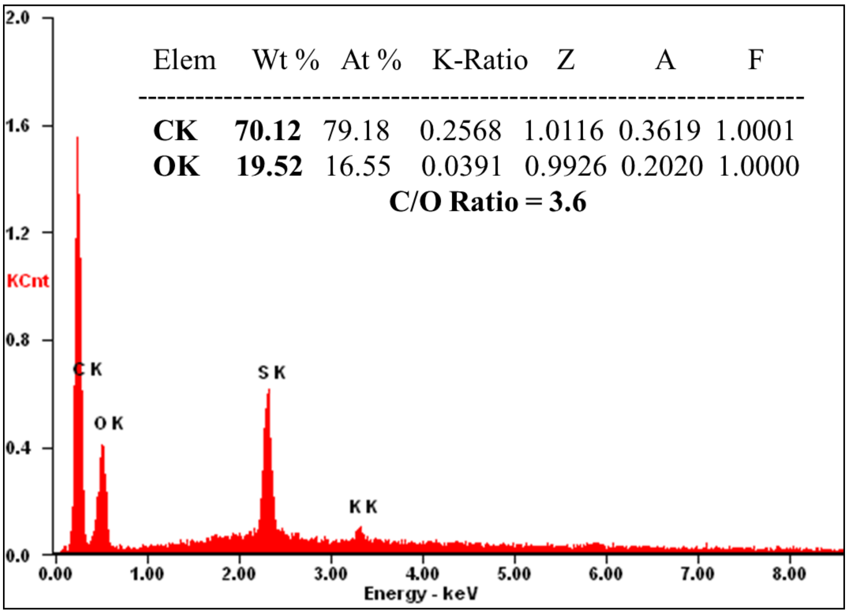
Fig. S1. SEM image of natural graphite flakes.



Elem	Wt %	At %	K-Ratio	Z	A	F
C K	91.19	93.24	0.8307	1.0016	0.9094	1.0001
O K	8.81	6.76	0.0159	0.9829	0.1833	1.0000

C/O Ratio = 10.3

Fig. S2. EDX spectrum of natural graphite flakes



Elem	Wt %	At %	K-Ratio	Z	A	F
CK	**70.12**	79.18	0.2568	1.0116	0.3619	1.0001
OK	**19.52**	16.55	0.0391	0.9926	0.2020	1.0000

C/O Ratio = 3.6

Fig. S3. EDX spectrum of GO

After oxidation processes of natural graphite in strong acidic medium, decreasing C/O ratio from10.3 to 3.6 shows the formation of GO as it can be seen in EDX spectra.

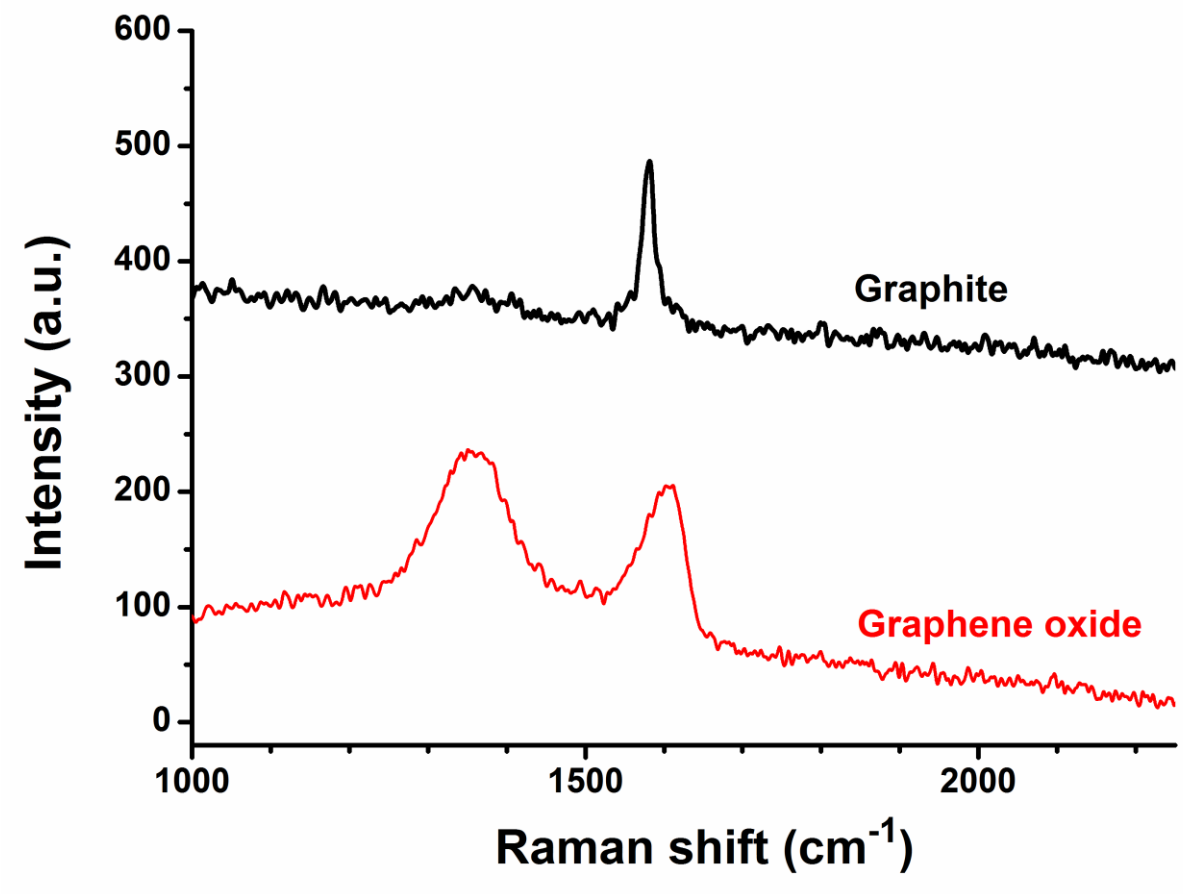


Fig. S4. Raman spectra of (a) Natural graphite flakes (b) GO

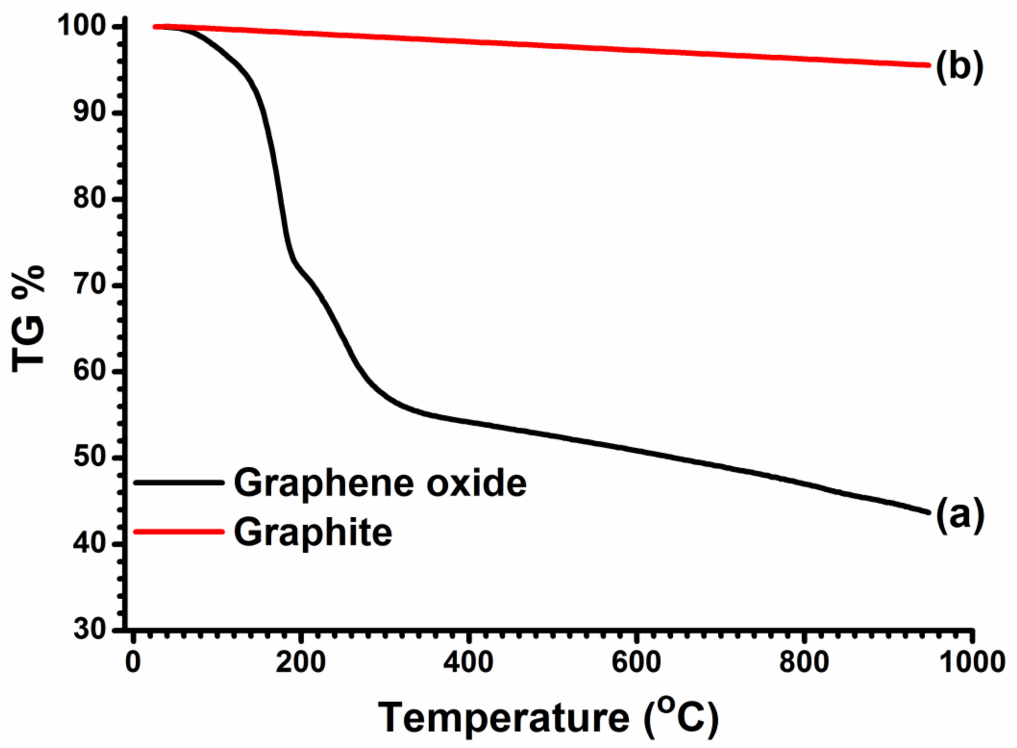
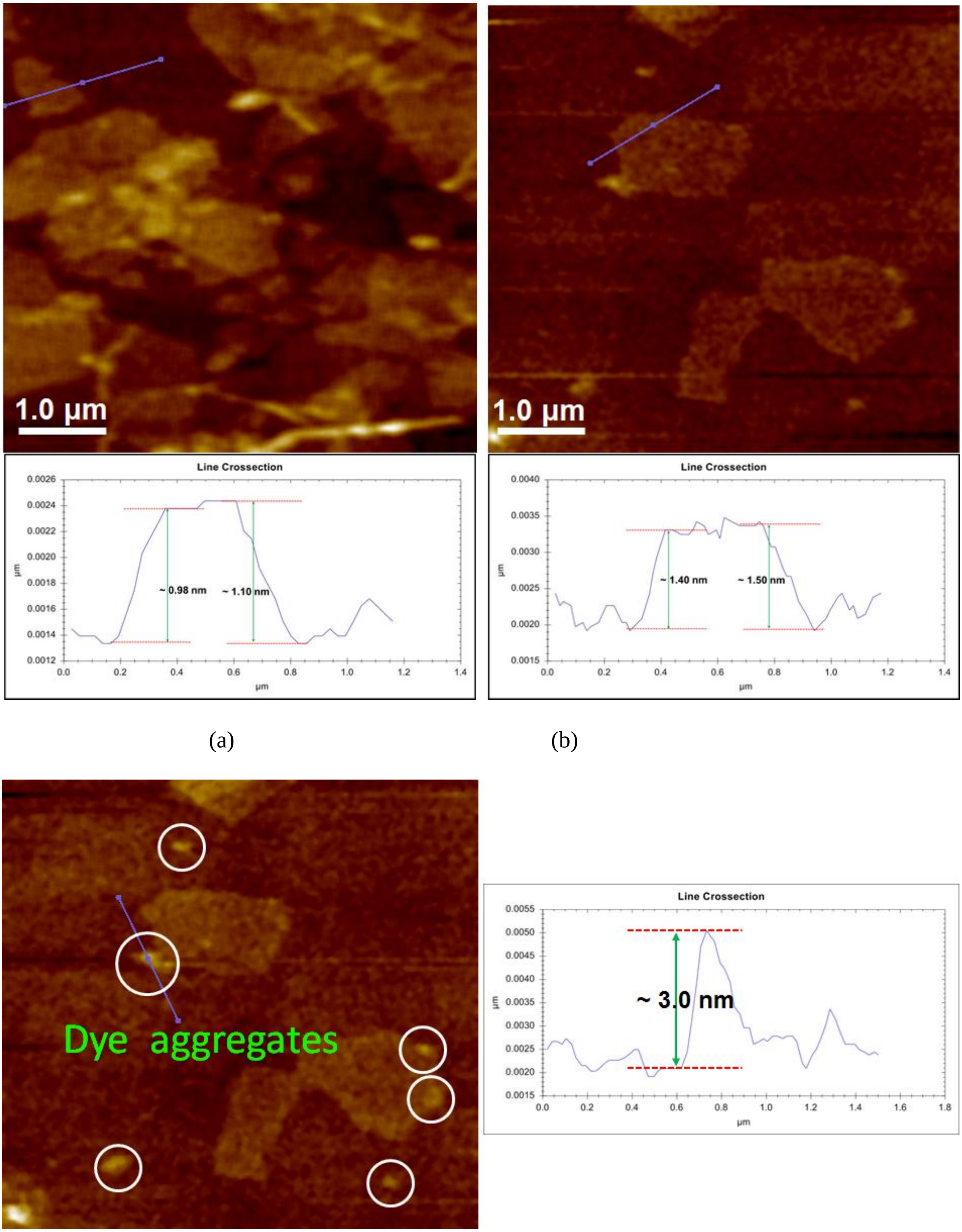


Fig. S5. TGA spectra of (a) GO (b) natural graphite flakes

Graphite flakes showed very low weight loss in the temperature range of 25-1000 ^{o}C. However, the GO showed a weight loss of 10% at 140 ^{o}C and then 30% at 200 ^{o}C corresponding to the removal of water and COOH groups, respectively. Next, the fast weight loss of GO took place until 360 ^{o}C due to the decomposition of the hydroxyl or epoxy groups. These results clearly reveal that the graphite flakes were effectively oxidized into the thin layered GO structure.



(a) (b)

(c)

Fig. S6. Cross-sectional analysis of AFM images of GO (a) and (b,c) PyY adsorbed GO on cleaved mica surface.

The GO and dye adsorbed GO samples were prepared by spin-coating on mica surface at 3000 rpm at 1 min. Figure S6 is the tapping mode AFM images of GO (a) and PyY-GO (b,c). As shown in cross–sectional analysis, the average thickness for GO sheets is found to be ~1.0 nm, whereas the GO–PyY composite show a higher step height at the edges (Figure S6c). This suggest the intense adsorption of PyY on the edges of GO.

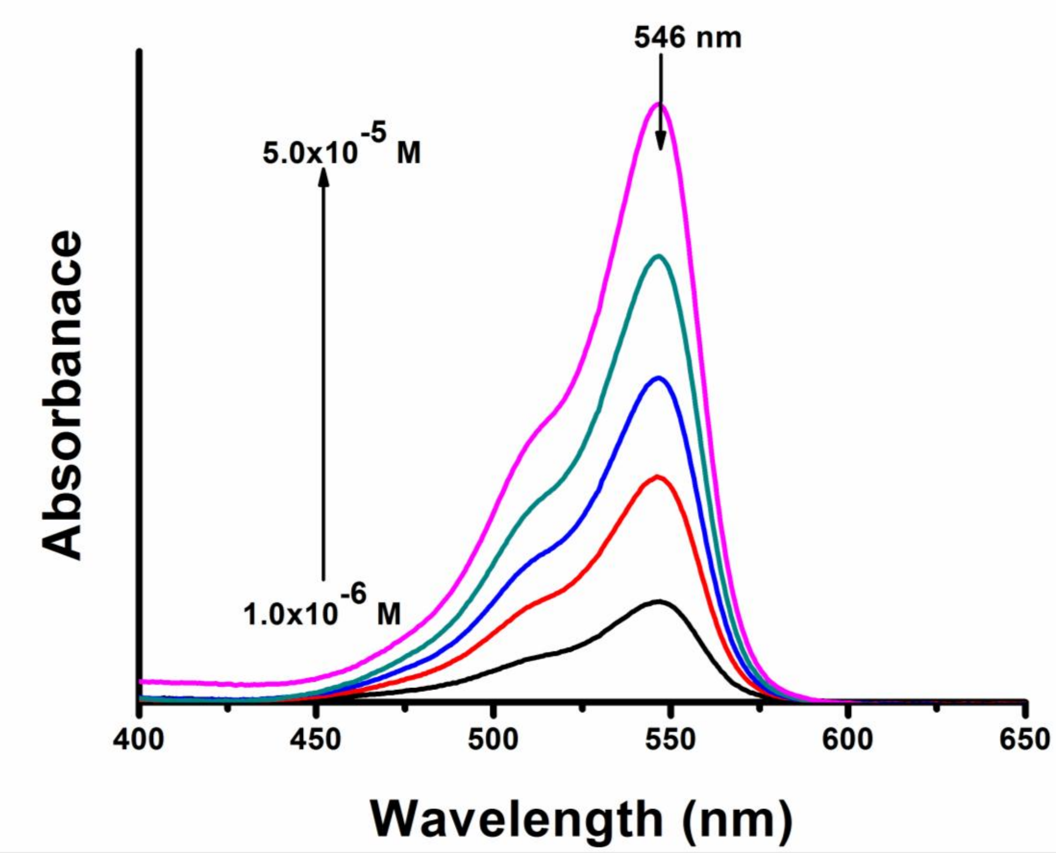


Fig. S7. Absorption spectra of PyY in deionized water at various dye concentrations.

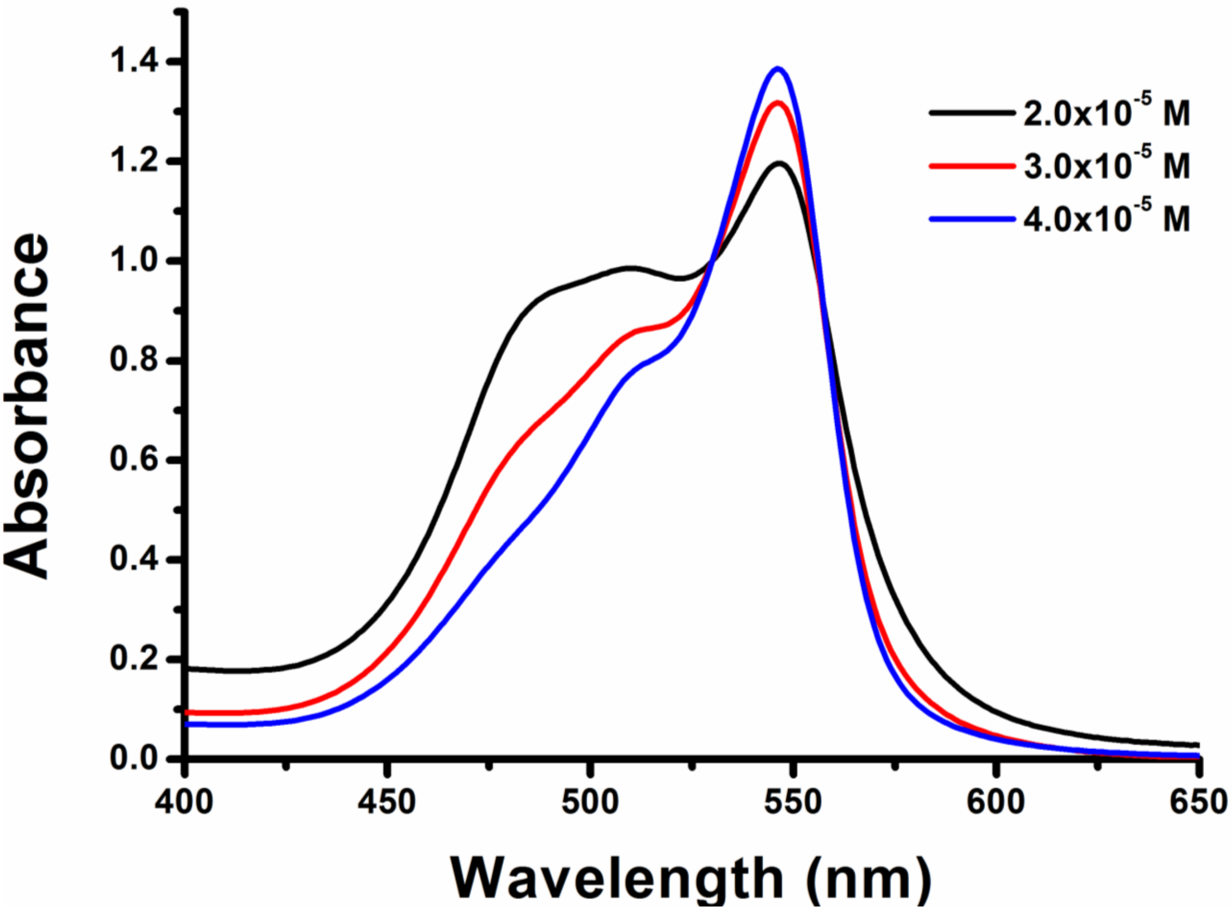


Fig. S8. Absorption spectra of PyY in 17 mg/L GO dipersion at various dye concentrations. (The spectra were normalized at the isosbestic point in order to show clearly the change of relative absorbance due to monomer and aggregates.)

The band maximum of PyY at higher concentration (>$1.5\text{x}10^{-5}$ M) are located at 546 nm as the monomer in water.

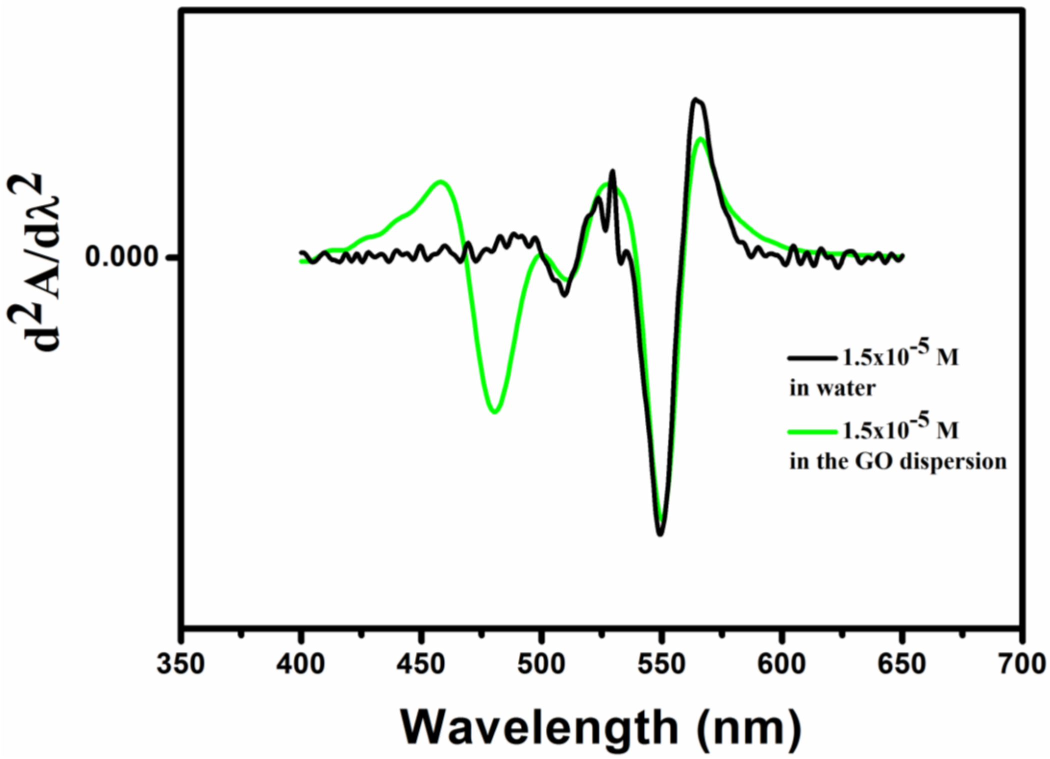


Fig. S9. Comparison of second derivative spectrum of 1.5x10^{-5} M PyY in water and the GO aqueous dispersion.

As can be seen from Fig. S9, the monomer maximum of PyY in water and the GO aqueous dispersion is 550 nm. This result implies that the absorption behaviors of the monomeric dye molecules in the GO aqueous dispersion resemble the monomer in water due to weak interaction of GO-PyY at higher dye concentration. This supports the presence of free PyY molecules (monomer) at higher dye concentration (>10^{-5} M).