Supplementary Information of
Highly Efficient Dye Adsorption and Removal: Functional
Hybrid of Reduced Graphene Oxide–Fe$_3$O$_4$ Nanoparticles as
An Easily Regenerative Adsorbent

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Figure S1. TEM of pure Fe$_3$O$_4$ NPs (a), as-prepared RGF-1 (b), as well as RhB adsorbed and then annealed (i.e., regenerated) RGF-1 (c). The preparation for the Fe$_3$O$_4$ NPs is almost the same as that for the RGO–Fe$_3$O$_4$ hybrid except that no GO is added.

Figure S2. SEM of pure Fe$_3$O$_4$ NPs (a), as-prepared RGF-1 (b), as well as RhB adsorbed and then annealed RGF-1 (c).
Figure S3. TEM of the hybrids with three different RGO/Fe$_3$O$_4$ ratios: (a) RGF-1, (b) RGF-2, and (c) RGF-3, where the C/Fe ratio are respectively estimated to be 1.7:1, 3:0:1, and 7.0:1 by XPS.

Figure S4. (a) Nitrogen adsorption–desorption isotherms of RGF-1, RGF-2, and RGF-3 obtained at 77 K, (b) pore size distributions of the three samples, together with those of the pure Fe$_3$O$_4$ NPs and the pure RGO.

Table S1. The respective C/Fe ratios obtained by XPS and BET surface areas calculated from Figure S4 for RGF-1, RGF-2, and RGF-3.

<table>
<thead>
<tr>
<th>Sample</th>
<th>C/Fe atomic ratio</th>
<th>BET surface area</th>
</tr>
</thead>
<tbody>
<tr>
<td>RGF-1</td>
<td>1.7 : 1</td>
<td>80.38 m$^2$/g</td>
</tr>
<tr>
<td>RGF-2</td>
<td>3.0 : 1</td>
<td>119.92 m$^2$/g</td>
</tr>
<tr>
<td>RGF-3</td>
<td>7.0 : 1</td>
<td>164.96 m$^2$/g</td>
</tr>
</tbody>
</table>
Figure S5. (a) Absorbance of RhB-adsorbed RGF-1 (which is extracted, washed and dried, and then redissolved in DI water) after step-by-step ethanol loading. (b) The absorbance at RhB’s characteristic absorption peak versus ethanol loading corresponding to (a).

Figure S6. The absorbance of RhB aqueous solution (black solid curve) and that after Fe$_3$O$_4$ NPs are added for 9 h (red dashed curve).

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Figure S7. Magnetic hysteresis loops ($M-H$ curves) for the as-prepared (black solid line) and regenerated (RhB-adsorbed and then annealed, red dash line) RGF-1 measured at 300 K. A saturation magnetization of ~75 emu/g is found for both samples, indicating the unchanged magnetic behavior through the regeneration process.

Figure S8. Calibration of the $K_b$ values of a series of dyes’ solutions. (a) RhB; (b) AB92; (c) R6G; (d) OII; (e) MG; (f) NC. For each dye, the $K_b$ is calibrated through measuring the absorbance of the dye’s solutions with a series of normal concentrations.
Figure S9. TGA curve for RGF-1 under inert atmosphere (nitrogen protecting). The effect of nitrogen gas desorption/re-adsorption during the heating/cooling process has been taken out by repeating the heating/cooling process for several cycles and measuring the TGA curves for each cycle.

Figure S10. The adsorption kinetics fitted by the pseudo-second-order adsorption equation for (a) equal-amount RGF-1 at the 1st and 2nd cycles, (b) equal-amount RGF-1 at different pH values (3.45, 7.55, and 11.45) while the initial adsorbate concentrations are kept the same (15.2 mmol/L), and (c) equal-amount RGF-1 at different adsorbate concentrations (11.4, 15.2, and 19.1 mmol/L) under a neutral pH condition of 7.55, respectively. RhB is served as the adsorbate for all the cases.
Figure S11. XPS N1s spectral scan for as-prepared RGF-1 (black hollow squares) and for that after one adsorption-regeneration cycle (experimental curve: the red solid line; fitting curve: the blue dashed line). For better comparison, each spectrum has been normalized to the corresponding integrated intensity of C1s peak. It is clear that the relative N content is more in the regenerated hybrid than that in the as-prepared one.

Figure S12. (a) The adsorption kinetics of RGF-1, RGF-2 and RGF-3 for RhB. (b) The corresponding adsorption kinetics fitted by the pseudo-second-order adsorption equation. The initial adsorbate (RhB) concentration is 18.3 mmol/L while the pH value of the solution is 7.55.
Figure S13. Absorbance of the three dyes’ (NCP, AB92, and MG) “cocktail” against different RGF-1 loading (from the topmost curve to the undermost one: 0, 0.25, 0.5 and 0.75 mg of RGF-1 are respectively loaded).