Supporting information for

Hydrophilic and strengthened 3D reduced graphene oxide/nano-
Fe$_3$O$_4$ hybrid hydrogel for enhanced adsorption and catalytic
oxidation of typical pharmaceuticals

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Table S1 Kinetic parameters for the adsorption of CIP and TC on 3D-rGO/Fe₃O₄

<table>
<thead>
<tr>
<th>Adsorbate</th>
<th>k (g/(μmol·h))</th>
<th>qₑ (μmol/g)</th>
<th>v₀ (μmol/(g·h))</th>
<th>R²</th>
</tr>
</thead>
<tbody>
<tr>
<td>CIP</td>
<td>0.00026</td>
<td>546.3</td>
<td>77.6</td>
<td>0.979</td>
</tr>
<tr>
<td>TC</td>
<td>0.00031</td>
<td>556.7</td>
<td>96.1</td>
<td>0.962</td>
</tr>
</tbody>
</table>

Table S2 Calculated parameter values of the Langmuir model for CIP and TC adsorption on 3D-rGO/Fe₃O₄, 3D-rGO, GO, rGO and EG in the whole concentration range

<table>
<thead>
<tr>
<th>Adsorbents</th>
<th>Adsorbates</th>
<th>Langmuir model a</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>b (L/μmol)</td>
<td>qₘ (mmol/g)</td>
</tr>
<tr>
<td>3D-rGO/Fe₃O₄</td>
<td>CIP</td>
<td>0.005</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.007</td>
</tr>
<tr>
<td>3D-rGO</td>
<td>CIP</td>
<td>0.018</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.028</td>
</tr>
<tr>
<td>GO</td>
<td>CIP</td>
<td>3.285</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.142</td>
</tr>
<tr>
<td>rGO</td>
<td>CIP</td>
<td>7.664</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.316</td>
</tr>
<tr>
<td>EG</td>
<td>CIP</td>
<td>0.219</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.137</td>
</tr>
</tbody>
</table>

a qₑ = qₘ Cₑ/(1/b + Cₑ)
**Table S3** Calculated parameter values of the Langmuir model for CIP and TC adsorption on 3D-rGO/Fe$_3$O$_4$, 3D-rGO, GO, rGO and EG at equilibrium concentrations below 100 μmol/L

<table>
<thead>
<tr>
<th>Adsorbents</th>
<th>Adsorbates</th>
<th>b (L/μmol)</th>
<th>$q_m$ (μmol/g)</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3D-rGO/Fe$_3$O$_4$</td>
<td>CIP</td>
<td>0.116</td>
<td>842.9</td>
<td>0.781</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.022</td>
<td>1891.6</td>
<td>0.994</td>
</tr>
<tr>
<td>3D-rGO</td>
<td>CIP</td>
<td>0.065</td>
<td>596.3</td>
<td>0.937</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.097</td>
<td>623.8</td>
<td>0.991</td>
</tr>
<tr>
<td>GO</td>
<td>CIP</td>
<td>1.599</td>
<td>567.6</td>
<td>0.861</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.168</td>
<td>557.2</td>
<td>0.973</td>
</tr>
<tr>
<td>rGO</td>
<td>CIP</td>
<td>6.178</td>
<td>237.6</td>
<td>0.965</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.439</td>
<td>115.2</td>
<td>0.913</td>
</tr>
<tr>
<td>EG</td>
<td>CIP</td>
<td>0.221</td>
<td>71.2</td>
<td>0.826</td>
</tr>
<tr>
<td></td>
<td>TC</td>
<td>0.143</td>
<td>73.7</td>
<td>0.599</td>
</tr>
</tbody>
</table>

\[ q_e = q_m C_e/(1/b+C_e) \]

**Table S4** Surface area and pore volume of 3D-rGO/Fe$_3$O$_4$, 3D-rGO, GO, rGO and EG

<table>
<thead>
<tr>
<th>Sample</th>
<th>Specific surface area (m$^2$/g) $^a$</th>
<th>Total pore volume (cm$^3$/g) $^b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3D-rGO/Fe$_3$O$_4$</td>
<td>66.8</td>
<td>0.285</td>
</tr>
<tr>
<td>3D-rGO</td>
<td>55.8</td>
<td>0.214</td>
</tr>
<tr>
<td>GO</td>
<td>15.0</td>
<td>0.017</td>
</tr>
<tr>
<td>rGO</td>
<td>24.1</td>
<td>0.011</td>
</tr>
<tr>
<td>EG</td>
<td>123.2</td>
<td>0.097</td>
</tr>
</tbody>
</table>

$^a$ The specific surface area was calculated by the BET method.

$^b$ The pore volume was calculated by the DFT method.
Table S5  Specific surface area and pore volume of different activated carbons

<table>
<thead>
<tr>
<th>Parameters</th>
<th>AC-1</th>
<th>AAC</th>
<th>CAC</th>
<th>AC-2</th>
</tr>
</thead>
<tbody>
<tr>
<td>SSA (m$^2$/g)$^a$</td>
<td>930.5</td>
<td>761.3</td>
<td>545.9</td>
<td>323.7</td>
</tr>
<tr>
<td>Pore volume (cm$^3$/g)$^b$</td>
<td>0.597</td>
<td>0.496</td>
<td>0.353</td>
<td>0.353</td>
</tr>
</tbody>
</table>

$^a$ The specific surface area was calculated by the BET method.

$^b$ The pore volume was calculated by the DFT method.

Fig. S1  Mass ratios of 3D-rGO/Fe$_3$O$_4$ to 3D-rGO prepared at different concentrations of Fe$^{2+}$.

Fig. S2  Raman spectra of GO (a) and 3D-rGO/Fe$_3$O$_4$ prepared by 0.0125 (b), 0.05 (c) and 0.25 mol/L Fe$^{2+}$(d).
Fig. S3 Adsorption of CIP and TC on the hydrogels prepared by different reductants.

Fig. S4 Strain-stress curves of untreated and strengthened 3D-rGO/Fe$_3$O$_4$ before and after drying for 1 h.

Fig. S5 XRD pattern (a) and hysteresis curve (b) of 3D-rGO/Fe$_3$O$_4$. 
**Fig. S6** Contact angles of 3D-rGO/Fe$_3$O$_4$ (a), 3D-rGO (reduced by NaHSO$_3$) (b) and corresponding freeze-dried 3D-rGO (c).

**Fig. S7** FTIR (a) and Raman spectra (b) of GO and 3D-rGO/Fe$_3$O$_4$.

**Fig. S8** XPS spectra of GO (a), 3D-rGO/Fe$_3$O$_4$ (b) and core-level Fe2p spectrum of 3D-rGO/Fe$_3$O$_4$. 
Fig. S9 Pore size distributions of 3D-rGO/Fe₃O₄ prepared at 0.0125 and 0.25 mol/L Fe²⁺ after 1 h heating.

Fig. S10 Adsorption kinetics of CIP and TC on 3D-rGO/Fe₃O₄.

Fig. S11 Adsorption isotherms of CIP (a) and TC (b) on 3D-rGO/Fe₃O₄, 3D-rGO, GO, rGO and EG at the equilibrium concentrations below 100 µmol/L.
**Fig. S12** Pore size distribution of different ACs.

**Fig. S13** TOC concentrations in CIP and TC solutions by Fenton-like (after adsorption), Fenton and H₂O₂ oxidation for 4 h.

**Fig. S14** XRD patterns of 3D-rGO/Fe₃O₄ and re-3D-rGO/Fe₃O₄ (a), as well as FTIR spectra of GO, 3D-rGO/Fe₃O₄ and re-3D-rGO/Fe₃O₄ (b).
Fig. S15 Concentration of Fe in solution in the Fenton-like regeneration process.