Supporting Information

Chemodynamic/photothermal synergistic therapy based on Ce-doped Cu-Al layered double hydroxide

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Scheme S1. Schematic illustration of the bottom-up method of synthesizing CuAlCe-LDH.

Table S1. The feed ratio and the actual ratio of LDH determined by ICP.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Feed ratio</th>
<th>Actual ratio</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu Al-LDH</td>
<td>2 : 1</td>
<td>2.47 : 1</td>
<td>this work</td>
</tr>
<tr>
<td>Cu Al Ce-LDH</td>
<td>2 : 0.5    : 0.5</td>
<td>2.32 : 0.57 : 0.43</td>
<td>this work</td>
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<tr>
<td>Cu Al Ce-LDH</td>
<td>2 : 0.67   : 0.33</td>
<td>2.21 : 0.65 : 0.34</td>
<td>this work</td>
</tr>
<tr>
<td>Cu Al Ce-LDH</td>
<td>2 : 0.75   : 0.25</td>
<td>2.37 : 0.75 : 0.28</td>
<td>this work</td>
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<tr>
<td>Mg Al Ce-LDH</td>
<td>3 : 0.8    : 0.2</td>
<td>0.73 : 0.24 : 0.014</td>
<td>1, 2</td>
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<tr>
<td>Ni Fe Ce-LDH</td>
<td></td>
<td>doping 5% Ce</td>
<td>3</td>
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</table>

Table S2. The $K_M$ and $V_{max}$ values of different Fenton catalysts.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$K_M$ (mM)</th>
<th>$V_{max}$ (M·s$^{-1}$)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>ICG/CuAlCe-LDH</td>
<td>1.57</td>
<td>4.88×10$^{-6}$</td>
<td>this work</td>
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<tr>
<td>FeAl-LDH</td>
<td>0.16</td>
<td>1.47×10$^{-6}$</td>
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<td>PEG/Fe-LDHs</td>
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<td>1.76×10$^{-6}$</td>
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<td>Fe$_3$O$_4$ NPs</td>
<td>26.08</td>
<td>6.17×10$^{-8}$</td>
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<td>Mn-NS</td>
<td>26.40</td>
<td>7.04×10$^{-8}$</td>
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<td>Fe$_3$O$_4$@PPy@GOD NCs</td>
<td>4.94</td>
<td>1.13×10$^{-8}$</td>
<td>7</td>
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</tbody>
</table>

Fig. S1 XRD pattern of CuAl-LDH (2:1) nanosheets after restocking.
Fig. S2 H$_2$O$_2$ reacts with CuAl-LDH and CuAlCe-LDH to oxidate TMB at pH=6.5, and the absorbance (650 nm) of reactants is determined via UV spectrum.

Fig. S3 TEM image of CAC-LDH nanosheets with corresponding EDX mapping images for Cu, Al, Ce and O, respectively.
Fig. S4 The hydrodynamic size of CAC-LDH.

Fig. S5 Zeta potential of ICG and CAC-LDH.

Fig. S6 The UV–vis–NIR of ICG aqueous solution before and after adsorption with CAC-LDH.
Fig. S7 FTIR spectra of CAC-LDH, ICG and ICG/CAC-LDH, respectively.

Fig. S8 (A) TEM and (B) AFM of ICG/CAC-LDH.

Fig. S9 Size distribution of ICG/CAC-LDH in water, PBS, and culture medium (DMEM).
Fig. S10 UV–vis–NIR spectra of ICG, CAC-LDH and ICG/CAC-LDH, respectively.

Fig. S11 Release profiles of copper (A) and cerium (B) from ICG/CAC-LDHs under various conditions. Error bars represented for standard deviation, \( n = 3 \).

Fig. S12 Cu (I) detected by the selective sequestering agent neocuproine.
Fig. S13 FL spectra of terephthalate (TA) oxidized by ·OH generated from the reactions between ICG/CAC-LDH and H$_2$O$_2$: (A) without 808 nm laser irradiation; (B) with 808 nm laser irradiation.

Fig. S14 TEM images of CAC-LDH after different treatments for various periods of time.
Fig. S15 Mass extinction coefficient of ICG (A) and ICG/CAC-LDH (B) at 808 nm. Normalized absorbance intensity at λ = 808 nm divided by the characteristic length of cell (A/L) at varying concentrations.

Fig. S16 Photostability tests of ICG and ICG/CAC-LDH for three cycles.
**Fig. S17** Normalized absorbance of ICG and ICG/CAC-LDH at 808 nm in solutions at different pH values with H$_2$O$_2$ (0.1 mM).

**Fig. S18** Cytotoxicity tests with different concentrations of CuAlCe-LDH and H$_2$O$_2$ in different pH conditions.

**Fig. S19** Relative viabilities of HepG2 cells after incubated with ICG and ICG/CAC-LDH at various concentrations (quantified by ICG: 0, 5, 10, 15, 20, 25 $\mu$g·mL$^{-1}$) at pH 6.5 with 808 nm laser irradiation.

**Fig. S20** GSH content of HepG2 cells treated with different concentrations of CAC-LDH (0–50 $\mu$g·mL$^{-1}$) for 24 h.
**Fig. S21** ROS levels of DCFH-DA stained HepG2 cells with different treatments.

**Fig. S22** Linear relationship between PA signal and ICG/CAC-LDH concentration under the conditions of (A) GSH (1 mM) and (B) H$_2$O$_2$ (0.1 mM). Linear relationship between T$_1$-MR signal and Cu(II) concentration under the conditions of (C) GSH (1 mM) and (D) H$_2$O$_2$ (0.1 mM).

**REFERENCES**