Supporting Information

A General Strategy for Ligand Exchange on Upconversion Nanoparticles

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**Figure S1.** XRD pattern of the as-synthesized NaYF$_4$: Yb/Er (38/2 mol%)@NaYF$_4$ nanoparticles.
Figure S2. A comparison of FTIR spectra for PAA-modified upconversion nanoparticles prepared with and without adjusting pH of the ligand solution. Without pH adjustment, the absorption peak of C=O vibration appears, which indicated the existence of –COOH groups. By adjusting the pH to 8, the absorption peak due to C=O vibration disappeared, indicating that the –COOH groups were transformed into –COO⁻ and coordinated with the nanoparticles.
Figure S3. TG curves for the upconversion nanoparticles modified with different types of ligands.
Figure S4. A comparison of upconversion emission spectra for the ligand-free upconversion nanoparticles and the different ligand-coated counterparts. The spectra were obtained from aqueous dispersion of the nanoparticles (12.5 mM) under 980 nm diode laser excitation at 1 W.
Figure S5. $^1$H NMR spectra of different kinds of biomolecules after solvothermal processing. The signals inside dashed blue squares for cysteine were due to impurities from the raw molecules.
Figure S6. A comparison of upconversion emission spectra for biotinylated upconversion nanoparticles and ligand-free nanoparticles after incubation with 60 µL of streptavidin-Cy3 (1 mg mL⁻¹). The spectrum of biotinylated nanoparticles alone is also shown for reference.
Table S1. The effect of experimental procedure on dispersibility of the PAA-modified nanoparticles.

<table>
<thead>
<tr>
<th>#</th>
<th>Experimental Procedure</th>
</tr>
</thead>
</table>
| 1  | ① Adjust pH of ligand solution  
    ② Add uncoated nanoparticles to ligand solution  
    ③ Solvothermal treatment |
| 2  | ① Adjust pH of ligand solution  
    ② Add uncoated nanoparticles to ligand solution  
    ⊙ Solvothermal treatment |
| 3  | ⊙ Adjust pH of ligand solution  
    ① Add uncoated nanoparticles to ligand solution  
    ⊙ Solvothermal treatment |
| 4  | ① Add uncoated nanoparticles to ligand solution  
    ② Adjust pH of mixture  
    ③ Solvothermal treatment |

Ligand-free nanoparticles at pH=8
Table S2. Experimental variables for coating different ligands and quantitative characterizations of the resulting nanoparticles.

<table>
<thead>
<tr>
<th>#</th>
<th>Ligand</th>
<th>Parameters for Ligands</th>
<th>Characterizations</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Amount of Ligand</td>
<td>pH of Solution</td>
</tr>
<tr>
<td>1</td>
<td>PAA</td>
<td>50 mg</td>
<td>8</td>
</tr>
<tr>
<td>2</td>
<td>PEI</td>
<td>50 mg</td>
<td>11.8</td>
</tr>
<tr>
<td>3</td>
<td>PVP</td>
<td>50 mg</td>
<td>8</td>
</tr>
<tr>
<td>4</td>
<td>Cysteine</td>
<td>100 mg</td>
<td>8</td>
</tr>
<tr>
<td>5</td>
<td>Glycine</td>
<td>100 mg</td>
<td>8</td>
</tr>
<tr>
<td>6</td>
<td>Citric Acid</td>
<td>100 mg</td>
<td>8</td>
</tr>
<tr>
<td>7</td>
<td>Biotin</td>
<td>50 mg</td>
<td>8</td>
</tr>
<tr>
<td>8</td>
<td>AEP</td>
<td>100 mg</td>
<td>8</td>
</tr>
</tbody>
</table>

\(^{a}\) The zeta potential of ligand-free upconversion nanoparticles is 33.6 mV.