Facile Preparation of Multifunctionalised ‘Stealth’ Upconverting Nanoparticles for Biomedical Applications


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Figure S1. Bright-field TEM images of (a) OA-, (b) alendronate-, (c) OPLT-, and (d) PEG-phosphate-coated NaYF₄:Nd³⁺/Yb³⁺/Er³⁺ (1/20/2%)@NaYF₄:Nd³⁺ (25%) core-shell UCNPs. The relative ratio of dopants in the NPs is given as mol%, as obtained by inductively coupled plasma mass spectrometry (ICP-MS) analysis on the OA-coated hydrophobic UCNPs. Transmission electron microscopy (TEM) images were acquired using an image Cs-corrected FEI Titan 80-300 electron microscope, operating at an accelerating voltage of 300 kV. Samples were prepared by drop-casting NP dispersions on carbon-coated TEM grids followed by drying in air.
Figure S2. Hydrodynamic diameter ($D_h$) of (a) OA-coated, (b) alendronate-, (b) PEG-phosphate, and (d) OPLT-coated NaYF$_4$:$\text{Nd}^{3+}$/Yb$^{3+}$/Er$^{3+}$ (1/20/2%)@NaYF$_4$:$\text{Nd}^{3+}$(25%) core-shell UCNPs, as measured by DLS (intensity-weighted size distributions, pH 7.4).

Figure S3. Digital photographs of alendronate-coated NaYF$_4$:$\text{Nd}^{3+}$/Yb$^{3+}$/Er$^{3+}$ (1/20/2%)@NaYF$_4$:$\text{Nd}^{3+}$(25%) core-shell UCNPs in fetal bovine serum (left) and in PBS buffer (right) after laser excitation at $\lambda = 793$ nm (cw diode laser, power output 100 mW).
Figure S4. Upconversion emission from colloidal dispersions of (a) alendronate-, (b) PEG-phosphate-, and (c) OPLT-coated NaYF$_4$:Nd$^{3+}$/Yb$^{3+}$/Er$^{3+}$ (1/20/2%)@NaYF$_4$:Nd$^{3+}$(25%) core-shell UCNPs in dd water ([UCNP] = 6 mg/mL) after laser excitation at $\lambda =$ 793 nm (cw diode laser, power output 100 mW).
Figure S5. SDS-PAGE analysis of the biomolecular corona formation on OPLT-coated NaYF₄:Nd³⁺/Yb³⁺/Er³⁺ (1/20/2%)@NaYF₄:Nd³⁺ (25%) core-shell UCNPs. Surface associated biomolecules were isolated after incubation with increasing concentrations of (a) heat-inactivated human serum for 1 h at 37 °C, and (b) human serum without pre-treatment for 1 h at 4 °C (temperature maintained by keeping on ice).
Figure S6. TLC analysis of alendronate-coated UCNPs after coupling reaction with fluorescein isothiocyanate (FITC). In a room temperature reaction, (I) 5, (II) 10 or (III) 20 nmol of FITC was coupled with 2 mg of UCNPs, and the samples were analysed by TLC before (a) and after (b) purification. For comparison, 20 nmol of FITC (CTRL) were also analysed. Fluorescence signals from the fluorescein unit were detected by using Amersham Typhoon 5 Scanner.
Table S1. Intensity-weighted DLS particle size distributions ($D_h$) for OPLT-, alendronate-, and PEG-phosphate-coated NaYF$_4$:Nd$^{3+}$/Yb$^{3+}$/Er$^{3+}$ (1/20/2%)@NaYF$_4$:Nd$^{3+}$(25%) core-shell UCNPs, measured in water at pH 7.4.

<table>
<thead>
<tr>
<th>UCNP samples</th>
<th>Intensity-weighted particle size ($D_h$) in nm (PDI)(^{[a]})</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>as synthesised</td>
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<tr>
<td>OPLT-coated</td>
<td>14.4 ± 3.1 (0.21)</td>
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<tr>
<td>Alendronate-coated</td>
<td>13.2 ± 1.8 (0.19)</td>
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<tr>
<td>PEG-phosphate-coated</td>
<td>16.2 ± 4.1 (0.34)</td>
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<tr>
<td>OA-coated(^{[b]})</td>
<td>10.3 ± 1.3 (0.10)</td>
</tr>
</tbody>
</table>

\(^{[a]}\) PDI = polydispersity index. \(^{[b]}\) The measurements were performed using NP solution in cyclohexane.