

Fig. S1. NaGdF₄:Dy@PPF solution with the concentration of 1000 µg mL⁻¹ (a) and 100 µg mL⁻¹(b) stored in water and PBS at room temperature and PBS at 4°C for 15 days.

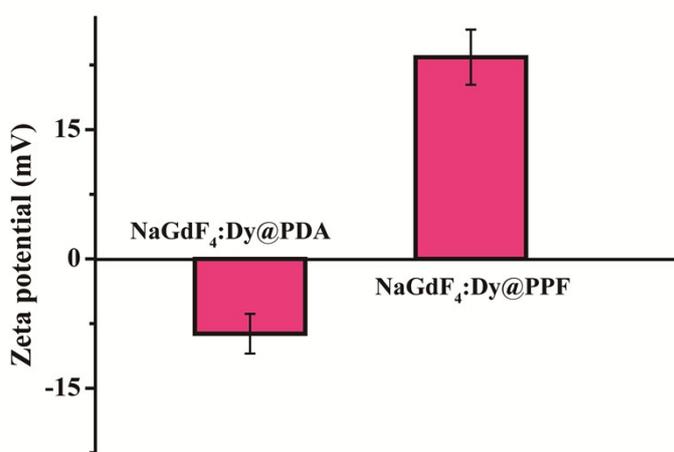


Fig. S2. Zeta potentials of the synthesized NaGdF₄:Dy@PDA and NaGdF₄:Dy@PPF.

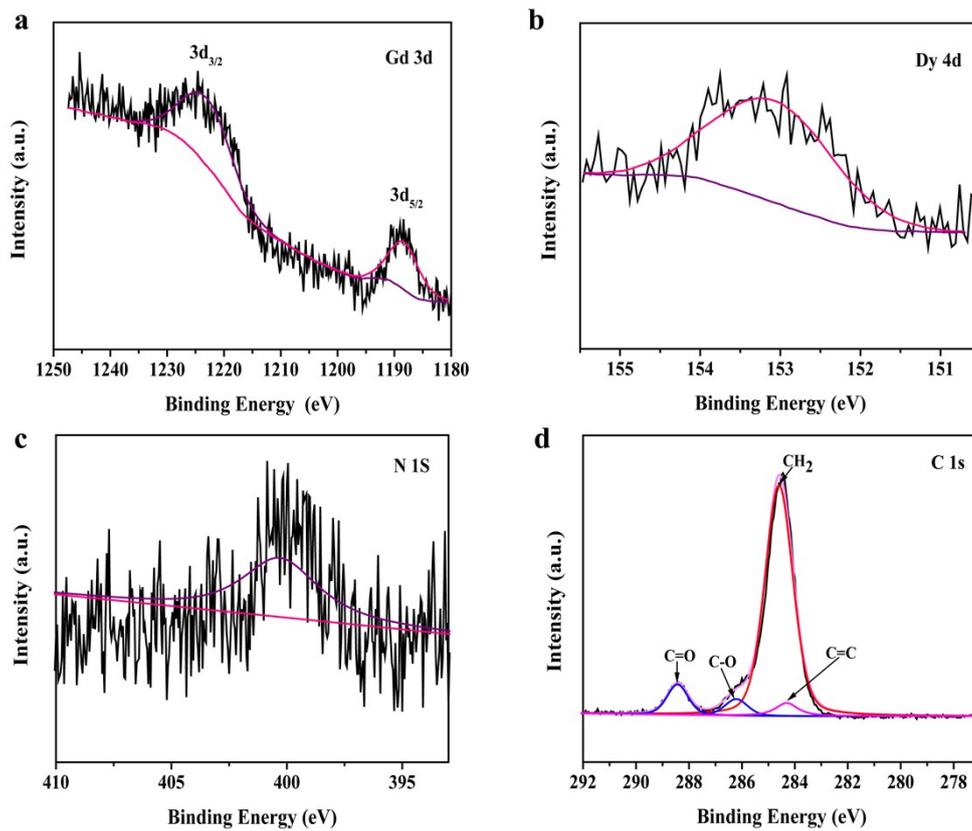


Fig. S3. High resolution XPS spectra of Gd 3d (a) and Dy 4d (b). High resolution spectra of N 1s (c) and C 1s (d).

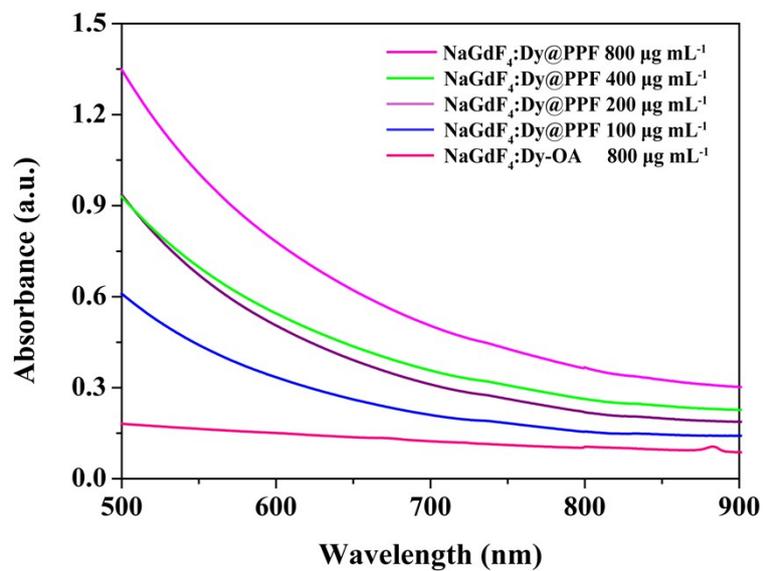


Fig. S4. UV-vis-NIR absorption spectra of different concentrations of NaGdF₄:Dy-OA and NaGdF₄:Dy@PPF sample.

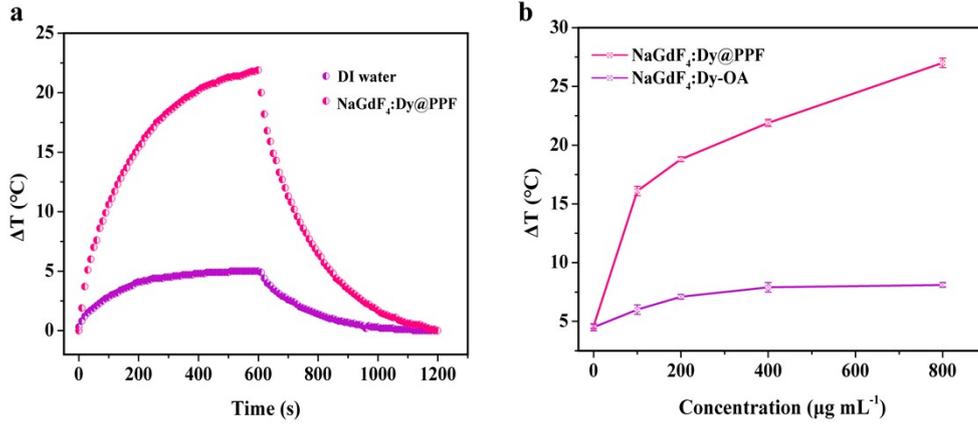


Fig. S5. Heating and cooling curve of NaGdF₄:Dy@PPF and water (a). Temperature changes of NaGdF₄:Dy-OA and NaGdF₄:Dy@PPF under 808 nm laser irradiation for 10 min (1.5W cm⁻²) (b).

Calculation of photothermal conversion efficiency:

The photothermal conversion efficiency was calculated according to the following Equations 1, 2 and 3^[1].

$$\eta = \frac{hA\Delta T_{MAX} - Q_S}{I(1 - 10^{-A_\lambda})} \quad (1)$$

where h is the heat transfer coefficient, A is the surface area of the vessel, ΔT_{MAX} is the maximum temperature change of NaGdF₄:Dy@PPF solution from ambient temperature to maximum temperature, Q_S is the heat associated with the absorbance of the solvent, I is the laser power, A_λ is the absorbance of the NaGdF₄:Dy@PPF solution at 808 nm (Fig. S4), and η is the photothermal conversion efficiency.

$$\tau_s = \frac{\sum m_i c_{p,i}}{hA} \quad (2)$$

The value of hA is calculated using Equation 2, where τ_s is the sample system time constant, m_i is the mass of the solvent, and $C_{p,i}$ is the heat capacity of the solvent. The value of τ_s is calculated from the mode of the cooling curve (Fig. 4d).

$$Q_S = \frac{cm\Delta T_{H_2O}}{t} \quad (3)$$

And ΔT_{H_2O} is the maximum temperature change of water from ambient temperature to maximum temperature, t is the time it takes for the water to warm up to the maximum temperature.

Table 1. Theoretical moles and actual moles of Gd and Dy in NaGdF₄:Dy@PPF NPs

Samples	Gd ³⁺ of NPs	Dy ³⁺ of NPs
Theoretical moles (mmol)	0.80	0.20
Actual moles (mmol)	0.76	0.24

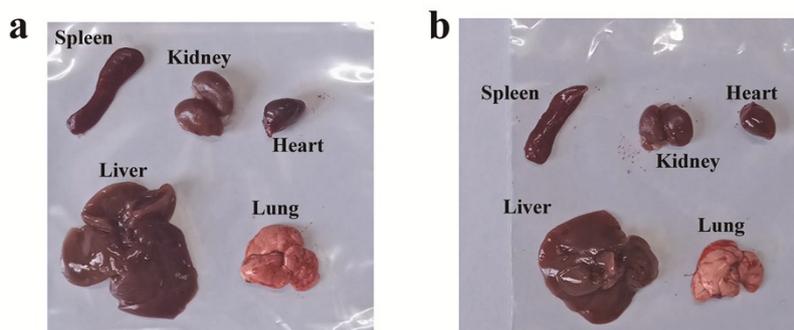


Fig. S6. The major organs (heart, liver, spleen, lung and kidney) of mice in the test (a) and control group (b).

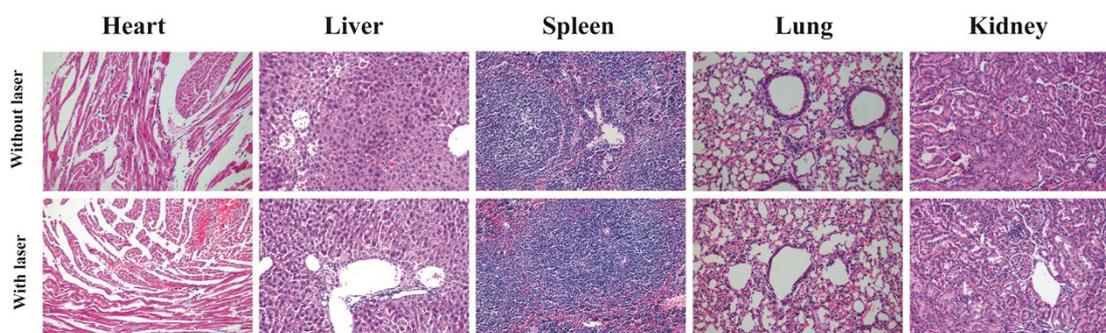


Fig. S7. H&E stained organ sections harvested from mice before and after laser irradiation

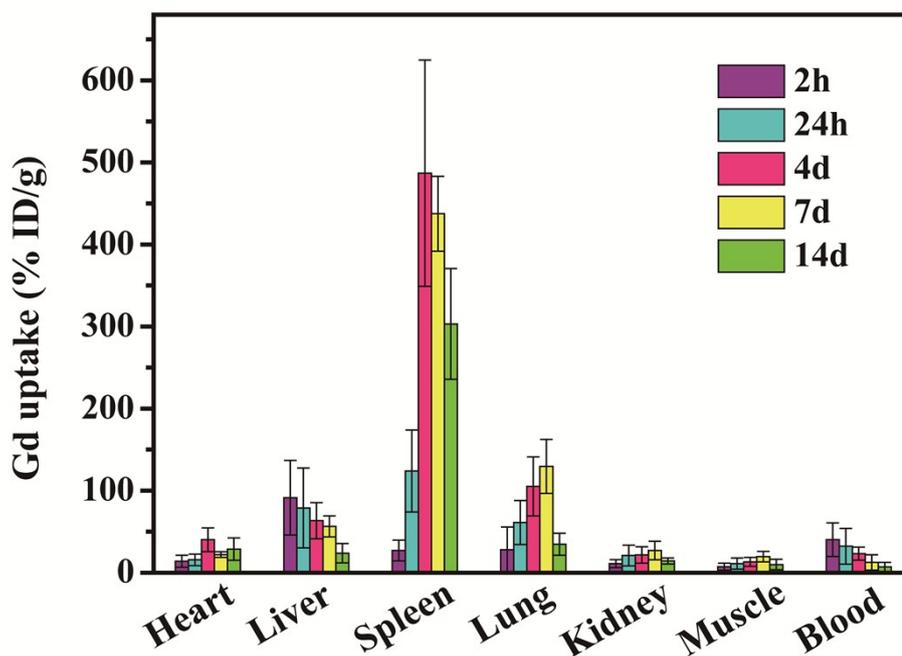


Fig. S8 Biodistribution of the NaGdF₄:Dy@PPF at different time points after injection(2 h, 24 h, 4 d, 7 d and 15 d).

For the application *in vivo*, the long-term biodistribution and metabolic process of the injected nanoprobes is an important concern. To determine the biodistribution and excretion pathway of NaGdF₄:Dy@PPF, the content of Gd in various organs (heart, liver, spleen, lung and kidney) and tissues (blood and muscle) was measured by ICP-OES at various time points (2 h, 24 h, 4 d, 7 d, 14 d) after injection. As shown in Fig. S8, at 2 h post administration, NaGdF₄:Dy@PPF mainly accumulated in liver (91.39 %ID Gd per g) and spleen (26.98 %ID Gd per g) through the mononuclear phagocytes of reticuloendothelial system (RES)^[2]. The higher uptake was also found in lung (27.92 %ID Gd per g), which might be attributed to the retention in the capillary vessels of lung owing to the aggregation of nanoparticles after injection. In contrast, the accumulations in heart, kidney, muscle and blood were less at any time post administration. At the fourth day, the uptake in liver (63.45 %ID Gd per g), spleen (486.97 %ID Gd per g) and lung (105.19 % ID Gd per g) increased to the maximum values, implying that the current nanoparticles might take a biliary elimination pathway. After 14 days injection, the % ID per g values gradually reduced, indicating that these injected nanomaterials could be completely excreted out from mice as time prolonged.

[1] Y. Dai, D. Yang, D. Yu, C. Cao, Q. Wang, S. Xie, L. Shen, W. Feng and F. Li, *ACS Appl. Mater. Interfaces*, 2017, **9**, 26674-26683.

[2] X. Jin, F. Fang, J. Liu, C. Jiang, X. Han, Z. Song, J. Chen, G. Sun, H. Lei and L. Lu, *Nanoscale*, 2015, **7**, 15680-15688.