## Ultra-small and Size Tunable PVP-NaGdF<sub>4</sub>:Dy Nanoparticles With High Biocompatibility for Multimodal Tumor Imaging

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Table S1. Parameters for controlling the size of OA-NaGdF<sub>4</sub>:Dy.

Temperature (°C)	Time (min)	Size (nm)
240	60	2.51
280	60	8.62

Table S2 The theoretical and actual molar ratio of Gd:Dy in OA-NaGdF<sub>4</sub>:Dy NPs.

Samples	2.51 nm NPs	8.62 nm NPs
Theoretical molar ratio of Gd/Dy	0.80:0.20	0.80:0.20
Actual molar ratio of Gd/Dy	0.783:0.217	0.774:0.226

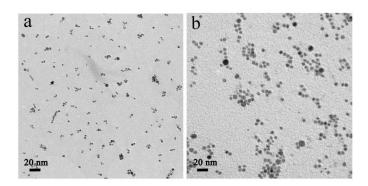


Fig. S1. TEM images of different PVP-NaGdF<sub>4</sub>:Dy NPs obtained at 240 °C (a), 280 °C (b).

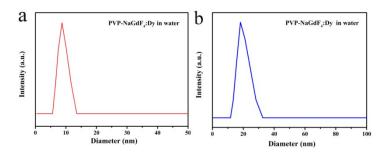


Fig. S2. Hydrodynamic size of different PVP-NaGdF<sub>4</sub>:Dy NPs obtained at 240 °C (a), 280 °C (b).

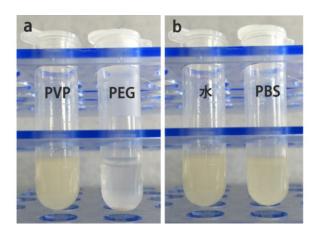


Fig. S3. The photographs of the aqueous solutions of PVP-NaGdF<sub>4</sub>:Dy NPs and PEG-NaGdF<sub>4</sub>:Dy NPs after standing 1 h (a), PVP-NaGdF<sub>4</sub>: Dy NPs in water and PBS after 30 days (b).

The preparation method of PEG-NaGdF<sub>4</sub>:Dy NPs: 5 mg NPs in 5 mL chloroform were added into 10 mL chloroform including 5 mg DSPE-PEG2000 in a 50 mL flask, the solution was stirred for 10 min at room temperature then evaporated using a rotary evaporator. The obtained mixture was maintained at 60 °C for 1 h under vacuum. Then, the product was re-dispersed in 5 mL deionized water.

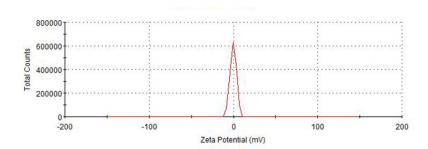


Fig. S4. Zeta Potential of PVP-NaGdF<sub>4</sub>:Dy NPs in water.

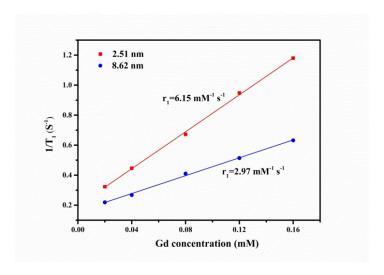


Fig. S5. The longitudinal relaxivity of different size PVP-NaGdF<sub>4</sub>:Dy NPs.

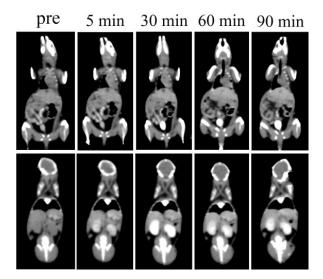


Fig. S6. CT images of the mice before and after injection of iobitridol at different time point through tail vein (100  $\mu$ L, 50 mg I mL<sup>-1</sup>).