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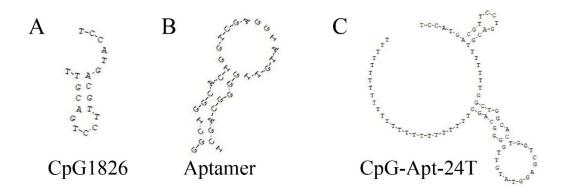


Figure S1. The secondary structures of nucleic acids as predicted by the DNAMAN® software. (A) CpG1826; (B)Aptamer; (C) CpG-Apt-24T.

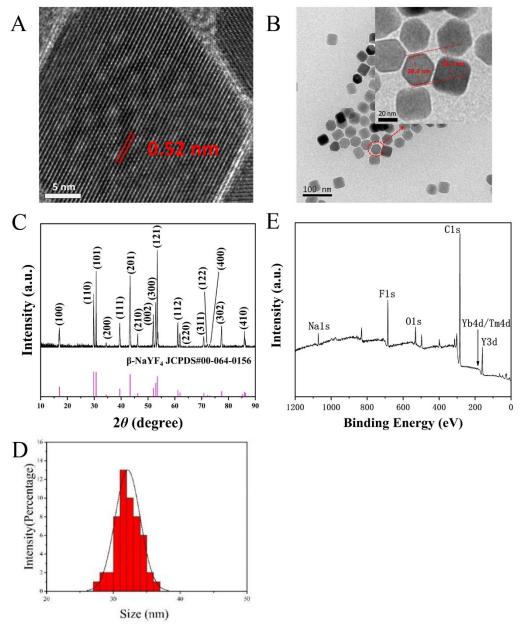


Figure S2. (A) HRTEM image of the NaYF<sub>4</sub>:Yb/Tm. The HRTEM image of UCNPs showed that the lattice distance was 0.52 nm, corresponding to the typical plane (110 and 010) of the hexagonal NaYF<sub>4</sub> structure. (B) HRTEM image of UCNPs. The shell thickness of the UCNPs was about 3.8 nm.(C) XRD patterns of UCNPs and the standard card of  $\beta$ -NaYF<sub>4</sub> (JCPDS: 00-064–0156). (D) XPS survey spectrum of UCNPs. (E)Size distribution of 50 UCNPs by ImageJ software.

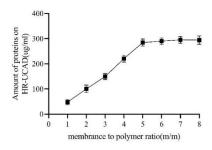


Figure S3:Determination of total membrane protein content on HR-UCAD.

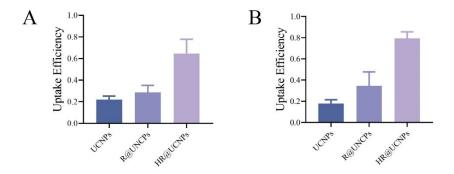


Figure S4: (A)The uptake percentages of UCNPs, R@UCNPs and HR@UCNPs in 4T1 cells were 22%, 29% and 65%, respectively. (B) The uptake percentages of UCNPs, R@UCNPs and HR@UCNPs in tumor tissues were 18%, 34% and 79%, respectively.

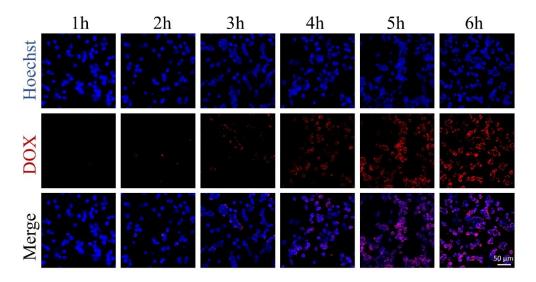


Figure S5 Fluorescence image of intracellular cellular uptake of HR-UCAD by 4T1 cells at different times. Nuclei were stained with Hoechst 33342 (blue). Scale bar:  $50 \mu m$ .