Electronic Supplementary Information

A Nanoprobe for Fluorescent Monitoring of MicroRNA and Targeted Delivery of Drugs

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Apparatuses and measurements

Transmission electron microscopy (TEM) (H600, Hitachi, Japan), scanning electron microscopy (SEM) (Hitachi SU8010, Hitachi, Japan) and high angle annular dark field scanning transmission electron microscopy (HAADF-STEM) (Tecnai™ G2 F20, FEI, USA) were used to investigate the size, morphology, and microstructure of nanoparticles. Elemental compositions were analyzed on Thermo Escalab 250Xi X-ray photoelectron spectrometer (Thermo Fisher, USA). Crystal structures were analyzed by Smartlab X-ray diffractometry (Rikagu, Japan). Zeta potential was measured using Nano-ZEN3600 zetasizer system (Malvern, UK) at 25 °C. Dynamic light scattering (DLS) experiments were performed using BI-200SM instrument (Brookhaven, USA) under scattering angle of 90° at 25 ºC. UV-Vis-NIR spectra were measured with a Shimadzu UV-2550 spectrophotometer (Shimadzu Co, Kyoto, Japan). Fluorescence measurements were carried out on a Cary eclipse fluorescence spectrophotometer (Agilent Technologies, USA) using a 0-2 W 980 nm CW laser (Hi-tech Optoelectronics Co., Ltd. Beijing, China) as excitation source. The confocal images were imaged by a two-photon excited confocal laser scanning microscope

(Leica SP8 DIVE, Germany). The cell viability was measured using a microplate reader (Multiscan GO, Thermo Scientific).

Table S1. Nucleic acid sequences in this work.

Oligonucleotide	Sequence
Anchor-DNA	5'-phosphate-(CH_2)6-TAGCTTATCAGACTG-(6-FAM)-3'
Capping-DNA	5'-GGTGGTGGTGGTTGTGGTGGTGGTGGTCAACAT CAGTCTGATAAGCTA-3'
MicroRNA-21	UAGCUUAUCAGACUGAUGUUGA
Scramble sequence	AUUGAAGGUUCGUAUUACGCAU
MicroRNA-200b	UAAUACUGCCUGGUAAUGAUGA
MicroRNA-15a	UAGCAGCACAUAAUGGUUUGUG
Let-7d	AGAGGUAGUAGGUUGCAUAGUU
MicroRNA-141	UAACACUGUCUGGUAAAGAUGG

*Underlined letters represent the sequence of AS1411.

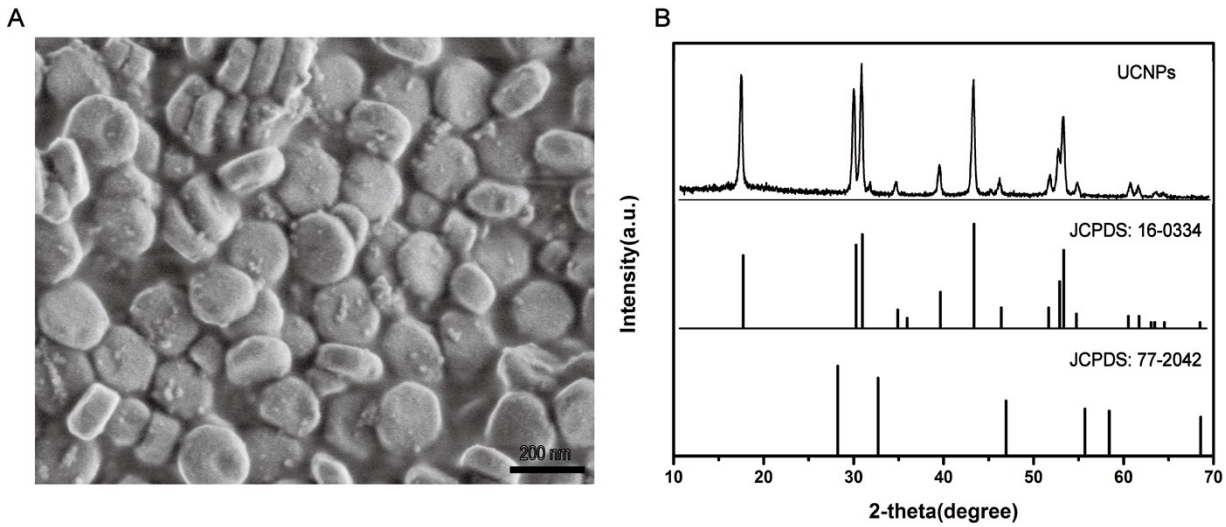

Figure S1. Scanning electronic microscopy (SEM) characterization (A) and X-ray diffraction (XRD) spectrum of UCNPs compared with hexagonal plate pattern (JCPDS: 16-0334) and cubic pattern (JCPDS: 77-2042).

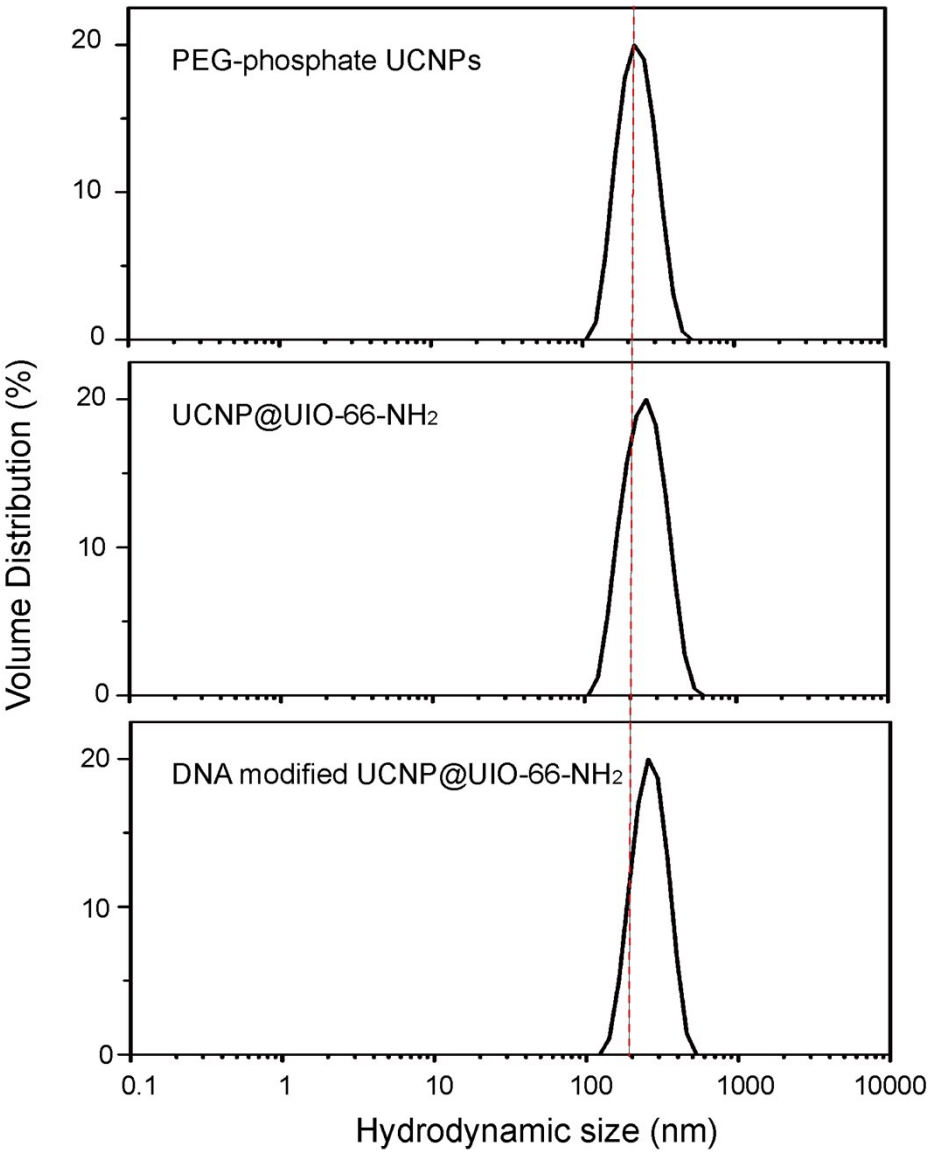

Figure S2. Hydrodynamic size distributions of PEG-phosphate coated UCNPs, UCNP@UIO-66-NH_2 NPs and DNA modified UCNP@UIO-66-NH_2 NPs in PBS buffer (pH =7.4).

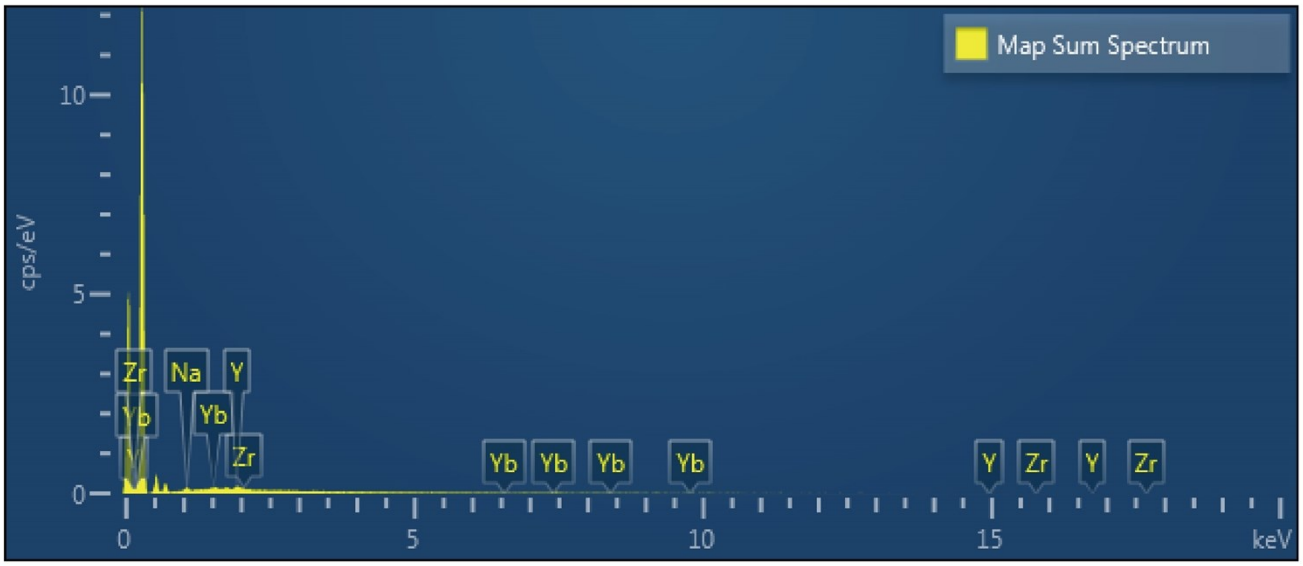

Figure S3. Energy dispersive X-ray spectrum (EDS) of UCNP@UIO-66-NH_2.

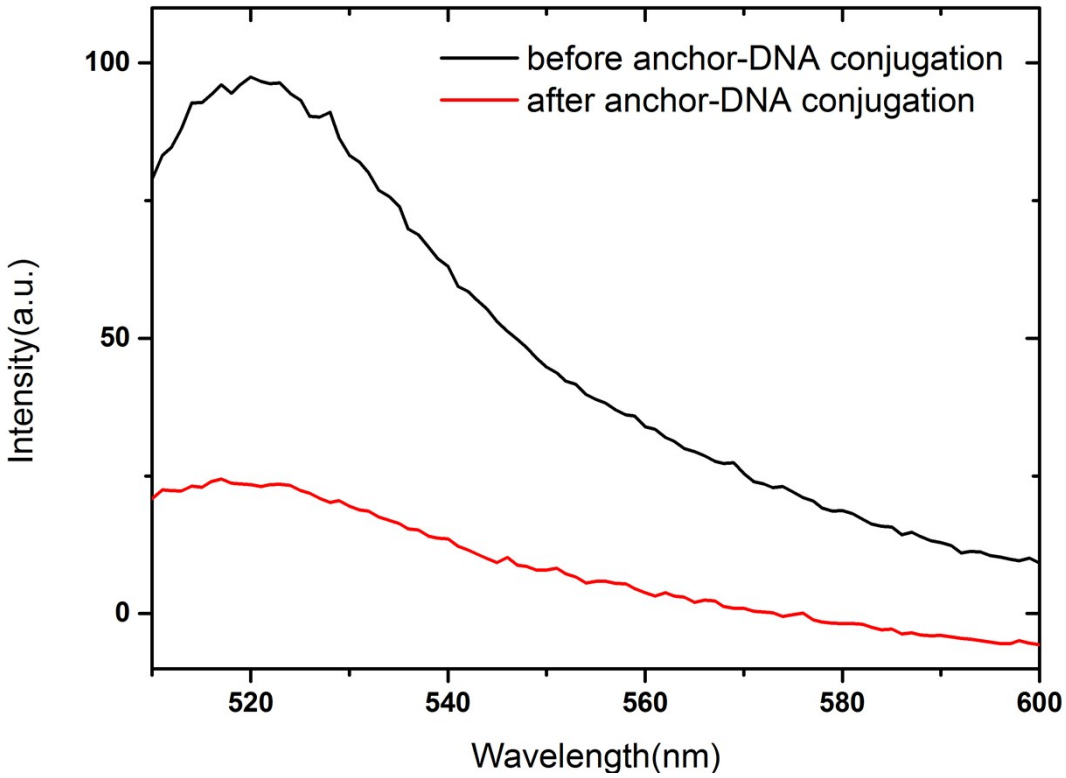

Figure S4. Fluorescence spectrum of 6-FAM in the supernatant before and after anchor-DNA conjugation.

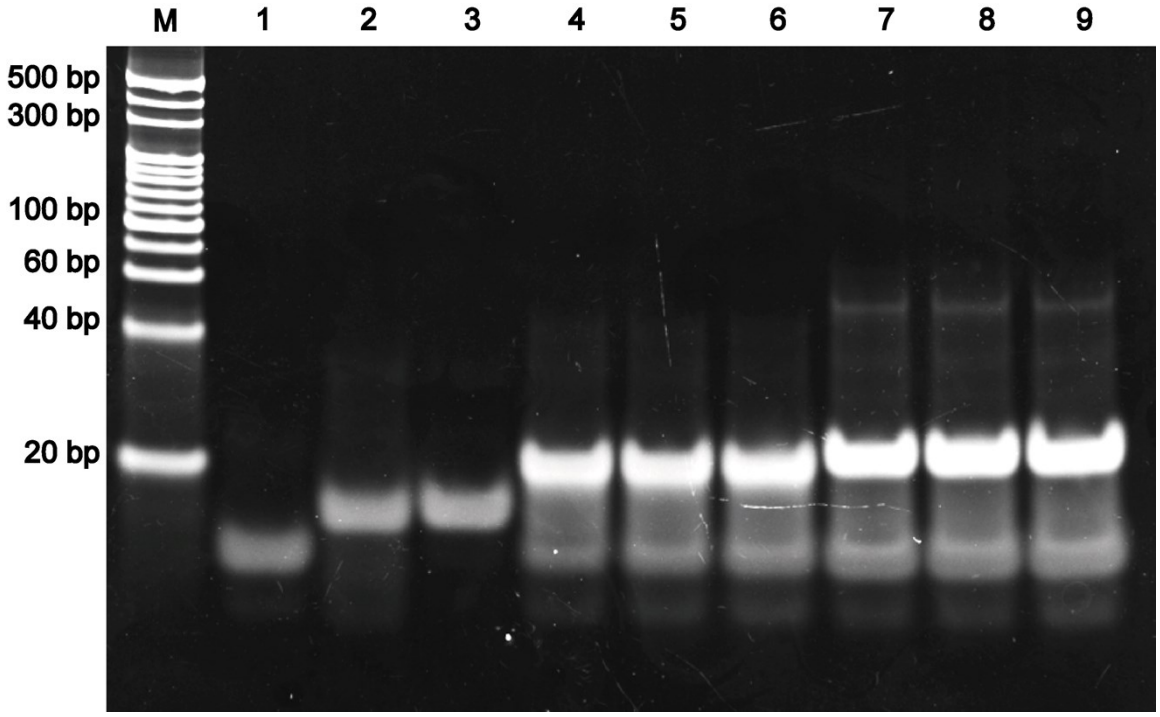

Figure S5. Optimization and verification of the ratio of anchor-DNA and capping-DNA (without AS1411) by PAGE (M: marker; lane 1: anchor-DNA; lane 2: capping-DNA (without AS1411); lane 3: miR-21; lane 4-lane 6: DNA duplexes hybridized by anchor-DNA and capping-DNA in the ratio of 1:1, 1:1.3 and 1:1.5; lane 7: Mixed system of DNA duplexes of lane 4 and miR-21; lane 8: Mixed system of DNA duplexes of lane 5 and miR-21; lane 9: Mixed system of DNA duplexes of lane 6 and miR-21).

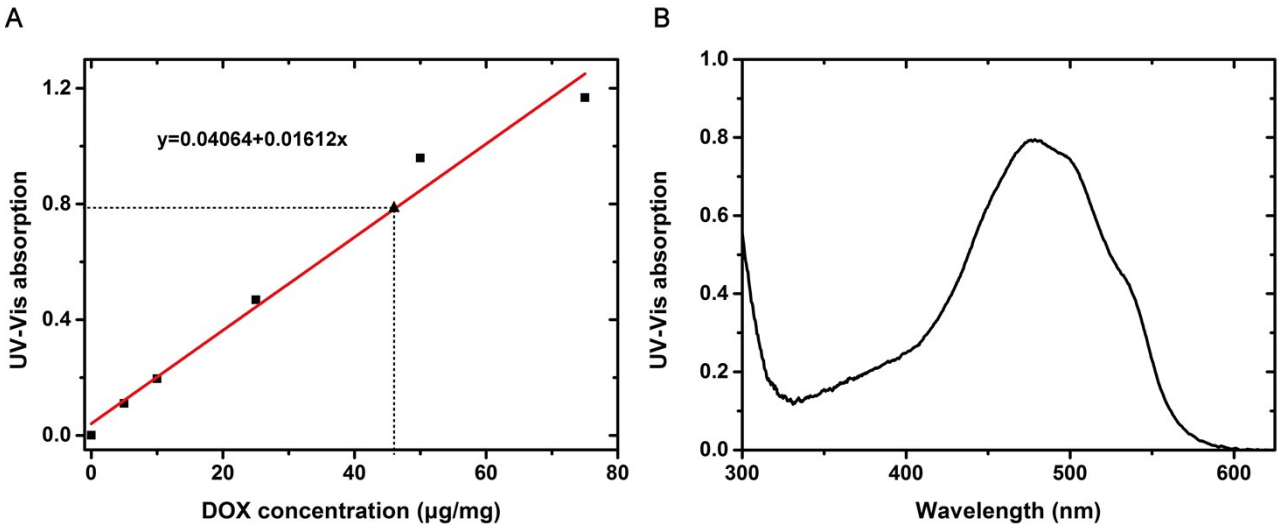

Figure S6. The DOX loading efficiency in DNA-hybrid-gated UCNPs@UIO-66-NH_2/DOX. (A) The calibration curve corresponding to the UV-Vis absorption as a function of the concentration of DOX. (B) The UV-Vis absorption of the supernatant diluted 10 times after loading DOX in nanoprobe.

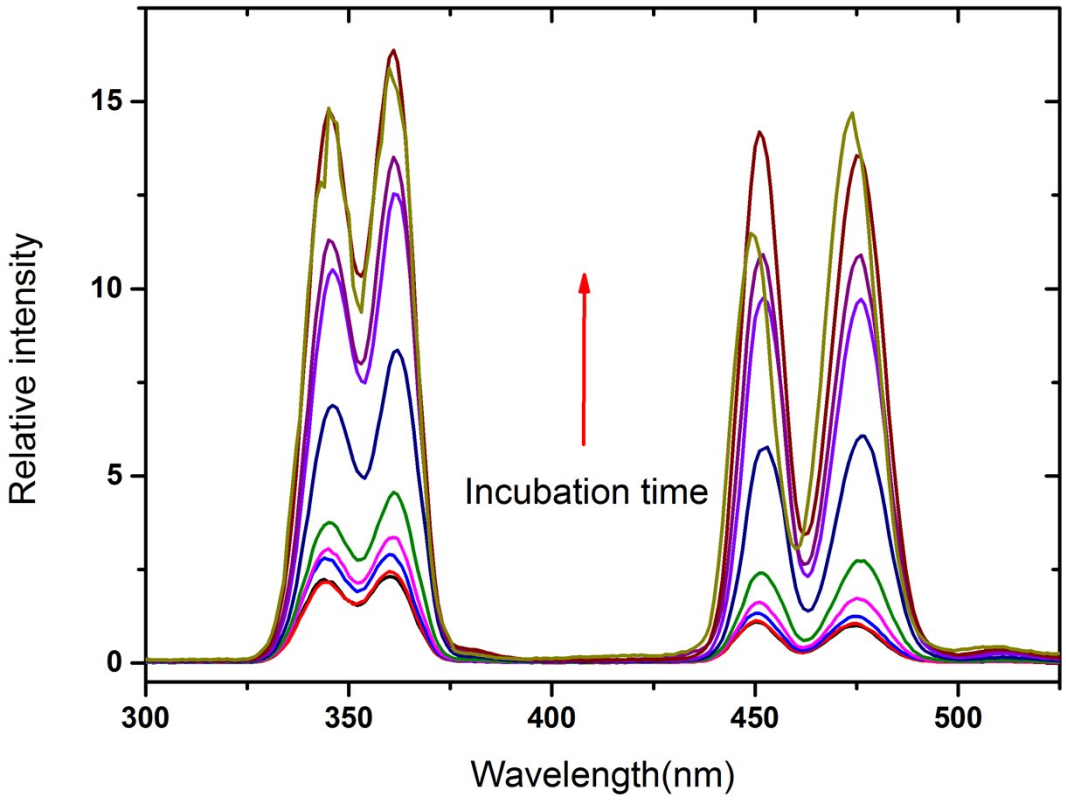

Figure S7. The UCF signal of the nanoprobe (0.75 mg mL^{-1}) incubated with miRNA-21 (100 nM) at different time intervals.