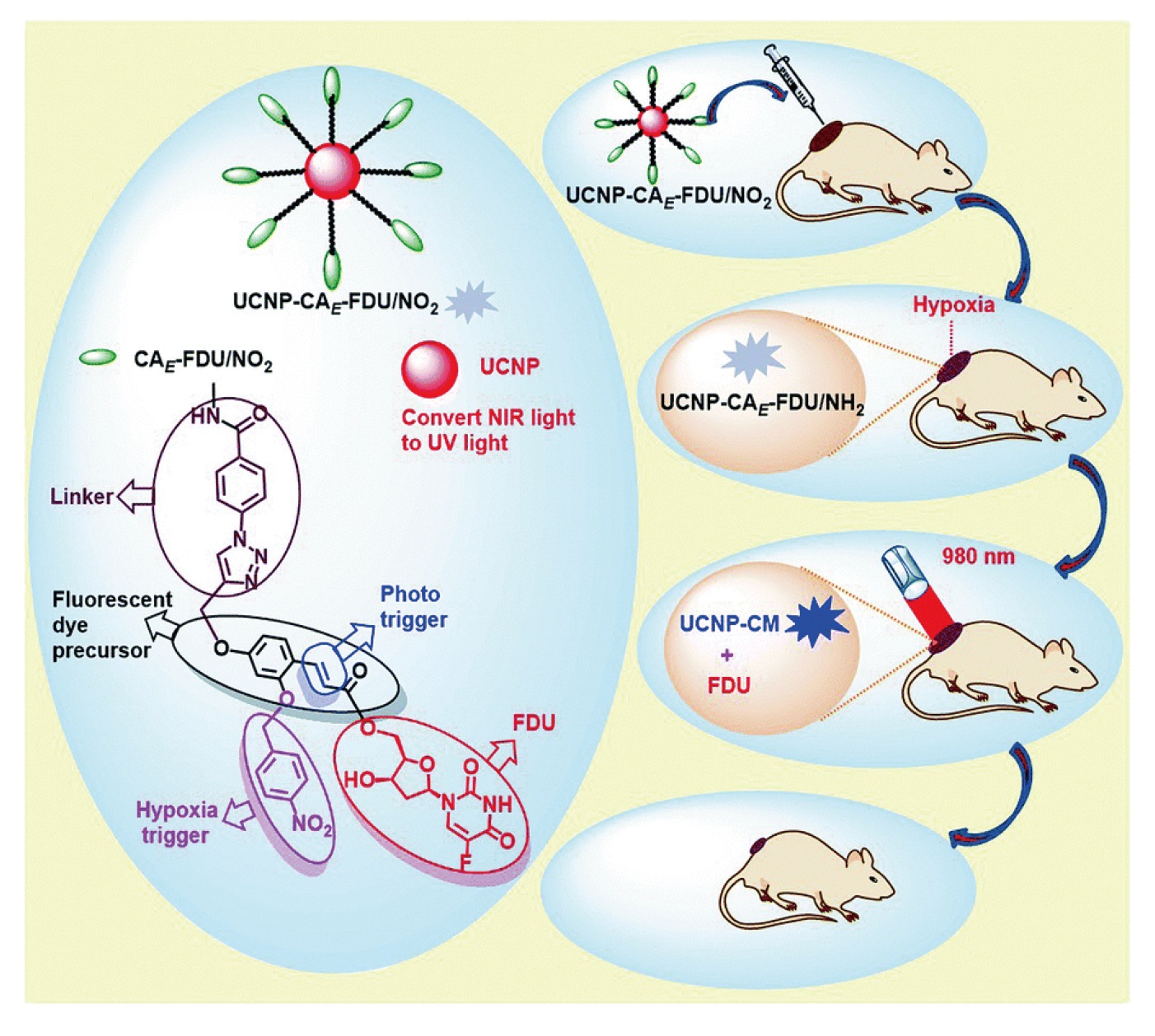
**Supplementary Information**



Supplementary Figure 1. Schematic of light-controlled liposome–membrane fusion process. UCNPs: Upconversion nanoparticles; DOX: Doxorubicin. Reproduced with permission from [1], licensed with CC BY 3.0.



Supplementary Figure 2. Schematic illustration of the UCNP-CA*E*-FDU/NO2-based theranostic nanoplatform under the hypoxia state. UCNPs: Upconversion nanoparticles; NIR: Near infrared; UV: Ultraviolet; CA*E*: (*E*)-*o*-hydroxycinnamic acid; FDU: Floxuridine; CM: 7-propargyloxycoumarin. Reproduced with permission from [2], © 2018 Royal Society of Chemistry.



Supplementary Figure 3. Schematic demonstration of NO-modulated and overloaded Ca2+-triggered tumor cell killing effect. UC: Upconversion; ZIF: Zeolitic framework; BER: Berbamine; NO: Nitric oxide; RyR: Ryanodine receptors. Reproduced with permission from [3], © 2021 Wiley-VCH GmbH.



Supplementary Figure 4. Illustration of the design for separating the imaging and PDT. UCL: Upconversion light; PDT: Photodynamic therapy; RB-HA: rose bengal hexanoic acid; BHQ: Black hole quencher. Reproduced with permission from [4], © 2019 Elsevier Ltd.



Supplementary Figure 5. Illustration of the pH dependent FRET process. Reproduced with permission from [5], © 2020 Elsevier B.V.



Supplementary Figure 6. Schematic illustration of the glycans detection on the cancer cell membrane. UCNP: Upconversion nanoparticle. Reproduced with permission from [6], licensed with CC BY-NC 3.0.



Supplementary Figure 7. Schematic overview of tumor immunotherapy activated by dual-mode ROS. AUNPs: AIEgen-coupled upconversion nanoparticles; NIR: Near infrared; PDT: Photodynamic therapy; ICD: Immunogenic cell death; TAA: Tumor-associated antigen; DLN: Draining lymph node; I.T.: Intratumoral injection. Reproduced with permission from [7], licensed with CC BY-NC 4.0.



Supplementary Figure 8. Schematic illustration of NIR-controlled dePEGylation for improving the tumor penetration. UCNPs: Upconversion nanoparticles; NIR: Near infrared; UV: Ultraviolet; PEG: Polyethyleneglycol; DOX: doxorubicin; PAE: poly(β-aminoester)s. Reproduced with permission from [8], © 2019 American Chemical Society.

**Supplementary Table 1.** A summary of UCNP-based nanoplatforms for light-controlled drug release.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Nanoplatform** | **UCNP** | **Photoresponsive moieties** | **Drug** | **Ref.** |
| UCNP–ONB–FU | NaYF4:Yb,Tm@NaYF4 | *o*-nitrobenzyl | 5-FU | [9] |
| DOX-UCNP@PNB-*b*-POEG | NaYF4:Yb,Tm@NaYF4 | *o*-nitrobenzyl | DOX | [10] |
| DOX-UCNP@mPEG-PEI-(NBS,OA) | NaYF4:Yb,Er | *o*-nitrobenzyl | DOX | [11] |
| UCNP@Azo-Lipo/DOX | NaGdF4:Yb,Tm@NaGdF4 | azobenzene | DOX | [12] |
| DOX-UCNPs@PAzo/MAA | NaYF4:Yb,Tm@NaYF4 | azobenzene | DOX | [13] |
| DOX-UCNPs@PMA | NaYF4:Yb,Tm@NaLuF4 | azobenzene | DOX | [14] |
| DOX-UCNP@mSiO2-Azo | NaYF4:Yb,Tm@NaYF4 | azobenzene | DOX | [15] |
| DOX-UCNP@mSiO2-DNQ-CD | NaYF4:Yb,Tm@NaYF4 | DNQ | DOX | [16] |
| UCNP@mSiO2(DOX)-S-BP@β-CD | NaYF4:Yb,Tm@NaYF4 | thioether | DOX | [17] |
| DOX-UCNP-TiO2@mSiO2-Ru2-Au | NaYF4:Yb,Tm,Er,Fe@NaYbF4:Tm | Ru2 | DOX | [18] |
| UCNP@mSiO2/HA-DOX@CNBA | NaYF4:Yb,Tm@NaYF4 | CNBA | DOX | [19] |
| UCNPs@SiO2@mSiO2-SP/Curcumin | NaYF4:Yb,Tm | spiropyran | Curcumin | [20] |
| UCNP: Upconversion nanoparticle; ONB: *o*-nitrobenzyl; 5-FU: 5-fluorouracil; DOX: Doxorubicin; PNB: Poly(4,5-dimethoxy-2-nitrobenzyl methacrylate); POEG: Poly(methoxy polyethylene glycol monomethacrylate); mPEG-PEI: mPEGylated polyethyleneimine; NBS: *o*-nitrobenzyl succinate; OA: Oleic acid; Azo: Azobenzene;PMAA: Poly(methacrylic acid); mSiO2: mesoporous silica; DNQ: 2-diazo-1,2-naphthoquinones; CD: Cyclodextrin; -S-BP: Thioether linkers; HA: Hypocrellin A; CNBA: 4-(2-carboxy-ethylsulfanylmethyl)-3-nitro-benzoic acid linker; SP: Spiropyran. | | | | |

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