

Supporting Information on

An Overview on Nanoparticles Commonly Used in Fluorescent Bioimaging

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Table S1. Representative examples of nanoparticles (NPs) for use in plain (non-bimodal) fluorescence (bio)imaging

<i>Material and application</i>	<i>Ref.</i>
Reviews on synthesis, characterization, properties and uses of P-dots in bioimaging and other fields	1,2
Review on carbonic nanomaterials for sensing and imaging	3
Review on nanoparticles and nanocomposites for fluorescence sensing and imaging	4
SiNPs doped with NIR dyes to enable imaging in the optical window of biomatter; allows simultaneous imaging with other commonly used dyes or fluorescent proteins	5
SiNPs wrapped with graphene oxide; this hybrid material exhibits good stability and protects the fluorescent dye in the NPs from external attack	6
Hydrophilic fluorescent core-shell SiNPs containing NIR cyanine chromophore	7
SiNPs doped with green fluorescence protein	8
Organic NPs containing a dye that undergoes aggregation-induced emission; applied to cell imaging	9
Photostable fluorescent organic NPs with aggregation-induced emission for noninvasive long-term cell tracing	10
Organic NPs with red fluorescent aggregation-induced emission dye, and cell imaging applications	11,12
Biocompatible organic polymer NPs prepared from a zwitterionic monomer; doped with red fluorescent dye that displays aggregation-induced emission; applied to cell imaging	13
Aggregation-induced emission based fluorescent NPs for cell imaging applications	14
Reversibly photoswitchable dual-color fluorescent NPs for live-cell imaging; uses photochromism and a FRET system; used in HEK-293 cells where it display red or green fluorescence depending on the wavelength of light to which the cells are highlighted	15
Carbon dots for multicolor patterning, sensors, and bioimaging	16
Mesoporous upconverting NCs coated with a shell of silica; bioimaging of living cells	17
Tm(III) doped NaYbF ₄ upconversion NPs coated with CaF ₂ ; excellent brightness and suitable for high-contrast deep tissue imaging	18
Silica NPs loaded with the sensitizer Pd(II)octaethylporphyrin and an annihilator; low-power excitation; used for bioimaging of lymph nodes of living mice	19
C-dots functionalized with and rhodamine B for imaging cells; excitation at 225 nm is disadvantageous	20
Near-infrared emitting gold nanoclusters for tumor imaging in vivo	21
Fluorescent gold nanoclusters used as biocompatible marker for tracking endothelial cells; delivered into cells via the liposome complex	22
Sandwich-like SiO ₂ @CdTe@SiO ₂ fluorescent NPs for cellular imaging	23
Q-dots loaded with fluorescent liposomes; fluorescence resonance energy transfer studies; NIR in-vivo imaging of mouse tissue	24
Clathrin- and caveolin-mediated endocytosis and macropinocytosis of 6-nm titanium dioxide (anatase) NPs in prostate cancer PC-3M cells	25
Biocompatible fluorescent hydroxyapatite NPs for live cell imaging	26
Calcium phosphate NPs doped with NIR emitting fluorophore; used for in vivo-imaging of human breast cancer	27
DNA-quantum dot sheath used to image and electrochemically detect cancer cells	28
Multicolor upconversion NaLuF ₄ NPs for plant cell imaging and detection of sodium fluorescein; NPs are paramagnetic in strong magnetic field	29
Comparative study on upconversion NPs prepared by hydrothermal synthesis and coated with poly(ethylene glycol), polyethylenimine or 6-aminocaproic acid; used to image HeLa cells	30
Upconversion NP based luminescence imaging of cells and small animals	31
Preparation of core-shell NaGdF ₄ upconversion NPs doped with luminescent lanthanide ions for used as NIR probes	32
NaYbF ₄ :Tm,Gd NPs with engineered NIR-to-NIR upconversion fluorescence for in-vivo imaging	33
10-nm sized lanthanide-doped NaLuF ₄ upconversion NPs for imaging of a whole-body black mouse with a penetration depth of ~2 cm	34
NaYF ₄ :Nd nanocrystals for in-vivo bioimaging with minimized heating	35

Table S2. Representative examples of fluorescent nanoparticles (NPs) for use in targeted imaging. Numerous others are known based on the use of various kinds of (bright) nanoparticles and surface-modified in order to target specific domains

<i>Method and nanomaterial</i>	<i>Ref.</i>
Quantum dots functionalized with streptavidin and a zwitterions used to track the cannabinoid receptor in live cells	36
Graphene oxide nanosheet modified with an aptamer and carboxyfluorescein used for intracellular molecular targeting	37
Silver cluster with aptamer used to specifically mark the nucleus of live cells	38
Two-photon imaging of immune-functionalized NPs targeted to breast cancer cell line SK-BR-3; alternating 780-nm two-photon and 488-nm single-photon excitations induce reversible on-off fluorescence switching	39
Antigen-loaded pH-sensitive hydrogel particles; used for T-cell activation in vivo	40
Semiconducting conjugated organic polymer NPs with folic acid on surface to target and image tumor tissue	41
Nanoprobe enables all targeted cells to be imaged (at 680 nm) and specific cells to be photoactivated using 405- nm light; photoactivated cells can then be tracked (at 525 nm) spatiotemporally	42
High contrast targeted imaging in-vivo using peptide labeled upconversion NPs	43
Poly(ethylene glycol)-co-poly(lactic acid) NPs dyes with NIR fluorophore and labeled with two targeting peptides; used to target amyloid plaques in the brains of Alzheimer's disease mice	44
Gold NPs modified with hairpin DNA to target mRNA	45
Aptamer-based silver NPs used to target and image intracellular proteins	46
Amino-modified lanthanide-doped zirconia NPs for use in time-resolved FRET assay and in targeted imaging	47
Hyperbranched dendritic NPs for sub-cellular targeting of doxorubicin and the sensitization breast cancer cells by circumventing microsomal glutathione transferase-mediated drug resistance	48

Table S3. Representative examples of fluorescent nanoparticles (NPs) for sensing and imaging of intracellular pH values

<i>Materials and Methods</i>	<i>Ref.</i>
Dually colored mesoporous SiNPs with pH activable rhodamine-lactam for ratiometric sensing of lysosomal acidity	49
CdSe/ZnS quantum dot photoluminescence lifetime-based pH nanosensors; capped with mercaptopropionic acid; lifetime depends on pH in the range from 5.2 to 6.9; applied to simulated intracellular media	50
Quantum dot-fluorescent protein FRET probes for sensing intracellular pH; used to visualize the acidification of endosomes in living cells following polyarginine-mediated uptake	51
C-dots labeled with fluorophores FITC and RBITC (acting as ratiometric probe and reference, respectively) for intracellular pH; applied to intact HeLa cells under oxidative stress	52
C-dots used for ratiometric (2-wavelength) sensing of intracellular pH; uses two types of C-dots, one exhibiting excitation-independent and pH independent blue emission, the other showing excitation-dependent and pH dependent full-color emissions	53
Polyurethane nanogel for ratiometric fluorescent sensing of intracellular pH values; uses FRET; three dyes are employed (the pH probe Bromothymol Blue, Nile Red and a coumarin)	54
Nanogel containing a ratiometric pH probe (8-hydroxypyrene-1-carbaldehyde) for imaging intracellular pH; pH induces and up to 100-nm hypsochromic shift	55
Fluorescent pH-sensitive NPs in an agarose matrix for imaging of bacterial growth and metabolism	56
Hydrogel particles made from a block copolymer (made from modified PEO, PMMA and PMA); multicolored; pH-activatable; pK_a values range from pH 5.2, 6.4, 6.9, to 7.2.; used to image the pH of lung cancer cells.	57
Mesoporous silica NPs (MSNs); pH can be probed by ratiometric fluorimaging; positively charged MSNs locate in higher pH regions (cytosol mostly), negatively charged MSNs mainly trapped in acidic endosomes.	58
Crosslinked polyacrylamide NPs used for time-resolved imaging; NPs contain two pH indicators; applied to sense pH in the endosomes-lysosome pathway in living cells	59
Mesoporous silica NPs to sense and image pH; dyed with FTC (pH responsive) and RITC (the reference dye); covers wide pH range	60
Ultrabright semiconducting P-dots labeled with the pH probe fluorescein; ratiometric (2-wavelength) sensing of pH in HeLa cells	61
Gold NP pH sensor based on ratiometric photoinduced electron-transfer and confocal microscopy to image localized intracellular pH	62
Indicator-labelled PAA particles used to image intra(sub)cellular pH via 2-photon-excited fluorescence; non-targeted nanosensors were modified with a peptide in order to target them to escape/avoid acidic compartments in cancer cells	63
Polystyrene nanosensors used for 2D luminescence imaging of pH in wounds in vivo	64
Q-dots based FRET sensing scheme for intracellular pH values; images obtained by ratioing the fluorescence intensities of the Q-dots (the reference) and a pH-sensitive fluorescent protein; FRET efficiency is highly pH-dependent	65
pH-sensitive CdSe/ZnSe/ZnS quantum dots used to image pH value in human ovarian cancer cells	66
Nanoprobes for sensing and imaging of pH based on resonance energy transfer from an UCNP to a pH-dependent dye covalently bound to the aminosilane surface	67

Table S4. Representative examples of fluorescent nanoparticles (NPs) for sensing and imaging of oxygen in biomatter

<i>Material and method</i>	<i>Ref.</i>
Nontoxic, targeted, and ratiometric 30-nm oxygen nanosensors made of polyacrylamide; uses two NIR luminescent dyes (probe and reference) and surface-conjugated tumor-specific peptides to target tumor cells; used to sense oxygen in tumorous tissue	68
Polystyrene nanosensors for sensing and imaging of extracellular, intracellular, and intra-mitochondrial oxygen; targeted sensing of O ₂ is accomplished by proper modification of the surface of the polystyrene NPs with either silica, polylysine, or triphenylphosphonium groups	69
Upconversion NP sensors for imaging oxygen (hypoxia) in malignant solid tumors upon NIR excitation	70
PS nanoparticles dyed with a ruthenium-based probe; applied to image cellular oxygen via 2-photon excitation	71
Sol-gel based spherical optical nanosensors; applied to ratiometric sensing of oxygen inside living rat C6 glioma cells	72
NPs made from a hydrophobic composite and having a PEG shell; membrane-impermeable; complexes of Ir(III), Ru(II), Pt(II) and Pd(II) used as indicator probes	73
Nanosensors consisting of a hydrophobic silica-rigidized core (Pluronic) and a hydrophilic PEG shell; labeled with a pH probe and an oxygen probe; applied to image intracellular oxygen and pH values	74
Polystyrene based luminescent dually responding nanosensors (for pH and oxygen) can visualize extracellular pH-gradients and hypoxia on chronic wounds that disrupt epidermal repair	75
Dendrimer probe (referred to as <i>Oxyphore</i> TM) used to sense local oxygen concentration in the bone marrow of live mice	76
Phosphorescent dendrimer probe used for functional imaging of cerebral oxygenation	77
Fluorescent NPs for sensing oxygen; surface modified with poly-L-lysine to favor internalization; contain a red fluorescent quenched probe for oxygen and a green emitting reference dye	78
Cationic hydrogel NPs dyed with a Pt(II)porphyrin oxygen probe and poly(9,9-dioctylfluorene) acting as a fluorophore in a FRET system and as a 2-photon antenna	79

Table S5. Selected examples of fluorescent nanoparticles (NPs) for use in intracellular sensing/imaging of ions, organic and other species

<i>Material and method</i>	<i>Ref.</i>
Indicator dye for Mg(II) immobilized in polyacrylamide NPs (40 nm i.d.); dynamic range 1 – 30 mM; 2-wavelength ratiometric sensing; used to monitor Mg(II) in C6 glioma cells	80
Ca(II)-sensitive indicator dye and Ca(II)-insensitive reference dye in poly(acrylamide-co-decylmethacrylate) NPs; responds with a K _d of 293 nM	81
Silica NPs dyed with the calcium probe Fluo-4 and an inert reference dye; coated with dextran; dissociation constant is 520 nM which covers the physiological range	82
Optical nanosensors ("PEBBLE" type) using fluorescent probes and organic polymer hosts; used to image both Ca(II) and pH values	83
Silica NPs functionalized with 8-aminoquinoline for imaging Zn(II) in yeast cell suspension	84
Silica-coated thiol-enriched and Zn-doped CdS quantum dots; applied as a turn-on fluorescent probe for imaging intracellular Zn(II) and Cd(II) in endophytes	85
Q-dots covered with a dendron and a Cu(II)-free derivative of zinc superoxide dismutase; used to image changes of intracellular Cu(II) levels with red-to-yellow color change	86
Gold NPs functionalized with BODIPY as a fluoroprobe for imaging Cu(II) in living cells	87
Chloride probe lucigenin self-assembled on mercaptopropionic acid capped QDs; lifetimes of both QD and lucigenin change with chloride concentration; applied to lifetime imaging in simulated intracellular media	88
Nanosensor synthesized by linking a chloride receptor to a water soluble CdSe/ZnS quantum dot; emission peaking at 620 nm quenched by a photo-induced electron transfer mechanism; applied to epithelial cells	89
C-dots acting as fluorescent probes for iodide	90
Iridium(III) complex coated onto a nanosystem for ratiometric luminescence bioimaging of cyanide anion	91
Probing phosphate ion via fluorescent gold nanoclusters capped with 11-mercaptoundecanoic acid and Eu(III) ion	92
Two-photon turn-on fluorescent probe based on C-dots for imaging H ₂ S in cells	93
NIR-fluorescent carbon nanotube nanosensors for spatiotemporal NO signalling in cell cultures	94
PEG-ligated single-walled carbon nanotubes injected into mice for determination of local NO concentration; detection limit is 1 mM; half-life for liver retention is 4 h; NPs clearing the lungs within 2 h after injection	95
C-dot based nanosensor for chromium(VI) and ascorbate based on an inner filter effect	96
Ratiometric fluorescent probe for the hydroxy radical in live cells based on gold nanoclusters	97
Glucose imaged by using polyacrylamide-based, ratiometric nanosensors (PEBBLEs) that incorporate glucose oxidase and an oxygen sensitive fluorescent Ru(II) indicator	98
Glucose imaged with a gold nanoprobe carrying apo-glucose oxidase; method is based on FRET between apo-GOx and dextran labeled with fluorescein isothiocyanate; no oxygen consumed, no H ₂ O ₂ formed	99

Glucose-responsive core-shell hybrid NPs with controllable metal-enhanced fluorescence effects; silver core and cross-linked poly(3-acrylamidophenylboronic acid-co-acrylic acid) shell; fluorescence of adsorbed porphyrin is sensitive to glucose but also to pH	100
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Table S6. Representative examples of fluorescent nanoparticles (NPs) for use in intracellular sensing/imaging of temperature (T)

<i>Methods and materials</i>	<i>Ref.</i>
Review on probes and NPs for sensing temperature; includes methods of imaging	101
Quantum dot based nanothermometers; used to image heterogeneous local thermogenesis in living cells	102
Luminescent Eu(III) complex in composite NPs made from 2-bis(trimethoxysilyl)decane and PMMA; used to image <i>T</i> in the physiological range;	103
Silica NPs doped with the <i>T</i> -probe Ru(bpy) ₃ used to image intracellular temperature	104
Silica nanoparticles and sensor films based on a terbium(III) complex probe	105
Semiconducting P-dots labeled with rhodamine; fluorescence is highly <i>T</i> -dependent	106
Dual-emitting quantum dot/quantum rod-based nanothermometer; delivered to live cells using a pH-responsive cationic polymer colloid	107
Upconversion NPs for ratiometric nanoscale thermometry of cellular systems	108
NPs made from a poly(methyl methacrylate)-co-1,2-bis(trimethoxysilyl)decane composite and a red-emitting Eu(III) dye and a green-emitting reference dye used to sense <i>T</i> in the 25 - 45 °C range with a sensitivity of -4.0 % per °C	109
Quantum dot nanothermometers used to map intracellular heat generation in NIH/3T3 cells following Ca(II) stress and cold shock	110
A molecular thermometer for nanoparticles for optical hyperthermia	111
High-spatial-resolution surface-temperature mapping using fluorescent thermometry	112
<i>T</i> measured on the surface of gold nanorods and nanostars along with magnetite NPs under near-infrared and radiofrequency excitation by monitoring the excited state lifetime of the <i>T</i> probe Rhodamine B placed some 20 nm from the NP surface; sensitivity is 0.029 ns/°C	113

Table S7. Representative examples of nanoparticles (NP) for use in multimodal imaging (i.e., fluorescence combined with MRI or other methods)

<i>Methods and materials</i>	<i>Ref.</i>
Polymer-functionalized NIR fluorescent dyes on magnetic NPs for optical bioimaging and MRI	114
Fluorescently doped SiNPs for use in bimodal (PET and fluorescent) imaging of lymph nodes	115
Dual-modal upconversion fluorescence and X-ray imaging using NaLuF ₄ :Gd,Yb,Er nanorods for blood vessel visualization	116
PEGylated NaLuF ₄ :Yb,Er upconversion nanophosphors for in-vivo fluorescence and X-ray imaging	117
Photosensitizer-incorporated G-quadruplex DNA-functionalized magnetofluorescent NPs for targeted magnetic resonance and fluorescence imaging and subsequent photodynamic therapy of cancer	118
Combined delivery of antigens and CpG by lanthanide-based core-shell NPs for enhanced immuno response and dual mode imaging	119
¹⁸ F-Labeled magnetic upconversion NPs prepared via rare-earth cation-assisted ligand assembly	120
Bimodal magnetic resonance (MRI) and fluorescence imaging of intracranial glioblastoma using NP of the type NaYF ₄ :Yb,Tm,Gd@oleate and a surface modified with HS-PEG-NH ₂	121
Upconverting NPs as optical imaging nanoprobe and T1 MRI contrast agents	122
Rhodamine dye conjugated to gold NPs; used as labels for bimodal SERS and 3D fluorescence studies on live endothelial cells; reveals inhomogeneous distribution in the cytoplasm	123
Hydroxyapatite NPs doped with Tb(III) and Gd(III) ions for fluorescence and magnetic bimodal imaging	124
Controlled synthesis of uniform and monodisperse upconversion NPs with a mesoporous silica shell for bimodal imaging	125
Upconverting NPs for optical imaging and as a T1 MRI contrast agent	126

Table S8. Representative examples of nanoparticles (NP) for use in drug release and gene delivery

<i>Methods and materials</i>	<i>Ref.</i>
Mesoporous titania NPs functionalized with flavin mononucleotide and loaded with the cancer drug doxorubicin used for imaging drug delivery in breast cancer cells BT-20	127
Mesoporous strontium hydroxyapatite; if capped with a respective aptamer, it serves as a nanovehicle for both cancer cell-responsive drug delivery and optical imaging	128
Upconversion luminescence cell imaging combined with drug-storage and drug release using particles consisting of an upconversion luminescent core and a mesoporous silica shell	129
Upconversion NPs for fluorescent labeling and targeted delivery of siRNA; folic acid and anti-Her2 antibody conjugated to the NPs and used to label folate receptors of cells	130

Upconversion NPs coated with mesoporous silica for imaging and PDT	131
Aptamer-directed synthesis of multifunctional lanthanide-doped porous nanoprobe for targeted imaging and drug delivery	132
Upconversion NPs in mesoporous silica used for plasmon-enhanced luminescence imaging and NIR light triggered drug release	133
Water-stable NaLuF ₄ -based upconversion nanophosphors for multimodal (fluorescence, CT, MR) imaging of lymph; stable for 6 months	134
Amino-modified lanthanide-doped KGdF ₄ NPs capped with PEI; acting as FRET bioprobes for avidin at a concentration of 5.5 nM; also acting as a MRI contrast agent in bimodal imaging	135
DNA-functionalized upconversion NPs for nanoassembly, DNA delivery, and fluorescence imaging	136
NIR-light triggered anticancer drug delivery by upconverting NPs with azobenzene-modified mesoporous silica	137
Doxorubicin loaded into NaYF ₄ :Yb,Tm NPs for drug delivery and imaging by luminescence resonance energy transfer	138
Tween-coated NaYF ₄ :Yb,Er@NaYF ₄ core/shell upconversion NPs for bioimaging and drug delivery	139
Upconversion luminescent NPs with mesoporous silica shell for anti-cancer drug delivery and cell imaging.	140
Upconversion luminescent NPs with two silica shells (the second being mesoporous) for imaging and drug storage/release	141
Magnetic resonance and fluorescence imaging of doxorubicin-loaded and dextrane coated NPs	142
Hydroxyapatite NPs doped with Eu(III) and Gd(III) ions dual imaging of drug delivery	143
LaF ₃ :Yb,Tm coated with SiO ₂ for folic acid-directed targeting of cancer cells; bimodal imaging by upconversion luminescence and X-ray computer tomography	144
Lanthanide-doped upconverting luminescent NPs for optical imaging, guided drug delivery and therapy	145
Urethane-doped biodegradable photoluminescent polymers; typical size 100 nm; obtained by nanoprecipitation; loaded with the drug 5-fluorouracil	146

Table S9. Representative examples of nanoparticles (NP) for use in photodynamic (PDT), photothermal (PTT) and other therapies

<i>Methods and materials</i>	<i>Ref.</i>
Review on upconversion nanomaterials for bioimaging and in vivo therapy	147
Upconversion imaging and photothermal therapy using NaYF ₄ :Yb,Er@oleylamine NPs with a shell of silver metal	148
Upconversion NPs coupled to a photosensitizers for NIR-triggered photodynamic therapy and imaging	149
Rare-earth functionalized reduced graphene oxide for tracking and photothermal killing of drug-resistant bacteria	150
30 nm-sized nanohybrid upconversion nanocrystals capped with a PEG derivative and loaded with a photosensitizer; imaging and PDT of cancer cells	151
Carbon nanodots for fluorescent bioimaging and targeted PDT; uses fluorescent, folic acid (FA)-functionalized carbon nanodots as carriers for the photosensitizer Zn(II)phthalocyanine	152
Aptamer-guided G-quadruplex DNA nanopatform for targeted bioimaging and PDT; capable of selective recognition and imaging of cancer cells, and of controllable activation of the photosensitizer	153
Targeted bioimaging and PDT of cancer cells with an activatable fluorescent probe; uses a new red-emissive bioprobe (a dicyanomethylene-tetraphenylethylene) and a cancer cell-specific peptide	154
Mesoporous silica-coated NaYF ₄ :Yb,Er nanocrystals for in vitro bioimaging and PDT of cancer cells	155
In-vivo PDT using upconversion NPs coated with dye-doped mesoporous-silica functioning as a remotely controlled nanotransducer	156
Core-shell hybrid nanogel capable of (a) optical sensing of T, (b) targeted tumor cell imaging, and (c) combined chemo-photothermal treatment	157

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