Multicolor Emission through Bi-directional Energy Transfer in Nd3+-Sensitized Gd3+-based Core/Shell/Shell Upconverting Nanoparticles

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Unique properties presented by Lanthanide (Ln3+)-doped upconverting nanoparticles (UCNPs) are at the core of the widespread applications in solar cells, security inks, biosensing, clinical applications and bioimaging (both in vitro and in vivo), for instance. The possibility of multicolor upconversion luminescence (UCL) under conventional single NIR wave-length excitation at ~ 980 nm, enriches these motivations. However, the 980 nm laser, generally applied to trigger the Yb^{3+}-sensitized upconversion processes is strongly absorbed by water molecules in biological structures causing severe overheating effects that damage cells and tissues. One strategy to overcome this problem is to shift the excitation wave-length to a shorter wavelength where the absorption of water molecules is greatly reduced. Moreover, in UCNPs, the energy transfer in bi-directional manner is more important than uni-directional one as the later induces detrimental cross-relaxation between the activator ions, leading to poor UCL. We demonstrate here the tailoring of multicolor UCL from a Nd^{3+}-sensitized Gd^{3+}-based core/shell/shell UCNPs with an architecture represented as $NaGdF_4$: Tm^{3+} (0.75) / Yb^{3+} (40) / Ca^{2+} (7)/ Nd^{3+} (1) @ $NaGdF_4$: Ca^{2+} (7) / Nd^{3+} (30) @ $NaGdF_4$: Yb^{3+} (40) /Ca^{2+} (7) /Nd^{3+} (1) /Er^{3+} (X = 1,2,3,5,7). Such UCNPs can be excited at a single wavelength (~ 808 nm) without generation of any local heating. Incorporation of substantial Nd^{3+}-sensitizers with proper concentration in the middle layer allows efficient harvesting of excitation light which migrates bi-directionally across the core/shell interfaces in sync to produce blue emission from Tm^{3+} (activator) ions in the core as well as green and red emission from Er^{3+} (activator) ions in the outermost shell. By simple and precise tuning of the content of Ln^{3+} ions in each domain, a multicolor UCL can be produced, ranging from blue to white. We envision that our Nd^{3+}-sensitized multicolor UCNPs are potential candidates for a variety of multiplexed biological applications, without impediment of any heating effect.

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