Supplementary Figures: 'FRET-reporter nanoparticles to monitor redoxinduced intracellular delivery of active compounds'



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Fig. S1 Electrokinetic measurements (zeta potential) of the plain silica cores ($nSiO_2$), coreshell particles ($nSiO_2@mSiO_2$), PEI-functionalized silica cores ($nSiO_2@PEI$) and PEIfunctionalized core-shell particles ($nSiO_2@mSiO_2@PEI$).



Fig. S2 Thermogravimetric analysis (TGA) of the plain silica cores $(nSiO_2)$, core-shell particles $(nSiO_2@mSiO_2)$, PEI-functionalized silica cores $(nSiO_2@PEI)$ and PEI-functionalized core-shell particles $(nSiO_2@mSiO_2@PEI)$.



Fig. S3 Loss of red emisson from the cleavable particles, while no loss of red emission from the non-cleavable control particles, was observed after treatment with reducing agents. Particles were incubated for one hour in test tubes with a concentration of a reducing agent (10 mM gluthatione, GSH) corresponding to intracellular conditions at endosomal pH. As comparision equal concentration of an artifical reducing agent (DTT) was introduced at pH 5 and pH 7. The particles were dried on glass slides and imaged with a Nikon Microphot-FXA light microscope using UV light for excitation and a dual FITC-TRITC filter, which simultaneously gave rise to emission from both of the fluorescenct dyes. Figure (a) represent the cleavable particles before introducing reducing agent, while (b-d) show the cleavable particles after introducing cleavable agents; DTT at pH 7 (b), DTT at pH 5 (c), and GSH at pH 7 (f), DTT at pH 5 (g), and GSH at pH 5 (h).



Fig. S4 No loss of FRET was observed for the non-cleavable control particles after uptake into HeLa cells. (a) Bright field images of cells incubated with silica particles that were labeled with a non-cleavable FRET-pair. The ratio donor:acceptor was kept 1:10. (b) FRET images of cells shown in (a) with FRET-level on silica particles color coded from yellow (low) to black (high). (c) Histograms of FRET-ratio per pixel of adjacent images shown in (b). Scale bars in (a) are also representative for (b) and are 10 µm.