

Electronic Supplementary Information

Experimental section

Synthesis of Pas-DTPA-APTS: Diethylenetriaminepentaacetic acid (DTPA) dianhydride (0.18 g) was dissolved in the mixture of 2.0 mL of dimethyl sulfoxide (DMSO) and 1.0 mL of triethylamine. A total of 0.0875 g of p-aminosalicylic acid (Pas) in 2.0 mL of DMSO was added to the above solution dropwise via a syringe. After 60 min of vigorous stirring, 100 μ L of 3-aminopropyltriethoxysilane (APTS) was added to the above solution. The reaction was stirred at room temperature for 5 h. Then the obtained precipitate was collected by centrifugation and further washed with DMSO and anhydrous ethanol. The solid product was dried in a desiccator and confirmed by mass spectroscopy (689, M^+ , ESI).

Synthesis of nanoparticle: Core-shell silica nanoparticles were synthesized in the w/o microemulsion system.^{1,2} The microemulsion consisted of a mixture of 8.9 mL of Triton X-100, 9.0 mL of **n-hexanol**, 37.5 mL of cyclohexane, 2.0 mL of deionized water, 2.0 mg of Tris (2, 2'-bipyridyl) dichlororuthenium (II) (RuBpy), and 0.75 mL of ammonium hydroxide that was stirred for 30 min, and then 0.25 mL of TEOS was added. After 24 h of stirring, 70 mg of Pas-DTPA-APTS dissolved in 1.5 mL of deionized water was added to the above solution and stirred for 30 min, and then 0.25 mL of TEOS was added. The mixture was then allowed to stir for another 36 h. After the reaction was complete, silica nanoparticles were isolated from the microemulsion using acetone, centrifuged and further washed with ethanol and deionized water several times to remove surfactant and superfluous dye molecules. The obtained nanoparticles were finally redispersed in 20 mL deionized water for further use.

Silica nanoparticles post-functionalized with Tb^{3+} ions: Different concentrations of Tb^{3+} ions were added to a plastic tube containing 0.2 mL of the above silica nanoparticles and 4 mL deionized water. The reaction was complete after approximately 10 h. Then the silica nanoparticles were collected by centrifugation and redispersed in deionized water for fluorescence measurements.

Silica nanoparticles post-functionalized with Gd^{3+} and Tb^{3+} ions: Different concentration ratios of Tb^{3+} and Gd^{3+} ions (0.1 mg, Tb^{3+} : Gd^{3+} = 0:100, 5:95, 10:90, 15:85, 20:80) were added to plastic tube containing 0.2 mL of the above silica nanoparticles and 4 mL deionized water. Then the silica nanoparticles were collected by centrifugation and redispersed in deionized water for fluorescence measurements.

Characterization of particles: A Shimadzu RF-5301pc spectrofluorometer was used for recording the fluorescence spectra. All fluorescence spectra were performed at room temperature by using a Xenon lamp as the source of excitation and a 3 mL standard quartz cuvette (10 mm $\times$ 10 mm). Particle characterizations were performed on a Tecnai F30 electron microscope and a LEO 1530 electron microscope. For the high **resolution** transmission electron microscopy (HRTEM), sample preparation was done by placing a drop of the freshly prepared colloidal solution on a carbon coated copper grid and the excess solution then removed. The distributions of the particle size were determined from TEM images of at least 100 particles.

ICP-MS measurements: Silica nanoparticles **post-functionalized** with different concentration ratios of Tb^{3+} and Gd^{3+} ions were centrifuged and further washed with deionized water several times to remove superfluous ions. The obtained silica nanoparticles were dissolved in 1 mL HNO_3 and HF mixture (2 wt.-%). Then the digested solution was diluted with 2 wt.-% HNO_3 solution for ICP-MS measurements. A HP 4500 ICP-MS (Yokogawa, Japan) was used for the measurements.

References

- 1) S. Santra, R. P. Bagwe, D. Dutta, J. T. Stanley, G. A. Walter, W. Tan, B. M. Moudgil and R. A. Mericle, *Adv. Mater.* 2005, **17**, 2165.
- 2) R. P. Bagwe, C. Yang, L. R. Hilliard and W. Tan, *Langmuir* 2004, **20**, 8336.