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## **Controlling the Dominant Magnetic Relaxation Mechanisms for Magnetic Hyperthermia in bimagnetic core-shell nanoparticles**

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Figure S1: TEM images of the Fe<sub>3</sub>O<sub>4</sub> core and Fe<sub>3</sub>O<sub>4</sub>/Zn<sub>x</sub>Co<sub>1-x</sub>Fe<sub>2</sub>O<sub>4</sub> core/shell samples.



Figure S2: Size distribution of the Fe<sub>3</sub>O<sub>4</sub> core and Fe<sub>3</sub>O<sub>4</sub>/Zn<sub>x</sub>Co<sub>1-x</sub>Fe<sub>2</sub>O<sub>4</sub> core/shell with x = 0.00, 0.25, 0.50, 0.75 and 1.00. The histograms where fitted with a lognormal distribution (red line) in order to determine the mean diameter and dispersion size which are shown in Table 1. The blue line is the size distribution calculated from  $f(T_B)$  of the ZFC and FC magnetization curves.



Figure S3: High Resolution TEM image of the core/shell sample with x = 1.00 (a), electron diffraction of the core/shell sample with x = 0.50 (b) and dark field TEM images reconstructed from a fraction of the (133) spinel diffraction ring of the core/shell samples with x = 0.25 and x = 0.50 (c) and (d), respectively, where the core-shell structure is evidenced.



Figure S4: Hydrodynamic diameter of as-made hydrophobic core-shell Nanoparticles measured by DLS in Hexane fitted with lognormal distribution.



Figure S5: (a) Hydrodynamic diameter of PEG-coated hydrophilic  $Fe_3O_4/Zn_xCo_{1-x}Fe_2O_4$  core-shell nanoparticles dispersed in DMEM measured by DLS.(b) Magnetic fluid hyperthermia measurement of 0.5 % wt nanoparticles dispersed in DMEM with an applied field of 200 Oe and 570 kHz. (c) SPA values as function of Zn content as obtained from hyperthermia experiments of hydrophilic nanoparticles in DMEM.



Figure S6: Diagram of the relaxation time mechanism for the Fe<sub>3</sub>O<sub>4</sub>/Zn<sub>x</sub>Co<sub>1-x</sub>Fe<sub>2</sub>O<sub>4</sub> core-shell nanoparticles dispersed in hexane ( $\eta \sim 0.3$  mPa.s), butter oil ( $\eta \sim 477$  mPa.s) and Fe<sub>3</sub>O<sub>4</sub>/Zn<sub>x</sub>Co<sub>1-x</sub>Fe<sub>2</sub>O<sub>4</sub> core-shell nanoparticles coated with PEG dispersed in DMEM ( $\eta \sim 0.94$  mPa.s).

Table 1: Values of  $h = \frac{H_0}{H_k(f,T)} \ll 1$  and  $\xi = \frac{M_S V H_0}{k_B T} \ll 1$  calculated for all the nanoparticles systems, where  $H_k(f,T) = \frac{2K}{M_S} \left( 1 - \sqrt{\frac{T}{T_B(f)}} \right).$   $\frac{\text{Sample } x = 0.00 \quad x = 0.25 \quad x = 0.50 \quad x = 0.75 \quad x = 1.00}{h \quad 8.6 \times 10^{-2} \quad 7.8 \times 10^{-2} \quad 8.8 \times 10^{-2} \quad 1.6 \times 10^{-1} \quad 7.3 \times 10^{-1}}{\xi \quad 1.47 \quad 1.27 \quad 1.15 \quad 1.08 \quad 1.30}$