Supporting Information
Iron Oxide Nanospheres: Dual Functionality as MRI Contrast Agents and Magnetic Fluid Hyperthermia Therapeutics

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Atomic Force Microscopy (AFM)

The Atomic Force Microscopy (AFM) images of all the studied samples are shown in Figure 1. Details about the instrumentation, technique, and quantitative analyses of the AFM images (Table 2), are reported in the main text.

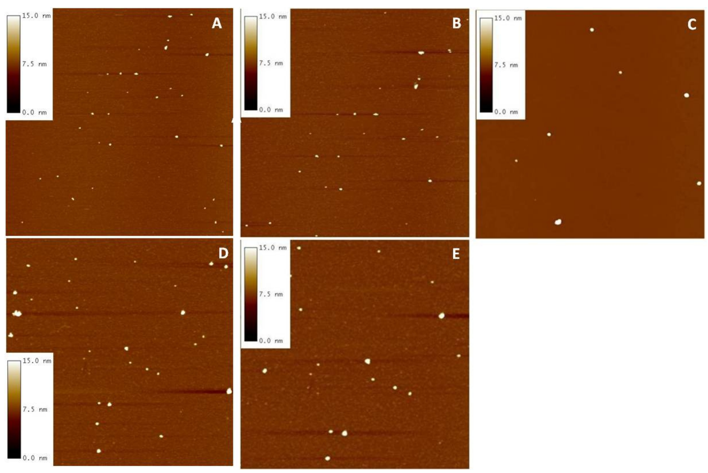


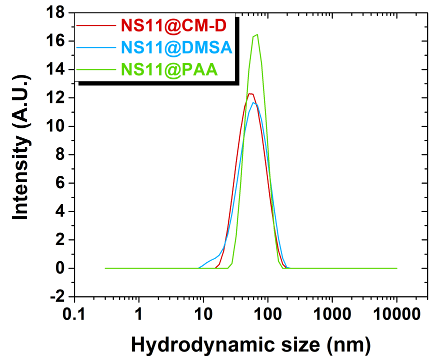
Figure 1 AFM topography images of all the studied MNPs: A) NS11@CMD, B) NS11@PAA, C) NS11@DMSA, D) NS14@CMD and E) NS14@PAA. Single particles, as well as MNPs small clusters, were observed on the mica support; all the images were collected in air, in tapping mode. Scan area: 1x1 μm^2. Range of the vertical color scale: 0–15 nm.

Dynamic Light Scattering (DLS)

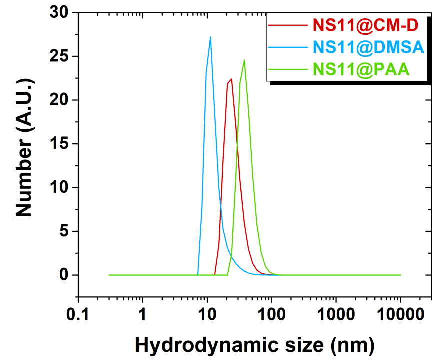
The Dynamic Light Scattering (DLS) acquisitions were performed with a Malvern Instruments Zetasizer Ultra equipped with a solid-state He–Ne laser (wavelength λ = 633 nm), to evaluate the hydrodynamic size of the studied MNPs and their surface charge. Samples for the hydrodynamic size evaluation are constituted of a rectangular box DTS1002 containing 1 mL of milliQ water and 1 drop of the investigated dispersion of MNPs. The acquisitions were repeated halving the dispersion concentration several times, and the recorded acquisition refers to a maximized number of counts and a minimized Polydispersity Index (PDI). The acquisitions were conducted at room temperature: 2 minutes of calibration and 1 minute of acquisition. Samples used to acquire the Z-potential at a pH $\sim$ 7 were prepared as follows: a few drops of MNPs dispersion were mixed with KNO_{-3} (10^{-2} M) within a glass tube, to enhance the volume without modifying the number of ions. The pH is decreased to $\sim$ 7 by means of HNO_{-3} and the dispersion is poured into a DTS1060C box. The measurements were performed at room temperature: 2 minutes of calibration and 3 minutes of measurements (3 acquisitions). The hydrodynamic size distributions are displayed in Figure 2, while the obtained values are listed in Table 1, together with the Z-potential results.

Table 1 Hydrodynamic size (intensity and number) with Polydispersity Index (PDI) and Z-potential at the listed pH value of the studied samples.

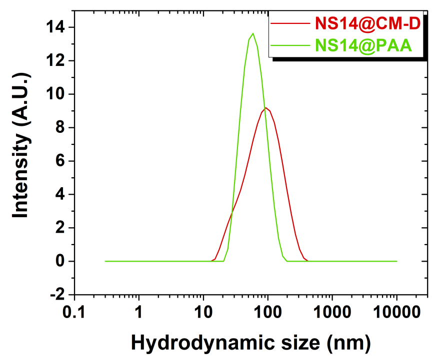
Sample	Hydrodynamic size (nm)			Z-potential (mV)	
	Intensity	Number	PDI	pH	Z-pot
NS11@CM-D	49	38	0.23	6.7	-32.7
NS11@DMSA	51	11	0.20	6.6	-43.4
NS11@PAA	64	24	0.22	6.9	-46.8
NS14@CM-D	71	20	0.27	6.2	-30.1
NS14@PAA	56	28	0.19	7.1	-46.2



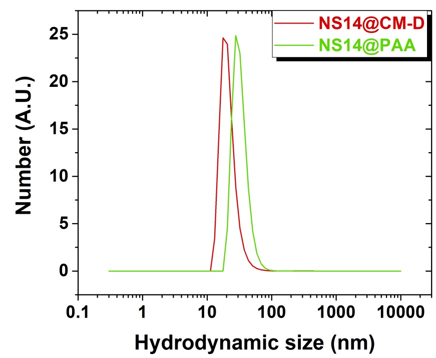
(a) NS11 - Intensity



(b) NS11 - Number



(c) NS14 - Intensity



(d) NS14 - Number

Figure 2 Hydrodynamic size distributions of the NS11 coated particles (a, b), and of the NS14 coated particles (c, d). Figures a and c show the intensity distribution, while b and d show the number distribution. The intensity distribution is the primary result. The number distributions are based on calculations, taking into account the optical properties of the particles measured.

Roch-Müller-Gillis heuristic model

The heuristic low-intermediate anisotropy model of Roch-Müller-Gillis was utilized to fit the experimental $^{1}H-NMR-d$ profiles. Here is the utilized expression of r_1:

$$r_1 = \frac{1}{T_1} = F \times \left(\frac{1}{C_{Fe}}\right) \times \{7P\frac{L(x)}{x}J_{F_1} + \left[7Q\frac{L(x)}{x} + 3\left(1 - L^2(x) - 2\frac{L(x)}{x}\right)\right] \times J_{F_2} + 3L^2(x)J_A\} \tag{1}$$

where C_{Fe} is the iron ions concentration and the pre-factor F is given by

$$F = \left(\frac{32000\pi}{135}\right) \times 10^{-14}\mu^2\gamma_I^2\left(\frac{C_{MNP}}{DR}\right) \tag{2}$$

with μ the magnetic moment of a single particle, γ_I the proton gyromagnetic ratio, C_{MNP} the number of magnetic nanoparticles per liter, D the diffusion coefficient, and R the minimum approach distance. The other functions contained in the r_1 expression are:

- the Langevin function $L(x)$ with $x = \frac{\mu H_0}{k_B T}$.
- the Freed spectral density functions $J_{F_1}\left(\Omega(\omega_S, \omega_0), \tau_D, \tau_N\right)$ and $J_{F_2}\left(\omega_I, \tau_D, \tau_N\right)$.
- the Ayant spectral density function $J_A\left(\sqrt{\omega_I \tau_D}\right)$.

The term $\Omega(\omega_S, \omega_0)$ is introduced to represent the dispersion at low fields, and its empirical formulation is:

$$\begin{aligned} \Omega(\omega_S, \omega_0) &= \left(\omega_S^{1/4} - \omega_0^{1/4}\right)^4 \quad \text{if } \omega_S > \omega_0 \\ &= 0 \quad \text{if } \omega_S < \omega_0 \end{aligned} \tag{3}$$

where ω_0 is a factor dependent on the anisotropy energy E_a and it is null for $E_a = 0$. The parameters P and Q quantify E_a, being $P + Q \leq 1$. $P = 1$ and $Q = 0$ mark $E_a = 1$, while $P = 0$ and $Q = 1$ represent a huge E_a amount[?] .