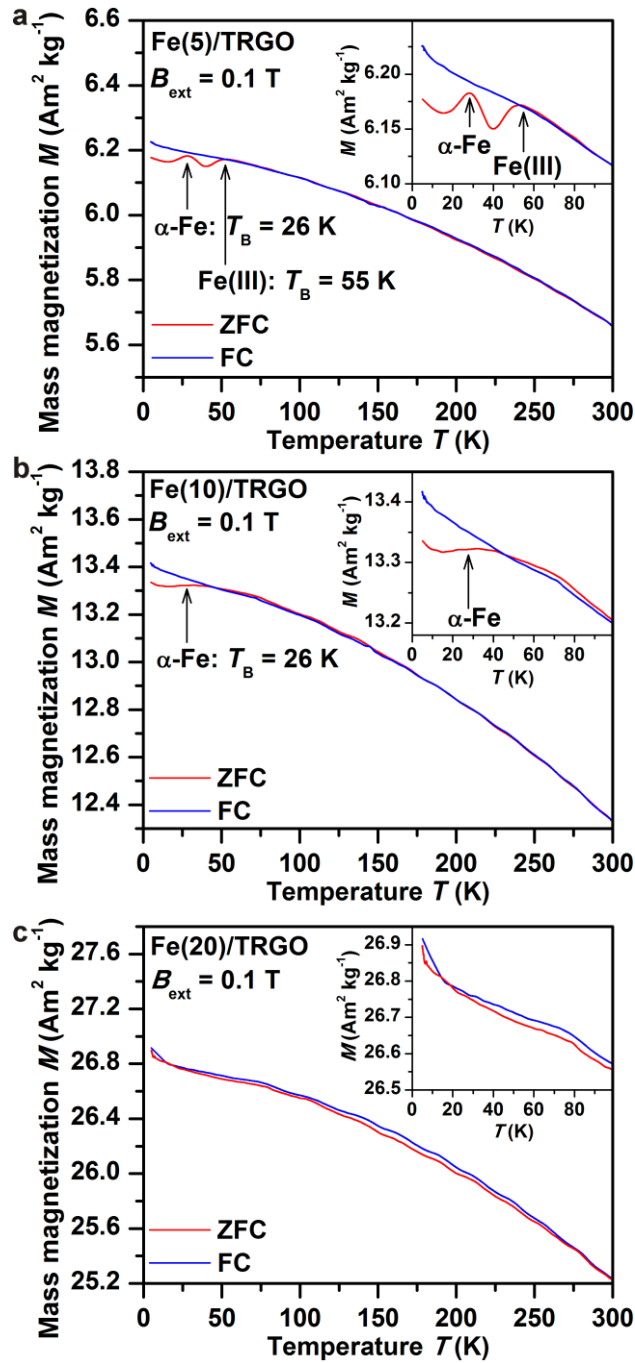
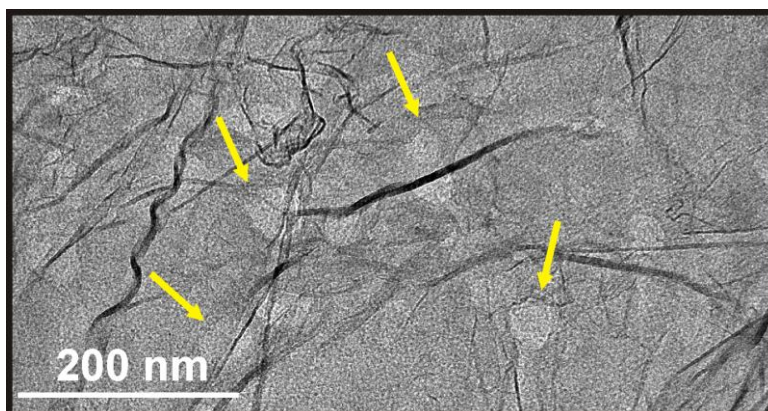


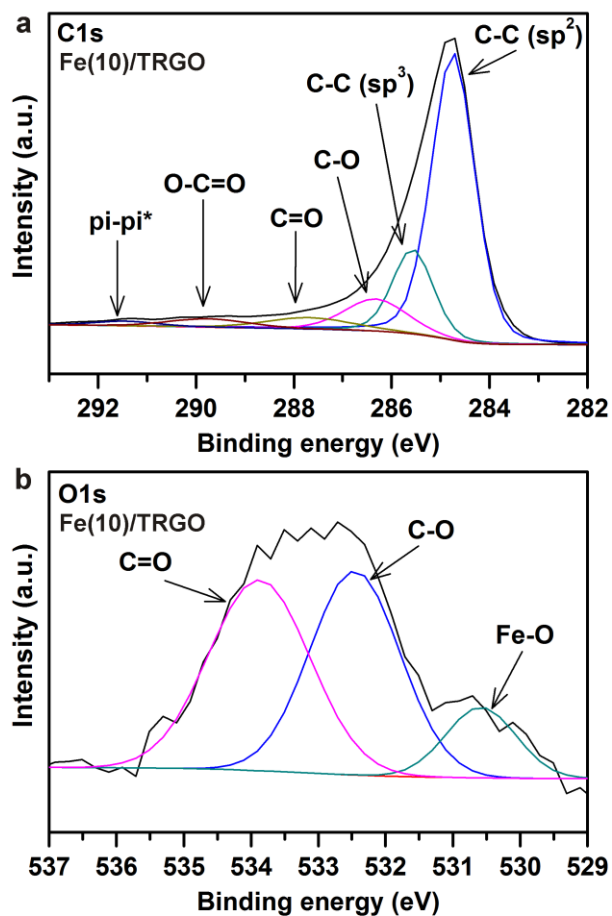


Supplementary Figure 1 | Results of magnetization measurements for the Fe(5)/TRGO, Fe(10)/TRGO, and Fe(20)/TRGO hybrids. (a) ZFC and FC magnetization curves of the Fe(5)/TRGO hybrid with two maxima identified in the ZFC magnetization curve. The maximum observed at a lower temperature ($\sim 26 \text{ K}$) corresponds to the magnetic blocking mechanism of the iron nanoparticle core with the most probable size in the system while the second maximum at

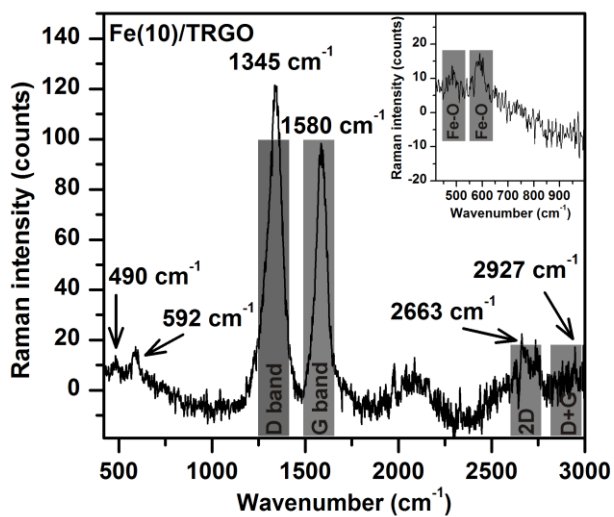
~55 K reflects the magnetic blocking of the nanoparticle surface shell of iron(III) oxide nature. (b) ZFC and FC magnetization curves of the Fe(10)/TRGO hybrid with one maximum at ~26 K related to the magnetic blocking of the iron nanoparticles. No other maximum is observed implying that Fe(III) shell is not completely developed and Fe(III) surface species undergo superparamagnetic/paramagnetic fluctuations even down to 5 K. (c) ZFC and FC magnetization curves of the Fe(20)/TRGO hybrid with profiles typical for bulk iron without any indication of transition to the superparamagnetic state.



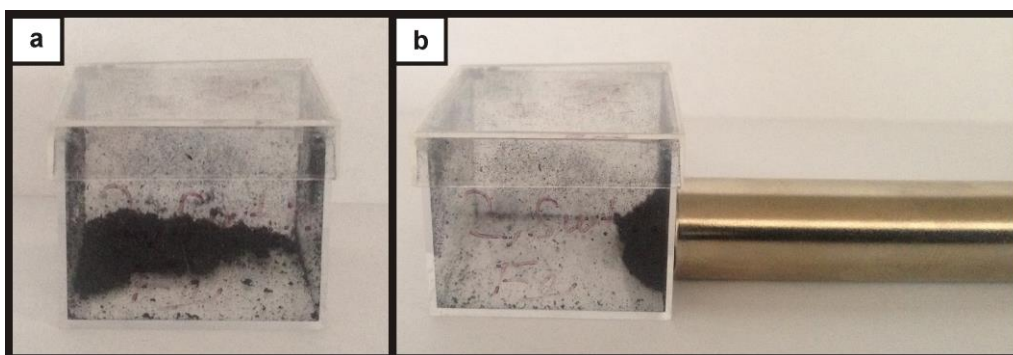
Supplementary Figure 2 | HRTEM image of cavities (indicated by yellow arrows) in the TRGO matrix (imaged without the use of iron precursor) that serve as nucleation sites for the formation/growth of iron (metal) nanoparticles.



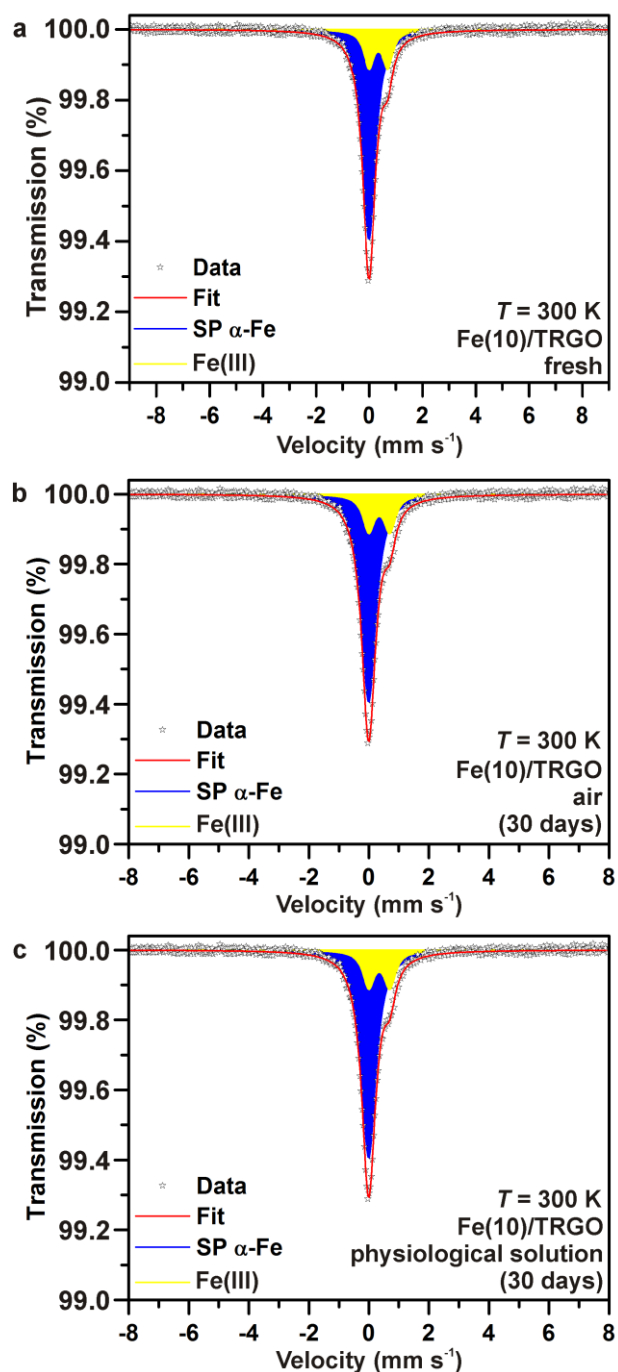
Supplementary Figure 3 | XPS spectra of the Fe(10)/TRGO hybrid. (a) C1s and (b) O1s XPS spectrum of the Fe(10)/TRGO hybrid.



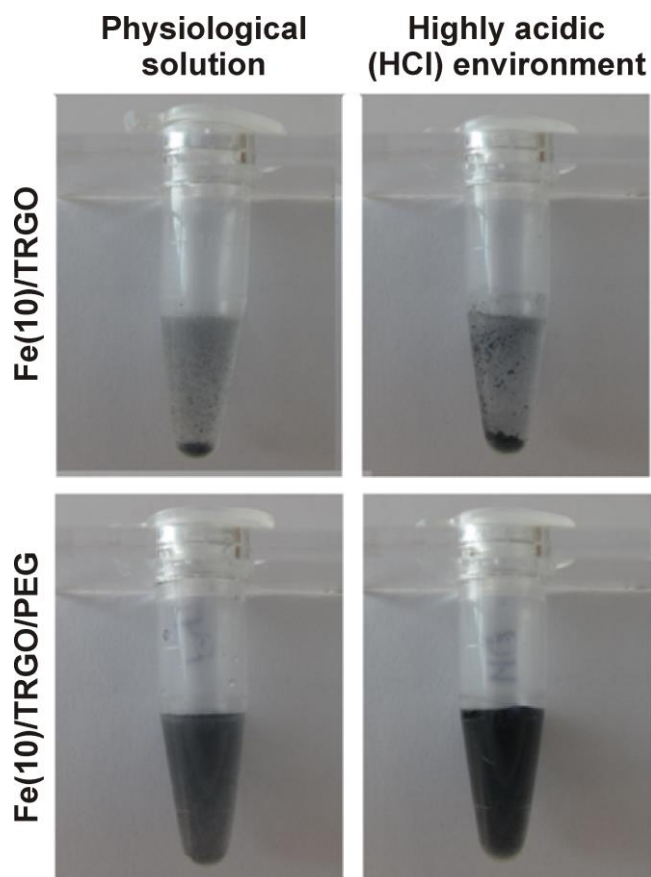
Supplementary Figure 4 | Raman spectrum of the Fe(10)/TRGO nanocomposite.



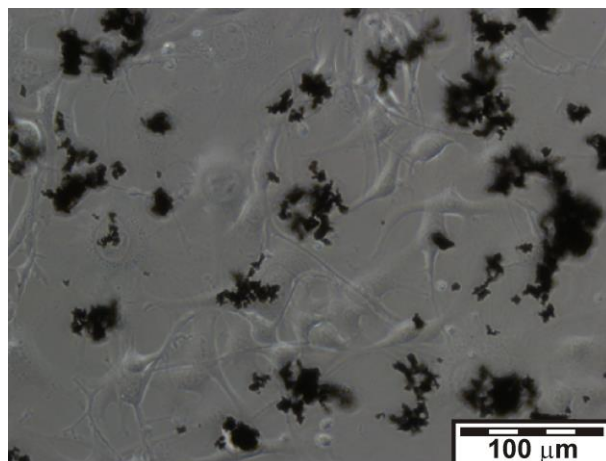
Supplementary Figure 5 | Photographs of the Fe(10)/TRGO hybrid. Photographs of the Fe(10)/TRGO nanocomposite **(a)** before and **(b)** after placing a simple hand magnet (with an induction of ~ 0.3 T) against the wall of the box.



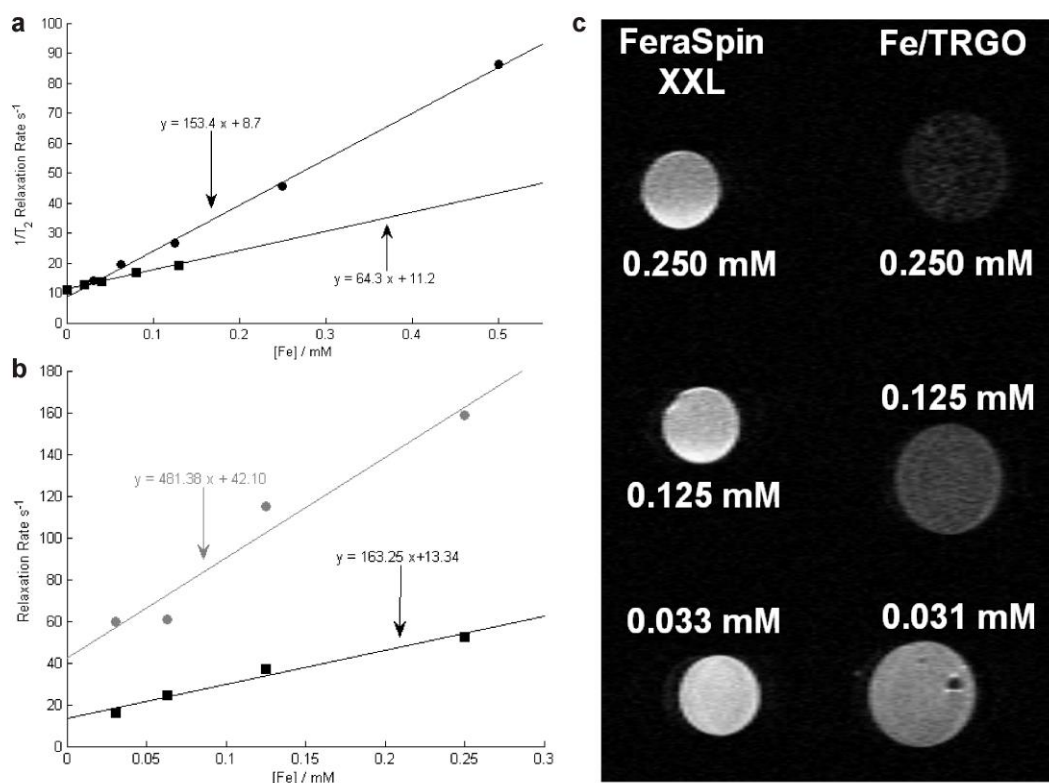
Supplementary Figure 6 | Chemical stability of the Fe(10)/TRGO hybrid under air conditions and in a physiological solution assessed by ^{57}Fe Mössbauer spectroscopy. Room-temperature ^{57}Fe Mössbauer spectra of (a) the freshly-prepared Fe(10)/TRGO hybrid after primary purification, (b) the hybrid after one month of storage under ambient laboratory conditions in the air, and (c) the hybrid after one month of storage in a physiological solution.



Supplementary Figure 7 | Colloidal stability of the Fe(10)/TRGO and Fe(10)/TRGO/PEG hybrid. Photographs comparing the colloidal stability of the freshly prepared Fe(10)/TRGO hybrid and a surface-modified Fe(10)/TRGO/PEG nanocomposite in physiological solution (left column) and a highly acidic (HCl) solution (right column). The concentration of the hybrid is 2 mg mL^{-1} in each case, and the pictures show the suspensions as they appeared 1 hour after hybrid dispersion. The lower images demonstrate the drastically improved colloidal stability of the PEGylated system.



Supplementary Figure 8 | Optical microscopy image showing adherent non-degraded NIH/3T3 mouse fibroblast cells after 24 hours of incubation with $200 \mu\text{g mL}^{-1}$ of Fe(10)/TRGO/PEG hybrid.

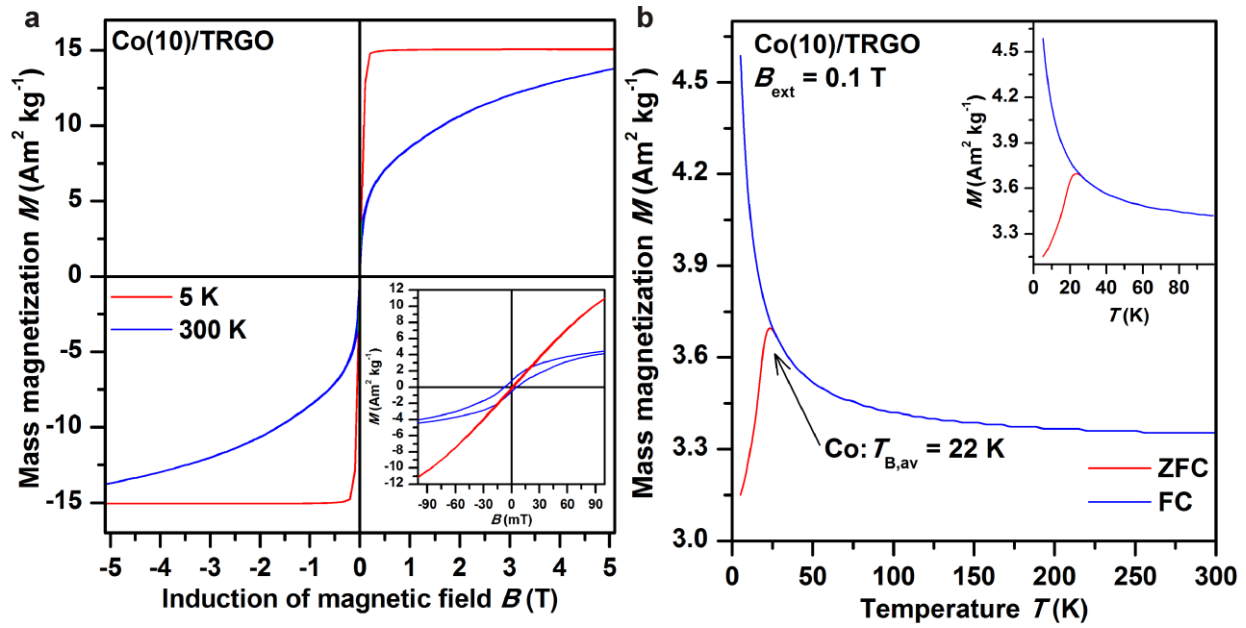


Supplementary Figure 9 | *In vitro* MRI measurements. (a) The transverse relaxation rates ($1/T_2$) versus iron concentration for Fe(10)/TRGO (circles) and commercial FeraSpin XXL (squares) nanosystems with corresponding linear fittings (solid lines) measured in a field of 9.4 T. The values of r_2 determined from these curves are $153 \text{ mM}^{-1} \text{ s}^{-1}$ for Fe(10wt.%)/TRGO and 64

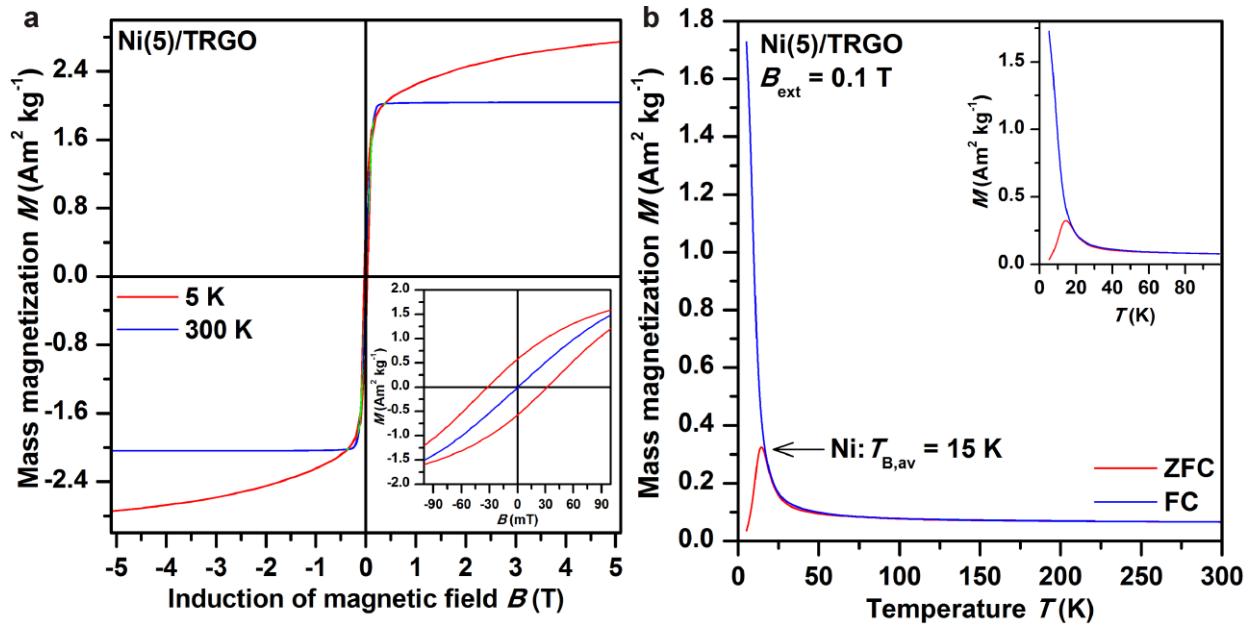
$\text{mM}^{-1} \text{ s}^{-1}$ for commercial FeraSpin XXL. **(b)** Plots of transverse relaxation rates ($1/T_2^*$ - grey circles) and ($1/T_2$ black squares) for Fe(10)/TRGO nanocomposite with corresponding linear fittings (solid lines) measured under a field of 4.7 T. The values of r_2^* and r_2 determined from these curves are $481.36 \text{ mM}^{-1} \text{ s}^{-1}$ and $163.25 \text{ mM}^{-1} \text{ s}^{-1}$, respectively. **(c)** Phantom experiments: comparison of negative contrast effect for FeraSpin XXL and Fe(10)/TRGO nanocomposite imaged by a 9.4 T MRI in T_2 weighted images and demonstrating remarkable differences in contrast properties.



Supplementary Figure 10 | Optical microscopy of Fe(10)/TRGO/PEG system before *in vivo* MRI application.



Supplementary Figure 11 | Results of magnetization measurements for the Co(10)/TRGO hybrid. (a) 5 K and 300 K hysteresis loops and (b) ZFC/FC magnetization curves recorded for the Co(10)/TRGO hybrid. The maximum in ZFC magnetization curve clearly demonstrating the passage of the Co nanoparticles from the high-temperature superparamagnetic state to the low-temperature magnetically blocked regime. The insets in panel a and b show behavior of the hysteresis loops of the Co(10)/TRGO hybrid around the origin and details of profile of the ZFC/FC magnetization curves of the Co(10)/TRGO hybrid at low temperatures, respectively.



Supplementary Figure 12 | Results of magnetization measurements for the Ni(5)/TRGO hybrid. (a) 5 K and 300 K hysteresis loops and (b) ZFC/FC magnetization curves recorded for the Ni(5)/TRGO hybrid. The maximum in ZFC magnetization curve clearly demonstrating the passage of the Ni nanoparticles from the high-temperature superparamagnetic state to the low-temperature magnetically blocked regime. The insets in panel a and b show behavior of the hysteresis loops of the Ni(5)/TRGO hybrid around the origin and details of profile of the ZFC/FC magnetization curves of the Ni(5)/TRGO hybrid at low temperatures, respectively.

Supplementary Table 1 | Mössbauer hyperfine parameter values. Mössbauer hyperfine parameter values determined by fitting the ^{57}Fe Mössbauer spectra of the unfiltered and filtered Fe(10)/TRGO nanocomposite samples, where T is the temperature of the measurement, δ is the isomer shift, ε_Q is the quadrupole shift (for doublet component), ΔE_Q is the quadrupole splitting (for sextet component), B_{hf} is the hyperfine field, and RA is the relative spectral area of individual spectral components.

Sample	T (K)	Component	$\delta \pm 0.01$ (mm s ⁻¹)	$\varepsilon_Q/\Delta E_Q \pm 0.01$ (mm s ⁻¹)	$B_{\text{hf}} \pm 0.3$ (T)	RA ± 1 (%)	Assignment
unfiltered Fe(10)/ TRGO	300	Sextet	0.00	0.00	32.9	31	α -Fe bulk
		Doublet	0.34	0.70	----	27	Iron oxide shell
		Singlet	0.00	----	----	42	α -Fe – SP
filtered Fe(10)/ TRGO	300	Singlet	0.00	----	----	79	α -Fe – SP
		Doublet	0.35	0.71	----	21	Fe ³⁺ environment
	50	Sextet	0.08	0.00	33.1	32	α -Fe – blocked
		Singlet	0.09	----	----	46	α -Fe – SP
		Doublet	0.43	0.75	----	22	Fe ³⁺ environment
	5	Sextet	0.11	0.00	33.2	65	α -Fe – blocked
		Singlet	0.11	----	----	13	α -Fe – SP
		Doublet	0.50	0.72	----	22	Fe ³⁺ environment

Supplementary Table 2 | Mössbauer hyperfine parameter values assessing the chemical stability of the Fe(10)/TRGO hybrid. Mössbauer hyperfine parameters derived from the room-temperature ^{57}Fe Mössbauer spectra of the as-prepared Fe(10)/TRGO hybrid, the Fe(10)/TRGO hybrid after one month of storage under ambient laboratory conditions in the air, and the Fe(10)/TRGO hybrid after one month of storage in physiological solution. δ is the isomer shift, ΔE_Q is the quadrupole splitting, and RA is the relative spectral area of individual spectral components identified during fitting.

Sample	Component	$\delta \pm 0.01$ (mm s ⁻¹)	$\Delta E_Q \pm 0.01$ (mm s ⁻¹)	RA ± 1 (%)	Assignment
as-prepared	Singlet	0.00	0.00	79	SP iron
	Doublet	0.35	0.71	21	Fe ³⁺ species
one month in air	Singlet	0.00	0.00	77	SP iron
	Doublet	0.34	0.72	23	Fe ³⁺ species
one month in physiological solution	Singlet	0.00	0.00	78	SP iron
	Doublet	0.35	0.69	22	Fe ³⁺ species

Supplementary Table 3 | Relaxivity indices (r_1 and r_2) of the most frequently used contrast agents under a field of 4.7 T. Data adopted from Rohrer, M., Bauer, H., Mintorovitch, J., Requardt, M. & Weinmann, H. J. Comparison of magnetic properties of MRI contrast media solutions under a field of 4.7 T. *Invest. Radiol.* **40**, 715-724 (2005).

Trade name or international code	r_1 (mM ⁻¹ s ⁻¹)	r_2 (mM ⁻¹ s ⁻¹)	r_2/r_1
MAGNEVIST	3.2 (3.0 – 3.4)	4.0 (3.8 – 4.2)	1.3
GADOVIST	3.2 (3.0 – 3.4)	3.9 (3.7 – 4.1)	1.2
PROHANCE	2.8 (2.7 – 2.9)	3.7 (3.5 – 3.9)	1.3
MULTIHANCE	4.0 (3.8 – 4.2)	5.0 (4.7 – 5.3)	1.3
DOTAREM	2.8 (2.7 – 2.9)	3.7 (3.5 – 3.9)	1.3
OMNISCAN	3.3 (3.1 – 3.5)	4.1 (3.9 – 4.3)	1.2
TESLASCAN	1.6 (1.5 – 1.7)	2.7 (2.6 – 2.8)	1.7
OPTIMARK	3.8 (3.5 – 4.1)	4.7 (4.5 – 4.9)	1.2
RESOVIST	2.8 (2.7 – 2.9)	176 (167 – 185)	62.9
FERIDEX	2.3 (2.2 – 2.4)	105 (100 – 110)	45.7
GADOMER	9.1 (8.6 – 9.6)	22 (21 – 23)	2.4
MS-325	5.5 (5.2 – 5.8)	6.9 (6.5 – 7.3)	1.3
PRIMOVIST	4.9 (4.7 – 5.1)	6.3 (6.0 – 6.6)	1.3
SH U 555 C	4.3 (4.1 – 4.5)	66 (63 – 69)	15.3