Cobalt Ferrite Supported on Reduced Graphene Oxide as a T_2 Contrast Agent for Magnetic Resonance Imaging

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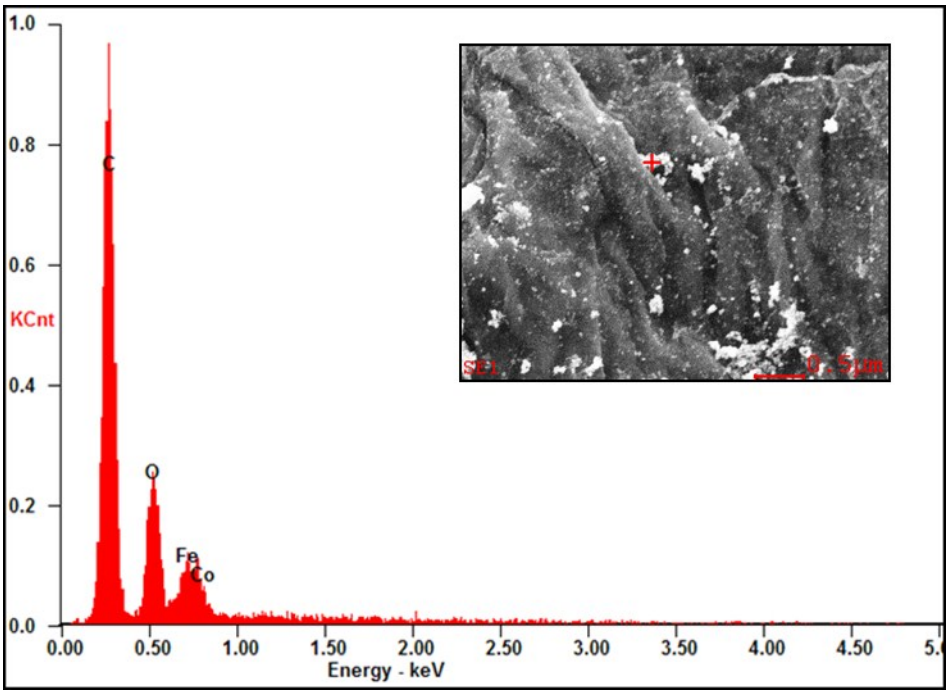

Figure SI1. EDS analysis of a representative $CoFe_2O_4$-rGO composite (the corresponding SEM image and probe marker are inset).

Table SI1. Concentration of Fe in the $CoFe_2O_4$ nanoparticles and composites, as measured by ICP-OES.

Sample	Fe (wt%)
$CoFe_2O_4$	36.5
rHGO- 5 %	2
rHGO – 10 %	3
rHGO- 16 %	5
rHGO -30 %	9
rIGO- 5 %	3
rIGO – 10 %	4
rIGO- 16 %	5
rIGO -30 %	15

Table SI2. Average particle size (Dav) and respective standard deviation (σ) for the $CoFe_2O_4$ NPs, $CoFe_2O_4$-rHGO composites and $CoFe_2O_4$-rIGO composites (as estimated from TEM images).

Sample	Particle size	
	Dav (nm)[a]	σ (nm)[b]
$CoFe_2O_4$	12.3	2.6
rHGO-5%	3	1
rHGO -10%	3	0.8
rHGO -16%	4	1
rHGO -30%	7	2
rIGO -5%	5	1
rIGO -10%	7	1
rIGO -16%	9	3
rIGO -30%	12	2

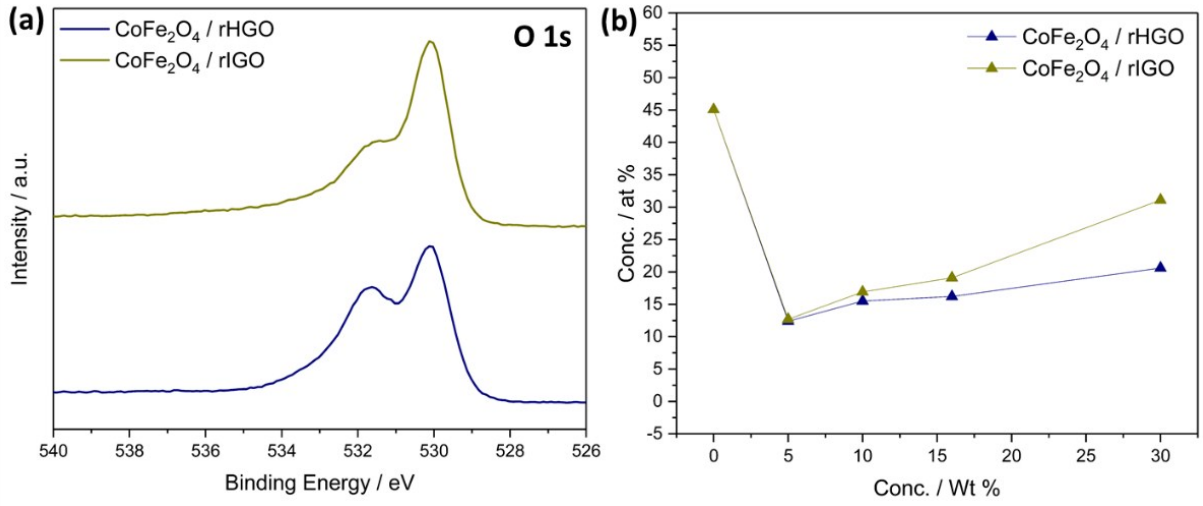

Figure SI2. (a) High-resolution O1s XPS spectra of the two 30% rGO composites; (b) Concentration of the surface oxygen atomic percentage as a function of the $CoFe_2O_4$ loading (data extracted from the corresponding XPS spectra).

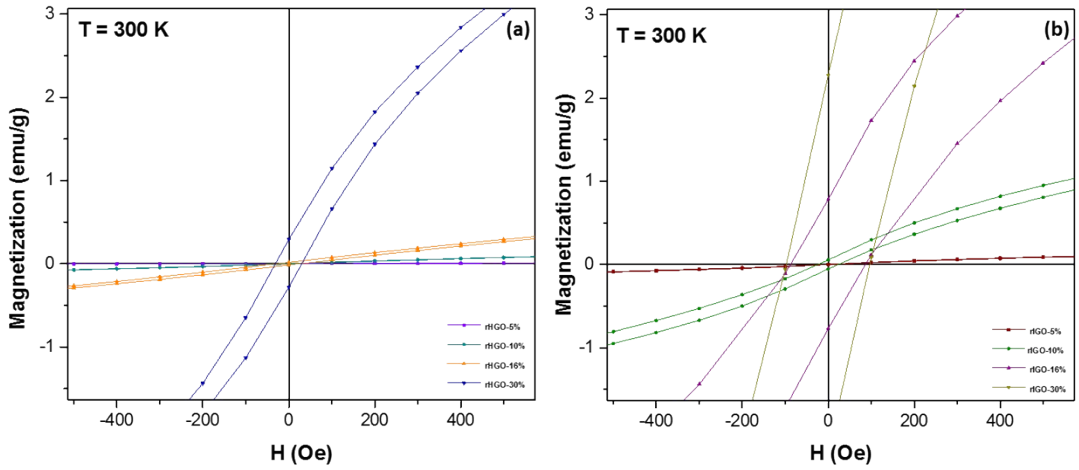

Figure SI3. Low-field regions of the hysteresis loops at room temperature for: (a) $CoFe_2O_4$-rHGO composites and (b) $CoFe_2O_4$-rIGO composites.

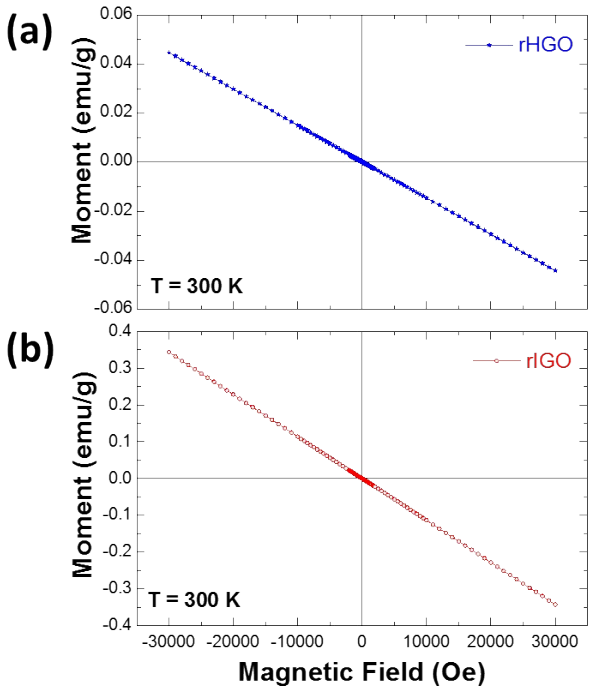

Figure SI4. Room temperature field-dependent magnetic measurements for: (a) rHGO and (b) rIGO.

Table SI3. Magnetization at 30000 Oe ($M_{H=30000\ Oe}$), saturation magnetization (M_r) and magnetic coercivity (H_c) for the $CoFe_2O_4$ nanoparticles, $CoFe_2O_4$-rHGO composites and $CoFe_2O_4$-rIGO composites (extracted from the M vs. H loops).

$CoFe_2O_4$ Loading wt%	$CoFe_2O_4$ 5%		$CoFe_2O_4$ 10%		$CoFe_2O_4$ 16%		$CoFe_2O_4$ 30%		$CoFe_2O_4$ 100%
Composite	rHGO	rIGO	rHGO	rIGO	rHGO	rIGO	rHGO	rIGO	-
$M_{H=30000\ Oe}$ (emu/g)	0.08	0.50	0.84	4.70	2.50	10.00	12.70	24.71	55.51
M_r (emu/g)	0.00	0.00	0.00	0.05	0.02	0.78	0.29	2.20	10.00
H_c (Oe)	18.00	8.00	20.00	23.00	28.00	87.00	30.00	97.00	199

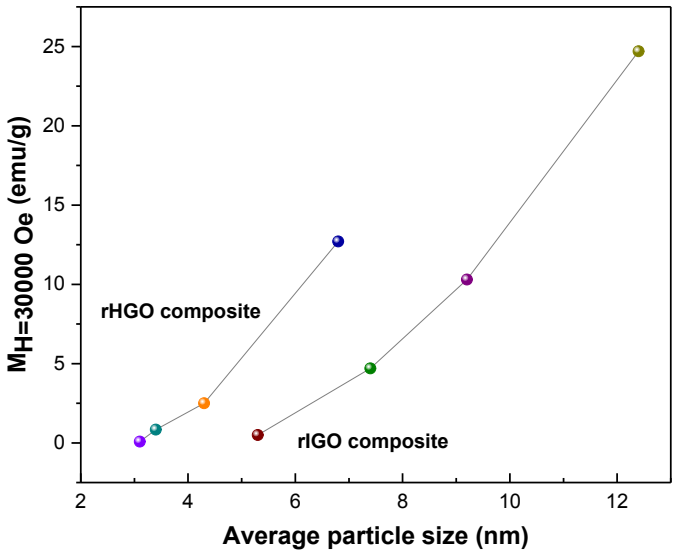

Figure SI5. Plot correlating the average particle size of $CoFe_2O_4$ with magnetization at 30,000 Oe for the two sets of rGO composites studied.

Table SI4. Particle size, with corresponding r_2 values, of two commercial contrast agents, two reported research samples and the present cobalt ferrite composites/nanoparticles.

Material	**Particle size (nm)**	**r_2 value ($mM^{-1}sec^{-1}$)**	**Reference**
Feridex (Ferumoxides)	120-180	98.3	Y.-X. J. Wang, Quantitative imaging in medicine and surgery, 2013, **3**, 1.
Resovist (Ferucarbotran)	45-60	151.0	Y.-X. J. Wang, Quantitative imaging in medicine and surgery, 2013, **3**, 1.
Au–Fe_3O_4 dumbbell	28	114.0	C. Xu, J. Xie, Angewandte Chemie International Edition, 2008, **47**, 173-176.
$CoFe_2O_4$-GO	5-13	72.7	G. Wang, Chemical Engineering Journal, 2016, **289**, 150-160.
$CoFe_2O_4$-rHGO 30%	7 (±2)	55.6	This work
$CoFe_2O_4$-rIGO 30%	12 (±2)	102.1	This work
$CoFe_2O_4$	12 (±3)	112.4	This work

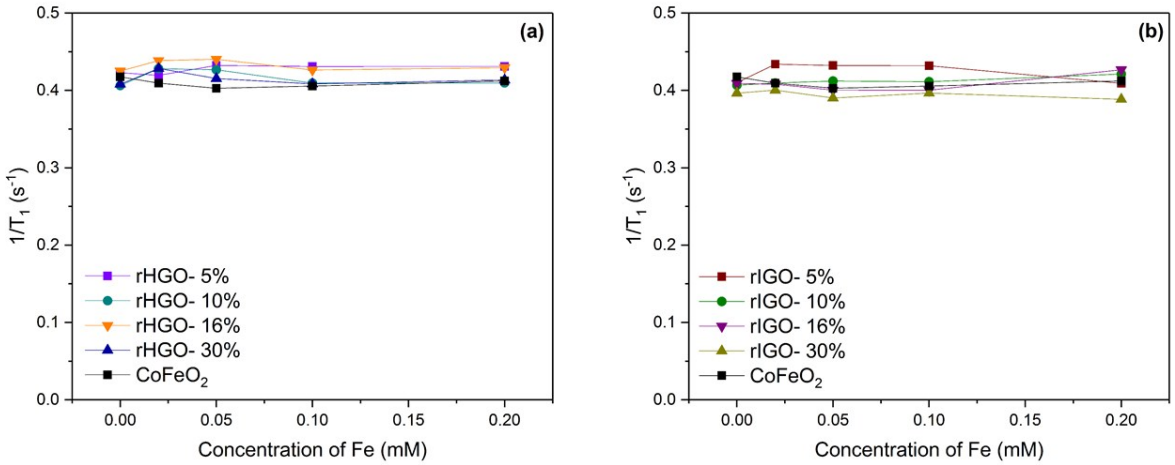

Figure SI6. Plot of the T_1 relaxation rate ($1/T_1$) for: (a) $CoFe_2O_4$-rHGO composites and pure $CoFe_2O_4$, (b) $CoFe_2O_4$-rIGO composites and pure $CoFe_2O_4$; all samples were suspended in aqueous solution at different Fe concentrations.

Table SI5. Chronological evaluation of the ζ-potentials for the 30 wt% $CoFe_2O_4$-rHGO and 30 wt% $CoFe_2O_4$-rIGO composites dispersed in aqueous PBS (samples measured at 70 μg mL^{-1}). Mean values and standard deviations obtained from triplicate measurements.

Samples	**ζ-potential in PBS (mV)**		
	0 time	**10 hours**	**24 hours**
30 wt% $CoFe_2O_4$-rHGO	-31.5 ±6.99	-31.8 ±5.38	-35.0 ±5.06
30 wt% $CoFe_2O_4$-rIGO	-36.4 ±6.09	-35.4 ±5.23	-39.2 ±5.60

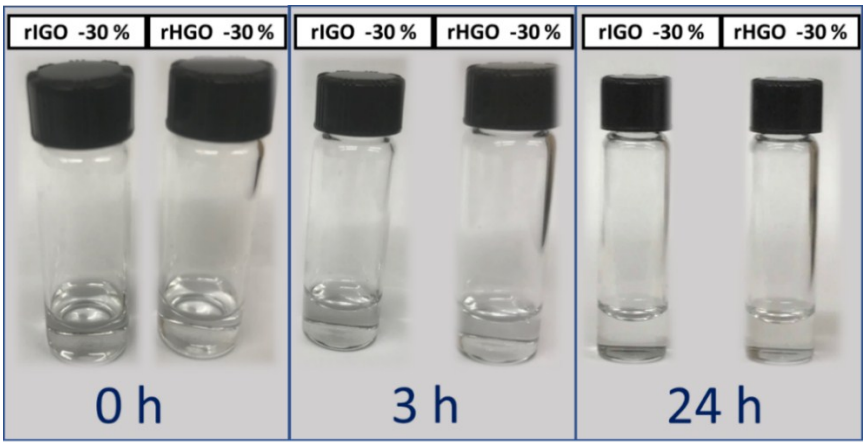

Figure SI7. Photographs of 30 wt% $CoFe_2O_4$-rHGO and 30 wt% $CoFe_2O_4$-rIGO composites, dispersed at 70 μg mL^{-1} in PBS, after various time points.

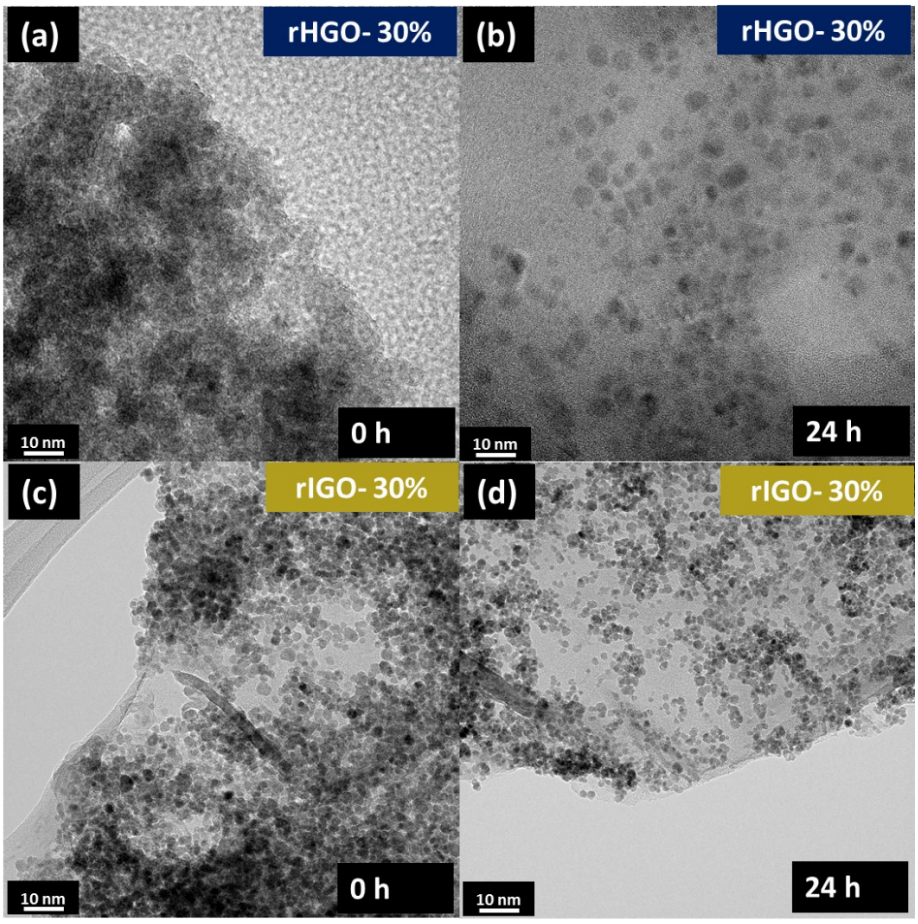

Figure SI8. (a-b) TEM images of the 30 wt% $CoFe_2O_4$-rHGO in PBS after 0 h and 24 h, respectively; (c-d) TEM images of the 30 wt% $CoFe_2O_4$-rIGO in PBS after 0 h and 24 h, respectively.