Fabrication of a Multifunctional Magnetic-Fluorescent Material for Medical Applications

Fernando Arteaga-Cardona,*^a J. Octavio Estévez ^b, Miguel A. Méndez-Rojas ^c, Silvia Hidalgo-Tobón^{d,e}, Pilar Dies-Suarez^e, N. Rutilo Silva-González^a, Justo Miguel Gracia y Jiménez^a, Gary N. Cherr^{f,g}, Ulises Salazar-Kuri^a

a. Instituto de Física, Benemérita Universidad Autónoma de Puebla, Apdo. Postal J-48, Puebla, Pue. 72570, México.

b. 2 Departamento de Materia Condensada/Instituto de Física UNAM, Circuito de la Investigación Científica Ciudad Universitaria, C.P. 04510, México.

c. Departamento de Ciencias Químico-Biológicas, Universidad de las Américas Puebla, ExHda. Sta. Catarina Mártir s/n, San Andrés Cholula, 72810 Puebla, México.

d. Departamento de Física, Universidad Autónoma Metropolitana, Avenida San Rafael Atlixco 186 Iztapalapa, México City, México.

e. Departamento de Imagenología, Hospital Infantil de México Federico Gómez, Dr. Márquez, Col. Doctores, México City, México

f. Bodega Marine Laboratory, University of California-Davis, Bodega Bay, California, USA.

g. Department of Environmental Toxicology and Nutrition, University of California-Davis, Davis, California, USA.

Simulation of X-ray diffraction patterns



Figure 1S. Simulation of pure non-interacting $CoFe_2O_4$ (red) and pure Gd_2O_3 (black) plotted in the same graph to show that the peaks of both crystalline structures overlap. The structures on the left are the ones used for the simulation.



Figure 2S. Simulation of a unit cell of $CoFe_2O_4$ (red) $-Gd_2O_3$ (black) with a unit cell relationship of 1:1 respectively. The structure used for the simulation of the diffraction patterns is on the left of the graph.



Figure 3S. Simulation of a unit cell of $CoFe_2O_4$ (red) $-Gd_2O_3$ (black) with a unit cell relationship of 1:2 respectively. The structure used for the simulation of the diffraction patterns is on the left of the graph.



Figure 4S. Simulation of a unit cell of $CoFe_2O_4$ (red) $-Gd_2O_3$ (black) with a unit cell relationship of 1:3 respectively. The structure used for the simulation of the diffraction patterns is on the left of the graph.



Figure 5S. Simulation of a unit cell of $CoFe_2O_4$ (red) $-Gd_2O_3$ (black) with a unit cell relationship of 1:4 respectively. The structure used for the simulation of the diffraction patterns is on the left of the graph.



Figure 5S. Simulation of a unit cell of $CoFe_2O_4$ (red) $-Gd_2O_3$ (black) with a unit cell relationship of 1:5 respectively. The structure used for the simulation of the diffraction patterns is on the left of the graph.

From figure 1S it can be seen that the peaks of the Gd_2O_3 overlaps with the peaks of $CoFe_2O_4$, this overlap makes extremely difficult to distinguish each peak of the diffraction pattern for the combined structures. When the relationship is 1:1 (Figure 2S) or 1:2 (Figure 3S) it may seem that there is not much change and the main peaks of the $CoFe_2O_4$ can be still distinguishable. Nevertheless, when the relationship increase of 1:3 and more, the main peaks of the $CoFe_2O_4$ cannot be distinguished from peaks of the Gd_2O_3 .

Fluorescence of Gd₂O₃:Eu

Peak	Position (nm)	Transition
1	426	${}^{5}D_{3} \rightarrow {}^{7}F_{2}$
2	434	${}^{5}D_{3} \rightarrow {}^{7}F_{3}$
3	450	${}^{5}D_{2} \rightarrow {}^{7}F_{0}$
4	466	${}^{5}D_{3} \rightarrow {}^{7}F_{4}$
5	471	${}^{5}D_{2} \rightarrow {}^{7}F_{1}$
6	488	${}^{5}D_{2} \rightarrow {}^{7}F_{2}$
7	498	${}^{5}D_{3} \rightarrow {}^{7}F_{5}$
8	512	${}^{5}D_{2} \rightarrow {}^{7}F_{3}$
9	535	${}^{5}D_{1} \rightarrow {}^{7}F_{1}$
10	551	${}^{5}D_{1} \rightarrow {}^{7}F_{2}$
11	565	${}^{5}D_{2} \rightarrow {}^{7}F_{5}$
12	586	${}^{5}D_{1} \rightarrow {}^{7}F_{3}$
13	600	${}^{5}D_{0} \rightarrow {}^{7}F_{1}$
14	611	${}^{5}D_{0} \rightarrow {}^{7}F_{2}$

Table 1S. Observed transitions in Figure 5.

15	630	${}^{5}D_{1} \rightarrow {}^{7}F_{4}$
16	650	${}^{5}D_{0} \rightarrow {}^{7}F_{3}$
17	660	${}^{5}D_{1} \rightarrow {}^{7}F_{5}$
18	709	${}^{5}D_{0} \rightarrow {}^{7}F_{4}$

Elemental analysis







Figure 7S. EDS mapping of the sample ML5 showing the corresponding SEM image and the distribution of elements Fe, Gd and O.

It is believed that the core-shell structure is formed by a $Gd_{2-x}O_3$: Eu_x matrix with the small $CoFe_2O_4$ inside the matrix. To probe it, EDS distribution line scan (Figure 6S) was performed to study the composition of the ML15 composites. Also on Figure 7S it is showed the EDS mapping of the same sample. Both figures measured using 25 kV. In Figure 6S it is observed that in the studied line we have the presence of the elements Gd, Eu, Fe and O. Gadolinium and oxygen with strong intensities due to the matrix $Gd_{1.95}O_3$ and europium with a weak signal due to the 5 % of nominal substitution in the gadolinium locations. On the other hand Fe has also a weak intensity and Co was not detected.



Figure 8S. Z-Contrast images of ML15 showing homogeneous brightness.

We tried to identify core-shell structures by Z-contrast, however, those couldn't be identified probably because two main factors. The first one is the enormous difference between the atomic number of Gd (64) and Co (27), and the second reason is because of the difference in mass, as $CoFe_2O_4$ are nanoparticles of sizes below 10 nm, while Gd_2O_3 are submicrometer particles, with sizes over 100 nm.