Supplementary Information

Topochemical reduction of FeCo-oxide to FeCo-alloy nanosystems into a SiO_2 matrix

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1. Magnetization curves of FeCo-oxides and alloys at room temperature

The magnetization *M* of FeCo-oxides samples (Figure S1, Table S1) and FeCo-alloys (Figure S2, Table S1) was estimated using the law of approaching to saturation reported in equation below[1].

$$M = M_s\left(1 - \frac{A}{H} - \frac{C}{H^2}\right)$$

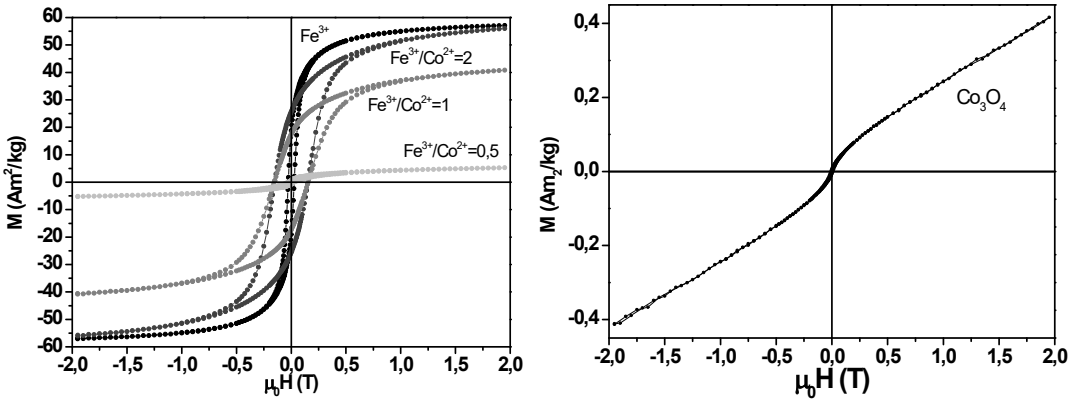

Figure S1 *Magnetization measured at 300K of the FeCo-oxides.*

Table S1 *Saturation magnetization and coercivity size of the oxide and alloy samples.*

Fe^{3+} at.%	***Oxides***		***Alloys***	
	M_s (Am^2kg^{-1})	$\mu_0 H_c$ (mT)	M_s (Am^2kg^{-1})	$\mu_0 H_c$ (mT)
100	59(1)	27(1)	219(4)	6(1)
67	67(1)	151(2)	242(6)	10(1)
50	46(2)	156(2)	242(5)	14(1)
33	6(1)	24(1)	222(1)	9(1)
0	-	8(1)	166(4)	10(1)

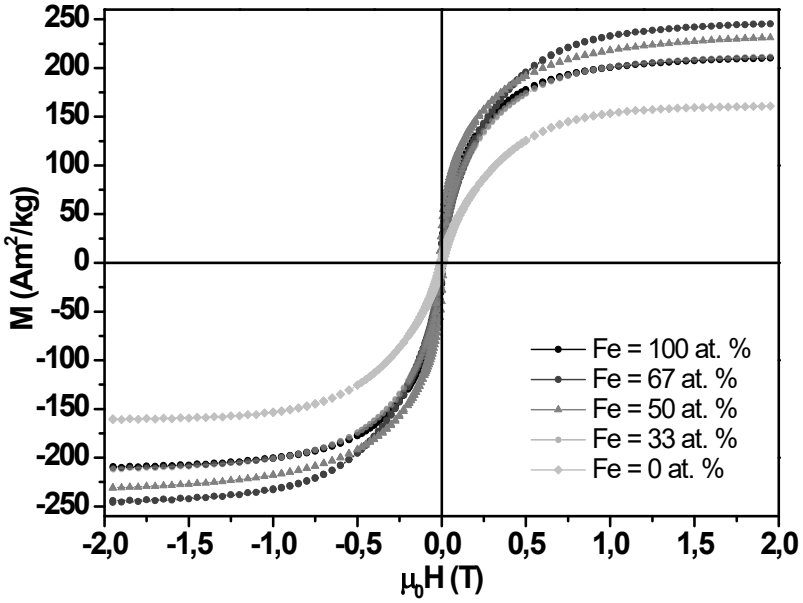

Figure S2 *Magnetization measured at 300K of the corresponding FeCo-alloys.*

2. Density functional theory (DFT) calculations $Fe_{1-x}Co_x$

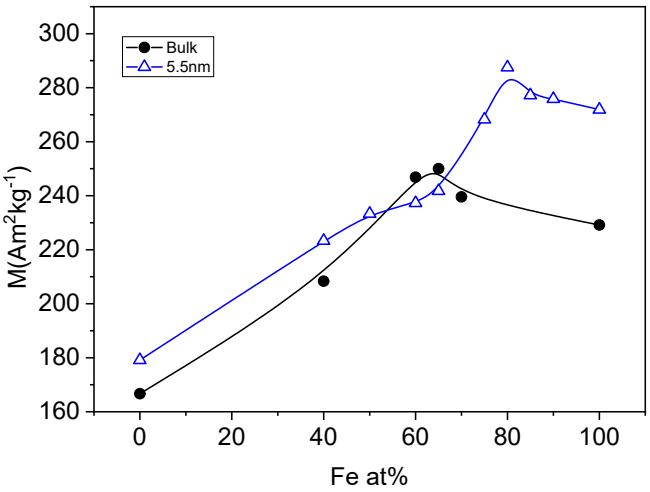

Figure S3 *Magnetization predicted by DFT calculations for bulk and NP systems.*

We have developed an advanced two stage process in order to perform Density Functional Theory (DFT) calculations on $Fe_{1-x}Co_x$ alloys that enables modelling of magnetic nanoparticles up to ~10 nm diameter. In the first stage collinear spin polarized Tigh Binding Density Functional (TBDFT) [2]. In the second stage spin-polarized density functional theory implemented by the Vienna Ab Initio Simulation Package (VASP)code were performed [3]. The maximum position changes almost with Fe concentration (see figure above). If we extrapolate the calculated data, for which a more in-depth study is being under submission [4], for a nanoparticle of 20nm in size we would expect the maximum magnetisation to be close to 41 Co at%.

3. Thermal gravimetry and differential thermal analysis of the sample of alloys

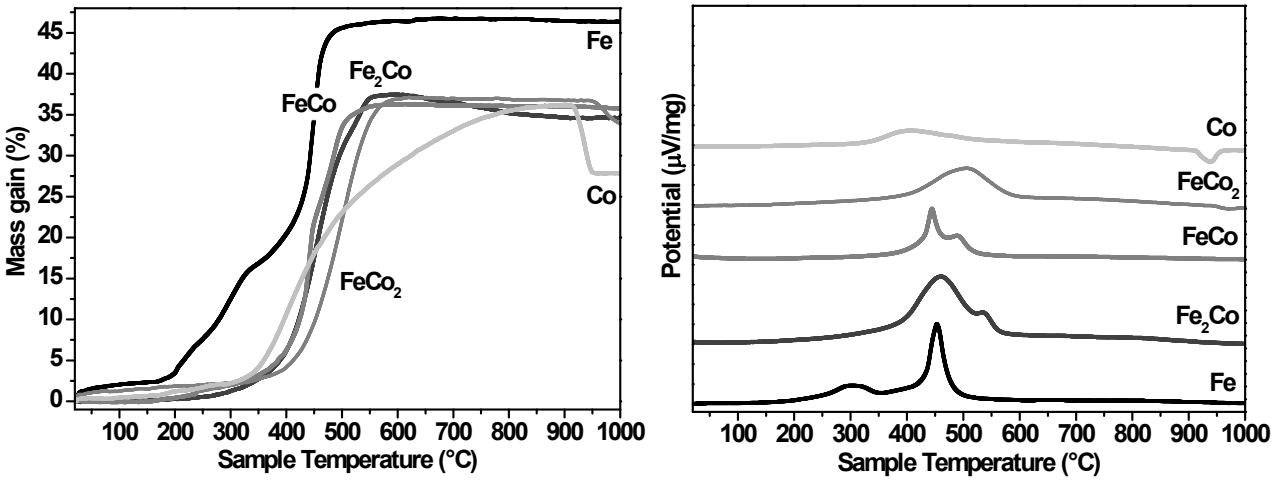

Figure S4 *Thermal behavior of the samples of alloy in the range 25°C – 1000°C with a heating rate of 10°C/min (left) and the registered heat flow during the heating ramp (right).*

Table S2 *Mass gain and onset temperature for each peak. The onset temperatures were measured at the center of the peak.*

Fe^{3+}/Co^{2+}	**Total mass gain [%]**	**First peak [°C]**	**Second peak [°C]**
Only Fe^{3+}	46.7	304	454
2	37.4	461	534
1	36.3	444	488
0.5	37.1	506	974
Only Co^{2+}	36.1	408	938

REFERENCES

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2. Hourahine, B., Aradi, B., Blum, V., Bonafé, F., Buccheri, A., Camacho, C., Cevallos, C., Deshaye, M.Y., Dumitrică, T., Dominguez, A., Ehlert, S., Elstner, M., van der Heide, T., Hermann, J., Irle, S., Kranz, J.J., Köhler, C., Kowalczyk, T., Kubař, T., Lee, I.S., Lutsker, V., Maurer, R.J., Min, S.K., Mitchell, I., Negre, C., Niehaus, T.A., Niklasson, A.M.N., Page, A.J., Pecchia, A., Penazzi, G., Persson, M.P., Řezáč, J., Sánchez, C.G., Sternberg, M., Stöhr, M., Stuckenberg, F., Tkatchenko, A., Yu, V.W. -z., and Frauenheim, T. (2020) DFTB+, a software package for efficient approximate density functional theory based atomistic simulations. *J Chem Phys*, **152** (12).

3. Kresse, G., and Furthmüller, J. (1996) Efficient iterative schemes for *ab initio* total-energy calculations using a plane-wave basis set. *Phys Rev B*, **54** (16), 11169–11186.

4. Ntallis, N., Peddis, D., and Trohidou, K. (2025) DFT calculations of high moment $Fe_{1-x}Co_x$ alloy and Fe/Co, Co/Fe core shell magnetic nanoparticles. **In submission**.