Magnetic Nanosheets: from iron oxide nanocubes to polydopamine embedded 2D clusters and their multi-purpose properties

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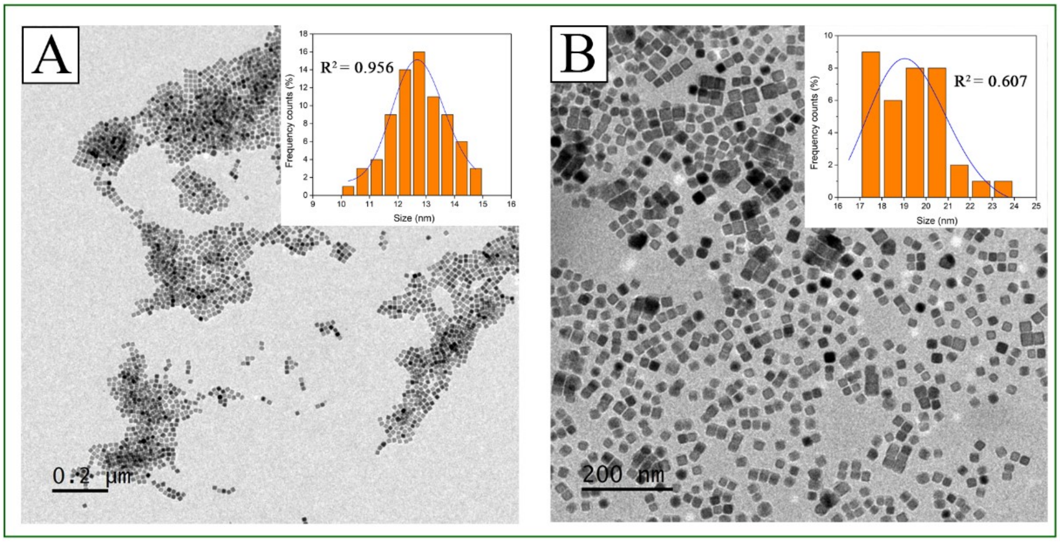
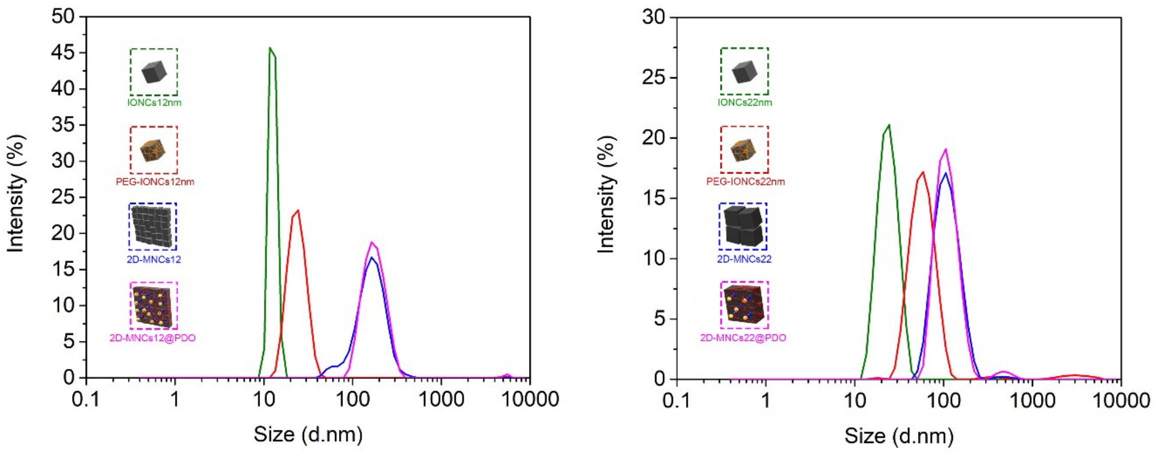


Figure S1. TEM images of IONCs with a corresponding diameter of A) PEG-IONCs 12nm and B) PEG-IONCs22nm. In inset are reported the particle size distribution histograms observed in TEM images.

A)



B)

Samples	Intensity (d.nm)	Z-Potential (mV)
IONCs-12nm	13 ± 1	11 ± 10
PEG-IONCs 12nm	23 ± 6	-34 ± 8
2D-MNCs12	146 ± 62	-41 ± 8
2DMNCs12@PDO	175 ± 51	-21 ± 8
IONCs-22nm	23 ± 6	32 ± 13
PEG-IONCs 22nm	58 ± 18	-39 ± 8
2D-MNCs22	106 ± 36	-54 ± 10
2DMNCs22@PDO	109 ± 62	-22 ± 9

Figure S2. A) Comparison of size distribution by DLS Intensity of: IONCs12nm dispersed in $CHCl_3$, PEG-IONCs12nm, 2D-MNCs12nm and 2D-MNCs12@PDO (left) and IONCs22nm dispersed in $CHCl_3$, PEG-IONCs22nm, 2D-MNCs22nm and 2D-MNCs22@PDO (right). Green line for IONCs dispersed in $CHCl_3$, red line for PEG-IONCs, blue line for 2DMNCs and purple line for 2DMNCs@PDO. B) Summary of average hydrodynamic sizes by intensity and ζ-Potential of all structures as measured by DLS

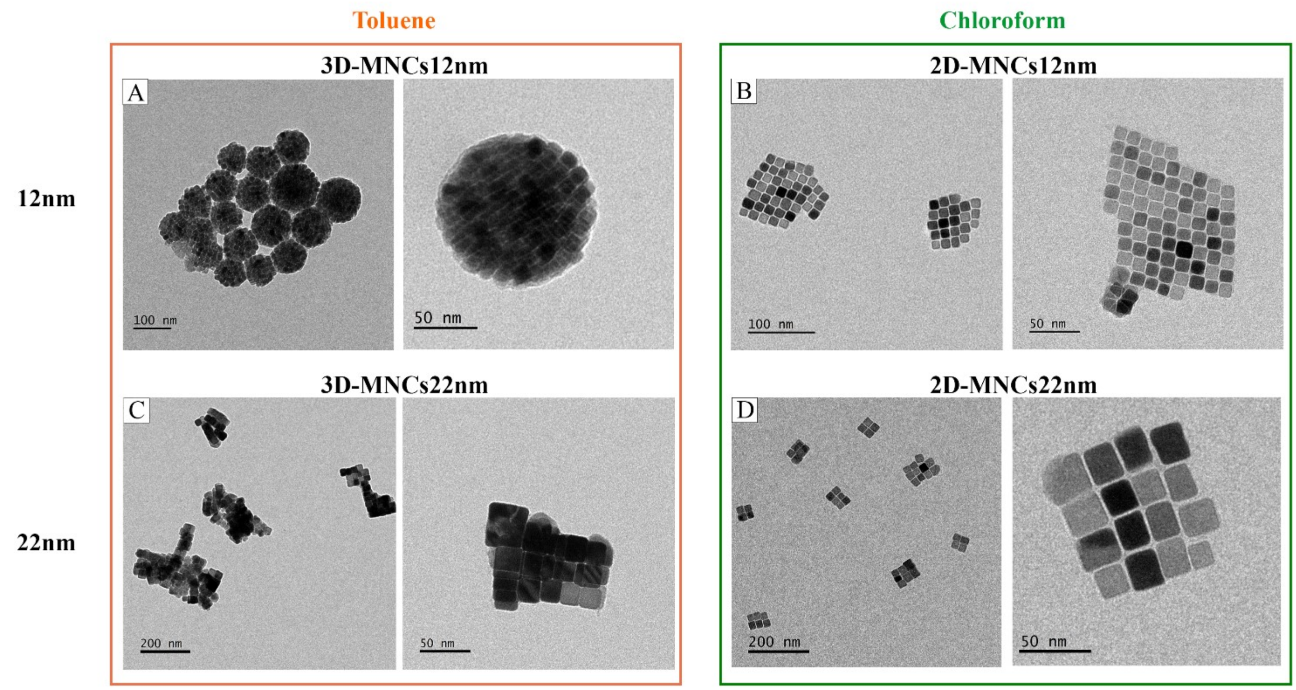


Figure S3. TEM images of: MNCs obtained by starting from IONCs12nm dispersed in A) toluene (3D-MNCs12nm) or in B) chloroform (2D-MNCs12nm), and corresponding clusters obtained by starting from IONCs22nm dispersed in C) toluene (3D-MNCs12nm) and D) chloroform (2D-MNCs12nm).

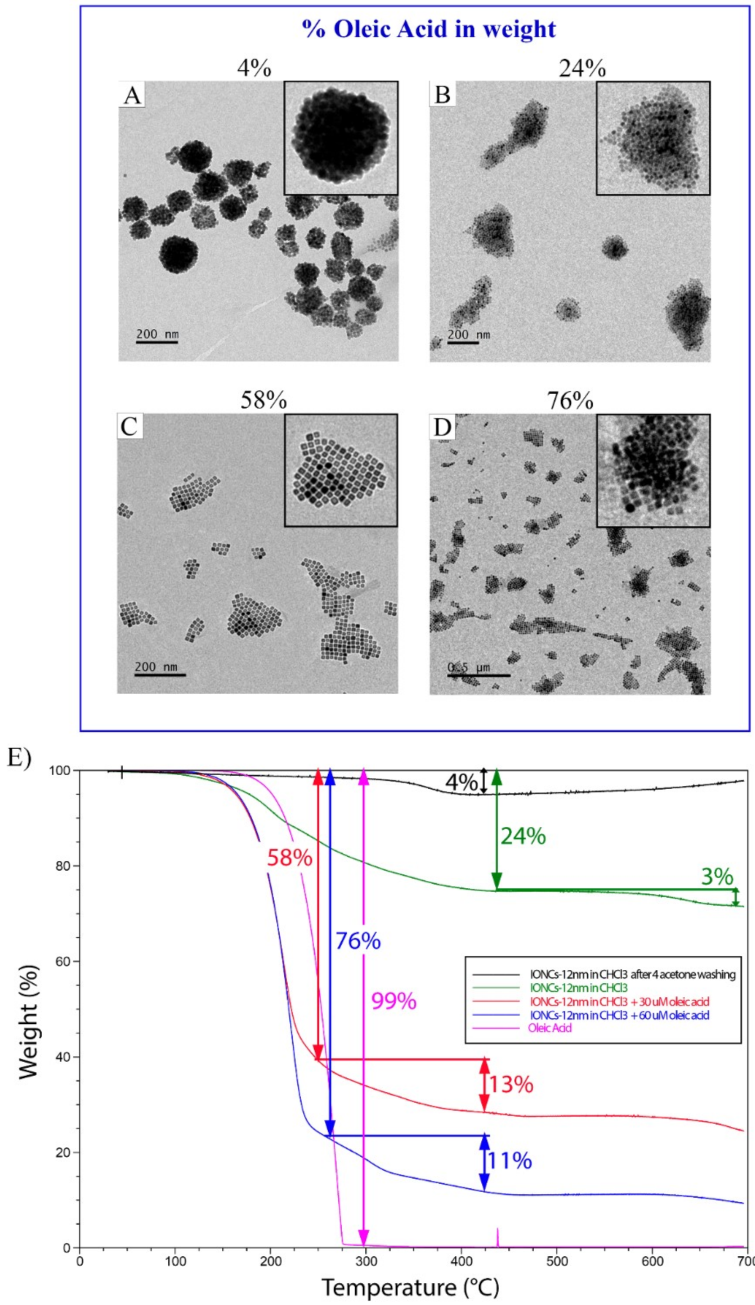


Figure S4. TEM images of MNCs12nm obtained using IONCs12nm: A) after four acetone washing to remove the oleic acid (4% in weight) and after the addition of oleic acid in the solution reaching three different percentage in weight, B) 24% C) 58% and D) 76%. E) TGA analysis of IONCs-12nm (green spectra) obtained after by adding different oleic acid concentration: 30 μM (red spectra) and 60 μM (blue spectra). In black spectra is shown IONCs-12nm after 4 acetone washing (black). Finally, TGA of free oleic acid is reported (purple).

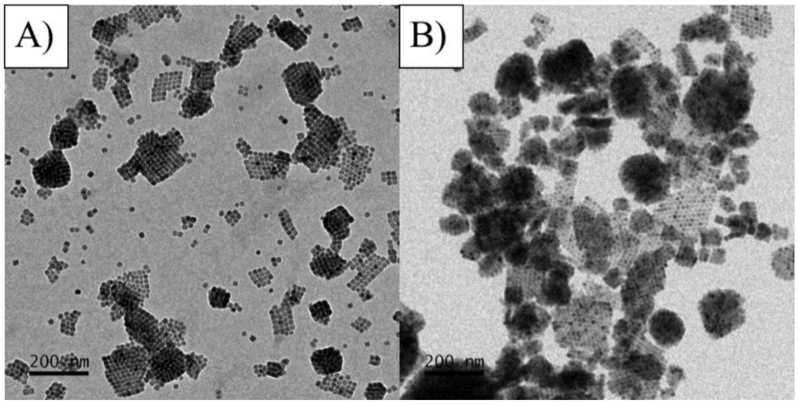


Figure S5. TEM images of: a) MNCs obtained by using SDS of 1.1mg/mL and b) MNCs by using SDS of 2.6 mg/mL. 2D and 3D clusters arrangements are both evident in both samples.

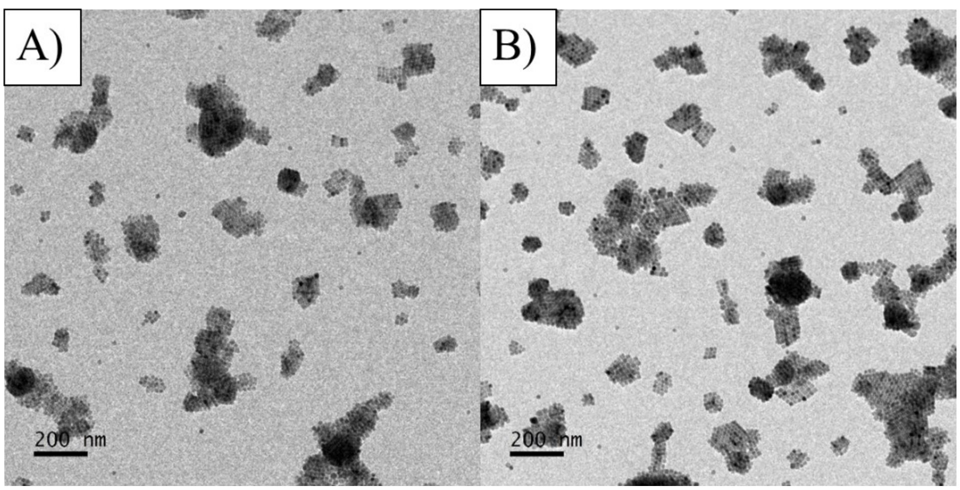


Figure S6. TEM images of MNCs obtained by setting the temperature of the water-bath sonicator at room temperature. Under these conditions 2D and 3D clusters are seen.

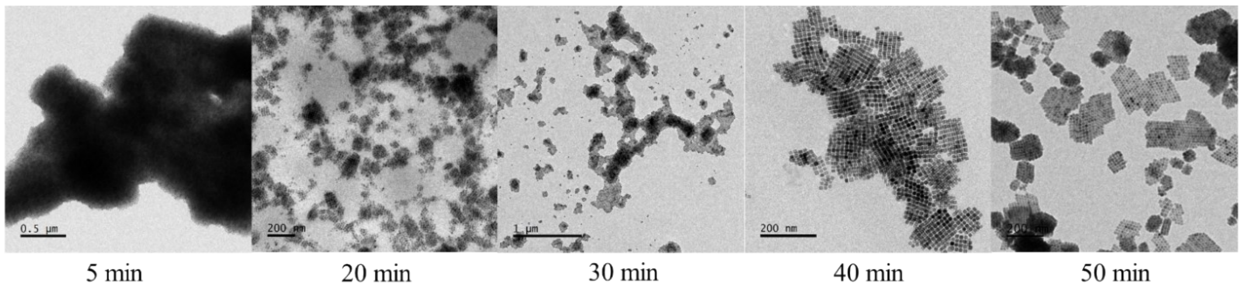


Figure S7. TEM images of aliquots collected during the formation of 2D-MNCs with 12nm iron oxide nanocubes at different time points, after: a) 5 min, b) 20 min, c) 30 min, d) 40 min and e) 50 minutes. The choice of 40 minutes was the selected one for better 2D MNC ordering.

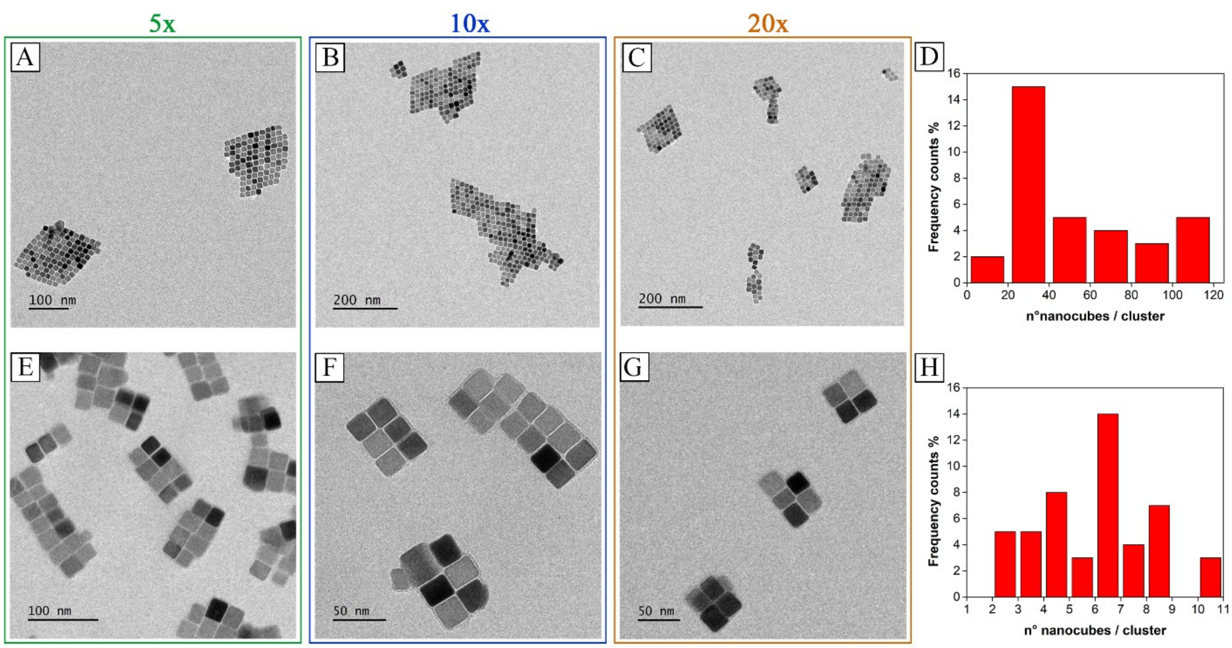


Figure S8. TEM images of 2D-MNCs12nm and 2D-MNCs22nm scaled up A and E) 5 fold; B and F) 10 fold , C) and G) 20 fold, respectively. Number of IONCs per clusters histograms of D) 2D-MNCs12@PDO and H) 2D-MNCs22@PDO observed in TEM images scaled up 20 fold.

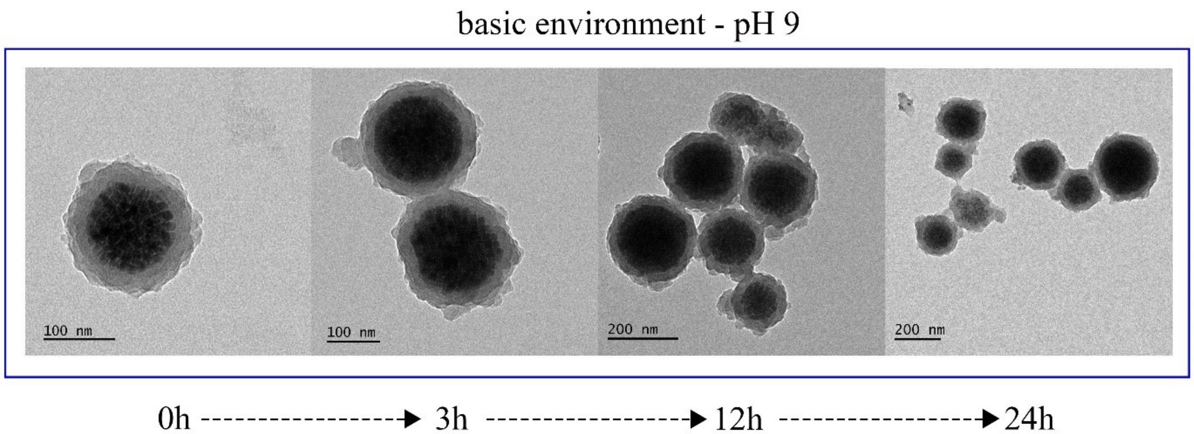


Figure S9. TEM images of polydopamine functionalized magnetic nanoclusters after incubation in saline PBS buffer, at pH = 9 at four specific time points: 0, 3, 12 and at 37°C for 24 h . No structural changes are observed overtime.

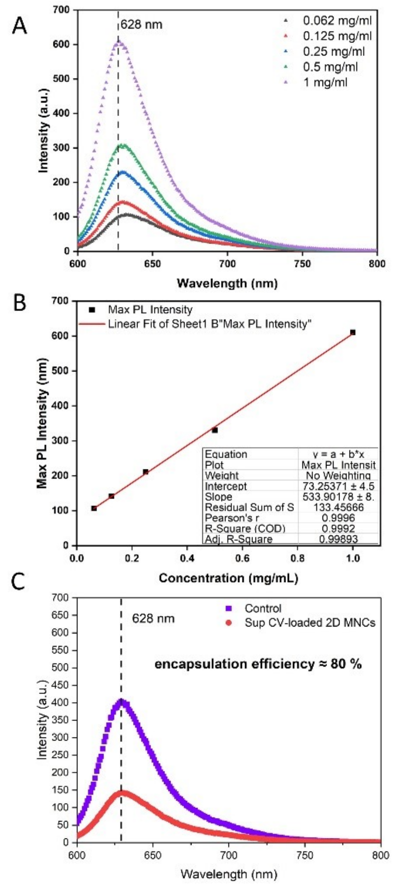


Figure S10. A) PL spectra of CV stock solutions at 1, 0.5, 0.25, 0.125, and 0.062 mg/mL used for recording the PL intensity of CV at 628 nm for the calibration curve. B) Linear fitting of the PL intensity at 628 nm (excitation 590 nm) versus CV dye concentration (R^2=0.9992). C) PL signals of the initial CV stock solution (DOPA + CV + water) and CV left in the supernatant after magnetic separation of the CV-loaded 2D clusters. The encapsulation efficiency of the CV dye in the 2D MNCs corresponded to about 80 %.

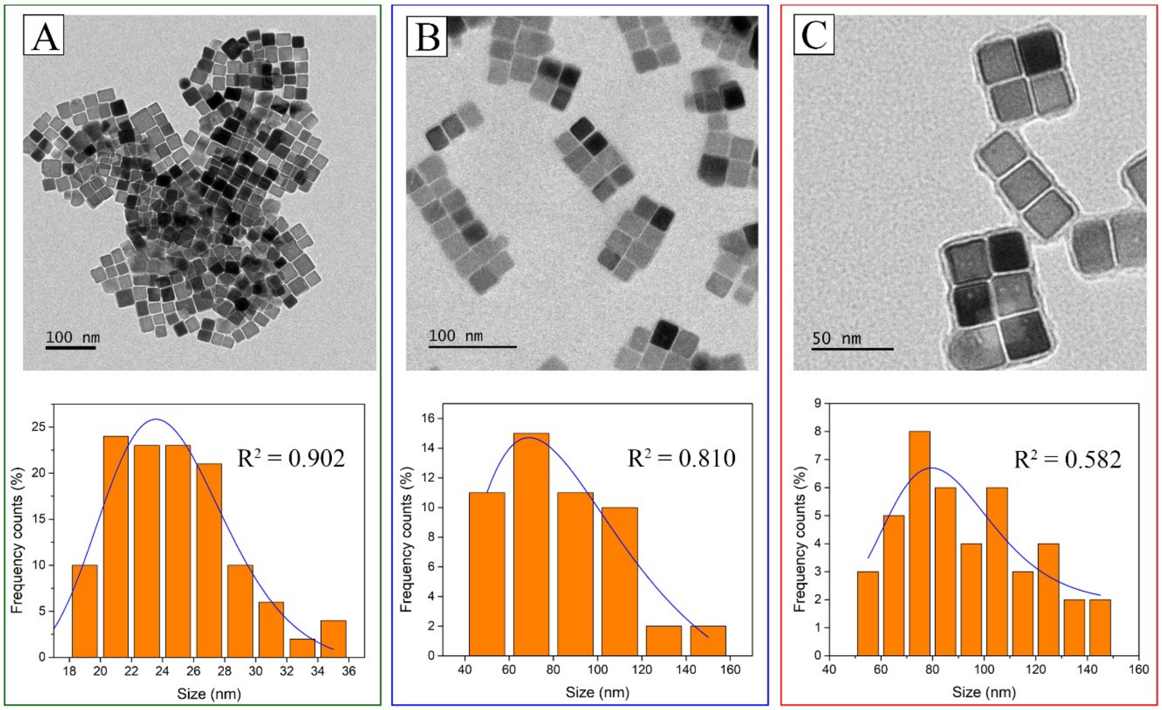


Figure S11. TEM images of A) IONCs24nm, B) 2D-MNCs24nm and C) 2D-MNCs24@PDO and their respective particle size distribution histograms as measured by statistics on TEM images.

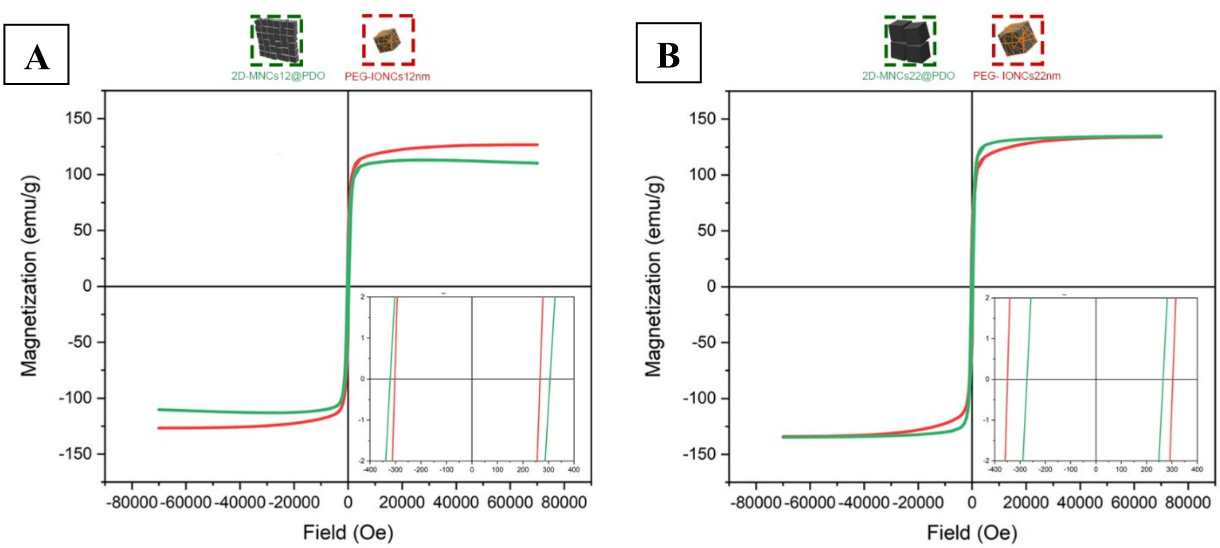


Figure S12. Normalized magnetization curves measured at 5K for A) PEG-IONCs12nm and 2D-MNCs12@PDO and B) PEG-IONCs22nm and 2D-MNCs22@PDO. The inset represents a detail of the low field region for each of the curves.

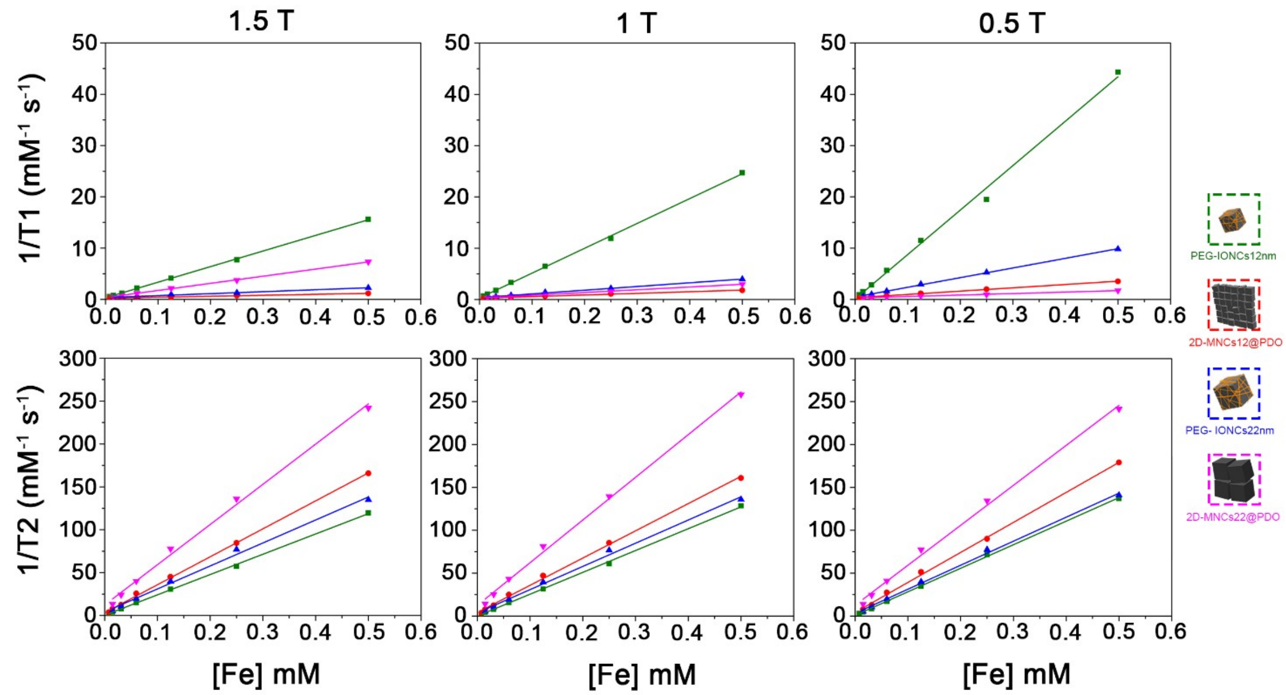
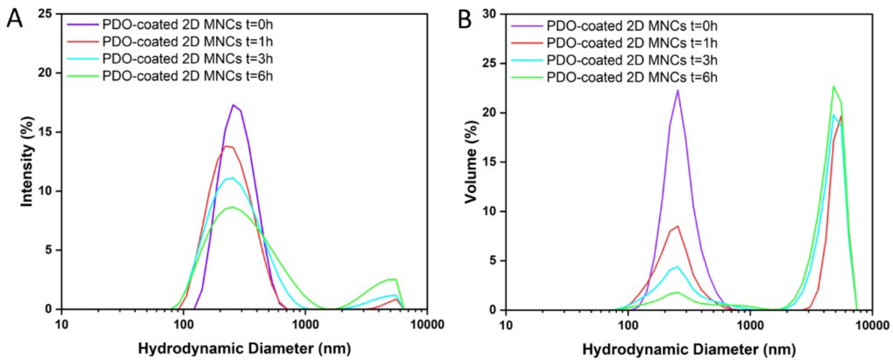
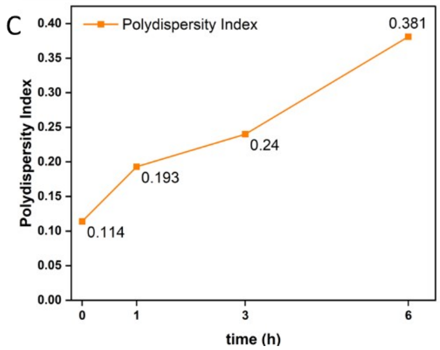


Figure S13. r1, r2 as a function of the applied magnetic field for PEG-IONCs12nm (green line), 2D-MNCs12@PDO (red line), PEG-IONCs22nm (blue line) and 2D-MNCs22@PDO (purple line). Data were fitted with linear fit and plotted as continuous lines.





D

Sample	Intensity (%) ± FWHM	Volume (%) ± FWHM	PDI
violet	288 ± 91	273 ± 84	0.114
red	259 ± 99	250 ± 91	0.193
blue	290 ± 141	278 ± 143	0.240
green	340 ± 203	359 ± 255	0.381

Figure S14. Hydrodynamic size evolution measured by dynamic light scattering (DLS) of PDO-coated 2D MNCs before acidic treatment (purple line) and after 1h (red line), 3 (light blue line), and 6h (green line) at pH=5 weighted by A) intensity percentage and B) volume percentage. C) Corresponding polydispersity index (PDI) evolution at 0,1,3 and 6 h. The progressive DLS peak broadening and the concomitant increase in the PDI values are suggesting gradual PDO polymer degradation and change in configuration of the PDO-coated 2D MNCs at pH 5 supporting TEM structural data (Figure 3) and the change in MPI and SAR signals at acidic environment (Figure 9). D) Table summary of the DLS data.