Electronic Supplementary Material (ESI) for RSC Advances.

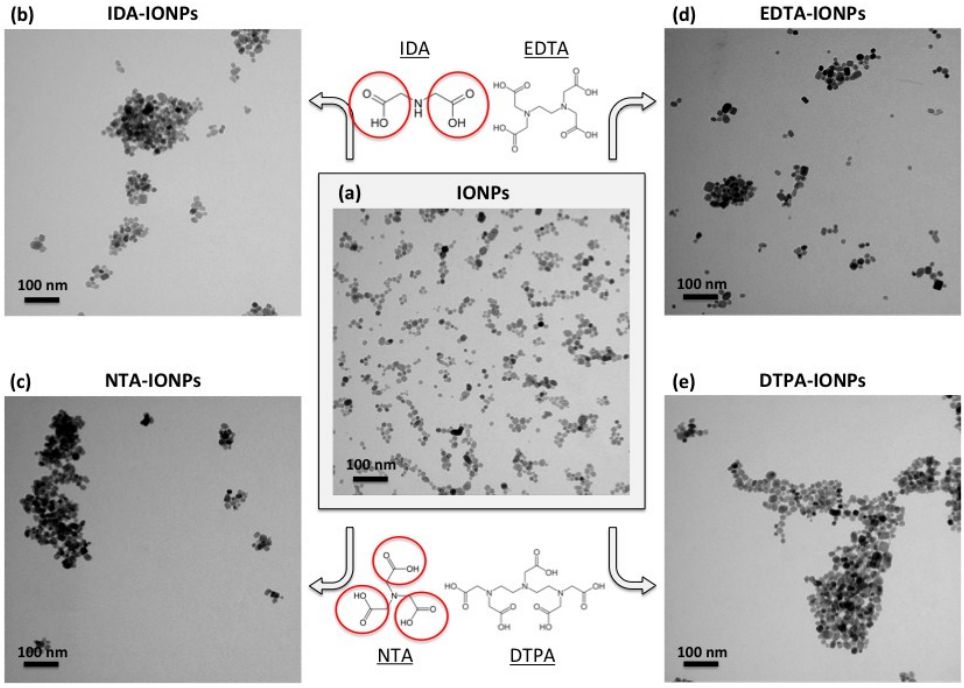

Fig. 1: Representative TEM micrographs of uncoated IONPs (a), IDA-IONPs (b), NTA-IONPs (c), EDTA-IONPs (d) and DTPA-IONPs (e), all kept in water. The insert in (a) shows the distribution of the *TEM diameter* of uncoated IONPs. The chemical structure of the four CAs, which were used in this study, is given in the middle part above and below panel (a), and as an example dents containing carboxylic groups are marked by red circles in IDA and NTA molecules.

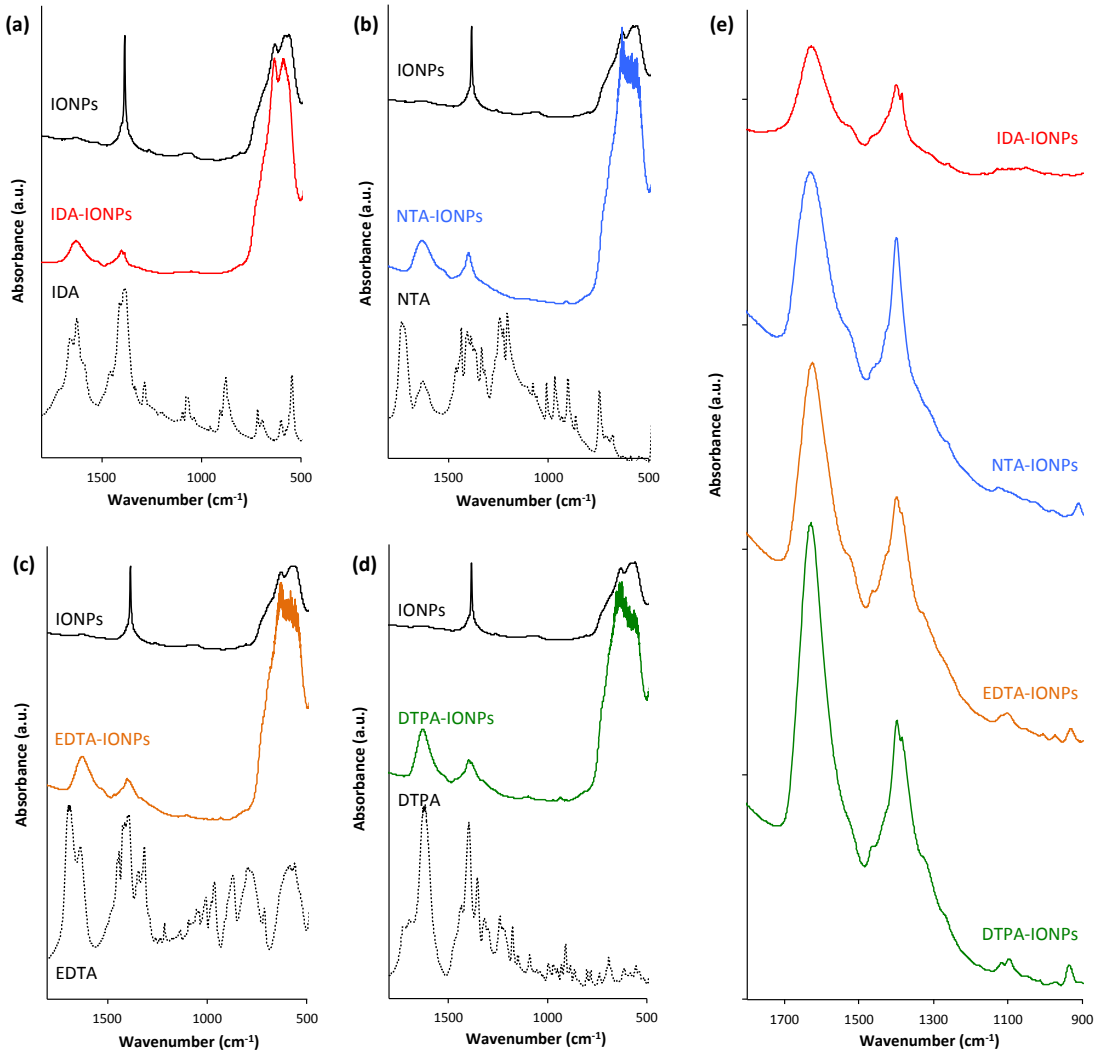

Fig. 2: Absorbance of IDA-IONPs (a), NTA-IONPs (b), EDTA-IONPs (c) and DTPA-IONPs (d) as compared to uncoated IONPs (highlighted with black lines) and to each of the coating molecules (IDA, NTA, EDTA and DTPA, respectively; highlighted with black dashed lines) measured by FTIR. (e) Absorbance of CA-coated IONPs measured by FTIR for a narrow wavenumber range (900 – 1800 cm^{-1}).

Table 1: Number-weighted distribution of hydrodynamic diameter (d_h), polydispersity index (PDI) and the zeta potential (ξ) measured by DLS of uncoated IONPs, IDA-IONPs, NTA-IONPs, EDTA-IONPs and DTPA-IONPs measured in water (pH ~6.1) or in RPMI medium (pH ~7.6). Their agglomerate density (ρ_{agglo}) measured in water are also given.

	d_h in H_2O (nm)	PDI in H_2O (-)	ρ_{agglo} in H_2O (g cm^{-3})	ξ in H_2O (mV)	d_h in RPMI (nm)	PDI in RPMI (-)	ξ in RPMI (mV)
IONPs	25 ± 2	0.13	1.62	6.3 ± 0.8	115 ± 1	0.47	-7.6 ± 0.6
IDA-IONPs	74 ± 27	0.31	1.41	26.2 ± 0.6	173 ± 180	0.25	-7.8 ± 0.3
NTA-IONPs	67 ± 27	0.46	2.24	-17.6 ± 4.8	364 ± 448	0.42	-7.5 ± 0.7
EDTA-IONPs	35 ± 11	0.77	1.49	-10.9 ± 1.0	429 ± 199	0.27	-7.9 ± 0.1
DTPA-IONPs	132 ± 89	0.34	3.47	-6.0 ± 4.3	1064 ± 683	0.41	-7.5 ± 0.2

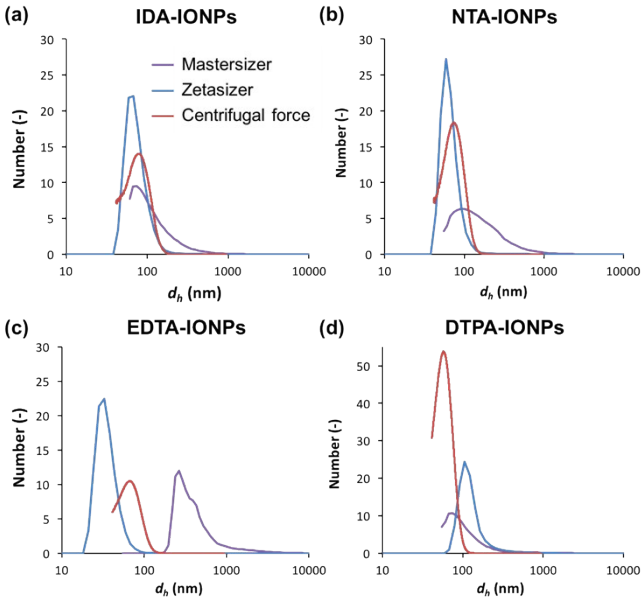

Fig. 3: Number weighted distribution of the hydrodynamic diameter, d_h, of IDA-IONPs (a), NTA-IONPs (b), EDTA-IONPs (c) and DTPA-IONPs (d) measured by dynamic light scattering (DLS) with two instruments (Mastersizer or Zetasizer), or by centrifugal force.

Table 2: MRI longitudinal relaxivity r_1, transverse relaxivity r_2 and the relaxivity ratio (r_2/r_1) measured at 3 T for uncoated IONPs, IDA-IONPs, NTA-IONPs, EDTA-IONPs and DTPA-IONPs.

	r_1 (mM^{-1} s^{-1})	r_2 (mM^{-1} s^{-1})	r_2/r_1 (mM^{-1} s^{-1})
IONPs	10.6 ± 0.2	1330.2 ± 33.5	125.4
IDA-IONPs	3.8 ± 0.1	1335.8 ± 64.0	353.9
NTA-IONPs	3.5 ± 0.1	1040.8 ± 27.3	293.2
EDTA-IONPs	5.5 ± 0.3	1543.6 ± 62.3	282.6
DTPA-IONPs	4.0 ± 0.1	1008.9 ± 24.4	249.7

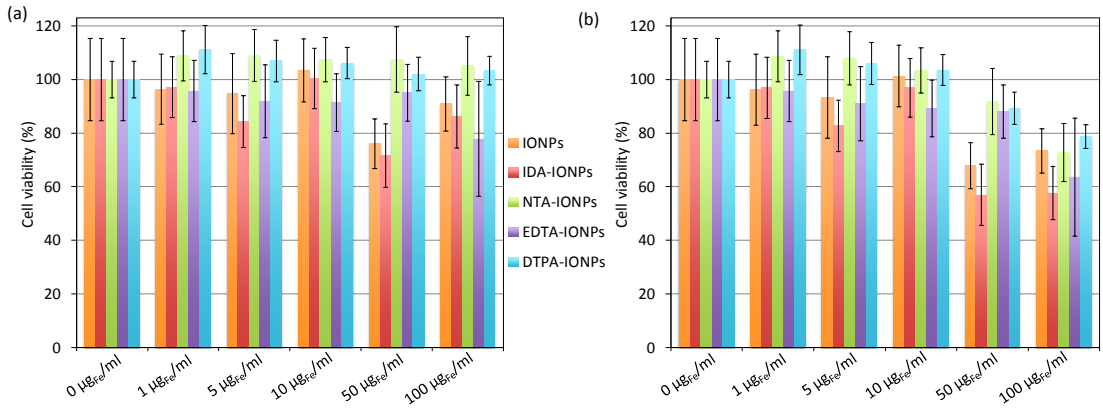

Fig. 4: Viability of LnCaP cells incubated for 24 h with different concentrations (0, 1, 5, 10, 50 and 100 μg_{Fe} ml^{-1}) of uncoated IONPs, IDA-IONPs, NTA-IONPs, EDTA-IONPs and DTPA-IONPs measured with the MTS test. The cell viabilities are the percentages obtained from the absorbance of cells treated with IONPs non-corrected (a) or corrected (b) for the absorbance of the administrated dose of the corresponding coated or non-coated IONPs and normalized with the absorbance of cells without IONPs (0 μg_{Fe} ml^{-1}). All values are given as average ± standard deviation.

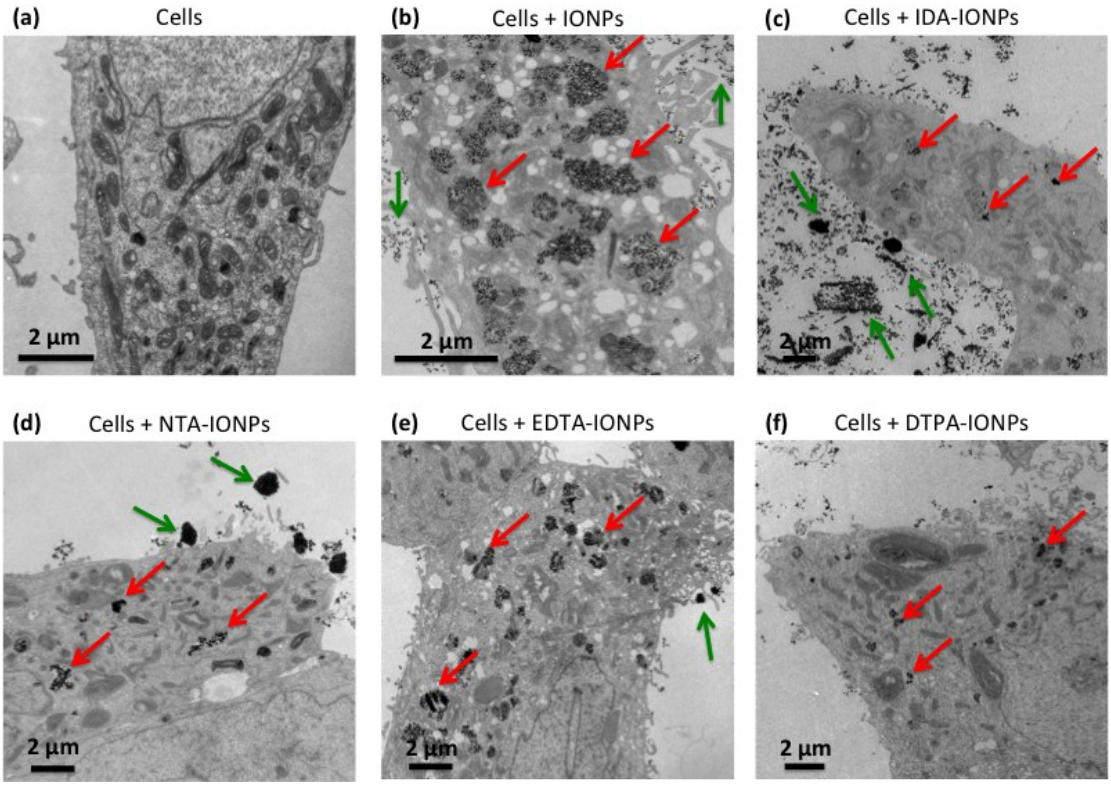

Fig. 5: Representative TEM micrographs of 50 nm-thick sections of LnCaP cells without IONPs (a), and LnCaP cells incubated for 24 h with uncoated IONPs (b), IDA-IONPs (c), NTA-IONPs (d), EDTA-IONPs (e) and DTPA-IONPs (f). Green and red arrows show IONPs outside and inside cells, respectively.