

Supplementary Information:

Determination of r_1 and r_2 relaxivities with 3 T MRI scanner:

To determine the relaxivities of the USPIOs using a 3T MRI scanner (TrioTim, Siemens, Germany), the particles were dispersed in deionized water in plastic vials. To avoid susceptibility artefacts from the surrounding air in the scans, all the samples were placed in a water-containing plastic container at room temperature. For the determination of the T_2 relaxation times, a clinical head coil and CPMG sequence were used with the following parameters: TR = 2000 ms; TE = 30-960 ms, 32 echoes; FOV = 100 mm; Matrix = 192×192 ; Slice thickness = 2 mm; Average 3. The relaxation times were calculated by a linear fit of the logarithmic region-of-interest signal amplitudes versus TE. T_1 relaxation times were obtained using an inverse saturation recovery sequence with different inversion times: TR = 500 ms; TE = 15 ms; Average 3; FOV = 100 mm; Matrix = 192×192 ; Slice thickness = 2 mm; Inversion time = 10 – 1000 ms.

Table S1 Relaxivities of USPIOs at different steps of probe preparation

Magnetic field (T)	Relaxivity ($\text{mM}^{-1}\text{s}^{-1}$)	PAA@USPIOs	After PEG modification	After RGD conjugation	RGD-Tc-PAA@USPIOs
1.41	r_1	8.67	9.12	8.92	9.34
	r_2	25.36	24.87	25.11	24.64
3	r_1	7.55	7.46	7.44	7.42
	r_2	32.81	33.52	32.90	32.14