Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.


**Supporting Information**

# Controllable Synthesis of Exceptionally Small-sized Superparamagnetic Magnetite Nanoparticles for Ultrasensitive MR Imaging and Angiography

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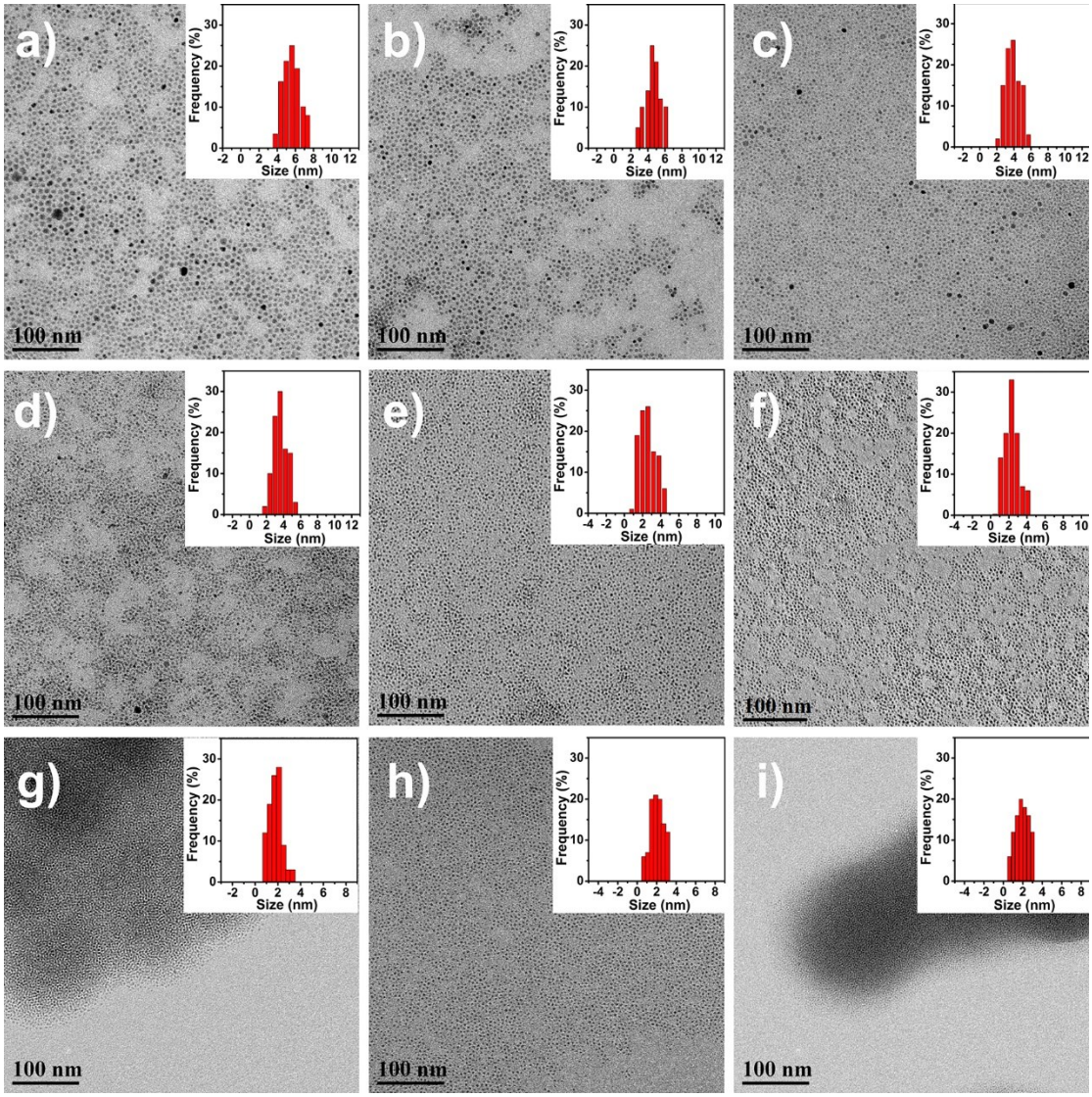


**Figure S1.** TEM images of a) 5.6 nm, b) 4.5 nm, c) 3.9 nm, d) 3.6 nm, e) 2.6nm, f) 2.3 nm, g) 2.1 nm, h) 1.9 nm, and i) 1.8 nm ESM NPs. The scale bar is 100 nm. Insets are the size distribution diagrams.

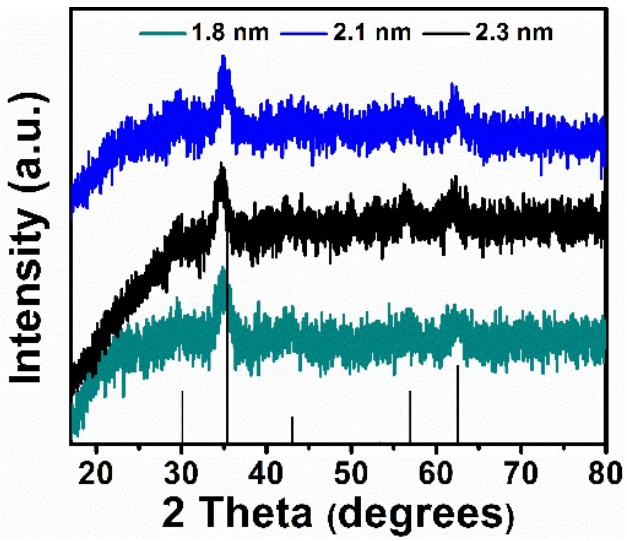


**Figure S2.** XRD patterns of 1.8 nm, 2.1 nm, and 2.3 nm ESM NPs.

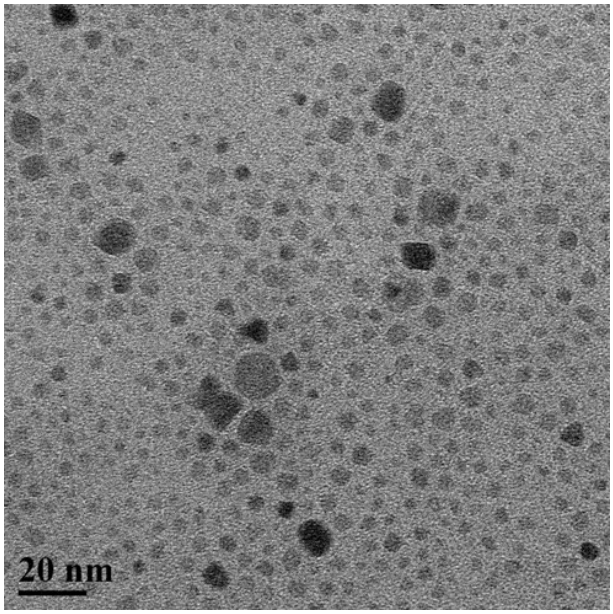


**Figure S3.** TEM images of ESM NPs obtained by reducing stabilizer and reducing agent in reaction solution by half.

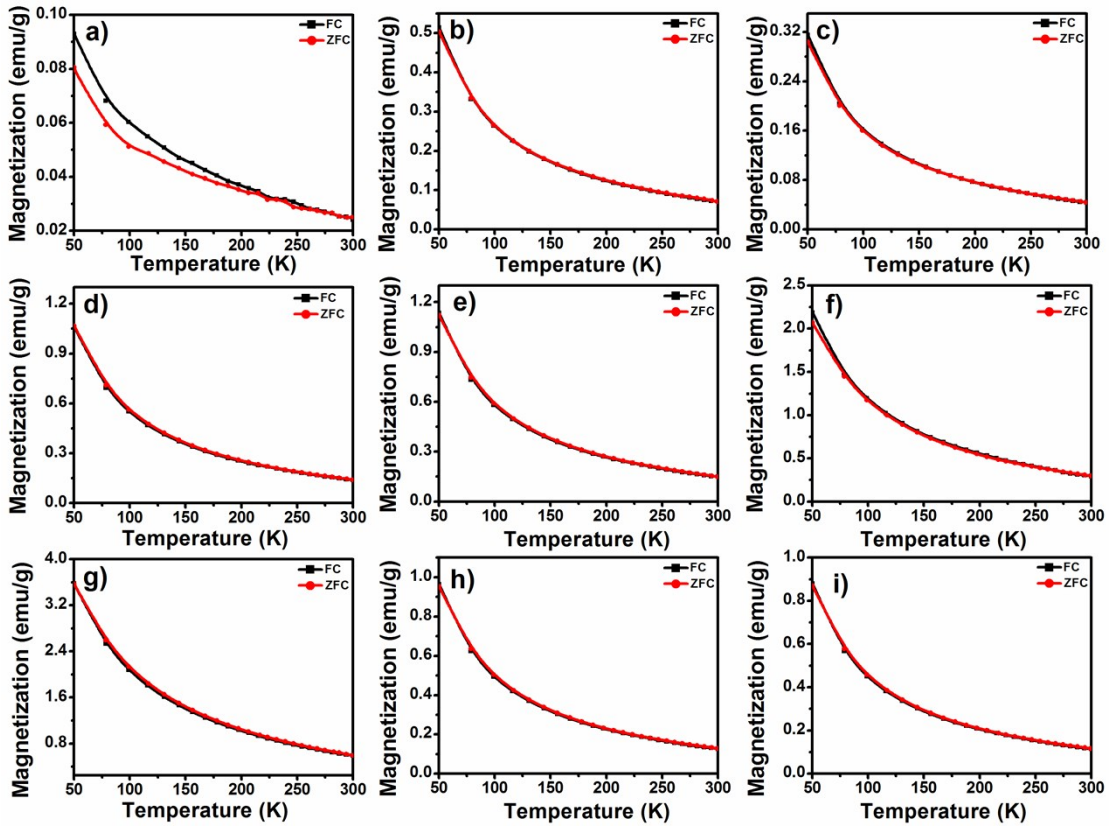


**Figure S4.** ZFC/FC curves of a) 5.6 nm, b) 4.5 nm, c) 3.9 nm, d) 3.6 nm, e) 2.6 nm, f) 2.3 nm, g) 2.1 nm, h) 1.9 nm, and i) 1.8 nm ESM NPs.

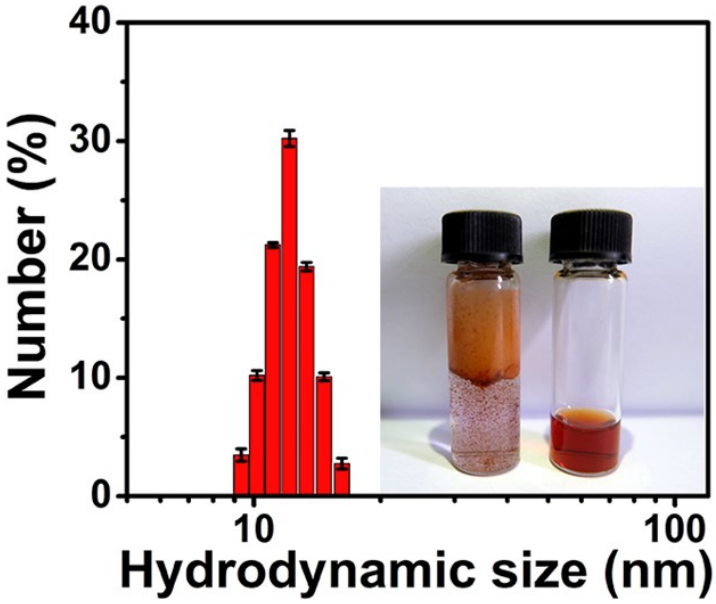


**Figure S5.** Hydrodynamic diameter of 2.3 ESM NPs. Inset: The as-synthesised ESM NPs are insoluble in water (left), and the ESM NPs are well dispersed in aqueous

solution after surface modified with carboxyl–polyethylene glycol–phosphoric acid ligand.

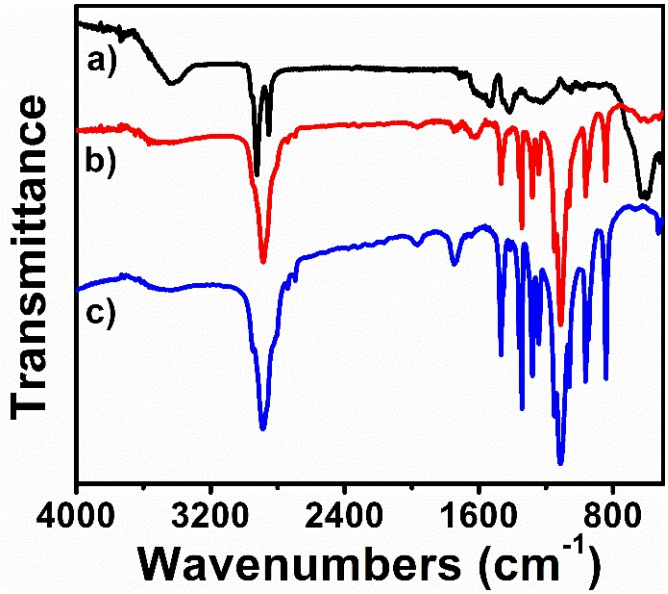


**Figure S6.** FT-IR spectra of (a) as-synthesised ESM NPs, (b) ESM NPs after ligand exchange process, and (c) carboxyl–polyethylene glycol–phosphoric acid ligand.

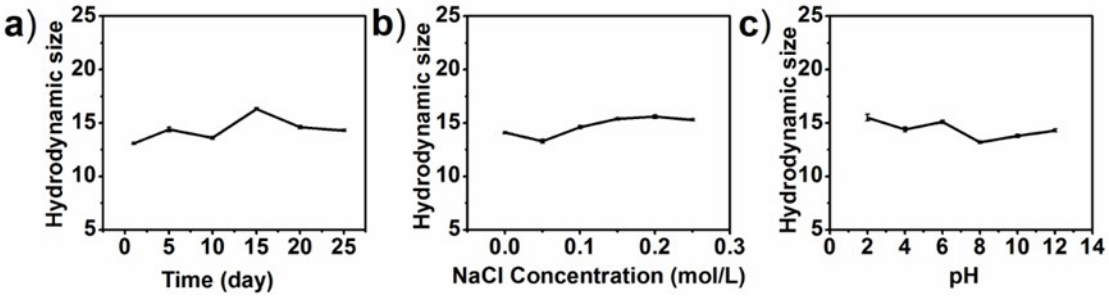


**Figure S7.** Hydrodynamic diameters of water-soluble ESM NPs (a) in deionized water within 30 days, (b) in NaCl solutions and (c) at different pH conditions.