[Supporting Information]

Low-Dimensional Nanoparticle Clustering in Polymer Micelles and Their Transverse Relaxivity Rate

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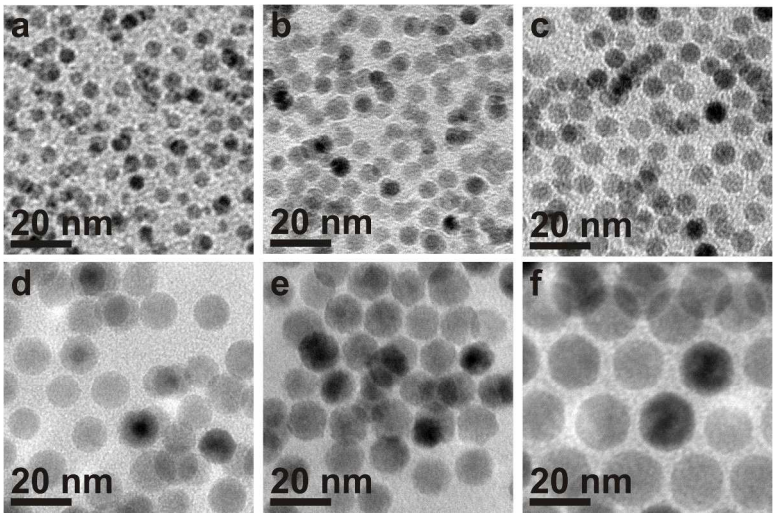

Figure S1. TEM images of synthesized iron oxide nanoparticles with measured diameters of 4.6 nm (a), 5.6 nm (b), 6.4 nm (c), 10.5 nm (d), 10.8 nm (e) and 14.9 nm (f).

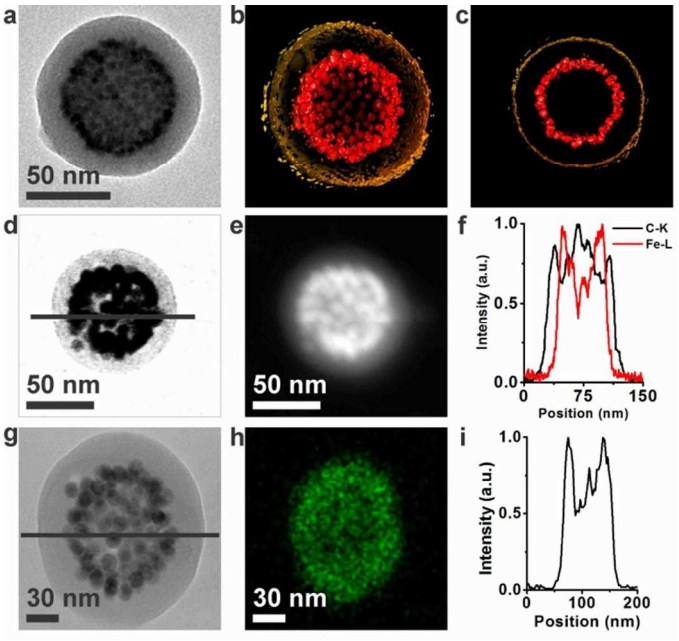

Figure S2. Characterization of magneto-core/shell assemblies using electron tomography, energy loss electron spectroscopy (EELS), and energy dispersive spectroscopy (EDS). (a-c) A TEM image (a), a 3-D surface rendering of the tomographic volume (b), and an X-Y computational slice of the 3-D volume (c) of a magneto-core/shell assembly prepared with 2.8 nm (r_{NP}) iron oxide particles and PAA_{38}-*b*-PS_{247} at 24 np wt%. (d-f) A bright field image (d), dark field STEM image (e), and EELS line scans for carbon-K and iron-L absorption edges (f) of a magneto-core/shell assembly formed with 4.4 nm (r_{NP}) iron oxide particles and PAA_{14}-*b*-PS_{250}.

(g-i) A bright-field STEM image (g), EDS mapping (h), and EDS line scan (i) for Fe of a magneto-core/shell assembly formed with 7.5 nm (r_{NP}) iron oxide particles and PAA_{38}-*b*-PS_{154}.

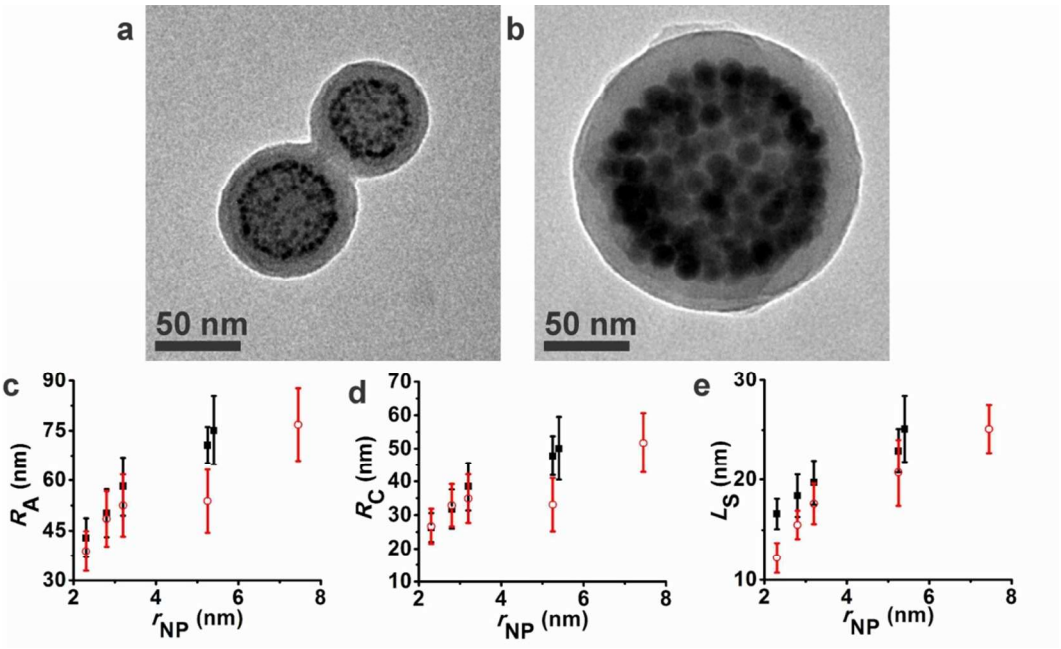

Figure S3. Nanoparticle-size dependent structural parameters of magneto-core/shell assemblies. (a-b) TEM images of magneto-core/shell assemblies formed with PAA_{38}-*b*-PS189 and r_{NP} = 2.3 nm (a) and r_{NP} = 7.5 nm (b). (c) R_A dependence on r_{NP} for PAA_{15}-*b*-PS_{107} (■) and PAA_{38}-*b*-PS_{189} (○). (d) R_C dependence on r_{NP} for PAA_{15}-*b*-PS_{107} (■) and PAA_{38}-*b*-PS_{189} (○). (e) L_S dependence on r_{NP} for PAA_{15}-*b*-PS_{107} (■) and PAA_{38}-*b*-PS_{189} (○).

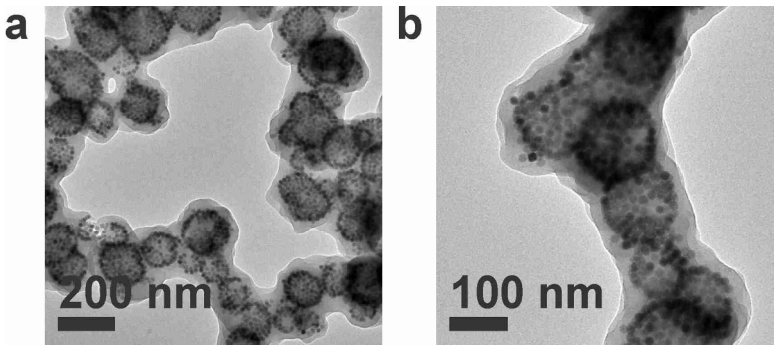

Figure S4. Additional TEM images of the nanoparticle/polymer aggregates shown in Figure 5d formed with nanoparticles with a radius of 7.5 nm and PAA_{38}-PS_{73}. When the relative size of nanoparticles to the length of the polymer exceeds a certain value, aggregates of assemblies were observed instead of discrete magneto-core/shell assemblies.

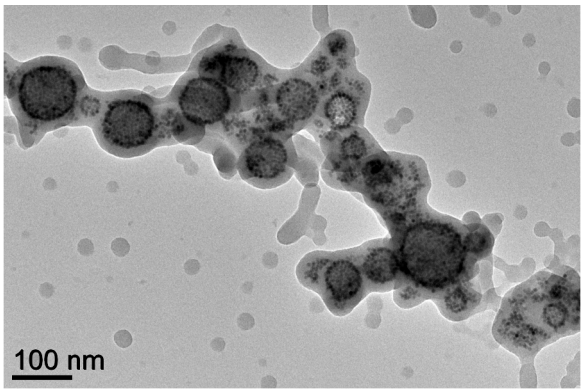

Figure S5. A TEM image of nanoparticle/polymer aggregates formed with 3.2 nm (r_{NP}) iron oxide particles and PAA_{38}-*b*-PS_{247}. The nanoparticle weight percent was 24 np wt%, and the polymer concentration was 0.23 wt %. When polymer concentration exceeds a certain threshold, large aggregates of polydisperse core-shell assemblies were formed instead of well-defined discrete assemblies.

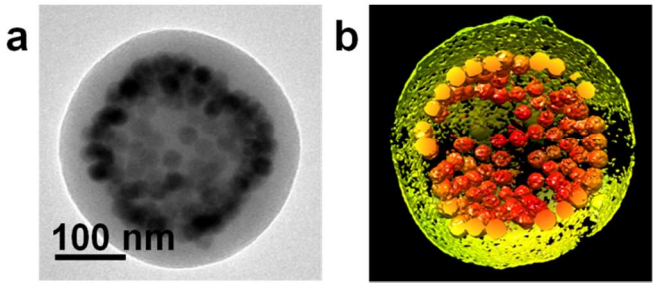

Figure S6. Electron tomography 3-D structural analysis of a magneto-core/shell assembly formed with 7.5 nm iron oxide nanoparticles and PAA_{38}-PS_{247}. (a,b) A TEM image (a) and a 3-D surface rendering of the tomographic volume (b). The assemblies were formed with the initial solvent mixture of DMF and THF (96.8 % DMF) at a polymer concentration of 0.01 wt% and a nanoparticle content of 24 np wt %.

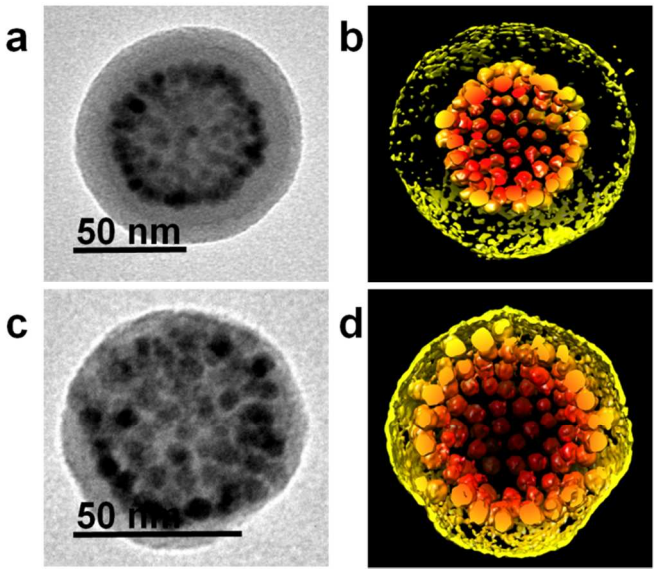

Figure S7. Electron tomography 3-D structural analysis of magneto-core/shell assemblies formed at two different np wt% using 2.9 nm iron oxide nanoparticles and PAA_{38}-PS_{247}. (a,b) A TEM image (a) and a 3-D surface rendering of the tomographic volume (b) of a magneto-core/shell assembly formed at 24 np wt%. (c,d) A TEM image (c) and a 3-D surface rendering of the tomographic volume (d) of a magneto-core/shell assembly formed at 62 np wt%. The assemblies were formed with an initial solvent mixture of DMF and THF (96.8 % DMF) at a constant polymer concentration of 0.01 wt %.

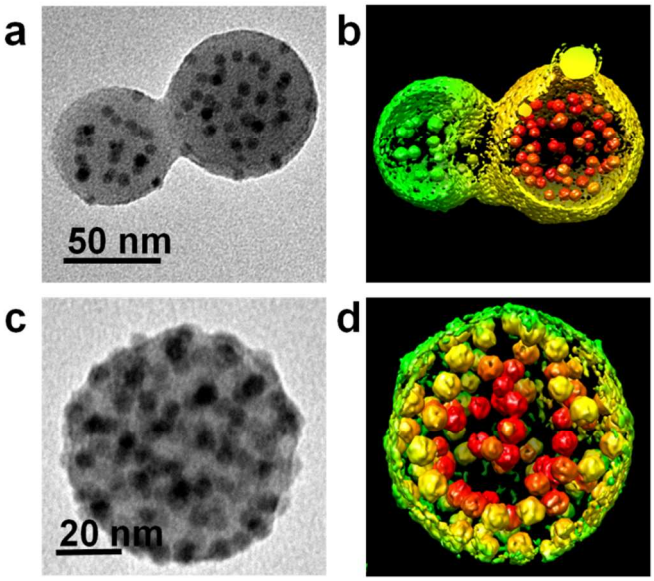

Figure S8. Electron tomography 3-D structural analysis of magneto-micelles formed at two different np wt% using 2.9 nm iron oxide nanoparticles and PAA_{38}-PS_{247}. (a,b) A TEM image (a) and a 3-D surface rendering of the tomographic volume (b) of a magneto-micelle assembled at 24 np wt%. The Movie 3 shows the reconstructed tomography data of this assembly. (c,d) A TEM image (c) and a 3-D surface rendering of the tomographic volume (d) of a magneto-micelle assembled at 62 np wt%. The assemblies were formed at a polymer concentration of 0.01 wt% with THF as the initial solvent. The Movie 4 shows the reconstructed tomography data of this assembly. The string formation is clearly seen in the Movie 4.