Supporting Information

Figures S-1 and S-2 show details regarding the transverse and longitudinal relaxivities of *MBICs* and *MBIClusters* as a function of hydrodynamic size. This material is available free of charge via the Internet at http://pubs.acs.org.

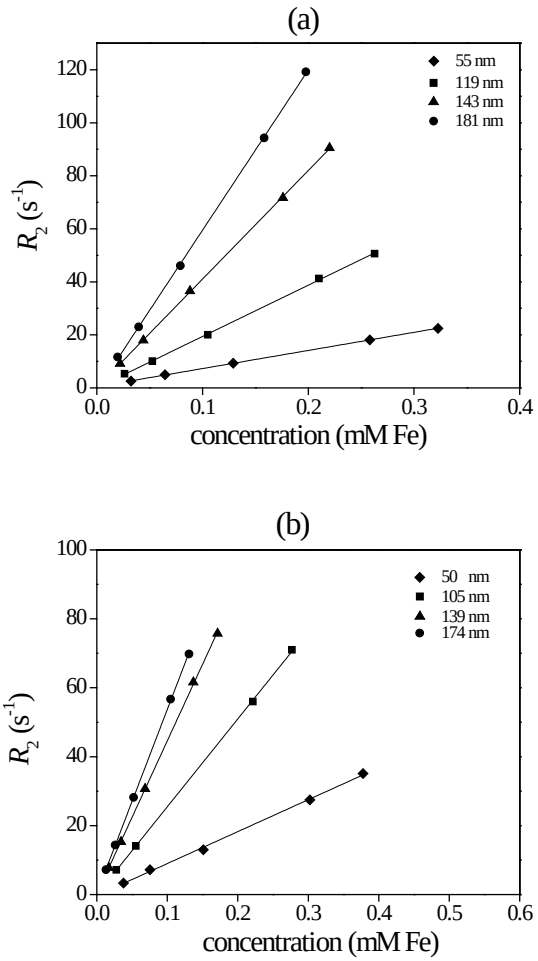

Figure S-1. Transverse relaxation rates of *MBICs* and *MBIClusters* with different intensity average diameters with a) 3.5K-6.8K H_2N-PEO-PAA, and b) 3.5K-9.5K H_2N-PEO-PAA as a function of iron concentration. Non-crosslinked *MBICs* are represented by the smallest of the average particle diameters in each case.

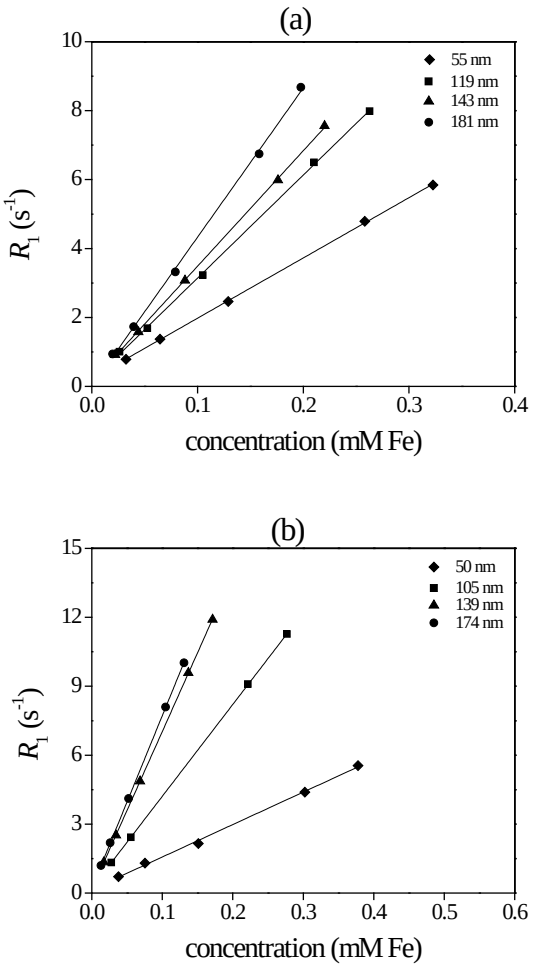

Figure S-2. Longitudinal relaxation rates of MBICs and *MBIClusters* with different intensity average diameters with a) 3.5K-6.8K H_2N-PEO-PAA, and b) 3.5K-9.5K H_2N-PEO-PAA as a function of iron concentration. Non-crosslinked MBICs are represented by the smallest of the average particle diameters in each case.

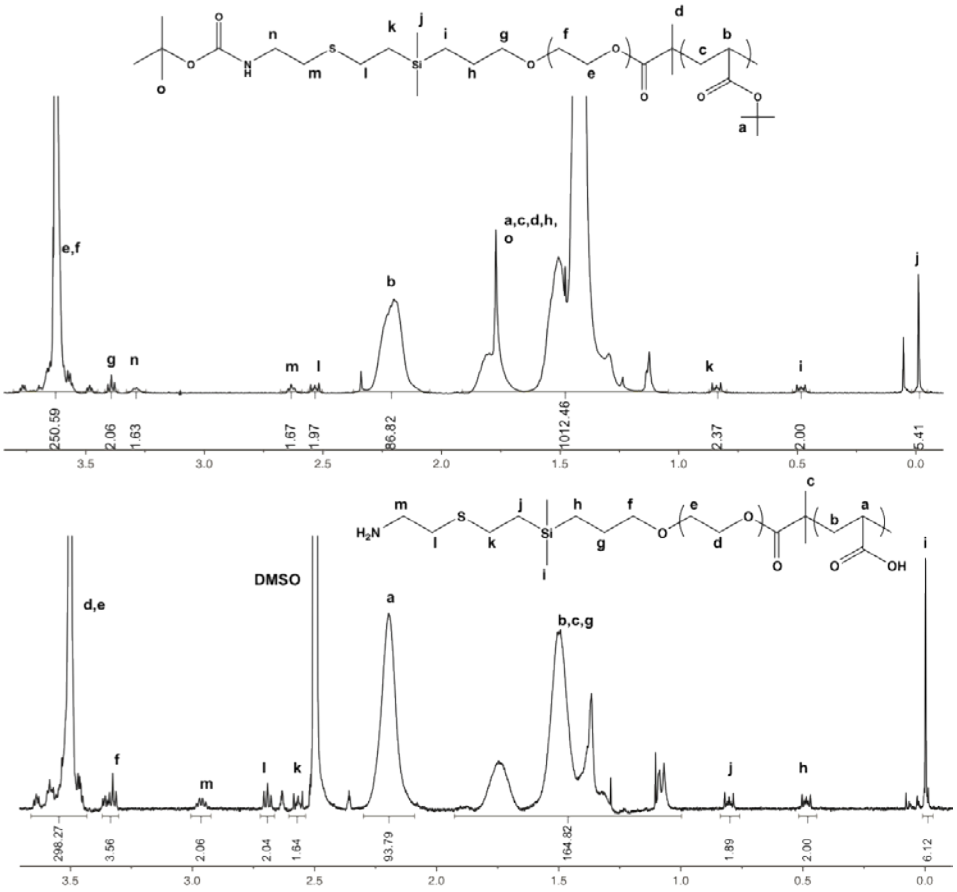

Figure S-3. ^{1}H NMR spectrum of *tboc*-NH-PEO-*b*-P*t*BA in $CDCl_3$ (top) and H_2N-PEO-*b*-PAA in D_6-DMSO (bottom)

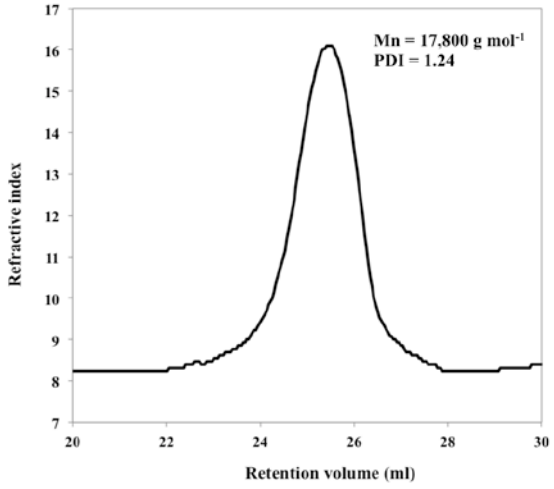

Figure S-4. SEC analysis of *tboc*-NH-PEO-*b*-P*t*BA

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