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 \url{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 H₂N-PEO-PAA, and b) 3.5K-9.5K H₂N-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$_2$N-PEO-PAA, and b) 3.5K-9.5K H$_2$N-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 $\text{tboc-NH-PEO-b-PrBA}$ in CDCl$_3$ (top) and H$_2$N-PEO-$b$-PAA in D$_6$-DMSO (bottom)

Figure S-4. SEC analysis of $\text{tboc-NH-PEO-b-PrBA}$
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