Supporting Information

(Facile synthesis of monodisperse poly(MAA/EGDMA)/Fe$_3$O$_4$ hydrogel microspheres with hollow structures for drug delivery systems: The hollow formation mechanism and effects of various metal ions on structural changes)

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Figure S1. OM images of synthetic process of the poly(MAA/EGDMA)/Fe$_3$O$_4$ composite microcapsules with four steps; (a) poly(MAA/EGDMA) microspheres dispersed in DI water, (b) in aqueous solution of pH 6.8 (c) after adding FeCl$_2$ solution and (d) after adding ammonium hydroxide.
Figure S2. XRD patterns of (a) pure poly(MAA/EGDMA) hydrogel microspheres and poly(MAA/EGDMA)/Fe₃O₄ composite microcapsules prepared using different concentration of FeCl₂ aqueous solution: (b) 0.0025M, (c) 0.0035M and (d) bare Fe₃O₄ nanoparticles prepared under same experimental condition without polymer template.
Figure S3. EDX profile of poly(MAA/EGDMA)/Fe$_3$O$_4$ composite microcapsules
Figure S4. SEM images of poly(MAA/EGDMA)/Fe$_3$O$_4$ composite microcapsules fabricated after iron ion diffusion process in aqueous solution of pH 5.0 at (a) low magnification and (b) high magnification.
**Figure S5.** Cross-sectional images of (a) pure poly(MAA/EGDMA) microspheres and (b) after adding 0.0025M iron ion solution.
**Figure S6.** (a) HR-TEM images of poly(MAA/EGDMA)/iron ions and (b) hollow poly(MAA/EGDMA)/Fe$_3$O$_4$ composite microcapsules. EDS elemental mapping of the spatial distribution of C (c and d), Fe (e and f) and O (g and h) atoms in poly(MAA/EGDMA) microspheres and poly(MAA/EGDMA)/Fe$_3$O$_4$ microspheres, respectively.
Figure S7. CLSM images of poly(MAA/EGDMA) microspheres after incubation with fluorescent dye (FITC-dextran) dispersed in aqueous solution with different pH values: (a) pH 11; (b) pH 5; (c) pH 2. Insets: fluorescence profiles along the line through the microspheres indicated in the confocal images. The image of magnification measured (a, b and c) x 3.5.
Figure S8. (a and b) SEM images of Fe$_3$O$_4$/poly(MAA/EGDMA) composite microcapsules synthesized with the monomer ratio of MAA and EGDMA; (a) 3.7:0.3 and SEM and (insets) FIB-SEM images of Fe$_3$O$_4$/poly(MAA/EGDMA) composite microcapsules synthesized with the monomer ratio of MAA and EGDMA; (c) 3.5:0.5 and (d) 3.4:0.6.
Figure S9. CLSM images of the poly(MAA/EGDMA) microspheres after incubation with fluorescent dye (FITC-dextran, green color) dispersed in aqueous solution of pH 7. The sequence was imaged upon adding the various metal ion solution: (a) CuSO$_4$; (b) FeCl$_3$; (c) SnCl$_2$. Insets: fluorescent intensity profiles along the line through the microspheres indicating that the fluorescent intensity is observed in the cavity as well as the shell of the capsule. The scale bar represents 5µm.