# Electronic Supplementary Information

**Vortex-Assisted, Nanoarchitectonic Manipulation of Microparticles with Flavonoid-$Fe^{3+}$ Complex in Biphasic Water-Oil Systems**

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## Experimental

**Materials.** Luteolin (LUT, 98%, TCI), quercetin (QUE, 95%, Sigma-Aldrich), naringenin (NAR, 93%, TCI), (+)-catechin hydrate (CAT, 98%, Sigma-Aldrich), myricetin (MYR, 97%, TCI), iron(III) chloride hexahydrate ($FeCl_3 \cdot 6H_2O$, ≥98.0%, Sigma), 1-octanol (ACS reagent, 99%, Sigma-Aldrich), Alexa Fluor 488-conjugated albumin from bovine serum (BSA-Alexa 488, Invitrogen), polystyrene microparticle (PS, diameter: 3.97 μm, Microparticles GmbH), ethanol (absolute for analysis, Supelco), tetrahydrofuran (THF, HPLC grade, Junsei), hydrochloric acid (HCl, 37%, Daejung), and fluorescein isothiocyanate-dextran (FITC-dextran, average MW: 20, 40, 150, 250, 500, and 2000 kDa, Sigma-Aldrich) were used as received. Deionized (DI) water (18.3 MΩ·cm) from Milli-Q Direct 8 (Millipore) was used.

**Synthesis of PS@LUT-$Fe^{3+}$ and Hollow Capsules.** To 5 mL of an aqueous suspension of $FeCl_3$ and PS particles (0.3% w/v) was added 5 mL of a 1-octanol solution of LUT, with predetermined concentrations of $Fe^{3+}$ and LUT. The mixture was stirred at 800 rpm at room temperature, transferred to a microtube, and centrifuged at 6000 *g* for 1 min to collect PS@LUT-$Fe^{3+}$. For the formation of hollow capsules, PS@LUT-$Fe^{3+}$ in THF (1 mL) was vortexed for 1 min and centrifuged at 2000 *g* for 1 min, and the supernatant was removed. After repeating the process three times, THF was added, and the suspension was incubated for 3 h for PS removal. For permeability test, 100 μL of a FITC-dextran solution (1 mg/mL) was added to the capsule suspension. The mixture was placed on a rotator and incubated at room temperature for 10 min, after which it was immediately analyzed for at least 100 capsules by confocal laser-scanning microscopy (CLSM).

**Characterizations.** Field-emission scanning electron microscopy (FE-SEM) imaging was performed with an FEI Inspect F50 microscope (FEI) with an accelerating voltage of 5 kV, after sputter-coating with platinum. The ζ-potential was measured with a Zetasizer Nano ZS (Malvern). Transmission electron microscopy (TEM) imaging and energy-dispersive X-ray (EDX) spectroscopy elemental analysis were conducted with a Talos F200X (FEI) with a carbon-support-film copper mesh, and CLSM imaging was performed with an LSM 700 (Carl Zeiss). Atomic force microscopy (AFM) images were taken in the QI mode with a NanoWizard4 BioAFM (JPK). X-ray photoelectron spectroscopy (XPS) spectra were obtained with a Sigma Probe (Thermo VG Scientific), and UV-Vis absorbance was measured with a SpectraMax iD5 microplate reader (Molecular Devices) in a 96-well plate (UVmax, SPL Life Science). Fourier-transform infrared (FT-IR) spectrum was obtained with an FT-IR spectrophotometer (Nexus 670, Thermo Nicolet).

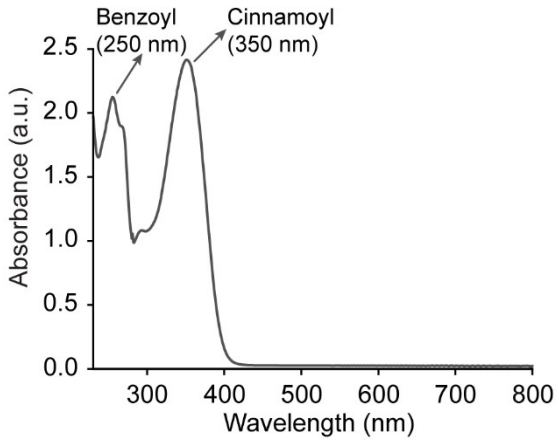


**Figure S1**. UV-Vis spectrum of LUT in water/ethanol (1/20 v/v).

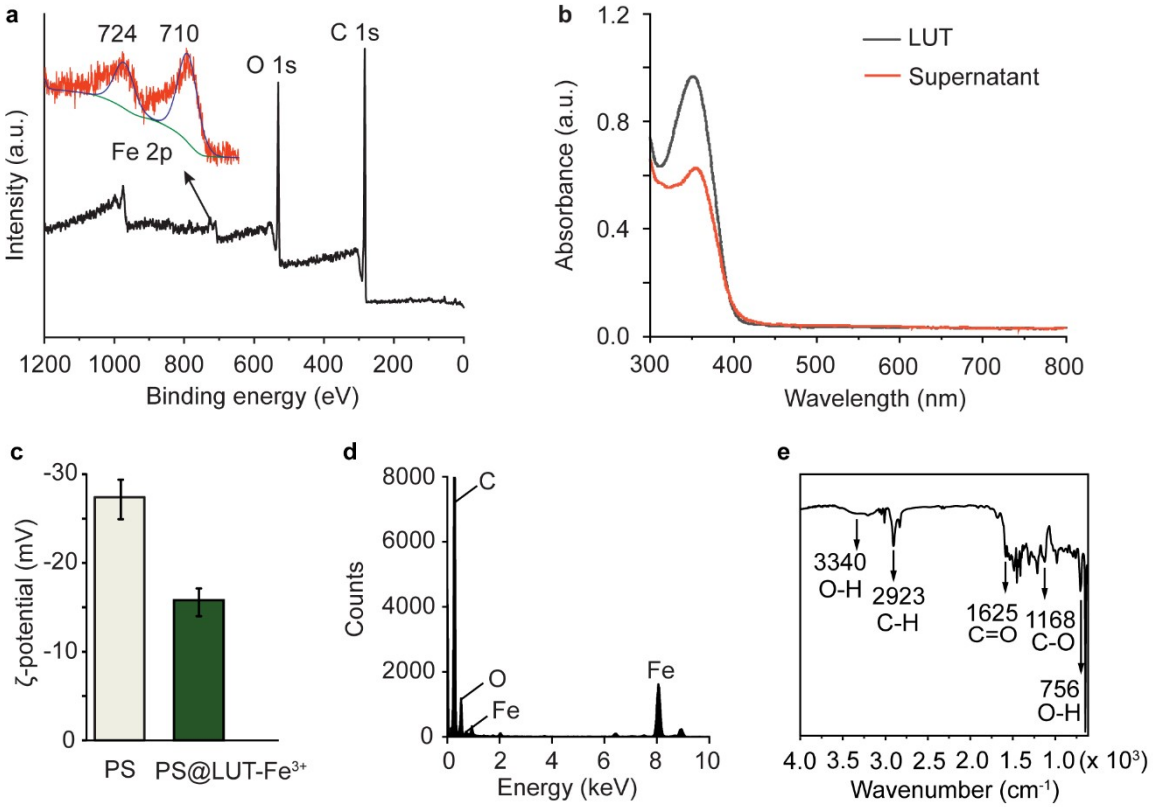


**Figure S2.** (a) XPS spectra of PS@LUT-$Fe^{3+}$. (b) UV-Vis spectra of (black) LUT and (red) supernatant after shell degradation of PS@LUT-$Fe^{3+}$. (c) ζ-potentials of PS and PS@LUT-$Fe^{3+}$. (d) EDX spectrum of LUT-$Fe^{3+}$ capsules. (e) FT-IR spectrum of LUT-$Fe^{3+}$ capsules.

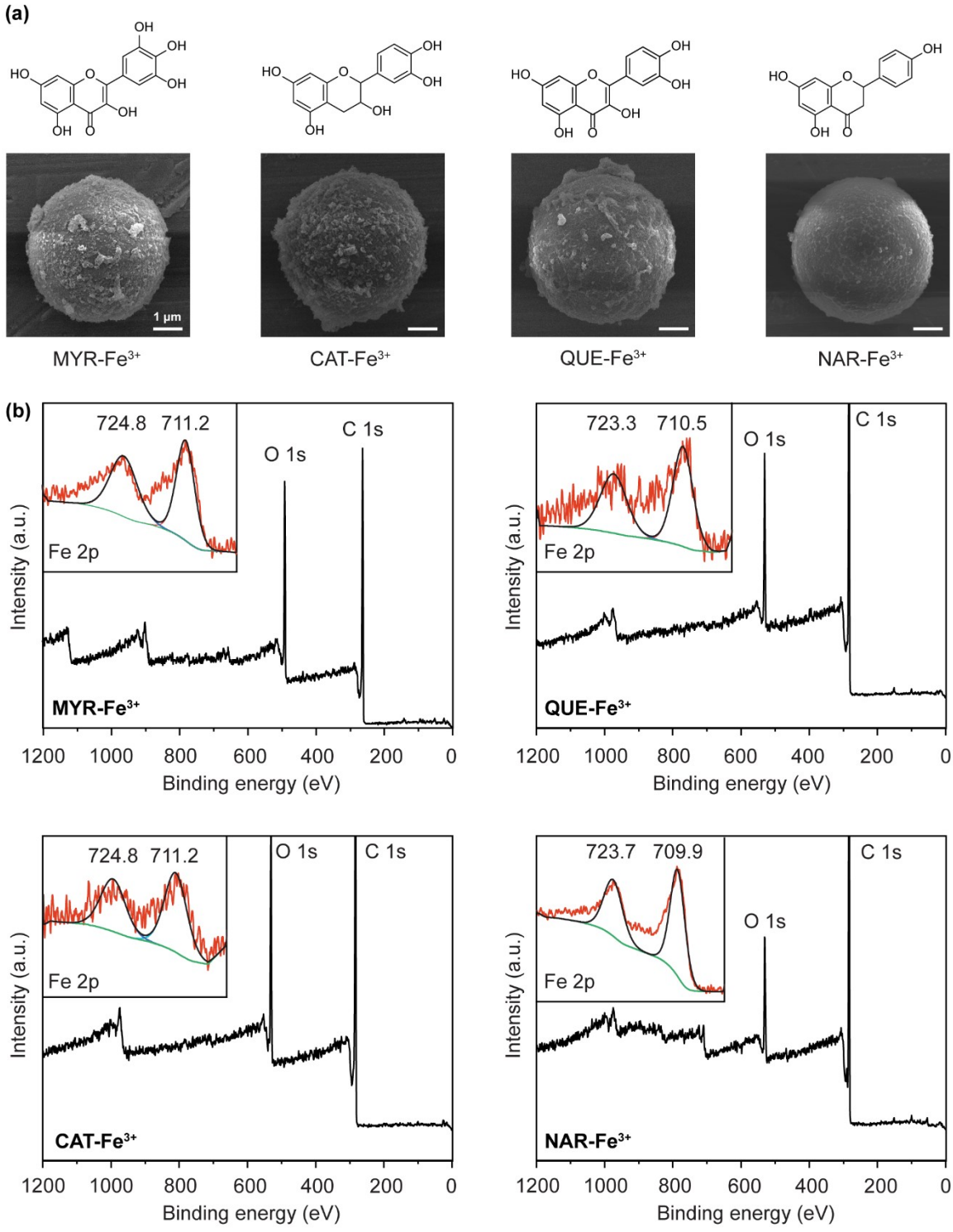


**Figure S3.** (a) FE-SEM images of PS@flavonoid-$Fe^{3+}$. (b) XPS spectra of PS@flavonoid-$Fe^{3+}$. MYR: myricetin; QUE: quercetin; CAT: catechin; and NAR: naringenin.

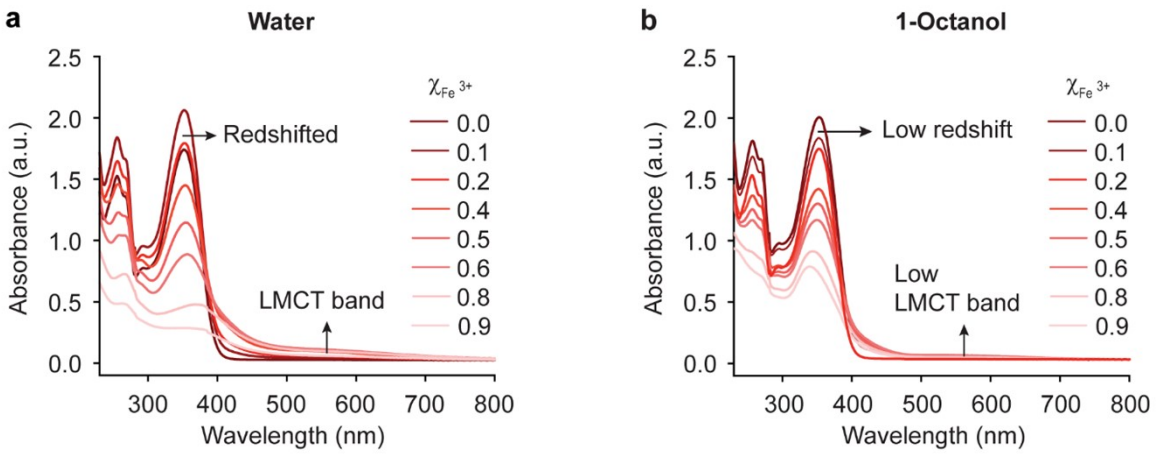


**Figure S4.** UV-Vis spectra of solutions containing LUT and $Fe^{3+}$ with varied $\chi_{Fe}$: (a) water and (b) 1-octanol phases.

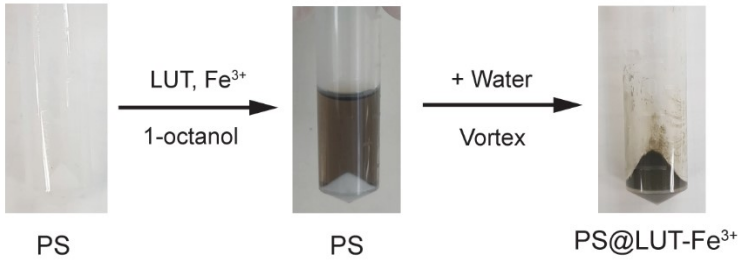


**Figure S5.** Control experiment with use of 1-octanol only. The white PS particles in the middle image indicate no shell formation, while the brownish color in the right image indicates the formation of LUT-$Fe^{3+}$ shells on the PS particles after the addition of water and vortexing.

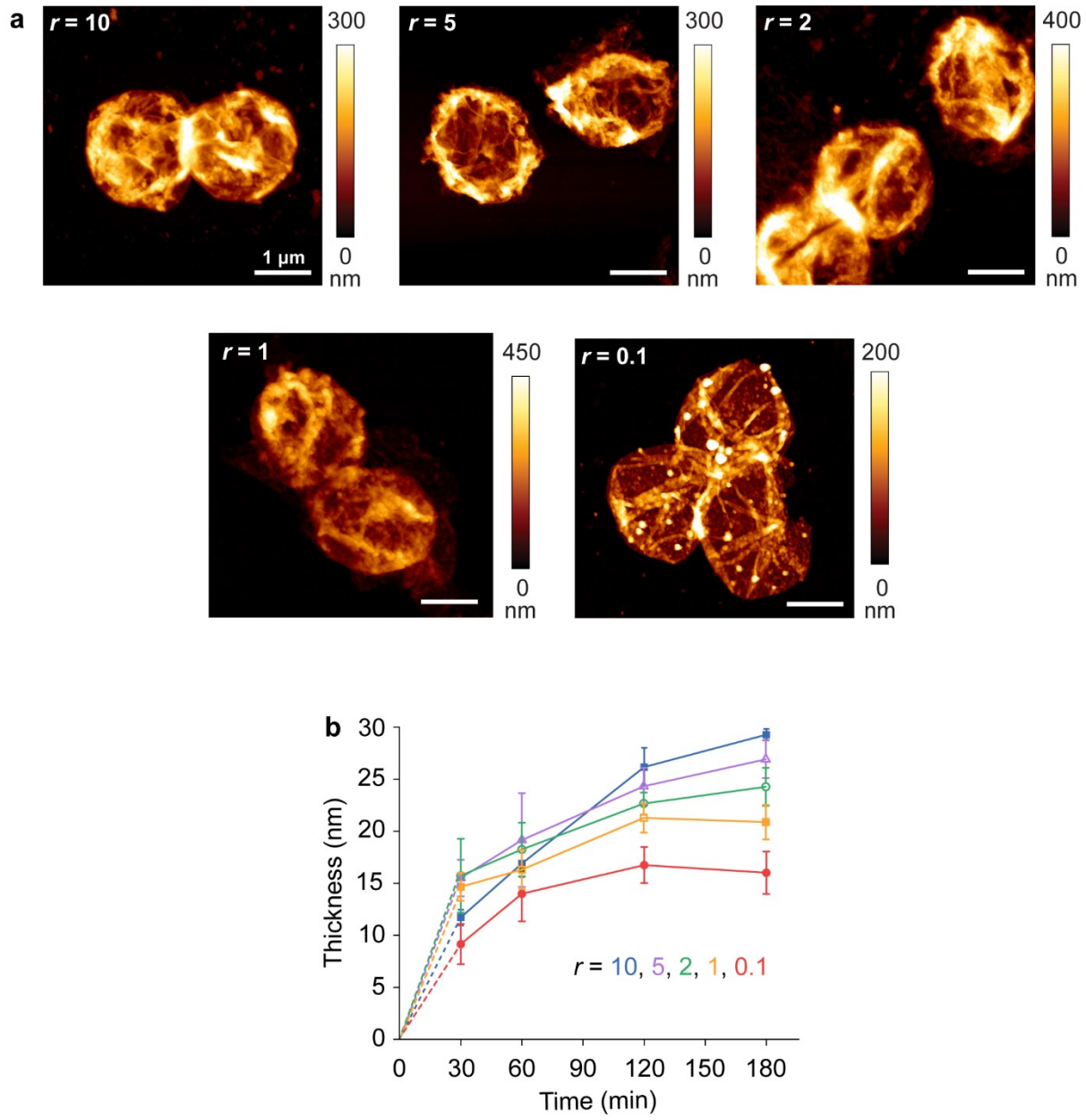


**Figure S6.** (a) AFM images of LUT-$Fe^{3+}$ capsules for various *r* values, after 3 h of reaction. (b) Time-lapse shell thicknesses of LUT-$Fe^{3+}$ capsules for various *r* values. *r* = 0.1, 1, 2, 5, 10.

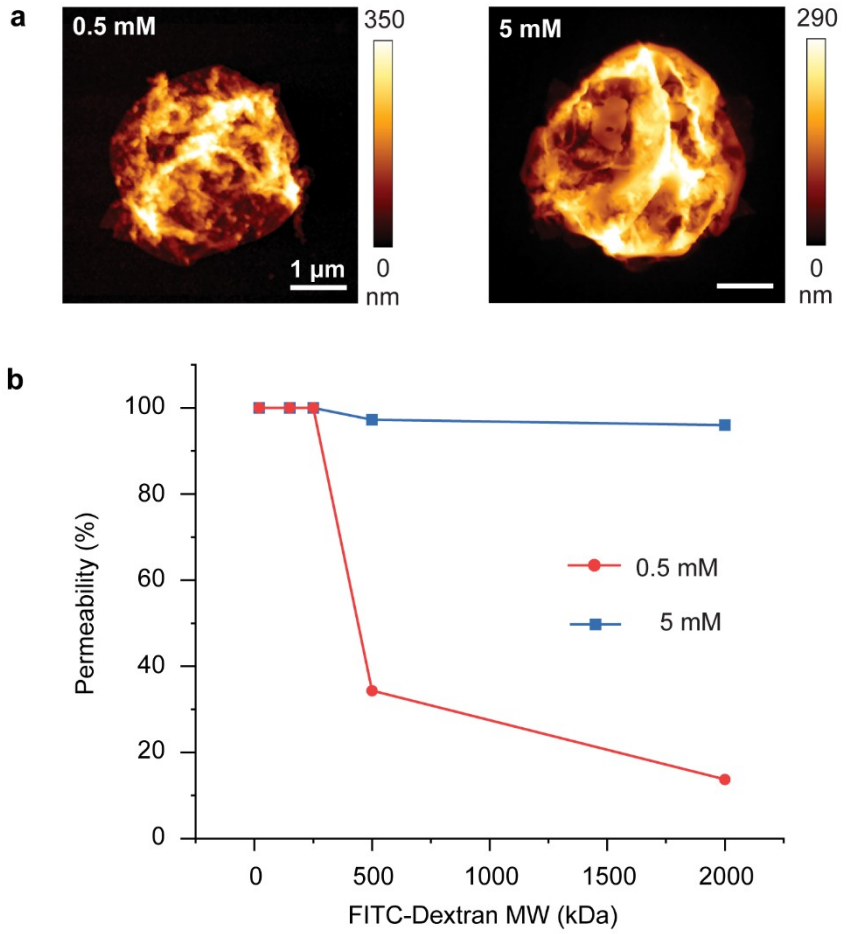


**Figure S7.** (a) AFM images of LUT-$Fe^{3+}$ capsules prepared with (left) [LUT] = [$Fe^{3+}$] = 0.5 mM and (right) [LUT] = [$Fe^{3+}$] = 5 mM. (b) Permeability of LUT-$Fe^{3+}$ capsules for FITC-dextrans: (red) [LUT] = [$Fe^{3+}$] = 0.5 mM and (blue) [LUT] = [$Fe^{3+}$] = 5 mM.