Supporting Information

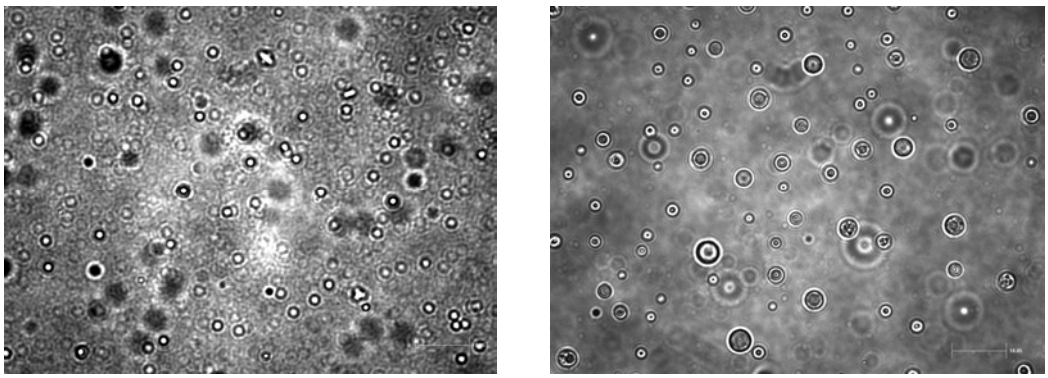
Materials and Methods

Materials and Reagents

Poly(methyl methacrylate) (PMMA) particles were obtained from Fluka (1 μm, 10 % solid content). Poly(styrene) (PS) particles were obtained from Seradyn (0.132, 0.301, and 0.520 μm, 10 % solid content). $Ru(dpp)_3Cl_2$ (Fluka) was dissolved in ethanol (anhydrous, VWR) at 1 mg/mL. Fluorescein isothiocyanate (FITC) and sodium dodecyl sulfate (SDS, 98%) were from Sigma-Aldrich. Methylene chloride (CH_2Cl_2, 99%) was from Fluka. Acetone (>99.5%) was purchased from Richard-Allan Scientific. Sodium poly(styrene sulfonate) (PSS, Mw ~70,000), poly(allylamine hydrochloride) (PAH, Mw ~70,000) were obtained from Sigma.

Loading of hydrophobic material into polymer particles

The particles were first diluted to 0.25-0.5 % solid content with DI water and 25 μL of 5 mg/mL SDS (98 %, Aldrich) was added to 100 μL of 0.5 % (w/w) PMMA (or PS) particles in microcentrifuge tube with gentle stirring for 15 min. Then, 25 μL of CH_2Cl_2 was added into the dispersion for another 30 min to allow the organic solvent to swell the particles. During this process, the surfactant prevents swelled particles from aggregation. Fluorescent dye was added into the dispersion as 25-50 μL of 1 mg/mL $Ru(dpp)_3Cl_2$/ethanol solution (or FITC/ethanol solution), followed by 50 μL of acetone. The dispersion was kept stirring for 30 min and then keep at room temperature with the microcentrifuge cap open for about 30 min to 1 hr to partially remove the organic solvent. The particles were then triple centrifuged at 10000 rpm at 10 °C and rinsed with DI water to remove unloaded Ru(dpp) or FITC. The 1 μm fluorescent PMMA particles were fixed in poly(acrylamide) gel to avoid motion of tiny particles during confocal imaging. Confocal fluorescence micrographs were collected with a Leica TCS SP2 laser scanning system equipped with a 63× oil immersion objective. A fluorescent microscopy (Nikon ECLIPSE TE2000-U, with 100× oil immersion objectives) was used to show the swelling of PMMA particles before and after adding of organic solvent. The following optical images clearly show that the PMMA particles could expand to about 2~3 times of their original size and keep separated after adding of CH_2Cl_2 and acetone.



(a) (b)

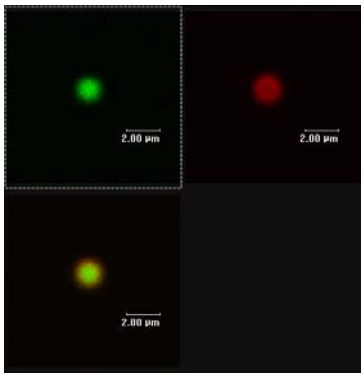
Optical images of PMMA particles (a) before and (b) after adding of CH_2Cl_2 and acetone.

Fluorescence spectrum study of Ru(dpp) in different medium and its oxygen sensing property

Fluorescence spectroscopy was used to check the emission wavelength of $Ru(dpp)_3Cl_2$-loaded PMMA particles compared with that of Ru(dpp) (1 mg/mL ethanol solution) in ethanol or DI water, using 460nm as excitation wavelength. The emission spectra were also collected for Ru(dpp)-loaded PMMA particles in DI water by alternate purging of O_2 and N_2, respectively, to check the oxygen quenching property of Ru-loaded particles.

Layer-by-Layer of polyelectrolytes on fluorescent dye-loaded particles

The PSS and PAH solutions used for alternating adsorption of {PAH/PSS} multilayers on fluorescent dye-loaded particles were prepared in DI water at 2 mg/mL with 0.5 M NaCl. The particles were suspended in DI water via vortexation and ultrasonication for 10 min prior to LbL assembly. There may still be some surfactant on the polymer surface. However, the existence of surfactant won't affect the assembly of polyelectrolytes according to our experiments. For deposition of each coating, 1 mL of polyelectrolyte solution was added to a microcentrifuge tube containing 100 µL of particle suspension. Adsorption was allowed to proceed for 20 minutes, after which the suspension was centrifuged to separate the particles from remaining un-adsorbed polyelectrolyte. The particles were then triple-rinsed with DI water by successive centrifugation cycles. The process was repeated for the oppositely-charged polyelectrolyte, and then alternated until a total of three bilayers of {PAH/PSS} films were obtained. The assembly of polyelectrolyte layers on Ru-loaded PMMA particles was monitored by electrophoretic mobility measurements (ZetaPlus Zeta Potential Analyzer, Brookhaven Instrument Corp.).



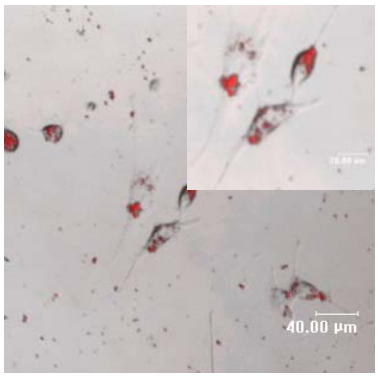
Confocal images of FITC loaded 1 µm PMMA particles, coated with PAH/PSS, PAH labeled with TRITC.

Co-culture of fluorescent particles with NIH-3T3 fibroblasts

Unless stated otherwise, all cell culture reagents were purchased from Atlanta Biologicals. Standard cell culture techniques were used for all cell experiments. The NIH-3T3 fibroblasts were cultured in a humid 37°C/5% CO_2 incubator in pH 7.4 growth media consisting of Dulbecco's Modified Eagle Medium (DMEM) high glucose supplemented with 10 % calf serum, 100 units/mL penicillin, 100 µg/mL streptomycin, and 10 µg/mL gentamycin. Cells were grown near confluence in Fisher cultureware, washed twice with warm Hank's balanced salt solution (HBSS), detached with trypsin (Sigma), and passaged once a week.

Cells were plated at a density of 2000 cells/well in 24-well TCPS plate, followed by adding 20 µL of Ru-loaded particles with/without {PAH/PSS} coating. The fibroblast and particles were cultured for 2 days

and imaged with confocal microscopy, equipped with a 10× objective.



Confocal images of fibroblast cultured with Ru(dpp)-loaded 0.3 µm PS particles, coated with $\{PAH/PSS\}_2$.