
S1: Product purification by an external magnetic field.  
a) The unpurified product comprises ball-in-ball particles filled with both, Alexa Fluor 488-HSA and TRITC-HSA (two-colored particles) and the coproduct containing only Alexa-HSA (small green particles).  
b) Magnetite containing ball-in-ball particles after collection by a magnetic field.

S2: Real time movie visualising the conversion of a ball-in-ball particle (type II) into a shell-in-shell capsule. Calcium extraction was performed by mixing equal volumes of the particle suspension and 0.2 M EDTA on a microscope glass-slide. The reaction was monitored by CLSM. The disruption of the imaging after 1 - 2 sec corresponds to the addition of EDTA.

S3: Principle of a coupled enzymatic test using glucose oxidase (GOD) and peroxidase (POD) in a shell-in-shell capsule. All substrates and products of GOD and POD can freely diffuse through the polyelectrolyte shells, whereas GOD and POD are retained in separated capsule compartments. Oxidation of glucose by GOD in the outer capsule compartment is accompanied by the generation of H₂O₂. After permeating the intersecting polyelectrolyte multilayer (PEM), accumulation of H₂O₂ is coupled to the conversion of Amplex Red to fluorescent resorufin by POD.
S4-S6: Real time movies visualising the enzyme catalysed resorufin formation within shell-in-shell capsules (recorded by CLSM). **S3**: GOD in the outer and POD in the inner compartment (according to S2): After preincubation with 50 mM glucose the POD reaction was started by adding 15 µM Amplex Red. Almost immediately the inner compartment fills with resorufin that subsequently spreads into the outer compartment. **S4 (negative control for POD)**: POD in the inner, but no GOD in the outer compartment. The same treatment as described above does not lead to a likewise formation of resorufin. Only a faint background fluorescence is visible in the vicinity of the intersecting polyelectrolyte shell. **S5 (positive control for POD)**: Within the very same capsule Amplex Red is converted into resorufin by POD immediately after the addition of 0.3 % H₂O₂. Capsules containing GOD in the outer, but no POD in the inner compartment, as well as capsules lacking both enzymes do not show resorufin formation under any of the described conditions (data not shown).