Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.

Electronic Supplementary Information

Enhanced Cytoplasmic Release of Drug Delivery Systems: Chloroquine as a Multilayer and Template Constituent of Layer-by-Layer Microcarriers

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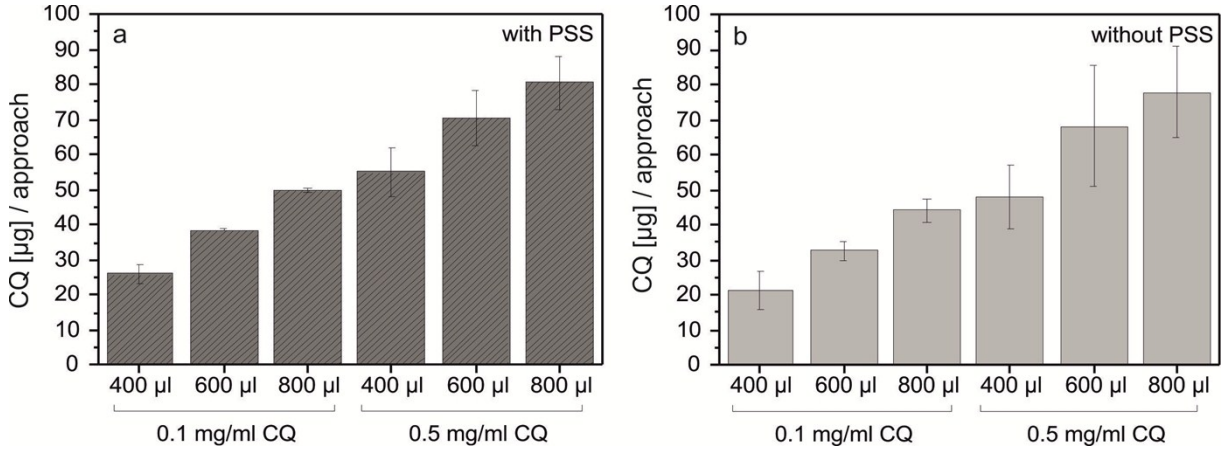

Figure S1: Assembly of CQ into the pre-fabricated $CaCO_3$ template. Different concentrations of CQ were applied in different NaCl concentrations. In **S1-a**, CQ was assembled into a PSS (negatively charged) doped $CaCO_3$ template, **S1-b** shows the CQ assembly without PSS.
Due to the variety in particles size (5µm +/- 1µm) after preparation, mean CQ assembly into particles of an entire preparation cycle was plotted (approach = 300µl $CaCl_2$ + 300µl Na_2CO_3 which correlates with about 5×10^8 $CaCO_3$ particles of 5µm in diameter).

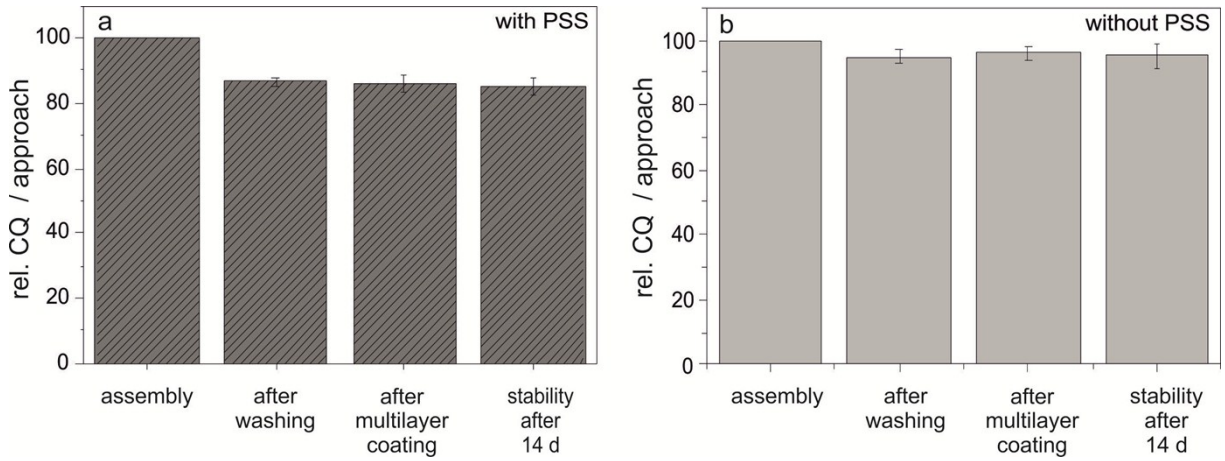

Figure S2: Template-assembled CQ shows no difference in CQ amount after washing, after virtual biopolymer multilayer coating and under storage conditions (14 d, 4°C). In case of biopolymer coating, coating procedure was simulated by applying all "coating steps" and "washing procedure" according to the assembly of 8 layers. Leaving out polymers ensured the photometric detection of CQ without interference of potential polymer material in supernatant. Data are presented in relation to CQ amount after assembly. In **S2-a**, $CaCO_3$ template was previously doped with PSS, in **S2-b** no PSS was used.

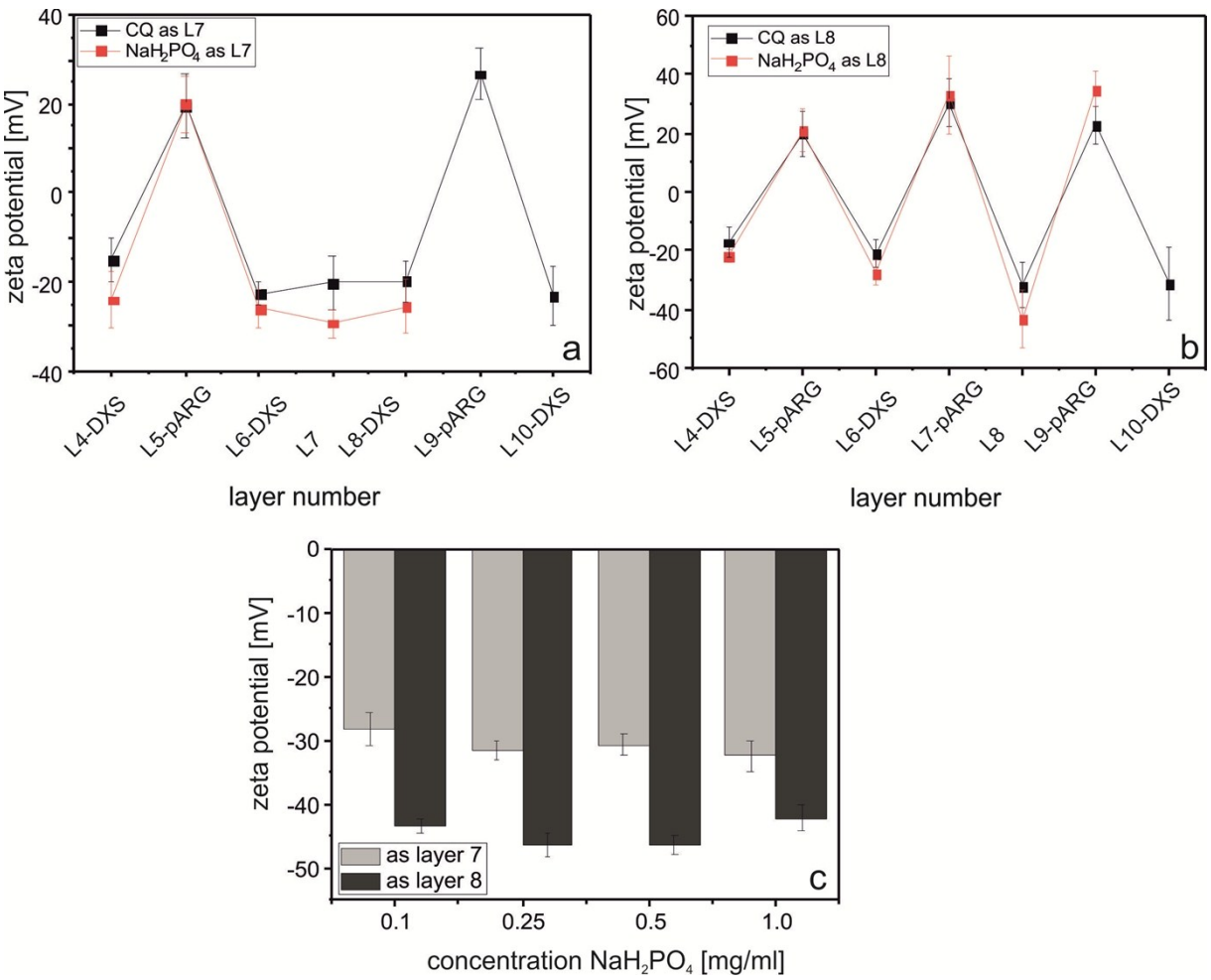

Figure S3: Zeta potential measurement of NaH_2PO_4 as an CQ component solely "assembled" in 0.5 mg/ml in different layer positions, either as 7th layer following negatively charged DXS (a) or as 8th layer following positively charged pARG (b). Three washing steps were performed after incubation with particles. In both cases, a surface layer with higher negative potential could be detected compared to CQ in the same position. Zeta potential values are independent of NaH_2PO_4 concentration (c).

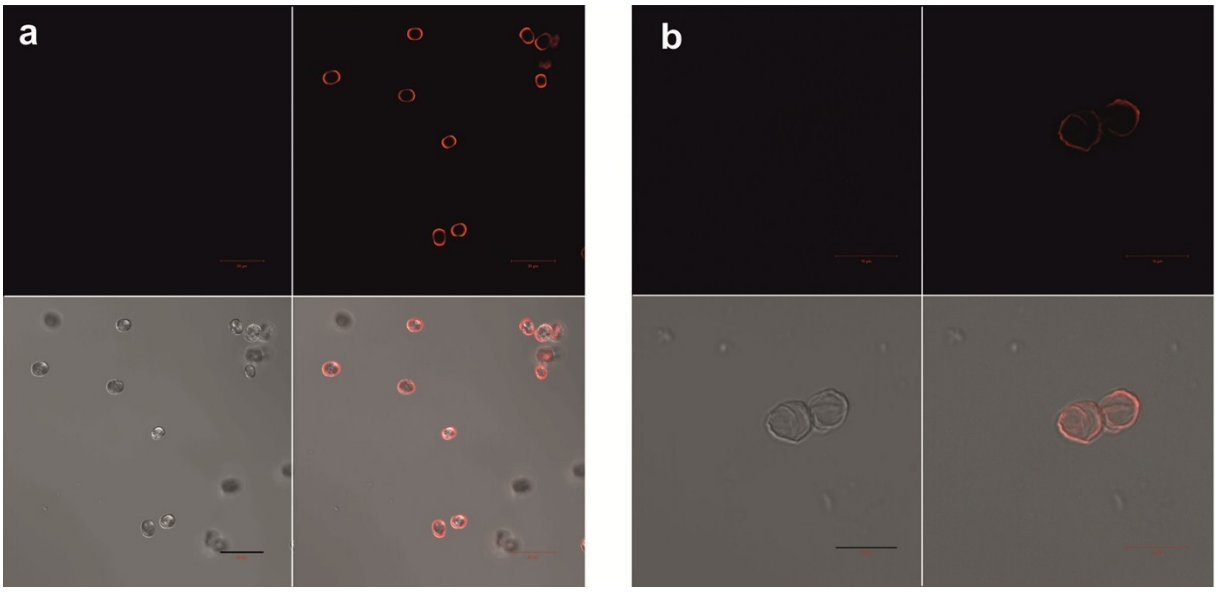

Figure S4: Investigations of LysoTracker Green staining of LbL-microcarriers. Carriers were stained for 1h according cell experiments. In a) LbL-microparticles, in b) LbL-microcapsules are shown. Both carrier types were equipped with RITC-labeled pARG as a multilayer constituent (upper panel, right). LysoTracker Green was applied in cell relevant concentrations (upper panel, left). No unspecific interaction of LysoTracker Green into the multilayer could be detected proved by the non-detectable green fluorescence intensity. Those results qualify the staining regarding real LbL carrier tracking within cells by allowing a clear distinction between carriers in phagolysosomes and cytoplasm. Transmission image and entire overlay are shown in lower panel. Scale bar (a): 20µm, scale bar (b): 10µm.