

# Architectural layer-by-layer assembly of drug nanocapsules with PEGylated polyelectrolytes

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## Supplementary Information

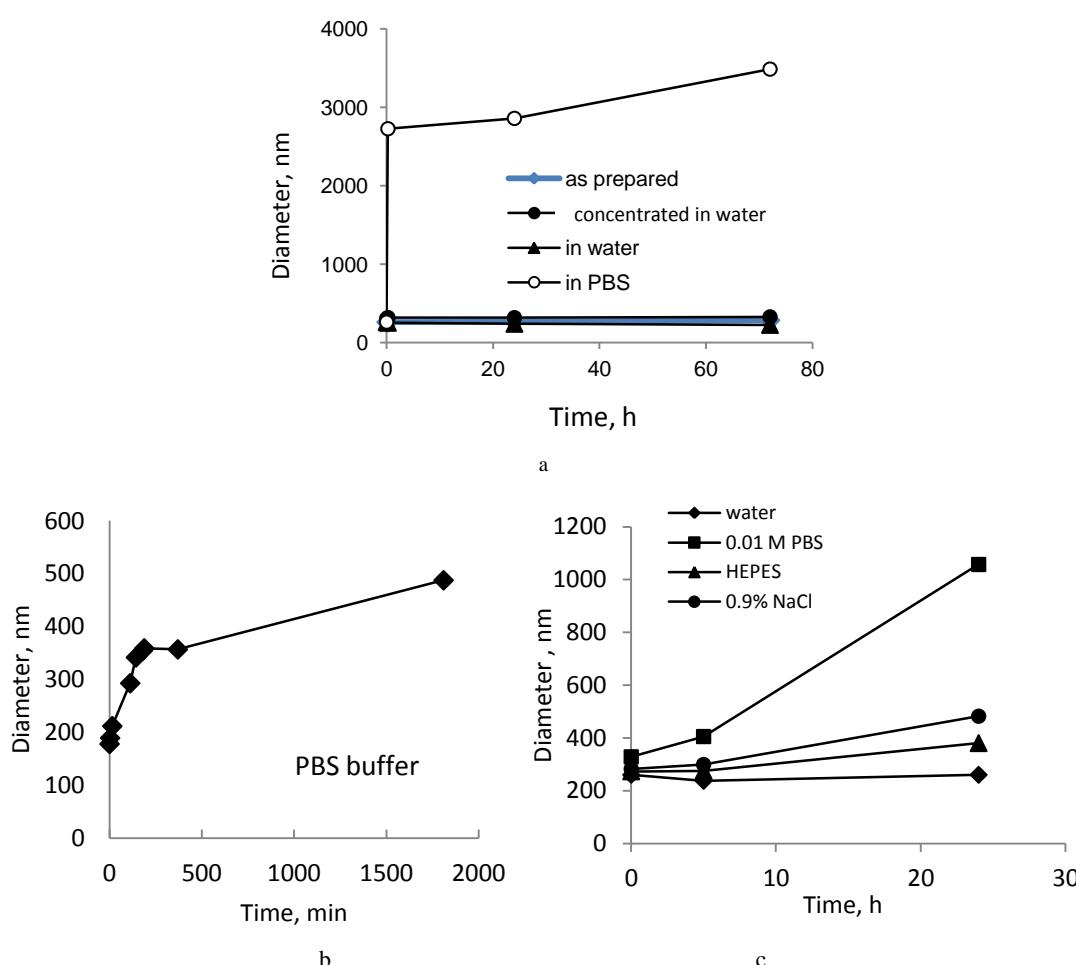


Fig. A Colloidal stability of LbL-coated paclitaxel nanoparticles (prepared in water) in different media. Shell: a) (PLL/Hep)<sub>4</sub>/PLL b) (PLL/Hep)<sub>4</sub>/PLG65[4.5]5/Hep, b) (PLG65[4.5]5/Hep)<sub>4</sub>

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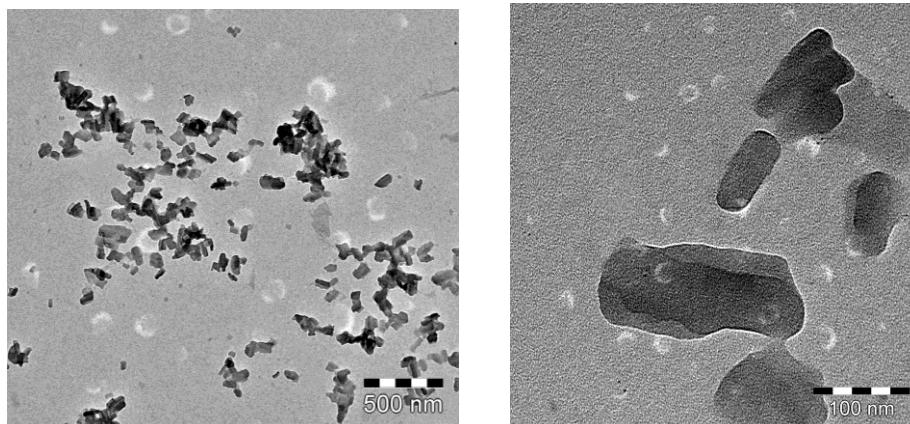


Fig.B TEM images of camptothecin nanoparticles coated with a Hep/(PLB16-5/Hep)<sub>4</sub> shell

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Table A. Content of surfactants remaining adsorbed on PXT colloids\*

Surfactant	Recovery of crystalline PTX nanoparticles, %	Amount of surfactant, remaining adsorbed on nanoparticles, $\mu\text{mol}/\text{mg}$ PTX	Hydrodynamic diameter, nm
Docusate sodium salt (AOT)	84.2 $\pm$ 7.4	1.03 $\pm$ 0.23	140-210
Glyclic acid ethoxylate oleyl ether (Oleth-6 carboxylic acid)	89	0.56	92.5 $\pm$ 2.5
Glycolic acid ethoxylate lauryl ether (Laureth-6 carboxylic acid)	64	0.65	126.6 $\pm$ 2.5
Dodecyl sulphate sodium salt (SDS)	76	0.17	2000-9000
SDS- no PVP	$\sim$ 100	0.27	$\sim$ 300
Poly(maleic acid-co-olefin) sodium salt	-	0.17-0.25	122.5 $\pm$ 2.0

25 \*The amount of surfactants adsorbed on PTX nanoparticles was estimated using the pinacyanol assay for negatively charged sites.

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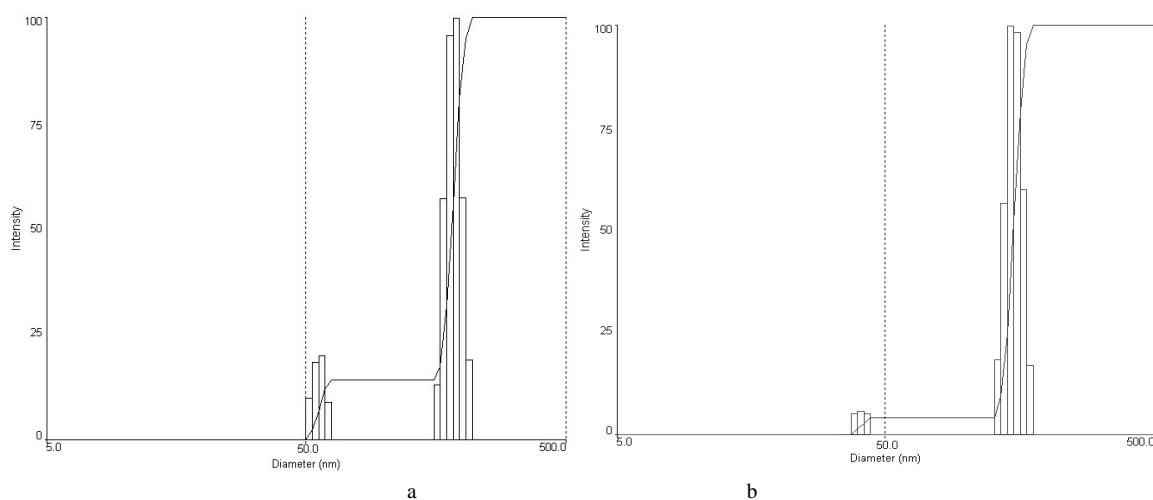


Fig. C DLS distribution of apparent hydrodynamic diameter of LbL coated PTX nanoparticles as prepared.

Shell: a) (PLB16-5/Hep)<sub>3.5</sub>; b) (PLB16-5/Hep)<sub>7.5</sub>

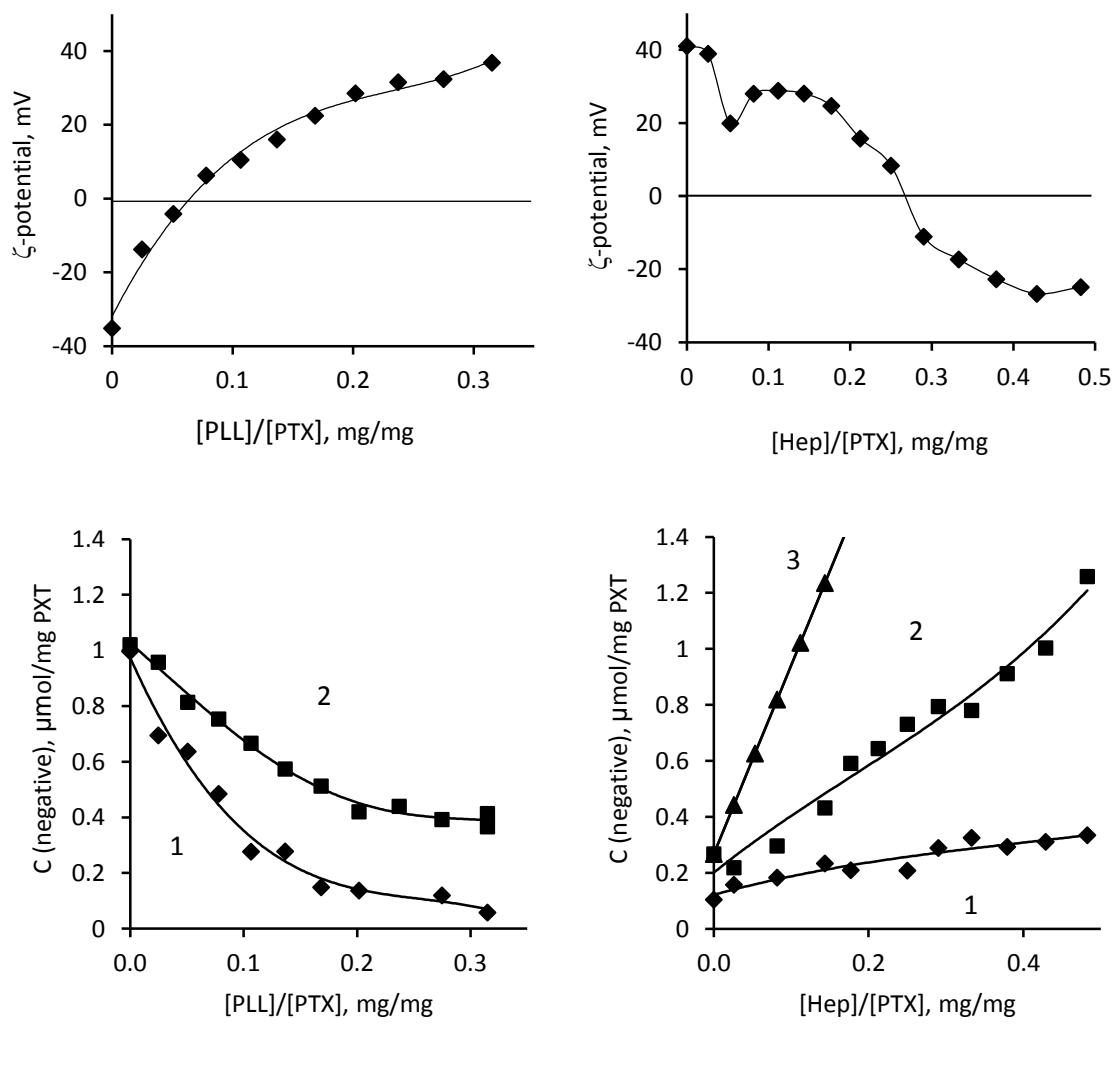


Fig. D Changes of  $\zeta$ -potential (upper row) and amounts of negative sites estimated by the pinacyanol assay (lower row) in the process of step-wise addition of a) PLL to PTX/AOT cores, b) Hep to PTX/AOT/PLL nanoparticles. Lower row: 1 – nanoparticles, 2 – total experimental, 3 – calculated.

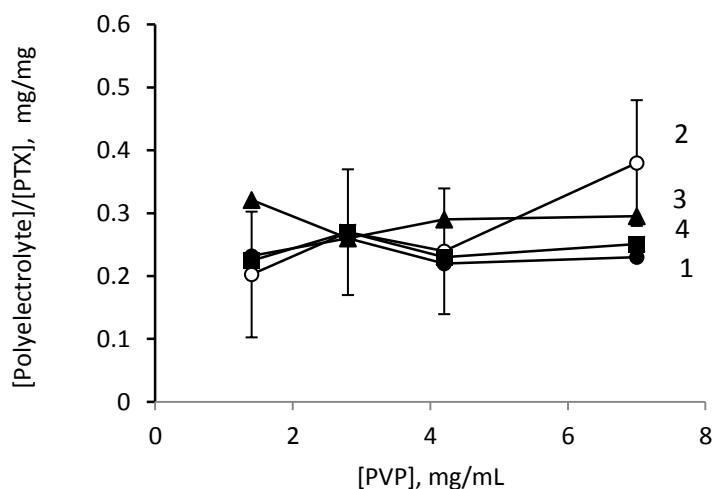


Fig.E Influence of PVP concentration on amounts of PLB16-5 (1, 3) and Hep (2, 4) added to reverse  $\zeta$ -potential of nanoparticles. The polyelectrolytes were added in the following orders: 1-2-3-4

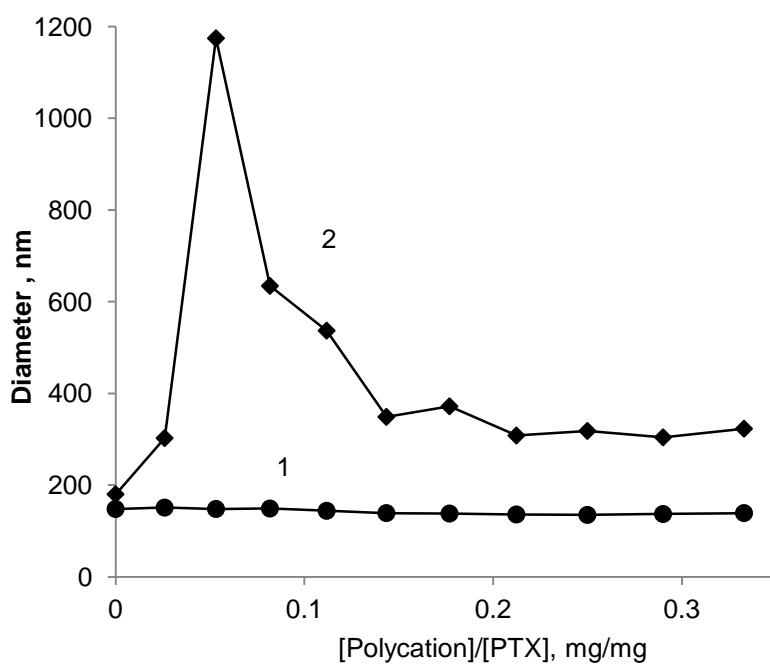


Fig. F Changes of apparent hydrodynamic diameter of PTX nanoparticles in the process of step-wise addition of heparin to nanoparticles coated with a PLB16-5 (1) and a PLL (2) layer

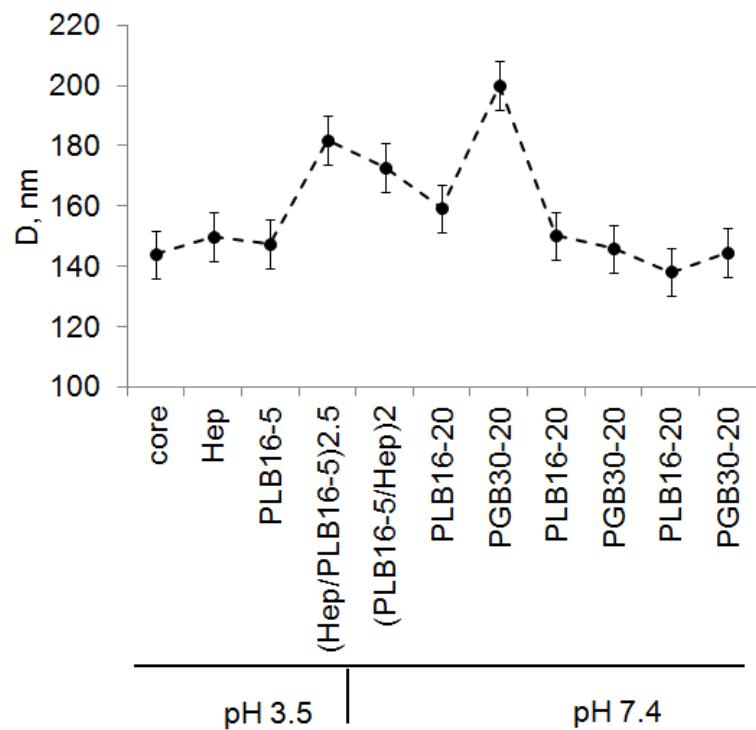


Fig.G Changes of hydrodynamic diameter of CPT nanoparticles upon adsorption of different polyelectrolytes on their surface (*a ZetaPlus Brookhaven instrument*)

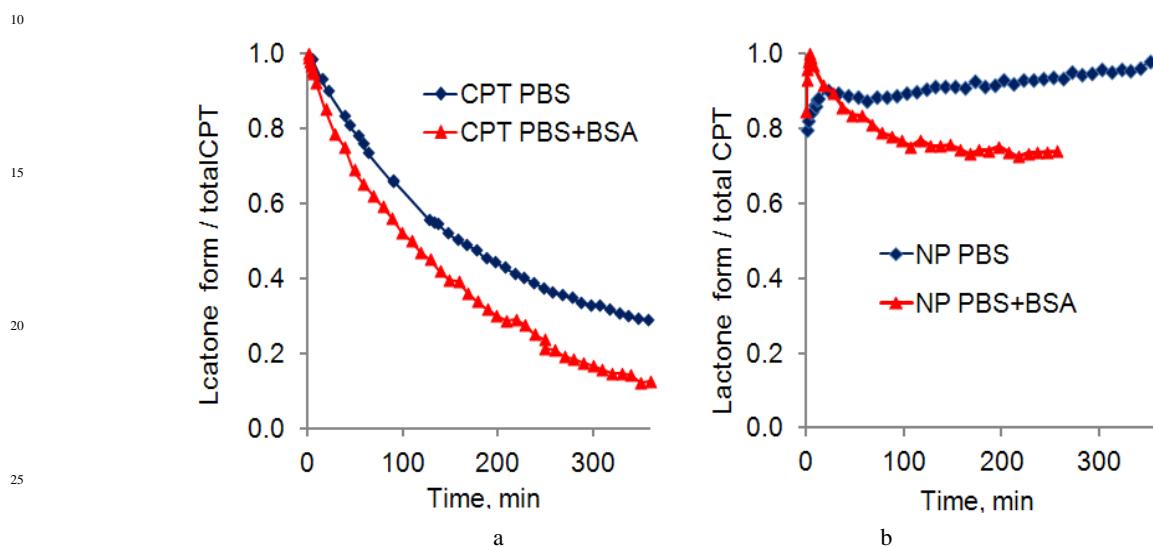
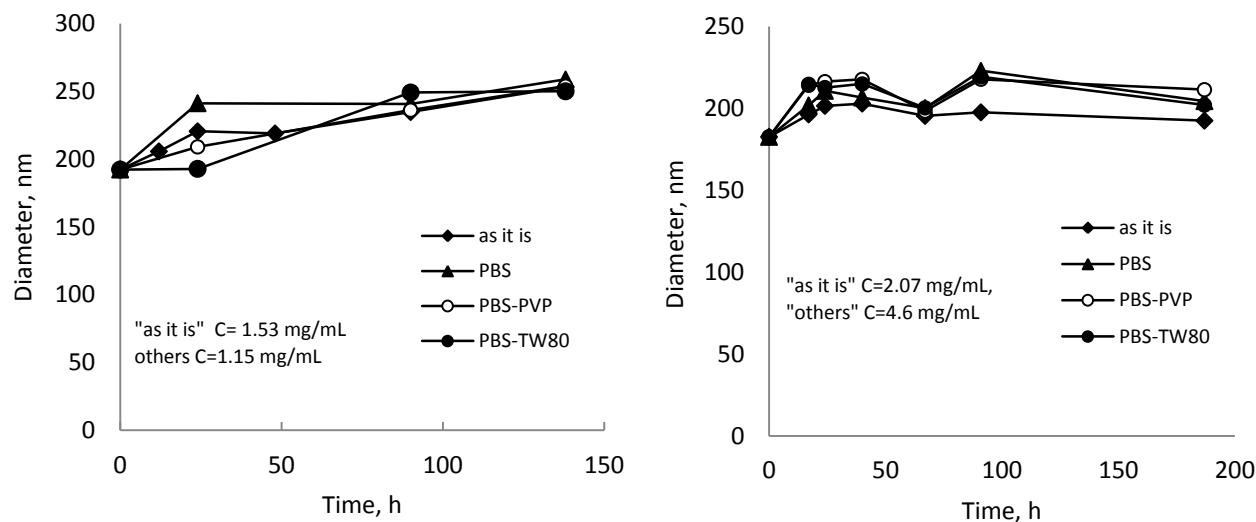


Fig.H Degradation of the lactone (353 nm) form of CPT in PBS buffer and a 40 mg/mL BSA solution. a) free CPT added in DMSO, b) 180 nm nanoparticles coated with a Hep/(PLB16-5/Hep)<sub>2</sub>/(PLB16-20/PGB30-20)<sub>2.5</sub> shell. 23 °C, pH 7.4, C(CPT)=3.5 µg/mL



5 Fig. I Apparent diameter of selected samples of PTX nanoparticles coated with a  $(PLB16-5/Hep)_{3.5}$  shell as a function of time. Samples were dispersed in: supernatant ("as it is"), PBS, PBS-0.7 mg/mL PVP, PBS+ 0.7 mg/mL Polysorbate 80. Concentrations as shown on the graphs

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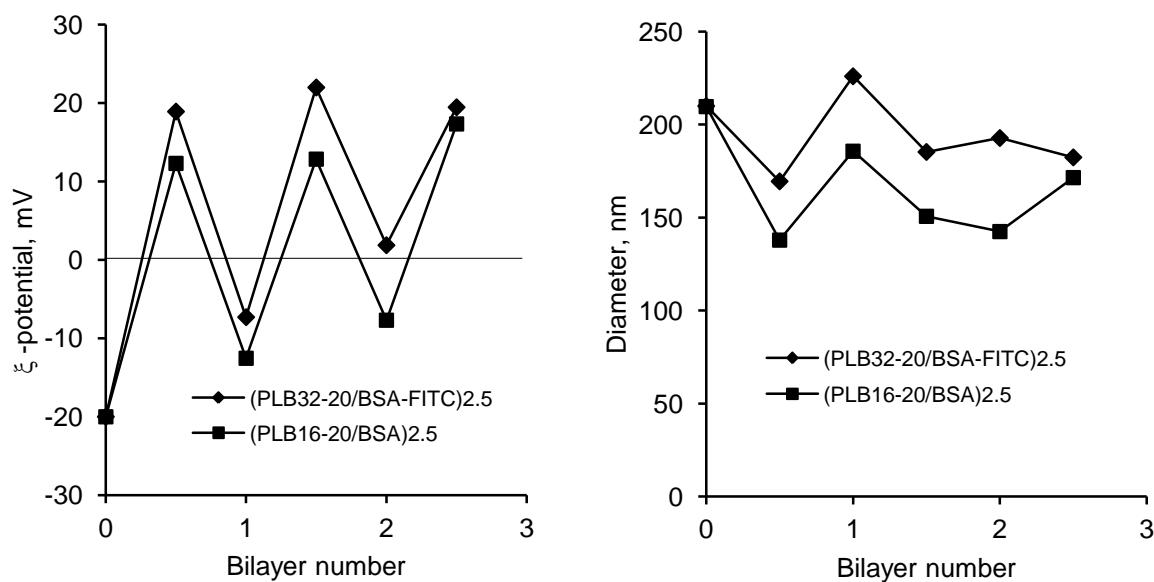


Fig.J Changes of  $\zeta$ - potential (a) and hydrodynamic diameter (b) in the process of LbL assembly using BSA. Shell architecture is shown on the graphs

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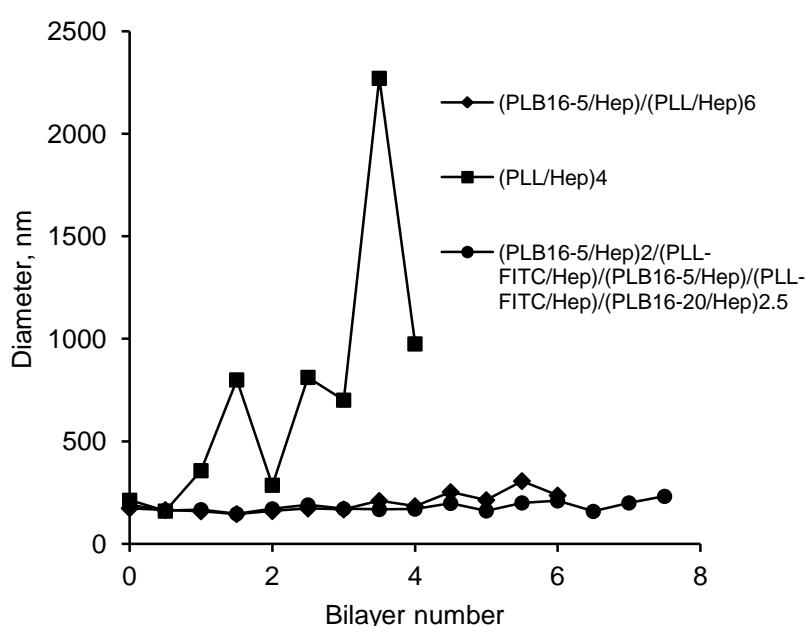


Fig. K Changes of hydrodynamic diameter in the process of LbL assembly of shells with different architecture using non-PEGylated and PEGylated polylysines.

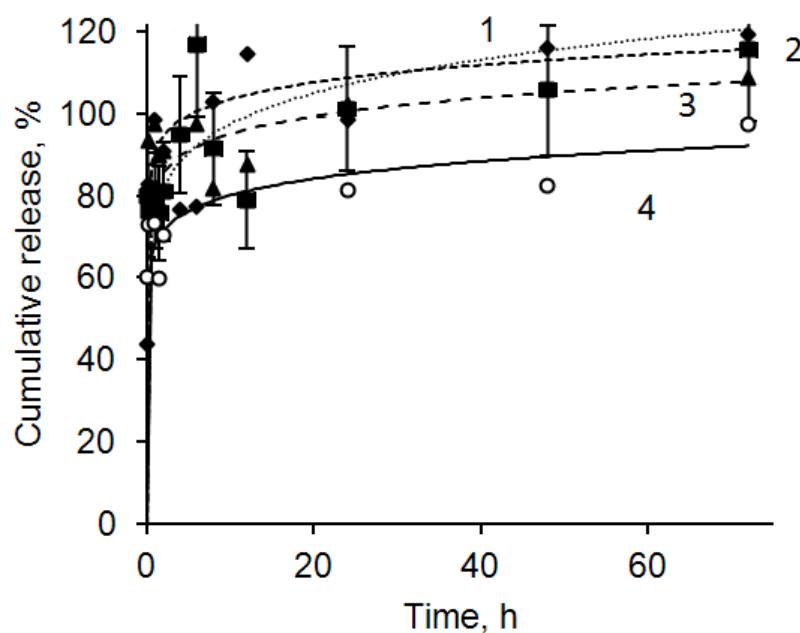


Fig. L Paclitaxel release from 170 nm nanocolloids coated with a  $(PLB16-5/Heparin)_n$  shell.

Number of bilayer in shell n: 1- 0.5, 2-1.5, 3-2.5, 4-3.5. C(PTX)= $2.0\pm0.4$   $\mu$ g/mL. 0.2% Polysorbate80 in PBS at  $37^{\circ}\text{C}$ .

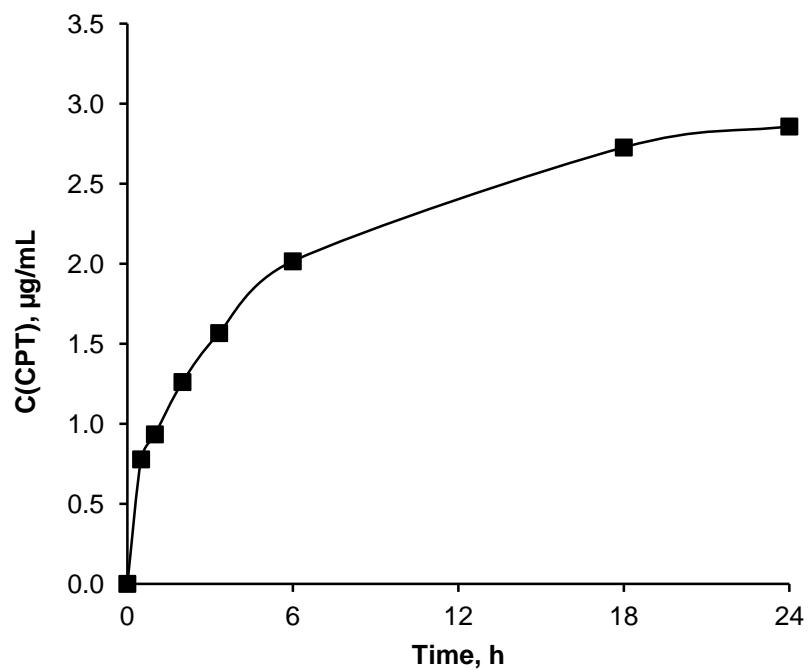


Fig. M Release of camptothecin from 160 nm nanocapsules with a (Hep/PLB16-5)<sub>4,5</sub> shell architecture.

PBS + 2% Polysorbate 80. C (CPT) = 9.3 µg/mL