Supplementary Material (ESI) for *Soft Matter*

Supplementary Material for "**Closed Nanoconstructs Assembled by step-by-step ss-DNA Coupling Assisted by Phospholipid Membranes**" by F. Baldelli Bombelli et al.

Synthesis of oligonucleotides. The list of all oligonucleotides used in this study is reported in Table 1: 5'. Cholesterol-tetraethylenglycol-modified oligonucleotide (Scheme 1) was synthesized "trityl-on" and unmodified oligonucleotides were synthesized "trityl-off". Standard DNA phosphoramidites, solid supports and additional reagents were purchased from Link Technologies and Applied Biosystems Ltd. The cholesterol-tetraethylenglycol phosphoramidite monomer was purchased from Glen Research Inc. An Applied Biosystems 394 automated DNA/RNA synthesizer was used and multiple 1.0μmole phosphoramidite cycles of acid-catalyzed detritylation, coupling, capping and iodine oxidation were employed to produce the required oligonucleotides on large scale. Convential monomers (A, G, C and T) were allowed to couple for 25 seconds. Stepwise coupling efficiencies and overall yields of monomers were determined by measuring trityl cation conductivity and in all cases these were >98.0%. Cleavage of the oligonucleotides from the solid support was carried out in concentrated aqueous ammonia at 55°C for 6 hours. After ammonia deprotection the cholesterol-tetraethilenglycol oligonucleotides were evaporated to dryness then treated with acetic acid/water (80/20) for 30 min at room temperature to remove the DMT-group from the cholesteryl moiety. After evaporation of the solvent the oligonucleotides were extracted with diethyl ether and then purified by reversed-phase HPLC. All other oligonucleotides were purified by reversed-phase HPLC directly after ammonia deprotection and evaporation of aqueous ammonia.

Purification of oligonucleotides. Purification of oligonucleotides was carried out by reversed phase HPLC on a Gilson system using an ABI Aquapore column (C8), 8mm x 250mm, pore size 300Å controlled by Gilson 7.12 software. The following protocol was used: run time 30 minutes, flow rate 3ml per minute, binary system gradient (time in minutes (% buffer B);0 (0); 3(0); 5(20); 21 (100); 25(100); 27 (0); 30(0)). Elution buffer A: 100mM ammonium acetate, pH 7.0, buffer B: 100mM ammonium acetate with 70% acetonitrile pH 7.0 (cholesterol-tetraethilenglycol oligomers). Elution was monitored by ultraviolet absorption at 295nm. After HPLC purification oligonucleotides were desalted

with disposable NAP 10 Sephadex columns (Pharmacia) using the manufacturer's instructions, aliquoted into Eppendorf tubes and stored at –20ºC in distilled deionised water.

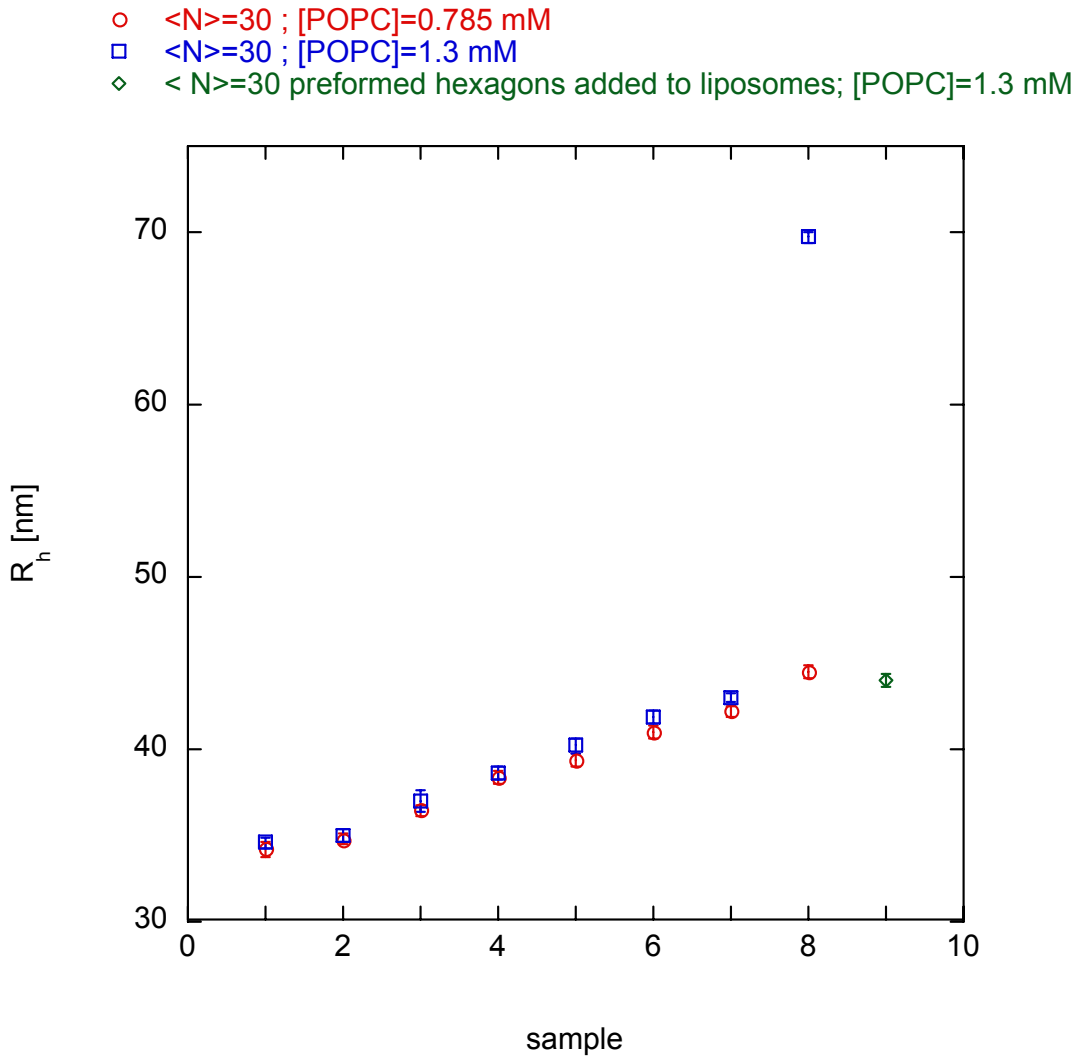
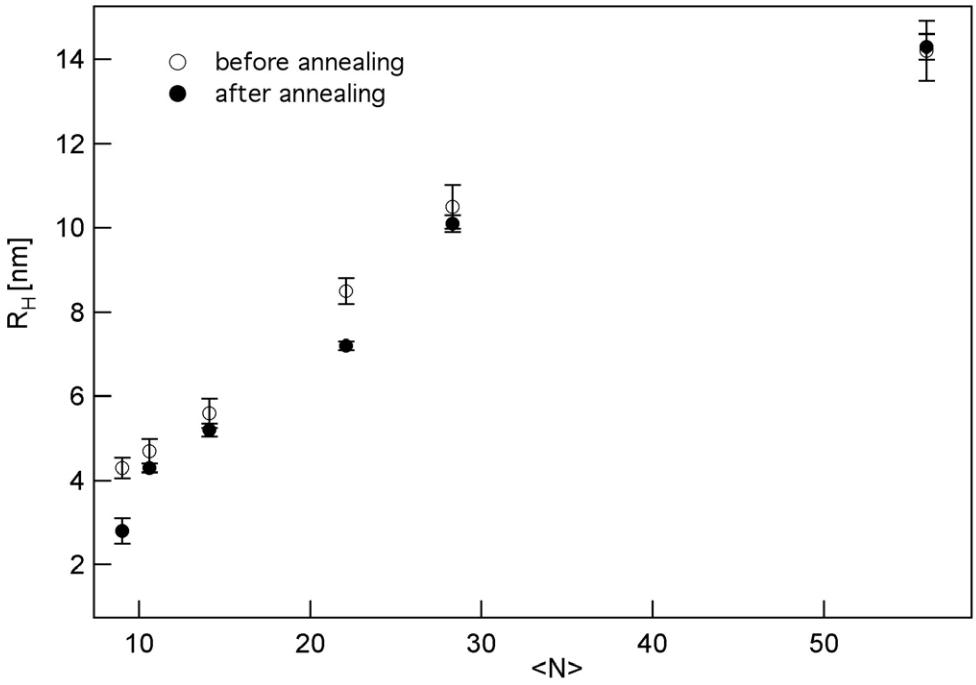


Figure 1_supplementary. Hydrodynamic radii of the lipid/DNA hybrids throughout the symmetric step-by-step build-up of vesicles decorated by DNA pseudohexagons for [POPC]=1.3mM and [POPC]=0.785 mM and <N>=30. The final results obtained for addition of preformed hexagons to liposomes is also reported for comaprison.



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