

Supporting information for

Hydrogen Bonded Capsules by Layer-by-Layer assembly of Tannic Acid and Poly(2-n-propyl-2-oxazoline) for Encapsulation and Release of Macromolecules

*Gokul Paramasivam*¹, *Maarten Vergaelen*², *Munuswamy-Ramanujam Ganesh*³, *Richard Hoogenboom*^{2*} and *Anandhakumar Sundaramurthy*^{1,4*}

¹SRM Research Institute, SRM University, Kattankulathur, Kanchipuram 603 203, Tamil Nadu, India

²Supramolecular Chemistry Group, Department of Organic and Macromolecular Chemistry, Ghent University, Krijgslaan 281 S4, B-9000, Ghent, Belgium *Email: richard.hoogenboom@ugent.be

³Interdisciplinary Institute of Indian System of Medicine, SRM University, Kattankulathur, Kanchipuram 603203, Tamil Nadu, India

⁴Department of Physics and Nanotechnology, SRM University, Kattankulathur, Kanchipuram 603203, Tamil Nadu, India

Table S1. Number average molar mass (Mn), dispersity (Đ) and degree of polymerization (DP) of the synthesized polymers.

| Polymer | Mn, kg/mol ^(a) | Đ ^(a) | DP ^(b) |
|----------|---------------------------|------------------|-------------------|
| PnPropOx | 16.2 | 1.23 | 100 |

a) Determined by size exclusion chromatography with N,N-dimethylacetamide containing 50 mM LiCl as eluent and calculated against poly(methyl methacrylate) standards. b) Theoretical DP based on the

actually weighed reagents for the polymerization leading to a monomer to initiator ratio of 100, which in combination with full monomer conversion leads to a theoretical DP of 100 which has been confirmed (within experimental error of 5%) by ^1H NMR spectroscopy.

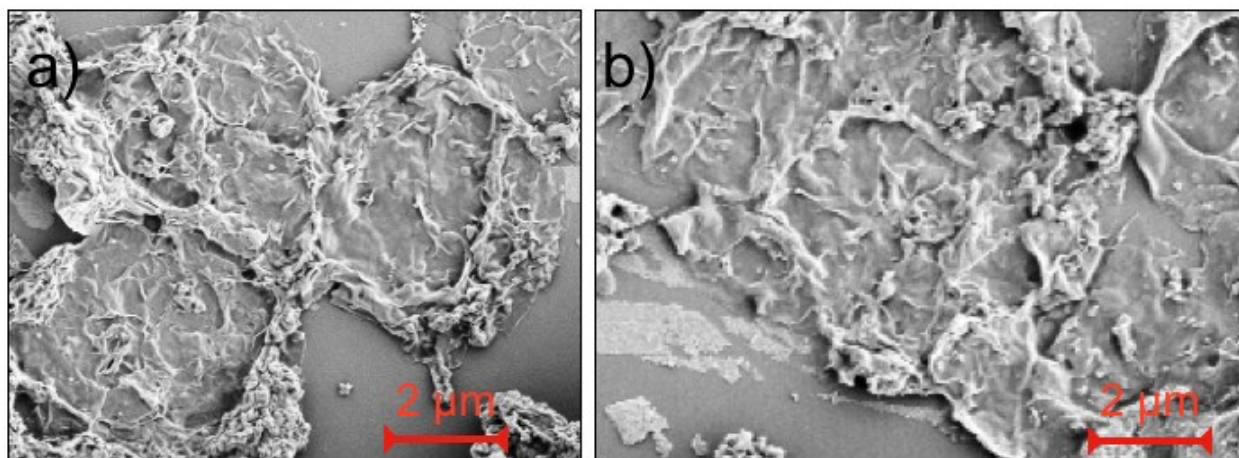


Fig. S1. SEM investigation to show the morphology of $(\text{TA}/\text{PnPropOx})_2$ capsules. Collapse and aggregation of capsules occurs due to drying. Direct exposure of electron beam on capsule surface results in capsules deformation as shown in Fig. S1b. Hence focus was done on adjacent features and then moved to capsules for acquiring the images.

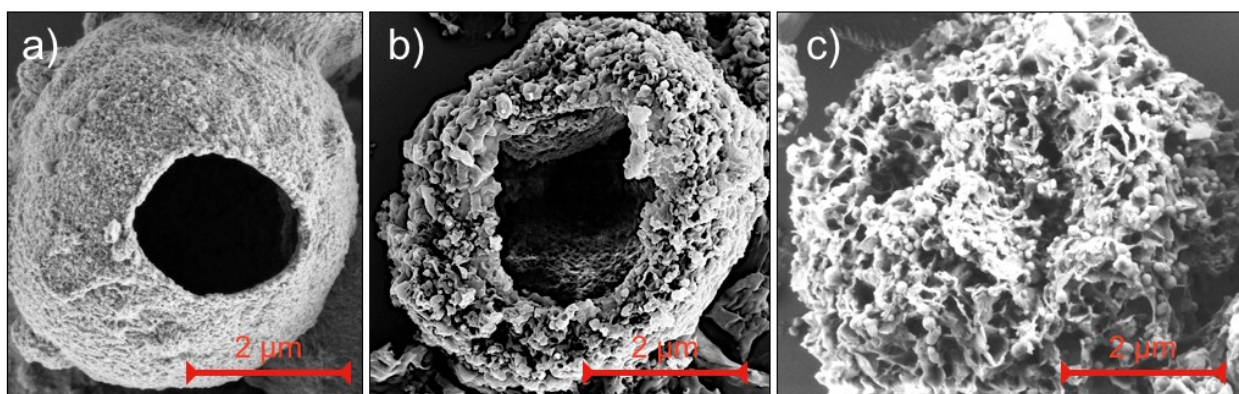


Fig. S2. SEM investigation to show the morphology of $(\text{TA}/\text{PnPropOx})_4$ capsules after heat treatment at 37°C . Control capsules (a), heat treated capsules for 15 min (b) and 1 h (c).

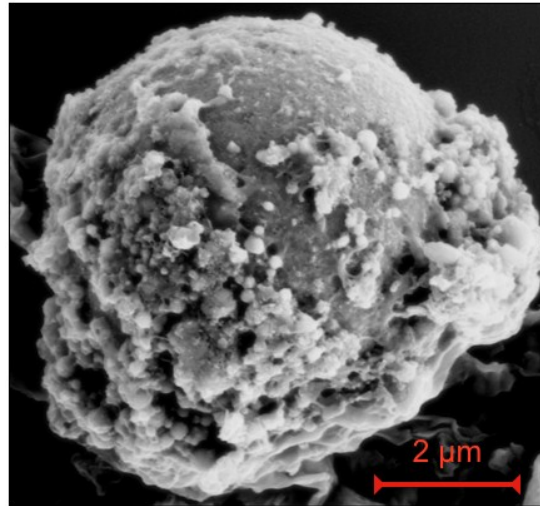


Fig. S3. SEM investigation to show the morphology of (TA/PnPropOx)₄ capsules. As its wall thickness is high, it maintains its spherical shape even after drying.

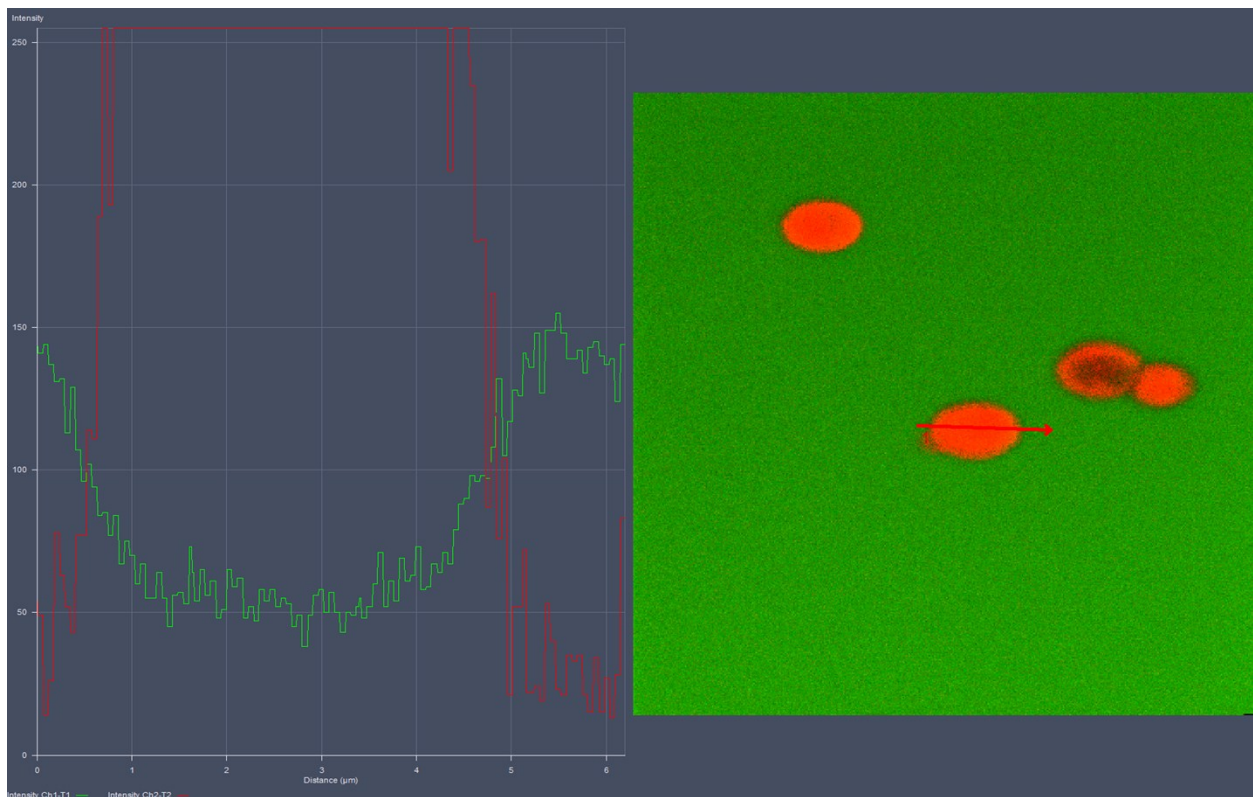


Fig. S4. CLSM investigation of heat treated capsules (at 37 °C for 15 min) with FITC-Dextran and TRITC-Dextran. Investigation shows that the capsules were permeable only to TRITC-Dextran.

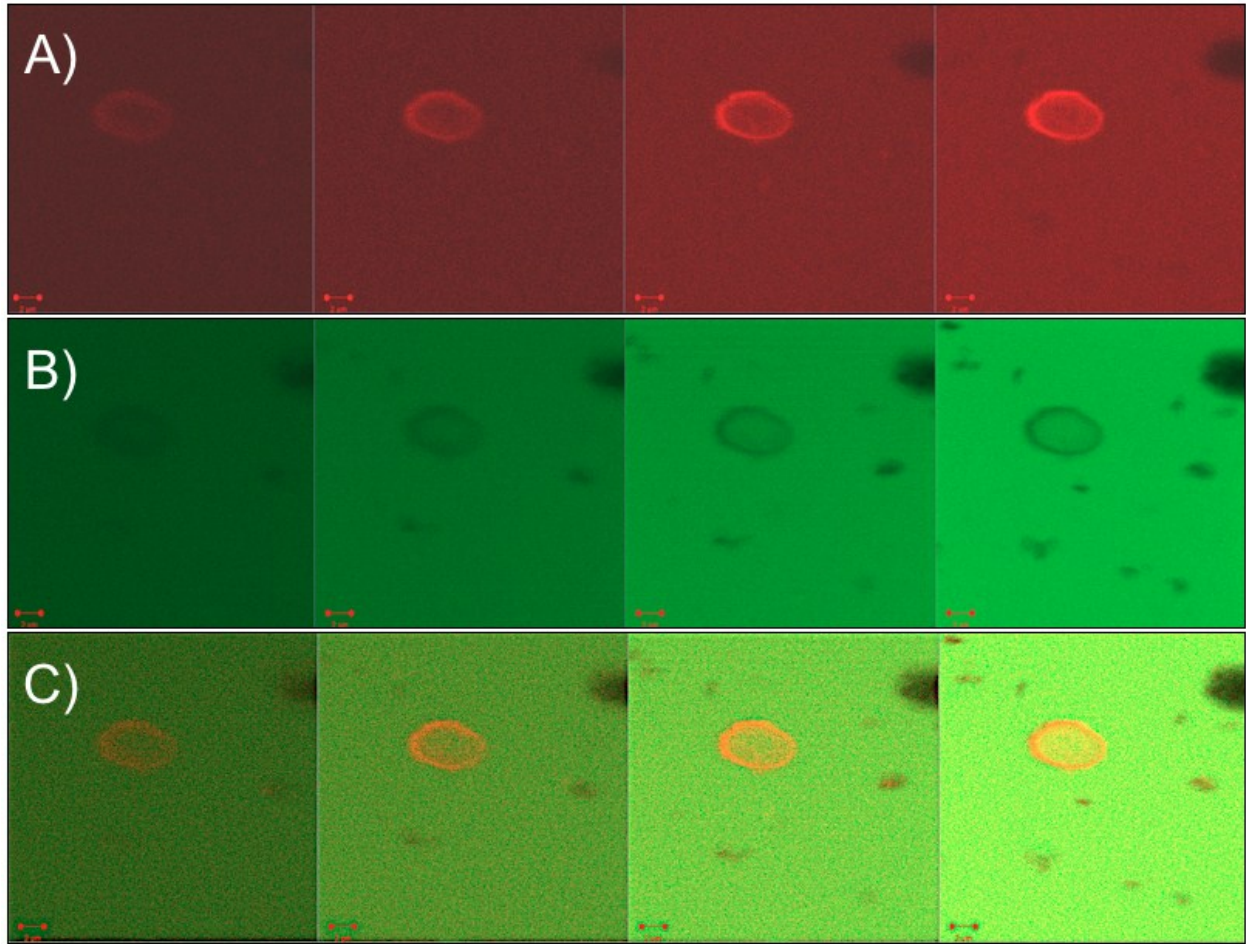


Fig. S5. CLSM investigation of heat treated capsules (at 37 °C for 1 h) with FITC-Dextran and TRITC-Dextran. Investigation shows that the capsules were permeable to both FITC-Dextran and TRITC-Dextran. Images were acquired at different planes (z-stack) for better understanding of permeability of the capsules. Red channel a), green channel b), and overlapped image c).

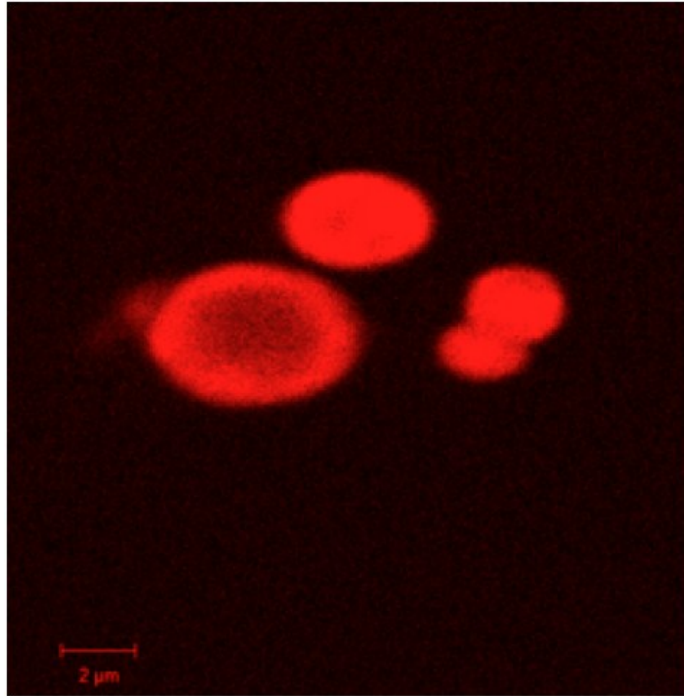


Fig. S6. CLSM image shows TRITC-Dextran loaded capsules (with PSS).

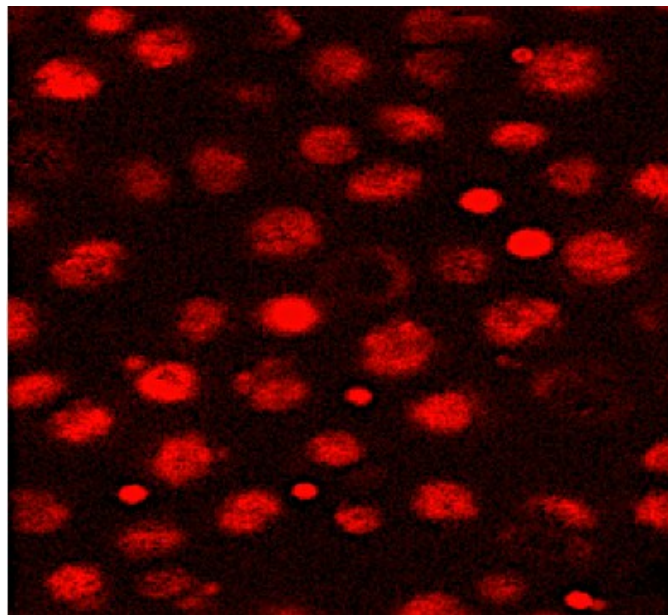


Fig. S7. CLSM image shows TRITC-Dextran loaded capsules (without PSS).