SUPPLEMENTARY INFORMATION

Synthesis of Poly(3-sulfopropyl acrylate) (PSPA) homopolymers.

3-sulfopropyl acrylate, potassium salt 97.7% (SPA) and 4,4'-Azobis(4-cyanopentanoic acid) (V-501) were purchased from Sigma-Aldrich. 4-cyanopentanoic acid dithiobenzoate (CPADB) was synthesised by following a protocol in the literature. The polymerization of SPA was achieved by first adding 4 g (0.017 mol) of SPA monomer, 52.9 mg (1.9×10⁻⁴ mol) CPADB and 5.6 mg (2×10⁻⁵ mol) V-501 initiator to a glass vial. These were dissolved in 9 g milliQ water with 2 g ethanol to assist solubilisation, yielding a pink, transparent solution. Approximately 1.5 g of this mixture was transferred to a glass test tube, which was flushed with N₂ gas for 15 minutes over ice. Polymerisation was performed, with the use of an oil bath maintained at 70 °C. The polymerisation was halted at 150 mins by exposing the system briefly to air, before re-sealing and then placing on ice. A small sample of the mixture was dissolved in D₂O, and characterised by proton NMR (Bruker Avance III 300 MHz spectrometer) to calculate the % conversion which was found to be 48%. The polymer was purified by dialysis in snakeskin dialysis tubing (3500 MWCO, Thermo Scientific) against milliQ water, and then recovered by lyophilisation. The dry polymer was dissolved in D₂O and evaluated by proton NMR in order to calculate the M_n , which was found to be 10800 g/mol. This was found to be in agreement with the theoretical M_n of 10200 g/mol for PSPA with 48 % conversion. The proton NMR spectrum of PSPA and the peak assignments are shown in Figure S1.

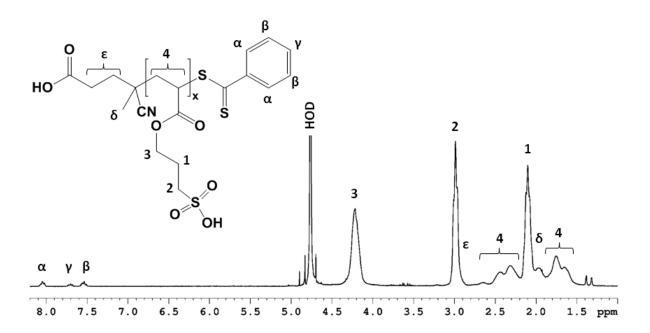


Figure S1: ¹H- NMR spectrum of PSPA, after dialysis, in D₂O.

Further results of crystal growth

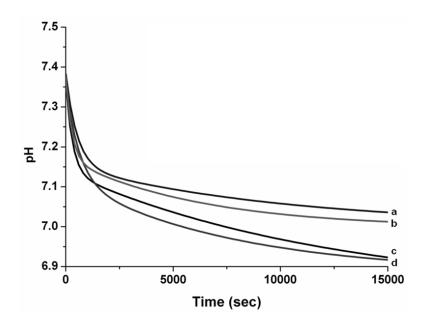


Figure S2: Change in pH after the addition of dry seed crystal to working solution with 10 mg alginate dissolved (a), addition of slurry with 1 mg alginate (b), addition of dry seed crystal to working solution with 1 mg alginate dissolved (c), addition of dry seed crystals to solution containing no additives (d).

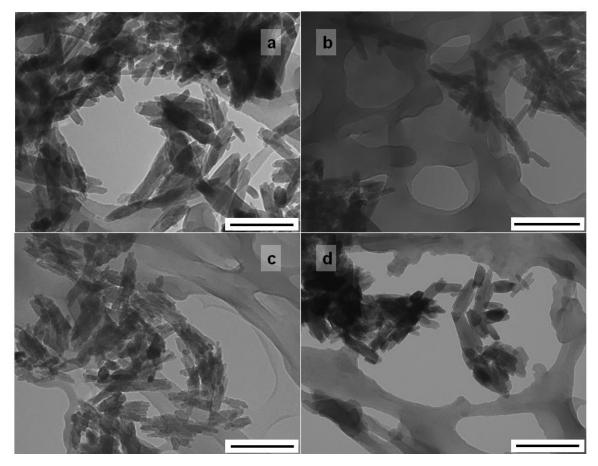


Figure S3: TEM images of (a) HAlg; (b) HPAlg; (c) HHep; (d) HPSPA. Scale bar denotes 200 nm.

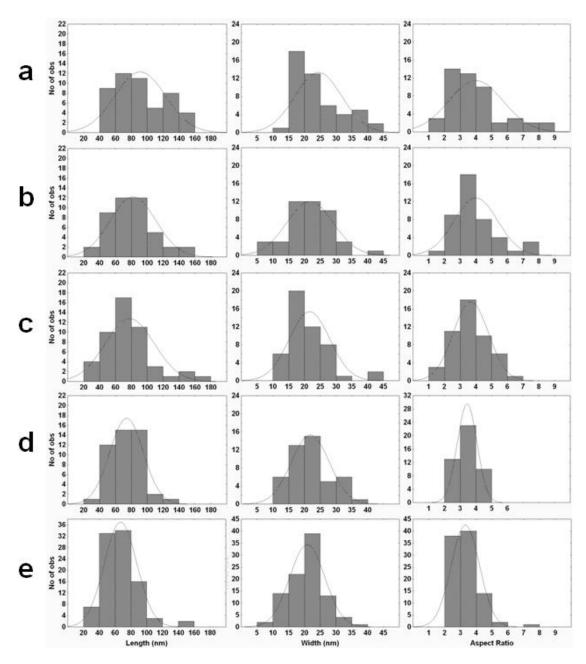


Figure S4: Histograms of the distributions of Length, Width and Aspect Ratio for (a) sHAP, (b) HPAlg, (c) HAlg, (d) HHep, (e) HPSPA crystals.

References.

1. Thang, S. H.; Chong, Y. K.; Mayadunne, R. T. A.; Moad, G.; Rizzardo, E., A Novel Synthesis of Functional Dithioesters, Dithiocarbamates, Xanthates and Trithiocarbonates. *Tetrahedron Letters* **1999**, *40*, 2435-2438.