Supplementary Information

**Figure S1** – (a)–(d) Hydrogelation of samples 1-4 respectively tested by vial inversion after 24 hours (samples include subtilisin and heat to dissolve as per experimental for subtilisin assisted hydrolysis of methyl ester protected starting materials).

**Figure S2** – (a)–(d) starting mixtures of 1-4 respectively in buffer. Starting materials precipitate from the aqueous environment without the addition of the subtilisin.

**Figure S3** – (a)-(d) sample preparation of of 1-4 respectively without the addition of subtilisin. The starting materials do not dissolve when heated to 55 °C in the aqueous solvent without the addition of subtilisin.
Figure S4 – (a)-(d) Fluorescence emission spectra of 1-4 respectively measure at 24 hours after enzyme addition overlayed with starting materials in buffer and sample preparation with no enzyme.

Figure S5 - CD spectra of 20 µL of subtilisin in sodium phosphate buffer, pH8, measured 24 hours after heat/cool cycle.
**Figure S6** – Further TEM images of self-assembled structures of (a)-(b) 1, (c)-(d) 2, (d)-(e) 3 and (e)-(f) 4, taken after 24 hours.

**Figure S7** - cryoTEM images of self-assembled structures 1 (a), 2 (b), 3 (c) and 4 (d), taken after 24 hours. Arrows indicate the formation of spherical aggregates.
Figure S8 – Rheological strain sweeps of (a) 1, (b) 2 and (c) 3 measured at 24 hours after treatment with subtilisin to ensure measurements are within the linear Viscoelastic Regime.
Figure S9 - (a) Peak Ratios of J-aggregate:Fmoc monomer (450 nm/320 nm) calculated from fluorescence emission spectra data monitored over time with no heat/cool cycles. (b)-(e) Fluorescence emission spectra of 1 – 4 respectively obtained over time.

Figure S10 - (a) CD spectra of the thermodynamically preferred states of 1, 2, 3 and 4. (b)-(e) CD spectra of initial formation against thermodynamic state of 1, 2, 3 and 4 respectively.
Figure S11 - Amide I region of FTIR absorbance spectra of 1, 2, 3, and 4 of thermodynamically preferred state.

Figure S12 – (a)–(d) visual images of samples 1-4 respectively in their thermodynamically preferred state, tested by vial inversion.
Figure S13 - TEM images of self-assembled structures 1 (a), 2 (b), 3 (c) and 4 (d), taken of thermodynamically preferred state.

Figure S14 - cryoTEM images of self-assembled structures 1 (a), 2 (b), 3 (c) and 4 (d), taken of thermodynamically preferred state.
Figure S15 - Rheological strain sweeps of (a) 1, (b) 2 and (c) 3 measured of thermodynamically preferred state to ensure measurements are within the Linear Viscoelastic Regime.

Figure S16 - Rheological data of materials in thermodynamically preferred state. (a) Viscosity and flow curves of 1. (b) Frequency sweep of 2. (c) Frequency sweep of 3. (d) Viscosity and flow curves of 4.