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Supplementary Information

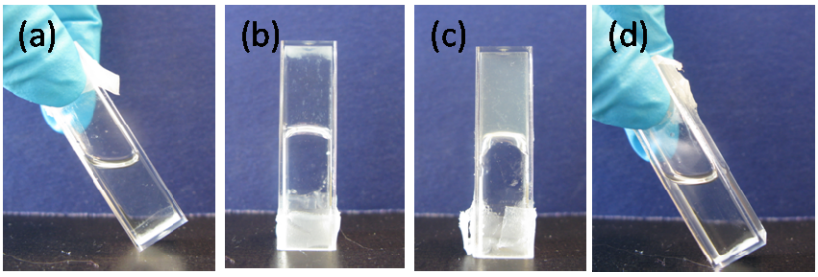

Figure S1 – (a)–(d) Hydrogelation of samples **1**-**4** respectively tested by vial inversion after 24 hours (samples include subtilisin and heat to dissolve as per experimental for subtilisin assisted hydrolysis of methyl ester protected starting materials).

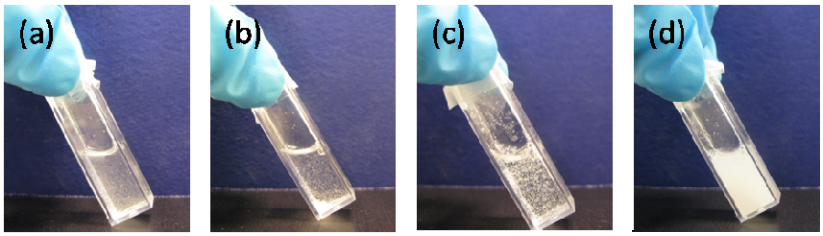

Figure S2 – (a)–(d) starting mixtures of **1**-**4** respectively in buffer. Starting materials precipitate from the aqueous environment without the addition of the subtilisin.

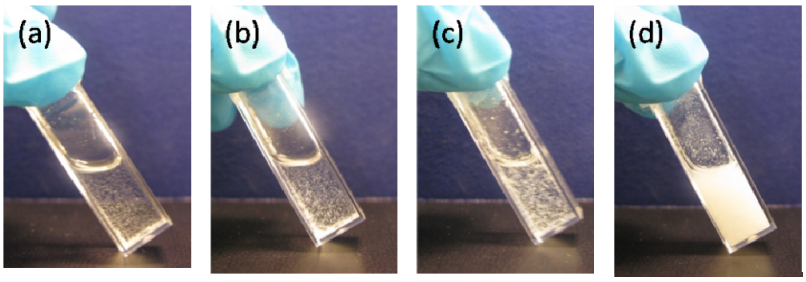

Figure S3 – (a)-(d) sample preparation of of **1**-**4** respectively without the addition of subtillisin. The starting materials do not dissolve when heated to 55 °C in the aqueous solvent without the addition of subtilisin.

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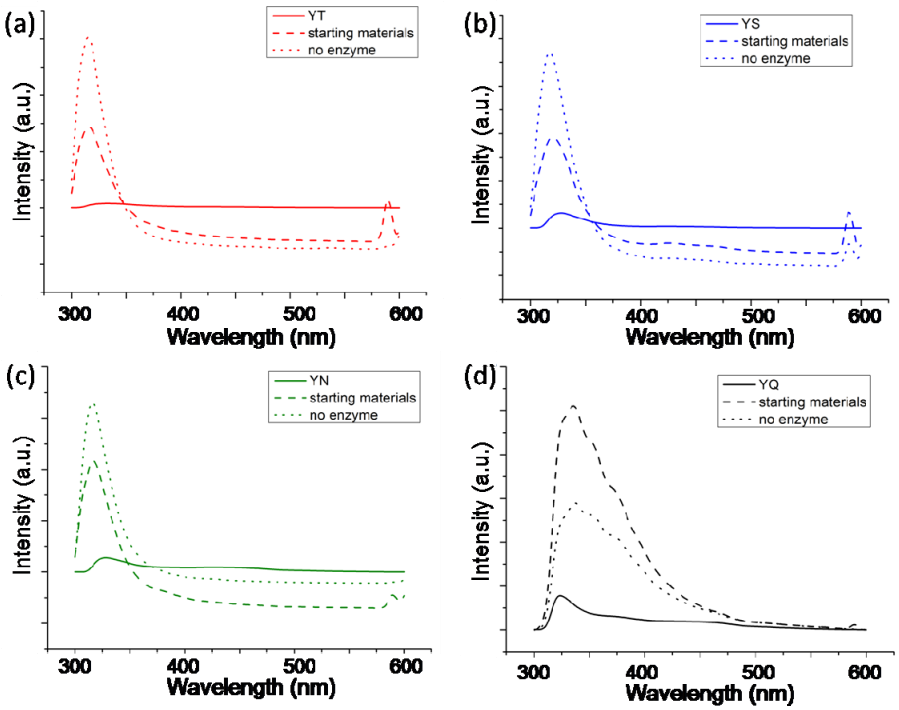

Figure S4 – (a)-(d) Fluorescence emission spectra of **1**-**4** respectively measure at 24 hours after enzyme addition overlayed with starting materials in buffer and sample preparation with no enzyme.

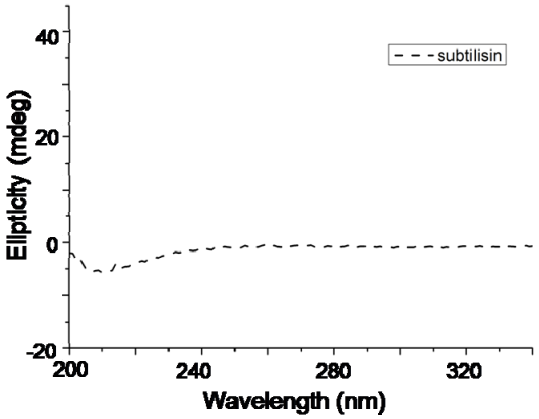

Figure S5 - CD spectra of 20 µL of subtilisin in sodium phosphate buffer, pH8, measured 24 hours after heat/cool cycle.

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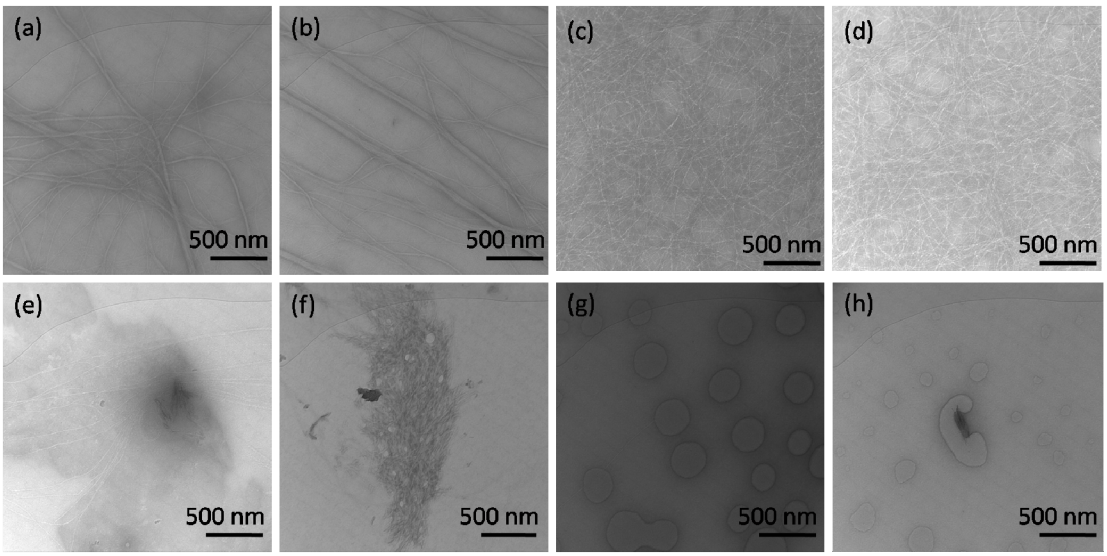

Figure S6 – Further TEM images of self-assembled structures of (a)-(b) **1**, (c)-(d) **2**, (d)-(e) **3** and (e)-(f) **4**, taken after 24 hours.

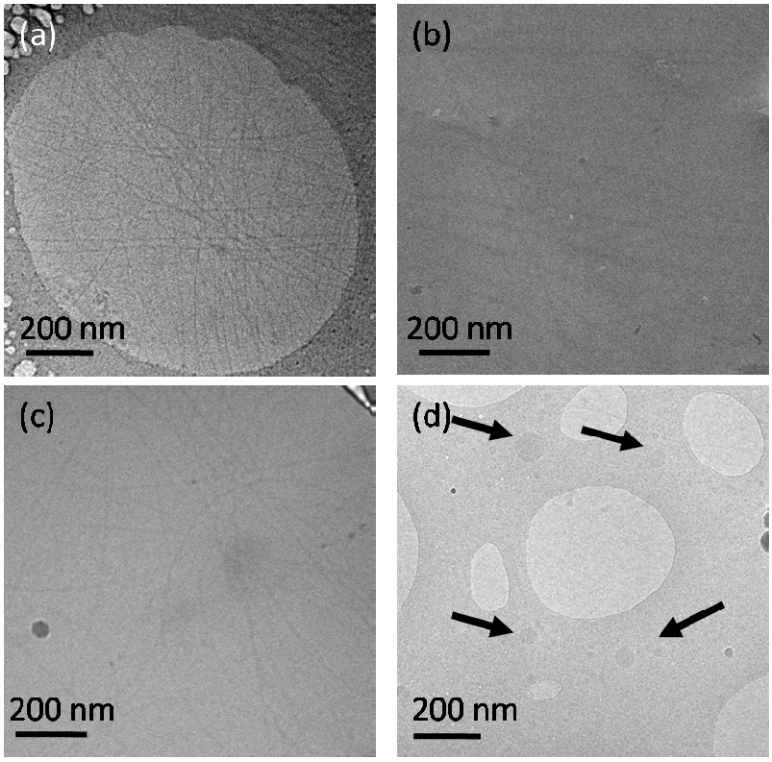

Figure S7 - cryoTEM images of self-assembled structures **1** (a), **2** (b), **3** (c) and **4** (d), taken after 24 hours. Arrows indicate the formation of spherical aggregates.

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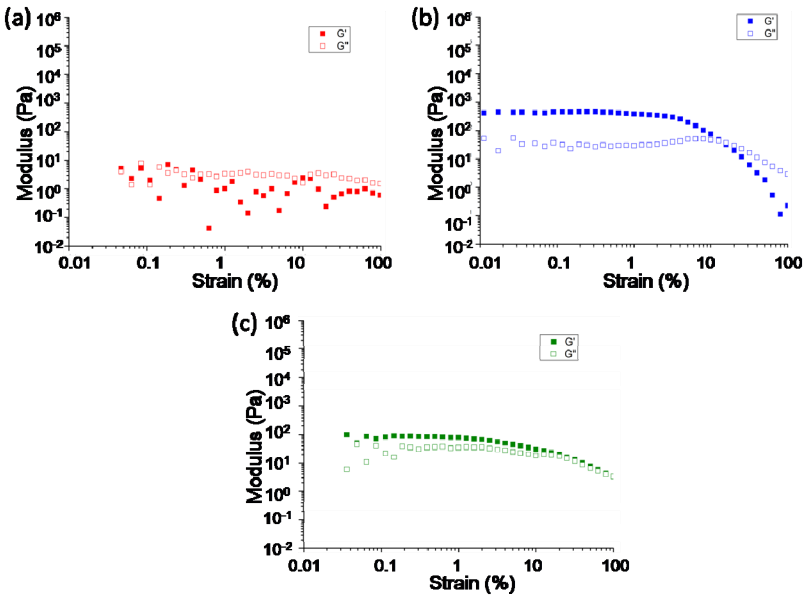

Figure S8 – Rheological strain sweeps of (a) **1**, (b) **2** and (c) **3** measured at 24 hours after treatment with subtillisin to ensure measurements are within the linear Viscoelastic Regime.

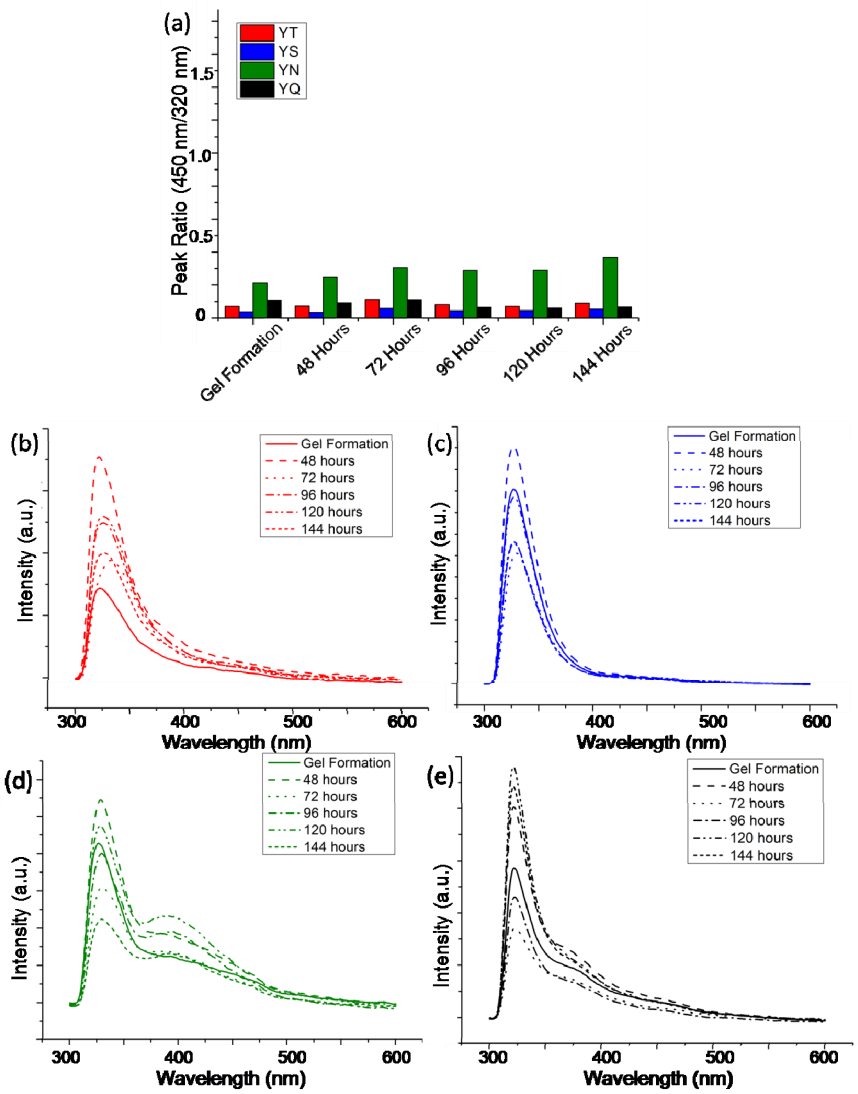

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Figure S9 - (a) Peak Ratios of J-aggregate:Fmoc monomer (450 nm/320 nm) calculated from fluorescence emission spectra data monitored over time with no heat/cool cycles. (b)-(e) Flourescence emission spectra of **1** – **4** respectively obtained over time.

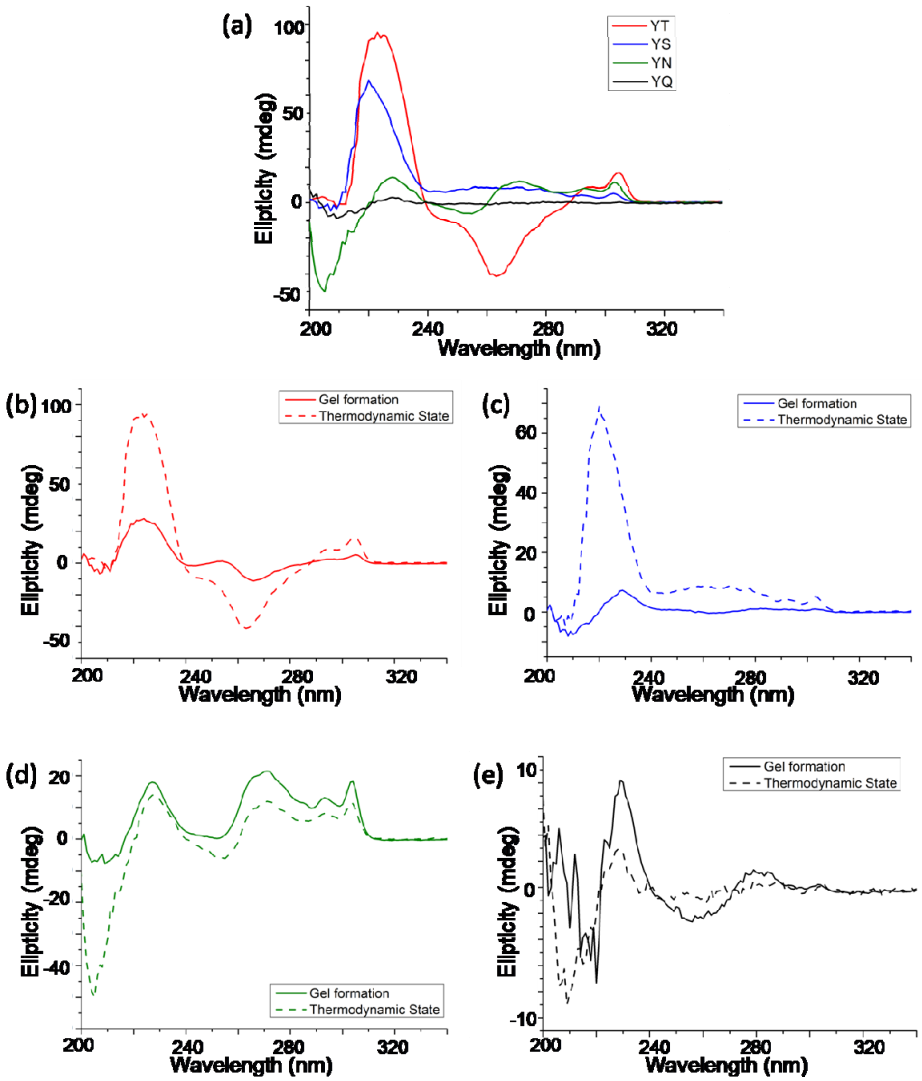

Figure S10 - (a) CD spectra of the thermodynamically preferred states of **1**, **2**, **3** and **4**. (b)-(e) CD spectra of initial formation against thermodynamic state of **1**, **2**, **3** and **4** respectively.

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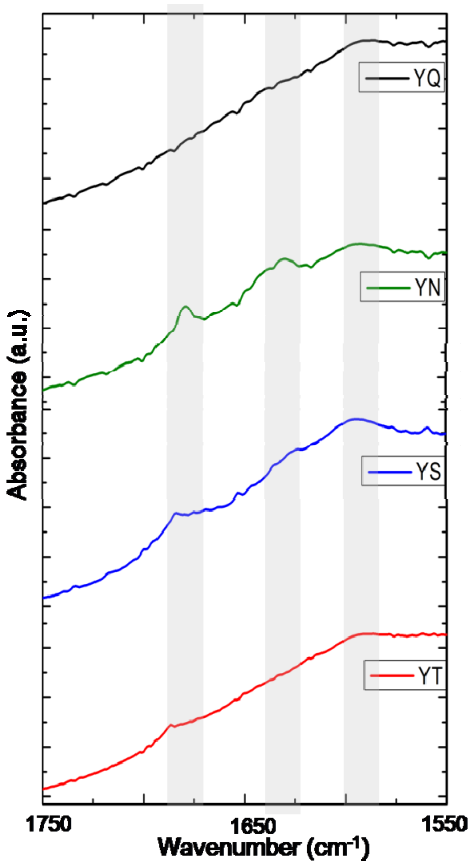

Figure S11- Amide I region of FTIR absorbance spectra of **1**, **2**, **3**, and **4** of thermodynamically preferred state.

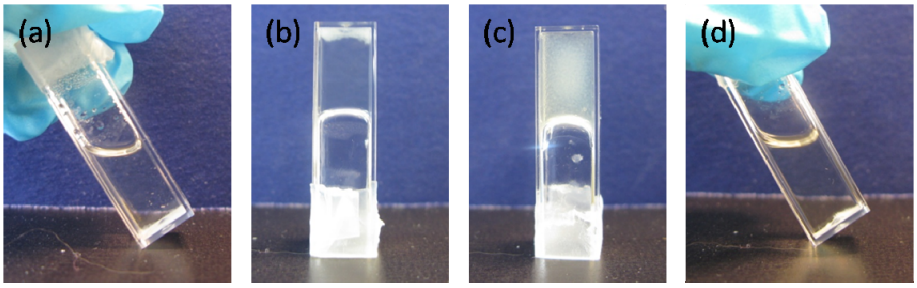

Figure S12 – (a)–(d) visual images of samples **1**-**4** respectively in their thermodynamically preferred state, tested by vial inversion.

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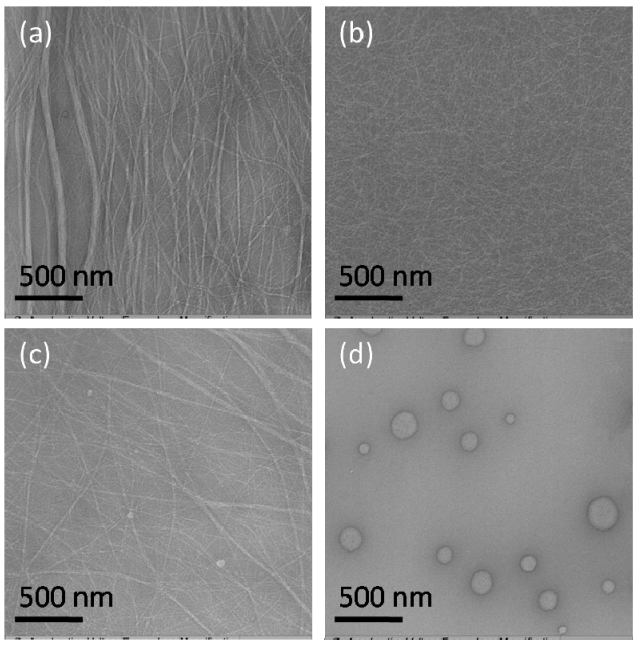

Figure S13 - TEM images of self-assembled structures **1** (a), **2** (b), **3** (c) and **4** (d), taken of thermodynamically preferred state.

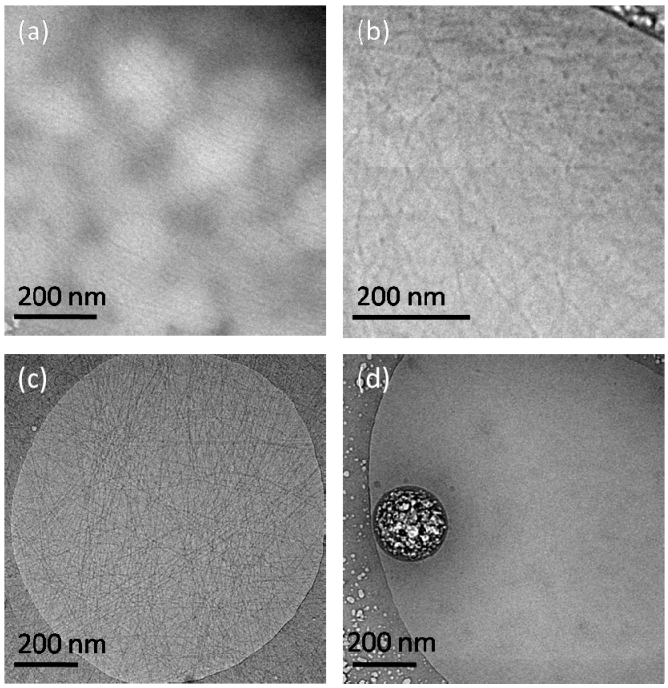

Figure S14 - cryoTEM images of self-assembled structures **1** (a), **2** (b), **3** (c) and **4** (d), taken of thermodynamically preferred state.

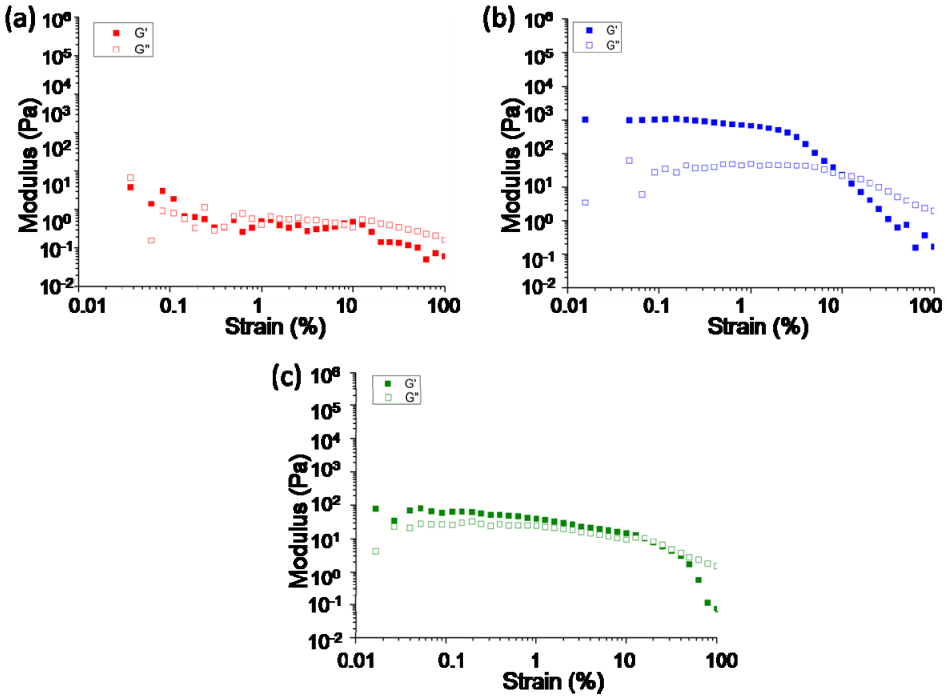

Figure S15 - Rheological strain sweeps of (a) **1**, (b) **2** and (c) **3** measured of thermodynamically preferred state to ensure measurements are within the Linear Viscoelastic Regime.

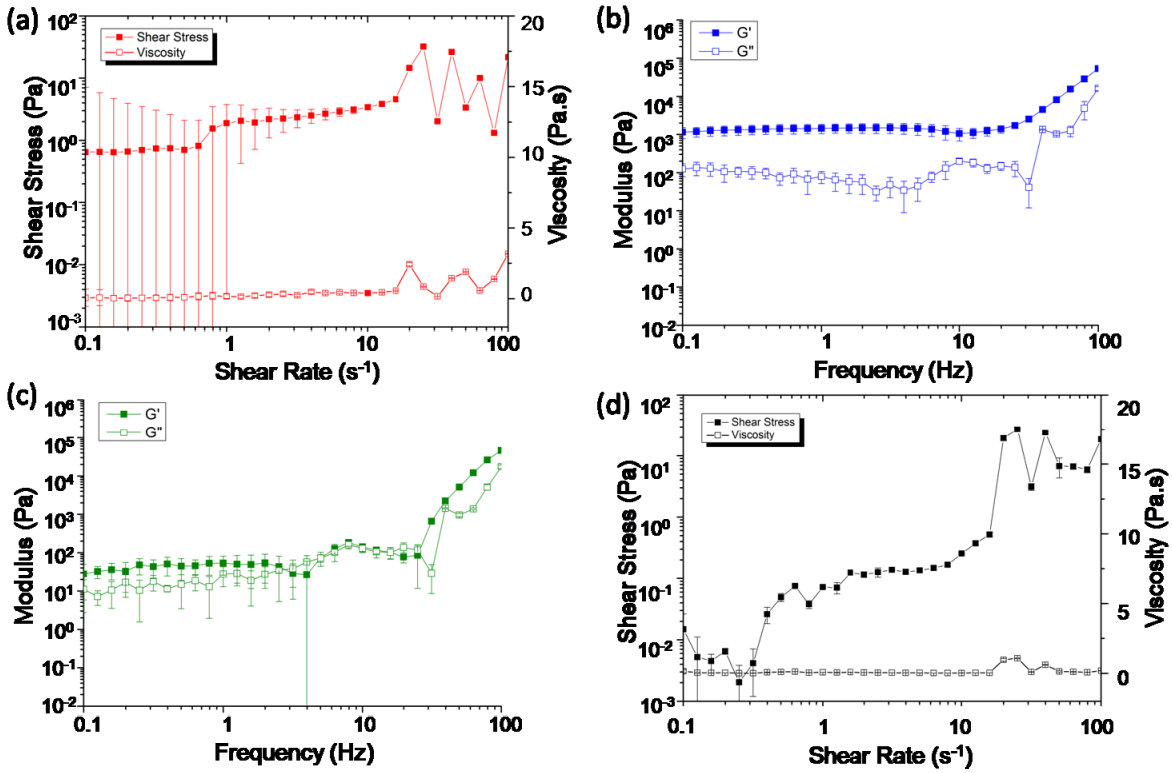

Figure S16 - Rheological data of materials in thermodynamically preferred state. (a) Viscosity and flow curves of **1**. (b) Frequency sweep of **2**. (c) Frequency sweep of **3**. (d) Viscosity and flow curves of **4**.