## Supporting Information

## Single Molecule Protein Stabilisation Translates to Macromolecular Mechanics of Protein Network

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Figure S1: The value of the power law exponent, extracted from the frequency dependence of the storage and loss moduli in Fig. 2a, as a function of maltose concentration.

Langmuir type equation S1:

$$X_{[Maltose]} = \frac{\Delta X \cdot [Maltose]}{K_d + [Maltose]} + X_0$$
(S1)

Where X in this study is substituted for either melting temperature, shear moduli or energy dissipated.



Figure S2: Strain amplitude ramps showing the (a) storage and (b) loss moduli of chemically crosslinked MBP hydrogels as a function of the maltose concentration. The large drop occurring at approx.  $300 \pm 60\%$  in both moduli is attributed to rupture of the gel and shows no statistically significant trend with maltose concentration. (c) Ratio of loss to storage modulus as a function of strain and maltose concentration, an upturn is noted at strains of approx. 300% after which tan( $\delta$ ) becomes larger than one, meaning the viscous forces now dominate the system, demonstrating

rupture of the gel network.



Figure S3: Stress-Strain curves of chemically crosslinked MBP hydrogels as a function of maltose concentration. Samples were strained to (a) 10% and (b) 20% at a rate of 1%/s and then unloaded down to 0% at the same rate. Similar trends are seen in these curves as shown in the main text

(Fig. 3a).



Figure S4: Exemplar stress-strain curves of chemically crosslinked MBP hydrogels. Red lines show linear fit below 15% strain, the gradient of which is interpreted as the storage modulus of the hydrogel in the linear regime.



Figure S5: Residual strain at OPa extracted from strain-strain curves upon unloading of MBP hydrogels strained to 50% as a function of maltose concentration.



Figure S6: Exemplar stress-strain load curves of MBP based hydrogels as a function of maximum strain. All strain loading was performed at a rate of 1%/s up to the maximum strain before unloading and restraining. Note that all curves overlay i.e. follow the same strain path, demonstrating that at strains up to 75% the gel is not damaged or permanently deformed.



Figure S7: (a) The effiency of MBP based hydrogels extracted from stress-strain curves as a function of both maltose concentration and maximum applied strain. Efficency is defined as

 $Efficency = 1 - \frac{Energy \ dissipated}{Strain_{Max} \cdot Stress_{Max}}$  and is a measure of the proportion of energy stored that is released and not lost to the internal energy of the system upon unloading of the system. Samples were strained loaded and unloaded at a rate of 1%/s. (b) Energy dissipated of MBP hydrogels

extracted from stress-strain curves as a function of maximum applied strain in the presence and

absence of 10mM maltose.



Figure S8: (a) SANS scattering curve of MBP in 25mM sodium phosphate buffer pH 7.4, at a concentration of 10mg/ml. (b) P(r) inversion analysis of the data in Fig. S1a, extracting an R<sub>g</sub> value of 21Å (c) Guinier plot of the data in Figure S1a, fitting this curve with a linear fit extracts a value for R<sub>g</sub> of (20±1)Å. (d) SAXS scattering curve of MBP in 25mM sodium phosphate buffer pH 7.4, at a concentration of 10mg/ml. Experiments performed in the presence and absence of maltose. (e) P(r) inversion analysis of the data in Fig. S1d, extracting a Rg value of 23Å (presence of maltose) & 24Å (absence of maltose). (f) Guinier plot of the data in Fig. S1d, fitting this curve with a linear fit extracts a value for Rg of (23±1)Å in the presence & (24±1)Å in the absence of maltose.



Figure S9: SANS curve of MBP hydrogel in the presence of 10mM maltose, taken on the instrument ZOOM. The red line showing the fit from equation (9) in Methods. Insert: Magnification of the scattering curve to clearly show the presence of Kiessig fringes and the peak

to peak analysis.



Figure S10: Gelation time,  $t_{gel}$ , of photo-chemcially crosslinked MBP hydrogels (final concentrations: 100mg/ml MBP, 30mM NaPS, 100µM Ru), illuminated at t=60s till t=360s, as a function of maltose concentration. (insert) Examplar gelation curve demonstrating the definition of  $t_{gel}$  used in this work.



Figure S11: (a) Examplar gelation curve (showing storage and loss moduli vs time) of photochemical crosslinked MBP hydrogels (final concentrations: 100mg/ml MBP, 30mM NaPS, 100μM Ru). Illuminated at t=60s till t=360s. (b) Measured force normal to the plane of shear (The positive direction is defined as upwards) during gelation of a MBP-based hydrogel.



Figure S12: Exemplar gelation curve (showing storage modulus vs time) of chemical crosslinked MBP hydrogels (final concentrations: 100mg/ml MBP, 30mM NaPS, 100µM Ru). Red line shows fit of empirical fitting function (eqn (1)) to the gelation curve data.