Supplementary Material

2 The lightly cross-linked monodisperse PNIPAM microgels were prepared by the surfactantfree emulsion polymerization (SFEP) method [14]. 100 ml of Type I water (18.2 MΩ cm) was filtered through a $0.2 \,\mu m$ Acrodisc syringe filter. Then, $146 \, mM$ (1.65 g) of N-4 isopropylacrylamide (NIPAM, 99 %, Acros) monomer was dissolved in filtered water. The monomer solution was again filtered through a $0.2 \ \mu m$ Acrodisc syringe filter into a 3-neck 6 round bottom flask. The solution was stirred at 500 rpm, purged with nitrogen, and heated to 8 68°C in a temperature-controlled oil bath until the temperature of the solution became stable (1 hour typically). We then injected a solution of $2.8 \ mM \ (80 \ mg)$ potassium peroxodisulfate (KPS, 99 %+, Sigma-Aldrich) dissolved in 1 ml of the pre-filtered Type 1 water through a 10 $0.2 \ \mu m$ Acrodisc syringe filter to initiate the polymerization. The mixture was left to react 12 under continuous stirring at 500 rpm in nitrogen atmosphere overnight. After the polymerization, the solution was cooled down to the room temperature and filtered with a glass 14 wool five times to remove large particulates. The microgel particles were then thoroughly purified via five cycles of a centrifuge/dispersion process. The centrifugation was done at 16 15000 xg of relative centrifugal force (RCF), and the dispersion was enabled by a mixed process of the ultrasonication followed by the magnetic stirring. The cleaned particles were then lyophilized for further characterization. 18



20 Figure S1 -Temperature dependence of the hydrodynamic diameter in the low concentration limit (0.04 wt%) of slightly charged microgels measured via DLS. As temperature increases in the

- 22 region T = 10 32 °C, there is a weak roughly linear decrease of the average hydrodynamic diameter. As the lower critical solution temperature (LCST) of pNIPAM microgels is crossed,
- 24 microgels become hydrophobic and undergo massive deswelling.





Figure S2 - At low concentrations, the relative viscosity $\eta_r = \eta_{\infty}/\eta_s$ at infinite shear rate (obtained using a Carreau-Yasuda model fits, $\eta(\dot{\gamma}) = \eta_{\infty} + (\eta_0 - \eta_{\infty}) [1 + (k\dot{\gamma})^a]^{\frac{n-1}{a}}$) agrees well with the

$$\frac{\eta}{m} = 1 + 2.5\phi$$

Einstein equation (^ηs). For dilute suspensions (c→0), the effective volume fraction
can be related to the mass fraction using, 2.5φ = [η]c, where [η] is the intrinsic viscosity (
[η] = 4.02 ± 0:45 wt%⁻¹). The solvent viscosity, ^ηs, is taken as that of deionized water (=
0.001 Pa.s). At higher concentrations (c > 0.35 wt%) the viscosity strongly deviates in an upward direction due to inter-particle repulsions, consistent with our observation of a measurable linear
elastic moduli at c = 0.4 wt%.

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Figure S3 - (A) Storage modulus, G['] and (B) Loss modulus, G^{''}, for various microgel
42 concentrations in the temperature range (10 - 15)°C probed at a fixed strain amplitude of γ₀ = 1% in the linear response regime at an angular frequency of ω = 1 rad/s The temperature
44 is increased at a rate of 1 °C/min. The rheological properties are temperature independent in the range of probed temperature.



Figure S4 - Cubic spline fits to the amplitude sweep data to extract the yield properties. The strain amplitude at which a cubic spline fit to $G_1^{"}$ achieves a maximum is taken as the dynamic yield strain and the point of intersection of cubic spline fits to $G_1^{'}$ and $G_1^{"}$ is taken as the absolute yield strain.

Parameter	Value	How determined
2 <i>R</i>	550 nm	Measured by DLS at low concentration (0.4 wt%)
<i>c</i> ₁	0.4wt%	Lowest concentration studied in experiments
<i>c</i> ₂	1.5wt%	Observed concentration where $G'(c)$ plot turns to linear dependence
$(\phi_{0.5 wt\%}, E)$	(0.5067, 30000)	Volume fraction $\phi_{0.5\text{wt\%}}$ and softness parameter E are determined simultaneously from the inset of Fig. 8 by requiring the theoretical prediction of modulus to match the modulus at 0.5wt% and additionally requiring that theory produces a similar order of magnitude growth in <i>G</i> ' as that of experiments before soft jamming occurs.

54 The volume fraction at all concentrations is obtained by using the relations:

56 $\phi \sim c^{1/2}$ for 0.5 < c < 1.5 wt%, and $\phi \sim c^0$ for 1.5 < c < 9 wt%

58 and $\phi_{0.5\text{wt}\%} = 0.5067$.

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