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Spontaneous reduction of polydispersity and self-healing colloidal crystals

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Crystallization is often suppressed by point defects due to larger impurity particles. Surprisingly, microgels can overcome this limitation: Large microgels can spontaneously deswell to fit into the crystal lattice of smaller but otherwise identical microgels. We find this unique reduction of polydispersity and particle deswelling to be triggered by a difference in osmotic pressure, Π, between the inside and the outside of the microgel particles. Π is set by the counterions of charged groups on the microgels. Although pNIPAM is uncharged, pNIPAM microgels carry charged groups originating from the starter for their synthesis. A Π-difference between inside and outside of a microgel builds up when the counterion clouds of neighboring particles overlap at high concentration. This causes an increase of Π in the space between particles, which is not compensated inside the particles. With increasing concentration, this Π-difference exceeds the bulk modulus of the softest and largest microgels and makes them deswell, enabling crystallization. We find the freezing point of polydisperse and bidisperse pNIPAM suspensions to be linked to particle deswelling: A reduction of polydispersity due to particle deswelling is required for crystallization. Compared to monodisperse suspensions, this causes the freezing point to shift to higher concentrations. In comparison to hard, incompressible colloidal particles, this particle deswelling mechanism fundamentally changes the role of polydispersity in microgel suspensions.

Position

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