Gelation in Colloidal Systems: Non-Equilibrium Phase Separation meets the Glass Transition

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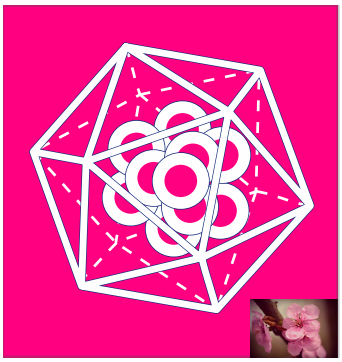

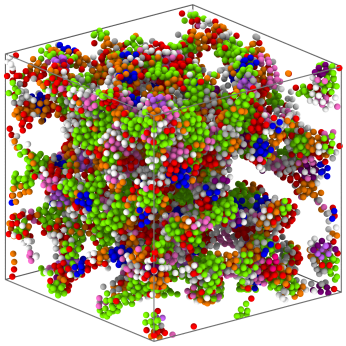

Gels are an everyday material, from foods to cosmetics to structures in living organisms. Yet gelation is among the most challenging phenomena in soft condensed matter to understand. Gels are non-equilibrium materials, which already puts them right at the limit of our understanding. As if the were not enough, gels sit at the intersection of the glass transition and failure in amorphous materials. The glass transition has been described as “the deepest problem in condensed matter physics” and failure of amorphous materials continues to evade a meaningful theoretical description [1,2].

Here we describe recent development in our understanding of the nature of colloidal gels [3,4] and explore mechanisms by which gels acquire their unusual “soft” solidity [3,4,6]. In particular, we elucidate the perceived wisdom of materials science that the *microscopic structure determines the dynamics and macroscopic behaviour of the material.* We develop an intuitive approach based on the local structure of constituent colloidal particles which provides a basic mechanism for local rigidity in the system. We show how this leads to a long—lived yet metastable solid material and provide approaches to understand the mechanisms of failure, which are key to applications in these complex yet everyday materials.

[1] Royall CP & Williams SR “The role of local structure in dynamical arrest”, *Phys. Rep*. **560** 1-75 (2015).

[2] Royall CP, “Hunting Mermaids in Real Space: Known Knowns, Known Unknowns and Unknown Unknowns” *Soft Matter* **14** 4009-4016 (2018).

[3] Royall CP, Williams SR, Ohtsuka, T and Tanaka H, “Direct observation of a local structural mechanism for dynamic arrest”, *Nature Materials* **7** 556-561, (2008).

[4] Royall CP, Williams SR and Tanaka H “Vitrification and gelation in sticky spheres”, *J. Chem. Phys*. **148** 044501 (2018).

[5] Richard D, Hallett JE, Speck T and Royall CP, “Coupling between criticality and gelation in “sticky” spheres: A structural analysis”, *Soft Matter* **14** 5554-5564 (2018).

[6] Royall CP, Eggers J, Furukawa A and Tanaka H, “Probing Colloidal Gels at Multiple Length Scales: the Role of Hydrodynamics”, *Phys. Rev. Lett.* **114** 258302 (2015).