

Attractive colloidal dispersions: flow, gelation, yielding and sensitivity to high power ultrasound

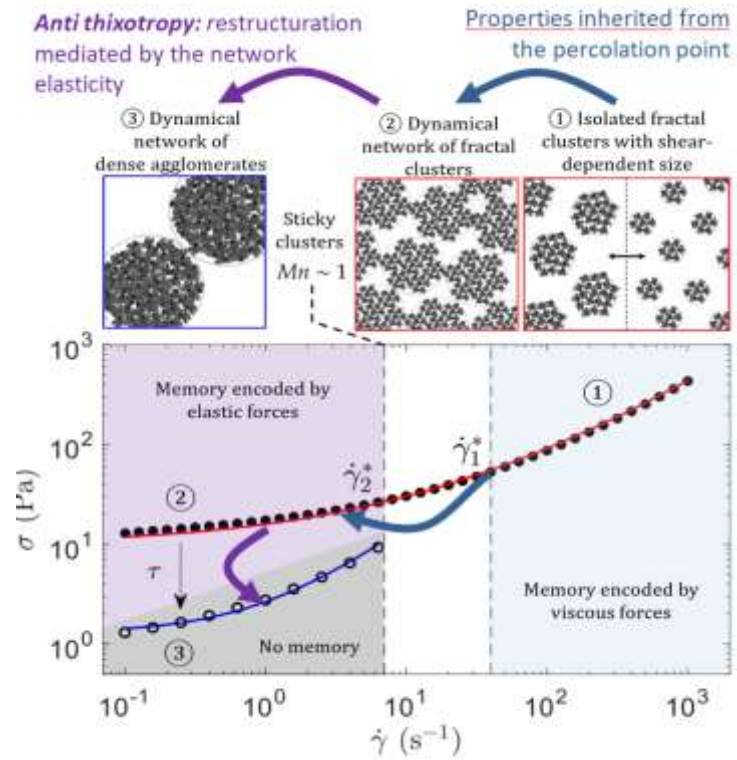
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The rheological behavior of colloidal dispersions is of paramount importance in a wide range of applications, including construction materials, energy storage systems and food industry products. Here we explore how dispersions of attractive colloids, namely carbon black particles (CB) in oil [1], flow, gel and yield and how those properties are affected by high power ultrasound. In this study, we adopt a synergistic approach, combining rheology to measure the mechanical properties of the dispersions, ultra-small-angle X-ray scattering (USAXS) to probe the structure of dispersion, ultrasonic velocimetry to access the local flow profiles and impedance to characterize the dispersion conductivity (connectivity).

Flow – Attractive colloidal dispersions consistently exhibit non-Newtonian behaviors, a consequence of intricate interplays involving colloids morphology, volume fraction, and inter-particle forces. Understanding how colloids structure under flow remains a challenge, particularly in the presence of attractive forces leading to clusters formation. First, we show that in the hydrodynamic regime, at high shear rate, CB particles structure into fractal clusters, which size conforms to a power law of the shear rate [2] aligning with simulation from Varga [3]. Second, drawing insights from the fractal structure of clusters, we compute an effective volume fraction in clusters and show that the clusters flow like hard sphere following Krieger Dougherty model [4]. Third, at low shear rate, we identify a critical shear rate at which the clusters percolate to form a dynamical homogeneous network. This low shear rate regime is not stationary. Using step down shear rate experiments, we show that CB dispersion display an anti-thixotropic behavior characterized by an initial decrease in viscosity before reaching a steady state as already observed by Wang [5]. We demonstrate that the anti-thixotropic behavior is accompanied by the restructuration of dynamical network of clusters into a dynamical network of dense agglomerates weakly connected. This transition is mediated by elastic stresses [6].



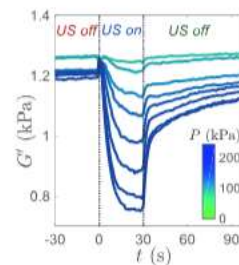
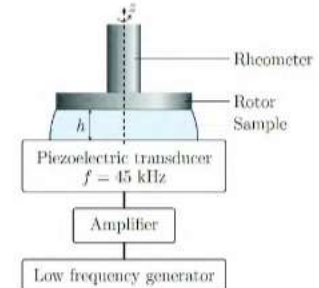
Flow cessation and gelation – Next, we focus on gelation. More precisely as we abruptly stop the flow the dispersion gels. We show that the resulting gels properties depends on the shear rate before flow cessation as already observed by Koumakis [7], therefore exhibiting shear memory [8]. When expose to high preshear using a scaling model inspired by [9] we are able to relate the gel structure to its elasticity.

Yielding – once the gels are formed, we show that their yielding properties are time dependent. Using creep experiments, we demonstrate that the solid to fluid transition display delayed yielding: the

fluidization time exponentially decreases with increasing stress [10-12]. This transition is spatially heterogeneous.

Rheo-acoustic gels – We show that high-power ultrasound above a critical amplitude leads to a complex viscoelastic transient response of the gels within a few seconds: a softening of its storage modulus accompanied by a strong overshoot in its loss modulus. Under high-power ultrasound, the gel displays a viscoelastic spectrum with glasslike features and a significant decrease in its yield strain. Those effects are attributed to the formation of intermittent microcracks in the bulk of the gel as evidenced by Time resolved USAXS. Provided that the shear rate is not large enough to fully fluidize the sample, high-power ultrasound also facilitates the flow of the gel, reducing its yield stress as well as increasing the shear-thinning index, thanks again to the formation of microcracks [13-14].

Rheology coupled to ultrasound



Ultrasound induces a strong softening

WHY?

- Not due to radiation pressure
 $\lambda_{US} \sim 10 \text{ mm} \gg L_{cluster} \sim 1 \mu\text{m}$
- Repetitive acoustical strain
 $\gamma_{US} \sim 0.4 \% \text{ VS } \gamma_y \sim 5\%$

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