

Supplementary Information

ROLE OF WALL SLIP

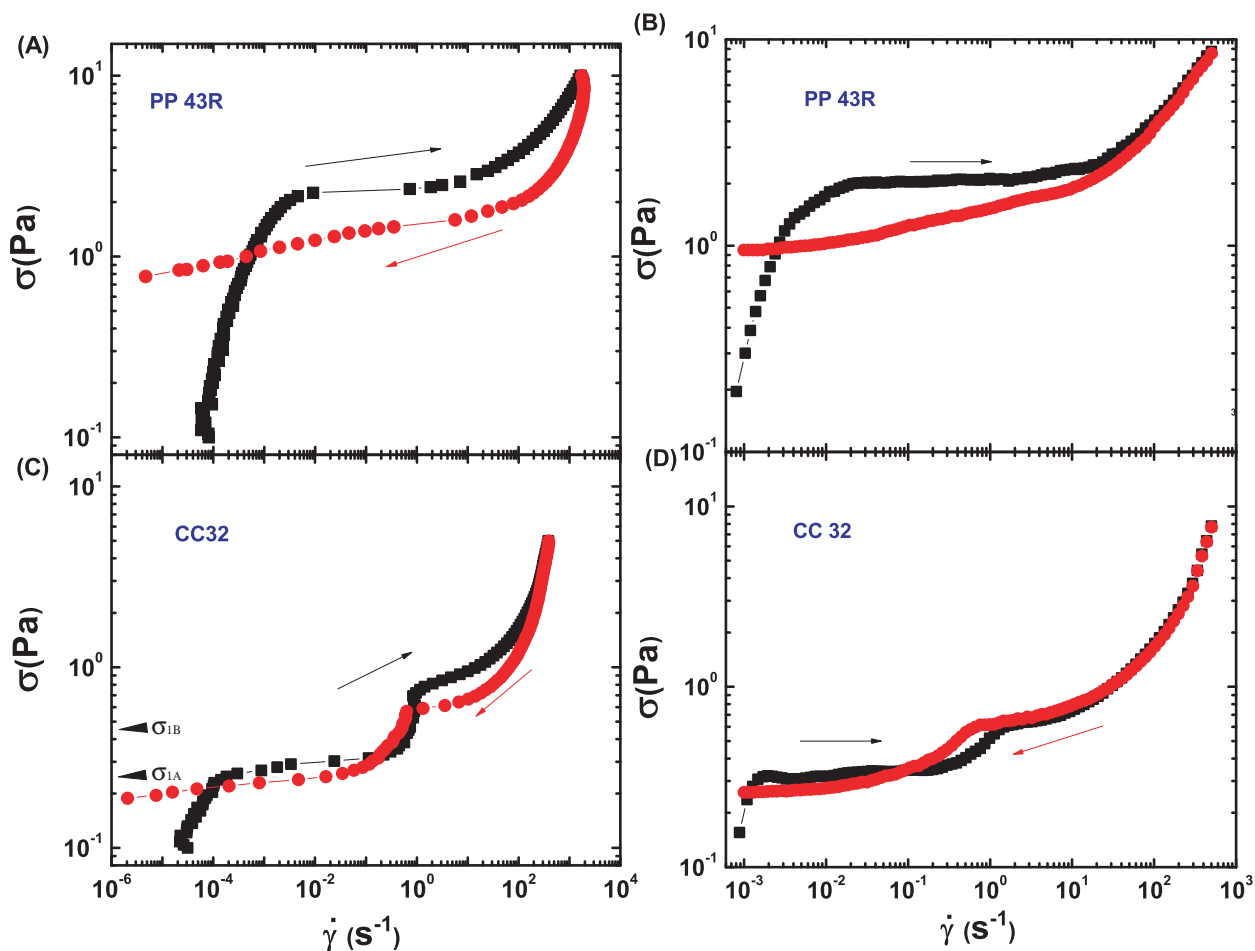


FIG. 1: Flow curve, shear stress (σ) versus the shear rate ($\dot{\gamma}$) obtained in the controlled shear stress (CSS) mode in A) Rough parallel plate (PP43R) at a gap of $100 \mu\text{m}$ and C) Couette geometry with a gap of 2mm . Flow curve in a controlled shear rate (CSR) mode in B) Rough parallel plate (PP43R) and D) Couette (CC32) geometries with the same specifications as CSS. The waiting time was 20 s for each data point. The volume fraction of the GO suspension is 0.00375 . The arrows indicate the forward and reverse measurements

A characteristic feature of the flow curves shown in Fig 1 for a GO suspension at $\phi = 0.00375$, is the appearance of two stress plateaus in a Couette geometry as opposed to a parallel plate geometry where only a single stress plateau occurs. Dual stress plateaus in the

flow curves can arise from inherent microstructural changes in suspensions giving rise to a flow instability [1] or can occur from wall slip [2] where the velocity at the walls of the stator or rotor does not match with that of the suspension immediately adjacent to the walls. For the GO suspensions, controlled shear stress flow curves (Fig 2A) indicate an unexpected sharp drop in viscosity at significantly lower stresses (marked as σ_{1A} in Fig 1B and 2A) in smooth plate and Couette geometries in comparison with the cone-plate or a rough plate geometry. However, particle velocimetry measurements in a Couette geometry indicates that no steady state flow occurs at stress values corresponding to σ_{1A} and the suspension remains jammed, moving around the rotor as a solid body. The onset of flow occurs only at stress values corresponding to σ_{1B} . (see Fig 1B and Fig 2A).

Creep measurements at different values corresponding to the stress plateau (where the temporal evolution of shear rate was monitored in flow geometries of varying roughness) revealed oscillations in shear rate with their time period corresponding to that of rotation of the rotor for geometries with smooth walls (Fig 2B). To further confirm the role of slip, stress relaxation measurements were carried out in different flow geometries, at shear rates corresponding to the plateau in the flow curve. A characteristic stress response at a shear rate of 0.5 s^{-1} in a Couette geometry after a steady state of flow is reached (Fig 2C), indicates large ($\frac{\delta\sigma}{\sigma} \sim 30 \%$) periodic fluctuations in stress reminiscent of flow instabilities [3]. However the time period of the fluctuations corresponds to the period of rotation of the rotor suggesting a correlation between the bulk flow and a stick-slip motion at the walls of the rotor. The scaling of the time period of the stress fluctuations with the globally imposed shear rate (Fig 2D) further substantiates the role of wall slip fluctuations. In addition, similar oscillations were also observed at shear rates well beyond the stress plateau, clearly indicating the significant role of wall slip even in a spatially homogenous steady shear flow. As proposed earlier in the context of dense emulsions, these oscillations can arise from spatial heterogeneities in the flow along the velocity direction, adjacent to the walls [4]. The stress relaxation and creep measurements carried out in a rough parallel plate and cone-plate geometries did not reveal any periodic oscillations in steady state flows.

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- [1] W. M. Holmes, P. T. Callaghan, D. Vlassopoulos, J. Roovers, 2004, *J. Rheol.*, **48**, 1085-1102.
[2] H. A. Barnes, 1995, *J. Non-Newtonian Fluid Mech.*, **56**, 221-251.

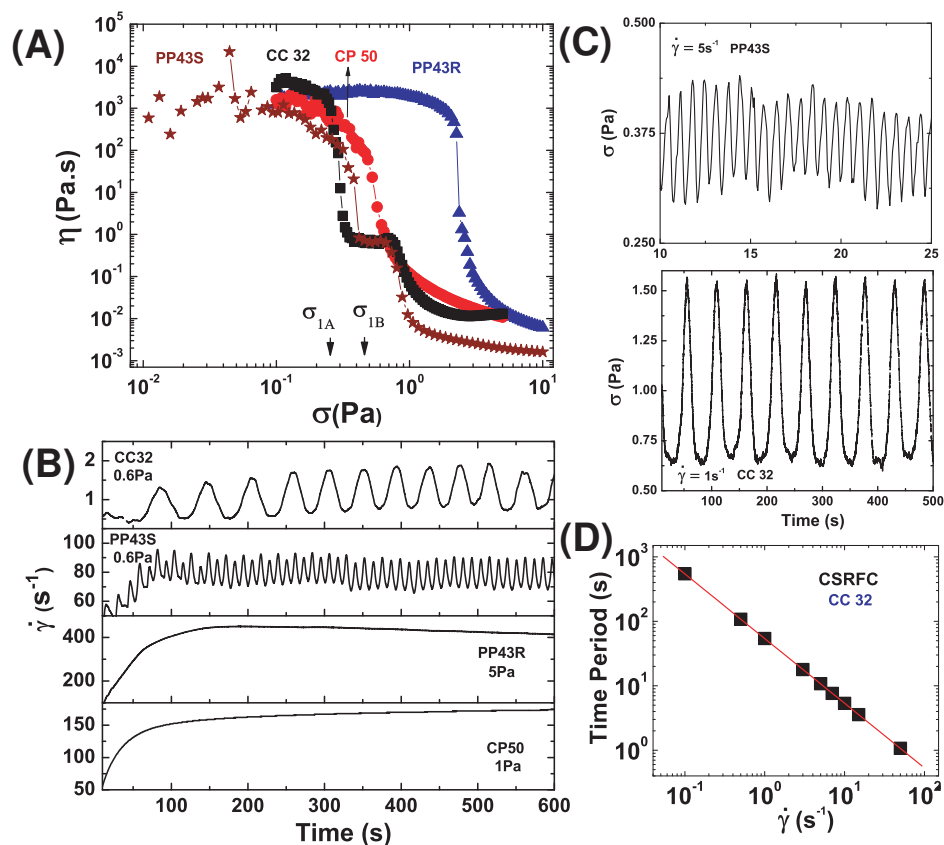


FIG. 2: A) Viscosity (η) vs. shear stress (σ) for the GO suspension ($\phi=0.00375$), measured in a couette (CC32), cone-plate (CP50) smooth parallel plate (PP43S) and rough parallel plate (PP43R) geometries. (B) Evolution of shear rate with time at an imposed stress, once the steady state is reached, indicates periodic oscillations in couette and smooth parallel plate geometries. The periodic oscillations are absent in a cone plate and rough parallel plate geometries. (C) Stress response at different imposed shear rates in a couette and smooth parallel plate geometries indicating periodic oscillations during a steady state of flow. D) The time period of oscillations scales inversely with the shear rate imposed as shown by the linear fit to the data.

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[4] L. Bècu, P. Grondin, A. Colin, S. Manneville, 2004, *Coll. and Surf. A*, **263**, 146-152.

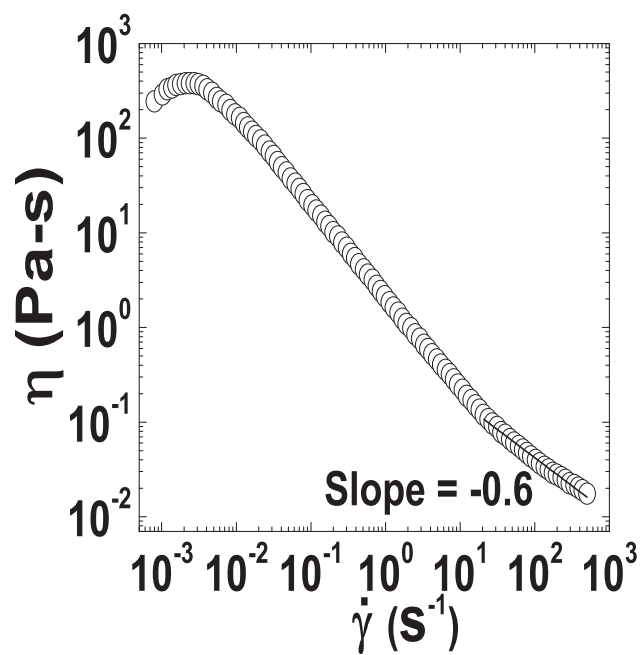


FIG. 3: Viscosity (η) versus the shear rate ($\dot{\gamma}$) obtained in the controlled shear rate (CSR) mode in a cone-plate geometry. The waiting time was 20 s for each data point. The volume fraction of the GO suspension is 0.00375. The solid line indicates a linear fit.