

## **Adjuvanted silica suspensions: from particles contact properties to rheology**

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### **ABSTRACT**

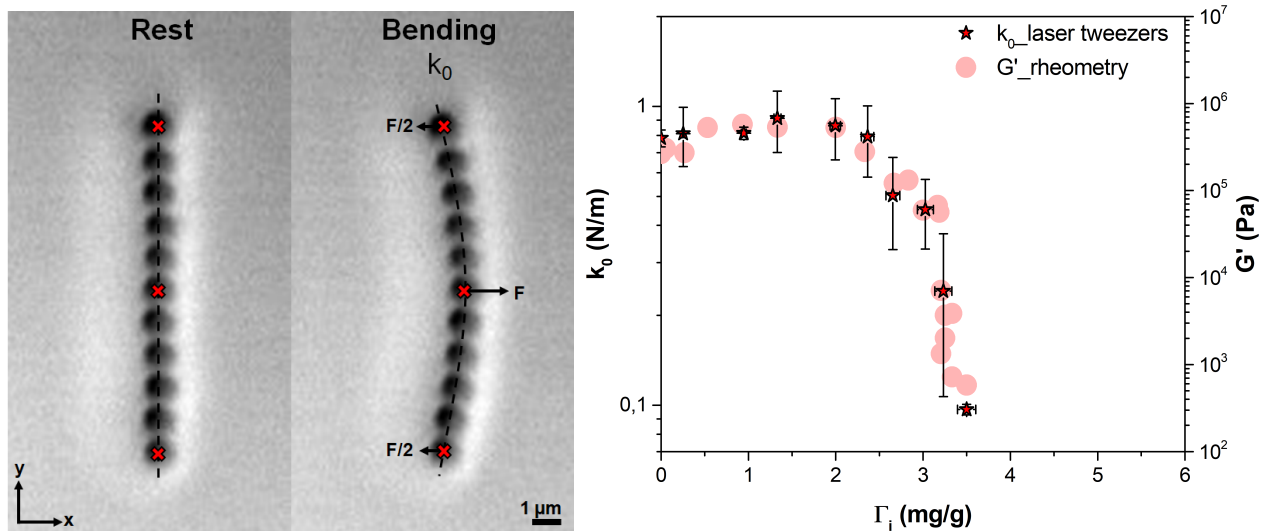
The behavior of dense aqueous silica suspensions is investigated through a comprehensive experimental approach that considers the material's properties at different length scales, from the contact level up to the overall properties. The first part of the study explores the ageing behavior of these suspensions and identifies a contact-controlled ageing process that governs both the shear modulus and yield strength ageing. Subsequently, the effect of a non-ionic surfactant on the yield stress and shear modulus of the same model suspension is studied. Results show that the addition of the surfactant significantly alters the interparticle contact properties, leading to modifications in the suspension's storage modulus and overall yield stress. Importantly, we establish a relationship between the interparticle contact properties and the overall properties of the suspension for both systems.

### **Ageing in silica suspension**

While the ageing behavior of dense suspensions or pastes at rest is almost exclusively attributed to structural dynamics, a set of experiments conducted on dense aqueous silica suspensions allowed us to identify another ageing process: contact-controlled ageing. To do so we have investigated the origin of shear modulus and yield strength ageing in dense aqueous silica suspensions at moderate ionic forces. We first showed by combining rheometry and confocal microscopy that the elastic modulus and yield stress of these suspensions at rest grow logarithmically with time while their structural evolution is rapidly arrested by the formation of thermally irreversible roll-resistant interparticle contacts. So these suspensions age in the absence of structural rearrangement. Then by performing three-point optical tweezer (OT) bending tests on particle rods, we showed that particle contacts resist rolling and yield by overcoming a rolling threshold. We also observed that both the rolling stiffness and rolling threshold grow logarithmically in time. By comparing the results of the measurements on the particle contact properties with the results of the rheometry tests, we were able to show that ageing of interparticle contacts governs both shear modulus ageing and yield stress ageing of these dense aqueous silica suspensions. We also identified simple constitutive relationships between contact-scale bending stiffness and rolling threshold, which transfer to macroscopic scale. This allowed us to propose a constitutive relationship between the macroscopic shear modulus and the yield strength of these silica suspensions that should be generic for an array of colloidal systems.

## Effect of an ionic surfactant

Dispersing agents are widely used to formulate high-solids pastes that remain fluid enough to be easily transported and placed. In the cement industry, superplasticizers are used to control the workability of fresh cement pastes containing high volume fractions of solid particles enabling the formulation of concretes with higher mechanical strength, better durability and also lower carbon impacts by substituting cement particles with mineral additions (fly ashes, slag, silica fumes...). Although dispersing agents are commonly used, the mechanism by which they impact the interaction between particles, and thus the paste rheology, remains obscure so that process optimization relies almost essentially on empirical or trial-and-error processes. In order to improve our understanding of these systems, we experimentally investigate the influence of a non-ionic surfactant that mimics superplasticizers on the yield stress and storage modulus of model suspensions containing silica beads suspended in an ionic solution. By combining rheometry, Total Organic Carbon, zeta potential, confocal microscopy and laser tweezers experiments, we show that adding non-ionic surfactant to the suspension strongly alters both the interparticle contact mechanical properties (stiffness and strength) and the overall suspension rheology (yield stress and storage modulus). Change of the contact rigidity is ascribed to the adsorption of the surfactant on beads, which induces a shift from an adhesive rolling resisting contact to a non-adhesive contact due to steric hindrance. As demonstrated by the perfect agreement between rheometry and 3-point bending tests, drop of the suspensions storage modulus originates from a particle contact shift. Same relationship is observed between the flexural contact resistance and overall suspension yield stress or the ageing of the material.



**Figure 1:** (Left: 3-point bending test on a particle rod using laser tweezers to probe the contact rigidity ( $k_0$ ). Red crosses represent optical traps used to move the beads and to measure the applied forces. Right: Contact stiffness measured through 3-point bending tests and shear elastic modulus ( $G'$ ) of the suspension (solid volume fraction: 45 % ; ionic strength: 0.15 M) as a function of the mass of surfactant added per unit of bead mass ( $\Gamma_i$ )

## REFERENCES

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