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Contact aging in dense and attractive colloidal suspensions

Dense colloidal suspensions constitute a broad class of materials found in areas ranging from environmental systems (silts, clays), to industry (ceramics, drilling muds, slurries), construction (plaster, cements), foodstuff, cosmetics and pharmaceuticals (toothpaste, medical ceramics). Their most remarkable feature is thixotropy and aging: a slow evolution of their rheological properties when switching from rest to flow and vice versa. While this behavior has been almost exclusively attributed to structural dynamics, a set of experiments conducted on dense aqueous silica suspensions allowed us to identify another aging process: contact-controlled aging. To do so we have investigated the origin of shear modulus and yield strength aging in dense aqueous silica suspensions at moderate ionic concentrations. We first showed by combining rheometry and confocal microscopy that the elastic modulus and yield stress of these suspensions grow logarithmically with the resting time, while their structural evolution is rapidly arrested by the formation of thermally irreversible roll-resistant interparticle contacts. 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 these results with those of the rheometry tests, we were able to show that aging of interparticle contacts governs both shear modulus and yield stress aging 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 suspensions that should be generic for an array of colloidal systems.

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