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Mechanical Consequences of Fluid Transport in Gels and Suspensions

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Mechanical consequences of fluid transport in gels and suspensions

In this thesis, we investigate the rheological behaviour of complex materials that are a mixture of a solid and liquid phases whose displacements can be decoupled. A particular emphasis is put on the relative motion between the two phases, and the consequences that a potential decoupling can have on the mechanical response of the material.

The first chapter is a brief introduction to the topic, providing a description of the materials used in the thesis, and defining the properties of visco-elastic materials with simple models as well discussing the normal stress differences. Classical evidences of the manifestations of normal stresses are shown, and the scope of the thesis is presented.

The second chapter focuses on the materials and methods used in the thesis. A first section is dedicated to rheology, both from a theoretical and a practical point of view. Subsequently, the principle of magnetic resonance imaging and its utility in local rheology measurements is described. Finally the the making of the different materials is detailed.

In chapter 3, we focus on hydrogels, materials made of water in a cross-linked network. The main purpose of this chapter is to understand why some synthetic gels such as polyacrylamide show positive normal stresses, and some others, made of biopolymers, exhibit the opposite tendency. We show that the main factor for this is the pore size of the network, which, in turn, controls the permeability of the solid material. If water is not allowed to flow through the pores, as it happens in polyacrylamide, the whole system will behave like a single phase incompressible material, and will expand upon shearing. To the contrary, if water is free to move, then then network can contract when deformed by expelling the fluid out of the gel, and will exhibit negative normal stresses. By tuning with the gel pore size, we find that it is possible to regulate the normal stresses, and reveal a transition in which the network initially expands, then contracts slowly as water is expelled out of the gel. The experimental data are discussed in the light of two models. This discovery paves way to new methods in material processing and control normal stresses, which could prevent industrial problems such as die swell to happen.

The fourth chapter is focused on another two phase material : a Dense Non-Brownian suspension. These are solid spheres floating in a density matched liquid. In spite of its apparent simplicity these materials exhibit non trivial physics, among which normal stresses. Previous reports however are in disagreement even on the sign of the normal stress, and never considered very low viscosity interstitial fluids, in which shear thickening takes place. By performing rheology experiments,we discover that a finite size effect appears at very low gap, which does not appear in high viscosity suspensions. We suspect that, once again, the solvent mobility through the pores, which is inversely proportional to viscosity, but also depends heavily of the beads size, plays a decisive role. The discovery of a characteristic relaxation time that

depends of these parameters is strong evidence for this theory.

Chapter 5 is a bit apart from the rest of the thesis since it is not about normal stresses. Instead we consider in the suitability of the fluidity model in predicting flow curves of suspensions or granular materials subjected to a shear rate gradient. In such materials, particle migration can happen and, sooner or later, inhomogeneities will appear. Macroscopic rheology is therefore invalid in this case, homogeneity being one of the main conditions of its application. We therefore probe the sheared suspension with magnetic resonance imaging, which gives the local volume fraction and flow velocity of the material. With these experimental data, we first build the flow curves for ideal homogeneous suspensions which are needed to apply the the fluidity model. We show that the model describes the experimental data, but requires the knowledge of a parameter, the flow cooperativity length, ξ which depends both on the particle size and the flow geometry. This prevents the model to be used for predictive purposes.

In conclusion, this thesis covers several aspects of complex materials in which the decoupling between the displacement of the two phases can lead to dramatic effects, such as negative normal forces and particle migration. The understanding of these non trivial phenomena has progressed, in particular for gels where a link between expansion/contraction and permeability was made. However, some questions also remain, especially in the domain of suspensions. Some leads are given for possible more in-depth studies in the near future.