



The Rheology of Jamming
M. Dinkgreve

Abstract

Traffic jams are a common phenomenon on highways; when there are too many cars on the road the traffic gets stuck. A similar jamming phenomenon also occurs in yield-stress fluids that consist of a dispersion of a material in a liquid, such as suspensions of particles or polymers, foams or emulsions. At high concentrations, these materials behave like solids (like in traffic jams there is no flow), and they only start to flow when enough stress is applied. For example, toothpaste behaves like a solid at rest but it starts to flow when you squeeze it out of the tube. This threshold stress that is needed to initiate flow is called the yield stress, hence the name yield-stress material. It is important to understand these kinds of properties and the flow behavior (rheology) of these materials since they are widely applied in the cosmetic, oil and food industry. We seek to understand the transition from mechanically solid-like to fluid-like behavior on a fundamental level. The associated jamming transition between solid and liquid "states" has similarities to classical phase transitions like those between solid, liquid, gas and plasma phases. However it is not completely clear how general the jamming description is, and to what extent the mechanical behavior of jammed materials can be fully described by considering the jamming transition to be analogous to a classical phase transition.

In this thesis we study the flow behavior (the rheology) of a variety of yield-stress materials in the aim of describing, understanding and predicting the rheology of jamming.

Chapter 1 gives the motivation for this research and a general introduction to the main topics related to this thesis are explained: the jamming phase diagram, rheology and yield-stress materials. In chapter 2 the experimental techniques and the different materials that were used are discussed. Rheology was the main method used to understand the flow properties of the different materials and a variety of rheological tests is described in detail.

Since the notion of a yield-stress fluid was introduced by Bingham in 1922, these fluids have become more and more popular. However, in the nineties Barnes and Walters published a provocative paper entitled "the yield stress myth?" from which a huge discussion arose whether the yield stress really existed or that it was just an experimental artifact. In chapter 3 we review the definition of the yield stress and demonstrate the significance of the pre-yielding behavior through a number of elementary measurements, showing that the yield stress is real. Secondly, in chapter 4 we compare different methods of measuring the yield stress with conventional rheometers that have been used in the literature on a variety of materials. Subsequently, an overview is given of the different values that are found for the yield stress and yield strain.

In chapter 5 we investigate the stability of Laponite-stabilized Pickering emulsions under shear. Often, colloidal particles are used to make Pickering emulsions that have been reported to be very stable. Commonly the stabilization is a combined effect of particles adsorbing at the fluid interface and a particle network in the continuous phase, however the contribution of each to the overall stability is difficult to assess. To clarify the structure of the emulsion and the role of the clay particles, we fluorescently label the clay particles by adsorbing the dye onto the particle surfaces. This allows us to show directly by using confocal microscopy, that the clay particles are not only located at the interface but also aggregate and form a gel in the continuous aqueous phase. This reveals that the formation of the emulsions with clay particles only, without surfactant, is mostly due to gel formation of the clay particles in the continuous phase, rather than that the clay is an emulsifier.

In chapter 6 we demonstrate experimentally that a simple yield-stress fluid, can undergo a transition to thermally induced shear banding. The experiments suggest that both types of behavior may be

found in the same type of material, however the presence of thermal parts in addition to athermal jamming leads to a depletion interaction that causes an apparent shear band to appear. A set of rheological tests are done to study this transition in a Carbopol gel. Flow visualization experiments, with fluorescently labelled Carbopol, elucidate the difference between simple and shear banding behavior and reveal the presence of thermal particles in the system. We therefore show that a simple yield-stress fluid can be transformed to a shear banding yield-stress fluid by different preparation protocols.

In chapter 7 we aim to describe, understand and predict the flow behavior (stress vs. shear rate) of simple yield-stress fluids. The experimental flow curves of four different athermal yield-stress fluids are studied near the jamming point. By scaling with the distance to the jamming volume fraction all rheology data can be collapsed onto master curves below and above jamming. A two-state microscopic theory of heterogeneous dynamics is presented to rationalize the observed transition. Finally the experimental data and the microscopic theory are compared with much of the literature data for yield-stress systems. We conclude that all our athermal yield-stress materials can be described by one universal scaling form, independent of the mechanical properties of the system, but a discrepancy between theory and simulations remains.

In this last chapter (chapter 8) we investigate the cross-over between thermal and athermal yield-stress regimes, by looking at the effect of volume fraction, particles size and the inter-particle interactions on the flow behavior. Interestingly, the flow curves of thermal systems can be scaled onto a universal curve in a similar way as the athermal systems, with respect to the glass transition instead of the jamming transition. In addition, we find that all yield stress flow curves of both thermal and athermal systems can be collapsed, using the Laplace pressure as stress scale for athermal systems and osmotic pressure as the stress scale for the thermal systems. In conclusion, we can predict, rather than fit, characteristic stress and rate scales from material properties.