

Oldroyd's model and the foundation of modern rheology of yield stress fluids

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Abstract: Oldroyd's 1946 formulation of the constitutive equation of yield stress fluids laid the foundation of modern rheology of yield stress fluids, by considering not just the simple shear behavior of yield-stress fluids in the plastic regime, but also by prescribing a simple expression for how they behave prior to yielding and a 3D expression for how they behave beyond yielding. We discuss the implications of Oldroyd's model, and the ways in which researchers have tested and refined his ideas. We also review the experimental methods that have been employed to test these implications, and to determine the rheological parameters it lays out. We finally discuss the impact of Oldroyd's model, and the future of investigations into what happens prior to yielding.

Introduction

In 1946 Oldroyd published a paper entitled "A rational formulation of the equations of plastic flow for a Bingham solid" [1]. This paper provides much more than suggested by its title, tending to suggest that yield stress fluids (YSFs) will be described by a pure plastic flow beyond some critical stress. As a matter of diction, Oldroyd uses the term "plastic", when the material can be considered to flow, i.e. being deformed indefinitely, in opposition to "elastic", when the material's deformation under stress is limited.

The final expression of the model, established by Oldroyd in the paper, is as follows:

$$\text{"Elastic region", for } \sqrt{-T_{II}} < \tau_c, \mathbf{T} = 2\mu\mathbf{E} \quad (1)$$

in which \mathbf{E} is the strain tensor, \mathbf{T} the deviatoric component of the stress tensor, T_{II} the second invariant of the deviatoric stress tensor, μ a "rigidity modulus" (our modern "shear modulus") and τ_c the "yield value" of the material.

$$\text{"Plastic region", for } \sqrt{-T_{II}} > \tau_c, \mathbf{T} = (2\eta + \tau_c / \sqrt{-D_{II}}) \mathbf{D} \quad (2)$$

in which \mathbf{D} is the rate of strain tensor

The content of Oldroyd's paper may appear as relatively obvious and standard to a modern reader, but one should realize that these results and the analysis leading to them provided in this paper tell us much more than these two simple equations. Let us review several of the main aspects of the model:

- 1) **Two regimes.** Oldroyd proposes to describe the behavior of a YSF by considering it exhibits two regimes, namely a solid one ("elastic"), in which the material behaves linearly elastically, and a liquid ("plastic"), in which the material effectively flows, i.e. it is continuously deformed so that its flow characteristics are solely described by a relation between the stress tensor and strain rates, and are time-independent.
- 2) **Tensorial constitutive equation.** Oldroyd presents a tensorial description of the behavior in each regime, which allows him to express the material behavior under any type of boundary conditions.

- 3) **Yielding criterion.** Oldroyd builds a criterion for the transition between the two regimes, which is consistent with the form of the constitutive equation in both regimes; this (von Mises) criterion relies on the idea that yielding occurs when the potential energy per unit volume due to deviatoric stresses reaches a critical value. The demonstration of the consistency of this criterion leading to a single yielding parameter, i.e. the yield value, occupies a large part of the paper. Additionally, Oldroyd makes consistent reference to a quasi-static approach to the yield criterion, allowing for the possibility of transient behaviors that would differ substantially from this steady-state treatment.

In this paper, we will discuss the ways in which Oldroyd's formulation was ahead of his time by considering not just the behavior of yield-stress fluids in the plastic regime, but also by prescribing a simple expression for how they behave prior to yielding. This simple extension has provided a framework that is still heavily used by modern researchers.

We will also discuss some of the implications of Oldroyd's model, and the ways in which researchers have tested and refined his ideas. The forward-thinking nature of Oldroyd's work is further shown by the 60+ year gap between the publication of his paper in 1946, and modern formulations that build upon his ideas published in 2007 [2].

The next major section of this paper will be a discussion of the experimental methods that have been employed to test the implications of Oldroyd's model, and to determine the rheological parameters he lays out. We finish the paper with a summary of the impact of Oldroyd's model, and the future of investigations into what happens prior to yielding.

2. Oldroyd's description of yield stress fluids ahead of his time

Most studies since the initial proposal of Bingham [3], including the works of Herschel and Bulkley [4] and Hohenemser and Prager [5], essentially developed descriptions of the properties in the liquid regime, neglecting the possible deformations in the solid regime. Oldroyd's is therefore an original perspective that considers the different facets of the material behavior above *and* below the yield condition. Previously, yield stress fluids were only considered as fluids with a strongly non-linear behavior, which only deformed or flowed once their yield stress was exceeded. The same conclusion holds about the advanced formulation of Schwedoff [6] about twenty years before Bingham's publication, which describes the transition to flow with a viscoelastic model, but just considered the solid regime as a no-flow regime. Even today, it is not so common to see treatments of YSFs that exhibit two such regimes of behavior.

It was therefore daring of Oldroyd to propose a description of behaviors above and below the yield condition. His proposal naturally leads us to consider that YSFs belong to both fluid and solid material classes, and that their description should involve both Lagrangian and Eulerian approaches.

It was not until the nineties, around five decades after Oldroyd's paper was published, that researchers revisited his proposal and began to reconsider, via various modelling approaches, the behavior of YSF in their solid regime. Even so, the number of works that have investigated the solid behavior of YSFs more deeply remains limited. It has been considered in the field of thixotropy [7,8] in relation with the description of concentrated suspension behavior. In such descriptions, the formation of flocs provides the mechanism for storing elastic energy (by deformation). These flocs become smaller (by erosion or breakage to viscous stress) with increasing shear rate [9]. In this frame, the continuity with the solid regime is natural: it corresponds to the case for which a floc percolates throughout the sample and has to be broken for flow to take place.

Another remarkable aspect of Oldroyd's work is the construction of a consistent tensorial form of the constitutive equation of a rather complex material, well before the developments of rational mechanics by Truesdell and Noll [10], theoretical viscometry by Coleman, Markovitz, and Noll [11], or the proper mathematical expressions of constitutive equations for complex fluids by Rivlin and

Ericksen [12]. It is even more remarkable that this was done for a material type whose behavior strongly differs from the polymeric materials which were to be studied and modelled in depth in the next decades (see e.g. Bird [13] and Ferry [14]).

The DNA of Oldroyd's model is apparent in many modern formulations. The ideas of separating YSF behavior into two distinct regimes with their own physical description and linking the regimes with the von Mises criterion can still be seen in models that were published more than half a century after Oldroyd's. The different forms of model put forward by Saramito [2] and de Souza Mendes and Thompson [15] follow the formulation of Oldroyd. The three main aspects of Oldroyd's work we identify; the concept of an elastic and a plastic regime, the tensorial description, and the inclusion of the von Mises criterion, therefore remain essentially unchanged in today's descriptions of YSFs.

Importantly, Oldroyd's expression relying on the von Mises criterion as the threshold between an elastic and a plastic regime is still the basic 3D expression of the constitutive equation of a simple yield stress fluid [16–18], perhaps with the simple extension to a Herschel-Bulkley term for the liquid regime [19]. The von Mises criterion had also been considered by Hohenemser and Prager [5] in their description of the yield condition of metals, though these authors assumed perfectly rigid behavior in the solid regime. Oldroyd's more complete formulation of the constitutive equation of YSF is likely at the origin of the naming, by some authors, of the ratio of the two terms of the Herschel-Bulkley model for the liquid regime as the Oldroyd number [20]. To properly recognize his contribution, the Oldroyd number should be seen as an estimation of "how far we are from the solid regime", through a comparison of the maximum elastoplastic component in the solid regime to the additional viscous term in the liquid regime. This number is typically referred to as the Bingham number elsewhere.

Despite the inclusion of ideas that have become standard inclusions of models of YSFs, Oldroyd's description of both regimes went under-discussed in the next 50 years, as people essentially focused on the liquid regime with simple tools. A new phase of strong interest in yield stress fluids was sparked by the debate about the existence of yield stresses initiated by Barnes and Walters in the eighties [21]. Ultimately, this debate reached the conclusion that the yield stress is an engineering reality [22,23]. Interestingly, some of the tests that were used to question the existence of a yield stress relied on data associated with only very limited total deformations that essentially placed the observations in the solid regime. It could be argued that such tests neglected Oldroyd's quasi-static condition and instead interpreted very slow transient behavior as representing the steady state. In these cases, it was shown that some extremely slow unsteady flow may be observed, whereas the total deformation remains limited, with some similarity with creep flows of solids. This further proved the need to clearly identify the two regimes to properly determine YSF behavior.

In the 2000s, further discussions on the existence of a yield stress or, more pragmatically, on its determination, relied on this frame of description [24–26]. Determination of the yield stress was shown to be difficult, and that different answers may be obtained depending on the choice of experiment that was performed. Dinkgreve et al went as far as to show how multiple measures of the yield stress have been obtained from the same test, and that there is still a lack of agreement as to when yielding occurs [26].

It is reasonable to question whether it is constructive to think that the yield stress varies with the technique or procedure used to determine it. For a homogeneous material prepared in a given state and undergoing a well-controlled stress field history, the path to yielding is deterministic and should lead to a single value of the yield stress associated to a given material state. For simple (non-thixotropic) yield stress fluids [25,27–29] it is thus necessary first to control the material homogeneity and initial state. This is generally done by imposing a given preshear, but such a procedure may also tend to induce the development of inhomogeneities (for example associated with particle migration or sedimentation effects). In some cases, it may then be preferable to properly mix the material before each test. In the next step it is necessary to be sure to impose a shear to the bulk, which means

precluding the possibility of wall slip. The stress field must also be as homogeneous as possible. At last, the question of the appreciation of the yielding transition is critical. Various techniques attempt to provide an answer through a fast, single test or ramp. Here, it is important to be able to distinguish between the transience of the methods used to determine yielding and Oldroyd's assumption of a quasi-static approach to the yield condition and a well-defined yield stress.

Although some of these techniques can, in some cases, give a good approximation of the yield stress value, fundamentally the exact value of the yield stress can be measured only through the observation of a transition between a finite deformation, i.e. no flow, below a critical stress, and a steady state flow in the liquid regime for a larger stress. Determination of yielding therefore rests on the ability to determine plastic flow. The highest precision in the yield stress determination thus requires the ability to observe the steady flow the closest to the yield stress value, which, for a simple yield stress fluid obeying Oldroyd's or Herschel and Bulkeley's models [4], means a flow at an infinitely low rate beyond the critical deformation. This implies the ability to observe the flow over an infinite time. Obviously in practice it is not necessary to be as strict, but these considerations at least illustrate how the precise determination of the yield stress value is necessarily associated with a sufficiently large time of measurement and observation.

For thixotropic materials, the problem is much more complex. To start with, by definition, no single yield stress value can be expected in that case, since the material structure evolves as a function of the flow history, in particular at rest [30,31]. The model of Renardy [32] provides a useful overview, by considering the evolution of the conformation tensor leading to yielding at different stresses at different times. Therefore, a clearly defined single value of a yield stress does not exist in this model. Instead, yielding behavior arises in a particular limit of how the conformation tensor evolves for non-monotonic constitutive relations. Additionally, Renardy's model predicts yield stress hysteresis, dependence of the yield stress on time scales, thixotropy and the Mullins effect. Given the experimental and theoretical arguments, one has then to consider the above points about the procedure for appreciating the yielding, and the impact of the time and flow history up to the yielding point, on the current yield stress of the material.

Finally, the rheological modelling of the behavior of YSFs has been the object of many important developments in the last 15 years. We can consider that these renewed approaches started with the work of Saramito [2], who proposed an elastoviscoplastic constitutive equation for YSF covering both the solid and the liquid regimes. Saramito's work also highlighted and resolved the discontinuity in stress contained in Oldroyd's model. By enforcing continuity of the stress at the yielding point, Saramito allowed for more realistic yielding responses at conditions that differ significantly from the quasi-static case assumed by Oldroyd.

Considering the above remarks, the evolution of the structure during the solid-liquid transition should be at the source of more complete models. This means that either viscoelastic effects associated with some storage of energy in the structure, or thixotropic effects, associated with the destruction or rebuilding of the structure, can be considered. The exact distinction between these two types of effects is not so obvious (see, for example Larson's work on thixotropy [33,34], and Joshi's paper on viscoelasticity [35]), but here we will simply assume this is readily possible.

The last decade has also seen the publication of a few reviews of yielding behavior, including works by Balmforth et al [29], Fraggedakis et al [36], Coussot [37,38], Denn and Bonn et al [39] and others. Dimitriou et al [40] have introduced ideas into the YSF arena from solid mechanics, constructing a model that is based on the idea of kinematic hardening. This concept posits that the yield surface moves as a function of all previous deformations and is contrasted to isotropic hardening, in which the yield surface gets larger without moving. Kamani and coworkers have recently produced a 5-parameter model that accounts for the major phenomena observed experimentally in yield stress fluids [41]. Their model incorporates a rate-dependent relaxation time into a framework in which plastic flow is enhanced by rapid elastic deformation. With a single yield stress determined under quasi-static testing

conditions, the model shows how yielding can manifest across a range of conditions under various time-varying protocols. These models show how modeling of yield stress fluids may move beyond the Oldroyd formulation, and unify the physics observed in the yielded and unyielded states.

The fields of yielding and thixotropy have also been closely linked by works from de Souza Mendes and Thompson [15,31,42–44], and Bonn et al [45,46]. The model developed by de Souza Mendes and Thompson uses the ideas of thixotropy and viscoelasticity, without the piecewise construction of Oldroyd. Their 7-parameter model makes a strong connection between the ideas of yielding and thixotropy. This connection is conceptually appealing as both sets of behaviors deal with structural and rheological changes in response to an applied stress or strain field. A common thread running through all these models is that there are different types of deformations that lead to different stress components inside the material as it flows. The relevance of these different models with regards to experimental data continues to be tested and studied.

3. Implications of Oldroyd's description

Implicit in Oldroyd's description are the following statements. Firstly, the material undergoes an abrupt transition from a solid to a "liquid" behavior, when a yield condition is met at a critical deformation associated with a critical stress. This transition is abrupt in terms of the deformation, but not necessarily in time, as Oldroyd assumes a quasi-static approach to the yield condition. More precisely, the critical deformation can be simply derived from the constitutive equation and is expressed as follows: $\varepsilon_c = \sqrt{-E_{II}} = \tau_c / 2\mu$. In this frame, the kinematics is strictly described through some deformation in the solid regime, and through the current strain rate in the liquid regime. As a consequence, the material exhibits no viscoelasticity or thixotropy. Thus the transition from the solid to the liquid regime is also abrupt in terms of the physical characteristics of the material, and beyond the critical deformation the internal elastic deformations, which were playing the major role in the solid regime, suddenly do not play any role. A particular implication is a problem emphasized (and solved) by Saramito [2]: according to the model (1-2) in simple shear, the stress should jump abruptly from τ_c to $\tau_c + \eta\dot{\varepsilon}$ at the yield transition when a finite deformation rate $\dot{\varepsilon}$ is imposed from the initial (equilibrium) state. As the deformation rate $\dot{\varepsilon}$ approaches zero, the conditions may be said to be closer to Oldroyd's assumption of a quasi-static approach to the yield condition, and the stress jump disappears.

Another implicit statement of the constitutive equation is that an initial state of zero deformation has to be defined, since in practice the material's behavior is *a priori* described first in its solid regime. It is assumed that this state is reached after some rest, as expressed by Oldroyd: "the displacement is taken from the equilibrium position which it would attain if, at a suitable time t_0 , the motion were instantaneously stopped and the stresses remaining at zero velocity were reduced reversibly to zero"; this in particular implies that the material must exhibit some convenient properties of self-relaxation or recovery.

At this point it seems important to note again that, from a physical point of view, the conditions of transition from the solid to the liquid state, yielding, are still quite unclear, and the above models do not seem to take this clearly into account. It's also worth revisiting Oldroyd's assumption in constructing his model that the yield point be approached quasi-statically. This assumption is most often violated in studies of yielding, as there is always some characteristic timescale associated with the approach to the yield point, which was not considered by Oldroyd. Moreover, as already mentioned above, from the point of view of the liquid regime, it is in theory necessary to wait an infinite time to observe a steady flow at a vanishing shear rate.

Another implicit assumption of Oldroyd's approach, and of most models proposed afterwards for describing yield stress fluid behavior, is that yielding takes the form of a smooth transition in which (for simple shear experiments under uniform stress) the material is first deformed homogeneously in

the solid regime. The deformation continues up to a critical point and then the material starts to flow, which is also assumed to be homogeneously sheared. Despite the appealing simplicity of these assumptions, there are several reasons to expect that the reality is much more complex. First, standard solids behave in a similar way but start to break (brittle structure) or shear-band (ductile structure) beyond some critical deformation [47].

A smooth transition as described above, or the abrupt transition that results from taking Oldroyd's quasi-static approach out of context, would mean that there exists some deformation at which the whole material instantaneously turns from a solid state to a liquid state. This is equivalent to assuming that the potential energy landscape of every constituent is identical, and that the same energies are required to enforce flow. Rather, it seems more realistic to consider that the transition is more complex, involving first some localization of the deformation in regions where the local energy potentials are shallower than elsewhere. These would be a consequence of distributions in sizes, shapes, charge, of other physical characteristics, for instance. This strain field will continue to evolve as some parameter is increased (deformation, stress, shear rate) until the entire system is flowing. Such an assumption is supported by observations of shear-banding in steady state simple shear flows of thixotropic YSFs, which exhibit an apparent yield stress increasing with the time of rest [48–50] or even in supposedly non-thixotropic fluids [51–53]. It was also shown that, in the apparent liquid regime, the thickness of the shear-band increases with the apparent shear rate, and that a slow imposed flow may induce a flow in a thickness of the order of the suspended element size, which induces additional instabilities [54]. From these observations we can hypothesize that for soft solids the transition from the solid to the liquid regime first involves some fractures or localization of the deformation in some regions followed by an evolution of the strain field. Being optimistic, we can consider that this is in fact globally what the above sophisticated models attempt to encompass when they consider viscous, elastic, and plastic events in the solid regime and beyond the solid-liquid transition.

The large deformation of a plastic material has been studied through numerical simulations to understand the physical mechanisms behind plasticity of amorphous solids, and the results suggest an even wider possible phenomenology for the solid-liquid transition in soft solids. For example, it was shown [55] that plastic deformation manifests as local rearrangements exhibiting a broad distribution of sizes and shapes [56], nonaffine displacements [57], and connectivity changes between particles [58] that lead to a redistribution of elastic stresses in the system [59]. Furthermore, the collective behavior of these reorganizations includes spontaneous strain localization, intermittent dynamics and power-law distributed avalanches [60]. Complex shear band patterns may also develop with fractal structure [61]. As already remarked [62,63] YSF (and *a fortiori*, pastes) present some analogy, in terms of behavior, with amorphous solids, as we expect essentially plastic rearrangements ultimately leading to yielding. Thus, the above observations should provide inspiring information and concepts. Finally, there have been a few original experimental approaches studying the plastic events in YSF. From diffusion-wave spectroscopy, reversible and irreversible events in an oscillating flowing emulsion were distinguished, and it was shown that yielding occurs beyond a small critical amount of irreversible motions [63]. Similar observations were obtained for a colloidal glass [64]. Direct 3D-imaging of the structure evolution finally showed localized irreversible shear transformation zones and allowed to determine their formation energy and topology [65] and growing clusters of non-affine deformation percolating at yielding [62]. However, these approaches (essentially numerical, but also some experimental) in general did not attempt to describe the associated (macroscopic) rheological behavior.

Experimental studies that go beyond traditional bulk rheological studies have also shed light on the behavior of plastic materials. Microscopy studies of monolayers of particles at an oil-water interface [66], concentrated emulsions [67], non-Brownian suspensions [68], and an amorphous bubble raft [69] have all shown that particles follow closed trajectories in oscillatory shearing below some yield threshold, while the trajectories are open orbits when the yield conditions are exceeded. The

observation of acquisition of plastic strain has therefore been clearly linked to rheological yield conditions. An interesting set of observations from microscopy studies has led to the conclusion that soft plastic systems can have their structures ‘trained’ by oscillating at some small strain amplitude before being read out at some later stage by sweeping the amplitude of straining from small to large [68–70]. These have also shown how the presence of noise in the system can allow for multiple memories to be encoded in the structure and successfully read out after a delay.

In addition to microscopy studies, scattering of light and X-rays has been used to investigate the dynamics of concentrated colloidal suspensions under oscillatory shearing, using dynamic light scattering and X-ray photon correlation spectroscopy, or rheo-DLS [64] and rheo-XPCS [71,72]. These techniques allow for structures to be correlated before and after some deformation protocol, so that plastic rearrangements can be directly determined. It was shown that below a yield threshold the structure recorreates, as expected, each period [73]. However, once the yield threshold is exceeded, the structural correlation does not immediately fall to zero, but rather drops over subsequent oscillations. Petekidis and coworkers’ rheo-DLS study [64] even observed non-zero correlations at long times. These observations indicate that yielding is heterogeneous and is consistent with the picture that there exists a distribution of local energy potentials. Further, the significant correlation observed by Petekidis et al at long times is evidence that rheological yielding does not imply that all parts of the material are flowing homogeneously.

4. Comparison between experimental results and models

We now address some of the methods and assumptions that underlie many of the comparisons between the predictions of Oldroyd’s model and experimental results. Comparisons abound between experimental data and model predictions in the liquid regime. Such comparisons have led to ever more accurate descriptions of the steady-state liquid behavior, as exemplified by a recent reformulation of the Hershel-Bulkley description of the liquid regime by Caggioni and Spicer [74]. Although the need for describing the rheological behavior in both regimes has progressively emerged from the lack of agreement between steady-state models and transient experimental data, the rheological behavior of YSF in the solid regime had not been the subject of much attention until the 2000s. The very nature of such studies is based on the ability to clearly define the solid regime, and when it ends. This, in turn relies on the ability to determine when yielding happens, or at least identify that yielding has happened. Given the variety of ways in which the yield point has been identified, there are a number of behaviors that may be ascribed by some metrics as being in the pre-yielded solid state, while not by other measures. A few works at the beginning of the 2000s looked at some characteristics in the solid regime of YSF: the solid-liquid transition [75], creep flow [53], aging and creep [24]. At the same time, more sophisticated models [76] were proposed, but without possible direct measurements of the parameters. As soon as one considers that YSF clearly exhibit a “solid regime” such models can in fact naturally be inspired from models developed for solids or soils [40,76].

A detailed description of the rheological behavior and its physical origin could help understanding and describing the transition from the solid regime to the liquid regime and the plastic behavior. The problem we wish to address here is the measurement of these properties, and more specifically the issue of the measurement of these different stress or deformation components. With YSFs we have the great advantage over studies of solids that the same material can be tested repeatedly as their behavior is fundamentally reversible. With careful consideration of shear histories, it is indeed possible to employ well-defined pre-shear steps that place YSFs in reproducible initial configurations, allowing for multiple yielding experiments to be carried out on the same sample.

One of the simplest tests that can be considered is a creep test, which is the standard test in the field of solids, and which consists of an imposition of a constant stress or an increasing stress ramp, and

follow the resulting deformation. A related component of the creep test is the recovery test, which is performed by removing the stress after some creep time and following the amount of deformation that is recovered elastically [77]. The recoverable strain is reminiscent of the comment made by Oldroyd noted previously, “the displacement is taken from the equilibrium position which it would attain if, at a suitable time t_0 , the motion were instantaneously stopped and the stresses remaining at zero velocity were reduced reversibly to zero”. It is possible to interpret Oldroyd’s note as relating directly to recoverable strains.

The first question to address relating to creep tests is how the resulting deformation should be interpreted. The evolution of the deformation in time for a given applied stress is typically interpreted in the following manner: the elastic and plastic components (that is, the recoverable and unrecoverable components) lead to an instantaneous deformation with no further deformation in time, while a delayed deformation can be associated with a viscous component. In practice, it seems that for some YSFs, such as a concentrated emulsion, no such viscous component is observed, whereas for other materials, such as Carbopol gels, we observed some slight residual motion in the solid regime suggesting the existence of some viscous component [78]. However, even when such an effect is observed we can remark that: i) the viscosity associated with this slow flow is rather high, ii) the apparent strain rate continuously decreases in time, so that no steady state flow can be considered to be reached, and, consistently, iii) the total deformation tends to saturate, i.e. remains below a critical value. Such observations allow us to distinguish between the solid and liquid regimes. In the liquid regime, a steady state flow is reached, and the deformation tends towards infinity.

The plastic and elastic components should thus ideally be studied over short times, i.e. before some residual viscous flow modifies the deformation values. In that case the stress vs strain curve is generally concave, i.e. the resulting strain increases faster than the stress. However, this does not tell us anything about the elasticity and plasticity of the system. The elasticity is strictly related to the reversibility of the deformation, and not about the linearity of the curve, although generally some correlation may be observed. This means that the elastic and plastic components can be measured by releasing the stress and observing the strain recovery, as shown in Figure 1. The recovered strain is the elastic component, while the non-recovered part is the plastic component. Recent systematic measurements of that type with YSF of various structures showed that the plastic and elastic components increase simultaneously, from very small deformations, both with the square of the shear stress [78], as shown in Figure 2.

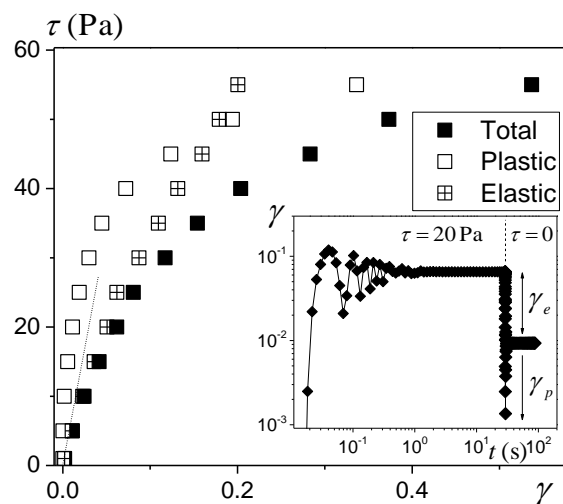


Figure 1: Shear stress vs induced deformation during recovery tests (stress release) for the emulsion: elastic (reversible) component (crossed squares), plastic (irreversible) component (open squares), total deformation (filled squares). The dotted line is the equation $\tau = G_{\infty}\gamma$, with G_{∞} taken as the constant G' (490 Pa for the emulsion) in small

oscillations tests. The inset shows a typical recovery test: deformation induced during creep test then deformation recovery for stress release, allowing to distinguish the two (elastic and plastic) components. *Reproduced with permission from [76]*

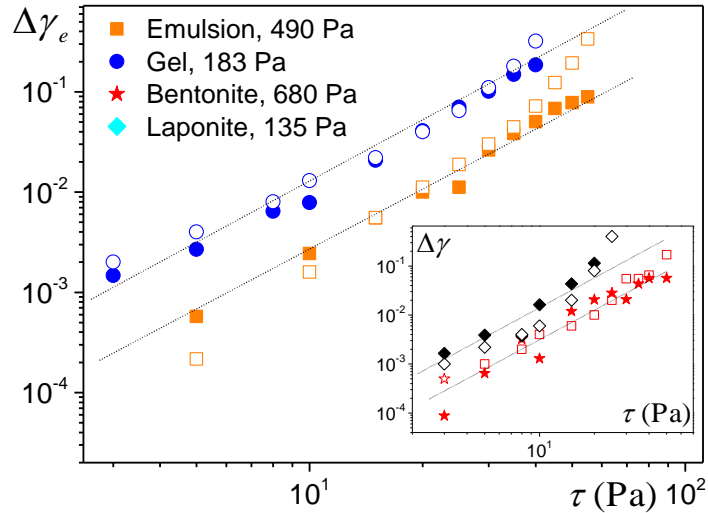


Figure 2: Additional elastic deformation (filled symbols) with regards to the deformation of the constant elastic network, and plastic deformation (open symbols), as a function of the stress applied for different materials (same type of data in the inset). The dotted lines (of slope 2) corresponds to the model fitted to data (see text) with $\alpha = 0.8$ (emulsion), $\alpha = 1$ (gel), $\alpha = 0.5$ (colloid (laponite)), $\alpha = 1.2$ (clay (bentonite)). *Reproduced with permission from [76]*

It was also suggested that further information on the structure behavior in the solid regime could be obtained by superimposing a stress oscillation of low amplitude to the main stress during a creep test. In that case it was observed for YSF of various structures that the solid state of the material is associated with a persistent elastic network of constant elastic modulus up to yielding (solid-liquid transition), while progressively more additional elastoplastic elements are involved.

Another simple rheological test that has been performed a lot on YSFs is that of steady-shear start-up tests. In this protocol, a material that is initially at rest is subjected to a constant shear rate at some start time, and the resulting stress is measured. In this protocol, an Oldroyd material would respond initially perfectly elastically, with the stress being proportional to the imposed strain, independent of the rate at which it is being imposed. Once the yield condition is exceeded, the response would immediately transition to the liquid response, where differences in the imposed rates would become clear. As noted by Saramito, the stress would jump from the value determined by the modulus and the yield stress to the value set by the plastic viscosity and the imposed shear rate. That this is never observed experimentally points to the limitations of applying an Oldroyd-type model, which assumes a quasi-static approach to the yield condition, to transient experiments where the point is specifically to violate the quasi-static assumption in a controlled manner.

Oldroyd's model has another prediction that was not acknowledged in his 1946 paper, but which has been used extensively in the literature since. In addition to employing a yield stress tensor, his use of a single modulus to describe the solid regime provides a prediction of the yield *strain*, $\epsilon_y = \tau_c/\mu$. This yield strain has been used to rescale steady-shear start-up experiments, which might otherwise be displayed as a function of time. By plotting the resulting stress as a function of the total applied strain,

many researchers have observed a nearly constant yield strain at small applied shear rates, with some observations of rate-dependent yield strains at higher rates [79–81].

In addition to applications of constant stresses or strain rates in creep and steady-shear start-up tests, many researchers have used oscillating stresses or strains to probe the yielding conditions [26]. In such tests, the applied stress or strain is oscillated at a particular angular frequency, ω , while the amplitude of the stress or strain is swept. Such experiments have been variously referred to as amplitude sweeps or LAOS experiments to reflect the large amplitude oscillatory shearing that is taking place. The convention of referring to LAOS experiments or to amplitude sweeps is typically associated with the method by which the results are displayed, though the experimental protocols are identical. Amplitude sweep data are typically presented in terms of the amplitude dependence of the dynamic moduli, G' and G'' , and rarely as the amplitude dependence of the dynamic compliances, J' and J'' [82]. These representations are commonly reported in commercial rheometry software and are also often the first figures used to situate LAOS studies. It is widely acknowledged that the dynamic moduli and compliances represent only the average responses during the experiments and are therefore incomplete representations [83]. In contrast, LAOS studies typically present some analysis of the full transient response.

Given the increased interest in large amplitude oscillatory shearing, it has now become common to refer to oscillatory linear viscoelastic measurements as being small-amplitude oscillatory shearing, SAOS. SAOS is limited to the case of small amplitudes, where the dynamic moduli or compliances are independent of the applied amplitude and has been used for over 70 years to measure the linear viscoelastic behaviors of polymeric and colloidal materials [84,85]. SAOS studies of YSFs are more recent and were not in existence at the time of Oldroyd's writing. It is well-known that the dynamic moduli represent the average energy stored per cycle and that the loss modulus represent the average energy dissipated per cycle, giving them their names [86]. In SAOS experiments, YSFs typically present dynamic moduli where both components are measurable, and the storage component is much larger than the dissipative component, sometimes by a decade or more. So we can consider that under oscillations of sufficiently small amplitude the material remains in its solid regime, but also that below the yield condition, most YSFs have some energy dissipation mechanisms that deviates from the ideal elastic response assumed by Oldroyd.

Dinkgreve et al. have gone into some depth in ways of experimentally determining the yield stress, as well as the yield strain, and present numerous measures that have been reported in the literature just for amplitude sweeps [26]. One of the cases we wish to highlight here is the determination of a yield strain, typically from the strain amplitude at which the dynamic moduli cross. In these experiments, as before, Oldroyd's assumption of a quasi-static approach to the yield point is violated. While in steady-shear start-up experiments the violation is driven by the choice of the shear rate that is applied, in the case of oscillatory experiments it is the choice of angular frequency that dictates how quickly the yield condition is approached. Higher frequencies violate the quasi-static assumption more than do low frequency experiments. Finally, Dinkgreve et al. noted that the yield strains determined from amplitude sweeps were consistently larger than those determined from other protocols that approach the yield condition more slowly.

Understanding LAOS by tracking the behavior *during* an oscillation has been intensively studied in the past decade. Numerous approaches have been proposed [83], and while discussion continues as how best to extract all the information contained in such responses, a few observations are consistent across different YSFs. A range of authors studying different YSFs have observed that at large amplitudes, the materials responses can be clearly broken down into a sequence of processes. It is commonly observed that a predominantly elastic response pre-yielding gives way to plastic flow over some short interval [87] after which nearly pure plastic deformation is observed. This cycle is repeated twice per period, and begins when the strain is reversed, or equally when the rate drops to zero and

reverses direction. During such a process, using the sequence of physical processes analysis, the predominantly elastic pre-yielding response of a soft colloidal glass has been shown to mimic the viscoelastic response in the linear regime [88], with similar amounts of energy storage and dissipation observed momentarily under LAOS.

Recently, a hybrid study from Donley et al [89] on a model YSF, Carbopol, coupled the LAOS/amplitude sweep tests with the zero-stress recovery steps typically associated with the steady creep tests explored in Figures 1 and 2. The Donley et al. study employed an iterative rheological protocol, shown in Figure 3, that provided information regarding recoverable and unrecoverable strains during an oscillation allowing for the explanation of all the features of the amplitude sweep data. The transient knowledge of how much deformation is recoverable elastic strain, and how much is unrecoverable plastic strain allowed for the identification of two distinct components of G'' . One component is determined by the rate at which recoverable strain is acquired and was called G''_{solid} , while the component proportional to the rate at which unrecoverable plastic strain is acquired was called G''_{fluid} . By making a detailed series of measurements across an amplitude sweep, Donley et al showed that the overshoot in G'' is associated with the acquisition of unrecoverable strains. It was further found that at stresses and strains below the yield condition determined from steady shearing that the Carbopol gel displayed measurable amounts of plastic deformation, and therefore had measurable values of G''_{fluid} across the entire range of amplitudes tested. A summary of these results is presented in Figure 3. It should be noted that the Donley et al. work, like many other studies that employ transient protocols to study yielding, was violating the quasi-static assumption mentioned previously.

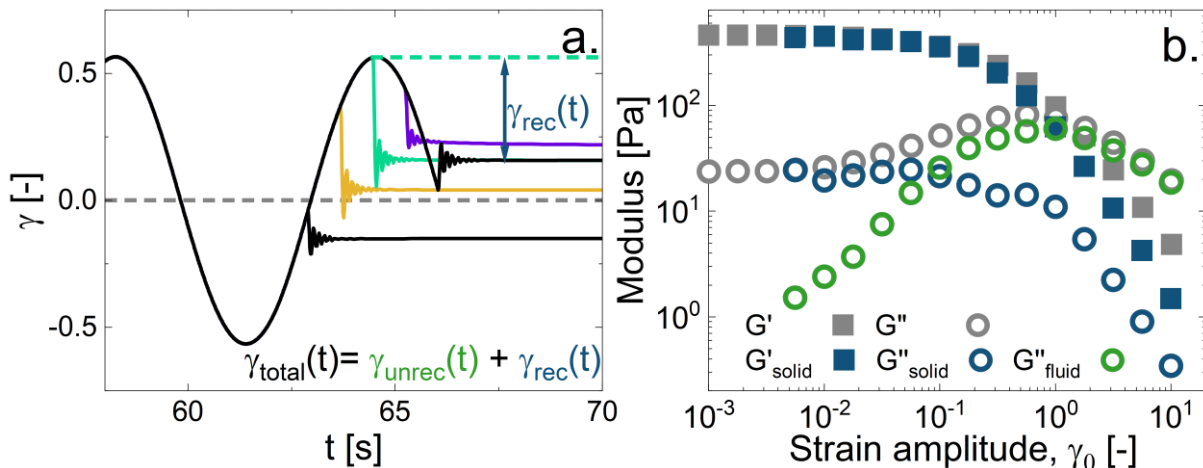


Figure 3 : Oscillatory shear and recovery tests on a Carbopol gel (a) are performed like a regular oscillatory shear test but with an iterative imposition of zero shear stress recovery tests. This protocol leads to transient measures of the recoverable and unrecoverable strains during oscillatory shearing that allow for the definition of two contributions to G'' : one each from the recoverable strain rate (G''_{solid}) and unrecoverable strain rate (G''_{fluid}). These measures can be used to show that the overshoot in G'' observed in an amplitude sweep comes from contributions from unrecoverable strains (b). Yielding, as interpreted as an acquisition of unrecoverable plastic strain, therefore occurs at amplitudes much smaller than typically assumed from the crossover point or the maximum in the loss modulus.

5. Conclusions

Oldroyd's model, published in 1946, has had a significant and lasting impact on our understanding of yield stress fluids. We delineate three main contributions from this paper. His separation of the

behavior into a pre-yielded elastic regime and a yielded plastic regime was the first time someone had addressed how YSFs behaved prior to yielding. His proposal of a full tensorial version of the model allowed decades worth of studies to use his formulation unchanged to examine the behaviors of YSFs in complex geometries. Further, he laid out the condition under which yielding takes place, which is consistent with the form of the constitutive equation in both regimes. The (von Mises) criterion adopted states that yielding occurs when the potential energy per unit volume due to deviatoric stresses reaches a critical value.

Oldroyd progressed through his study under a very clear assumption of a quasi-static approach to the yielding condition. This, above all his other assumptions, seems to be the least well-known. As it is very rarely, if ever, acknowledged, it may be easy to suggest that Oldroyd's model is critically flawed because it does not describe the transient nature of yielding well. While the model does not tell us about the process of yielding, it appears that the transient nature of yielding was never the goal of this particular formulation. Rather, the focus was always on describing the steady-state behavior of YSFs in a fully tensorial manner.

Oldroyd's ideas have greatly influenced generations of experimental, computational, and theoretical soft matter scientists and can still be seen in many modern models and experimental studies. Each of his three main contributions continues to be tested and improved upon using a range of experimental and computational techniques that continues to uncover the complexity of yielding and of yield stress fluids.

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