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## Bingham's heritage

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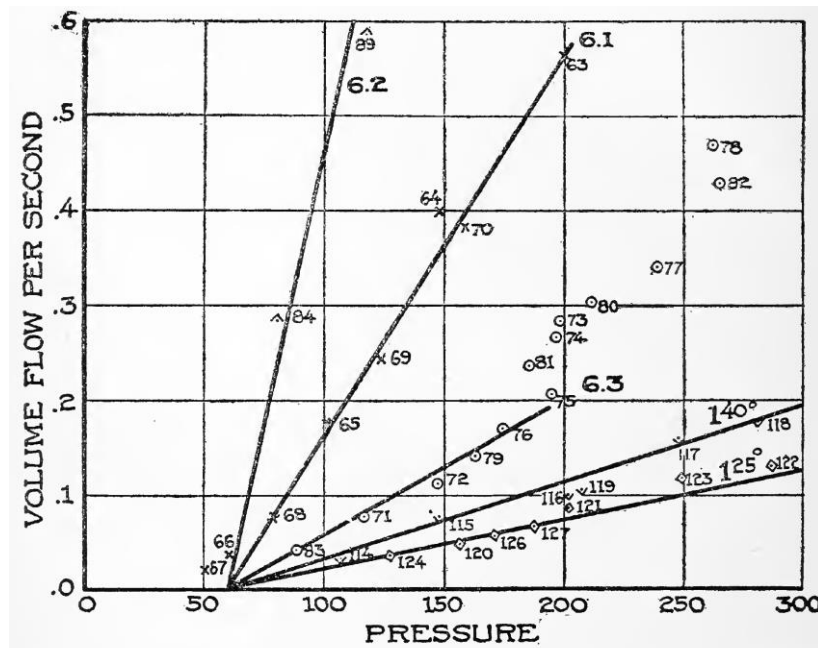
**Abstract:** A hundred years after the founding paper by E.C. Bingham, we briefly review the impact of the yield stress concept and current interest in it in the scientific community. We show that yield stress fluids have only emerged as a relevant fluid type, in both mechanics and physics, over the past twenty years, opening the way to a broad range of new study areas.

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#### 1. The founding paper

When considering the concept of a material which appears to have either a solid or a liquid behavior depending on circumstances, one generally refers to the book published by Bingham in 1922, i.e., *Fluidity and Plasticity* (McGraw Hill, New York, 1922). Actually, as early as 1916, E.C. Bingham, former professor of chemistry in Richmond, Virginia (until 1915), then assistant physicist at the Bureau of Standards in Washington, and so far essentially working on the improvement of viscosity measurement and on the viscosity of solutions, mixtures, and emulsions, published a paper entitled “An investigation of the laws of plastic flow” (Bulletin of the Bureau of Standards, 13, 309-353). In this paper Bingham presents, probably for the first time, the concept of yield stress fluid. The paper shows a series of flow experiments in capillaries of different sizes with kaolin-water suspensions (“English China clay”). It appears that the pressure needed for flow to take place must exceed a critical value, and then, in his range of flow rates, the flow velocity approximately increases in proportion to the difference between the current pressure and this critical value (see Figure 1). Bingham thus concludes that the stress between adjacent material layers gliding over each other during flow contains a constant “friction term” plus a term depending on the velocity. As he himself recalls, since 1864 and the first publications by H.E. Tresca on solid metal flows (Osakada 2010) scientists were already aware of the existence of plastic materials that could be deformed at will beyond some critical stress, and thus flow. The originality of his work was to demonstrate that such materials can flow at an increasing velocity as the stress increases beyond this critical value.



**Figure 1:** Key figure of Bingham’s paper: Flow rate ( $\text{mL}\cdot\text{s}^{-1}$ ) as a function of the pressure (in  $\text{g}\cdot\text{cm}^{-2}$ ) for a clay suspension flowing in capillaries of different diameters. The small numbers correspond to the test number, and the large number to the capillary diameter (see Bingham’s paper for details).

One could imagine that a hundred years ago such an original observation would be essentially phenomenological. Reading this manuscript it is impressive to see that Bingham goes much farther than the basic phenomenological observation. Indeed, he analyzes different aspects of the physics of this flow. First he provides a physical origin for this friction term, which up to now remains the basic explanation for the shear-thinning tendency and thixotropy of yield stress fluids (Michaels et Bolger 1962, Firth and Hunter 1976, Wildemuth and Williams 1985, Tsenoglou 1989,1990, Coussot et al. 1993, Coussot and Ovarlez 2010, Mewis and Wagner 2012): the solid structure of the material finds its origin in a network of particles in interaction, extending throughout the sample; as a result of large deformations (and then flow), this structure breaks into smaller structures, but reforms larger structures when the smaller ones come into contact as a result of material flow, until they reach on average an “equilibrium” size. Moreover, Bingham, probably wondering whether such a “discontinuous” material could be considered as a fluid, discusses the local structure of the flow, to conclude finally that it can indeed be considered as a continuous medium. Remarkably, Bingham also suggests that various other problems should be considered with this type of material, including wall slip, shear-banding, and phase separation, which are all very “modern” problems (see below).

Note that the term by which one should refer to such materials is uncertain in Bingham’s paper. He obviously did not call them “Bingham fluids”, and did not in fact suggest any term himself. This hesitation over the most appropriate term remained until the 90s (see below): Bingham solids, Bingham plastics, viscoplastic fluids, Bingham fluids, fluids with a yield stress, and finally yield stress fluids have all been used.

## 2. Status of this concept in physics and mechanics

The new concept is thus that there exist materials which are only slightly deformed when the applied stress is below some critical value, i.e. they are solid, and flow when the stress is beyond this value. In physics, flow is associated with liquid or gas state. As a consequence, from the point of view of physics this concept poses a problem, as it suggests that there exist materials which can be in either a solid state or a liquid state depending on the (shear) stress applied to them, whereas phase changes are usually considered to depend only on temperature and pressure (Tabor 1991). Moreover, the solid state in physics is associated with an ordered (crystalline) structure while the liquid state is associated with disorder. But we cannot rely on such a classification with yield stress fluids as (i) they are generally highly disordered: foams, cement, mud, paints, etc., are made up of many elements of various types and sizes suspended in a liquid; and (ii) depending on the stress applied, they behave either as solids or as liquids. In fact the disorder allows the structure to be deformable at will, while keeping (except for significant thixotropy) its mechanical properties whatever the deformation, since in the end the disorder, and thus the structure, can remain the same after a large deformation of the material.

On the other hand, in mechanics, this concept also poses a problem, in particular with regard to rational mechanics (Truesdell 1991), as developed by Walter Noll and Clifford Truesdell. Taking advantage of the usual classification in physics, but also on the basis of a natural classification of their mathematical properties, rational mechanics offers a rigorous and complete mathematical framework for describing the mechanical transformations of materials. It also suggests a separation of simple materials into two broad classes, *solids* and (*simple*) *fluids*, each associated with a specific simple mathematical property of its "peer group" (i.e., roughly speaking, the set of transformations of a material leading to indistinguishable materials) (see Truesdell 1991). According to these definitions, solids are materials which have a preferential, natural configuration, relative to which their behavior must be defined. More precisely, at any time the current stress tensor can be determined from knowledge of the current deformation relatively to the preferential configuration. Fluids are materials that do not have a preferential configuration; they may be deformed at will but then always relax. This means, for example, that if after any type of flow the material remains a sufficient time at rest, it eventually completely forgets its previous deformations, i.e., its final configuration does not depend on the previous flow history.

Because of its potentially interesting simplifications of the treatment of constitutive equations, theoretical rheometry (or viscometry) (Coleman et al. 1966) has so far been dealt with in the framework of *simple fluids*, which have a vanishing memory. These materials are such that, if one waits long enough after some reference time, they will have completely forgotten their history prior to that reference time. The establishment of mathematical tools for rheometry, including in particular the expression for the material functions in different flow geometries (see Coleman et al. 1966), relies on this assumption. It is particularly important to keep this in mind since it explains some unexpected difficulties we may have with yield stress fluids during rheometrical tests. Indeed, yield stress fluids are not simple fluids: they behave partly as solids, because for some flow histories they have a preferential configuration, and partly as fluids, because they are able to permanently forget this configuration after a particular deformation history, or may recover this initial configuration after coming to rest for a while. It thus appears necessary to distinguish a third (or intermediate) class of materials, which borrow their behavior partly from fluids and partly from solids. Finally, for such materials, rheometrical problems are treated as experimental artefacts, whereas in fact the usual material functions depending only on the shear rate, i.e. apparent viscosity and normal stress differences, have been clearly defined only under the assumption of simple fluid.

For example for a material being solid under some conditions these material functions are certainly not the appropriate way to describe the behavior. On the other hand, since a full treatment is rather difficult (see Coussot 2005), this may also be the best approach.

### 3. Short-term impact of the concept

In fact, this novel and problematic “intermediate” behavior of yield stress fluids would not appear to have particularly worried anyone working in these different fields up to the 80s (see below). It seems likely that they were still viewed as “exotic”, not deserving any particular scientific interest. However, if we trust in the idea of natural scientific development, we may also suggest that scientists at the time considered it more important to develop the theoretical foundations of mechanics for the two basic branches of materials, rather than picking up on these complex intermediate materials, which could not be classified in the usual way, either in physics or in mechanics.

Maybe the greatest impact of Bingham’s findings in the first half of the 20th century was the emergence of the idea that the behavior of materials was something more complex than had been previously thought, at least as treated within the standard theoretical framework, whence all these materials which differ from simple liquids, gases, and solids needed to be studied with a specific scientific approach. This led to the creation of the first national rheology society, namely the Society of Rheology, and more importantly to the creation of a new word, associated with a new scientific field, i.e., rheology. In this endeavour, Bingham certainly played a major role, as in 1924, he could already make the following statement in the introduction to the first “Plasticity Symposium” at Lafayette College (Nadai 1947):

“Our discussion of plasticity therefore concerns itself with the flow of solids [...], for the Greek philosopher Heraklitus was literally correct when he said that “everything flows”. It is therefore necessary to limit our discussion by excluding the flow of those things which we are accustomed to refer to as fluids, the pure liquids and gases. But the circle of our lives is not concerned principally with the fluids, even air and water, but with plastic materials. Our very bodies, the foods we eat, and the materials which we fashion in our industries are largely plastic solids. Investigation leads us to the belief that plasticity is made up of two fundamental properties which have been named *yield value* and *mobility*, the former being dependent upon the shearing stress required to start the deformation and the mobility being proportional to the rate of deformation after the yield value has been exceeded.”

Then D. Doraiswamy (2002) gives the following description of what happened at the third Plasticity Symposium in 1929:

“The decision was made to form a permanent organization for the development of the new discipline of rheology. The preliminary scope of The Society of Rheology was set up by a committee which then met on April 29, 1929 at Columbus, Ohio, and some of the luminaries who participated in this pioneering event included Eugene C. Bingham, Winslow H. Herschel, Marcel Brillouin, Herbert Freundlich, Wolfgang Ostwald, Ludwig Prandtl and Markus Reiner. The name ‘rheology’ was proposed to describe ‘the study of the flow and deformation of all forms of matter’ by E.C. Bingham and M. Reiner; Heraclitus’ quote ‘παντα ρει’ or ‘everything flows’ was taken to be the motto of the subject (Reiner 1964).”

Paradoxically, considering that he was the main founder of the society, the motto chosen seems somewhat in contradiction with Bingham’s findings. In fact, as explained during this meeting, the

word “flow” in the motto must be understood in the larger sense of deformation, to include all material types.

#### 4. Life of the concept

Was the new concept fully accepted? This is not so clear if we note the celebrated attack against it by H.A. Barnes and K. Walters in their 1985 paper entitled “The yield stress myth” (Barnes and Walters 1985). This paper claimed that, in materials considered so far as “Bingham plastic materials”, one could observe very slow flows for very low stress, which meant that they did not exhibit a true yield stress. In fact several experimental aspects were unclear in this paper, such as (i) the existence or otherwise of wall slip, which, as is now well known, can lead to an apparently Newtonian or power-law behavior for a range of stresses below the yield stress (see, e.g., Bertola et al 2003, Meeker et al 2004), and (ii) the fact that the total deformation induced by an extremely low shear rate (typically as low as  $10^{-5}$  or  $10^{-6}$  s $^{-1}$ ) over a reasonable experimental time is so small that one could hardly expect to reach the liquid regime if the material was initially in its solid state (Coussot 2005). And in fact the apparent viscosity in the solid regime tends to infinity with time (Coussot et al 2006, Moller et al 2009), which means that this is not simply a question of Deborah number, as suggested by Reiner 1964; for a stress below the yield stress the deformation seems to have an upper bound, and mountains flow because they are submitted to sufficiently large stresses.

However, the publication of this paper caused some agitation in the scientific community, and a series of papers questioned the validity of this assessment, most of them published as letters to the editor of the Journal of Rheology in the early 90s (Hartnett and Hu 1989, Schurz 1990, Astarita 1990, Schurz 1992, Evans 1992, de Kee and Fong 1993). The fundamental idea which emerged from this debate is that the yield stress is an “engineering” reality: various pasty materials in our everyday life can flow like liquids if submitted to sufficient stress, while they can keep their shape over long times (at least as compared to our usual time of observation) if they are left under the action of a small stress such as can be induced by gravity under some conditions. A nice straightforward demonstration was also provided by J.M. Piau (personal communication): take a hair gel bottle, which usually contains some bubbles, a few millimeters in diameter, suspended in the gel. This gel can flow like a liquid if mixed with a spoon. But if left at rest, the bubbles, although much less dense than the gel, will maintain their position (or remain very close to it) for years. This means that the gravitational force due to the density difference can only impose an extremely slow flow with completely negligible practical impact, especially when compared to the velocity of displacement observed if the critical stress is overcome.

In the end, the question of whether or not there is steady flow below the yield stress is essentially a philosophical one. However, this debate probably had two important impacts on the scientific community. The first was to throw doubt on the effective existence of a yield stress, and in particular for scientists not directly working in the field, to suggest that, since even the experts did not agree on the effective properties of such materials, it would be difficult to get reliable data or measure something that would then be recognized by all concerned. On the other hand, a more constructive consequence of this debate is that it has encouraged people working in the field to reinforce their experimental characterization and be as clear as possible about what they are measuring.

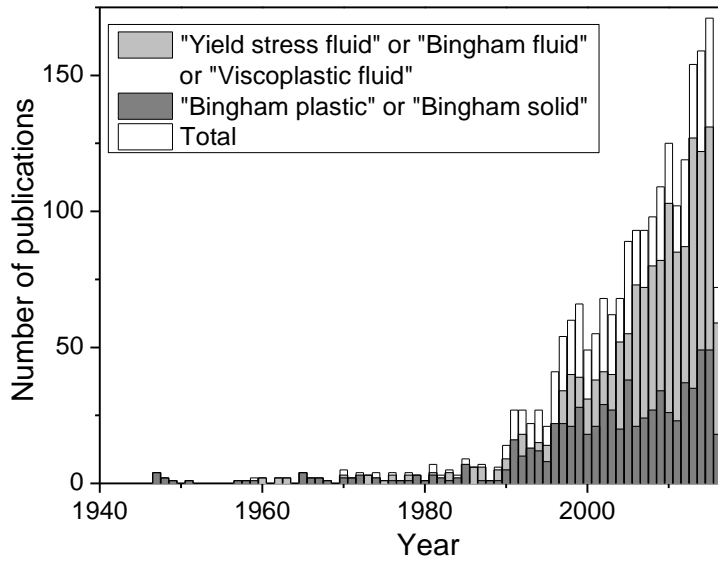
Independently of this controversy, various studies have been carried out in the field of yield stress fluids since the origin of the concept. However, contrary to what one would expect for a new and useful concept, Bingham’s findings and the creation of the Society of Rheology did not lead to an

explosion of publications concerning yield stress fluids. Instead, the number of studies in this field remained rather limited during the first 70 years.

Our modern tools for monitoring references can provide us with an overview of the evolution of the studies carried out in any given field over the last hundred years. Here we will refer to Web of Science (Science Citation Index of Thomson Reuters' Web of Science). First of all, it appears that the term "yield stress fluid" is a rather recent one. Apparently, the first publication mentioning this expression either in the title, in the abstract, or in the key words, is the one by Nguyen and Boger 1987: "Characterization of yield stress fluids with concentric cylinder viscometers". Before that the expression "fluid with a yield stress" had already been employed, first by Hanks 1963 in "The laminar-turbulent transition for fluids with a yield stress", while the expression "viscoplastic fluid" was employed by Andres 1960, in "Equilibrium and motion of a sphere in a viscoplastic fluid", and the expression "Bingham fluid" is even older, first used by Hirai 1959 in "Theoretical explanation of heat transfer in laminar region of Bingham fluid".

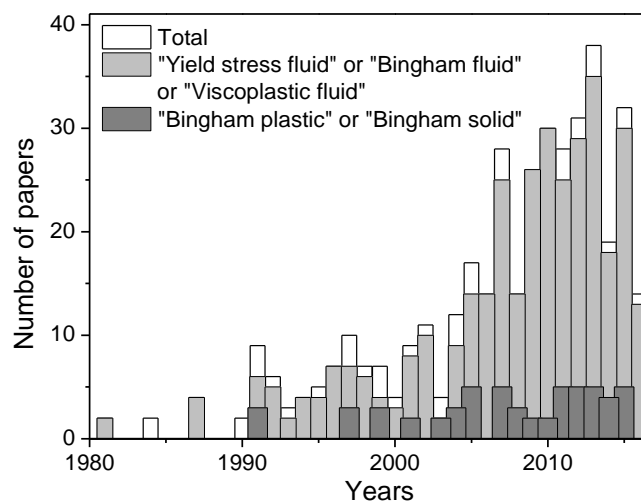
Thus the association of the word "fluid" with a concept of yielding or plasticity appeared only rather late on, almost 50 years after Bingham's paper. Before that, it seems that the tendency was to refer to "Bingham solids" or "Bingham plastics". It was J.G. Oldroyd who apparently inaugurated this usage in his two papers: "A rational formulation of the equations of plastic flow for a Bingham solid" (Oldroyd 1947), and "Rectilinear flow of non-Bingham plastic solids and non-Newtonian viscous liquids" (Oldroyd 1949). Before the latter publications no expression seems to dominate reference to such materials.

Actually the publications concerning yield stress fluids, whatever name was used to specify them, were rather scarce for a long time. After a period with numerous fundamental publications by Oldroyd around the 50s, it was only at the beginning of the 60s that the number of publications on this subject began slowly to increase (see Figure 2). Finally, the publications on yield stress fluids (referred to by any of the above-mentioned terms) started to grow significantly at the beginning of the 90s, and at a strong rate. However, it must be kept in mind that this growth may be partly explained by different factors, independently of a simple interest in such materials, e.g., an increase in the number of referenced publications (in particular conferences), an increase in the number of journals, an increase in the number of publications per author, an increase in the number of people publishing around the world, all factors that are difficult to take properly into account. A quick overview shows that the number of publications also grew more or less in the same way in other fields of rheology (considering all journals). But at least we can conclude that yield stress fluids constitute a lively field, as the number of publications there is growing rapidly, and the terms involving "fluid" to describe these materials have become more frequent than "Bingham solid" or "Bingham plastic" (see Figure 2). The latter point shows that yield stress fluids are gradually becoming more commonly accepted as fluids, and that their properties are most often studied by people working in that field.



**Figure 2:** Number of publications per year concerning yield stress fluids, named in different ways.

Another way to assess the relative importance of yield stress fluids and the way it has evolved in scientific studies is to focus on publications in the main rheology journals (*Journal of Rheology*, *Rheologica Acta*, *Journal of Non-Newtonian Fluid Mechanics*) since the 70s. The total number of papers in these journals per year has remained remarkably stable since 1978, i.e., it stands at around 280, but the number of papers mentioning yield stress fluid behavior significantly increases from 1990, with a first jump to between 5 and 10 papers per year up to 2005, then a second jump to 25-30 papers per year since 2005 (see Figure 3). This means that 100 years after Bingham's paper and the subsequent foundation of the Society of Rheology, yield stress fluid behavior has become a major field of rheology. And we can note that the terms "Bingham plastic" or "Bingham solid" are now more marginally used in this area than terms involving "fluid".



**Figure 3:** Number of publications per year concerning yield stress fluids, named in different ways, among all publications of the main rheology journals.



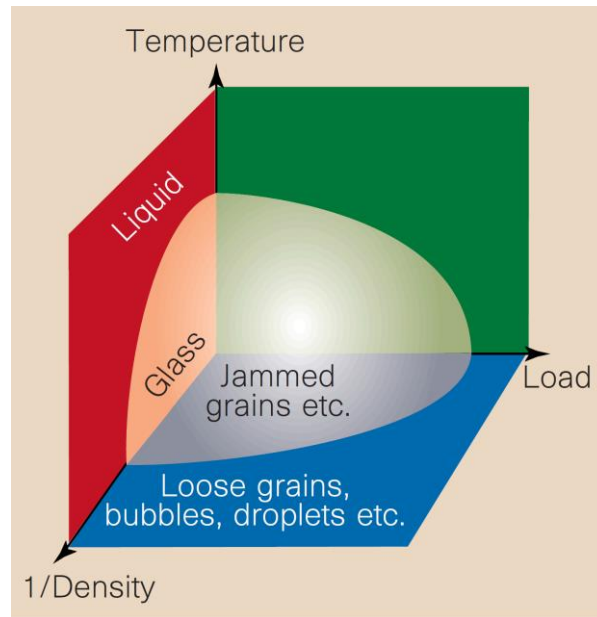
Another sign of this new interest in yield stress fluids is the setting up of a biennial international conference on viscoplasticity, initiated by Ian Frigaard and Neil Balmforth, with a first meeting in Banff (Canada) in 2005. These conferences have been extremely successful and now currently bring together about 100 participants from all over the world, each giving rise to a special issue of the Journal of Non-Newtonian Fluid Mechanics or *Rheologica Acta*.

### **5. Yield stress fluids as “jammed systems”**

The progress of science has also finally led to the consideration of these “intermediate” materials in physics. A lot of interest was generated at the end of the 90s for what became known as “jammed systems”. In 1998, Liu and Nagel, generalizing the concept suggested by Cates et al 1998 for a specific class of material, suggested that a wide range of materials retain their structure under certain conditions, i.e., they jam, while this structure breaks for other conditions (i.e., they unjam). A new type of phase diagram was proposed for such materials, in which the three variables governing the material state are the stress, the temperature, and the reciprocal of the density (see Figure 4). These variables play an equivalent role with regard to the state of the material: in fact, it can unjam only beyond a critical set of values of these variables. This description is quite consistent with the basic property of yield stress fluids, whose structure breaks down only beyond a critical stress, and with our common observation in rheology that, for a colloidal system, a foam, or an emulsion, there exists a critical concentration beyond which the apparent yield stress grows rapidly from zero. Such a diagram is an extension of the conventional phase diagram for pure materials, allowing us to include materials that are intermediate between simple solids and simple liquids, through the introduction of an additional state variable, namely the stress. By this means, yield stress fluids finally gained recognition among the physics community.

This suggestion very likely stimulated a new interest in these materials. In particular, theoreticians came up with the idea that jammed systems might be strongly analogous with glasses as soon as the temperature is replaced by the stress, whence it became possible to derive modelling approaches by considering the evolutions of an energy landscape, as in the recent trap model developed for glasses (Monthus and Bouchaud 1996). This gave rise to the SGR (Soft Glassy Rheology) model (Sollich et al. 1997, Hébraud and Lequeux 1998, Fielding et al. 2000), which was able to predict various behavioral trends observed in practice.

Note that basically jamming is associated with a critical concentration of elements beyond which their spatial configuration cannot significantly evolve under the action of thermal agitation alone. Thus, this cannot be strictly associated with the formation of a percolating network of links or contacts between the elements, as one also needs to have a sufficiently strong structure formed.



**Figure 4:** Possible phase diagram for jammed systems. Reprint from Liu and Nagel 1998.

Recently, the Bingham approach found a new field of application. Although some of its mechanical properties were already well known in mechanics, in particular for quasi-static flows, granular materials have been the focus of much quite independent interest in physics over the last 30 years. Such materials were seen as a potentially new state of matter which might be described using a novel thermodynamics approach, since it can behave either as a solid or as a liquid, depending on the circumstances. Various novel behavioral trends, in particular in the liquid regime, have been observed (see, e.g., Andreotti et al 2013). For example, inertial flows, with such a high level of particle agitation that collisions are the dominant vector of stress transmission, have been successfully described by analogy with the kinetic theory of gases (see, e.g., Goldhirsch 2003). In contrast, flows with negligible inertia effects seem to be similar to the flow of yield stress fluids: they can form a deposit at rest on a certain slope and start to flow if the slope is increased beyond some critical value (see, e.g., Nedderman 1992, Coussot 2005). However, for such flows one aspect distinguishes the behavior of this type of material from that of a typical fluid: the pressure now plays a critical role in the behavior, since the force required to move two solid particles tangentially when they are in contact is proportional to the normal force exerted between them. Moreover, information about the apparent viscosity of such a material as a function of the flow rate remained limited for a long time, even though some scaling laws were found (Pouliquen 1999).

It was finally shown, only quite recently, that compact or non-inertial free surface granular flows may be described by a viscoplastic-type constitutive equation, in which the yielding term is taken to depend on a dimensionless number involving the pressure (Jop et al 2006). Thus the Bingham approach is the basic concept, but a refinement is needed in order to describe this specific type of behavior: the apparent viscosity is now a function of the pressure through a dimensionless (inertial) number proportional to the shear rate and to the reciprocal of the square root of the pressure. For slow flows, the stress tends to the yield stress, which is here proportional to the pressure.

## 6. Contemporary subjects of interest in the field of yield stress fluids

Here, to conclude this paper we look at the present situation by discussing some contemporary subjects of further research in the field of yield stress fluids. This review is certainly not exhaustive, there are likely various other research questions, more or less tightly connected to this field, which are ignored from the author.

### *Measurements*

The question of measurement is inherent in the determination of yield stress fluid behavior. It is indeed only through careful measurements that one can clearly identify the two regimes (solid and liquid) and the stress value associated with this transition, and finally conclude as to whether the material is a yield stress fluid, and if so, determine its yield stress value. The most precise technique, but also the most tedious one, consists in carrying out creep tests at different stress levels, and following the deformation vs time curves: below the critical stress, the deformation tends to saturate, and above the critical stress, it ultimately follows a straight line of finite slope. In theory, this makes it possible to clearly distinguish the solid and the liquid regime (Coussot 2005). However, even with this technique there is some uncertainty over the determination of the yield stress, since an ideal measurement requires one to be able to precisely measure the stress for which the flow switches from a plateau of deformation at long times to a constant increase in the deformation at an infinitely low rate. A simplified approach is to apply a constant, low shear rate and observe the stress plateau reached beyond a sufficiently large deformation. The level of this plateau is close to the yield stress, as long as the shear rate is sufficiently low. Another possibility is to impose a large shear rate and then decrease the stress or the shear rate down to a low value: the stress plateau at low shear rate corresponds to the yield stress. These different measurements provide a single yield stress value, as long as the material is not thixotropic (Ovarlez et al 2013). A few years ago, a new technique appeared, known as LAOS (Large Amplitude Oscillations Shear) (Hyun et al 2011). This exploits the principle of oscillating flows, which proved so useful for viscoelastic liquids, as a way to investigate the complex behavior of yield stress fluids. Here the large amplitude provides a way to escape from the solid regime, at least for part of the motion. However, during such a process, the material experiences a complex flow history made up of undetermined periods in the solid and liquid regimes. This significantly complicates the interpretation of data, but recent work has provided some possibilities for a qualitative and sometimes quantitative interpretation of such results (Rogers et al 2011, Dimitriou et al 2013).

For a thixotropic material the behavior depends on the shear history: the structure breaks with flow and does not immediately recover. As a consequence there is a discrepancy between the yield value associated with flow start from rest ("static yield stress"), i.e., from a solid state, and the value associated with stoppage from the liquid regime ("dynamic yield stress") (Kraynik 1990, Bonnecaze and Brady 1992, Cheng 2003). Under these conditions the yield stress is not unique, but depends on the procedure used to measure it.

Actually, a lot of work has tended to be more focused on the yield stress determination than on other aspects of the behavior. There are several reasons for this:

- The yield stress value is the main indicator of yield stress fluid behavior: if it is equal to zero, the material is not a yield stress fluid.
- In many flow situations the yield stress is the dominant term in the stress expression, which means that the viscous dissipation (i.e. mechanical work per unit time) can be roughly estimated solely from information about the flow rate and the yield stress value. More precisely, such a situation is encountered when the Bingham number, which is the ratio of the yield stress to any additional viscous effects, is sufficiently large; this means that the yield stress is the basic rheological parameter for such a material. The above description a priori concerns flows in the liquid regime, but we can

also consider the energy dissipated during a finite deformation in either the solid or the liquid regime. In that case this energy may be estimated as the yield stress times the total deformation.

- Since it is directly related to the strength of the internal structure resulting from the interactions between the components of the material, the yield stress value can be used to characterize the material obtained by mixing various components in a liquid (formulation).

- Since it is related to the critical force making it possible to switch from rest to flow, or the opposite, it underlies various situations of flow start or flow stoppage observed in practice. As a consequence, it may be roughly estimated from “simple” measurements, i.e., not requiring an expensive rheometer; moreover these techniques generally involve a large volume, much larger than the size of the suspended elements, so that the continuum hypothesis is respected, and that is not always easily achievable with conventional laboratory rheometers.

In fact, in industry, in particular in the food industry and in civil engineering, there has long existed a great number of tests aimed at determining certain critical conditions relating to flow stoppage or flow start with pasty materials. Typically, some volume of material is left to spread over a solid surface until it stops, and some characteristic length of the final shape of the material (thickness, extent) is measured. Another possibility is that some object is pushed against or through it, and the corresponding force measured (squeeze test, penetrometry). In industry these tests are in general not rationalized, i.e., the measurements are not interpreted in terms of an intrinsic rheological parameter of the fluid. However, in essence, Bingham’s initial publication was already concerned with the determination of the rheological parameters of the material, and in particular the yield stress, from simple tests. His initial suggestion was to use a series of capillaries of different sizes and/or lengths to estimate the yield stress (Bingham 1916, Bingham et al 1922). Since then, most tests have been the subject of research attempting to establish a theoretical correspondence between the quantity measured in practice for each test and the material yield stress: penetrometer (Kaufman et al 1939, Uhlherr et al 2002, Lootens et al 2009, Tikmani et al 2013), L-box (Khayat et al 2004, Nguyen et al 2006), Bostwick consistometer (Rao and Bourne 1977, Perona 2005, Balmforth et al 2007), slump test (Murata 1984, Rajani and Morgenstern 1992, Pashias et al 1996, Roussel and Coussot 2005, Staron et al 2013), squeeze test (Covey and Stanmore 1981, Sherwood and Durban 1996, Adams et al 1997, Meeten 2000, 2010, Rabideau et al 2009), inclined plane test (Coussot and Boyer 1995, Khayat et al 2010), ball displacement (Schatzman et al 2009, Kashani et al 2015), vane test (Nguyen and Boger work in 1987, Ovarlez et al 2011), etc. The scientific analysis of these tests has thus progressed significantly over the last twenty years. However, there is probably still a need to transfer this knowledge to industry, in order to define more appropriate boundary conditions and procedures for ascertaining intrinsic rheological parameters directly, the idea being that these will constitute more relevant references for characterizing pasty materials.

The measurement of flow characteristics is also a subject of discussion, as certain problems often seem to arise. Typically, these problems come from the fact that flow heterogeneities sometimes develop, as anticipated by Bingham. The first such was already identified a long time ago: this is wall slip, which, for yield stress fluids, is generally due to the fact that the material can remain in a solid state while moving over an extremely thin layer of liquid (the interstitial liquid of the material) sheared along a smooth wall. The impact of wall slip on rheometrical measurements is particularly significant for yield stress fluids, since it can give the impression that the material flows under very low stresses, in complete contradiction with the yielding behavior expected for a much larger stress value. At first, techniques aimed essentially at analyzing data to extract the intrinsic behavior of the material despite wall slip, or investigating the modification or disappearance of wall slip by changing

the wall surface properties (wetting, roughness) (Walls et al 2003), but there were scarce few attempts to quantify and observe this effect at a local scale (see a review in Coussot 2005). Recently, a series of careful experiments along with microscopic modelling has provided a full understanding and quantification of the phenomenon for model materials (Meeker et al 2004, Seth et al 2008, Seth et al 2012). The next step might be to generalize such a description to more complex materials containing various element types and sizes, in order to be able to predict wall slip occurrence in industrial processes.

Another effect of flow heterogeneity which can impair measurements is shear-banding. In that case several regions with different flow rates coexist in the material although the shear stress is homogeneous. The typical situation is the existence of an unsheared region in contact with a region that is sheared at a finite shear rate. In that sense this effect must not be confused with the simple coexistence of sheared and unsheared regions in yield stress fluid flows when a heterogeneous stress distribution is imposed (Ovarlez et al 2009). This shear-banding effect for yield stress fluids (and more particularly colloidal systems) was first observed by Pignon et al 1996 in a Laponite suspension, and since then by different groups for various materials (Raynaud et al 2002, Holmes et al 2004, Becu et al 2006, Ragouilliaux et al 2006, 2007, Jarny et al 2008, Rogers et al 2008, Gibaud et al 2009, Divoux et al 2012, Malkin et al 2012). It was considered as the result of thixotropy (Coussot et al 2002, Moller et al 2008): as a result of competition between destructuring due to imposed shear and spontaneous restructuring of the material, the material is unable to flow steadily below a critical shear rate (Coussot and Ovarlez 2010), because below this shear rate, restructuring is faster than destructuring, whence the viscosity increases, and the flow rate decreases, and so on until complete stoppage. Note that this effect in itself does not alter the static yield stress measurements more than the above-mentioned problem of the dependence on the flow history: as long as the thickness of the sheared band is much greater than the size of the suspended elements, the apparent yield stress for the flow to start is just what is required to break the structure at that time, whatever form this breakage takes. Note that things become more complex when the band thickness is of the order of the element size: the stress needed to impose a flow may be greater or less than the yield stress of the homogeneous material (Pignon et al 1996). Since shear-banding had previously been observed for micellar solutions, this subject has generated much theoretical interest in recent years (Fielding 2007, Dhont and Briels 2008, Olmsted 2008, Fielding et al 2009, Fielding 2014, Moorcroft and Fielding 2013).

These discoveries came about through research interest in the internal flow characteristics of yield stress fluids. This interest seems to have begun with the remarkable work by Magnin and Piau (1990), who observed the deformation of a Carbopol gel along the periphery at different (apparent) shear rates. From the beginning of the 2000s, the flow properties inside flowing yield stress fluids, and in particular the spatial distribution of solid and liquid regions, could be observed more systematically with ultrasound and MRI (Magnetic Resonance Imaging) velocimetry, the latter technique being the most suitable for any non-transparent material containing a liquid. With the help of such techniques, it became possible to determine directly the “local” constitutive equation of the material, i.e., local shear stress vs local shear rate, in a flow with a heterogeneous stress distribution: local measurements of the velocity field provide the local shear rate corresponding to the local shear stress (found from torque measurements, along with our knowledge of the theoretical distribution of the tangential stress in the material) (Raynaud et al 2002, Goyon et al 2008, Ovarlez et al 2006). Why were people so motivated by such observations with yield stress fluids? Actually, this is logical enough. We are dealing with materials which are solid under some conditions, and as a consequence

they are expected to break or localize the deformation when they yield, as for brittle or ductile solids; this is indeed the most natural situation before reaching a fluid regime in which the material flows homogeneously. With these techniques various problems of flow or material heterogeneities which typically occur for yield stress fluids (migration, yielding behavior, thixotropy, shear-banding, wall slip) can be clarified or solved whereas it is often much more difficult to study them through conventional (macroscopic) rheometry.

Finally, it has been suggested recently that, for homogeneous flows of yield stress fluids, there may exist a “confinement” effect when the characteristic thickness of the flow is not much greater than the element size (say typically less than 50 times). In that case the apparent flow curve of the material may differ significantly from what is measured with greater flow thicknesses (Goyon et al 2008, Géraud et al 2013). Roughly speaking, this is because, due to the limited volume available to the jammed structure, the relative motions of its elements are rather restricted. This has led to the development of a new modelling approach, called the “fluidity model” (Bocquet et al 2009), and many recent theoretical and numerical studies (e.g., Benzi et al 2016, Nicolas and Barrat 2013) to describe such situations.

### *Modelling*

Considering the limited range of shear rates covered by the original Bingham data, the initial model proposed by Bingham to represent his data was the most natural one. This model is indeed sufficient to get a good representation of data in a range of the order of one decade of shear rates. Actually, this is also the most natural model when one expects the different terms of the constitutive equation to be able to represent distinct physical phenomena, but unfortunately, this is not the case. First of all, when data are obtained over a wide range of shear rates, typically covering four decades of shear rates, the Bingham model does not provide a good representation of the data. One must then use a HB (Herschel-Bulkley) model, as suggested by Herschel and Bulkley in 1926. Secondly, although the yield stress corresponds to the critical stress required to break the structure, which is thus a clear physical trend, the additional term in the constitutive equation does not in general correspond to a distinct physical effect, such as the additional stress required to move the suspended elements relative to one another after structure breakage; the sum of the two stress terms generally reflects the total viscous dissipation associated with the complex relative motions of elements during flow. Note that in order to represent the tendency of the second term of the constitutive equation to decrease for increasing shear rate using only two parameters (instead of 3 for the HB model), the Casson model has been widely used in the food industry (Missaire et al 1990, Rao and Cooley 1992, Wilson et al 1993), but it is no better placed to represent the data over a wide range of shear rates. Finally, although Bingham was the initiator of the concept and the basic model, we now know that the Bingham model generally provides only a very rough approximation, and that the HB model is more appropriate to represent the flow characteristics of yield stress fluids in a consistent way.

The initial Bingham approach and the above discussion assume a simple shear. However, proper modelling of complex flows requires a 3D constitutive equation. The basic form of the 3D expression which is used nowadays was provided by Oldroyd (1947), with a subsequent modification to take into account the power-law variation of the additional viscous term. It assumes that yielding occurs when the limit of a von Mises criterion is reached (i.e., the second invariant of the stress tensor is equal to a critical value), and in the liquid regime the stress is simply proportional to the strain rate tensor via an apparent viscosity that is formally similar to the one in simple shear (i.e., HB model), but with the

second invariant of the strain rate tensor now replacing the shear rate. The global validity of this 3D constitutive equation has been checked to some extent by comparing simulations and experiments (Rabideau et al 2010), but its full validity remains to be proved (see Coussot 2014). A straightforward means would be to look at pure elongational flows, but so far it has not been possible to obtain such flows experimentally: when a small heap of yield stress fluid is stretched between two solid plates, it soon separates into two conical parts and the corresponding flow does not at all correspond to a pure elongation (Boujlel and Coussot 2013). Besides, in order to explain some discrepancies between the HB model predictions and experimental observations it was suggested to use an elasto-viscoplastic model, the main aspect of which being to introduce some significant elastic component in the liquid regime. The modelling approaches in that field were recently reviewed by Fraggedakis et al. (2016) and concluded at the superiority of the Saramito (2009) model that combines both the Oldroyd viscoelastic model and the HB model and is derived to satisfy the second law of thermodynamics. In several cases it was also considered that the viscoplasticity (with thixotropy) could be seen as a special case of viscoelasticity with a flow history dependent structure parameter (Coussot et al (1993), De Souza Mendes (2011), Renardy and Renardy (2016)). However, relying on a framework adopted from plasticity, it was also suggested to base the description of the behavior of such materials on an additive strain decomposition into characteristic reversible (elastic) and irreversible (plastic) contributions, including the concept of kinematic hardening. This approach proved to be able to capture all most important features of non-linear rheology of Carbopol gels and waxy crude oil (Dimitriou and McKinley 2014).

on the idea of strain decomposition, capture in a compact manner the important features of the nonlinear rheology of the microgel. The second constitutive model, which incorporates the concept of kinematic hardening, embodies all of the essential behaviors exhibited by Carbopol. These include elasto-viscoplastic creep and time-dependent viscosity plateaus below a critical stress, a viscosity bifurcation at the critical stress, and Herschel-Bulkley flow behavior at large stresses on a framework adopted from plasticity theory and implements an additive strain decomposition into characteristic reversible (elastic) and irreversible (plastic) contributions

Over the last 30 years methods have been developed to simulate the flow of such complex fluids under arbitrary boundary conditions. One fundamental problem for these simulations is to properly describe and follow the position of the interface between the solid and liquid regions, in order to apply the specific constitutive equation of each regime in each of these regions. The basic approach consists in “regularizing” the behavior: in order to avoid the problem of determining this interface at each step of the flow and then applying the specific behavior type in each region, it is assumed that the material has instead a high viscosity below some critical (low) shear rate, while exhibiting the apparent viscosity of the HB model beyond this shear rate (Papanastasiou 1987). Under such conditions, there is only one (liquid) regime, with a constitutive equation valid throughout the sample volume; the theoretically solid regions flow very slowly and the approximation provides valuable data, especially when there exists some region flowing at a shear rate much higher than the critical one. A wide range of simulations have been carried out using this approach, in particular by E. Mitsoulis, J. Tsamopoulos, and their coworkers (e.g., Abdali et al 1992, Beaulne and Mitsoulis 1997, Karapetsas and Tsamopoulos 2006, Tsamopoulos et al 2008). These have provided interesting information about flows with complex boundary conditions.

In parallel, more sophisticated numerical modelling has been developed with the aim of providing more precise solutions, in particular for the boundary between the solid and liquid regions. These

approaches avoid any regularization and aim to solve the associated non-smooth variational problem using different optimization techniques, the main example being the augmented Lagrangian approach (Fortin and Glowinski 1982, Saramito and Roquet 2001, Zhang 2010), although another approach has also been used (Beris et al 1985). Mesh adaptive strategies have also been proposed to enhance the prediction of the liquid-solid boundaries (Saramito and Roquet 2001). A review of such techniques can be found in Dean et al 2007. Another recent technique consists in formulating the minimum principle for Bingham and Herschel-Bulkley yield stress fluid steady flows as a second-order cone programming (SOCP) problem, for which very efficient primal-dual interior point solvers are available (Bleyer et al 2015).

One of the next challenges in the field of modelling may be to properly represent and take into account the rheological behavior of the material in the solid regime. The simplest assumption would be to treat the material behavior as linearly elastic up to a critical deformation associated with the yield stress, where it starts to flow in the liquid regime. But things are generally more complex. Yield stress fluids are linearly elastic only for rather small deformations, and beyond that range, but still for stresses below the yield stress, they may exhibit some viscoelasticity and some plasticity (see Maimouni et al 2016) or some aging (see the paper on this topic in the present issue). The other challenge will be to take into account the behavior of the material in the solid regime in numerical simulations. Indeed, this aspect has been left aside up to now, but it may play a critical role in flows for which the solid-liquid boundary moves through the fluid (e.g., extrusion, displacement of an object), since in that case there is a need to continuously store new energy in the solid region before yielding (see Coussot 2014). Such approaches are in general more complicated and time-consuming, but they provide more trustworthy results when a precise description of the solid and liquid distribution is sought.

In fact, our above description essentially concerns “simple yield stress fluids” (Coussot et al 2009, Ovarlez et al 2013). Many yield stress fluids are also thixotropic, i.e., their behavior depends on the flow history. This typically appears as an apparent yield stress (static yield stress) varying with the rest time, and an apparent viscosity decreasing in time during flow in the liquid regime. There have been many studies in this field, and various model types have been proposed to represent the observed trends. However, we can hardly consider that there is a consensus on the most appropriate constitutive equation to be used to represent data. We can suggest several reasons for this:

- The thixotropy of a material necessarily makes the description of its rheological behavior much more complex; in addition to some steady state yielding behavior modelled with the help of several parameters as described above, the apparent viscosity must be expressed as a functional of the flow history, which will require several additional parameters (if this functional of flow history can be “rationalized” in simple terms, which also seems to be a difficulty).
- The form of this functional may vary from one material to another.
- There is a lack of data that could provide the main trends of thixotropic behavior for model systems to be used to determine or validate the constitutive equation; this is in part due to the absence of a clear set of procedures that could be used to achieve that, in relation with the problem of rationalizing the functional of flow history.
- As already mentioned, rheometrical problems are often encountered with thixotropic materials, in particular shear-banding, and these complicate the interpretation of rheological data in terms of the intrinsic behavior of the material.

Thus, understanding, controlling, and modelling thixotropy remains a major challenge for the future.



### *Microstructural origin of the behavior*

The link of the mechanical behavior of the fluid with the interactions at a local scale (i.e. between the fluid components) has soon been a subject of research (see Section 1). Most works attempted to predict the yield stress value as a function of the interactions between the suspended elements and their concentration in the suspending liquid. The first approach in that field is likely that of Princen (1983), who showed that the yield stress of foams should scale with the ratio of surface tension to bubble size (the same if expected for emulsions) and discussed the value of the factor as a function of structure and concentration. The value of this coefficient is still a subject of debate (Mason et al 1996, Rouyer et al 2005). Other works focused on the yield stress of colloidal suspensions as a function of the concentration, for example assuming a fractal structure of the flocs of aggregated particles (Shih et al 1990, Manley et al 2005). Alternate approaches assumed a yield stress proportional to a power of the difference between the concentration and a critical (percolation) value (Chen and Russel 1990, Trappe et al 2001), but for concentrated thermoreversible gels a dependence on the ratio of the concentration to a critical one was found (Rueb and Zukoski 1997).

Recent works in that field focused on the visualization of the flow structure evolution at the particle scale via different techniques (confocal microscopy, scattering) which provided new direct information on the physical origin of the yield stress (Hsiao et al ), the structure around the solid-liquid transition (Brunel et al 2016), the flow-induced structures in the liquid regime (Vermant and Solomon 2005, Masschaele et al 2011), or specific effects occurring in suspensions such as shear-banding (Shereda et al 2010), concentration gradient and shear-banding (Besseling et al 2010), shear-thickening (Hermes et al 2016, Gurnon and Wagner 2015).

One more issue has been the subject of attention recently. Concentrated suspensions generally contain a wide range of elements with different sizes and interactions, and in order to predict the rheological behavior as a function of the suspended elements, one approach is to assume that a “separation of scales” is relevant, which leads one to treat the coarsest elements as suspended in a homogeneous yield stress fluid made of the smallest elements suspended in a liquid. The problem then becomes one of predicting the rheological behavior of a suspension in a yield stress fluid, just as one would try to describe the viscosity of a suspension in a Newtonian fluid. This problem has been addressed recently, both theoretically and experimentally, by X. Chateau and G. Ovarlez (Mahaut et al 2008, Chateau et al 2008, Kogan et al 2013, Ducloué et al 2015, Ovarlez et al 2015). In particular, it has been shown that the behavior of a suspension is governed by that of the suspending yield stress fluid, i.e., the form of the constitutive equation is basically the same, with a yield stress function of the concentration of suspended elements and of the capillary number for the suspension of bubbles, but in addition, there are finite normal stress differences even if the yield stress fluid does not exhibit this trend. The next challenge in this field might be to try to describe the behavior of materials for which no obvious scale separation is possible.

## **7. Conclusion**

After a long period during which little work was done on yield stress fluids, and they were barely recognized in either fundamental physics or mechanics, there was a sudden surge of interest at the beginning of the 90s. At the same time, these materials have become the focus of attention of many physicists who see here a new class of materials, i.e., the jammed systems, requiring a new kind of

physics. Progress in our understanding of yield stress fluids has led to the emergence of new fields of research associated with different aspects of their behavior. These are areas in which many things remain to be done, including numerical modelling, characterization and modelling of thixotropy, and understanding behavior in the solid regime, confinement effects, shear-banding, and concentrated suspensions in yield stress fluids.

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