AADE-20-FTCE-078

# Arrested phase separation and gel collapse in oil-continuous drilling fluids; a route to barite sag

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## Abstract

Static barite sag can occur through a range of mechanisms. In extreme cases and under quiescent conditions in oil-continuous drilling fluids, a free oil layer forms at the upper fluid surface. Such oil-continuous formulations form a strong gel which exhibits the phenomenology of delayed gel collapse commensurate with much simpler depletion flocculated systems described in the recent literature. Here we present a range of data illustrating drilling fluid gel collapse through simple disperse-phase height evolution and x-ray tomography, consistent with arrested phase separation and through intermittency in the gel collapse process consistent with subsequent structural coarsening. Thus, we interpret our results, and therefore one static sag behaviour, as resulting from a complex interplay of structures within the gel.

## Introduction

Drilling fluids are highly engineered formulated products designed to fulfil many functions over a wide range of temperature and pressure. Key amongst these functions are (i) providing hydrostatic balance between the wellbore and the formation fluids during the drilling process, and (ii) carriage of the drilled cuttings to surface. Hydrostatic balance is commonly achieved by adding high density mineral particles to the fluid so that the overall density of the fluid is comparable to the formation density. Hence the drilling fluid formulation contains both dense particles that are part of the formulation and less dense particles, i.e. cuttings, that are produced during the drilling process. In drilling any well, it is routine that the drilling process is suspended for a period time, this could be for hours, days or even weeks. In such circumstances it is desirable that the particles dispersed in the drilling fluid do not settle within the well. If the dense weighting particles settle ("barite sag"), it can lead to unacceptable density variation that may allow formation fluids to enter the well potentially leading to a dangerous situation. If the cuttings settle, then the drillstring may become stuck in the resultant bed requiring expensive remedial action. Hence, the settling of particles within drilling fluids has been a concern within the industry for very many years. Typically, particle settling is controlled by formulating the drilling fluid to have a yield-stress. We are therfore interested in the prototypical problem of a particle, falling under gravity, in a viscoplastic material.

The usual assumptions when considering a spherical particle falling in a viscoplastic fluid are to consider the system to have full scale-separation. That is, the system comprises a solid sphere within a continuum fluid so that all usual hydrodynamic limits may be taken. This problem has been studied by (Beris, Tsamopoulos et al. 2006) for a Bingham background fluid and more recently both experimentally and theoretically for a Hershel-Bulkley background fluid by (Tabuteau, Coussot et al. 2007). In both cases a yield number, $Y$, is considered that compares the force exerted by the yield-stress over the surface of the particle with the gravitational force exerted on the particle,

$$Y = \frac{3\tau_c}{g.a.\Delta\rho} \tag{1.1}$$

with $\tau_c$, the yield-stress, $g$, gravitational acceleration, $a$, particle radius and $\Delta\rho$ the density difference between the particle and surrounding fluid. Whereas this criteria might be expected to provide a unique value to describe the boundary between particles that settle and particles that are suspended, more recently (Emady, Caggioni et al. 2013) have shown that even if the yield-stress is matched between differing fluids and scale separation is satisfied, the detailed microstructure of the fluids plays an important role. Indeed, different critical yield numbers are found for the two fluids investigated despite them having matched yield-stresses. Of course, a Bingham or Hershel-Bulkley analysis takes no account of creep or thixotropic behaviour present in a wide range of materials and in drilling fluids in particular.

Here we are interested in the evolution of mass density gradients that form under quiescent conditions in oil-continuous drilling fluids, i.e. static sag. In particular, we are interested in the specific observation of fluid syneresis, that is the appearance of a free oil layer at the surface of some oil-continuous formulations. For any operation there are defined acceptable limits as to how much density is allowed to vary in a given interval. As the industry has moved to formations that are more difficult to drill this specification has become progressively more stringent. Today a typical specification would be that the density change at the bottom of the standard test cell (about 150mm deep) over 7 days should be no more than 0.1ppb or about 0.8%.

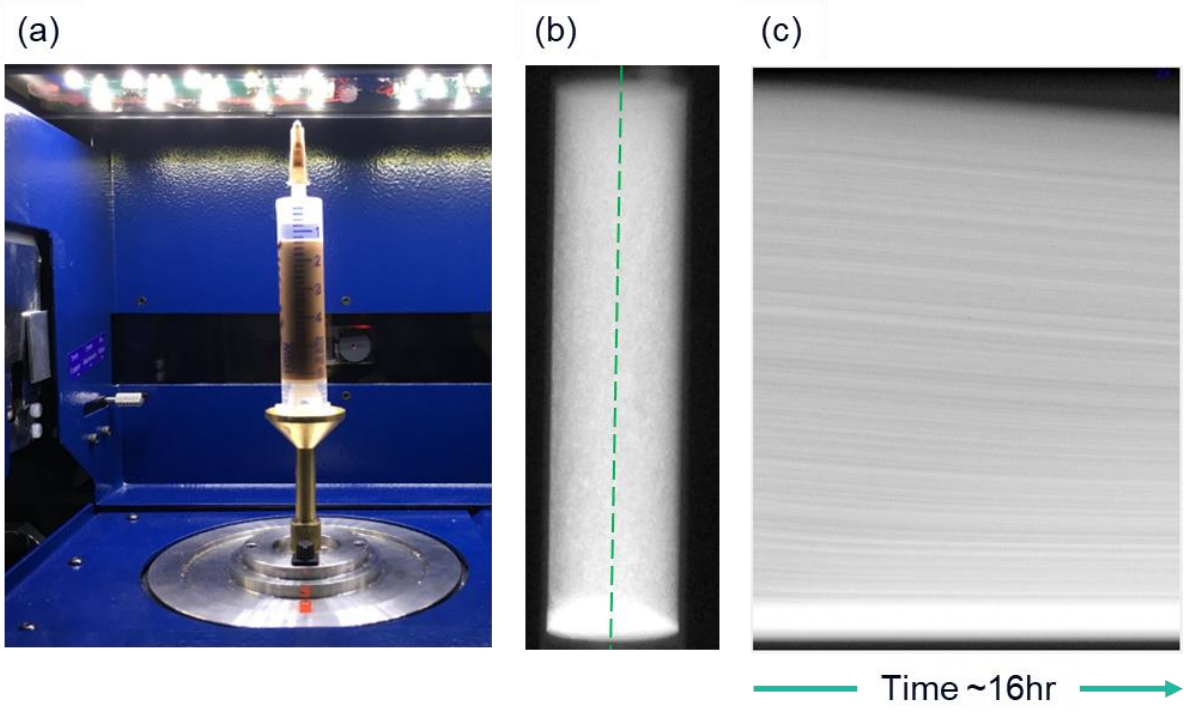


**Figure 1 – Model drilling fluid sample, constructed to show rapid syneresis, in a standard 5ml syringe. (a) sample mounted in a syringe tube within the x-ray microtomograph instrument (Bruker, SkyScan 1275 micro-CT scanner) (b) shadowgraph image taken at start of experiment (white is dense barite) showing initial distribution of barite particles. (c) time evolution of centerline of image (b) as indicated.**

## Gel collapse in drilling fluids

Oil-continuous drilling fluids (or oil-based mud, OBM) typically comprise a base oil with brine emulsion drops. The brine volume fraction is typically about 0.25. Additionally, the formulation also has a hydrophobically modified clay, excess surfactants (emulsifiers and dispersants to ensure both the stability of the emulsion and dispersion of drilling solids) and weighting mineral. The brine drops are typically about 1μm radius and the barite 3-80μm radius depending on grade and grinding polydispersity. There are also several minor components added for a variety of purposes. The whole formulation may be characterised as a colloidal attractive gel.

Oil-based drilling fluids, whilst variable, typically have a yield-stresses greater than about 1Pa. For the largest barite particle of 80μm and a density difference of about 2.5g/cm$^3$, the yield number is calculated as $Y \approx 3$. Since this number is greater than 1 we expect that yield-stress dominates and the particle is suspended. For smaller particles, the number is even larger (more dominant yield-stress). Hence the simplistic analysis suggests that no barite sedimentation occurs, in which case how and why do measured density changes occur?

To track the density distribution as a function of time it is instructive to use x-ray shadowgraphy. In Figure 1(a) is shown a model drilling fluid sample (brown fluid), formulated to show rapid syneresis, contained in a standard syringe and mounted internal to an x-ray microtomograph imaging instrument (Bruker SkyScan 1275 micro-CT scanner). In Figure 1(b) is shown an initial shadow image. In this case the denser the particle the brighter it is, hence the white spots are barite particles.

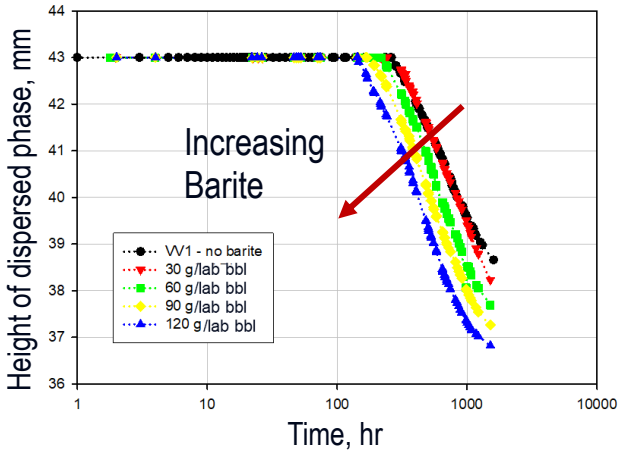


**Figure 2 – Height of interface of model OBM gel with supernatant as a function of time for several barite loadings measured using a Turbiscan (Formulaction) instrument. Initially there is no free oil. Free oil appears for this formulation after about 110hr. Subsequently the height of the gel phase drops in an exponential fashion.**

Images are acquired at successive times. In Figure 1(c) is plotted a line from each of the sequence of images (shown as dashed line on (b)) and arranged to generate an image corresponding to height on the vertical axis and time on the horizontal axis. The horizontal axis extends about 16hr. Note that the top surface falls, along with the obvious traces of particles so that the overall system appears to "squash" rather

than particles falling through a background. The bright base is likely due to a small amount of sedimentation that occurs during filling and before gelation.

The obvious initial way to model the process is as poroelastic collapse (Buzzaccaro, Secchi et al. 2012) whereby the gel structure is modelled as a gravitationally settling structure with porous flow through the structure in the opposite direction. However, such a model predicts collapse starting at time zero, followed by the interface between gel and free oil decreasing linearly with time. In general, neither feature is observed for gelled drilling fluid formulations.

In Figure 2 are shown measurements of the gel height as a function of time after shearing and loading the cell for a variety of barite loadings. It is obvious that there is a significant time delay before the gel collapses. Further, once collapse starts the height falls exponentially rather than linearly. These features suggest that the underlying mechanism is not poroelastic collapse. Note that recently a hydrodynamic instability model based on poroelasticity has been reported which shows a similar behaviour of the gel height (Varga, Hofmann et al. 2018), however there is no evidence in our systems of the modelled instability. There is experimental evidence in the literature that such a process occurs in moderate volume fraction gels (Harich, Blythe et al. 2016), but much lower volume fraction than drilling fluids.

### *Effect of colloidal interaction strength*

The problem of delayed gel collapse has been studied extensively in the academic literature. The phenomenon has relevance to a wide range of formulated products in industries spanning foods, personal care, cosmetics and agrochemicals. In general, studies have been reported for depletion-flocculated gels where detailed control over the colloidal attractive interaction strength and range can be exerted. It is found that such systems gel via an arrested phase separation process (Harich, Blythe et al. 2016) whereby the dispersed material starts to phase separate but a system spanning glassy structure is rapidly formed which prevents further separation. This network then undergoes a coarsening process, observed experimentally by (Bartlett, Teece et al. 2012), and directly modelled by (Padmanabhan and Zia 2018). The model, despite ignoring hydrodynamics, qualitatively reproduces in the gel: the delayed gravitational collapse, the exponential height evolution and ensuing density gradient we observe in x-ray tomography. Microscopically the process is driven by the particle osmotic pressure drawing the gel together and is therefore a syneresis process.

For a formulated drilling fluid, the detailed studies presented in the academic literature are not possible. However, we can probe OBMs in related ways. In (Bartlett, Teece et al. 2012) the depletion interaction strength was varied and it was observed that the delay time to gel collapse increased with increasing interaction strength. In our model drilling fluid we can change one element of the interaction strength by changing the brine salinity of the dispersed phase emulsion. Increasing the salinity increases the refractive index of the brine bringing its value toward match with the oil-continuous phase. At match (salinity approx. 5M) the drop-drop Van der Waals interactions are their weakest. In Figure 3 we see that the time delay to gel collapse is, as with the literature system, increased by increasing the interaction strength.

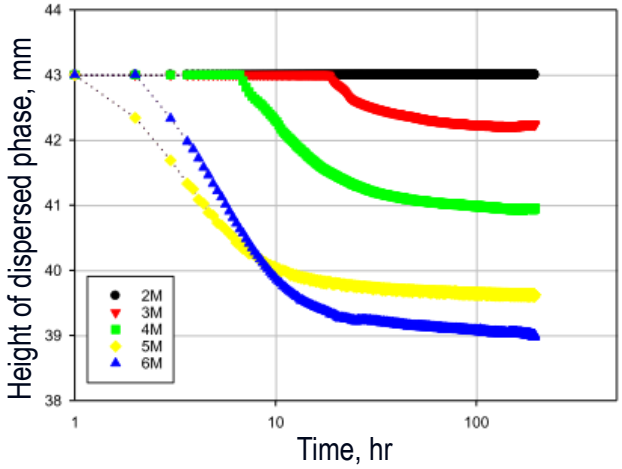


**Figure 3 – Height of interface of model OBM gel with supernatant as a function of time for varying brine salinity. The delay time is minimized at the minimal Van der Waals interaction strength (5M).**

### *Observations of intermittency*

Although OBM formulation complexity precludes direct confocal imaging of structural coarsening as reported by (Teece, Faers et al. 2011, Bartlett, Teece et al. 2012, Teece, Hart et al. 2014) we are able to measure the resulting intermittency in the collapse dynamics as seen by (Bissig, Romer et al. 2003, Kajiya, Narita et al. 2013).

In Figure 4a we show, for a model drilling fluid, the collapse velocity of the colloidal gel as a function of time. The velocity is measured using a spatial speckle correlation technique. Rather than a continuous slow velocity (as expected for poro-elastic collapse) we see complex dynamics with periods where the gel is almost stationary followed by intermittent very large velocities. On integrating the velocity we see an average movement of the gel downwards but predominantly as a series of steps (Figure 4b). The probability distribution function of event amplitudes shown in Figure 4a is plotted in Figure 4c where the solid black line is a gaussian fitted to the central peak. The broad non-gaussian wings are due to these intermittent events and are characteristic of avalanche-like statistics. Hence the probability distribution of times between these intermittent events is well described by a power law (Figure 4d) commensurate with that observed for glassy emulsions seen by (Kajiya, Narita et al. 2013).

A general explanation for these statistics together with the gel collapse observations is provided, for single-component systems, by (Padmanabhan and Zia 2018) using molecular dynamics simulation. An attractive colloidal system is unstable to phase separation. At low concentrations flocs will form which, if denser than the continuous phase and when they have grown to a non-brownian size, will settle.

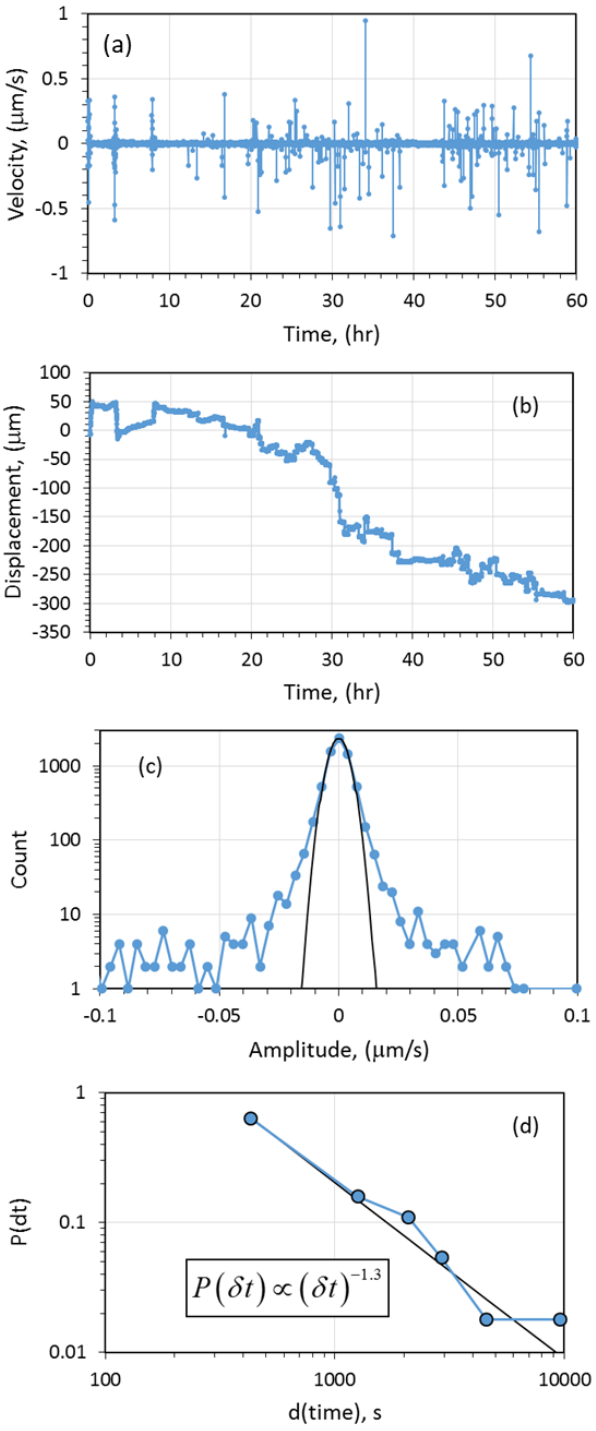


**Figure 4 – Speckle velocimetry of a model drilling fluid gel collapse (a) velocimetry as a function of time, intermittent high velocity events are observed rather than steady settling, (b) integration of the velocity for displacement, (c) probability distribution function of the data in (a), and (d) probability distribution of times between events.**

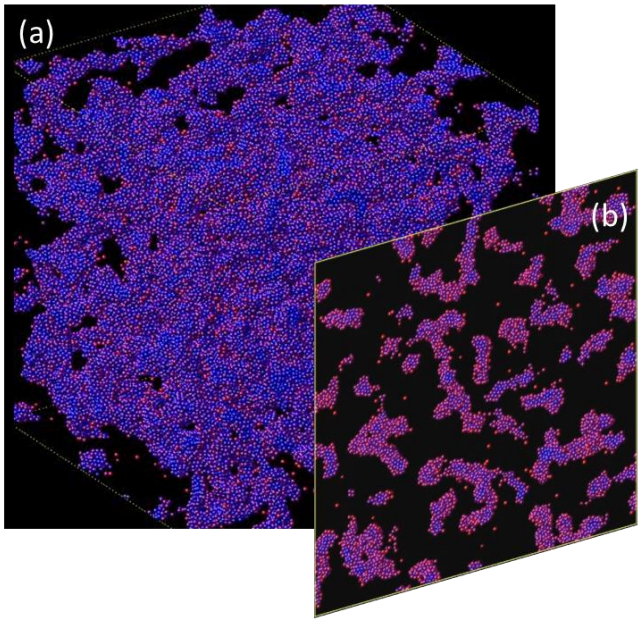


**Figure 5 – LAMMPS (Plimpton 1995) calculation of simple single component attractive system having undergone arrested phase separation. Initial volume fraction 0.2. The particles cluster forming a system spanning 3D network of glassy strands. (a) a projection of the 3D computational cell with periodic boundary conditions, (b) a single slice from the same calculation illustrating the clusters more clearly; in 3D these are fully connected.**

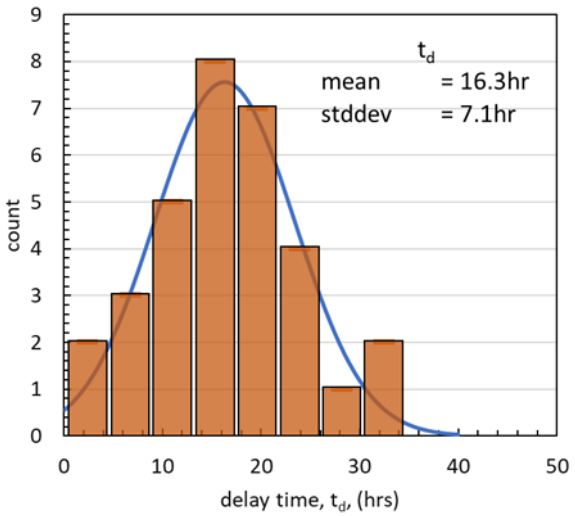


**Figure 6 – Thirty-two identical drilling fluid samples were allowed to stand quiescently. The gel interface height was measured optically and each individually followed the behavior illustrated in Figure 2. Plotted here is a histogram of the observed delay time before free oil was observed.**

At higher concentrations the flocs will join to form a system-spanning network which can be self-supporting. This is a gel. An illustration of the structure that is formed is shown in Figure 5 where a simple attractive system is allowed to evolve using LAMMPS (Plimpton 1995).

The particles on the surfaces of the strands can diffuse more

easily and on average move from the bridge regions to the node regions. Over time the bridge regions thin and eventually break. When this happens, the forces supported by that strand are re-distributed on neighbouring strands which in turn may trigger their breakage and a subsequent avalanche. The whole network is non-equilibrium and under compressive force due to the particle osmotic pressure.

The data in Figure 4 show that the collapse process is essentially stochastic in nature. This is further illustrated by repeating the gel collapse observations may times for a single formulation. We took a drilling fluid sample and prepared thirty-two identical samples. We then, in parallel, measured the time until a free oil layer was first observed (at 20°C and 1bar). A histogram of these times is plotted in Figure 6. A very broad distribution of times is observed. These data are strikingly similar to data obtained for a simple depletion gel in (Teece, Hart et al. 2014).

## Conclusions

Oil-based drilling fluids by formulation design are complex colloidal systems. The colloidal components are, at least in part, attractive systems and they exhibit time-dependent gel structures. Some formulations over long periods of time exhibit syneresis, that is the formation of an oil layer at the upper surface with the dispersed components forming a gel phase below. Given the presence of an oil layer, the density at the bottom of the sample necessarily increases and this can be one form of static sag. For systems behaving in this way, barite particles are seen to be locally suspended and it is instead the gel-structure that is evolving.

Thus, we find that for some, though not all, oil-based drilling fluids formulated with an emulsion the following,

- Over long periods, the gel is seen to separate leaving a free oil layer at the surface. Non-obviously, the process is associated with a significant time delay before any separation is seen.
- Whereas chemical degradation is always a concern, our observations are not associated with chemical changes.
- The time delay is shortened for higher barite loadings, but the particles move with the gel rather than through the gel.
- Increasing the attractive potential(s) within the formulation leads to longer delay times.
- The gel collapse process proceeds as a series of micro-avalanches rather than a slow continuous evolution implying a stochastic process.

These observations have a strong correlation with observations of the evolution of depletion gels reported in the literature. Hence our conjecture is that the OBM systems, where these phenomena are seen, may be usefully considered in the same theoretical framework.

## Acknowledgements

The authors would like to thank Dr. S. Lovett for calculation of figure 5.

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