Experimental observation of shear thickening oscillation

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Abstract – We report experimental observations of the shear thickening oscillation, i.e. the spontaneous macroscopic oscillation in the shear flow of severe shear thickening fluid. Using a density-matched starch-water mixture, in the cylindrical shear flow of a few centimeters flow width, we observed that well-marked vibrations of frequency around 20 Hz appear via a Hopf bifurcation upon increasing externally applied shear stress. The parameter range and the frequency of the vibration are consistent with those expected by a simple phenomenological model of the dilatant fluid.

Introduction. – Dense colloids and dense granule-fluid mixtures are often called dilatant fluids and known to show severe shear thickening, i.e. its viscosity changes discontinuously by orders of magnitude upon increasing the shear rate [1–4]. This is a source of a variety of un-intuitive behaviors of the media [5–9], and might be used for interesting applications like a body armor [10], but it often causes problems in industrial situations, such as the damage of mixer motors due to overloading [1].

Although the severe shear thickening is a property that can be demonstrated easily with common materials like starch and water, physicists have not reached a general agreement on its microscopic mechanism. Originally, the order-disorder transition of the dispersed particles was proposed [1,11–13], but later the hydrodynamics cluster formation by the lubrication force is considered to be more consistent with experimental observations and numerical simulations [14–17]. More recently, the shear thickening transition is discussed in connection with the jamming and/or the compaction of granules [18–21].

Recently, the present authors constructed a phenomenological model that describes macroscopic flowing properties of a shear thickening fluid [22,23]. By analyzing the model, they pointed out that the discontinuous thickening upon increasing the shear rate signifies that the shear thickening fluid is actually shear stress thickening, not shear rate thickening because only the shear stress thickening can produce the S-shaped flow curve with an unstable branch, that causes the discontinuous transition experimentally observed (fig. 1). Such a flow curve has been suggested also by a semi-microscopic theory of shear thickening fluid [24].

An interesting consequence of the existence of this unstable branch is that the flow may show a spontaneous oscillation, i.e. the shear thickening oscillation, when the applied external shear stress is in the unstable region because a uniform shear flow is unstable. It was predicted using the phenomenological model [21,22], and its basic dynamics can be understood as follows: A uniform shear flow in the unstable branch can be destabilized by an

![Fig. 1: (Color online) Schematic flow curve for a dilatant fluid that causes a discontinuous thickening. For the shear stress in the shaded region, a uniform shear flow is unstable.](image-url)
infinitesimal disturbance toward the thickened state with small shear rate flow. However, the shear rate increases again as the medium relaxes because the external stress is not strong enough to maintain the thickened state. Such cycles of alternation between the thickened state and the relaxed state is the shear thickening oscillation. In the deceleration phase, additional inertial stress is imposed on the fluid to boost further thickening. This makes the deceleration steeper than the acceleration. Such a effect is larger for the wider flow, thus the oscillation becomes of sawtooth shape for the flow with large width as is shown in refs. [21,22]. On the other hand, the oscillation may be suppressed in the flow with small width due to the momentum diffusion by the viscosity. By estimating the parameters using experimental data for the cornstarch-water mixture [2], the oscillation frequency is expected to be of the order of 10 Hz when the flow width is several centimeters. This oscillation is macroscopic, and is not due to a discreteness of granules.

The shear thickening oscillation is a salient property predicted by the phenomenological model and it is actually not difficult to demonstrate the vibration by just pouring the starch-water mixture out of a container, but we could not find any published literature which reports such a clear oscillation except for noisy fluctuations [18,25,26]. One of the reasons for this might be that a typical size of rheometer sample for a precision measurement may be too small; the oscillation is suppressed by the viscosity when the flow width is narrower than the length scale related to the momentum diffusion length scale associated with the shear rate in the unstable branch. In this paper, using containers with several centimeters flow width, we report experimental observations of spontaneous oscillation in the cylindrical Couette flow of the starch-water mixture.

**Experimental setup.** – Figure 2 illustrates our experimental setup. The fluid flows in Taylor-Couette geometry between the outer cylinder and the coaxial rod at the center. The cylindrical container is 24 cm tall, but is filled with the fluid up to 22 cm deep. The upper surface of the fluid is open to the air although the container is capped with an aluminum plate. The diameter of the center rod is 5 cm; We use several outer cylinders with different inner diameters, so that we can have the flow of the width \( h = 1 \sim 5 \text{ cm} \). The outer cylinder and the rod are both acrylic, and their surfaces are lined with water-proof sand paper in order to enforce the no-slip boundary condition. The center rod rotates under an externally applied constant torque with the outer cylinder being fixed. The external torque is applied by a weight through a steel wire wound on the rod; the weight is hung through a spring and damper system (Samini Co., Ltd.), and the mass of the weight is in the range of 0.5 ~ 10 kg, which gives the external stress \( \sigma_e = 0.14 \sim 2.8 \text{ kPa} \) at the surface of the rod. The moment of inertia of the rod is sufficiently small relative to those of the fluid in the case the fluid rotates as a solid.

For each weight, we choose an appropriate spring and damper system to minimize the vibration transmits to the weight from the rod. The intrinsic frequency of the system is within the range from 3 Hz to 5 Hz. We observe no oscillation in the experiment with honey. The angular speed of the center rod \( \omega \) is recorded by a rotary encoder at the sampling rate \( 2 \times 10^3 \text{ 1/s} \) (SP-405ZA, Ono Sokki). As for the fluid, we use the suspension of potatostarch (Hokuren) density-matched with the aqueous solution of CsCl of the density 1.68 g/cm\(^3\). The viscosity of the suspension in relaxed state is about 20 Pa·s for the suspension of 41 wt% and 30 Pa·s for the suspension of 42.5 wt% in the relaxed state.

**Shear thickening oscillation.** – Figure 3 shows the time evolutions of the angular speed of the rod for the potatostarch suspension of the concentrations 41 wt% (a) and 42.5 wt% (b) with the flow width \( h = 4 \text{ cm} \). The plots show the oscillations for the various external shear stresses \( \sigma_e \) at the surface of the rod. For both of the concentrations, clear oscillations about 20 Hz are observed for the external stress \( \sigma_e \gtrsim 0.2 \text{ kPa} \). As the external stress decreases, the frequency remains almost constant around 20 Hz, but the amplitude of the oscillation decreases and vanishes at \( \sigma_e = 0.1 \text{ kPa} \), thus the transition is consistent with a Hopf bifurcation as has been expected for the phenomenological model [22,23]. In the case of the thinner fluid of the 41 wt% concentration, a slower oscillation around 5 Hz remains at \( \sigma_e = 0.1 \text{ kPa} \) (fig. 3(a)). For the large external stress, we expect that the rod will be stuck due to the severe shear thickening, but in the present experiment, for \( \sigma_e \gtrsim 2.0 \text{ kPa} \), the rod starts slipping suddenly after initial transient. The shape of the oscillation wave looks rather symmetric and sinusoidal for the 41 wt% suspension, while for the 42.5 wt% suspension the oscillation is somewhat asymmetric even at smaller stress \( \sigma_e = 0.2 \text{ kPa} \). The oscillation is of sawtooth type that consists of a gradual increase of the angular speed followed by a sudden drop, and becomes asymmetric further for larger \( \sigma_e \).

These features can be seen also in the Fourier space. Figure 4 shows the Fourier spectra of the time sequences.

![Figure 2: Schematic illustration of the experimental setup consisting of a cylindrical container, a rotating rod, and a rotary encoder.](image-url)
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Fig. 3: (Color online) Time evolutions of the angular speed of the rod for the flow of $h = 4$ cm thickness with various applied stresses. Suspension densities are 41 wt% (a) and 42.5 wt% (b). (c), (d): enlarged waveform of the angular speed for each suspension density.

Fig. 4: (Color online) Fourier spectra of the oscillation for the time evolutions of the oscillation in fig. 3 for $S_e = 0$.1 (bottom), 0.2, 0.45, 0.7, 0.9, and 1.1 kPa (top). The absolute values of the spectra are plotted in logarithmic scale, shifting by the factor 100 for the upper plots to avoid overlapping.

in fig. 3. For larger $S_e$, there are peaks around 20 Hz accompanied by the peak of its higher harmonics around 40 and 60 Hz. For the 41 wt% suspension, the amplitude for the higher harmonics decreases faster than that for the peak at 20 Hz as $S_e$ decreases, showing that the oscillation becomes closer to the sinusoidal form. For the 42.5 wt% suspension, this tendency is not so clear, and the basic frequency is not stable but changes between 15 Hz and 25 Hz.

The amplitude and the frequency of the peaks around 20 Hz are plotted as a function of $S_e$ for some flow widths $h$ in fig. 5(a) and (b), respectively. There exists a threshold of $S_e$ around 0.1 kPa toward which the amplitude vanishes continuously as $S_e$ decreases. On the other hand, the frequency stays almost constant near the threshold, suggesting that the bifurcation is of the Hopf type.

It should be notable that both the amplitude and the frequency barely depend on the flow width $h$ within the parameter range studied in the present experiments. Figure 6 shows the oscillation frequency as a function of the flow width $h$ for various external stresses $S_e$. One can see that the frequencies are around 20 Hz for all the cases, and we could not find any systematic dependence on either $h$ or $S_e$. For $S_e$ higher than 1.0 kPa, the center rod starts slipping and we could not obtain clean data, but we find no clear sign of a systematic change in the frequency up to $S_e = 2.3$ kPa.

The maximum and minimum angular speed of the rod, $\omega_{\text{max}}$ and $\omega_{\text{min}}$, during a single cycle of oscillation are plotted against $S_e$ in fig. 7 for some values of the flow width $h$ for both of the concentrations. Each data point represents an average over a single run of the experiment, which contains typically $10^4$ cycles of the oscillation. Neither $\omega_{\text{max}}$ nor $\omega_{\text{min}}$ depend on the flow width $h$ in any.

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appreciable way. As for the maximum angular speed $\omega_{\text{max}}$, the plots show almost linear dependence on the external shear stress $S_e$:

$$\omega_{\text{max}} \approx a S_e + \omega_0$$

(1)

with $a \approx 11 \text{ rad)/(kPa} \cdot \text{s}$ and $\omega_0 \approx 10 \text{ rad/s}$ for the 41 wt% suspension (fig. 7(a)), and $a \approx 15 \text{ rad)/(kPa} \cdot \text{s}$ and $\omega_0 \approx 0$ for the 42.5 wt% suspension (fig. 7(b)). On the other hand, the minimum angular speed $\omega_{\text{min}}$ stays roughly constant for $S_e \gtrsim 0.5 \text{kPa}$ after the initial decrease for smaller $S_e$. In the case of the 42.5 wt% suspension, the oscillation disappears around $S_e \lesssim 0.2 \text{kPa}$, for which cases the terminal angular speeds are plotted with open marks in fig. 4(b).

The linear dependence of $\omega_{\text{max}}$ on $S_e$ leads us to introduce an average viscosity $\eta_{\text{relax}}$ for the flow during the relaxed state. If we assume the steady cylindrical Couette flow with a constant angular velocity $\omega$, the viscosity would be given by

$$\eta = \frac{S_e}{2\omega} \left[ 1 - \left( \frac{R_1}{R_2} \right)^2 \right],$$

(2)

where $R_1 = 2.5 \text{ cm}$ is the radius of the center rod and $R_2 = R_1 + h$ is the radius of the outer cylinder. Substituting $\omega_{\text{max}}$ of eq. (1) into $\omega$ in eq. (2), we have $\eta_{\text{relax}} \sim 30 \text{ Pa} \cdot \text{s}$ for the 42.5 wt% suspension, and $\eta_{\text{relax}} \sim 20 \text{ Pa} \cdot \text{s}$ for the 41 wt% suspension. This is consistent with the previous viscosity measurement before the thickening transition [2].

For the 41 wt% suspension, fig. 7(a) shows a substantial non-zero extrapolation $\omega_0$ at $S_e = 0$. This may be due to the inertia effect in the oscillatory state, in which case our steady-state formula, eq. (2), is not valid for this case.

In the thickened state, $\omega_{\text{min}}$ is nearly constant around $0.5 \text{ rad/s}$ for $0.5 \lesssim S_e \lesssim 1.5 \text{kPa}$ for the 42.5 wt% suspension. The estimated values of the viscosity in the thickened state are of the order of $10^3 \text{ Pa} \cdot \text{s}$, which seems to be somewhat smaller than the value observed in ref. [2]. This may suggest that the whole system is not thickened uniformly, but the thickening region is localized as we will discuss later.

Discussions. – We experimentally observed the spontaneous oscillation of macroscopic shear flow in the shear thickening fluid. The shear flow starts oscillating with a finite frequency through a Hopf bifurcation upon increasing the externally applied shear stress. The oscillation is asymmetric in the sawtooth shape, i.e. a gradual increase followed by a sudden deceleration, but seems to become more symmetric for the stress near the threshold. The
observed frequency of the oscillation is about 20 Hz, which is roughly in the range expected by the model for the flow width of several centimeters. These features are consistent with those of the shear thickening oscillation predicted by the phenomenological model [23].

On the other hand, there are some other features that seem difficult to interpret in terms of the simple shear flow oscillation as has been analyzed for the model. The oscillation frequency does not depend significantly neither on the external stress $S_e$ nor on the flow width $h$, whereas the simulations of cylindrically symmetric oscillation show that the frequency tends to become slightly higher as $S_e$ increases, and that the frequency decreases significantly as the flow width $h$ increases. It is also tricky to interpret the behavior of the maximum $\omega_{\text{max}}$ and minimum angular speed $\omega_{\text{min}}$ of the oscillation: $\omega_{\text{max}}$ is proportional to $S_e$ while $\omega_{\text{min}}$ stays almost constant, and neither of them depends on the flow width $h$. We find that these features are not easy to reproduce by the model as long as we assume that the flow is cylindrically symmetric.

One possibility is that thickening does not occur uniformly, thus these experimentally observed parameter dependences cannot be interpreted using the cylindrically symmetric flow. At the moment, we cannot determine the symmetry of the flow in our experiments because we cannot observe the thickening region in the cylinder. We performed numerical simulations in two dimensions on the phenomenological model, and we found that the cylindrical Poiseuille flow is destabilized for larger shear stress and a localized thickened band region appears diagonally to support most of the shear stress [27]. We have not performed simulations in three dimensions yet, but it is natural to expect such localized thickened bands to appear also in three dimensions because there is a natural tendency for the shear stress thickening fluid to form a thickened band; once a thickened region appears, it supports larger stress, which thickens the region further.

The oscillation we report here is not subtle but quite evident under an appropriate condition, yet we could not find any previous reports on this phenomenon. Some remarks on noisy fluctuations [18,25,26] are the only data that we could find in the published literature\(^1\). We suspect that this is because most of the previous measurements on the shear thickening fluid have been done on samples with the flow width of the order of millimeters, while the macroscopic oscillation is suppressed in the shear flow narrower than the length scale related to the momentum diffusion length scale, which is estimated to be of the order of centimeters in the present material [22,23].

Recently, von Kann et al. reported that the velocity of a sphere settling in a cornstarch suspension exhibits a steady oscillation [7,8]. This phenomenon is caused by the constant gravitational force, and the frequency that they observed is similar to our results. Thus, we consider that their observation can be explained by a shear thickening oscillation.

It should be mentioned that there is a report on an oscillatory shear flow of a lyotropic lamellar phase [28]. The phenomenon is analogous in the sense that the internal structure of the fluid changes during a period of oscillation, and the change in the viscosity associated with the structural change seems to cause the oscillation, thus a similar phenomenological model may be used to describe the behavior. On the other hand, the differences from the present oscillation are that the material shows shear thinning and the period of oscillation is of the order of a thousand seconds, thus no fluid dynamical inertia effect should be involved in the oscillation dynamics, unlike in the case of the shear thickening oscillation.

**Summary.** – In summary, using the starch-water mixture, we observed the shear thickening oscillation with the frequency around 20 Hz in the shear flow of the Taylor-Couette geometry with several centimeters flow width. The oscillation starts through a Hopf bifurcation with the frequency on increasing the external shear stress. The frequency and the amplitude of the oscillation barely depend on the flow width for the range 1–5 cm of the present experiments.

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