Persistent Holes in a Fluid at Different Conditions

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Abstract

It is known that perturbed holes in a vertically vibrating shear thickening fluid filled in a container become persistent at a range of acceleration and frequency. We show experimentally that these range of acceleration and frequency shifts to another range when fixed parameters such as container size, depth, viscosity are set to a different value. The results provide a broader understanding of persistent hole conditions for our shear thickening fluid (polypropylene and fumed silica mixture) that was used. It was additionally discovered by this experiment that polypropylene and fumed silica mixture has a required time for the particle structure to relax and the persistency of the holes also depend on the Faraday waves.

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I. INTRODUCTION

Merkt et al. discovered an interesting phenomenon in a vertically vibrated (sinusoidal oscillation) cornstarch and glass microsphere suspension [1]. It was discovered that holes, which were created by applying a perturbation to the layer were persistent under certain situations. The conditions for holes to be persistent requires a certain range of frequency and acceleration. The frequency and acceleration range, which can be either stable (holes that are persistent) or unstable (holes that close) was described as a domain in a phase diagram as a function of acceleration and frequency. These persistent holes were attributed to shear thickening of a fluid. Shear thickening is an increase of viscosity with increasing shear rate. Merkt et al. discovered that fluids which experience shear thickening during an increasing shear stress only maintains holes to not close during vertical vibration.

Here we compared the shift in domain by varying the parameters such as depth, container size, and rheological behavior. Instead of using cornstarch or glass microspheres, a mixture of polypropylene glycol and fumed silica were used for the suspension material in our experiment. This suspension is advantageous over other suspensions that were used in the past because of its transparency and the constant physical properties (concentration, viscosity, etc.) over time. Transparent fluid is useful since optical properties of the fluid can be explored easily. Also, cornstarch had difficulties of evaporation of water. This resulted in change of density and viscosity (at a given shear rate). However, this new mixture did not encounter such problems and were able to be kept at a long period of time. For actual experiments, we explored the phase diagram similar to that of Merkt et al. at which accelerations against frequency were plotted for each state. Additionally, which is the main task of this experiment, parameters were varied such as depth, container size (diameter), and viscosity and the phase diagrams were compared to each other.

II. EXPERIMENT

A. Preparing Suspensions

For suspensions, a mixture of polypropylene glycol (liquid) of molecular mass 725 and fumed silica (solid) was used in our entire experiment. The formula of concentration of this mixture is given in Equation (1).
\[
\text{concentration(\%)} = \frac{\text{Fumed Silica}(g)}{\text{Fumed Silica}(g) + \text{Polypropylene glycol}(g)} \times 100
\] (1)

If the concentration is not high enough, the change of viscosity at increasing shear rate (shear thickening) is too small that the perturbed holes do not result in persistent holes when the mixture is vertically vibrated. On the other hand, if the concentration is too high, the two substances do not mix properly to form into a liquid state. The mixture forms into a gel state that is inappropriate for our task. Several different concentrations of fumed silica were made and compared. The maximum concentration was 8%.

The rheological quantities were measured with a stress controlled rheometer (AR-2000EX, TA-instruments) with an acrylic cone of diameter 60mm and an angle \( \alpha = 1^\circ 58'59'' \) at temperature 25°C. Another problem was found while rheometric measurements were taken. As shown in Figure 1, exploring above a certain shear rate, the results of viscosity as a function of shear rate \( (\eta(\dot{\gamma})) \) shifted down and no longer followed the previous path. The higher the maximum shear rate was experienced to the fluid, the larger downshift of viscosity as a function of shear rate was resulted in. Several attempts were therefore made to get rid of this unexpected behavior. Eventually, using a commercial blender for mixing the material at a sufficiently high shear rate solved the problem. We set the standard to 2 minutes of blending time at highest speed (Frappe mode). After, 2 minutes of blending, the viscosity versus shear rate no longer shifted down during measurements with the rheometer (measuring up to 300 s\(^{-1}\)). The underlying reason is not clearly known, but we guess that the structure of particles in the mixture must have changed by high shear rate. Once the mixture experiences very high shear rate by the blender, the viscosity does not shift down during measurements by the rheometer since it does not apply higher shear rate than the blender applied. For additional blending time, however, the viscosity as function of shear rate does shifts down even more. This fact was used in our experiment in subsection G.
Figure 1: Viscosity of polypropylene and fumed silica mixture as measuring during an increasing shear stress ramp. Polypropylene and fumed silica mixture at 8% weight ratio (Based on Equation (1)). Before blending the mixture with a commercial blender, the plot shifted down as shown in the graph after a high shear rate (100s⁻¹) was experienced. The error bars correspond to variation of two measurements of the same sample.
B. Rough Sketch of the Phase Diagram

![Phase Diagram](image)

Figure 2: Phase diagram for a vibrated aqueous cornstarch suspension as a function of acceleration and frequency. In either the metastable or stable region, a surface perturbation forms a persistent hole. In a metastable regime these holes always collapse within $10^5$ container oscillations. Stable regime last more than $10^6$ oscillations. In the delocalized regime the surface is irregular. Faraday waves appear with increasing acceleration at $\Delta$, and disappear with decreasing acceleration at $\nabla$. (Courtesy: Merkt et al. [1])
Figure 3: Phase diagram for polypropylene glycole and fumed silica mixture suspension (8%) as a function of acceleration and frequency explored at several states. The acceleration is the root mean squared value. The stable points (persistent holes) are denoted as O (blue) and unstable points (holes that close) are denoted as X (red). Stable points were defined when the perturbed holes do not close for 5 minutes. Unstable holes were defined when the holes close before 5 minutes. The boundary between stable and unstable states were derived by the average of the lowest stable point and the highest unstable point.

Before examining different conditions, it is important to know how the phase diagram looks like in general. The container with an aluminum base plate, acrylic sidewall (inner diameter 126mm) was used and the liquid was filled in with a depth of 6.164 mm. The initial perturbation to first create holes were generated by a puff of air of 40 psi for 3 seconds. Stable points at which the perturbed holes are persistent were defined by an empirical decision as vibrating condition (acceleration and frequency) of holes not closing for 5 minutes. If the
hole closes before 5 minutes, it is defined unstable. Figure 3 shows a coarse result of the phase diagram. Through this, we concluded that above some boundary, perturbed holes result in persistent holes (stable region), whereas below the boundary the perturbed holes result in closing (unstable region). Comparing to the results of cornstarch from Merkt et al., delocalized region were not able to be observed since the electrodynamic shaker had a maximum acceleration limit of 40 g peak to peak.

C. Faraday Wave Dependance

It is known that Faraday waves appear on a surface of the liquid when vertical motion is sinusoidal [2]. For cornstarch, Faraday waves do not play any role for persistent holes [1]. Figure 2 shows that the onset and extinction of Faraday waves at each acceleration and frequency are unrelated to the stability regime of holes because persistent holes also exist in a regime where Faraday waves do not exist. For PPG+FS 8%, this was not the case. Using the container with a diameter of 126 mm and PPG+FS 8% suspension of 5.03 mm depth, the result of the boundary between stable and unstable and the onset and extinction of Faraday waves are shown in Figure 4. Persistent holes always entails Faraday waves. Also, the plot shows that the trend of the stability boundary and onset and extinction of Faraday waves are similarly increasing. This does not mean that Faraday waves are solely responsible for persistent holes. Faraday waves, though, must be taken care of with consistency. When Faraday waves form a symmetric circular shape (Figure 5 (a)), which depends on the boundary of the container, holes closed in a few seconds. This can be thought as the pressure caused by the Faraday waves is uniform in all directions and greatest. Holes that were confirmed to be persistent were always in a condition when Faraday waves were no longer followed a shape of the container (Figure 5 (b)). When Faraday waves are in a pattern no longer following the container shape, interference between Faraday waves from different directions generate a relatively weak pressure at a certain direction. In order to be consistent, we took every measurement after waiting 30 seconds until the Faraday waves become perturbed and resulted in wave patterns shown in Figure 5 (b).
Figure 4: Phase diagram for polypropylene glycole and fumed silica mixture suspension (8%) as a function of acceleration and frequency explored at several states. The acceleration is the root mean squared value. The container diameter is 126 mm and the depth of the suspension is 6.164 mm. Faraday waves appear with increasing acceleration at $\Delta$, and disappear with decreasing acceleration at $\nabla$. The boundary when perturbed holes starts to become persistent has an increasing trend. This graph implies that it is possible that persistent holes are Faraday wave dependent.
Figure 5: Two different examples of Faraday waves that appeared frequently during the experiment. (a) Symmetric. The pattern follows the shape of the container. Perturbed holes are never persistent under this condition. (b) Faraday waves that are container shape independent. When vibrating motions are waited long enough, Faraday waves no longer follow a circular shape (container shape) and forms to a shape as above.

D. Relaxation Time of Fluid

When the acceleration at the boundary of the phase diagram (acceleration and frequency) was re-measured, the values varied significantly. Possible factors that might cause this effect were explored. We prepared PPG+FS suspension in the container (d=126 mm) at a depth of 3.75 mm. For this setup, the minimum acceleration to create stable holes at 45 Hz was 17.02g. First, the vertical vibration time was varied and the minimum acceleration was re-measured to see if the measuring time would be a reason. The apparatus requires sufficient time for the values to be read off. So this was reasonable to be guessed as a cause. However, it turned out that this was not the reason since no difference were observed at low accelerations.

Eventually, we found that at a given frequency, the lowest acceleration that creates
persistent holes when increasing the acceleration starting at a low acceleration was different from lowering the acceleration starting at a high acceleration. Thus, the stability of holes depend on the path. It is thought that the structure of particles in the suspension change when high acceleration is experienced. To avoid this effect, we needed to know how long it takes for the suspension to recover its original behavior. To measure this time value, a certain high acceleration above 17.02g at 45 Hz was set constant and was vibrated at a fixed amount of time (5 minutes). Then, the equipment was turned off for an amount of time and was turned on again at 17.02g with 45 Hz. From this procedure, we found that an amount of time for the liquid to relax is required for a perturbed hole to be persistent again at 17.02 g. The minimum amount of time needed to relax for each acceleration values of 5 minute vibration was found by repeating the experiment with different relaxation times (time of vibration turned off). The result is shown in Figure 6. Figure 6 shows that for very high accelerations, the time it requires to recover its original value takes roughly 100 seconds. Based on this knowledge, our measurements that were taken subsequently, were turned off for a sufficient amount of time (2 minutes) whenever the fluid was previously experiencing high acceleration.
Figure 6: Relaxation time requirement for polypropylene glycole and fumed silica mixture suspension (8%) that experience a certain acceleration at frequency 45Hz. The acceleration is the root mean squared value. If the suspension experience higher acceleration than the boundary point (boundary between stable/unstable) in the phase diagram as a function of acceleration and frequency, the boundary point no longer becomes stable. Relaxation time is required when the acceleration of the suspension is above the boundary point in the phase diagram. For example, if the suspension experienced an acceleration of 19 (g) for 5 minutes, the amount of time that the vibration should be turned off is 40 ± 5 (seconds).

E. Phase Diagrams for Different Container Diameters

When perturbed holes created by a puff of air become persistent, the initial hole stabilizes at a size comparable to the size of the container (~centimeters). This is different from cornstarch, which the size of holes are in orders of millimeters and is independent of the container size[1]. Figure 7 shows examples of how the holes look like at stable conditions in
different container sizes. Also, unlike cornstarch, the holes are not circular. The net pressure
is different at different directions since the conditions are not homogeneous at all directions
(e.g. Faraday wave patterns are not symmetric under all directions).

Figure 7: Examples of persistent holes of polypropylene glycol and fumed silica mixture
suspension (8%) vertically vibrated. (a) Container diameter 126mm, depth 5mm (49Hz 22.5
g) (b) Container diameter 165mm, depth 5mm (49Hz 25.1g). Notice that the hole sizes are
large (order of centimeters) compared to cornstarch (order of millimeters[1]).

Phase diagrams as a function of acceleration and frequency were obtained by measure-
ments of different container size. The depth was kept identically at 5 mm. And the same
batch of PPG+FS (8%) suspension was used. First, for the container size of d=126mm
puff of air was blown at pressure of 40 (psi) for 3 seconds. The result is shown in Figure
8 (dotted line, blue). Next, the container size of d=165mm was set at the same condition
(depth, blowing air pressure and time). Unfortunately, even at maximum acceleration at
which the equipment can hold (40g), the air pressure was too weak that holes closed in a
few seconds. Higher blowing air pressure at 100 psi for 3 seconds was used instead for the
large container. The result is shown in Figure 8 (solid line, red). Figure 8 illustrates that
different container sizes result in different stability boundaries in the phase diagram as a
function of acceleration and frequency.
Figure 8: The boundary line of stable (above) and unstable (below) in a phase diagram for polypropylene glycole and fumed silica mixture suspension (8%) as a function of acceleration and frequency for two different container diameters. The acceleration is the root mean squared value. The boundary line for large container (diameter 165mm) is indicated in a red solid line and small container (diameter 126mm) is indicated in a blue dotted line. The large container requires higher acceleration than the smaller container at a given frequency.

According to Deegan [3], the hydrostatic pressure pushes the fluid into the hole. Since the pressure is an intensive variable, the volume of the container should have no effect. Several explanations were proposed, however, yet not confirmed. This still remains to be solved (Possible explanation is provided in Appendix I).
F. Phase Diagrams at Different Depths

![Phase Diagram](image)

Figure 9: Phase diagram for polypropylene glycol and fumed silica mixture suspension (8%) as a function of acceleration and frequency at different depths. The acceleration is the root mean squared value. The container diameter is 126 mm. Stability boundary at depth 4.03 mm is illustrated in a green dashed line. Stability boundary at depth 5.03 mm is illustrated in a red dotted line. Stability boundary at depth 6.05 mm is illustrated in a red solid line.

A container with a diameter 126 mm was used and a puff of air was blown at 40 psi for 3 seconds. PPG+FS (8%) suspension was filled into the container at three different depths: 4.03 mm, 5.03 mm, and 6.05 mm. The phase diagram as a function of acceleration and frequency is shown in Figure 9. The stability boundary for the smallest in depth at 4.03 mm is illustrated in a green dashed line. The depth at 5.03 mm is illustrated in a blue dotted line. And depth at 6.05 mm is illustrated in a red solid line. The result shows that the stability boundary shifts upwards as the depth of suspension increases.
As the fluid in the container becomes larger in depth, the pressure that pushes the holes to close increases. This pressure is hydrostatic pressure due to gravity

\[ P_{in} \sim \rho g h \]  

(2)

Where, \( \rho \) is the density of the fluid, \( g \) is the gravitational constant, \( h \) is the depth of the fluid. Using dimensional analysis, the pressure that pushes the fluid to not close can be roughly understood as

\[ P_{out} \sim \frac{M \times a}{A} \sin(2\pi f t) \]  

(3)

where, \( a \) is the acceleration \( f \) is the frequency of the vertical vibration, \( A \) is the area where the pressure is experienced, thus the boundary of the hole, and \( M \) is mass of the fluid. Thus, higher acceleration must be applied to generate higher pressure that equal to \( P_{in} \) and remains as a hole.
G. Phase Diagrams for Different Rheological Values

Figure 10: Viscosity of polypropylene and fumed silica mixture (8%) as measuring during an increasing shear stress ramp. The blue solid line indicates the suspension prepared for 2 minute blending time and the red dotted line for 3 minute blending time. The error bars correspond to variation of two measurements of the same sample.
Figure 11: Phase diagram for polypropylene glycol and fumed silica mixture suspension (8%) as a function of acceleration and frequency. The acceleration is the root mean squared value. Container size diameter was 126 mm and depth 5 mm. The blue solid line is the stability boundary line for batch of 2 minute blending time. The red dotted line is the stability line for batch of 3 minute blending time. Faraday waves appear with increasing acceleration at $\Delta$, and disappear with decreasing acceleration at $\nabla$. The boundary when perturbed holes starts to become persistent has an increasing trend. The error bars correspond to variation of highest measured unstable point and lowest measured stable point.

Merkt et al. concluded that persistent holes are created by shear thickening of the fluid. If the suspension has a different shear thickening behavior the stability boundary of the phase diagram as a function of acceleration and frequency would change as well. The viscosity as function of shear rate was measured with a stress controlled rheometer (AR-2000EX, TA-instruments) with an acrylic cone of diameter 60mm and angle $\alpha = 1°58'59''$ at temperature 25°C. We found that different blending time when preparing the PPG+FS
mixture results in different rheological behavior (subsection a). Generally, the viscosity shifts down with less dramatic shear thickening when the blending time with the commercial blender was increased. We used the PPG+FS (8%) suspensions that were prepared for previous experiments: blending time 2 minutes at highest speed. The data of the phase diagrams and rheological data were first collected. Then, this suspension were blended another extra minute at highest speed with the blender. The shear rate versus viscosity by the rheometer is shown in Figure 10. The original PPG+FS (8%) batch prepared with 2 minutes of blending time is illustrated in a blue solid line and batch blended of total 3 minutes is illustrated in a red dotted line. As expected, the viscosity is shifted down and the shear thickening is less drastic. Since the shear thickening is weakened, higher acceleration at a given frequency is required for large enough force (or pressure) to balance the hydrostatic force (or pressure). The result of the phase diagram shown in Figure 11. Also, the original PPG+FS (8%) batch prepared with 2 minutes of blending time is illustrated in a blue solid line and batch blended of total 3 minutes is illustrated in a red dotted line. As Merkt et al. predicted, less shear thickening leads to higher stability boundary. This is because higher acceleration is required to compensate the low shear thickening.

III. CONCLUSION

With a mixture of polypropylene glycol (PPG) and fumed silica (FS), measurements were made under various conditions. Unlike cornstarch, difficulties occurred since we could not disregard Faraday waves and a certain amount of time was required once the fluid exhibited higher accelerations than the minimum acceleration for persistent holes. A larger size container required a higher acceleration to create persistent holes when the other conditions are identical. This was unexpected and needs appropriate theoretical explanation. Phase diagrams for different depth resulted as we qualitatively predicted. It the liquid is deeper, larger force from the hydrostatic pressure is created and higher amount of force by vibrations is required to balance. Also, the relationship between shear thickening and persistent holes were understood by comparing the phase diagrams with two different rheological behaving liquids.

Still, reproducing the exact behaving fluid still remains a problem. A slight amount of blending time difference leads to large deviations of viscosity shifts. Also, more set of data
for each section need to be gathered in order to understand the results not only qualitatively, but also quantitatively. Also, varying the frequency while the acceleration is fixed could be done to see if there is any path dependence.

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