Settling and spreading behaviour of particle clusters in quiescent liquids in confined vessels

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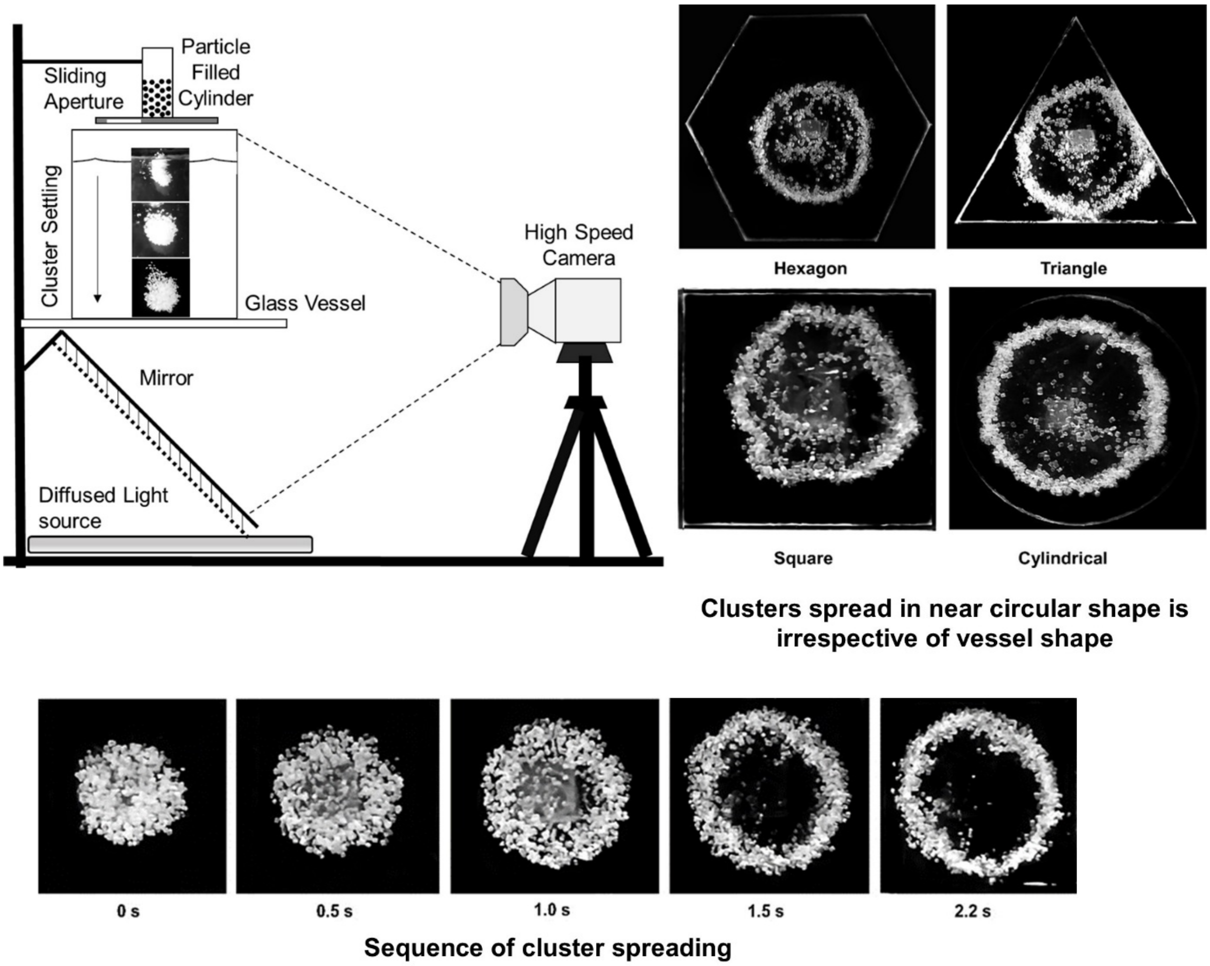
Abstract

Here we report experiments on particle cluster settling at high Reynolds number in quiescent liquid contained in a vessel. The particles were observed to spread at the vessel bottom surface in a near-circular annular shape after settling irrespective of the shape of the vessel cross-section and particle shape, size, and types. Effect of different parameters such as mass, type and aspect ratio of the particles, height, and viscosity of liquid was investigated on spreading behaviour. Formation of the hemispherical bottom cap of the cluster that bounces upon hitting the vessel bottom surface was found to be responsible for the final circular annular shape of the settled structure. Particle leakage from the cluster was seen in the form of a tail. In the liquid having viscosity beyond 100 cP, cluster breakage was observed that resulted in hindered settling and asymmetric shapes of finally settled particles. The observations are useful to understand the overall area over which settling and spreading of such clusters can be observed.

Keywords: Particle cluster, Temporal evolution, Settling, Spreading

Graphical abstract:



Highlights

- Particle cluster settles at high Reynolds number in quiescent liquid in a vessel
- Particles spread in a near-circular annular shape at the vessel bottom surface
- The near-circular shape doesn't depend on vessel cross-section particle properties
- Hemispherical bottom cap of the cluster was responsible for circular annular shape
- Effect of different parameters was investigated on spreading behaviour
- For high viscosity liquid, cluster breakage was responsible for asymmetric shapes

1. Introduction

The motion of single particles or particle assemblies in a liquid in a constant gravity field has been studied extensively over the last 170 years (Adachi, Kiriyama, & Yoshioka, 1978; Jayaweera, Mason, &

Slack, 1964; Metzger, Nicolas, & Guazzelli, 2007; Richardson & Zaki, 1954; Stokes, 1851). Several processes involving the motion of particle clusters in liquids are found in nature. Such systems are pertinent to the dynamics of particulate gravity currents which have applications in oceanographic (turbidity currents and study of carbon fluxes to the sediments in the ocean, lakes and reservoirs), geological (debris flows, pyroclastic density currents; lava flows from volcanic eruptions), and environmental (pollutant-laden wastewater treatment) scenarios. Moreover, the settling study of such particle-fluid systems has significant commercial applications in fields as diverse as effluent dispersal, food processing, dredging waste removal, and mine tailings (Daniel, Ecke, Subramanian, & Koch, 2009; Hanson, 1985; Huppert, 2006; Johnson, Li, & Logan, 1996; Peakall, Felix, McCaffrey, & Kneller, 2001). Understanding the nature of settling and spreading of such clusters in water is also important for undesired situations like airborne accidents of flights over the sea as rapid settling can restrict the area of investigation. This work performed using controlled experiments gives some insights in cluster sedimentation and settling in liquids.

When a swarm of particles is allowed to settle in quiescent fluids, it forms a cohesive entity called as cluster (also referred as blob or drop) (Adachi et al., 1978; Alabrudziński, Ekiel-Jeżewska, Chehata-Gómez, & Kowalewski, 2009; Kaye & Boardman, 1962; Machu, Meile, Nitsche, & Schaflinger, 2001; Nitsche & Batchelor, 1997). The motion of the cluster is often considered to be analogous to the settling of a drop of a viscous fluid in a comparatively lighter liquid where a jump in the value of the particle concentration is related to the liquid-liquid interface (Adachi et al., 1978; Alabrudziński et al., 2009; Machu et al., 2001; Metzger et al., 2007). The ambient flow within such a settling cluster geometry was reported to be similar to the toroidal circulation observed in a settling liquid drop (Abade & Cunha, 2007). The behaviour of a cluster of particles is different from a single particle in such scenarios, mostly because of the relative motion of particles in the cluster and the relative motion of clusters. The liquid motion at any point in the cluster is dependent on the relative velocity of the constituent particles and how distant the particles are from the point. Each constituent particle carried some amount of fluid along with it and convected by the velocity fields produced by the other constituent particles. This results in velocity enhancement of constituent particles and of the whole cluster as well (Adachi et al., 1978). The fall velocity of the cluster was observed to be higher at all times than the individual particles, and the enhancement in the falling rate increased when the particles were more closely packed (Jayaweera et al., 1964). Settling velocities of particle clusters were found increase than the single sphere stokes law model based on cluster size and number of particles. As the cluster takes a macroscopic identity, cluster Reynolds number (Re_c) based on the macroscopic scales, i.e., cluster radius and velocity, was often used for flow characterization instead of the particle Reynolds number (Adachi et al., 1978; Alabrudziński et al., 2009; Machu et al., 2001). Moreover, the added mass force plays an important role in the overall force balance.

At very low/zero *Re,* a cluster would maintain spherical shape while with increasing *Re* significant temporal evolution of the cluster is observed. For a certain time, depending on the specific system, the particles recirculate and stay together within a single almost spherical cluster/blob, after which the cluster forms a torus, then a ring-like structure and then eventually disintegrates. This phenomenon has been reported by several authors in different settling systems over a range of Reynolds number (i.e. 1×10^{-4} to 10) for particle sizes over a range of few microns to millimetres (Adachi et al., 1978; Machu et al., 2001; Metzger et al., 2007). The particles were observed to move with respect to each other inside the cluster, and the cluster significantly changed its shape (Alabrudziński et al., 2009) while responding to the various forces acting on it. Machu et al. (Machu et al., 2001) reported the lower part of the cluster takes the shape of a roughly hemispherical cap, while the upper part resembles the conical shape of the laminar jet formed during the injection process. At comparatively higher *Re*, i.e., 93 to 425, Daniel et al. (Daniel et al., 2009) observed that the variance of the cluster diameter grows quadratically at the beginning and attain a slower sublinear regime after some time. The temporal evolution of cluster was reported to be significantly dependent on the cluster *Re* and the initial number of particles in the cluster.

During the past few decades, there have been several notable experimental as well as theoretical advances in the understanding of the dynamics of motion of cluster/blob made of particles made of micron to milli meter ranges (Adachi et al., 1978; Alabrudziński et al., 2009; Jayaweera et al., 1964). These investigations were focused on $Re \leq 1.0$ in the creeping flow regime, where the inertia of particles and fluid and hydrodynamic interactions can be neglected. For $Re > 10$), both viscous, inertial, and buoyancy effects become significant and relative motion of the particles increases (Jayaweera et al., 1964). Wake mediated interactions starts becoming dominant with increasing Re_c of the system (Daniel et al., 2009; Feng, Hu, & Joseph, 1994). It can be said on the basis of the reported literature that beyond the creeping flow regime, the hydrodynamic interactions between particles mediated by the fluid become non-linear and demonstrate complex dynamics, for which our understanding of such systems is still rudimentary.

In the recent years, considerable attention has been focused on understanding the motion and evolution of fluid-particle systems such as a spherical settling cluster at low Reynolds numbers (Abade & Cunha, 2007; Adachi et al., 1978; Alabrudziński et al., 2009; Daniel et al., 2009; Di Felice & Kehlenbeck, 2000; Machu et al., 2001; Metzger et al., 2007; Nitsche & Batchelor, 1997) but the spreading behavior of particle cluster upon hitting the bottom surface of the vessel apparently received little attention. Daniels et al. (Daniel et al., 2009) only reported that even a localized release of particles into a quiescent fluid might give rise to sediment spreading on a greater section of the vessel bottom surface due to radial cluster expansion resulting from the source-flow interactions of the particles. To the best of our knowledge, to date, a detailed study of the corresponding observations of particle spreading from the clusters is not reported. Instead of settling at the centre of the vessel, particle clusters were observed to form a ring-like structure around the vessel centre in our experiments. Therefore the goal of this paper is to perform a detailed experimental analysis of settling at short time scales and spreading nature of the solid particle clusters in quiescent liquids in confined vessels. The focus is to investigate the solid deposition patterns at the vessel bottom surface. The effect of different parameters such as vessel geometries, wall interactions, and liquid properties over the settled geometry was studied experimentally. This is a purely experimental work, and in-depth theoretical understanding by means of numerical simulations is out of the scope of this study.

In view of this detailed introduction, the manuscript is organized as follows: the next section details out the experimental methods and the implemented data processing techniques in this work. Subsequently, a description of cluster settling and spreading phenomena and effects of different parameters on the spreading behaviour were discussed in detail. Finally, important findings are summarized.

2. Experimental

2.1. Experimental setup

Particles filled in a small cylindrical cavity were released instantaneously at the center of the vessel filled with a quiescent Newtonian liquid from an elevation of 2 cm and were allowed to settle. The particle trajectory and settling pattern at the vessel bottom surface were monitored. Glass vessels of different sizes and shapes were used for performing the experiments. An in-house developed motorized system was used for the release of particles to eliminate manual variations in particle release. Particles were loaded into a cylindrical reservoir, and a thin plastic sheet holding the particles was slid instantaneously using a servo-controlled motor to release all the particles so that the cluster descents in the vessel. Schematic of the experimental setup is shown in Fig. 1. The particles travel about 2 cm in the air before entering the liquid. The time needed to release the particles was less than 50 ms. Due to the rapid movement of the sheet, no lateral movement of particles was observed, and this technique significantly reduced the entrainment of air bubbles inside the cluster. Particle trajectory inside the liquid phase after feeding during its settling and the final settling pattern formed was monitored using a Chronos 1.4 high speed camera (Kron Technologies Inc., Canada). The evolution of the settling cluster was observed via an angled mirror placed directly below the cluster's path of travel. The mirror was kept at 45° angle at the bottom of the vessel to observe the side-view and bottom-view of the glass vessels. Visualization of settling and spreading time and pattern of the solid particles in water was performed by video processing (Virtual Dub and iMovie software), and image analysis was performed using Image J and Matlab softwares.

Fig. 1

Typical cluster settling studies will involve solid particles, liquid and a vessel in which the particles settle. Considering this simple set of variables, the parameters that are relevant can be varied to understand cluster settling in general, include: (i) particle size and shape, (ii) mass of particles, (iii) vessel liquid level, (iv) vessel shape and shape, (v) viscosity of liquid.

The choice of particles was based on the need to have dense particles that settle and secondly, the shape can be almost consistent within a cluster. For these experiments it was necessary to have particles having different shapes viz. cubical, spherical and ellipsoidal, which was made possible by choosing materials like sugar, glass beads and rice grains (see Fig. 2) , respectively. This allowed the aspect ratio to be varied from 0.22 to 1, which is defined as the ratio of largest diameter and the smallest diameter orthogonal to it.

The initial mass of particles varied from 6 g to 16 g. Since our interest was in exploring the patterns of sedimented cluster after it settles, it was expected that the height of liquid and also the shape of vessel would make a different in the final nature of particle settling. The liquid height in the vessel was varied from 6 cm to 14 cm. It is known that when a liquid fall on a flat surface through a faucet, the splashing takes the shape of the vessel in which it is falling. For the case of settling cluster such effects are not known and it all would depend on the relative wave dynamics of liquid and the cluster. Hence vessels having various cross-section shapes viz. square, circular, triangular, rectangular, hexagonal, etc. were used in two different sizes (full dimensions in the Supplementary Data) while other parameters were kept constant. When not studying the effect of different parameters, typical experiments were carried out in a 15 cm × 15 cm square cross-section glass vessel with 10 cm liquid level and using 10 g cubical sugar particles of 2.4 mm average size. In all the cases, the equivalent diameter of each vessel was at least 6 times larger than the cluster diameter.

The choice of liquid was primarily in terms of viscosity as interaction of cluster with viscous liquid is expected to be different in liquids of higher viscosity. Galileo Number (*Ga*) is a dimensional group representing a ratio of gravitational forces and viscous forces present in the system. The Galileo Number of the system is varied from higher values (150–180), where viscous forces were negligible compared to gravitational forces to lower values (<1) where viscous forces are comparable to the gravitational forces. Solutions of UCON (water soluble lubricant from Dow Chemicals) in water in different ratios were used for varying viscosity of the liquid in the range of 1–516 cP, without changing the density. The liquid viscosities were measured by a cone and plate viscometer.

Experiments were performed in triplicate to ensure reproducibility.

2.2. Analytical Methods

Settling time, spreading time, average spreading size (viz. diameters of the inner and outer circles of the annulus, for the cases where it forms a circular annular shape) were measured as a response. Settling time was defined as the time needed for the cluster to reach the bottom of the vessel from the time it enters the liquid phase and spread time was defined as the time required to form the final settled shape after the cluster reaches the bottom surface. The time-averaged cluster settling velocity was determined as the total vertical displacement of the cluster divided by the settling time. Image processing was performed by Matlab in order to detect the inner and outer edges of the of the settled shapes. The minimum circumscribed circle (MCC), defined as the smallest circle, which encloses the whole of the roundness profile and the maximum inscribed circle (MIC), defined as the largest circle that can be inscribed inside the roundness profile. A schematic representation of the same has been shown in Fig. 3. The average diameter and roundness were checterised as the mean and ratio of the MCC and MIC, respectively. The diameters are presented in a dimensionless form throughout the manuscript by dividing it with the hydraulic diameter of the vessel cross-section.

As different types of particles having a wide range of aspect ratios were used for the experiments, the equivalent diameter of the particles (Jennings & Parslow, 1988) was used to characterize the particle length scale instead of particle size. The cluster Reynolds number (Re_c) was estimated as $\left(\rho_f V_{Cluster} D_{cluster}\right)/\mu_f$. Gravitational velocity scale is defined by $V_g = (|\rho_p/\rho_f - 1|Dg)^{½}$ and gravitational time scale can be defined as $\tau_g = D_{cluster}/V_g$ (Uhlmann & Doychev, 2014).

Fig. 3

3. Results and Discussion

3.1. Settling of clusters

While falling under gravity in a quiescent liquid, the group of particles is found to form a compact cluster under a wide range of experimental conditions. The cluster of particles was observed to rapidly evolve into a nearly spherical shape upon entering the liquid surface. It was also observed that the cluster undergoes deformation thereby losing the cohesiveness of a cluster and it undergoes transition in a very short time before it settles. To understand the settling behavior of the cluster it is important to characterize the settling and terminal settling velocity. It is known from the literature that the motion and velocity of a settling particle cluster are different from the individual particles in the creeping flow regime (Adachi et al., 1978; Alabrudziński et al., 2009; Machu et al., 2001; Schaflinger & Machu, 1999). However, Daniel et al. (Daniel et al., 2009) reported a similar order of magnitude between the settling velocity of the cluster and individual particles outside the creeping flow regime. In the current work, the settling velocity of the cluster never reaches terminal settling velocities of the individual particles (0.33 to 0.93 times) or the terminal settling velocities of the clusters (0.28–0.5 times, assumed spherical and treated as single particles). The estimated terminal settling velocity of the clusters is found to be 2.22–3.09 times of the average cluster velocity for our experiments, which is higher that available literature (On average 1.58 times) (Bhatty, 1986). Significantly lower cluster velocities in comparison to the respective terminal settling velocities in our experiments can be explained by significantly lesser cluster travelling path in the liquid (settling distance is 1.71 to 5 times the cluster diameter compared to a minimum of 9.38 times in Daniels et al. (Daniel et al., 2009)).

All the experimental data was carefully analysed using the images obtained from a Chronos 1.4 high-speed camera at 1000 fps to monitor the transient variation in the velocity of cluster during its settling. Since the impact of the cluster on the air-liquid interface followed by continuous displacement of water actually helped in generating a vortex with transient variation in the fluid velocity. Typical variation in the transient cluster velocity is shown in Fig. 4, which includes the settling velocity of the cluster from the time it touches the air-liquid interface, followed by its motion through the liquid and finally the collision with the bottom wall. Four different regimes (R1-R4 in Fig. 4) are evident in the travel path of the cluster. Initially, the velocity continues to increase (R1) due to inertia until a hemispherical cap gets formed in the lower half of the cluster. Then velocity gradually decreases (R2) due to loss of energy while crossing the interface as well as due to viscous dissipation of cluster momentum. While the cluster still in suspended form reaches close to the bottom surface some particles come apart from the cluster which momentarily increases the velocity of the cluster (R3). Finally, a sudden drop in velocity is observed (R4) due to the buoyancy of the liquid coming from the bottom wall surface that leads to disintegration of cluster eventually subjecting individual particles to the motion. Among the four regimes of the cluster travel path, the time needed for the viscous dissipation region was the found to be the highest.

Fig. 4

The time scale for the initial rise in the velocity was seen to be a function of the mass of cluster, while the time scale for viscous dissipation of cluster energy was seen to depend primarily on the liquid level. The time scale for the regime of cluster fragmentation was always the smallest among all. This particular trend was observed for all the experiments. The gradient of the velocity with time was seen to be a typical quadratic polynomial having two points of inflection and no symmetry.

The cluster diameter in our experiments was in the range of 2 cm to 4.6 cm. The recirculation of particles was observed in the cluster, which also provided a macroscopic identity to it. We have explored this feature by observing uniformly mixed distribution of settled particles of different colors when the reservoir was filled up in layers with different colored particles (see Fig. 5(a)). The cluster was seen to undergo internal circulation as depicted in Fig. 5(b) thereby mixing particles of different layers. Further the phenomena continue and particles of different colours were seen to settle along the annulus clearly indicating that significant mixing of particle layers happens in the cluster before it settles. While settling in the liquid, the lower part of the particle cluster forms a roughly hemispherical cap. While most of the particles were retained together, a few particles tend to leave the cluster during settling and remain in the tail portion, which is known as particle leakage (see Fig. 5(b)) (Abade & Cunha, 2007; Adachi et al., 1978). Particle leakage from the cluster primarily occurs because of the instabilities in the cluster when interacting particles undergo transient variation in the velocity, trajectories, and waves generated in the fluid due to entry of the cluster. The particles which are located in the outer layer of the toroidal circulation, tend to escape from the toroidal circulation and are dragged by the outside flow to form a tail of particles at the rear section of the cluster (see Fig. 5(c)). With the increasing initial mass of the particles, the tendency for particle leakage is observed to decrease and the clusters move downward with lesser deviation to its initial configuration. This is due to the lesser departure from closed streamlines of the toroidal circulation with increasing number of particles (Metzger et al., 2007). An important quantity which characterizes the near-spherical cluster formation is a critical mass (or number of particles) required to form a coherent spherical cluster. We had observed that when the mass of cluster was lower than the critical mass, the clusters were having a distorted/asymmetric shape and tended to disintegrate significantly and rather rapidly before reaching the vessel bottom.

Fig. 5

3.2 Spreading of clusters at vessel bottom

The entrance of the cluster in the liquid created ripples on the water surface, and as it travels towards the vessel bottom, the displacement of liquid in the downward direction along with the cluster helps generate a temporary circulation in the liquid. A strong interaction between sediment transport and the fluid dynamics due to the spatiotemporal displacement of water was observed during the spreading and settlement of the cluster particles. As the cluster hits the vessel bottom, the constituent solid particles spread out almost symmetrically under the influence of particle driven gravity currents under the balance of inertial and buoyancy forces. Particle driven gravity currents are generated by the release of a swarm of particles along with the interstitial liquid into a lighter ambient fluid. In such a scenario, the flow is driven by the difference between the bulk density of the particle current and the density of the ambient fluid in the vessel. The length of the particle gravity current is determined by the balance of buoyancy and inertial forces. The current length is very much higher than its thickness. Though it is reported that a propagating gravity current without particles shows a uniform velocity profile for $Re \gg 1$, when particles drive the flow in addition to the advective effects, they fall out of the flow losing some of the energy to viscous dissipation, and the buoyancy force continually decreases (Huppert, 2006). First, the particulate current propagates through an inertia dominant regime, then as the current reaches a certain length, the velocity and height at the tip of the current decreases. As a result, the viscous forces acting along the vessel bottom surface become more important, and the carried particles are tended to settle from the current there (Bonnecaze, Huppert, & Lister, 1993). Fig. 6 shows the sequences of the cluster spreading with time, after colliding with the vessel bottom surface. In the following section, effects of different parameters viz. initial cluster mass, vessel liquid level, liquid viscosity, vessel shape and size, particle size, shape, and type on the spreading behavior has been presented and discussed in detail.

Fig. 6

3.2.1 Effect of the mass of particles

Due to the interparticle interaction in water and due to viscous effects, solid particles come near to each other and form a nearly spherical shape. A variation in the mass/number of solid particles affects the settling and spreading time. Increasing the mass of the solid particles resulted in a decrease in the settling time and an increase in the settling velocity due to higher inertia. Here the effects of added mass come become critical, which is the additional mass that an object appears to have when it is accelerated relative to a surrounding fluid (Brennen, 1982). With an increasing number of particles the added mass increases and adds to the effective inertia.

The spreading time, as well as the average inner and outer diameter of the final shape of settled particles, were found to increase with increasing mass of solid particles irrespective of different vessel sizes (see Fig. 7(b)). With an increasing number of particles and the size of the cluster, the bulk density of the resulting particulate current increases. This increases buoyancy, which is the driving force for the spreading of particles. With the increasing initial mass of particles, the roundness of the settled structure becomes closer to unity, i.e., the settled shape becomes closer to a circular annulus.

Fig. 7

3.2.2 Effect of vessel liquid level

The settling and spreading time were observed to increase with the increasing liquid level. This is due to the increased hindrance provided by the liquid on the motion of particles. This hindrance is due to pressure force and also the viscous force. Though Bonnecaze et al. (Bonnecaze et al., 1993) found that lengths of the particulate currents in shallow surroundings were greater than compared to deep surroundings, Fig. 8 shows that the variation in liquid level has no significant effect on the inner and outer diameter of the settled shape. The roundness value of the settled shape tends closer to unity with an increasingly fluid level, which represents that the settled shape becomes more symmetric. Higher settling time provides more time to recirculate and forms a cluster of symmetric shapes. When the cluster shape is closer to a perfect sphere, with an impact on the bottom surface, the particles settle more symmetrically to form a near circular annulus.

Fig. 8

3.2.3 Effect of Vessel shape and size

Vessels of different geometries in two different sizes were used to carry out particle settling experiments. As the water level was kept constant in all the experiments, the liquid volume in the vessel and cross-sectional area of the bottom surface was dependent on the size and shape of the bottom surface. It was previously observed that the dimensionless diameter of the settled circular pattern increases linearly with increasing mass of particles (see section 3.2.1). However, there is a limiting value for the mass of particles to form a symmetrical settled shape where the wall effect is insignificant, beyond which the particles gets inhibited and settle along the wall (see the Supplementary Data). The mass of the particles used to form clusters were changed for different vessel sizes as the limiting value for the mass of particles is dependent on the size of the vessel bottom surface area or the proximity of the vessel wall. Keeping this in consideration, in order to eliminate the effect of different mass of particles in the current set of experiments, providing a better understanding for the effect of vessel shape and size, the outer dimensionless diameter used in the following analysis was normalized by the mass of particles used for the experiments. This is to be noted that the normalization of the dimensionless diameter was performed only to separate out variation in the mass of particles in these set of experiments. Fig. 10(a) shows that irrespective of the vessel shape, the normalized diameter and normalized area of the settled shape decrease with increasing cross-sectional area of the vessel bottom surface. With an increasing cross-sectional area of the bottom surface, the proximity of the vessel walls decreases and waves formed by the cluster movement are strongly reflected resisting the spreading of the particles towards the wall. However, it was interesting to observe that the settled shape is found to be close to a circular annulus despite the different shapes of the vessels (see Fig. 9), if the mass of the particles is lower than the limiting value. Upon entering quiescent liquid, the swarm of particles forms almost like a sphere-like cluster and carries with it a wake that sets circulating flow past the cluster. Although the cluster disintegrates, the wake facilitates a circular vortex that makes these particles undergo a circular motion and hence they settle in identical circular shapes once the effect of settling velocity becomes more prominent than the fluid velocity in the vortex. This implies that though the wall proximity influences the normalized diameter of the settled shape, the spreading pattern is not altered. It is also evident from Fig. 10(b) that the square shape vessel results in enhanced spreading of the particles in comparison to other vessel shapes. The extent of reflection of the waves after the cluster enters the liquid is stronger for the geometries with a lesser number of reflecting walls and hence when compared to other geometries, square shape vessel results in enhanced spreading, while for the triangular vessel the secondary waves that reflect due to lesser confinement limit the spreading of particles.

Fig. 9, 10

3.2.4 Effect of particle size, shape and type

To understand the effect of particle type, size, and shape on the settling phenomena, different particle types of various sizes and aspect ratios were used for the settling experiments when other parameters were kept constant. Fig 11(a) shows no significant deviation in the dimensionless outer diameter of the settled shape for particles of various diameters (ranging from 492 μm to 3643 μm average equivalent diameter). In general, at ambient conditions, the settling velocity of a particle decreases if the shape of a single particle deviates from a sphere. (Komar & Reimers, 1978). The drag force on the individual particles would change with a significant variation in the aspect ratio. However, despite these facts on the settling behaviour of individual particles, it was interesting to observe that the spreading behaviour is not being affected by the varying particle type, size and aspect ratio. Despite using round shaped glass particles and elongated rice grains ($0.22 <$ aspect ratio < 1), the dimensionless outer diameter of the settled shape only varied over a close-range of 0.742 ± 0.035 (4.7% deviation; see Fig. 11(b)). We can speculate that the macroscopic identity of the cluster was the reason why the dimensionless diameter of the settled cluster was independent of the shape, size, and types of the constituent particles. Though the temporal evolution of the cluster was affected to some extent with different sized and shaped particle used for settling, the hemispherical bottom shape of the cluster (which is the crucial criteria for the symmetric shape formation) was not affected, despite using particles of different shape, size, and type.

Fig. 11

3.2.5 Effect of fluid viscosity

Cluster settling experiments were performed with ambient liquids of increasing viscosity. The settling velocity of the cluster was found to decrease with increasing viscosity of the liquid, and even the spreading time was observed to increase (see Fig. 13). This is due to higher friction/ resistance provided by the liquid to the particle cluster. It is also evident from Fig. 12 that with increasing viscosity of the liquid the terminal settling velocity of the cluster in the presence of buoyancy force (definition and equations in the Supplementary Data) becomes comparable to the stokes flow terminal settling velocity, indicating a shift of flow behaviour towards stokes flow or creeping flow. The dimensionless diameter of the settled shape also decreased with increased viscosity until a certain viscosity value, and then it was found to increase. The existence of a limiting viscosity ($\sim$100 cP) was witnessed beyond which near-spherical cluster formation was not observed. When the fluid viscosity is increased over the limiting viscosity value, destabilization of the cluster was found to occur, and the cluster was not able to maintain spherical shape (see Fig. 13(a)). Galileo Number is a dimensional group representing a ratio of gravitational forces and viscous forces present in the system. For sugar particles Galileo Number (*Ga*) ranges from 166 -180 in water and with increase in viscosity of the liquids it decreases up to 0.23. The regime change and the limiting viscosity is observed when the *Ga* comes close to unity, which means viscous forces in the system becomes comparable with the gravitational forces, resulting in destabilization of the clusters.

As the hemispherical bottom cap of the cluster was not formed, when hitting the vessel bottom surface, the constituent particles of the cluster spread in an asymmetric manner (see Fig. 13(b)). A significant deviation from a circular settled structure was observed. The initial decrease in the dimensionless diameter was due to a lesser density/intensity of the particulate gravity current, but after a limiting viscosity value when *Ga* values close to unity (see Fig. 14(b)), due to asymmetric spreading, the dimensionless diameter of the settled shape increased (see Fig. 14(a)) and the shapes deviate further from a perfect circular annulus. The limiting viscosity value ($\approx$100 cP) of the ambient liquid was required for the fluid in order to form a cluster with a hemispherical cap, which resulted in a uniform settled structure. This is due to a regime change of cluster motion around the cluster Re_c of 60 and *Ga* close to unity (see Fig. 14(b)), which can be linked to the shift of flow behavior towards stokes flow or creeping flow.

Fig. 12, 13, 14

4. Conclusions

The experimental investigations patterns formed from settling of a particle cluster falling in a quiescent liquid at high Re_c conditions, and it’s spreading at the vessel bottom surface is presented. The significant finding is that a cluster having an initially hemispherical bottom cap settles in an axisymmetric shape upon hitting the vessel bottom surface irrespective of the vessel cross-section shape. The formation of the hemispherical cap and the subsequent axisymmetric shape formation is found to be a robust feature of the system as it nearly independent of the particle shape, size, and types. With the increasing initial mass of the particles, the cluster size was found to increase, the tendency for tail formation was reduced, and the settled shape became closer to a circular annulus and of higher dimensionless diameter. Despite using vessels of different sizes, the settled shape was close to a circular annulus. However, the square shape vessels resulted in enhanced spreading behavior. It is worth noting that a limiting viscosity of the ambient liquid of 100 cP was observed beyond which the cluster shape was deformed, and the particles spread in an asymmetric manner. The experimental analysis presented here should be taken as a step towards understanding the effect of high Reynolds number hydrodynamic interactions between settling particle clusters on its spreading behaviour upon hitting the bottom surface.

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Conflicts of interest statement

The authors declare no competing financial interest.

Appendix A. Supplementary data

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Figures:

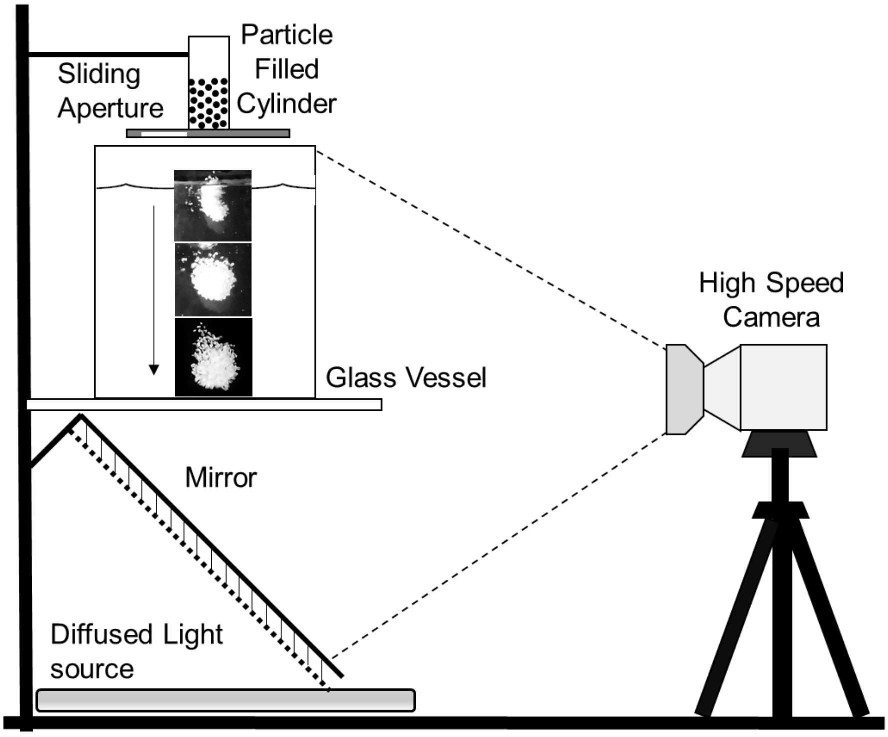


Fig. 1. Schematic illustration of the experimental setup used for settling of solid clusters. Particle settling and spreading was acquired as video in a mirror.

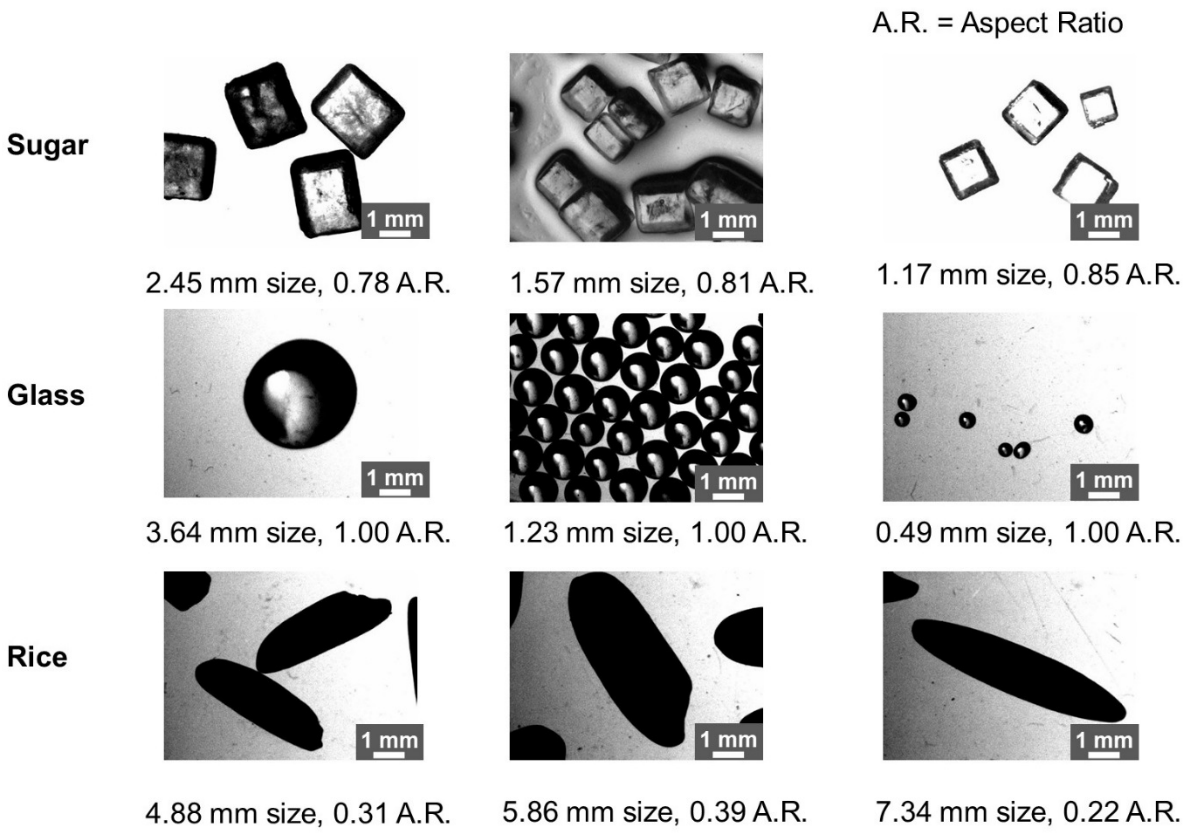


Fig. 2. Particle types and particle sizes used for settling experiments.

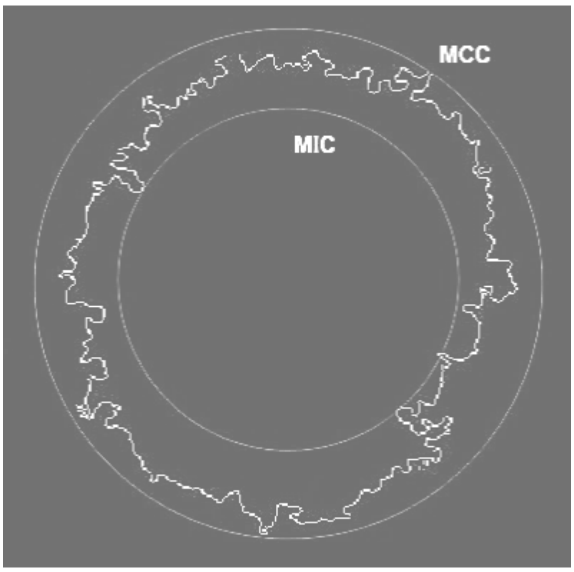


Fig. 3. Schematic representation of minimum circumscribed circle (MCC) and maximum inscribed circle (MIC).

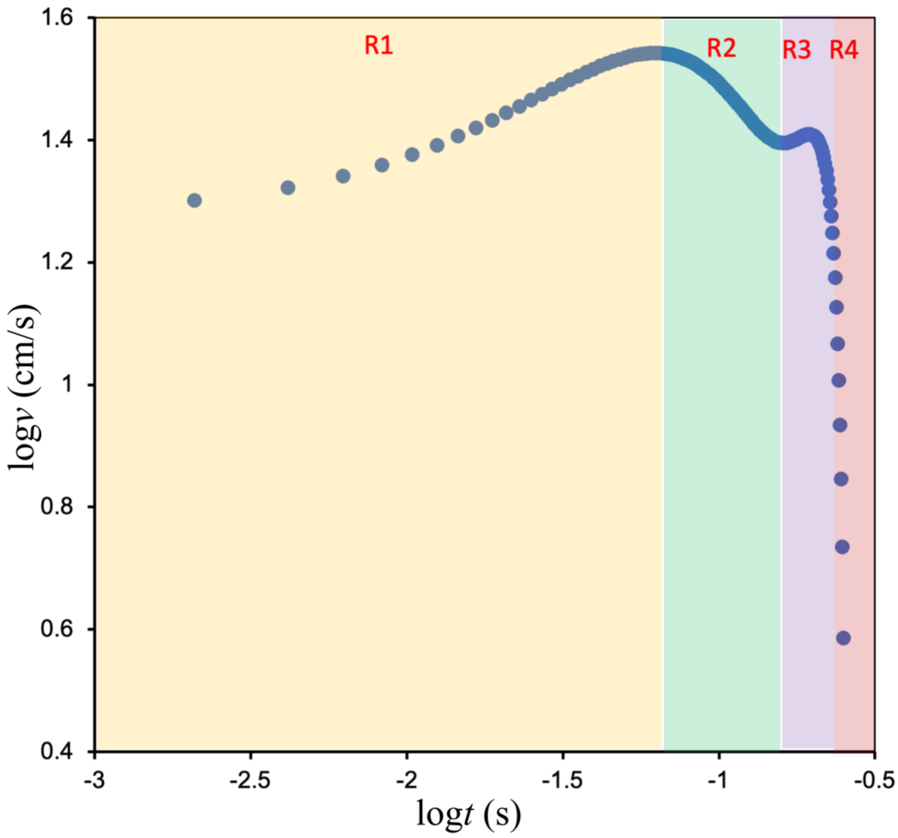


Fig. 4. Variation in cluster velocity as it travels through the liquid. Slope of different regimes (R1-0.142857, R2 - 0.52632, R3 - 0.14, R4 - 20). 15 cm × 15 cm square cross-section glass vessel with 10 cm liquid level was used for experiments with 10 g cubical sugar particles of 2.4 mm average size.

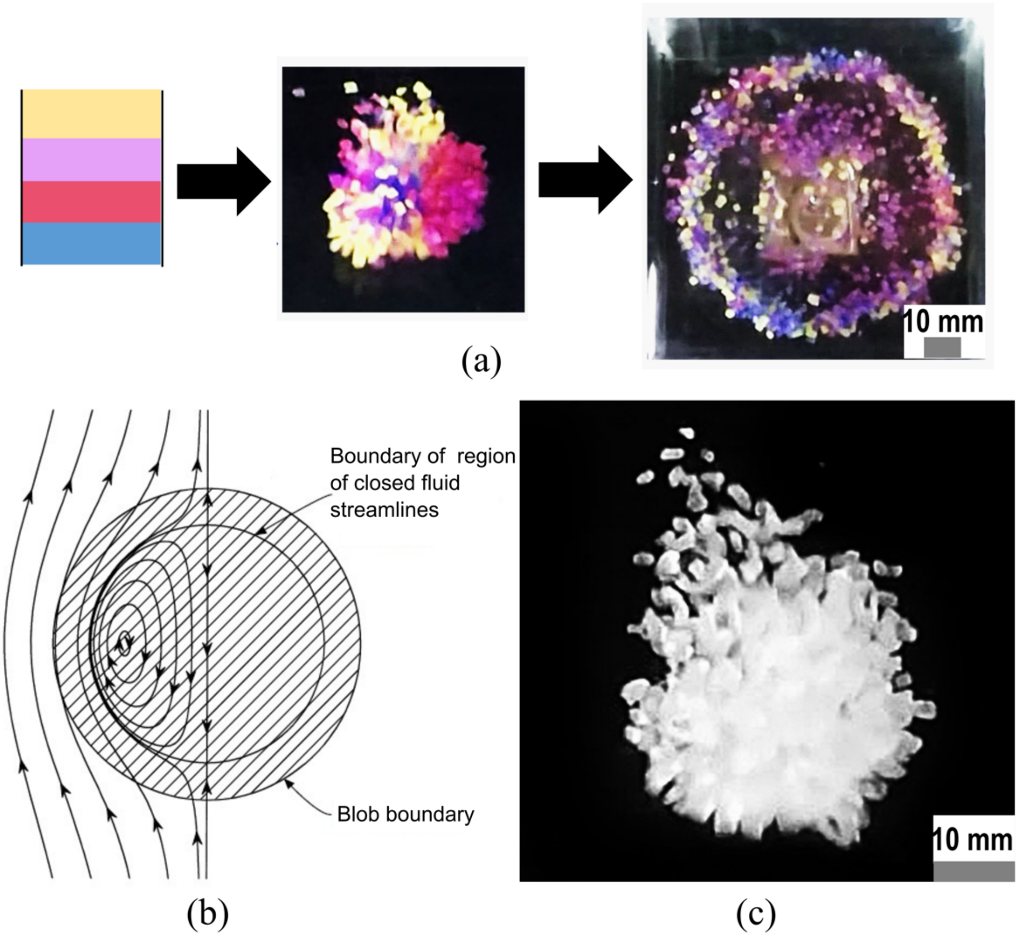


Fig. 5. (a) Typical stack of different colored particles before release, cluster formation inside the liquid and final settled shape for stacked colored particles; (b) Schematic of open and closed-loop streamlines of toroidal circulation - reason behind Particle leakage and tail formation (Reprinted from Nitsche et al. (Nitsche & Batchelor, 1997) with permission from Cambridge University Press), (c) Settling cluster with tail formation at the rear end.

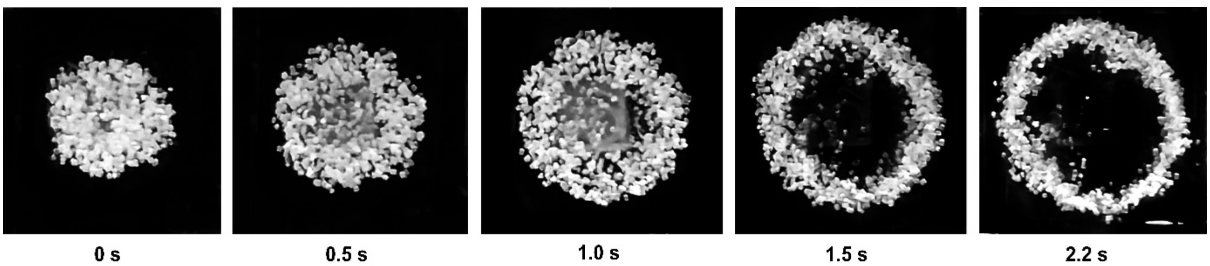
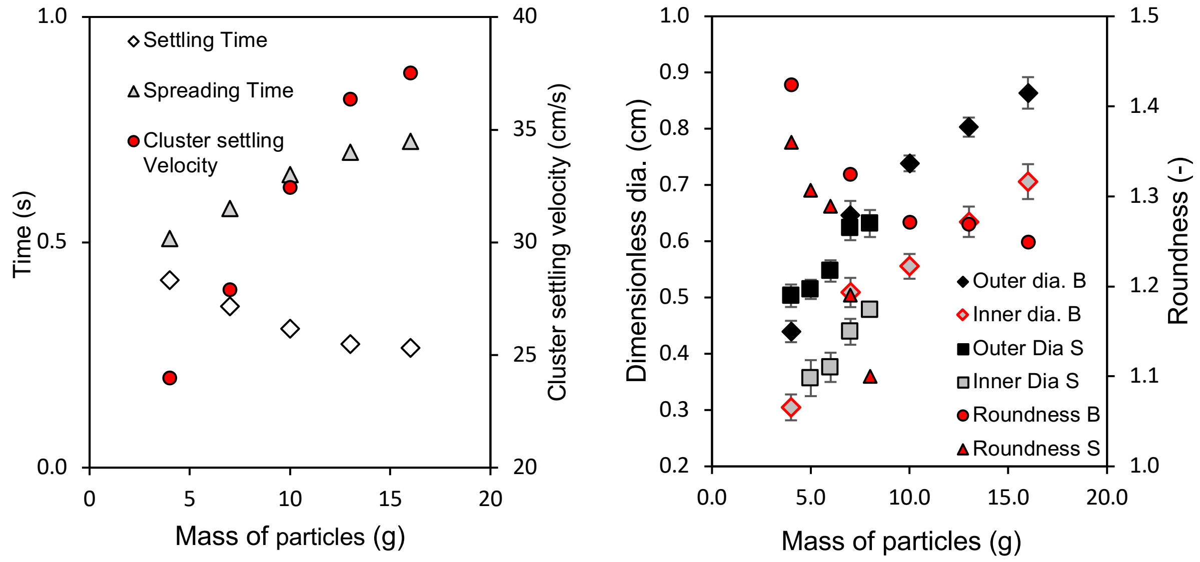


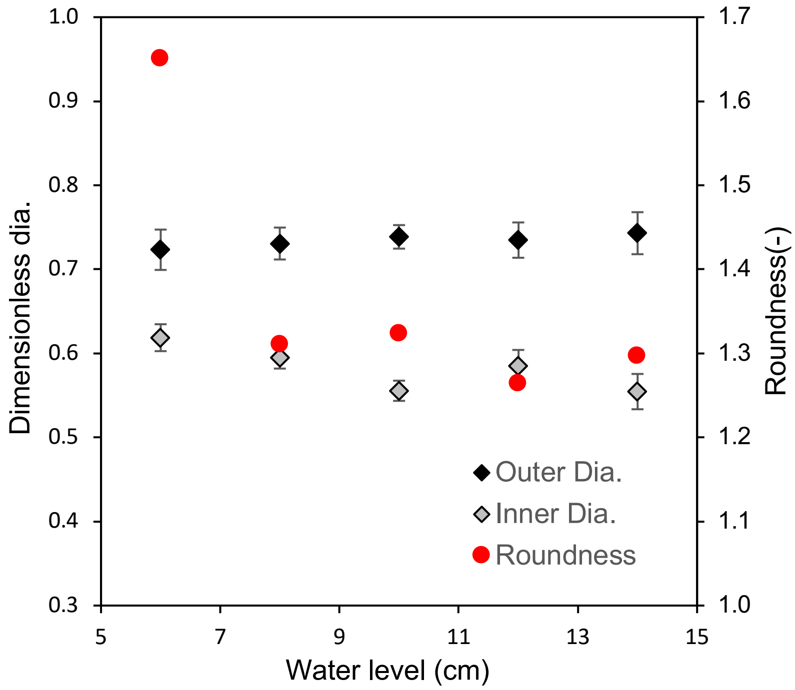
Fig. 6. Sequence of cluster spreading after collision with the vessel bottom surface. The zero time scale starts from the instant when the cluster touches the bottom of the vessel. A cluster comprising of 10 g of

2.45 mm sized sugar particles was allowed to settle in a 15 cm ×15 cm square cross-section vessel filled with water (14 cm height).

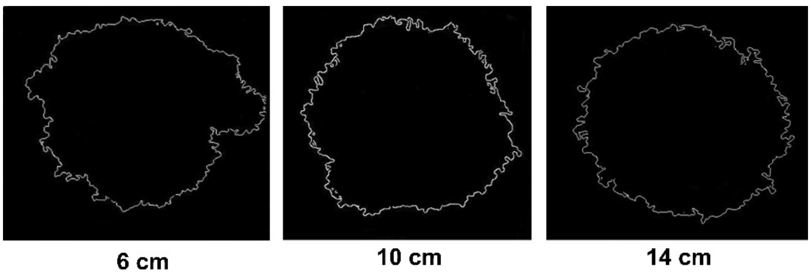


(a) (b)

Fig. 7. Effect of the total initial mass of particles on (a) Settling and spread time in big square vessel; (b) Dimensionless diameter of the settled shape in small and big square shape vessels (B- big square vessel, 15 cm ×15 cm cross-section; S- Small square vessel, 10 cm ×10 cm cross-section)



(a)



(b)

Fig. 8. (a) Effect of the vessel fluid level on the settled pattern in the big square-shaped vessel (15 cm ×15 cm cross section), (b) Outer diameter of the settled shape for different vessel fluid levels (6–14 cm).

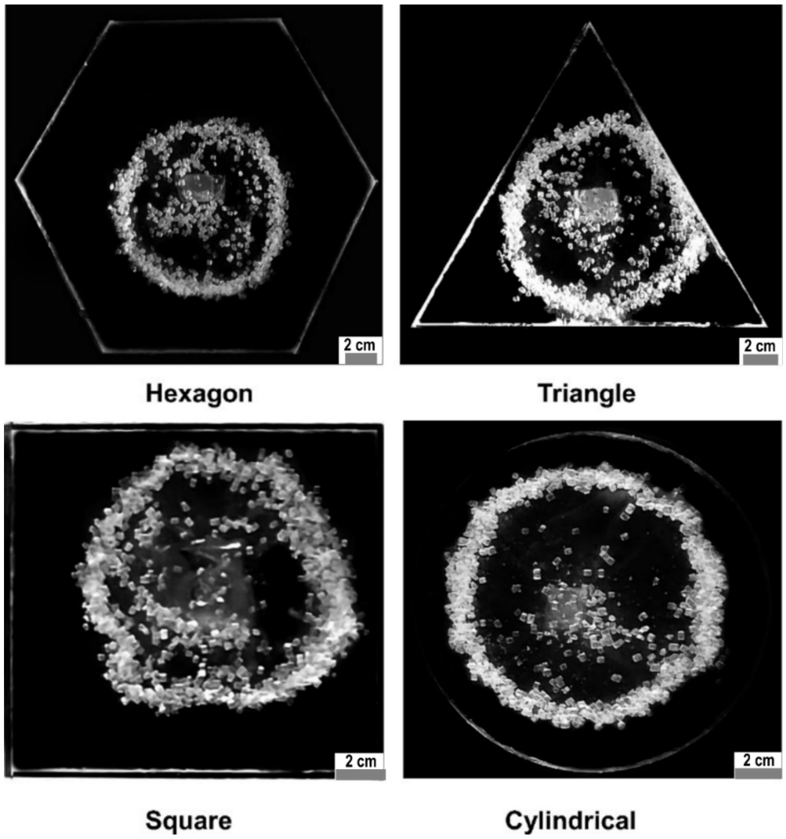


Fig. 9. Effect of vessel shape on the settled pattern.

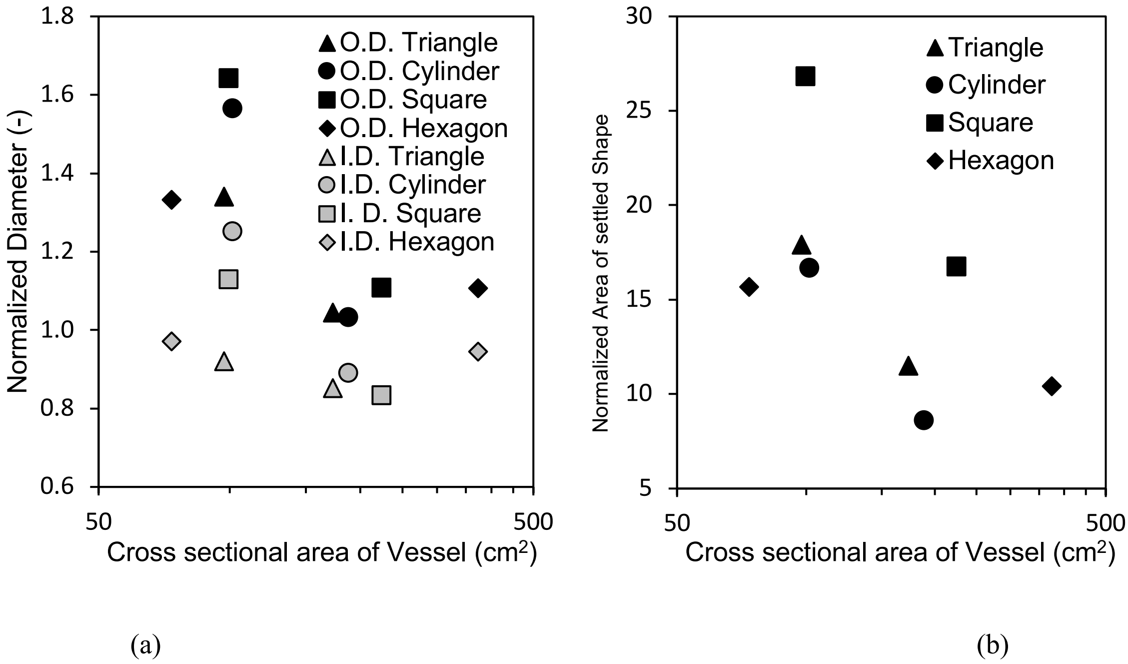


Fig. 10. Effect of vessel shape and size on the dimensionless diameters of the settled pattern.

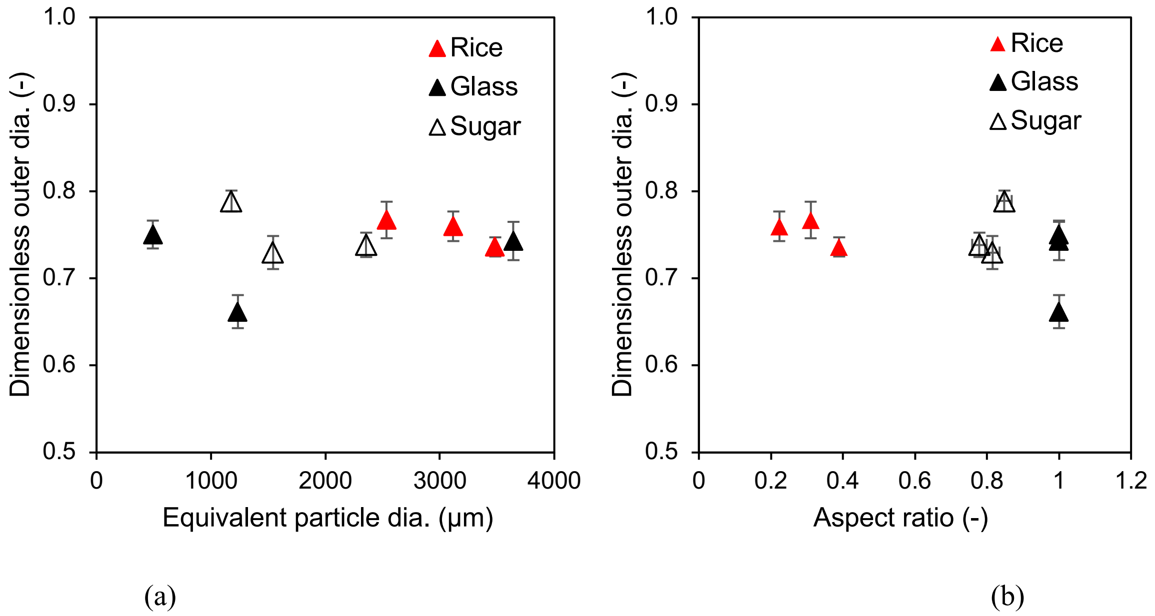


Fig. 11. Effect of (a) equivalent particle diameter and (b) aspect ratio of different particle types on the dimensionless outer diameter of the settled pattern. 15 cm ×15 cm square cross-section glass vessel with 10 cm liquid level was used for experiments.

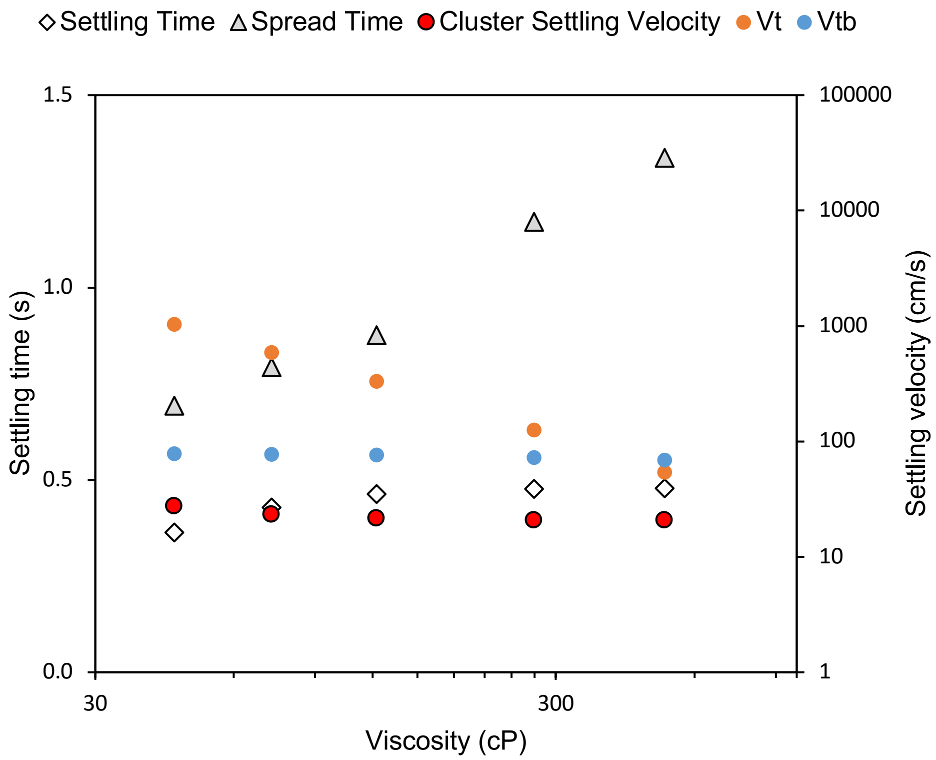
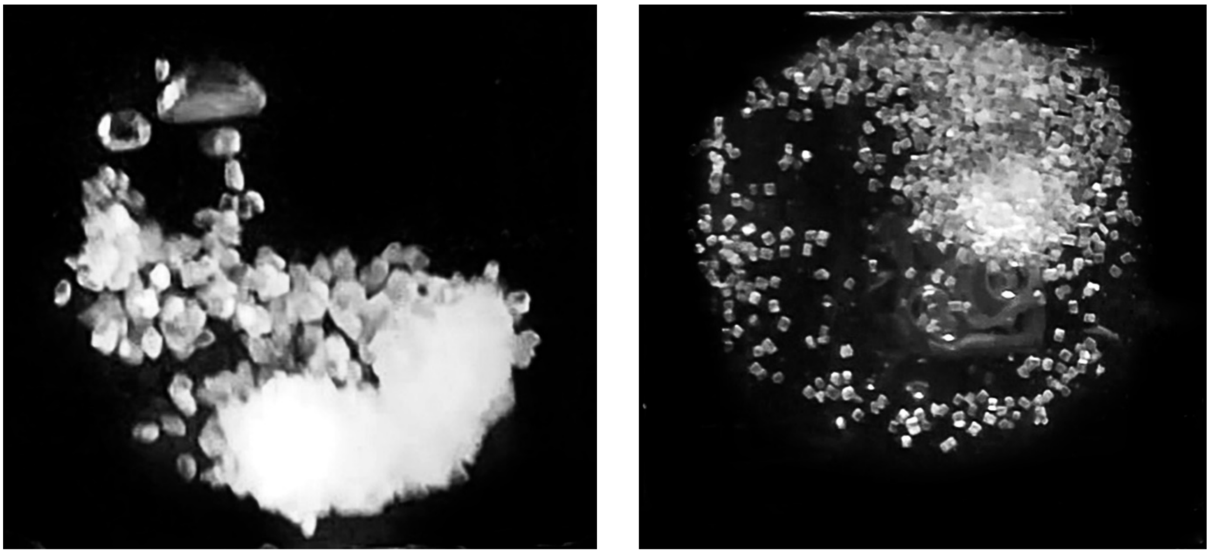
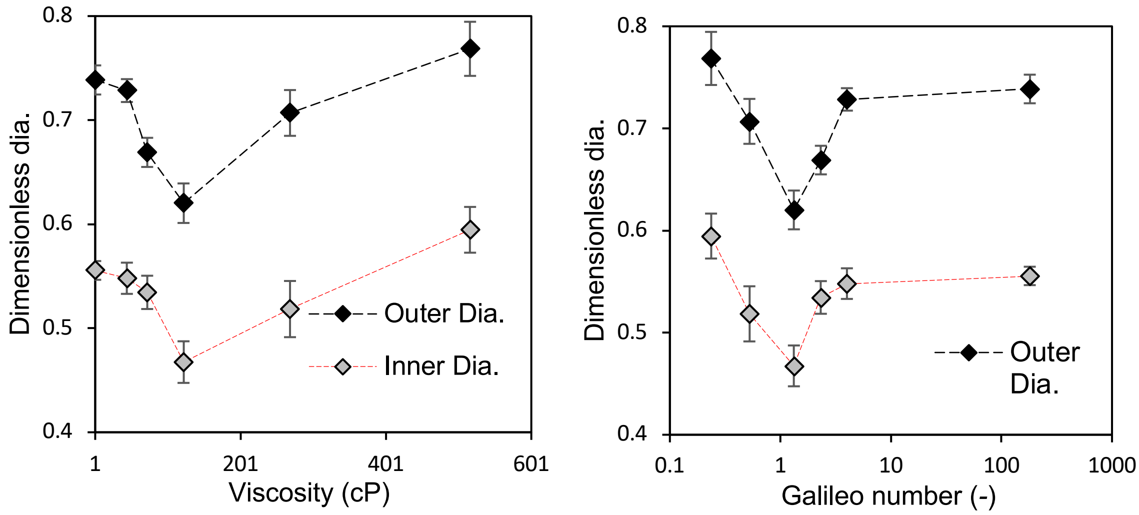


Fig. 12. Effect of fluid viscosity on the settling time, spread time, cluster settling velocity, Stokes flow terminal settling velocity (V_t) of the cluster and terminal settling velocity of the cluster in the presence of buoyancy force (V_{tb}) (terminal settling velocity equations in the Supplementary Data). 15 cm ×15 cm square cross-section glass vessel with 10 cm liquid level was used for experiments.



(a) (b)

Fig. 13. Settling and spreading of sugar particle cluster in viscous liquid: (a) Front view during cluster settling showing breakage of cluster; (b) Bottom view of the asymmetric spread of the particles on vessel bottom. 15 cm ×15 cm square cross-section glass vessel with 10 cm liquid level was used for experiments with 10 g cubical sugar particles of 2.4 mm average size.



(a) (b)

Fig. 14. (a) Effect of fluid viscosity; and (b) Effect of Galileo Number on the dimensionless diameter of the settled pattern.

Supplementary data

S1. Details of vessel dimensions

When not studying the effect of different parameters, typical experiments were carried out in a 15 cm × 15 cm × 20 cm square cross-section glass vessel with 10 cm liquid level and using 10 g cubical sugar particles of 2.4 mm average size. The different vessels of following dimensions are used to study the effect of vessel shape and size in section 3.2.3.

Vessel Size and Shape	**Vessel side length/ vessel diameter (cm)**	**Vessel height (cm)**	**Area of the vessel bottom surface (cm^2)**
Big - Triangle	20	15	173
Small - Triangle	15	15	97
Big - Cylindrical	15.5	15	189
Small- Cylindrical	12	15	102
Big - Square	15	15	225
Small - Square	10	12	100
Big - Hexagonal	12	15	374
Small- Hexagonal	6	12	74

S2. Effect of mass of particle swarm on cluster tail formation

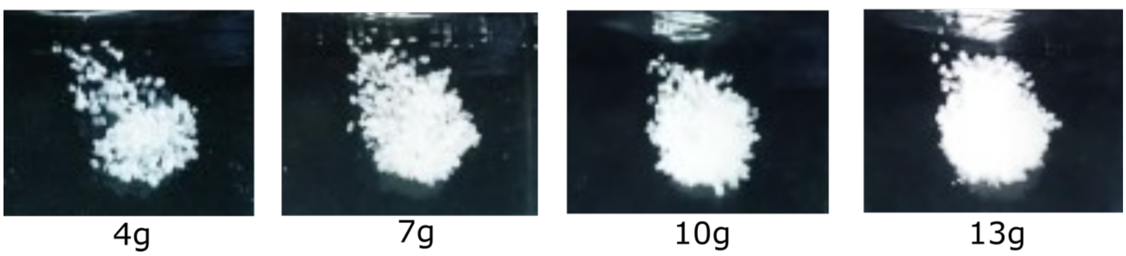


Fig. S1. Effect of the total initial mass of particles on extent of cluster tail formation

S3. Terminal settling velocity of clusters

When the buoyancy effects are taken into account, an object falling through a fluid under its own weight reaches a terminal settling velocity, if the net force acting on the object becomes zero. When the terminal settling velocity is reached, the weight of the object is exactly balanced by the upward buoyancy force and drag force, and can be estimated by the following equation

Terminal settling velocity in the presence of buoyancy force $V_{tb} = \sqrt{\dfrac{4gd_c\,(\rho_s - \rho_l)}{3C_d\rho_l}}$

For very slow motion of the fluid, the inertia forces of the fluid are negligible in comparison to other forces (Reynolds Number <<1) . Such flows are called Stokes flows and the settling velocity can be estimated by the following equation

Terminal settling velocity in Stokes Flow $V_t = {gd_c}^2(\rho_s - \rho_l)/18\mu$

Where d_c is the diameter of the cluster, is the gravitational acceleration, ρ_s is the density of the cluster, ρ_l is the density of the fluid, and C_d is the drag coefficient.