Hierarchical Structures in Granular Matter

J González-Gutiérrez¹, J L Carrillo-Estrada¹ and J C Ruiz-Suárez²

¹ Instituto de Física, Benemérita Universidad Autónoma de Puebla, Apartado Postal J-48, Puebla 72570, México.
² CINVESTAV-Monterrey, Parque de Investigación e Innovación Tecnológica km 9.5 de la Autopista Nueva al Aeropuerto, CP 66600 Apodaca NL, México
E-mail: carrillo@ifuap.buap.mx

Abstract. Granular matter, under the proper conditions of vibration, exhibits a behavior that closely resembles that of gases, liquids or solids. In a vibrated mix of glass particles and magnetic steel particles, it is also possible to observe aggregation phenomena, as well as, processes of reconstruction of the generated clusters. In this work we discuss the effects of the so called granular temperature on the evolution of the agglomerates generated by the magnetic interactions. On the basis of a fractal analysis and the measured mass distribution, we analyze experimental results on the static structural aspects of the aggregates originated by two methods we call: granular diffusion limited aggregation (GDLA) and growth limited by concentration (GLC).

1. Introduction

Granular fluids composed by a mix of glass particles and magnetic steel particles inside a vibrated container, exhibit structural transformations and changes in their physical properties that, at certain extent, can be modulated by the external vibration amplitude, the concentration of magnetic particles and the magnitude of their dipole [1, 2]. In the presence of a magnetic field the structural changes generated by the aggregation of the magnetic particles can be roughly described as the formation of rings, chains and more complex clusters. There has been discussed in the literature that the structure and some of the growth characteristics of the clusters generated by magnetic interactions in granular fluids are consistent with some recently reported numerical simulations based on the dipolar hard sphere model [3, 4]. However, these extensive numerical simulations and some theoretical works, have not been able to conclusively determine the existence of the experimentally observed phase transitions.

A vibrated system composed by a mix of glass particles and paramagnetic iron particles, with no applied magnetic field, does not exhibits any aggregation phenomenon. When an external magnetic field is applied, a magnetization is induced in the steel paramagnetic particles, then, they may interact and for suitable amplitude and frequency of vibration some aggregation phenomena could occur. It has been shown that in magnetorheological dispersions of non Brownian magnetic particles in the presence of an external field, the predominant interaction driving the aggregation processes is the many body dipolar magnetic force among the particles. In those dispersions there occurs some well defined sequential stages in the aggregation processes that generate multifractal clusters. In these stages the viscosity of the liquid matrix has also a sequentially increasing influence [5, 6]. This paper analyzes experimentally the effects of the granular temperature on the characteristics of the structures generated by aggregation.
of magnetic particles due to the application of an external static magnetic field. This is done by taking different values for the ratio between the vibration acceleration and the gravity acceleration. This ratio, usually denominated Γ parameter is directly related to the so called granular temperature. Here we present a comprehensive discussion of the structural characteristics of the aggregates obtained by following two experimental methods. The processes of aggregation are recorded by using a digital camera. Films and photographs obtained during the formation of the clusters allow us to perform a detailed analysis of the structures in terms of some measures of the complexity, in particular in terms of the mass fractal dimension and the mass spatial distribution. In section II, we briefly describe the experimental procedures and setups. In Sec. III the experimental results of the evolution of the structure in the granular fluid are presented, and the discussion is made by comparing the clusters grown by the GDLA and GLC methods. Finally in Sec. IV, some remarks and comments are made.

2. Experimental Procedures and Setups
Our system is composed by a mix of steel and glass spherical particles inside a vibrated container. The apparatus consists of a three-dimensional cell (15x15x3 cm). The measured acceleration of the cell, relative to the gravity acceleration is quantified by the parameter Γ = (Aω²)/g, where A and ω are respectively the amplitude and frequency of the vibration, and g is the acceleration due to gravity. The parameter Γ was varied between 1.67 and 2.56. The system is horizontally leveled to ensure that the plate is uniformly accelerated. The particles used in our experiments are paramagnetic steel spheres with a diameter of d=1.61 mm and mass m=0.017 gr. We also introduce in the container 2027 glass particles with a diameter of d=2 mm and mass m=0.0074 gr. This is done to incorporate additional randomness into the system by increasing the number of collisions. This also ensures that as the steel particles aggregate (condense), the "granular temperature" of the system does not go to zero. The glass particles play the role of a thermal bath because a large amount of glass particles collide with the paramagnetic particles, allowing reach small fluctuations in the value of the mean kinetic-energy per particle. This last quantity is associated with the so called granular temperature.

Thus, the granular temperature is defined as \( T_g = \frac{1}{2} m \langle v^2 \rangle \). Where \( m \) is the mass of a granular particle and \( \langle v^2 \rangle \) is the mean square speed of the particles. Blair and Kudrolli [5] found that for small values of parameter Γ, \( T_g \) depends linearly with Γ.

The concentration, \( \phi \), is defined as the ratio between the number of paramagnetic particles \( n_1 \) and the total number of the particles \( n_2 \).

A magnetic interaction among the steel particles was induced by placing a spherical magnet centered under the base of the cell. The magnetic field strength of this magnet is \( \vec{M} = 12 \) kG. In this configuration the magnetic field lines has approximately a radial configuration. Image data are acquired by means of a digital camera Pixelink 2000. The experimental setup is illustrated in Figure 1.

In this work we study, varying the Γ parameter, the structural characteristics of the aggregates grown by means of two different methods: granular diffusion limited aggregation (GDLA) and growth limited by concentration (GLC). We name GDLA the process whereby when a paramagnetic particle undergoing a random walk becomes adhered to another particle or to a cluster by the action of a potential, another particle is immediately introduced into the cell. This process was carried out for 325 paramagnetic particles. One by one, we introduce paramagnetic particles within the system through an orifice located at a height \( h = 0.5 \) cm on a side wall of the cell. On the other hand, GLC is that process in which a set of 325 paramagnetic particles are introduced simultaneously into the vibrating cell. Of course, the most remarkable difference with the first growth method appears at the early stages of the aggregation process, at those stages many particles become aggregated due to the dipolar interaction induced by the external field. We explore the patterns of aggregation generated for GDLA and GLC with a robust
coverage of values of the parameters of growth, keeping the number $n_1 = 325$ of paramagnetic particles. The observed aggregation processes in the GLC case are as follows. When the spherical magnet is placed under the cell center, the particles quickly form chain configurations around the magnet because locally this head-to-tail alignment is energetically favorable. However, many particles during the growth become aggregated laterally to the primary chains. Depending on the conditions of granular temperature and concentration, two additional phenomena can be observed in this processes: When the length of the primary chain is larger than 4 particles, the chain bends into a more compact structure. This effect can be seen as a widening in the primary chain. On the other hand, when the length of the primary chain is $< 4$ particles, it is possible to observe a side branching. Therefore new chains grow laterally. These observations agree with those discussed by Daniel Blair and A. Kudrolli [1]. They report that the formation of rings for aggregates larger than 4 particles are energetically more favorable than the formation of chains of equal length.

The comparison between Figure 2(a) showing a fibrous structure generated with GDLA and Figure 2(b) showing the structure generated GLC with the same value of the other physical parameters, allows to distinguish some significative differences: for instance, the structure generated by GDLA shows branches more elongated than GLC. One may infer that GLC is more affected by lateral aggregation. It can be physically explained as follows: when magnet is applied under the cell, it can be interpreted as if the kinetic energy of a magnetic particle were suffered an abrupt decrement, under these conditions the paramagnetic particle becomes unable to explore its whole phase space and quickly it felt stucked to the chains sides. Thus, this is why one may expect that the process of lateral aggregation should be affected by the particle concentration. On the other hand, the collisions among the glass and the steel particles introduce stochasticity into the system which could detach particles from the cluster introducing a limit in the mean size of the aggregate. Only the most bonded configurations remain unchanged. Therefore, when in the system there are not many collisions, some other different configurations could appear. Is in this context that we wish to explore the effects of the granular temperature on the characteristics of the structure.

By varying the $\Gamma$ value we change the granular temperature. We found that in certain interval, relatively high values of the granular temperature induce a higher degree of compaction in the clusters. Figure 2(c) and Figure 2(d) shows this effect on a fibrous structure generated by

![Figure 1. Sketch of the experimental set-up.](image.png)
Figure 2. Figure 2(a) shows the fibrous structures generated with GDLA, \( n_1 = 325 \) steel particles, volume fraction \( \phi = 0.138 \) and \( \Gamma = 1.67 \). Figure 2(b) shows the fibrous structures generated by GLC under the same physical conditions we used to generate the clusters of Figure 2(a). Figure 2(c) shows the static structure generated by GDLA with \( \Gamma = 2.12 \). Figure 2(d) shows the structure generated by GDLA with \( \Gamma = 2.56 \).

GDLA with \( \Gamma = 2.12 \). At a much higher granular temperature, \( \Gamma = 2.56 \) (see Figure 2(d)), the cluster never reaches a static structure, namely, as soon as the magnetic particles aggregate they are rapidly detached by effects of collisions with the glass particles.

3. Static Structure
To study the pattern formation, we firstly discuss the fractal aspects of the fibrous structure generated by GDLA and GLC. In this analysis, the mass fractal dimension and the radial distribution function of the cluster structure were measured on digital pictures. The method used in this paper to measure the mass fractal dimension is a variation of the standard box counting method, adapted to explore radial and angular correlations in systems affected by potentials with radial or axial symmetries [5, 6]. In this method, the mass fractal dimension is defined by the power law: \( M \propto r^D \) where \( M \) is the average mass contained in a circle of gyration radius \( r \). This mass is measured in an ensemble to properly extract the information of the correlations. If a scaling relation of this form fits well the experimental data, then the exponent \( D \), with a value in the interval \( 1 < D < 2 \), is called the mass fractal dimension. On
the other hand, the two-dimensional radial distribution of mass, for discrete values of the radius is defined by:

\[ g(r_i) = \frac{M(r_i + \Delta) - M(r_i)}{\pi[(r_i + \Delta)^2 - r_i^2]} \]  

(1)

Here \( \Delta \) is a small increment in the radius, \( r_i \), or equivalently, the separation between two contiguous circles of the set used to determine \( D \) and \( g(r_i) \). In this procedure the magnitude of \( \Delta \) can be varied to evaluate the correlations with more spatial detail. This quantities as well as some other quantitative measures of the complexity have been used to investigate the fractal characteristics of the complex structures generated in rheological suspensions by the application of external fields. A detailed discussion on the application of this method can be seen in reference [6].

![Figure 3. The mass fractal dimension \( D \) as function of \( \Gamma \).](image)

We measure these quantities in photographs obtained by means a digital camera Pixelink 2000. Operatively, the procedure is as follows: digital photographs of the structure are obtained from the whole structure of the granular media. First we make a high-contrast treatment of the images to define a single plane in the structure. Afterwards, we numerically draw a set of equally separated concentric circles, starting with a circle with a diameter of the order of the size of the steel particles, up to circles as large as the observation field allows. The separation between contiguous circles, \( \Delta \), also can be varied. Experimentally we have found that if \( \Delta \) is taken to be of the order or less than the diameter of the particles, and if the ensemble has at least ten clusters, quantitatively the results are stable. Obviously, in a given circle the number of pixels, \( N \), spanned by the particles is proportional to the mass contained in that circle.

To observe the characteristics of the structure at larger scales, we show our measurements extended up to 20 circles. This procedure was repeated, selecting randomly different sites of the photograph as centers of the set of circles. Then the average of the number of pixels for corresponding circles are taken in order to capture the correlations existing in the structures formed by the particles. The mass fractal dimension is determined by fitting a straight line to
Figure 4. The behavior of the mass distribution of the aggregates is shown for $\Gamma = 1.67, 1.82, 2.12$ and $2.56$. The squares correspond to GDLA and the circles correspond to GLC.

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the graph of $\log_{10} N_i$ versus $\log_{10} r_i$, where $N_i$ is the average number of pixels contained in the circle of radius $r_i$. The fitting was done by means of a standard less squares procedure, taking the dispersion of the number of pixels as the tolerance parameter.

Our results indicate that the agglomerates formed by both procedures GDLA and GLC are mostly monofractal structures. The numerical value of the mass fractal dimension depends strongly and non linearly on the $\Gamma$ parameter value. Figure 3 shows the behavior of mass fractal dimension, $D$, for several values of the granular temperature parameter $\Gamma = 1.67, 1.82, 2.12, 2.56$. The red curve corresponds to GDLA and the black one corresponds to GLC. Both curves decrease as the temperature increases, however, granular temperature affects less the mass fractal dimension of the clusters grown by GDLA than those grown by GLC. Thus, for both procedures of aggregation, higher values of temperature propitiate the formation of less ordered clusters. Nevertheless, the relatively slower formation of clusters as it occurs in the GDLA method in comparison to GLC, propitiates the formation of more stable, and for high granular temperatures, more ordered structures.

Finally, we quantify by means of the expression (2), the spatial mass distribution of the aggregates generated in the system at different values of the granular temperature parameter $\Gamma$. Figure 4 shows the behavior of the mass distribution, $g(r)$, for the final configurations of the clusters attained for different values of $\Gamma$. As it was expected, the behavior of the distribution indicates that the compactness of the aggregates attains shorter values as the granular temperature parameter increases.

4. Conclusions
We have experimentally studied aggregation phenomena in a vibrated granular system. The procedure here proposed allows us to analyze the effect of the magnetic interactions on the kinetics of the aggregation of magnetic particles immersed in a thermal bath formed by glass particles. The structural properties of the cluster generated by two different methods were studied. We have shown that this granular system exhibits a rich variety of complex patterns of
aggregation. Clear differences in the degree of order of the aggregates generated by GDLA and GLC procedures were found. Clusters generated by the GDLA procedure are, comparatively, more stable and at high granular temperatures more ordered and compact than those generated by GLC.

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