Three-dimensional visualization of contact networks in granular material
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In this chapter, the contact network of a granular system was reconstructed using solvatochromic dyes attached to the surface of PMMA particles. The position of the particles and the contact points between them were reconstructed with the help of specially designed software. We reconstructed the contact network of four granular systems with varying density. Valuable information on contact number ($Z$), angular distribution of the contacts ($\theta$), and packing structure was readily obtained with the software.

The average contact number $<Z>$ became higher when the sample is more dense. Notably, the densest systems exhibits regions of regular packing (HCP), which was evidenced by angular distribution of the contact points, showing peaks at $\theta = 90^\circ$ and $35^\circ$. Visual inspection of the complete contact network showed that the regular packing was indeed present at the bottom of the sample.
8.1. Introduction

Referring back to chapter 1, the ultimate goal of the present project is to visualize three-dimensional force networks in granular materials. Recapitulating, granular materials such as sand may behave as a solid, as a liquid or as a gas, depending on the conditions. Jamming occurs when granular material undergoes a transition between flow (liquid-like) and rigidity (solid-like).\(^1,^2\) Static friction forces between two touching granules force the system into a jammed state, and only when an external stress is applied which exceeds these friction forces, the granular material will start to flow.

Granular materials, such as sand, typically occur in the solid-like state, but relatively small external forces make them flow. Due to the complexity of the interactions of the particles, the properties of granular materials are still poorly understood, especially regarding this transition. In fact, one of the goals of modern physics is to find a unifying theory, which describes the properties of this transition for all soft matter.

This chapter describes a new experimental technique to visualize the interactions of particles and to reconstruct the three-dimensional contact network of a granular system. Section 8.2 describes the development of a software program that enables the reconstruction of the contact network in a granular system of PMMA particles labeled with a solvatochromic fluorescent dye, that were studied in chapter 6. Section 8.3 gives some examples of analyses that can be performed using this software, and the insight that is provided in the structure of the granular systems. Before going in detail on the development of the software, the next section gives some information on the packing structures that are possible for granular systems.

Systems of many particles, such as granular systems, may form several different packing structures. For perfectly monodisperse particles, several packing scenarios are known. Mostly, the particles in granular systems pack randomly, having a volume fraction \(\phi\) between 0.555 (random loose packing, RLP)\(^3\) and 0.634 (random close packing, RCP).\(^4\) The volume fraction is the volume of the particles divided by the total volume of the system. When the spheres are not perfectly monodisperse, the RCP limit of \(\phi = 0.634\) may be exceeded, as smaller particles may reside in the spaces between the larger particles.

Next to the disordered structure of random packing, ordered structures may occur in a packing of monodisperse particles. This is referred to as regular packing, in which the particles form periodic lattices. The three most common regular packings are depicted in figure 8.1. Face centered cubic (FCC) and hexagonal close packing (HCP) are the densest packings known for monodisperse systems, having a volume fraction of 0.740. Also shown is body centered cubic (BCC) packing, which is less dense with a volume fraction of 0.680 for monodisperse particles.\(^5\) Table 8.1 gives an overview of the most
important characteristics of the packings described here. For the regular packings, the number of contact points \( Z \) per particle and the angle(s) \( \theta \) at which the particles touch are given.

<table>
<thead>
<tr>
<th>Structure Type</th>
<th>( \phi )</th>
<th>( Z )</th>
<th>( \theta^{[a]} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Face Centered Cubic (FCC)</td>
<td>0.740</td>
<td>12</td>
<td>35.26°, 90°</td>
</tr>
<tr>
<td>Hexagonal Close Packing (HCP)</td>
<td>0.740</td>
<td>12</td>
<td>35.26°, 90°</td>
</tr>
<tr>
<td>Body Centered Cubic (BCC)</td>
<td>0.680</td>
<td>8</td>
<td>54.74°</td>
</tr>
<tr>
<td>Random Close Packing (RCP)</td>
<td>0.634</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Random Loose Packing (RLP)</td>
<td>0.536</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

[a] as defined in figure 8.7.

8.2. The software

We have developed software that reconstructs the contact network of a granular system of particles labeled at their surfaces with a (sub)monolayer of a fluorescent dye.\(^6\) The software uses the raw confocal microscopy images and transforms them into a digital contact network in the following five steps:

1. **Confocal Microscopy scanning**
   In the first step, fluorescence intensity images of the granular system are recorded in the \( x,y \)-plane, such as the one depicted in figure 8.2 (A). Images are recorded at increasing heights in the sample, with an interval of 0.2 \( \mu \text{m} \) between them (figure 8.2 (B)). These images are loaded in the software and analyzed in the subsequent steps.

2. **Determination of the \( x,y \)-coordinates of the particles**
   From the confocal images, the centers and contact radii of all particle cross sections are identified throughout the stack. The software recognizes the circular shaped cross-section of each particle and establishes an inner circle with radius \( r_{in} \) and gives the \( x,y \)-coordinates of the center. The centers of these inner circles are represented in figure 8.2 (C) with red dots.
3. Determination of the z-coordinate of the particles

Cross sections that belong to the same particle have the same x,y-coordinates in adjacent images. These cross sections are collected, allowing for small deviations in the x,y-coordinates, visualized as a cylinder in figure 8.2 (D). For each particle, the \( r_\mu \) is plotted against the z-coordinate, as is depicted in figure 8.2 (E) (blue points). The z-coordinate of the particle center is defined by the maximum of the circle fit (red line) through the data points in figure 8.2 (E) (green point).

4. Determination of the radius of the particles

For the determination of the radius of a particle, the x,y-image that is closest to the z-coordinate of the center is used. Then, radial fluorescence intensity profiles, analogous to figure 6.12, are evaluated. Starting from the center, these profiles are taken in 36 different directions (figure 8.2 (F)). All positions of the maxima of the fluorescence intensity are averaged to give the radius \( r_{\text{par}} \) of that particle (figure 8.2 (G)). Due to polydispersity in the PMMA particles, these radii are not all 5.0 \( \mu \)m. In fact, the average radius of the particles was determined to be 5.2 \( \mu \)m with a polydispersity of 2.5 %. While the vast majority of the particles were close to 5 \( \mu \)m in radius, there was a small fraction of substantially larger particles (ca. 10 \( \mu \)m radius).

5. Identification of the contact points

The software now knows the coordinates of the center of each particle, as well as its radius, which enables the localization of contact points between particles. In principle, two particles touch when the distance \( d \) between their centers equals the sum of their radii. However, the software allows the distance to be slightly greater to eliminate any error in the center coordinate and particle radius.

Finally, all potential contacts are checked via intensity profiles taken across the particles. The software establishes a fluorescence intensity cross section between the centers of the two touching particles, and decides from the occurrence of one or two maxima whether these two particles are actually in contact or not.

In case there are two maxima, the particles do not touch, but are close by. Only when one maximum is observed, the particles are truly in contact. An example of such a cross section is given in figure 8.2 (H), depicting particles close to each other that are not in contact.

Thus, using the software as described above, a 3-dimensional reconstruction of the granular system can be obtained with help of confocal microscopy images. In figure 8.3, a complete reconstruction of a granular system of PMMA particles is shown.
Figure 8.2. Schematic representation of the reconstruction of the contact network by the developed software; (A) fluorescence confocal image of the x,y-plane; (B) stacks of x,y-images with an interval of z = 0.2 μm; (C) identification of the centers of the particles; (D) determination of the x,y-coordinates; (E) determination of the z-coordinate; (F) determination of the radius of the particle; (G) radial intensity cross sections; (H) contact point identification.
Chapter 8

Figure 8.3. A reconstruction of the granular system of solvatochromic PMMA particles; the color varies with z. The size of this reconstruction is 266 × 266 × 90 µm.

8.3. Statistical analyses of granular systems

A rich amount of information on granular systems can be obtained using the software described in section 8.2. This section describes two scenarios, in which granular systems are analyzed with respect to their contact number $Z$, i.e. the number of contact points per particle, and the angular distribution of the contacts.$^7$

Thus, volumes of $106 \times 106 \times 90$ µm, each in four different granular systems, were imaged by confocal microscopy and analyzed. The four systems differ in the way they are prepared; their characteristics are given in table 8.2. Centrifuging the particles increases the volume fraction $\phi$, thus increasing the number of particles identified in the scanned volume by the software. Agitation of the sample with sound (“shaking”) loosens the packing and decreases $\phi$.

Notably, the $\phi$ of 0.681 for the centrifuged granular system is higher than the $\phi$ for a random closed packed system ($\phi$(RCP) = 0.634). This could be attributed to the polydispersity of the particles and the formation of local ordered structure, i.e. close packed regions such as HCP ($\phi$(HCP) = 0.740). Table 8.2. also gives the mean contact number $<Z>$.

<table>
<thead>
<tr>
<th>Sample preparation</th>
<th>Number of particles</th>
<th>$\phi$</th>
<th>$&lt;Z&gt;$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Sedimented</td>
<td>5725</td>
<td>0.598</td>
<td>5.94</td>
</tr>
<tr>
<td>2. Sedimented + shaken</td>
<td>5583</td>
<td>0.593</td>
<td>5.66</td>
</tr>
<tr>
<td>3. Centrifuged (500 rpm)</td>
<td>6211</td>
<td>0.681</td>
<td>7.17</td>
</tr>
<tr>
<td>4. Centrifuged (700 rpm) + shaken</td>
<td>5744</td>
<td>0.616</td>
<td>6.15</td>
</tr>
</tbody>
</table>

8.3.1. Contact number distribution

For each particle in the system, the contact number $Z$ can be determined from the contact reconstruction. The distribution of $Z$ for each of the four systems is given in
Visualization of the contact network in a granular system

Figure 8.4. The average contact number shifts to a higher value the denser the system (see also table 8.2). Apparently, the distribution of $Z$ around the mean remains closely the same for all four systems. Notably, some particles are observed to have 13 – 16 contacts, while the maximum number of contacts in a monodisperse granular system is 12. However, the present systems are not monodisperse, and the particles having $Z > 12$ have a significantly larger diameter.

![Figure 8.4](image.png)

**Figure 8.4.** Mean coordination number versus volume fraction $\phi$.

Formation of local order such as HCP may be induced by the bottom of the sample cell. This bottom is formed by a smooth glass cover slip, which may induce layering and an ordered packing. To investigate this phenomenon, the average $Z$ is plotted as a function of height in the sample (the $z$-coordinate of the particle) in figure 8.5 (A). The first layer of particles is found at $z = 5 \mu m$. Because of the presence of the boundary, these particles have fewer contact points with other particles. No contacts are recognized with the bottom cover slip. From the second layer upwards, the average $Z$ gradually decreases with increasing $z$ for all four samples. For the densest system 3, the second and third layers of particles ($z \approx 15 \mu m$ and $z \approx 25 \mu m$) have an average contact number of about 8, which decreases to about 6.5 in the seventh layer. Also for the other systems, $\langle Z \rangle$ starts to decrease from the fourth or fifth layer upwards. The decrease in $\langle Z \rangle$ is too big to be accounted for by a weight effect only. This weight effect is caused by the fact that the particles near the bottom of the system experience a greater pressure, caused by weight of the system above, than particles higher in the system. However, if only the weight effect would play a role in the decrease of $\langle Z \rangle$, the particles at $z > 200 \mu m$ would have no contact points any more. Thus, a decrease in structuring in the system causes the decrease in $\langle Z \rangle$, which is also apparent from figure 8.5 (B). This histogram shows the number of particles versus their $z$-coordinate for the densest sample 3. Clearly visible is the peak of the first layer at $z = 5 \mu m$. Subsequent layers can be identified at $z \approx 15 \mu m$, $z \approx 25 \mu m$.
µm and \( z \approx 35 \) µm, but the peaks clearly broaden with increasing \( z \). Thus, with increasing height the structure of the packing is gradually lost and the boundary effect of the bottom diminishes.

![Figure 8.5](image)

**Figure 8.5.** Structuring in the granular systems; (A) the mean coordination number \( \langle Z \rangle \) vs. the \( z \)-coordinate of the particles; (B) the distribution of particles \( z \)-coordinates for sample 3.

The software is able to reconstruct the entire contact network of a granular system, such as those depicted in figure 8.6. Here, the black dots represent centers of particles, blue lines represent contacts between two particles, with a red dot at the contact point where the particles touch. The contact network of figure 8.6 (A) belongs to sedimented granular system 1, which is randomly packed and does not show any structure. The dense packing of system 3 is represented in figure 8.6 (B), which clearly shows the ordered hexagonal close packing at the bottom few layers of the system. Further up in the system, the ordering gets lost and the packing becomes random, which is in accordance with the results given in figure 8.5.

![Figure 8.6](image)

**Figure 8.6.** Contact networks of granular systems, as reconstructed by the software; (A) contact network of system 1; (B) contact network of system 3.
8.3.2. Angular distribution of contacts

In the previous section, the numbers of contacts \( Z \) per particle were analyzed. This section focuses on the orientation of the contacts, i.e. the angular distribution of the contact points over the surface of a particle. The exact position of a contact point can be described with two angles: the angle \( \theta \) with the z-axis, and the angle \( \phi \) in the x,y-plane (figure 8.7).

![Spherical particle with the position of a contact point defined by \( \theta \) and \( \phi \).](image)

In figure 8.8, the distribution of the contact angle \( \theta \) for the four samples with different volume fractions is given. An angle \( \theta \) = 0° means that two particles are positioned exactly above each other, while \( \theta \) = 90° indicates particles that are in contact in a horizontal plane.

The first notable feature of the histograms in figure 8.8 is that the amount of contacts generally increases with \( \theta \) with a sizeable peak at \( \theta \approx 90^\circ \), thus there is a strong preference for the alignment of particles in horizontal planes. However, it should be taken into account that at \( \theta \approx 90^\circ \), there is room for six contacts per particle, whereas closer to the top of the particle there is only room for five or fewer contacts, and finally at \( \theta = 0^\circ \), there is room for only one contact.

The distribution of system 3 in figure 8.8 (C) is distinct from the other three distributions, showing an additional peak at \( \theta \approx 35^\circ \), which again implies the presence of HCP or FCC packing in the densest system. With increasing height in the sample, the peak at \( \theta \approx 90^\circ \) significantly decreases, as well as the peak at \( \theta \approx 35^\circ \) for system 3, indicating that the ordered packing mainly occurs at the bottom of the granular systems.

The distribution of \( \phi \) is uniform (histograms not shown), i.e. all values for \( \phi \) occur in more or less equal amounts.
Chapter 8

Figure 8.8. Histograms for the distribution of $\theta$; (A) system 1; (B) system 2; (C) system 3; (D) system 4.

8.4. Conclusions and outlook

This chapter describes the successful three-dimensional reconstruction of contact networks of granular particles with fluorescent dyes at their surface using specifically designed software. With such a digitally reconstructed contact network, a plethora of statistical analyses can be performed, which allow detailed investigation of the granular system. We exemplified the determination of packing structures, contact numbers and angle distributions including their dependency on the z-coordinate.

The question now arises whether this software could be used to visualize flow in granular material and the distribution of forces between the particles when an external force (e.g. shear) is applied to the system. Flow may be visualized by scanning the system before and after the application of a small amount of shear. Particles could be traced during shear, as long as their displacement is small enough so that their center after shear is still within the volume occupied by that same particle prior to shear.

Even more challenging is the visualization of a force network within a granular system, which was the ultimate goal of the present project. Force dependent fluorescence was not observed for the particles labeled with a solvatochromic dye (see chapter 6), but
was observed for the particles equipped with a rigidochromic fluorescent probe (see chapter 7). These particles showed a fluorescence intensity increase with increasing force applied to the system. In view of the results of this chapter, the developed software is expected to be able to reconstruct granular contact networks of rigidochromic particles without further modification. Further research may afford the implementation of an additional feature in the software, that is able to quantify the fluorescence intensity at contact points and translate that value to a local force experienced in that contact point. As such, the three dimensional visualization of force networks may be within reach.

8.5. Experimental section
Solvatochromic PMMA particles with a diameter of 10 μm were suspended in DMSO containing 0.1 g/mL NaI. The density of the particles was 1.18 g/cm³ and the density of the medium was 1.15 g/cm³, which allowed slow sedimentation under gravity. Systems 3 and 4 were centrifuged for 30 min at 500 rpm (19.6 g) and at 700 rpm (38.3 g) respectively. Systems 2 and 4 were “shaken” with sound of 400 Hz, using 10 bursts of sound (5 s) with intervals of 30 s. See section 3.2.5. for a detailed description of the sample cell.

See section 3.2.3.3. for a detailed description of the set-up of the laser and confocal microscope. Images in the x,y-plane were 106 × 106 μm and 450 images were recorded at intervals of 0.2 μm in the z-direction. Systems 1, 2 and 4 were recorded in twelve-fold, and system 3 in eleven-fold.

8.6. Acknowledgement
Marthe Schut and Hugo Doeleman are kindly acknowledged for performing major work underlying this chapter. Marthe developed the software described in section 8.2, and Hugo analyzed the granular systems as described in section 8.3.

8.7. References