Doctoral Thesis

Granular Matter: Electrostatics

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Publication Date:
2017

Permanent Link:
https://doi.org/10.3929/ethz-b-000196492

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Granular Matter: 
Electrostatics

A thesis submitted to attain the degree of
DOCTOR OF SCIENCES of ETH ZURICH
(Dr. sc. ETH Zurich)

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2017
Acknowledgment

I would like to thank Prof. Dr. Hans J. Herrmann for proposing me the topic of this thesis. His supervision and criticisms had inspired and motivated me throughout my Ph.D. Special thanks goes to Prof. Dr. Nuno A. M. Araújo for his supervision and for his role as a supportive mentor. I am grateful to Prof. Dr. Troy Shinbrot. I have learnt so much from the insights he brought into the projects and his truly professional work ethics. I would also like to thank him for accepting the request to co-examine this thesis. I appreciate David Parra with whom I had fruitful discussions about research and many other things. Special thanks goes to my colleagues in IfB for the wonderful company throughout my Ph.D. Last but not least, I want to thank my girlfriend and my family with whom I shared good times during my PhD.
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Abstract

Granular systems are known to self-charge upon collision. There has been some progress in understanding the fundamental mechanism of granular charging, but still a large part remains elusive. A recent model requires an external electric field to charge grains in collisional flows. In the first part of this dissertation, we study the charging of a fluidized granular bed in the presence of an external field. A simple model that has previously described charging of colliding polarized grains in the presence of a pre-existing field is extended by including more realistic physical features: e.g. grain rotations, heterogenous surface charge distribution, and electrostatic interactions. We find from our model that strong charge heterogeneities can occur inside an agitated bed and we predict that shielding due to these heterogeneities can dramatically alter the charging rate in such beds.

Charge induction in the presence of an external field and neutralization upon collision describe how grains can acquire strong charges, but it does not explain where the field comes from. Interestingly, however, experiments show that such field is not needed and charges on grains can grow on their own. In the second part of this work, we investigate the charging of an agitated granular bed in the absence of an external field using simulations, mathematical modeling, and experiments. We simulate charging with a discrete-element model including electrical multipoles and find that infinitesimally small initial charges can grow exponentially rapidly. We propose a mathematical model that defines conditions for exponential charging and provides insights into the mechanisms involved. We confirm the predicted exponential growth
in experiments using vibrated grains under microgravity. We also describe novel predicted spatiotemporal states that merit further study.

For the dynamics of grains, segregation plays an important role. Segregation of charged particles is especially relevant in many industries, for example in printing, pharmaceutical, plastics recycling, etc., where handling of charged particles is essential. However, comparatively little is known about the segregation of charged particles under controlled circumstances. In the final part of this dissertation, we study segregation of a binary mixture of mechanically identical but differently charged particles under shear. We simulate particle dynamics using a discrete-element model including Coulomb interactions. Here, we find that particles segregate according to their net charge. Particles that are charged twice as strongly as other particles with the same electrical sign migrate toward the insulating boundaries, with which we shear the system. Weakly charged particles, on the other hand, stay more in the center of the sheared bed. We propose a simple model based on electrostatic potential energy to understand this segregation. The model shows that the segregated system is indeed the most favorable configuration in terms of electrostatic potential energy. Our simulations further show that for a given packing fraction there is an optimal shear velocity at which the segregation maximally intensifies. We show that this maximum results from a competition between diffusional and Coulomb fluxes. For a larger shear velocity, diffusion suppresses segregation.
Related Publications

This dissertation contains content of the following published or submitted articles:


For each of these articles, the author contributed to: design, implementation, and execution of simulations; analysis and interpretation of data generated from simulations; conceptual and mathematical derivations; execution of numerical analyses; literature research; and writing of the manuscript.
Chapter 1

Introduction

1.1 Background

Despite being an insulator, granular materials have long been known to develop strong charges. For example, in volcano plumes and in sandstorms, agitation of ash or sand lead to contact charging of these particles, which in turn lead to spectacular electrical discharge [1, 2, 3, 4, 5, 6, 7, 8, 9]. Granular charging is also important in many industries. In xerography, toner particles are charged by rubbing against metal beads, and then attracted to an oppositely charged drum [10, 11]. In pharmaceutical formulation, where many pharmaceuticals are processed in the form of powder, contact charging occurs during a flow and leads to an undesirable blending [12, 13, 14]. In an extreme case, clouds of powders form during the transportation, in which particles mutually charge through contacts, and lead to a strong electric discharges resulting in devastating explosions known as dust explosions [15, 16, 17]. In spite of the paramount importance and prevalence of granular charging, its underlying causes remain controversial [18, 19].

To understand contact charging of granular media, past studies have largely focused on geometric or material differences between grains. It is often suggested that when two particles of the same material collide, charge transfer occurs such that smaller particles charge negatively and large particles positively [20, 21, 22], but the reason for this is still unclear and a few experiments even indicate charge transfer in the opposite direction [23, 24].
Other explanations claim that, in case of two dissimilar materials coming into contact, the direction of the charge transfer is determined by the chemical compositions of the two surfaces [25, 26, 27]. However, often the direction of charge transfer cannot be reasonably explained by the chemical compositions of the materials.

Surprisingly, however, grains that are identical in shape, size and chemical composition have also been found to spontaneously break symmetry and charge one another [28, 29, 30]. Experiments even show that this charging inexplicably grows with repeated contacts [28, 29].

Another important feature of granular media is segregation. Grains are known to segregate or de-mix [31], based on size [32, 33, 34, 35, 36, 37, 38], shape [39, 40, 41], density [42, 43], friction coefficient [44], or other material properties [45]. De-mixing is commonly seen both in natural processes: for example, in segregation of geological debris [46, 47, 48, 49, 50], and in industrial systems, for example, again in pharmaceutical mixing where uniformity is crucial to producing safe and effective medicines [51, 52, 53].

Segregation based on particle charge has also been widely applied in industry. For example, in plastics recycling, differential charging is used to separate different types of plastics [54]. In electrospraying and xerography, electrostatic forces adhere and localize charged grains [10, 11]. In a biological context, cell charging is used to separate dead from living and healthy from cancerous cells by applying non-uniform electric fields [55]. Additionally, it has been shown that mechanically identical grains that are charged with two different magnitudes of the same electrical sign spontaneously segregate while sliding down a chute [13] and another study has found that charged particles in a fluidized granular bed migrate toward the walls when all particles are charged equally [56]. Recently, it has been shown that a binary mixture of mechanically dissimilar particles otherwise known to segregate can be made to mix by charging particles with opposite electrical signs [24].
1.2 Motivation

1.2.1 Granular Charging in the Presence of an External Field

Pähtz et al. proposed a simple model in which repeated collisions between polarized particles transfer net charge along the direction of an external field [30]. This simple model captured the essential of the charging dynamics of fluidized beds in the presence of an external field, but neglected some important physical aspects. Specifically, the model did not consider the rotational motion of particles, hence, dipole moments were always aligned with the external field. The model also neglected all electrostatic interactions between particles under the assumption that the external field was overwhelmingly stronger than that due to nearby charges. However, it has been shown that the interaction between charged and polarized particles play an important role in the dynamics especially in close distances [57], and the amount of charge being transferred during a collision increases with the field strength [58]. In Chapter 2, we study the charging dynamics of a fluidized bed in the presence of an external field using a model which includes Coulomb interactions, polarization field effects, and heterogenous surface charge distribution. The goal is to understand how particle interactions among other added features affect the collective dynamics and charging behavior of the granular system from which we may gain insights into charging and discharging dynamics in natural and industrial granular flows.

1.2.2 Granular Charging in the Absence of an External Field

In Chapter 2, we show that an external field can indeed feed electrification and increase the bed charge. However, granular charging is observed for identical materials even in the absence of an external field and this charging surprisingly increases with repeated collisions [28, 29]. Previous work has shown that collisions between polarized particles can charge a system
even in the absence of a pre-existing field [59]. In that work, a static one-dimensional model was used to show that infinitesimal charges on grains can induce charges on their neighbors, bootstrapping one another to grow rapidly. In Chapter 3, we construct a dynamical three-dimensional model, in which thousands of particles interact, and study the charging of an agitated granular bed in the absence of an external field. Our goal is to understand how electrostatic interactions between particles permit initially nearly neutral system to build up significant charges. The answer may contribute to improved understanding of the long-standing question: how multi-million volt potentials observed in volcano plumes and sandstorms are generated from initially nearly neutral dusts and sands.

1.2.3 Segregation of Charged Particles under Shear

Segregation of charged particles is important in many industries: for example in printing, pharmaceutical, plastics recycling, etc., where charged grains are mixed or separated using electrostatic forces. Despite the prevalence and practical importance of particle segregation, relatively little is known about segregation of charged grains under well controlled conditions. In Chapter 4, we study segregation of mechanically identical but differently charged particles in a simple shear flow. Our goal is to gain understanding into the fundamental mechanics underlying segregation of charged particles.

1.3 Overview

Chapters 2, 3, and 4 start with an introduction to the topic discussed. In Chapter 2, we discuss the charging of fluidized granular beds in the presence of an external field. In section 2.2, we present our model and in section 2.3, we show that the charging mechanism proposed by the previous simplified model continues to function even under more detailed considerations. We show that strong charge heterogeneities can be established inside the bed and we present our prediction that shielding due to these heterogeneities can dramatically alters the charging rate in the bed.
1.3. OVERVIEW

In Chapter 3, we investigate the charging of an agitated bed in the absence of an external field. Section 3.2 consists of three parts. In the first part, we describe our particle-based simulations and present their results. Here, we show that particle charge can grow exponentially rapidly. In the second part, we introduce a mathematical model to provide a framework to understand the observations of the simulations. In the last part, we confirm the predictions of the simulations and the mathematical model in experiments using vibrated grains under microgravity.

In Chapter 4, we study segregation of a binary mixture of differently charged particles under shear. In section 4.2, we describe our simulation: e.g. a discrete-element simulation coupled with Coulomb interactions. In section 4.3, we show that particles segregate according to their net charge. We then construct a simple model to describe the spatial pattern of this segregation. We discuss a non-monotonic dependence of the intensity of segregation on the shear velocity. Here, we show that the optimal shear velocity, the point at which the segregation maximally intensifies, results from a competition between diffusional and Coulomb fluxes.

Detailed conclusions and outlooks of the studies of Chapters 2, 3, and 4 can be found at the end of the Chapters in the Conclusion sections. An overall conclusion of this dissertation is in Chapter 5.
Chapter 2

Field Driven Charging
Dynamics of a Fluidized Granular Bed

2.1 Introduction

Pähtz et al. proposed a simplified model [30] in which an external electric field - as might be produced by a nearby electrical storm [60] - can induce polarizations in grains to explain the charging of identical grains. In their model, an electric field polarizes initially electrically neutral grains as depicted in Fig. 2.1 (a) and (b). Polarizations due to a downward pointing electric field produce negatively charged upper and positively charged lower hemispheres. When two polarized grains collide, contacting hemispheres of both grains neutralize, as indicated in Fig. 2.1 (c). The result of such binary collision is that the top and the bottom hemispheres of the top and the bottom grains, respectively, retain a charge, whereas the contacting hemispheres become neutral. After the collision, as shown in Fig. 2.1 (d), each grain is again polarized according to the field and so charges are added to the top and bottom hemispheres. The result of this whole process is to increase the negative charge of the upper particle, and the positive charge of the lower one. If the same process occurs for every collision, collisional granular flows should pump negative charges up and positive charges down, as depicted in
Figure 2.1: Caricature showing the inductive charging of identical grains in the presence of an external field. (a) Without an external field, grains are electrically neutral and unpolarized. (b) In the presence of an external field, grains are polarized. (c) When grains come into contact, the touching hemispheres of the grains neutralize. (d) Grains exposed to an external field again polarizes.
2.1. INTRODUCTION

Figure 2.2: Caricature of a granular bed in collisional flows pumping negative charges up and positive charges down in the presence of a downward external electric field. Here, the bottom is insulating.

Fig. 2.2. The validity of the model was demonstrated in a table top experiment. Details can be found in Ref. [30], but in short, the experiment was contained in a glass jar, in which glass beads were fluidized by air blown from below. The airflow was set to the lowest pressure so that the grains just above the bottom plate become fluidized. By design, grains could only charge by contact with one another or with the grounded bottom. An electric field was applied by placing a metal plate at the top of the glass jar which was connected to a van de Graaff generator.

When the generator was turned on, grains floated energetically within the glass jar and hovered or bounced against the upper surface. And, subsequently, when the generator was turned off, grains remained adhered both to the top surface and to the side of the glass jar. The upper plate was at a high positive potential, so only negatively charged grains could remain adhered to the nearby glass. The bottom plate was grounded, and so there was no source of negative charge anywhere within the glass jar. This was an evidence of grains at the top attaining strong negative charges during the collisional flows.

This simplified model, shown in Fig. 2.1, provided a prediction whose validity was confirmed experimentally, but neglected some important physical
Figure 2.3: Caricature of the expected charge heterogeneities. (a) For a shallow granular bed, we expect a monolayer of negative charges at the top of the bed and (b) for a thicker bed, we expect a charge inversion due to the field induced by the negative layer.

aspects. Namely, the model only considered vertical dipole moments and neglected any electrostatic interactions between grains. The model assumed that the external field was overwhelmingly stronger than that due to nearby grain charges. In this chapter, we study the charging of fluidized granular bed in the presence of an external electric field by removing these assumptions and refining the model by including higher order electrical moments on grains and by allowing grains to interact through Coulomb forces and other electrostatic interactions.

Our goals in performing these more detailed simulations are twofold. Interactions between charged and polarized grains, which the prior work neglected, has been shown to play an important role in the dynamics of grains, especially in close distances [57], and it has been shown that when grains collide in the presence of an external field, the amount of exchanged charges increases with the strength of the field [61]. So, first we include these inter-
actions and evaluate how they affect subsequent grain charging.

Second, the charging mechanism described in Ref. [30] should produce significant charge heterogeneity that has not been quantitatively examined. In particular, if the bottom of the bed is grounded, then positive charges will be drained from beneath the bed, leaving growing negative charges near the top of the bed, as sketched in Fig. 2.3 (a). Significantly, we can expect these negative charges to shield the applied field within the bed and so to reduce granular charging. Moreover, as this shielding layer of negative charges grows sufficiently large, it can exceed the influence of the external field and induce its own charges within the bed - in this case of opposite, positive sign. This would produce a double layer, with negative charge at the top of the bed, overlying an induced positive charge, sketched in Fig. 2.3 (b).

We therefore present a detailed simulation of granular charging both to provide a more accurate investigation into the mechanism of charge transfer than previously possible, and to quantify expected charge heterogeneities in granular beds. In section 2.2 we describe this simulation, then in section 2.3, we analyze results, and in section 2.4, we draw conclusions.

2.2 Model

We simulate charging of grains in an agitated bed of spherical grains. Grains can accumulate charges on their surface, and since they are insulators, these charges are heterogeneously distributed.

Surface charges: To account for such heterogeneity, we model the surface as six independent, orthogonally placed charges as sketched in Fig. 2.4 (a), which allows us to define complex electrical moments.

Grain sizes: To prevent crystallization, we consider a polydisperse distribution of grain radii, $R_i$, following a Gaussian distribution with variance 10% of the mean $\bar{R} = 0.75$ mm. However, as we are interested in the dynamics of identical grains, we assume that all grains have the same mass, $m = 4.239 \times 10^{-6}$ kg, corresponding to a glass of average radius $\bar{R}$ and density $\rho_g = 2.4 \times 10^3$ kg/m$^3$. 
Figure 2.4: (a) Schematic representation of a grain with six constituent charges. Each charge is centered on a surface domain delineated here by broken lines. (b) Illustrative dipole moment calculated from charges on panel (a).

Equations of motion: Numerically, we solve the equations of motion for each grain by means of the discrete element method [62]. We track both translational and rotational motions of the $i^{th}$ grain and include mechanical and electrostatic forces and torques:

$$m \frac{d\vec{v}_i}{dt} = [\vec{F}_g + \vec{F}_{ela,i} + \vec{F}_{f,i}] + \vec{F}_{ele,i},$$

$$I_i \frac{d\vec{\omega}_i}{dt} = [\vec{T}_{m,i}] + \vec{T}_{e,i}. \quad (2.1)$$

Mechanical forces: The mechanical terms, in square brackets, are as follows. The gravitational force, $\vec{F}_g$, is defined in the usual way using earth’s gravity: $g = 9.807 \text{ m/s}^2$.

For the elastic force, $\vec{F}_{ela,i}$, we use the model of Walton and Braun [63]. In this model, the force acting on the $i^{th}$ grain resulting from a collision with the $j^{th}$ grain is given by:

$$\vec{F}_{ela,ij} = \begin{cases} 
0, & \vec{\epsilon}_{ij} = 0 \\
k_l\vec{\epsilon}_{ij}, & \frac{d|\vec{\epsilon}_{ij}|}{dt} \geq 0 \\
k_u\vec{\epsilon}_{ij}, & \frac{d|\vec{\epsilon}_{ij}|}{dt} < 0 
\end{cases} \quad (2.2)$$
where $\vec{\epsilon}_{ij}$ is the overlapping vector defined as:

$$
\vec{\epsilon}_{ij} = \begin{cases} 
0, & \text{if } (R_i + R_j - |\vec{r}_{ij}|) \leq 0 \\
(R_i + R_j - |\vec{r}_{ij}|)\hat{r}_{ij}, & \text{otherwise.}
\end{cases}
$$

(2.3)

Here, $\vec{r}_{ij}$ is the vector connecting the centers of the two grains from $j$ to $i$. $k_l$ and $k_u$ are the elastic coefficients when the colliding grains are approaching or moving away from one another, respectively. We use $k_l = 0.07$ and $k_u = 0.08$, thus fixing the restitution coefficient to $\sqrt{k_l/k_u} = \sqrt{0.07/0.08} \approx 0.935$. This value is the same as was used by Ref. [30], and is deliberately chosen to be large because we agitate beds of varying depths from below (described shortly). Lower restitution coefficients require either that we increase the agitation strength with bed depth or that we maintain strong constant agitation. The first alternative introduces a new parameter that changes with depth, while the second produces very different states for shallow and deep beds. Neither is desirable, whereas by choosing a high restitution coefficient, we are able to produce a nearly uniformly colliding state for all beds studied without changing the agitation strength [64].

We use a standard kinetic friction model for $\vec{F}_{f,i}$ [65]. The frictional force acting on the $i^{th}$ grain resulting from a collision with the $j^{th}$ grain is given by:

$$
\vec{F}_{f,ij} = -\mu_k |\vec{F}_{ela,ij}|\hat{v}_{ij},
$$

(2.4)

where $\mu_k = 0.4$ is the kinetic friction coefficient, and $\hat{v}_{ij}$ is the unit vector with the direction of the relative velocity of the contact points of two colliding grains:

$$
\vec{v}_{ij} = (\vec{v}_i + \vec{\omega}_i \times \vec{R}_{cont,i}) - (\vec{v}_j + \vec{\omega}_j \times \vec{R}_{cont,j}),
$$

(2.5)

where $\vec{R}_{cont,i,j}$ are the position vectors of the contact point measured from center of the $i^{th}$ and the $j^{th}$ grains.

As for the torque equation, $I_i = \frac{2}{5}m\bar{R}^2$ is the moment of inertia of the $i^{th}$ grain, and the mechanical torque, $\vec{T}_{m,i}$, is determined from the kinetic friction at the contact point of two colliding grains [65].
We agitate the bed by using a “splash function” to re-inject grains that hit the bottom of the computational domain. Such grains acquire velocity \( \vec{V}^s = 2.7\sqrt{2gR\hat{z}} \), which has long been used to model saltation and fluidization [66].

**Electrostatic forces:** To calculate the electrostatic terms in Eq. (2.1), we track all six charges shown in Fig. 2.4 (a) for every grain. Instead of a fully detailed calculation of the electrostatic interactions [67], we only use mono and dipole terms to compute Coulomb forces, \( \vec{F}_{\text{ele,i}} \), and electrostatic torques, \( \vec{T}_{\text{e,i}} \).

The mono terms are calculated from the net charge of the grain defined as:

\[
q^m_i = \sum_{n=1}^{6} q_{i,n}, \quad (2.6)
\]

where \( q_{i,n} \) is the \( n^{th} \) constituent charge of the \( i^{th} \) grain. The dipole terms are defined from the dipole moment of the grain:

\[
\vec{d}_i = \sum_{n=1}^{6} q_{i,n}\vec{r}_{i,n}, \quad (2.7)
\]

where \( \vec{r}_{i,n} \) is the position vector of the \( n^{th} \) constituent charge measured from the grain center. A virtual charge representing the net charge is fixed at the center of the particle and dipole forces and torques are defined via two virtual charges displaced by \( \frac{2}{3}R_i \) from the center of the grain, as sketched in Fig. 2.4 (b). This is to prevent divergence when two grains come into contact. Each virtual charge: \( q^d_i \), of the dipole moment is obtained from:

\[
q^d_i = \frac{3|\vec{d}_i|}{4R_i}, \quad (2.8)
\]

We avoid far-field dipole approximations and calculate all Coulomb forces exactly by evaluating \( k_e q_1 q_2 / r_{12}^2 \) for all charges, \( q_1 \) and \( q_2 \), and all distances \( r_{12} \) between these charges, where \( k_e \) is the Coulomb constant, as shown in Fig. 2.5.

We account for long-range electrostatic interactions between charged grains by using the Particle-Particle Particle-Mesh (PPPM) method [68]. In this method, the computational domain is divided into three dimensional grid
2.2. MODEL

Figure 2.5: Cartoon showing Coulomb interaction between the $i^{th}$ and $j^{th}$ grains in a short distance.

cells, and net charges of the grains are assigned to the closest intersections between cells. Accordingly, the total charge assigned to the $k^{th}$ grid intersection is:

$$Q_k = \sum_{i=1}^{N} q_i W(\vec{r}_k - \vec{r}_i),$$

(2.9)

where $N$ is the total number of grains, $q_i^n$ is the net charge of the $i^{th}$ grain, $\vec{r}_k$ is the position vector of the $k^{th}$ grid intersection and $\vec{r}_i$ is the position vector of the center of the $i^{th}$ grain. $W(\vec{r}_k - \vec{r}_i)$ is the charge assignment function defined as:

$$W(\vec{r}_k - \vec{r}_i) = \begin{cases} 
1, & \text{if } -2\bar{R} \leq (x_k - x_i) < 2\bar{R}, \\
-2\bar{R} \leq (y_k - y_i) < 2\bar{R}, \\
\text{and } -2\bar{R} \leq (z_k - z_i) < 2\bar{R} \\
0, & \text{otherwise}
\end{cases}$$

(2.10)

where $4\bar{R}$ is the linear length of the cubic cell. The electric field at the $k^{th}$ grid intersection is:

$$\vec{E}_k = k_e \sum_{l \neq k}^M Q_l \frac{\vec{r}_{kl}}{|\vec{r}_{kl}|^3},$$

(2.11)
where $\vec{r}_{kl}$ is the vector connecting the two grid intersections from $l$ to $k$. For long-range interactions, we approximate the field at the center of a grain to be the field at the closest grid intersection. We include torques on dipoles due to Coulomb interactions within a grain’s grid cell and its nearest neighboring cells, and neglect Coulomb dipole forces due to charges outside of this domain because the field due to dipole moment decays rapidly with $\frac{1}{r^3}$: the electric field due to a dipole moment beyond that domain is at most less than 1 % of the strength of the field due to the dipole moment within the same cell.

Coulomb forces for grains within any grid and its nearest neighbors are calculated in the usual way according to:

$$\vec{F}_{ele,ij} = k_e q_{i,1} q_{j,2} \frac{\vec{r}_{12}}{|\vec{r}_{12}|^3}$$ \hspace{1cm} (2.12)

where $q_{i,1}$ and $q_{j,2}$ are all net or dipole charges of both grains and $\vec{r}_{12}$ is position vectors between these charges, as shown in Fig. 2.5.

Finally, Coulomb forces including both short- and long-range terms are calculated according to:

$$\vec{F}_{ele,i} = q_i \vec{E}_{ex} + \sum_{i \neq j}^{n,n.} \vec{F}_{ele,ij} + q_i \vec{E}'_{k},$$ \hspace{1cm} (2.13)

where the first term is the net charge coupling with the applied external field, $\vec{E}_{ex}$, the second term is the short-distance interaction with the grains within the same or nearest neighbor grid cells and the last term is the long-range interaction with distant grid intersections. To prevent double-counting, we subtract the contribution to $\vec{E}_k$ from the closest grid intersection and its nearest neighbors (thus, $\vec{E}'_k$ in Eq. (2.13)): these terms are calculated exactly using Eq. (2.12). Barros et al. proposed an alternative method to numerically calculate the electrostatic interaction between dielectric objects by meshing each grain surface into hundreds of polarizable patches [69], which is rather expensive to apply to a system consisting of thousands of polarizable dielectric particles.

We duplicate the computational domain ten times in both horizontal directions for a total of 440 duplicates surrounding a central domain, and we
2.2. MODEL

use the electric field, $\vec{E}_i$, from these duplicates to calculate Coulomb forces and induced charges on each grain. In this way, we produce nearly periodic boundary conditions in a finite computational domain.

**Charge transfer:** We simulate charge transfer similarly to Ref. [30]: each grain receives an induced polarization proportional to the electric field that it is subjected to, and charges can be transferred between grains due to neutralization events during contact.

In detail, a local electric field of amplitude $E_i$ at the center of the $i^{th}$ grain, calculated by Eq. (2.13), induces a dipole moment:

$$\vec{P}_i = \alpha \vec{E}_i.$$  \hspace{1cm} (2.14)

Here $\alpha$ is the polarizability in cgs units. Unlike Ref. [30], $\vec{E}_i$, is not only the applied external electric field but also includes the field due to surrounding grains. The induced dipole moment is assigned to the nearest of the three pairs of charges shown in Fig. 2.4 (a). In order to compute neutralization in a well-defined and charge-conserving manner, during collision permanent charges on the contacting sectors neutralize according to:

$$q_{i,\text{after}} = q_{j,\text{after}} = \frac{1}{2} [q_{i,\text{before}} + q_{j,\text{before}}],$$  \hspace{1cm} (2.15)

where $q_i$ and $q_j$ are the charges on contacting domains of the $i^{th}$ and $j^{th}$ contacting grains. During a collision, we add the induced dipole charges to the nearest sector of the three pairs of charges to produce new permanent charges analogous to the repolarization process shown in Fig. 2.1 (d).

**Time step:** The time step for the numerical integration is set to $5 \times 10^{-5}$ seconds. This produces, for the most energetic grains, a minimum of more than 100 time steps per collision, allowing the dynamics to be stable.

**Initial configuration:** We obtained the initial configurations of the simulation by dropping the grains freely onto the bottom while switching off the splash function and the external field. Grains are all neutral during initialization and there is neither charge neutralization nor polarization. We wait $10^6$ time steps (50 sec.), by which time grain velocities become negligibly small (less than $|\vec{V}^s| \times 10^{-3}$, where $\vec{V}^s$ is the splash function defined
earlier). At this point, we reinstate the full collision, including the splash function, external field, Coulomb interactions, etc. From this point on, it takes about another 50 seconds for the largest system with 1500 grains to reach an asymptotic steady state, meaning a state whose mean charge per grain reaches an asymptote.

**Other technicalities:** Finally, we mention four technical points needed to close the description of the simulation. First, we model granular charging using a grounded bottom surface. This is the boundary condition used in Ref. [30], which permits the injection of charge into the bed to mimic field and laboratory observations [70, 71, 72, 60]. Second, to prevent spurious repetition of charging, we only apply induction and neutralization operations (Eqs. (2.14) and (2.15)) once per contacting grain pair, at the moment when the grains separate. Third, the algorithm that we have described applies to binary collisions. In rare cases when a grain simultaneously loses contact with multiple neighbors, we perform neutralization sequentially in random order. Fourth, grains at the top of the bed acquire sufficient charge to levitate against gravity. As in prior work Ref. [30], we remove these grains from the simulation once they lose contact with other grains. To keep the number of grains constant, we re-inject these grains at the bottom of the bed. We note that injecting charged grains at the bottom of the bed would introduce a spurious electrical current: to prevent this, we neutralize these grains before re-injection.

**Validation:** To validate the model, we count the numbers of levitated grains and compared them to the values reported by Pähtz et al. [30], which were already validated by comparing to experimental results. In particular, it was shown that it reproduces the dependence of the number of levitated grains on the total number of grain layers obtained experimentally. With our model, we also reproduced this dependence. See Fig. 2.6, where we show our numerical results (back) and the ones by Pähtz et al. (red).
2.3. RESULTS

Figure 2.6: Number of levitated grains versus bed depth comparing the results reported by Pähtz et al. [30] (red) and obtained using our simulations (black). Each data point is an average of 200 measurements taken at sequential times in the asymptotic state and the error bars show the standard errors of the mean.
Figure 2.7: Visualization of the bed with (a) nominal bed depth $n_L = 5$ and (b) $n_L = 13$ in asymptotic states. Grains are colored according to their net charge, where a unit charge corresponds to $1.3 \times 10^{-12}$ C. One particle layer here consists of approximately 100 grains.
2.3 Results

Using this simulation that we have described, we first evaluate the extent to which heterogeneities in the bed appear as discussed in the introduction, and we second dissect the mechanism of granular charging in greater detail than previously possible.

Charge heterogeneities: Qualitative assessment of expected charge heterogeneities can be seen from Fig. 2.7, where we visualize the granular beds in their asymptotic states. For a shallow bed, we see negative charges accumulating at the top, and for a deeper bed, we see strong negative charges at the top, overlying positive charges.

We quantitatively assess these observations by performing independent simulations for different bed depths. We then bin charge and electric field values as a function of height, as shown in Fig. 2.8 where we display charge and field values as a function of bed depth, \( n_L \). To bin charges and fields, we divide the computational domain into horizontal slices of thickness \( 2\bar{R} \), and integrate the charge or the vertical component of the field over each slice. In agreement with Fig. 2.7, Fig. 2.8 (a) shows that for \( n_L \geq 3 \), a layer of negatively charged grains forms at the top of the bed, and Fig. 2.8 (b) confirms that the field beneath this layer drops significantly. The horizontal components of electric field are invariably two orders of magnitude smaller than the vertical component, \( E_z \), so we report only \( E_z \) in Fig. 2.8 (b). Once the bed depth exceeds about 9 grain diameters, a second layer of positively charged grains emerges, shown in red in Fig. 2.8 (a). There is also a suggestion of a third layer, of negative charges, beneath this. These results suggest that, as the number of layers increases, the top layer of negative charges eventually exceeds the influence of the external field and it induces charges of opposite sign beneath it, as shown in Fig. 2.8.

Thus our results appear to confirm expectations that there should be significant electrical heterogeneity in agitated granular beds, with charges concentrating near the top of the bed, and electric fields within the bed being strongly shielded by these charge concentrations.

Charging mechanism: As we have mentioned, our simulations are more de-
Figure 2.8: (a) Net charge in different height bins (see text) versus bed depth. Black line shows top of granular bed and cells are colored according to mean charge of grains within each bin. Color bar shows charge magnitude, 1 unit charge is $1.3 \times 10^{-12} \text{C}$. (b) Vertical component of field versus bed depth. The applied vertical external field is 300 kV/m pointing down, and color bar shows vertical field magnitude in kV/m. Black line again indicates top of granular bed.
2.3. RESULTS

tailed than those performed previously, and so we can probe the essential mechanism that Ref. [30] sought to elucidate, namely how charging of identical grains occurs and what it depends on.

We investigate the charging mechanism by performing our simulations under strategically differing conditions. First, we repeat the results of Ref. [30] at multiple bed depths by excluding multipoles on grains, Coulomb forces between grains, and field-dependent polarization (Eq. (2.14)). Second, we include multipoles and repeat the same simulations, and finally we run the full simulation that we have described up to this point. In all cases, we evaluate the grain charging, explicitly the mean absolute value of the grain charge, \(<|q|>\). Results are shown in Fig. 2.9.

Red symbols in Fig. 2.9 show simulation for conditions of Ref. [30], blue symbols include multipoles that rotate with grains, and black symbols indicate the full simulations.

From Fig. 2.9 (a), we see that including multipoles has little effect: simulations with dipoles oriented only in \(\hat{z}\) direction (red), and those with multipoles that rotate with grains (blue) produce largely similar charging behaviors. This is unexpected: one might anticipate that rotating dipoles would orient in arbitrary directions, as likely building as diminishing ultimate charging, but in fact this only slightly reduces ultimate charges on grains. From this perspective, prior work using a simplified model appears to be essentially unchanged by more careful simulations. On the other hand, including Coulomb forces and field-dependent charge induction (black) dramatically reduces charging rates, by as much as a factor of five. This again is unanticipated.

To establish the cause of this reduction, we recall that it has previously been determined (Ref. [30]) that charging is proportional to collision rate. It is therefore plausible that Coulomb forces could prevent collisions between like-charged particles and so could lead to the reduced charging shown in Fig. 2.9 (a). To assess this possibility, we evaluate the collision rate, summed over each of our simulations, and plot these in Fig. 2.9 (b). Evidently the collision rate reaches a maximum at different bed depths for each simulation scenario,
Figure 2.9: (a) $|\langle q \rangle| >$ vs. bed depth in several scenarios. (Red) Simulations lacking multipoles, Coulomb forces, and field-dependent polarization (similar to Ref. [30]). (Blue) Simulations with multipoles that rotate with grains, but lacking Coulomb forces and field-dependent polarization. (Black) Full simulations described in text. (b) Rate of grain collision events vs. bed depth in each of these scenarios. Each data point is an average of 200 measurements taken at sequential times in the asymptotic state and the error bars show the standard errors of the mean (smaller than the symbols).
2.4. CONCLUSION

but each scenario reaches the same maximum, and at $n_L = 8-10$, where from Fig. 2.9 (a) we see that granular charging differs most dramatically between the full simulation and its simplified models, collision rates shown in Fig. 2.9 (b) are nearly identical for all three cases. Evidently then, changes in collision rates due to Coulomb forces cannot account for the change in charging seen in Fig. 2.9 (a).

Thus including Coulomb forces has a weak effect on collision rate, but this does not account for the reduction in bed charging seen. On the other hand, we have already seen from Fig. 2.8 (b) that the bed is strongly shielded from applied fields by the overlying charge layers shown in Fig. 2.9 (b). Since the more complete model represented by black symbols in Fig. 2.9 (a) generates charging in proportion to local electric fields, it seems likely that this shielding effect is responsible for reduced charging.

2.4 Conclusion

We performed detailed simulations of charging in agitated granular beds in the presence of an external field. These simulations confirm that in the presence of a vertical external electric field, colliding grains pump negative charge from a grounded bottom surface to the top of the bed. The simulations also reveal that the charges at the top of the bed can grow until they shield the interior of the bed from the external field. At this point, logically enough, granular charging is suppressed. We find that although the essential mechanism previously investigated continues to function in the presence of the more detailed considerations that we have described, the shielding observed can reduce ultimate granular charge levels by as much as a factor of five from those calculated using a more simplified model. We have also found that significant charge heterogeneities can be established in agitated beds that are exposed to external fields, and we anticipate that future studies into heterogeneous charge distributions may provide insights into both charging and discharging dynamics in natural and industrial granular flows.
Chapter 3

Self-charging of identical grains in the absence of an external field

3.1 Introduction

Granular charging is observed for identical materials and experiments show that this charging grows with repeated contacts [28, 29]. Previous work has shown that one origin of this energy can be an external electric field that feeds electrification [30, 73, 74, 75, 76].

In this chapter, we demonstrate that an external field is not needed: infinitesimal charges on grains themselves can induce charges on their neighbors, bootstrapping one another to grow exponentially rapidly in agitated beds. Unlike prior work [59], we show that the energy for this charge growth can arise strictly conservatively, trading mechanical work for electrical energy. It is by no means self-evident whether or when this could cause a buildup of significant charges, but as we will show, this feedback mechanism can generate very large electrical charges and can promote a rich set of macroscopic charge patterns within the bed.

The underlying mechanism that we explore here is that the electric field from charged particles induces a polarization on a neighbor. An induced
Figure 3.1: Typical snapshot of simulation showing the charging of an agitated granular bed. Here we use $\chi_c = \eta = 1$ as defined in text, and show time after 5 seconds. Grains are colored according to their net charge defined as: $\sum_{k=1}^{6} q_{i,k}$ (blue: negative, red: positive); with grains with an absolute net charge magnitude below 20 pC in transparent grey.
polarization of the $i^{th}$ grain is aligned with and proportional to the electric field, $\vec{E}_i$, at the center of the grain. We compute the polarization density as:

$$\vec{p}_{i}^{\text{ind}} = \epsilon_0 (\epsilon_r - 1) \vec{E}_i,$$  

(3.1)

where $\epsilon_0$ is the vacuum permittivity and $\epsilon_r$ is the relative permittivity of dielectric particles. We then integrate $\vec{p}_{i}^{\text{ind}}$ over a surface domain (described shortly or see Fig. 3.2) and obtain the induced surface charge:

$$q_{i}^{\text{ind}} = \oint_S \vec{p}_{i}^{\text{ind}} \cdot d\vec{s} = \frac{4\pi R_d^2}{6} |\vec{p}_{i}^{\text{ind}}|,$$  

(3.2)

where $R_d$ is an effective dipole radius, from which we calculate the dipole moment of the particle using two surface charges:

$$\vec{P}_{i}^{\text{ind}} = \frac{\chi e R_d^3}{k_e} \vec{E}_i,$$  

(3.3)

where $k_e$ is Coulomb’s constant, and $\chi_e$ is the grain polarizability which we define as:

$$\chi_e = \frac{\epsilon_r - 1}{3}.$$  

(3.4)

The relative permittivity of dielectric materials we are interested typically ranges from 1 to 4 (for example, PTFE : 2.1, polyethylene : 2.25, silicon dioxide (sand) : 3.9), and so we fix the parameter range for $\chi_e$ to zero to unity.

In a bed of colliding grains, we also assume that when the $i^{th}$ and the $j^{th}$ particles come into contact, charges $q_{i,k}$ and $q_{j,l}$ on the grain surfaces can partially neutralize [30, 73] according to:

$$q'_{i,k} = \left(1 - \frac{\eta}{2}\right) q_{i,k} + \frac{\eta}{2} q_{j,l},$$

$$q'_{j,l} = \left(1 - \frac{\eta}{2}\right) q_{j,l} + \frac{\eta}{2} q_{i,k},$$  

(3.5)

where the prime denotes the constituent charges after collision, $\eta$ is a neutralization efficiency which ranges from zero to unity, and $k$ and $l$ denote the two constituent charges of the $i^{th}$ and $j^{th}$ grains, respectively, that are in contact upon collision. We assume that particles initially have charges of the order of $10^5$ elementary charges and we observe a growth by three orders of magnitude as we will show in the next section.

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Figure 3.2: Schematic representation of grains with six independent charge domains of area $\frac{1}{6}\pi R_i^2$ immediately following the neutralization process. Each domain charge is fixed at $2/3$ of the mean grain radius from the center of the grain. Two contacting domain charges, $q_i$ and $q_j$, neutralize at a contact point to become $q_i'$ and $q_j'$. Size of the domain charges here represent the magnitude of the charges.

3.2 Results

We describe a particle-based simulation of charging of agitated grains subject to Eqs. (3.3) and (3.5) followed by a continuum mathematical model, with the goal of establishing whether induced polarization combined with contact neutralization can amplify small initial charges. We conclude with an experiment confirming that this amplification occurs as predicted.

Discrete element simulation

We begin by performing discrete-element method (DEM) [62] simulations of mechanically and electrically interacting dielectric particles. Grain size: The particle sizes are polydisperse to prevent crystallization: that is that the radius of each particle, $R_i$, is Gaussian distributed with standard
deviation 10% of the mean radius, $\bar{R} = 0.75$ mm, and each grain has the density of glass, $\rho_g = 2.4$ g/cm$^3$.

**Surface charge:** As in Chapter 2, to account for a heterogeneous charge distribution on the surface of insulating grains and to calculate polarizations and Coulomb forces between them, we embed within each particle three pairs of orthogonally placed constituent charges (see Fig. 3.2) at fixed separations $2R_d$, where $R_d$ is 2/3 of the grain radius. To compute forces and torques, we then consider a multipole expansion and truncated after the second-order term.

**Equations of motions:** We track translational and rotational motions of each grain by evaluating is dynamics due to mechanical and electrical forces and torques acting on it. We use the model of Walton and Braun [63] for the elastic force and fix the two elastic coefficients, $k_l = 0.07$ and $k_u = 0.08$, to achieve the restitution coefficient, $\sqrt{k_l/k_u} = \sqrt{0.07/0.08} \approx 0.935$. For friction, we use a standard kinetic friction with kinetic friction coefficient, $\mu_k = 0.4$.

We then solve the equations of motion for each grain by means of the DEM on a domain that is periodic along the horizontal directions. We achieve periodicity by surrounding the computational domain by 8 copies in the horizontal plane, which we use for field and energy calculations. The time step is set to 50 $\mu$sec., which produces over $10^2$ steps per collision for the shortest time collisions between grains. The top of the computational domain is free, and the bottom is fixed and insulating. Any grain that hits the bottom acquires additional upward energy defined by a kick velocity, $\vec{V} = 2.7\sqrt{2g\bar{R}}\hat{z}$, which maintains the granular bed in a collisional state.

**Initialization:** We initialize each simulation by dropping $10^3$ grains onto the fixed bottom, of area $20\bar{R} \times 20\bar{R}$. No energy is injected (through kicks by the bottom surface) while particles settle, and grains are all initially neutral. We wait ten seconds until grain velocities become negligibly small (kick velocity/$10^3$). We then add charges uniformly distributed on $[-10^{-2}, 10^{-2}]$ pC to all six domains of all grains and thereafter kick particles at the bottom surface.
Figure 3.3: Schematic description of an interaction between a permanent net charge and an induced dipole (red arrow). Here, particle $i$ has a net charge $q_i$ which is inducing a dipole in particle $j$ with the surface charge magnitude $q_{ji}$.

**Polarization:** Particles polarize according to Eq. (3.3), where we emphasize that $\vec{E}_i$ is the electric field at the center of the $i^{th}$ grain due to all pre-existing permanent charges in the system. The distinction between permanent and induced charges is important because induced charges are slaved to the external field, and cannot themselves do work. For example, induced charge dipoles always point in the direction of an external field and so cannot exert torque on a grain. Permanent charges, on the contrary, are fixed on a grain and exert forces on other charges \cite{77}. An induced polarization, given as a superposition of multiple orders of electrostatic interaction, could point at any arbitrary direction, and appropriately retains the effects of higher order polarizations. It is only when particles collide that induced charges are mapped onto the domains to become permanent (described shortly).

**Energy conservation:** We enforce energy conservation in two ways. First, we compensate for the energy associated with assembling the induced dipole moment prescribed by Eq. (3.3) by integrating the work needed to bring the
3.2. RESULTS

Figure 3.4: Schematic description of an induced dipole of grain \( i \) inducing a dipole in grain \( j \). Grain \( i \)'s dipole moment shrinks in its size from \( 2\bar{R}q_{i}^{\text{ind}} \) to \( 2\bar{R}q_{i}^{\prime \text{ind}} \) to induce a dipole in grain \( j \) of size \( 2\bar{R}q_{ji}^{\text{ind}} \). \( U_{ij}^{\text{ind}} \) is the interaction energy between the two induced dipoles.

induced charges to their positions from infinity [78]. We then evaluate the gradient of this energy, which gives a mechanical force that we apply to each particle. This is the force that must be exerted to polarize the particle, and the spatial integral of this gradient is mechanical work that exactly equals the required electrical energy. For example, as shown in Fig. 3.3, when grain \( i \) has a net charge, \( q_{i}^{m} \), it induces a polarization in grain \( j \), whose surface charge magnitude is:

\[
q_{ji} = \frac{\chi_{e}R^{2}_{d}}{2k_{e}}|E_{ji}| \quad (3.6)
\]

Here, \( E_{ji} \) is the field produced by \( q_{i}^{m} \) at the center of grain \( j \). We compute the electrostatic energy needed to assemble the configuration:

\[
U = -k_{e}\frac{q_{i}^{m}q_{ji}}{x-R_{d}} + k_{e}\frac{q_{i}^{m}q_{ji}}{x+R_{d}} - k_{e}\frac{q_{ji}^{2}}{2R_{d}} \quad (3.7)
\]

and apply a repulsive force on the center of both grains, which we obtain as:

\[
F_{ij}(x,q_{i}^{m}) = \frac{\partial U}{\partial x} \quad (3.8)
\]

The same computation is done when permanent dipoles induce polarizations in neighboring grains.

Second, we note that an induced dipole moment changes the electric field of neighboring grains, and this change in turn induces secondary dipole mo-
ments according to Eq. (3.3). To conserve energy, we account for secondary dipoles by reducing each primary moment by exactly the energy associated with every secondary moment. For example, when an induced dipole of grain \( i \) induces a dipole in grain \( j \), we reduce the dipole of grain \( i \), as shown in Fig. 3.4. The reduced dipole moment is obtained by solving the following quadratic equation for \( q'_{\text{ind}} \):

\[
U_{\text{total}} = -\frac{k_e q_{\text{ind}}^2}{2R_d} = -\frac{k_e q'_{\text{ind}}^2}{2R_d} - k_e \frac{q_{\text{ind}}^2}{2R_d} + U_{\text{ind}}^{ij},
\]

(3.9)

where \( q_{\text{ind}}^{ji} \) here is the induced dipole charge of grain \( j \) given by the field of due to the induced dipole of grain \( i \), and \( U_{\text{ind}}^{ij} \) is the interaction energy between the two induced dipoles calculated in the normal way. This process feeds back iteratively, so that every secondary moment in turn induces another moment on the originating dipole. We have numerically confirmed that this feedback converges rapidly, and after two iterations the error in neglecting higher order terms is less than 0.8%. Consequently, in our simulations we perform two iterations of inducing new additive dipole moments based on this feedback process.

Neutralization: Constituent charges shown in Fig. 3.2 take part in neutralization events defined by Eq. (3.5). At the instant when two grains collide, neutralization is imposed between contacting charge domains of colliding grains \( i \) and \( j \) according to Eq. (3.5). This permits charge transfer between grains, so for \( \eta = 0 \) all charges remain unchanged, and grains increasingly transfer charges as \( \eta \) grows. Equation (3.5) is applied during binary collisions, and whenever a grain contacts multiple neighbors during a single time step, we perform this operation for all pairs in random order.

Both permanent and induced charges take part in neutralization events, and to keep accounts straight, we add exactly the fraction of induced charges needed to conserve charge to the permanent charges. That is, if an induced charge \( \Delta q \) is added to one domain of a grain due to Eq. (3.5), then \( -\Delta q \) will be made permanent on the opposing domain of the same grain to guarantee overall charge neutrality. Finally, we prevent spurious repetition of charging by only applying neutralization and induction operations at the moment.
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when two grains first touch one another.

Note that neutralization events also involve energy considerations: when a particle of charge \( q_i \) fully neutralizes \((\eta = 1)\) during collision with a neighbor, each particle will leave the collision with a charge of up to \( q_i/2 \). This produces repulsion between the particles that was not present prior to the collision. This repulsion is a real physical effect that is seen in experiments [79], and so we include the repulsion in our simulations.

To quantify charge growth, we evaluate the evolution of the absolute value of all charges averaged over all grains:

\[
\bar{q} = \frac{1}{N} \sum_{i=1}^{N} \left| \sum_{k=1}^{6} q_{i,k} \right|,
\]  

(3.10)

as a function of model parameters \( \chi_e \) and \( \eta \), as shown in Fig. 3.5 using \( N = 10^3 \) particles.

Figure 3.5 (a) shows that for large constant polarizability, \( \chi_e \), \( \bar{q} \) typically does grow roughly exponentially following an initial transient and continuing up to an asymptote that we discuss shortly. We plot \( \bar{q} \) here, but remark that polarizations, and charges of both signs, also grow with the same exponential rate. For fixed neutralization, \( \eta \), \( \bar{q} \) also exhibits an exponential growth period, however for small \( \chi_e \), \( \bar{q} \) decreases in time, as shown in Fig. 3.5 (b).

Figure 3.6 collects growth rates obtained from the slopes of least-squares fits in linear regions of \( \log_{10}(\bar{q}) \) vs. time plots for \( \chi_e \) and \( \eta \) ranging from zero to one. Evidently, the growth rate increases with both \( \chi_e \) and \( \eta \).

Figures 3.5 and 3.6 contain several features that we discuss next. First, exponential growth is only seen for sufficiently large \( \chi_e \) and \( \eta \): Fig. 3.6 suggests that there is an onset criterion for exponential growth. Below this onset, \( \bar{q} \) shrinks monotonically. Second, for most parameter values, exponential growth is preceded by a transient during which \( \bar{q} \) drops. Third, \( \bar{q} \) reaches an asymptotic value for long times. And fourth, Fig. 3.6 indicates an oscillatory “breathing” state that we will describe.

To understand the first two of these observations, we will define a mathematical model that captures the problem’s essential dynamics. This will...
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Figure 3.5: Time evolution of mean charge amplitude, $\bar{q}$, in log-linear plots. 
(a) $\chi_e$ is fixed and $\eta$ is varied. (b) $\eta$ is fixed and $\chi_e$ is varied.
3.2. RESULTS

Figure 3.6: Exponential growth rates from the slope of linear fits of $\log_{10}(\bar{q})$ vs. time plots for an array of parameters. Growth rates are defined as: $a = \frac{1}{t} \log_{10} \left( \frac{\bar{q}(t)}{b} \right)$, where $b$ is a constant. Colored spots correspond to colors used in panels (a) and (b) in Fig. 3.5, and breathing modes are discussed in text.
Figure 3.7: Spatiotemporal evolution of $\bar{q}$ and ratio between the magnitude of typical Coulomb and gravitational forces, $R_{cg}$. The ratio is given in cgs units by $R_{cg} = \frac{\bar{q}^2}{\bar{r}_{min}^2 \bar{m} g}$, where $\bar{r}_{min}$ is the mean of the distance to each grains nearest neighbor, $\bar{m}$ is the mean grain mass, and $g$ is gravity. (a) Spatiotemporal evolution of bed charges using $\chi_e = 0.6$ and $\eta = 1$. Inset shows the same plot over longer time, (to 14 sec.). (b) Time evolution of $\bar{q}$ and $R_{cg}$ (blue: averaged over the entire bed, green: averaged over the layers 14-16), using $\chi_e = 0.6$ and $\eta = 1$. Note that although $R_{cg}$ is only 20% when averaged over the entire bed, in the fastest charging region, around height = 15, $R_{cg} = 1$. Data in (b) is obtained by dividing the bed into one-mean-grain-diameter slices and calculating the sum of the absolute values of charges in each slice.
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Figure 3.8: Spatiotemporal evolution of $\bar{q}$ and ratio between the magnitude of typical Coulomb and gravitational forces, $R_{cg}$. (a) Spatiotemporal evolution of $\bar{q}$ using $\chi_e = \eta = 1$: again inset shows same plot over longer time, (to 14 sec.), highlighting periodic oscillations in charge. Notice that for $\chi_e = \eta = 1$, the entire bed becomes highly charged. (b) Here, $R_{cg}$ averaged over the entire bed exceeds one, at which point the charge starts to plummet.
involve some analysis, so we first discuss the simpler asymptotic and breathing states.

We begin with the asymptotic behavior, which provides insight into the bed’s charging dynamics. The origin of this behavior can be established by comparing the magnitudes of typical Coulomb and gravitational forces. In Figs. 3.7 and 3.8, we plot two representative cases, one with moderate charging, \( \chi_e = 0.6, \eta = 1 \) and one with rapid charging \( \chi_e = \eta = 1 \). Figures 3.7 (a) and 3.8 (a) show color-coded charge densities, and Figs. 3.8 (b) and 3.8 (b) show corresponding bed charges alongside the ratio \( R_{cg} \) between characteristic Coulomb and gravitational forces (defined in the figure caption).

From Fig. 3.7 (b), we see that for moderate charging, \( \bar{q} \) reaches a noisy asymptote after about 5 seconds, at which point \( R_{cg} \) averaged over the entire bed approaches 20%. Figure 3.7(a), by comparison, shows that the bed charge is dominated by grains in the middle of the bed: we measure that 70% of the charge is contained in the 14\textsuperscript{th} through 16\textsuperscript{th} layers. If we evaluate \( R_{cg} \) in these central layers that dominate bed charging, we find that the charge saturates when \( R_{cg} \) reaches one, as shown in Fig. 3.7(b). We conclude that the charging asymptote coincides with \( R_{cg} \) approaching one in the fastest charging region. At this point, grains levitate or stick together - either of which will prevent the collisional charging mechanism that we have described.

As for the breathing state identified in Fig. 3.6, the same behavior occurs, but throughout the entire bed. Figure 3.8 (a) shows that for \( \chi_e = \eta = 1 \), where breathing occurs, the highly charged region extends over most of the bed. In this case, Fig. 3.8 (b) shows that \( R_{cg} \) averaged over the entire bed reaches one so the whole bed must levitate or stick together. Indeed, Fig. 3.8 (b) shows that when \( R_{cg} \) exceeds one, charging stops, bed charge reduces, and simultaneously the bed contracts. This of course causes densities and collision rates to increase, which in turn must increase charging rates.

We confirm the link between charge oscillations and mechanical breathing in Fig. 3.9, where we compare power spectra of charge and bed expansion.
3.2. RESULTS

Figure 3.9: Power spectra of time series obtained by Discrete Fourier Transform. (a) Power spectrum of $\bar{q}(t)$; (b) power spectrum of $\Delta z(t)$. Main plots in (a) and (b) show breathing state: $\chi_e = \eta = 1$, and insets show the non-breathing state: $\chi_e = 0.6$, $\eta = 1$. Horizontal axes of power spectrum plots are identical.
in breathing ($\chi_e = \eta = 1$) and non-breathing ($\chi_e = 0.6$, $\eta = 1$) states. To evaluate bed oscillations, we average displacements of grain heights, $z_i(t)$ from the center of mass height, $z_c(t)$: $\Delta z(t) = \langle |z_c(t) - z_i(t)| \rangle$. As shown in the main plots of Figs. 3.9 (a)-(b), for the breathing state, both charges and average displacements of grains oscillate at the same frequency, while as shown in the insets, the non-breathing state exhibits broad spectrum noise, with no dominant frequency and much smaller peaks.

Therefore we propose that the cause of both asymptotic charge and breathing oscillations is that the region of the bed that dominates charging reaches $R_{cg} = 1$, at which point particles cannot collide and so cannot charge. For moderate charging, this occurs over a limited bed height that we presume cannot lift overlying particles; for strong charging, this occurs over the entire bed, which appears to cause global oscillations.

The breathing state indicates that there can be substantial temporal and spatial variations in charging: moreover our DEM simulations also suggest spatial variations in charge density, as shown in Figs. 3.10 (a)-(b) (left panels) for two values of $\chi_e$ and $\eta$. In these plots, to identify spatial patterns we enlarge our simulations to $4 \times 10^3$ grains on a horizontal domain of $20 \times 20$ mean grain diameters squared.

**Mathematical model**

To better understand these spatial variations as well as the transient charge growth, we provide a mathematical model for bed charging and discharging. The model is simplified but captures the essential physics that we have described so far. That physics consists of an iterative growth in polarization, defined by Eq. (3.3), combined with a neutralization in charge, Eq. (3.5).

Let us define functions $P(\vec{x}, t)$ and $C(\vec{x}, t)$ to represent the polarization in units of polarization per length and charge as functions of position, $\vec{x}$, and time, $t$:
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Figure 3.10: Comparison between DEM and Turing model. (a) Color coded (blue: negative, red: positive) charges in both models using DEM parameters $\chi_e = 1$, $\eta = 1$, and Turing parameters $A = 0.0005$ and $A - B = -0.0001$; DEM shows slice through center of simulation, and Turing results show height average (left) and 3D view (right) of charges. (b) The same views as panel (a), but using DEM parameters $\chi_e = 1$, $\eta = 0.6$, and Turing parameters $A = 0.0005$ and $A - B = -0.0005$. The spotted patterns here are typical, with wavelength that decreases as $A$ and $A - B$ grow.
Here, the rate of polarization grows iteratively with electric field in proportion to a single parameter, $A$. And the charge is reduced in proportion to another parameter, $B$. We have included diffusivity terms $D_p$ and $D_c$ to account for migration of the agitated grains. We anticipate that since polarized grains tend to align and attract one another $D_p$ should be smaller than $D_c$.

We acknowledge that this model is simplified in several respects: it represents polarization as a scalar, neglects particle motion, and Coulomb forces. Nevertheless, as we will show, Eq. (3.12) reproduces much of the charging dynamics in the bed shown in Figs. 3.5 and 3.6, it allows us to explain the charging transient that we have observed, it predicts conditions for the onset of exponential charging, and it permits us to analyze spatial variations. Moreover, Eq. (3.12) is nothing more than the linear reaction-diffusion equation - arguably the simplest of Turing models, whose features have been fully analyzed elsewhere [80].

We begin our discussion of this mathematical model by reiterating that the parameter $A$ codifies the exponential rate of growth of polarization, as can be seen in the first line of Eq. (3.12). Additionally, considering the equations together, we note that $-B$ causes a reduction in charge, so difference $A-B$ determines a net growth. Neutralization is a physical mechanism that produces charge transfer allowing particles to both gain or lose charge. The physical model, Fig. 3.2, relies on neutralization to permit charge transfer between particles, and the equivalent term in the Turing model, Eq. (3.12), that allows charge transfer is $A-B$: because when $A-B = 0$, neither $P$ nor $C$ grows. Accordingly, we associate $A$ with polarization growth - analogous to $\chi_e$ in our DEM simulations - and $A-B$ with neutralization - analogous to $\eta$.

This correspondence is not exact, but as shown in Fig. 3.12, plotting the
Figure 3.11: Time evolution of mean charge amplitude, $\bar{C}$, in log-linear plots from solutions of Eq. (3.12) obtained through a finite difference integration. As in Fig. 3.5, polarizations and charges of both signs grow similarly. (a) $A$ is fixed and $A-B$ is varied. (b) $A-B$ is fixed and $A$ is varied.
Figure 3.12: Exponential growth rates from the slope of a fit of linear regions of $\log_{10} \bar{C}$ vs. time plots for an array of parameters. Exponents are defined as: $a = \frac{1}{b} \log_{10} \left( \frac{C(t)}{k} \right)$, where $b$ is a constant. Colored spots correspond to colors used in panels (a) and (b) in Fig. 3.11, and violet region should not grow according to stability analysis of Eq. (3.12).
3.2. RESULTS

exponential growth rates of the charge amplitude averaged over all grains, $\bar{C}$, as a function of $A$-$B$ and $A$ produces substantially similar behavior to that seen in Fig. 3.6. Moreover, if we plot the growth in $\bar{C}$ as a function of time, we obtain panels Figs. 3.11 (a)-(b) for the values indicated. Again the behavior is similar to the growth seen in DEM simulations, Figs. 3.5 (a)-(b). Here we have evolved Eq. (3.12) using a finite difference model integrated with Euler’s method with time step 0.5 on a domain of $20 \times 20$ horizontal elements by 10 vertical elements. Top and bottom conditions are free and, as in our DEM simulations, horizontal boundary conditions are periodic.

We can go further with Eq. (3.12) and evaluate when exponential growth should be seen and determine the cause of the initial transient observed both in DEM and the Turing simulations.

It is known that the Turing instability (at which point patterned growth emerges) occurs for Eq. (3.12) when $A \cdot D_c - B \cdot D_p > 0$ [80] (see also Appendix in the end of this Chapter). In Figs. 3.11 and 3.12, we set $D_p = 1 \cdot 10^{-5}$ and $D_c = 4 \cdot 10^{-5}$, as we mentioned to mimic the fact that charged grains are expected to diffuse faster than polarized ones. So we expect the onset of growing patterns to occur when $A > B/4$. We indicate parameters for which this inequality fails - and so growth is not expected - as a violet shaded region in Fig. 3.12.

From stability analysis of this model, we predict that the onset of charging in the DEM simulations is associated with a simple Turing mechanism. The transient in charging is also amenable to analysis using Eq. (3.12). Our finite difference solutions in Fig. 3.12 are initialized with small random charges (uniformly distributed on $[-0.025,0.025]$), and since diffusion constants are $O(10^{-5})$, gradients that would trigger diffusion are negligible. If we remove the diffusive terms from Eq. (3.12), we obtain a pair of coupled ordinary differential equations (ODEs) with off-diagonal terms (i.e. $A \cdot C$ in the $\dot{P}$ equation, and $-B \cdot P$ in the $\dot{C}$ equation) that are of the same magnitude as the diagonal terms. This is a recipe for non-normal growth [81] in which transient contraction can occur in a system whose eigenvalues indicate growth, or vice versa.
CHAPTER 3. SELF-CHARGING OF IDENTICAL GRAINS IN THE ABSENCE OF AN EXTERNAL FIELD

Figure 3.13: Comparison between DEM and Turing model. Transient from DEM (blue) and Turing simulations (magenta); both ordinate axes are logarithmic; DEM parameters: $\chi^e = 0.9$, $\eta = 0.2$; Turing parameters: $A = 2.25$, $A - B = -4$ (with diffusive terms removed as described in text).

In the case of Eq. (3.12), non-normal growth occurs when random orientations of vectors ($P, C$) re-orient along the expanding eigendirection, approaching smaller values as they do so. This can be confirmed by taking a cluster of points, $(P_i, C_i)$ near the origin and evolving them according to Eq. (3.12) with diffusive terms removed. We use $A = 2.25$, $B = 6.25$ for convenience; other values behave similarly. This produces a characteristic transient reduction in $\sum_i \sqrt{P_i^2 + C_i^2}$ followed by growth, as shown in Fig. 3.13. Here we evolve 36 points in a grid with $P$ and $C$ between $-10^{-6}$ and $10^{-6}$, and plot $\sum_i \sqrt{P_i^2 + C_i^2}$ to display the transient decrease. The same is seen for either $\sum_i |P_i|$ or $\sum_i |C_i|$, separately. In Fig. 3.13, we compare this non-normal transient with the transient seen in DEM simulations for the example $\chi^e = 0.9$, $\eta = 0.2$.

Apparently the transients shown in Figs. 3.5 (a)-(b), and presumably as well in Figs. 3.11 (a)-(b), are associated with a simple mathematical behavior that occurs while small random vectors align with their ultimate eigendirections. Once alignment occurs, exponential growth can proceed, causing strong local
heterogeneities, and at this point, the approximation of neglecting diffusion can no longer be made.

To study these charge heterogeneities, we consider patterns produced both by DEM simulations and by the reaction diffusion model Eq. (3.12), summarized in Fig. 3.10. In Fig. 3.10, we show a top view of color-coded charges from a DEM simulation that resemble horizontal stripes, along with a comparable Turing pattern from Eq. (3.12). In all of these plots, red indicates positive charge and blue indicates negative. We emphasize, however, that the dominant patterns seen in solutions of the Turing model consist of irregular spots, as shown in Fig. 3.10 (b). These plots reveal that for aspect ratio 1:2 (i.e. width = 2·height), the charge patterns extend throughout the thickness of the bed.

This occurs for all parameter values shown in Fig. 3.12, although we note that this is a property that results from the assumption, $D_c > D_p$, which inhibits spatial growth of patterns. As with other reaction-diffusion models, much richer behaviors are also possible: for example relaxing the $D_c > D_p$ constraint leads to temporally oscillatory charges, strong vertical gradients in charge, and other states such as tubes, labyrinths, etc. [82]. Similarly, changing boundary conditions has a strong and well documented effect on the patterns expressed [83] - for example modeling a tall thin column of grains rather than a broader bed produces horizontal charge striations [59].

Our primary goal for presenting a simplified Turing model has been to show that Eq. (3.12) can reproduce charging seen in detailed DEM simulations, as evidenced by comparisons shown in Figs. 3.6, 3.10 and 3.12. The essential thing that the Turing model appears to reveal is that granular charging follows a stereotypical evolution due to straightforward mathematical causes, including a transient decrease in charge followed by exponential growth, a well-defined onset criterion for growth of charging, and emergence of patterned states. The robustness of the mathematics underlying these behaviors suggests that these findings should be common in practical problems, and calls for experimental verification in laboratory and field experiments.
Figure 3.14: Charging of vibrated hollow glass spheres under microgravity produced in the Bremen drop tower [84]. Gravity is about $10^{-6}$g and pressure $= 1$ mbar. Spheres have density $0.14$ g/cm$^3$ and diameters between 125 and 150 $\mu$m (Cospheric LLC, Santa Barbara, CA). (a) Typical time-lapse images showing 200 ms superpositions of video frames taken at 110 frames per second. At time $= 0$, vibration of the transducer shown is initiated, after which the apparatus is rapidly accelerated by catapult, and by several tens of milliseconds, microgravity is achieved. Gravity is nearly nonexistent, so parabolic trajectories can only be due to electrostatics.

**Experiment**

The underlying hypothesis of both DEM and Turing models is that an iterative process leads to exponentially rapid growth of initially infinitesimally small charges, so we close by testing this hypothesis in experiments. In these experiments, shown in Fig. 3.14, hollow glass spheres are vibrated on a grounded metal plate at 2 kHz by a piezoelectric transducer. The thickness of the particle bed is close to that used in our DEM simulations (under 1 mm, or about 9 particle diameters). These experiments are performed under microgravity (see figure caption), yet as shown in Fig. 3.14, particles return to the plate along curved trajectories. Crucially, since gravity is essentially absent, the only known force that can act at a distance in this way is electrostatic. Moreover, the heights of particle flights diminish with time as can be seen in Fig. 3.14, which we can use to obtain a quantitative evaluation of our DEM and Turing predictions, as follows.

The maximum height, $h$, of a particle ballistically ejected from the bed is simply its kinetic energy, $KE$, divided by the force, $F$, attracting the particle
Figure 3.15: Horizontal projections of grayscale of time-lapse images are used to evaluate height of bed of agitated particles. Inset shows enlargement just beyond noisy region associated with irregular large particle flights; intersections with dotted line used to estimate bed heights.
Figure 3.16: Estimates of bed height obtained from intersections shown in panel before, along with exponential fit. Inset shows second experiment where metal plate is covered by insulating tape.
to the bed: \( h \sim KE/F \). We have seen that our model predicts exponential growth in charges of both signs, so the force, \( F \), associated with these charges must also grow exponentially in time. Since \( h \sim 1/F \), we predict that \( h \) will decrease exponentially in time: \( h \sim e^{-a\cdot\text{time}} \) where \( a \) defines the charging rate shown in Figs. 3.6 and 3.12.

We assess this prediction by evaluating heights reached by particles near the top of the bed. As shown in Fig. 3.15, we horizontally sum the grayscales of pixels from successive 200 ms superpositions (as in Fig. 3.14). The transducer could drift away from the agitated particle bed and change the particle number density that would affect the brightness of the images. However, the timescales for particle losses qualitatively are on the order of 10s while the timescales for charging are a fraction of a second. Keeping these two different timescales in mind, we consider the brightness as a suitable measure of the charging.

Our procedure is as follows. High flying outlier particles produce noisy variations in grayscale, so we exclude the noisy region identified in Fig. 3.15, and select a moderate grayscale that shows little noise but provides the largest available height discrimination between superpositions. This grayscale is boxed in the main plot and enlarged in the inset to this figure. We evaluate the grayscale at the center of this region (broken line in the inset), which we plot in Fig. 3.16, along with a least-squares fit to the predicted exponential, \( h = h_0 + h_1e^{-a\cdot\text{time}} \). We find that a fit can be made using \( a = 1.31 \pm 0.03 \) sec\(^{-1}\) with correlation coefficient, \( r^2 = 0.997 \). We repeat the experiment with the metal plate covered with insulating tape, and obtain the height vs. time plot shown in the inset to Fig. 3.16: here we obtain \( a = 1.40 \pm 0.03 \) sec\(^{-1}\) and \( r^2 = 0.998 \). Both of these fits have growth rates, \( a \), in the range expected from Fig. 3.6.

We are aware that other dependencies can also be fitted. However, this would require an alternative competitive idea to be tested, which does not currently exist. Consequently we find that the experimental results are consistent with the theoretical prediction and we avoid drawing conclusions about the functional dependence just from the analysis of the experimental data.
These results seem to confirm our predictions of exponential charging, however other possibilities deserve mention.

First, most bouncing particles return to the bed, yet some particles near the edges escape. It might be argued that loss of particles could account for the decrease in bed height, however we note that fewer bed particles would cause particles above the bed both to be less strongly attracted to the bed and to rebound more elastically, both of which would increase, rather than decrease, the measured heights shown in Fig. 3.16.

Second, it is possible that particles have been tribocharged by the vibrating plate. Although tribocharging doubtless occurs, we remark that (1) particles ejected from the bed are attracted back to the bed, so particles cannot simply be tribocharged with the same sign, which would cause repulsion; (2) spheres landing on grounded metal and on insulating tape produce nearly indistinguishable results; and (3) charging appears to occur exponentially in time. None of these results are consistent with tribocharging as it is traditionally understood [9]. It remains conceivable that particles near and far from the vibrating plate could acquire opposite charges [56], however, this would not explain the apparent exponential charging.

Third, several groups have described charging models for particles differing in size [20, 21, 22], and indeed our hollow spheres range from 125 µm to 150 µm. Again, size-dependent charging doubtlessly occur, however such mechanisms invariably produce monotonically decreasing charging rates, rather than the exponential growth that we observe.

Based on these considerations, we conclude that our agitated granular bed appears to produce exponential growth in charges of both signs, which to our knowledge our model is unique in predicting.

3.3 Conclusion

We have performed simulations, modeling, and experiments of charged grains in an agitated bed. The simulations show that grains can charge exponentially rapidly by feeding back their electric fields through their neighbors.
3.3. CONCLUSION

The Turing model provides a simple framework to understand the exponential growth in polarization and charge as well as more detailed predictions such as an onset criterion and non-normal charging transients. The microgravity experiments confirm that charging of agitated beds of insulating grains does appear to grow exponentially. Finally, the simulations and the Turing model predict a previously unreported oscillatory state and complex spatiotemporal charging dynamics, both of which merit further study.

We propose that our findings of exponential growth of charging may account for the generation of multi-million volt potentials observed in nature, and may contribute to improved understanding of electrical charging in mining [15], and industrial powder handling [13, 18, 56].

Appendix

In this section, we derive the instability condition for our reaction-diffusion equations, Eq. (3.12). We start with our set of equations as:

\[
\begin{align*}
\frac{\partial P}{\partial t} &= f(P, C) + D_p \nabla^2 P, \\
\frac{\partial C}{\partial t} &= g(P, C) + D_c \nabla^2 C,
\end{align*}
\]

(3.12)

where we define: \( f(P, C) := A(P + C) \) and \( g(P, C) := -B(P + C) \). We assume, in the absence of the diffusive terms \( (D_p = D_c = 0) \), a spatially uniform and stable solution in a steady-state exists for Eq. (3.12):

\[ f(P_0, C_0) = g(P_0, C_0) = 0. \]

(3.13)

We write \( P \) and \( C \) using \( P_0 \) and \( C_0 \):

\[
\begin{align*}
P &= P_0 + \tilde{P}, \\
C &= C_0 + \tilde{C},
\end{align*}
\]

(3.14)

where \( \tilde{P} \) and \( \tilde{C} \) are small. We expand \( f(P, C) \) and \( g(P, C) \) around the fixed point \( (P_0, C_0) \) and obtain:
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\[ f(P, C) = f(P_0, C_0) + \tilde{P} \frac{\partial f(P_0, C_0)}{\partial P} + \tilde{C} \frac{\partial f(P_0, C_0)}{\partial C} + \cdots, \]

\[ g(P, C) = g(P_0, C_0) + \tilde{P} \frac{\partial g(P_0, C_0)}{\partial P} + \tilde{C} \frac{\partial g(P_0, C_0)}{\partial C} + \cdots, \]

(3.15)

with which we linearize Eq. (3.12):

\[ \frac{\partial \tilde{P}}{\partial t} = f_P \tilde{P} + f_C \tilde{C} + D_P \nabla^2 \tilde{P}, \]

\[ \frac{\partial \tilde{C}}{\partial t} = g_P \tilde{P} + g_C \tilde{C} + D_C \nabla^2 \tilde{C}, \]

(3.16)

where \( f_P := \frac{\partial f}{\partial P}, \ f_C := \frac{\partial f}{\partial C}, \ g_P := \frac{\partial g}{\partial P}, \ \text{and} \ g_C := \frac{\partial g}{\partial C}. \)

We rewrite the above equations using a matrix notation:

\[ \frac{\partial}{\partial t} \begin{pmatrix} \tilde{P} \\ \tilde{C} \end{pmatrix} = \left( D \nabla^2 + J \right) \begin{pmatrix} \tilde{P} \\ \tilde{C} \end{pmatrix}, \]

(3.17)

where we define:

\[ D := \begin{pmatrix} D_P & 0 \\ 0 & D_C \end{pmatrix}, \]

(3.18)

and:

\[ J := \begin{pmatrix} f_P & f_C \\ g_P & g_C \end{pmatrix}. \]

(3.19)

Turing instability occurs when a spatially uniform steady state solution (in the absence of diffusion) becomes unstable upon introduction of the diffusive terms \( (D_P \neq 0 \ \text{and} \ D_C \neq 0) \). Patterns are time independent and spatially heterogeneous solutions of Eq. (3.17) with non-zero diffusive terms.

Thus, first, without the diffusive terms:

\[ \frac{\partial}{\partial t} \begin{pmatrix} \tilde{P} \\ \tilde{C} \end{pmatrix} = \begin{pmatrix} f_P & f_C \\ g_P & g_C \end{pmatrix} \begin{pmatrix} \tilde{P} \\ \tilde{C} \end{pmatrix}, \]

(3.20)

must be stable, which defines conditions for trace, \( \tau \), and determinant, \( \Delta \):

\[ \tau = f_P + g_C < 0, \]

\[ \Delta = f_P g_C - g_P f_C > 0. \]

(3.21)
3.3. CONCLUSION

Next, we assume that the spatially heterogenous solution of Eq. (3.17) can be split and has the form:

\[
\tilde{P} = p(t)P^{i\mathbf{q}\mathbf{x}} \\
\tilde{C} = c(t)C^{i\mathbf{q}\mathbf{x}},
\]

where \( q \) is wave number. We plug these into Eq. (3.17) and obtain:

\[
\frac{\partial}{\partial t} \begin{pmatrix} \tilde{P} \\ \tilde{C} \end{pmatrix} = \begin{pmatrix} f_P - D_Pq^2 & f_C \\ g_P & g_C - D_Cq^2 \end{pmatrix} \begin{pmatrix} \tilde{P} \\ \tilde{C} \end{pmatrix}.
\]

Likely, the conditions for the above to be stable is:

\[
\tau' = f_P + g_C - q^2(D_P + D_C) < 0,
\]
\[
\Delta' = (f_P - D_Pq^2)(g_C - D_Cq^2) - f_Cg_P > 0.
\]

Notice that the first condition in Eq. (3.24) is already met in Eq. (3.21). Therefore, to have an unstable system, we need the second condition in Eq. (3.24) to be false. We define \( q_{\text{min}}^2 \) as \( q^2 \) that gives the smallest \( \Delta' \):

\[
q_{\text{min}}^2 = \frac{f_PD_C + g_CD_P}{2D_PD_C},
\]

which we plug back into Eq. (3.24) and obtain the condition for instability:

\[
f_PD_C + g_CD_P > 2\sqrt{D_PD_C(f_Pg_C - f_Cg_P)}.
\]

From the definitions of \( f \) and \( g \), we get:

\[
A \cdot D_C - B \cdot D_P > 0.
\]
Chapter 4

Segregation of Charged Particles in Shear Induced Diffusion

4.1 Introduction

Despite the prevalence and practical importance of particle segregation, comparatively little is known about the effects of particle charge under controlled circumstances. In this chapter, we study the segregation of mechanically identical but charged particles in a shear flow using particle-based simulation, with the goal of understanding the fundamental mechanics underlying segregation of charged particles. We use discrete-element method (DEM) [85] simulations and demonstrate that a granular flow with a binary mixture of similarly charged particles segregates under shear according to a precise formula.

The chapter is structured as the following: in section 4.2, we describe the simulation, then in section 4.3, we analyze results, and in section 4.4, we draw conclusions.
Figure 4.1: Snapshot of the discrete-element method (DEM) simulation. Red particles are electrically charged twice as strongly as blue particles with the same sign. Grey particles are electrically neutral and embedded into the plates. Arrows show the directions of the shear.
4.2. Model

We perform DEM simulations of spherical particles in three dimensions, as sketched in Fig. 4.1.

Grain size: The grain sizes are polydisperse to prevent crystallization: the radius of each grain is normally distributed around the mean radius, $\bar{R} = 0.5$ mm, with standard deviation 5%. We are interested in electrostatic effects between mechanically identical grains, so all grains have the same mass: $m = \frac{4}{3}\pi\bar{R}^3\rho_g$, where $\rho_g = 2.4$ g/cm$^3$ is the density of glass. We have confirmed, using our simulation, that the polydispersity that we introduce produces no segregation on its own.

Equations of motions: As in Chapter 2, we use the standard procedures to evaluate forces and torques on interacting particles. In short, for the elastic force, we use the model of Walton and Braun [63] with two elastic coefficients, $k_l = 0.07$ and $k_u = 0.08$, to achieve the restitution coefficient, $\sqrt{k_l/k_u} = 0.935$. For friction, we use the standard kinetic friction model with kinetic friction coefficient, $\mu_k = 0.4$. We compute Coulomb forces directly using a virtual charge fixed at the center of each grain. Long-range electrostatic forces between grains are computed using the Particle-Particle Particle-Mesh Method described in Chapter 2. Unlike prior works [73, 86], dipole moments are neglected, and we compute electrostatic forces only using grains’ net charges. The time step for the numerical integration is 50 $\mu$sec., which produces more than 100 steps per collision for the fastest moving grains. The boundaries in horizontal directions are periodic with respect to both mechanical and electrical forces.

Initialization: To obtain an initial configuration with a desired solid fraction, $\phi$, we place $10^3$ grains randomly in a three dimensional box with stiff boundaries. The top and bottom boundaries are $20\bar{R} \times 20\bar{R}$ in size, into which we embed neutral insulating grains of radius $\bar{R}$ on a fixed grid. We wait until the initial packing relaxes and grain velocities become negligibly small under gravity. We then turn off the gravity and compress the system vertically by lowering the top boundary with a constant slow speed. Once a desired solid fraction, $\phi$, is achieved, we stop the compression. Grains are
all electrically neutral during the initialization process and gravity is always
turned off thereafter.

Shear: Fixed boundary conditions are implemented at the top and bottom. We shear the packing by moving the top and bottom boundaries horizontally in opposite directions with a fixed speed, $v_s$ (in units of $2\bar{R}$/sec.). We apply shear until the system reaches a steady velocity field, then we distribute charges on all non-boundary particles with values randomly chosen to be either $+q$ or $+2q$, where $q \sim 1.8 \times 10$ pC. We set this time to be $t = 0$ in the results that follow. Note that the transition from an unjammed to a jammed state occurs at $\phi > 0.6$, below which the packing remains collisional even for a small shear velocity [87].

4.3 Results

Figure 4.2 (a) shows a snapshot of a simulation at $t = 500$ sec. for packing fraction $\phi = 0.55$ and shear velocity $v_s = 10$ (in units of $2\bar{R}$/sec.). Qualitatively, more strongly charged particles (red) gather closer to the shearing boundaries than more weakly charged particles (transparent blue): this is quantified in Fig. 4.2 (b), where for the simulation shown in Fig. 4.2 (a), we plot a histogram of the mean number of strongly (red) and weakly (blue) charged particles as a function of height (details in figure caption).

We evaluate the degree of segregation in our sheared bed using the segregation parameter, $S$, defined as:

$$S = \frac{1}{N} \sum_{i=1}^{N} \frac{(n_{i,e} - n_{i,d})^2}{(n_{i,e} + n_{i,d})^2},$$  \hspace{1cm} (4.1)

where $N$ is the total number of particles, $n_{i,e}$ is the number of like-charged particles around the $i^{th}$ particle within the cutoff radius, $r_{cut} = 3\bar{R}$, from the center of that particle, and $n_{i,d}$ is the number of differently-charged particles within the same area. For a perfectly segregated system, $S = 1$, because particles only have like-charged neighbors, and for a perfectly mixed case, $S = 0$, because the number of similar and dissimilar neighbors is equal.
Figure 4.2: (a) Snapshot of charged particles in sheared bed at $t = 500$ sec. for $\phi = 0.55$ and $v_s = 10$. Weakly charged particles (blue) are displayed transparently to demonstrate the separation of particles. (b) Histograms of the numbers of strongly charged particles (red) and weakly charged particles (blue) as a function of height for $\phi = 0.55$ and $v_s = 10$. We divide the system into 10 horizontal slices of thickness $2\bar{R}$ and count the number of red and blue particles every 50 seconds after the system reached an asymptotic steady state. We then average these measurements over 5 configurations and compute the ratio between red and blue particles.
Figure 4.3: Time evolution of the segregation parameters, $S$, with a fixed packing fraction, $\phi = 0.55$, and varying shear velocities, $v_s$. Each curve is an average over five independent simulations using the same parameters but different initial configurations. Black lines show the mean values, $S_\infty$, calculated after $t = 300$ sec., a time where we assume $S$ appears to have reached its asymptotic value.

Figure 4.3 shows the time evolution of $S$ for fixed $\phi = 0.55$ and several shear velocities $v_s$. In all cases, $S$ grows with time until it saturates at $S_\infty$, but $S_\infty$ depends non-monotonically on $v_s$: i.e. sheared beds segregate maximally at moderate shearing velocities. Similar non-monotonic behaviors are seen for different packing fractions, as shown in Fig. 4.4. Apparently segregation produced by dissimilar charges exhibits nontrivial dynamics, which we explore next.

To understand the segregation observed, we note that since all charges are of the same sign, the electrostatic energy of the particle bed is minimized when the most highly charged particles are as far away from other charges.
Figure 4.4: Phase diagram showing $S_\infty$ for different $\phi$ and $v_s$. Red spots correspond to the maximum $S_\infty$ values for a given $\phi$. For $\phi > 0.6$, the bed attains a nearly jammed state, and segregation does not occur.
Figure 4.5: Cartoon showing the initial fully mixed configuration and two configurations with the lowest electrostatic potential energy of a one-dimensional chain of ten charged particles. Red particles are charged $+2q$ and blue particles $+q$, and the separation distance between particles is fixed.
4.3. RESULTS

Figure 4.6: Time evolution of $S$ (red) and total electrostatic potential energy, $E$ (green), defined as: $E = \frac{k}{2} \sum_{i \neq j}^{N} \frac{q_i q_j}{r_{ij}}$, where $r_{ij}$ is the distance between the particles $i$ and $j$, for $\phi = 0.55$ and $v_s = 10$. $E$ is normalized by its initial value, $E_{ini}$. Inset shows the same measurements for $\phi = 0.55$ and $v_s = 60$, where $S$ does not grow much.
Figure 4.7: $|\Delta E|$ (blue) and $S_\infty$ (red) versus $v_s$ for $\phi = 0.55$. $|\Delta E|$ is the absolute difference in $E$ between the initial and the final configurations: $|\Delta E| = |E_{ini} - E_\infty|$, where $E_\infty$ is $E$ in an asymptotic steady state. Grey curve to aid the eye is the same shown in Fig. 4.9, rescaled.
as possible. This is a simple consequence of the fact that the self-energy of a charge density, $\rho(\vec{x})$, is:

$$E = \frac{ke}{2} \int \int \frac{\rho(\vec{x})\rho(\vec{x}')}{|\vec{x} - \vec{x}'|} dx^3 dx'^3$$

[88], where $k_e$ is Coulomb’s constant, hence locating the largest charges along boundaries, beside which $\rho(\vec{x}) = 0$, minimizes the energy. In our simulations, the boundary particles are insulating, however, we remark that for conducting boundaries, image charges would be of opposite sign and so there would be even stronger energetic advantage when locating the highest charges near the boundaries [19]. To study if such spatial arrangement of particles actually minimizes the electrostatic potential energy of the system, we construct a simple model consisting of ten charged particles forming a one-dimensional chain. Half of the particles are strongly charged and the other half are weakly charged like in our simulations. Depending on the ordering of the particles, the chain takes different electrostatic potential energies, which we compute as:

$$E = \frac{ke}{2} \sum_{i \neq j}^10 q_i q_j/r_{ij},$$

where $q_i$ and $q_j$ are the charges of the $i^{th}$ and $j^{th}$ grains, and $r_{ij}$ is the distance between the centers of the two grains. A comparison of energies of different configurations shows that the most favorable configurations have strongly charged particles right next to boundaries and more weakly charged particles in the center as shown in Fig. 4.5. Notice that the model does not exhibit a perfect de-mixing of strongly and weakly charged particles, since the lowest energy configurations also contain some strongly charged particles in the center.

To examine if the same argument holds for our sheared system, we fix $\phi = 0.55$ and measure the time evolution of the total electrostatic potential energy computed the same way. As shown in Fig. 4.6, $E$ decreases as $S$ grows for a system that strongly segregates with $v_s = 10$ (main plot), and stays almost unchanged for a system that does not segregate with $v_s = 60$ (inset). Furthermore, the drop in the electrostatic potential energy:

$$|\Delta E| = |E_{ini} - E_{\infty}|$$

where $E_{ini}$ is the energy at $t = 0$ and $E_{\infty}$ is the energy in an asymptotic steady state, behaves similarly to $S_{\infty}$. This is shown in Fig.4.7, where we plot $|\Delta E|$ as a function of $v_s$ for $\phi = 0.55$. Note that for $\phi = 0.55$, $S_{\infty}$ takes its maximum at $v_s = 10$, in agreement with Fig.4.4, which shows that the maximally segregated system has the lowest electrostatic potential energy.
We have measured the mean inter-particle distance: \( <r_d> = \frac{1}{N} \sum_{i=1}^{N} r_{ik} \), where \( r_{ik} \) is the distance between the centers of particle \( i \) and its closest neighbor particle \( k \), and we confirm that this remains unchanged for an electrically neutral and a charged system. This implies that the packing does not expand due to charging and so the cause of the drop in the electrostatic potential energy is the rearrangement -i.e. the segregation- of particles. Moreover, we have also employed Lees-Edwards boundary conditions [89], which connect the lower and the upper boundaries via a periodic boundary. In this case, the energetic advantage of placing highly charged particles near the boundaries is removed, and segregation does not occur.

Next, we discuss the non-monotonic dependency of \( S_\infty \) on \( v_s \). Figure 4.8 (a) shows \( S_\infty \) versus \( v_s \) for several \( \phi \). Each curve has its distinct maximum \( S_\infty \) and optimal \( v_s \), a shear velocity where \( S_\infty \) maximizes. Both the maximum \( S_\infty \) and the optimal \( v_s \) decrease with \( \phi \). To further investigate the relationship between the packing fraction and the segregation, we note that the maximum \( S_\infty \) and the optimal \( v_s \) vary nearly linearly with \( \phi \), as shown in Fig. 4.8 (b). Consequently, we scale \( S_\infty \) and \( v_s \) using simple linear functions, \( \phi: \alpha \phi + \beta \), where \( \alpha \) and \( \beta \) are obtained from least-squares linear fits of the maximum \( S_\infty \) vs. \( \phi \) (for the vertical axis), and the optimal \( v_s \) vs. \( \phi \) (for the horizontal axis). After the scaling, curves collapse onto a single curve with a peak close to unity, in keeping with the observation that the maximum segregation and the optimal shear velocity both decrease linearly with the packing fraction, as shown in Fig. 4.9. We also did simulations of two dimensional systems. Figure 4.10 shows the scaled \( S_\infty \) vs. scaled \( v_s \) from the results obtained with two dimensional DEM simulations, where we have also observed a similar spatial segregation. Note that the two dimensional result is not trivial for it is not always obvious that particles produce the same segregation in two and three dimensions [52].

We propose that the maximum segregation results from a competition between mixing and segregational fluxes. At low shear, particles rearrange in response to electrostatic repulsion, whereas at high shear these rearrangements become overwhelmed by diffusional mixing [90, 91]. This is essentially
4.3. RESULTS

Figure 4.8: (a) $S_\infty$ versus $v_s$ for several choices of $\phi$. Error bars indicate the standard deviation (over 20 trials). (b) Optimal $v_s$ versus $\phi$ and a linear least-squares fit. Inset shows maximum $S_\infty$ versus $\phi$ with a linear fit obtained in the same way.
Figure 4.9: $S_\infty$ versus $v_s$, scaled using linear functions of $\phi$: $\alpha \phi + \beta$, where for the vertical axis, $(\alpha, \beta) = (-0.4, 0.28)$, and for the horizontal axis, $(\alpha, \beta) = (-170, 107)$. Black curve is a fit to Eq. (4.3).
Figure 4.10: Scaled $S_\infty$ versus scaled $v_s$ from two dimensional DEM simulation. The axes are scaled by linear functions as before, where for the vertical axis $(\alpha, \beta) = (-0.53, 0.45)$ and for the horizontal axis $(\alpha, \beta) = (-360, 300)$. Again the black curve is a fit to Eq. (4.3).
Figure 4.11: $S_\infty$ versus $v_s$ for fixed $\phi = 0.56$ and different charge magnitudes: (strong : weak) = (1:0.5) for red, (2:1) for green, and (3:1.5) for blue.
Figure 4.12: Optimal $v_s$ versus charge magnitudes for several $\phi$. 
Figure 4.13: Kinetic energy: $E_k = \sum_{i=1}^{N} \frac{1}{2} m_i v_z^2$, versus $v_s$ From three dimensional simulations for $\phi = 0.55$ (red dots). The black curve is a fit to a quadratic function with correlation coefficient $R^2 = 0.92$. Inset shows diffusion coefficient: $D_z = \langle (z_i(t) - z_i(0))^2 \rangle / t$, versus $v_s$ (red dots) also measured in the DEM simulations fitted using a linear function (black line) for $\phi = 0.55$. 

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the same competition historically analyzed by Einstein & Sutherland [92, 93].

Testing this hypothesis is straightforward. We carry out simulations with
different charge magnitudes while keeping the ratio between strongly and
weakly charged particles unchanged. Increasing the charge magnitudes strength-
ens the electrostatic repulsion and higher shear is needed to mix particles
that rearrange in response to the stronger electrostatic repulsion. Conse-
quently, if the proposed mechanism is correct, increasing the charge mag-
nitude should increase the optimal \( v_s \) and vice versa for weaker charges.
This is shown in Fig. 4.11, where \( S_\infty \) is plotted against \( v_s \) for different mag-
nitudes of charges. The optimal \( v_s \) is increased for a system with higher
charges. The same tendency is seen for different packing fractions as shown
in Fig. 4.12. Evidently the maximum segregation results from a competition
between Coulomb flux and diffusional flux.

In kinetic theory, the diffusion coefficient is expressed as:

\[
D_z = \mu k_B T, \tag{4.2}
\]

where \( D_z \) is the spanwise diffusivity, \( \mu \) is the mobility, \( k_B \) is Boltzmann’s
constant, and \( T \) is the (granular) temperature, which in our system is the
kinetic energy: \( T = E_k = \sum_{i=1}^{N} \frac{1}{2} m_i v_i^2 \). Notice that we are only interested
in the perpendicular motions of particles since the segregation we want to
understand is perpendicular to the shearing direction. \( \mu \) is a measure of
particles’ motion through a medium in response to an electric field, and so we
use \( \mu \) as a surrogate for the segregation for our system: \( S \sim \mu = D_z/(k_B T) \).
The diffusion coefficient is known to grow linearly with the applied shear
rate, which in our case is \( v_s \) [94] (also see the inset of Fig. 4.13, where the
diffusion coefficient is obtained by: \( D_z = \langle (z_i(t)-z_i(0))^2 \rangle /t \), hence \( D_z \sim v_s \). For \( T \), we find that a suitable fit can be made using a quadratic function
with correlation coefficient \( R^2 = 0.92 \), as shown in the main plot in Fig. 4.13,
and so \( T \sim av_s^2 + bv_s + c \). Consequently, we use a function of the following
form to fit the curves in Fig. 4.9:

\[
S \sim \frac{v_s}{av_s^2 + bv_s + c}. \tag{4.3}
\]
The function produces the fit shown in Fig. 4.9 with correlation coefficient $R^2 = 0.98$ for $\phi = 0.55$. This simple model predicts a maximum as seen in the simulations, but underestimates the decrease in segregation after the maximum. There certainly is a need of additional work to refine our understanding. Evidently from our simple simulations, we find that there is a maximum of segregation as shear grows, and it appears that the competition between the mixing and segregational fluxes can largely account for this maximum.

4.4 Conclusion

We found that a binary mixture of similarly charged particles segregate according to their net charge in a simple shear flow. This segregation occurs due to strongly charged particles repelling others and moving outwards to minimize the electrostatic potential energy of the system since the boundaries are insulating. However, we expect the same to happen for conducting boundaries. Our direct simulations showed that for a given packing fraction there is an optimal shear velocity where the segregation maximally intensifies and for a larger shear velocity, diffusion suppresses the segregation.
Chapter 5

Conclusion

We first studied the charging of a fluidized granular bed in the presence of a pre-existing field. Here, we used a refined model which captures important physics that the previous model neglected and found that the essential mechanism of inductive charging continues to work: e.g. colliding polarized grains transfer net charges along the direction of the external field. However, we found that strong charge heterogeneities can be established within the bed due to electrostatic interactions, and we further predict that shielding of an external field due to the heterogeneities could dramatically suppress the charging rate in the bed.

We then investigated the charging of an agitated bed of identical grains in the absence of an external field. The simulations showed that grains can charge exponentially rapidly by feeding back their electric fields through their neighbors: that is an initially polarized grains polarize neighboring grains which, in turn, feeds back to the polarization of the first one. The mathematical model provided a simple framework to understand the exponential growth as well as more detailed predictions such as an onset criterion and initial transient of the charging. The experiments carried out in microgravity confirmed that charging of agitated beds of insulating grains does appear to grow exponentially. We presented our prediction of a previously unreported oscillatory state and complex spatiotemporal charging dynamics.

Finally, we studied segregation of charged particles in a shear flow. We used particle-based simulations and found that a binary mixture of mechanically
identical but differently charged particles segregate according to their net charge under shear. This segregation occurs due to strongly charged particles repelling others and moving outwards to minimize the electrostatic potential energy of the system. For a given packing fraction, there is an optimal shear velocity, at which the segregation intensity maximizes. We showed that this maximum results from a competition between diffusional and Coulomb fluxes. For a larger shear velocity, diffusion suppresses the segregation.

Overall, we studied electrostatic interactions of charged and polarized grains of the same size, shape, and material. We developed theoretical models and methods to study the charging and dynamics of interacting particles. Our simulations consisting of thousands of particles revealed novel collective effects which have never been studied before. Some of these predictions were confirmed by experiments, but others await experimental verifications. We expect that future studies into these predictions may provide us with more fundamental understandings of granular dynamics and charging.
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