Towards Understanding Powder Behavior Via Simulation

by

Simo-Matti Siiriä

ACADEMIC DISSERTATION

To be presented, with the permission of the Faculty of Pharmacy of the University of Helsinki, for public criticism in auditorium 1 at Viikki Infocentre (Viikinkaari 11) on September 24th 2011, at 12 noon.

Helsinki 2011
Supervisors: Professor Jouko Yliruusi  
Division of Pharmaceutical Technology  
University of Helsinki  
Finland

Reviewers: Professor Veli Pekka Tanninen  
Orion Pharma  
Finland

Professor Ville Alopaeus  
Department of Chemical Technology  
Aalto University  
Finland

Opponent: Professor Jukka Rantanen  
Faculty of Pharmaceutical Sciences  
University of Copenhagen  
Copenhagen

©Simo-Matti Siiriä 2011  
ISBN 978-952-10-7156-0 (paperback)  
ISBN 978-952-10-7157-7 (pdf)  
ISSN 1799-7372

Unigrafia  
Helsinki 2011  
Finland
Acknowledgements

This study was performed at the Division of Pharmaceutical Technology, Faculty of Pharmacy, University of Helsinki, during the years 2005–2011.

First and foremost, I wish to express my sincere thanks to my supervisor, Professor Jouko Yliruusi. Without his support and faith in this project, it would probably never have been begun, let alone completed. Your support has helped me to grow, both as a scientist, and as a person.

Secondly, I wish to thank Professor Veli Pekka Tanninen and Professor Ville Alopaeus for reviewing this thesis and giving last-minute valuable suggestions for improving it. I also wish to thank Doctor Clare Strachan for proof reading my first article despite the terrible hurry I had at the time and Kalle Ojala for helping with several articles and other texts later on. I also wish to thank Jouko Yliruusi, Osmo Antikainen and Jyrki Heinämäki for their work as my coauthors in this work.

I am very grateful for the opportunity I have had to share my workroom with Osmo Antikainen and Satu Lakio. You have been one of the key factors in making our division for me a place I have been happy to spend my time, day after day, year after year. While our conversations may not always have been purely work-related, they have made the work itself much more enjoyable. At the same time, I wish to thank Natalja Genina for brightening the work environment and introducing me into a new hobby.

I thank our division secretary, Inkeri Eskola, for the numerous times she has helped me to find and figure out whatever number or paper was needed for various reasons, and generally keeping the division actually working.

I also wish to thank all the past and present coworkers at the Division of Pharmaceutical Technology. You are too numerous to mention all here, but you have created a very special and enjoyable working environment, a place where it was fun to work, and which I have felt and will always feel as another home.

Lastly, I wish to thank my dear parents, Heikki Siiriä and Pirkko Siiriä, who have always supported me in everything and encouraged me to find and reach for my dreams. Kiitoksiini teille, Isä ja Äiti, ei ole sanoja.

Helsinki, 2011
Simo Siiriä
Towards Understanding Powder Behavior Via Simulation

Simo-Matti Siiriä

The aim of this study was to investigate powder and tablet behavior at the level of mechanical interactions between single particles. Various aspects of powder packing, mixing, compression, and bond formation were examined with the aid of computer simulations. The packing and mixing simulations were based on spring forces interacting between particles. Packing and breakage simulations included systems in which permanent bonds were formed and broken between particles, based on their interaction strengths. During the process, a new simulation environment based on Newtonian mechanics and elementary interactions between the particles was created, and a new method for evaluating mixing was developed.

Powder behavior is a complicated process, and many of its aspects are still unclear. Powders as a whole exhibit some aspects of solids and others of liquids. Therefore, their physics is far from clear. However, using relatively simple models based on particle-particle interaction, many powder properties could be replicated during this work. Simulated packing densities were similar to values reported in the literature. The method developed for describing powder mixing correlated well with previous methods. The new method can be applied to determine mixing in completely homogeneous materials, without dividing them into different components. As such, it can describe the efficiency of the mixing method, regardless of the powder’s initial setup. The mixing efficiency at different vibrations was examined, and we found that certain combinations of amplitude, direction, and frequencies resulted in better mixing while using less energy. Simulations using exponential force potentials between particles were able to explain the elementary compression behavior of tablets, and create force distributions that were similar to the pressure distributions reported in the literature. Tablet-breaking simulations resulted in breaking strengths that were similar to measured tablet breaking strengths.

In general, many aspects of powder behavior can be explained with mechanical interactions at the particle level, and single particle properties can be reliably linked to powder behavior with accurate simulations.
Contents

Acknowledgements .......................................................... iii

Abstract ............................................................................. iv

Table of contents ............................................................. v

List of original publications ................................................ vii

1 Introduction ...................................................................... 1

2 Theory and literature review ............................................. 2
  2.1 Powder simulations ....................................................... 2
    2.1.1 Finite element models ............................................. 2
    2.1.2 Discrete element models ....................................... 3
    2.1.3 Joined models ....................................................... 5
  2.2 Powder packing ............................................................ 5
  2.3 Powder mixing ............................................................. 6
  2.4 Determining mixing ...................................................... 7
  2.5 Tablet compression ....................................................... 7
  2.6 Tablet crushing ........................................................... 8

3 Aims of the study ............................................................ 9

4 Simulation environment .................................................... 11
  4.1 Elements of the software .............................................. 11
  4.2 Forces included .......................................................... 11
    4.2.1 Overview .......................................................... 11
    4.2.2 Spring forces ....................................................... 12
    4.2.3 Exponential forces ............................................... 12
    4.2.4 Bond forces ....................................................... 13
  4.3 Time ........................................................................... 13
  4.4 Energy within the simulations ........................................ 15
    4.4.1 Potential energy .................................................... 16
    4.4.2 Kinetic energy ...................................................... 17
    4.4.3 Rotational energy .................................................. 17
    4.4.4 Spring energy ...................................................... 17
    4.4.5 Lost energy ........................................................ 18
  4.5 Particles ..................................................................... 18
  4.6 Elementary testing of the system ..................................... 18
  4.7 Mixing degree ............................................................. 21
  4.8 Tablet breaking ........................................................... 25
List of original publications


1 Introduction

The great majority of active drug substances and excipients are dry crystalline materials and are used as dry powders. As such, powder behavior in various instances, such as flow, packing, mixing, segregation, and compression, plays a vital role in drug processing. Many aspects of these phenomena are still largely unknown. Already, such aspects as when the bulk of the powder can be said to be well mixed and which attributes of the powder affect this and how much, are far from clear. The same can be said for segregation. It is generally known that large grains tend to rise above smaller ones during vibration. Then again, Li and McCarty showed in their study [1] that the amount of moisture can cause segregated mixtures to mix, and vice versa.

Powder behavior has been studied widely, both with simulations and experiments, but is still far from being explained. While the grains forming the powder are certainly solid, the powder itself exhibits several characteristics that are closer to a liquid. Pharmaceutical studies have often classified materials as plastically deforming and fragmented materials [2, 3, 4, 5]. This classification, however, is rather problematic, since ideally fragmenting materials, such as sodium chloride, are often described as plastic.

One of the reasons why powder behavior still poses many challenges for researchers is that it is extremely challenging to obtain detailed information inside the actual processes. Determining mixing completely, for example, would require representative samples from the powder, but how to acquire them without disrupting the process? Simulations are one solution to this problem. If we can simulate a studied phenomenon accurately enough so that it reproduced the observed effect, we have a way of gaining insight into the functioning of the process. A simulation once completed can be studied grain by grain without affecting the process. Every detail of the process can be acquired in great detail, which is always a challenge in a laboratory experiment.

In pharmaceutics, simulations can often refer to machines built to emulate several different types of tableting machines. Here, simulation always means a computer simulation, an attempt to model a real-life situation on a computer. When referring to mixing, it should always be put in the context of powder mixing. While there are various other mixing processes, such as liquid mixing, these are not considered here.
2 Theory and literature review

2.1 Powder simulations

Despite the rapid development of computer calculation capacity, powder particle simulations are still very time-consuming. It is important to be able to simulate large numbers of particles. Much has been done to develop faster computational algorithms. For example, in 2004 Sobolev and Amirjanov [6] described a very fast algorithm that can be used to simulate the dense packing of large assemblies of particulate spherical materials that may contain millions of particles, and Fu and Dekelbab [7] used a two-stage kinematics and dynamics simulation method for random packing of particles with variable sizes in three dimensions. Later Han et al. [8] demonstrated a very fast simulation algorithm for sphere packing. Using a normal personal computer, the packing of 27,000 particles in a cylindrical domain can be made in 3 min. Mueller [9] also developed a new method to study the numerical packing of spheres in cylinders.

It is not realistically possible to solve analytically any problem involving more than \( n \) bodies [10] when \( n \) is more than 3. Even for \( n = 3 \), only some special cases can be solved. For this reason, every simulation system involving powders must make some kind of approximations of the interactions. Simulations used in powder modeling can be roughly divided into two different approaches: finite element models (FEMs) and discrete element models (DEMs).

Developments in particle simulations are well described by Zhu et al. in an interesting two-part review [11, 12]. W. R. Ketterhagen [13] published an excellent review of DEM usage in the pharmaceutical industry, covering the areas of transport, blending, granulation, milling, compression, and film coating. Typical applications and approaches for simulations concerning pharmaceutical material science have been presented by Wassgren et al. [14].

2.1.1 Finite element models

FEM methods are used when the material studied is approximated as a flowing mass, rather than singular granules. The behavior of the mass is formulated in the form of differential equations describing the parameters studied. These equations are usually impossible to solve in open form, and thus the FEM is used to achieve a numerical solution. The core of a FEM is the grid that divides the simulated area into smaller parts, in which numerical solutions for the equations can be found. The grid generated is generally called a mesh (figure 1).

O. C. Zienkiewicz published an interesting review article on the state and history of FEM methods in 2000 [16]. From this, it was shown that the idea of a FEM dates as far back as 1910, although the first actual models could be said to have been developed during World War II.
Figure 1: Example of a FEM grid. Image shows a hopper and the borders of the FEM grid used by Kamath and Puri [15] to model the powder mass in it. Image modified from [15]. This FEM grid comprises 84 powder elements and one powder-wall interface element.

Software for powder simulations with FEMs is available. In their work, Rombarch et al. compared commercial and research software for simulating granular flow in silos [17].

FEMs have also been used to estimate the changes in compacted mass [18, 19, 20, 21, 22, 23]. However, due to their nature they cannot model interparticulate bond formation.

One advantage of a FEM in comparison to a DEM, is its capacity to handle large amounts of powder. The requirement for computing power is more dependent on the complexity of the mesh than the actual amount of material. This makes it possible to develop large-scale simulations. Since the simulation does not include single particles, it cannot give much information on how changes in the properties of the grains would change the flow properties of the powder. The FEM method can, however, be used to study the theories of those flow properties.

2.1.2 Discrete element models

In a DEM, the powder is modeled one grain at a time. The rules that determine the simulation are based on the interactions between each grain. One of the biggest challenges to this approach is that since each particle and each interaction between particles must be considered separately, the required computational power is high. Then again, the model itself does not require input of the powder behavior as a whole. Rather, the powder behavior will follow from the inter particle interactions. DEMs still can be divided roughly into two further categories: event-driven (ED) models and molecular dynamics (MD) models.
Molecular dynamics models  MD models were originally used for simulating inter-
molecular behavior. Some of the first occurrences of the concept are Alder and Wain-
wright’s work in 1959 [24], which describes a general method for using computers to
simulate interactions of several hundreds of molecules, and Rahman’s work in 1964 [25],
in which he studied the properties of liquid argon with a model of 864 particles.

In these models the interactions between particles or atoms are considered continuous
and soft. Collision based on such interactions takes a given amount of time and is
calculated during several time steps. This causes the MD approach to require much more
computing power for each collision than ED models, which calculate each collision in
one step. Furthermore, it has been reported that MD models experience difficulties with
the so-called detachment effect, in which the energy dissipation between densely packed
particles can be underestimated [26]. MD models with low force constants and thus high
penetration distance with long contact times can cause unrealistic braking of particles
[27].

The time in an MD DEM is divided into steps of certain lengths (which may or may
not vary, depending on the model). The interaction forces between each body within the
simulation are calculated and applied to each body during a time step. Since the changes
in the interaction forces are dependent largely on the places of the bodies, the end result
is not accurate. The longer the steps are, the more inaccurate the simulation will be. In
contrast, the shorter the time steps, the longer time the simulation will take.

The purpose of this study was to determine the effects of particle properties on powder
behavior. This requires simulation of single particles in tightly packed situations. This
made the MD-based DEM our choice of approach for the simulations.

Event-driven models  ED models consider each collision as an instantaneous event.
The particles in such a simulation are considered to be hard. The simulation advances
by moving the particles in their given routes until two of them collide. Then, new routes
are calculated and this is continued until the next collision, and so on. This method
makes calculation of single collisions much faster than with MD models. However, when
the collisions become more frequent (as they do when simulating powder packing, for
example), the interruptions in the simulations become increasingly frequent. Ultimately
this will lead to a problem called inelastic collapse [28, 29, 30]. In its simplest form, this is
caused when one particle is between two that approach each other. The collisions between
the particles keep occurring more and more frequently, until theoretically there would be
an infinite number of collisions before all three particles are connected. This, naturally,
would cause the simulation to freeze, since it cannot calculate an infinite amount of
collisions.

There are some studies in which ED models have been used with granular material.
For example, Esipov and Pöschel [31] studied how the granular temperature allows the
particles to move and used the areas where inelastic collapses occurred as a sign of a
solid area. Alam and Luding [32] determined the rheology of binary mixtures via the ED model and found good agreement with kinetic theory.

2.1.3 Joined models

FEMs and DEMs have completely different approaches to the simulation, although they both have their strengths and weaknesses. There are some works that have combined these two approaches. For example, Ransing et al. [33] simulated the compaction of metal powder, using a model that handled the insides of particles as a FEM, and the actual particles as a DEM. In 2005, R. W. Lewis et al. published a work [34] in which similar types of FEM-DEM was applied in pharmaceutical powder compression. Munjiza et al. studied fracturing, in which the finite element representation of the solid region was combined with progressive fracturing [35]. In this model, the fracturing leads to the formation of separate areas, that are handled with a DEM.

Usually, FEMs are better suited for simulations of clearly continuous cases, such as liquid, gas, or internal strains on a physical object. DEMs are, on the other hand more appropriate in situations where interacting objects are more defined, such as in molecular dynamics or cases with definite numbers of bodies. The fragmentation simulations of F. Kun et al.[36] are examples of such applications of a DEM.

2.2 Powder packing

The value of powder packing simulations is clear. Numerous promising simulation results have been described in the literature. The complex nature of particle systems is clearly seen in the formation of granule piles. Fortunately, it is possible to obtain valuable information on the packing of heaps by simulations, as described by Matuttis et al. [37].

Methods for evaluating powder packing have also been developed. For example Yu et al. [38] presented a way to estimate powder packing density, based on the initial porosity and a derived packing size ratio.

Cunningham, Sinka, et al. published two interesting papers on their FEM simulations of powder compaction. In the first [39], they used the model to determine the mechanical properties of microcrystalline cellulose. In the second [18], they studied the effect of lubrication on walls and the relative density inside the compaction. Landry et. al. [40] used discrete element simulations of stress distributions in silos. They compared 2D and 3D packing of particles.

Chuan-Yu Wu studied the die-filling process with a 2D DEM [22]. Nolan and Kavanagh [41] formed various particle shapes from assemblies of component spheres. They modeled the packing of cylindrical, beamlike and nailike particles and concluded that the simulated packing densities were similar with the experimental values. Cheng et al. [42] showed that the influence of van der Waals interaction was crucial in the packing of spherical particles, using a 2D distinct element method simulation. Silbert et al. [43]
studied frictionless and frictional packing of spheres in three dimensions, using monodisperse particles and molecular dynamics simulations. The packing of wet particles was also modeled with a DEM [44].

Z. Lu, C. Negi, and J. C. Jofriet simulated a granular-flow model of a silo in their work [45, 46, 47]. Here, a silo is modeled mainly with a FEM, but areas near the hopper and near the intersection of the vertical part of the silo with a DEM.

2.3 Powder mixing

Mixing has been widely studied experimentally. For example, Brigdwater [48] published an interesting review considering the understanding of rotary drums and mixing in them. In addition, Edwards and Gine [49] studied the packing and jamming of powder and proposed a mathematical method to approximate powder behavior with a few key parameters. Ding et al. [50] examined tumbling mills and developed a method to estimate the turnover time of the granular bed. Various blenders have also been studied widely with DEMs, e.g Lemieux et al. performed research on V-blenders [51, 52] and Muguruma et al. on a rotating mixer with baffles [53].

Xu et al. [54] performed both simulated and experimental studies of mixing and segregation in a rotating drum. A vibrated granular bed was simulated [55, 56] and the convections and ratio of the convective flow to the diffusive flow were monitored. Particle mixing in a rotating fluidized bed was studied with simulations [57], while the kinetics and mixing of the particles were simulated in a rotary dryer[58].

A very interesting paper by Abreu et al. [59] describes the vibration-induced segregation of binary mixtures, using different particle dimensions and volumes. Powder mixing was studied with DEMs [60, 61] and a DEM was applied for studying the effect of particle shapes on granular system behavior [62]. Particle movement in spouted beds [63] was also studied with DEMs, as were the viscoelastic and plastic behaviors of particles [64], granular convections in wall-induced cases [65] and mixing in tumbling mills [66, 67, 68, 69].

One approach for modeling a fluidized bed containing fine particles was made by S. Limtrakul et al. [70]. Particles were simulated with a DEM, and the forces caused by the flow were calculated, using equations of momentum. In their recent work, K. W. Chu and A. B. Yu simulated [71] a particle-fluid flow with a combination of DEM-like and FEM-like systems called the combined continuum and discrete method (CCDM).

Rhodes et al. [72, 73] used FEM methods to handle air viscosities and DEM methods for the granular material. In another example [74], the magnetic field around particles was modeled with a FEM, while the particles themselves were modeled with a DEM.
2.4 Determining mixing

Mixing can be performed with several different methods. These include shaking, rotating, and screw mixing. All of these have different parameters and have been widely studied. Simulations of various mixing systems were also demonstrated [75].

Mixing is a process with wide applications, varying from manufacturing of concrete [76, 77] to pharmaceutics [78]. However it is still difficult to define clearly the efficiency of the mixing process or the quality of the resulting mixture. Most of the methods applied today are based on Lacey’s index [79, 80], published in 1943. This method is based on taking a sample from the mixture and determining how well each component of the mixture is presented in each sample.

Another way to describe the quality of the resulting mixture was presented by Cleary et al. [81]. This method is based on calculating the probability distribution of the desired property (such as mass or color) within the powder. Moakher et al. [82] used a method in which the property studied was the original location of the particle. McCarthy et al. [83] demonstrated a method for estimating mixing efficiency, which is based on calculating the avalanches on a rotary drum.

There are several ways to apply these models in practice. For example, Kaneko et al. [84] marked in their simulations all particles starting from a certain area as trace particles. During the mixing, the number of these trace particles within different areas was monitored to determine the quality of the resulting mixture. Masiuk and Rakoczy [85] presented another method for monitoring the mixing, based on monitoring the entropy within a given sample.

Yet another approach to determine the quality of the mixture was suggested by Lai and Fan [86], in which the variation in the amount of different compounds was analyzed with a stochastic model.

2.5 Tablet compression

The effect of powder compaction behavior on the breaking strength of the resulting tablet has been studied widely [87, 88, 89, 90], as have the effects of powder properties. For example, the effect of particle size on the tablet breaking strength [91] and the effect of various tablet shapes on the breaking strength have been examined [92].

Michrafy et al. [23] demonstrated that die wall friction largely determines the density distribution of pharmaceutical compacts, using FEM. Kadiri et al. applied a FEM for powder compaction simulation [93].

Khoei et al. introduced a software environment [19] to study powder compaction with a FEM, which was further applied in a subsequent study [20]. Wu et al. used a FEM to study tablet compression and density distribution during compaction [21]. They later applied [94] a FEM to study situations in which the cap of the tablets fails to remain attached to the rest of the tablet (the capping problem). Michrafy et al. [23] studied the
relationship between wall friction and density distribution in tableting with a FEM.

Samimi, Hassanpour et al. used DEMs to study the compression of soft granules [95, 96]. In these studies, the authors applied the DEM to determine how much the bulk behavior of the material correlates with its particle properties. Mehrotra et al. [97] used a DEM to study the effect of cohesion and compression speed on the compression process. Sheng et al. used another DEM in their study of the compaction process [98], which incorporated periodic boundaries in a unit cell. Another approach combined several spherical particles into grains and followed their compression [99].

Another interesting study [100] introduced a 2D-DEM-FEM, in which every granule of the DEM is further modeled with a FEM. The model was used to study the compaction forces and particle deformation within tablet compression. Lewis et al. combined the FEM and DEM methods in 2D simulations in order to model the strains inside particles during compaction [34].

### 2.6 Tablet crushing

An interesting theoretical model for analysis of tablet tensile strength was introduced [101]. Here, the properties affecting tablet fracturing were divided into structural and interactional fractions. The structural factors were described with a simplified, theoretically based tablet model.

The feasibility of predicting the tensile breaking strength of tablets has been studied widely. For example, the relationship of compaction speed and compression pressure on the tablet tensile strength was examined [102]. Another study attempted to determine the tablet breaking strength on the binary mixtures of well and poorly compactable material components [103]. A later study suggested a model that is able to predict the tensile strength of tablets compressed from binary mixtures [104]. Attempts to predict the tensile strength of tablets formed from binary mixtures were based on the properties of the components, applying both linear and power law mixing rules [105]. In another study, the effect of binary mixture distributions inside a tablet on the breaking strength were analyzed [106].

Simulation studies in tablet crushing have been scarce, although the cracking behavior of agglomerates has been examined [107, 108, 109, 110, 111]. An attempt to model the crushing of rocks in a jaw crusher was also performed by Refahi et al. [112].
3 Aims of the study

The purpose of the present study was to develop and apply an environment for simulating powder behavior. The aim was to further the understanding of powder packing, mixing, compression, and tablet breaking. These aims were approached by generating a simulation environment based on simple physical interactions and examining how closely such systems describe the powder behavior observed. This study focused on the following topics:

- the effect of friction and elasticity on powder packing
- determining the theoretical degree of mixing
- powder mixing with one directional force
- tablet formation during compression
- breaking strength of a tablet

Using simulations accurate enough as an aid, we can obtain additional information on several still puzzling phenomena in powder behavior. In this work the following areas were examined:

**Packing** How do single changes in grain properties affect the packing density of the powder? In laboratory experiments it is challenging to change only one aspect of the grains without risk of modifying others. Within a simulation, once the environment is established, any aspect of the grains can be changed without risk of affecting others.

**Mixing** How does steady vibration mix powder and how to determine the mixing value for the given powder? Performing steady vibrations is feasible in laboratories, yet it is challenging to determine objectively the end result of the mixing, especially when it is desired to determine the mixing efficiency of the machine in homogeneous powder. During simulation it is possible to track each particle separately throughout the process and thus to develop a method for defining the mixing.

**Mixing energy** How much energy, and how far in general, does each grain move during mixing, and how does it correlate with the mixing degree obtained? In a laboratory experiment, it would be really challenging to follow some, let alone all, particles during a mixing process. Thus, it is difficult to know the average distances they have traveled or the total kinetic energy they have received during the mixing. Knowing these can give insight into the efficiency of the mixing. This kind of information can be obtained via simulations.
**Compression**  What kind of interactions determine the interaction between grains when they are compressed into a tablet? Total pressure, speed, and many other variables can be studied from tablet compression in the laboratory. However, it is really challenging to obtain any information on the internal phenomena in the tablet. Simulation of tablet compression can give insight into the force distribution within the tablet and what phenomena cause certain areas of the tablet to remain weaker.

**Breaking strength**  How do bonds and the arrangements between grains create tablet strength, and what kind of theoretical model could describe it? Once a simulation recreates the behavior of a compressed tablet, it offers one theoretical model of the interactions that determine the tablet. In studying the breaking strength of a tablet, one big disadvantage is that the same tablet cannot be broken more than once; thus, it is impossible to derive an estimate of the error on its breaking strength, without relying on a series of tablets. Within a simulation, the theoretical variation in breaking strength of a single tablet can be estimated.
4 Simulation environment

4.1 Elements of the software

The principal idea in the simulation system is to model the interactions between single particles and, through these interactions, to obtain realistic behavior for the powder. The software itself was written in C++, and visualizations were done with either Matlab (MathWorks) or Blender (Blender Foundation). The simulation environment consists of several elements, most obvious of which are the particles themselves and the walls that define the containers in which they reside. The bonds between particles are also handled as separate entities. New bonds may form and old ones break during the time, while every particle in the simulation always exists from start to finish.

In addition, the volume simulated is divided into smaller volumes. These volumes are used to determine which particles can collide with each other. The volumes are always large enough that their diameters are larger than the largest particles. In this way it is sufficient enough to check the collisions between the particles in the same volume, and in the volumes adjacent to that volume, for a particle.

Time steps are also handled as separate entities holding information on particle locations and forces, and their maximum changes during the given time. These are saved at preset intervals and later used in either Matlab or Blender to visualize and analyze the given simulation.

4.2 Forces included

4.2.1 Overview

The system developed during this work is a DEM simulation environment. The interactions between particles are here based on Newtonian mechanics. The forces included for consideration are:

- **Gravity** affects every particle, imparting an acceleration of $9.81 \, \text{m} \, \text{s}^{-2}$ downwards.

- **Support forces between particles** are potentials that are zero when the particles are not touching, and increase the more the particles overlap.

- **Support forces between walls and particles** are similar to the support forces between two particles, with the exception that the mass of the walls is considered infinite, so that wall movement is unaffected by the interaction.

- **Frictional forces** affect the area between particles and walls, increase as the particles are pressed more strongly against each other or walls, and stop the particles from rotating or moving in reference to each other.
- *Bond forces* are formed when particles press against each other strongly enough. Once formed, the bond creates an attractive/repulsive force that keeps the particles together.

The support forces between particles can be considered as spring forces, as long as the forces are relatively small and the deformations in the particles are reversible. These assumptions were reasonable during the mixing and packing cases, and thus we kept the support forces linear in these simulations. In compaction and breaking, however, these assumptions no longer held. The particles go through heavy deformation and, as could be seen from the compaction experiments, the resistance to compaction was more exponential than linear. For this reason, the support force model was changed to an exponential form during the compaction and breaking simulations.

### 4.2.2 Spring forces

The interaction between the particles and walls is described as a potential that is formed between them. In the mixing and packing simulations, the overlapping was small. Thus, an approximation of an ideal spring of form

\[ F = k \Delta x, \]  

was used. Here, \( F \) is the force induced in a particle when it has overlapped another (or a wall) by the amount \( \Delta x \) and \( k \) is the spring constant determined for the particles. If the collisions were handled like this, they would be completely elastic and no energy would be lost. A typical way to avoid this is to use some kind of spring-dash pot approximation. In such case, a force proportional to the speed of movement, which is opposite to the direction of movement, is introduced to remove a portion of the energy. Here, the solution desired was one that would give a clearly defined elasticity factor for the particles. For this reason, we chose to implement this by making the potential asymmetric. Elasticity for the particle was defined as \( B \), which would be 0 for completely inelastic behavior and 1 for a completely elastic. The \( F \) from equation 1 is then multiplied with \( B \) when the particles retract from each other. This will cause the collision to lose a fraction of \( 1 - B \) of its energy during the collision.

### 4.2.3 Exponential forces

The spring potential was sufficiently accurate for the packing and mixing simulations, since the forces and overlapping between particles remained relatively low. In these cases it was acceptable to assume that the material would remain within its ideal spring area. However, in the compaction simulations, in which the material was compressed into nearly half of its volume, the overlapping and thus forces included exceeded the limits of this approximation. We noted that the form of the resistance to compaction was closer to exponential (IV) than linear in these cases.
For these reasons, in simulations that include compaction or tablet breaking, a potential of form

\[ F = k \Delta x + Ae^{a\Delta x} - A \]  \hspace{1cm} (2)

was used. Here \( k \), \( A \), and \( a \) are constants determined by the properties of the material and particle size. \( \Delta x \) is the overlapping distance of the two interacting particles. For closer details of application see (IV) and (V).

4.2.4 Bond forces

Bond-forming simulations were applied in compaction and breaking simulations, in which the interactions between particles may cause considerable deformation. Available results and experiments both show that even if bond formation occurs, the force increases exponentially (IV). As such, the effect of the bonding force on its particles was modeled so that compression of the powder would give similar, exponentially increasing resistance with and without bond formation. The bonding force was also chosen to be symmetrical in relation to the rest distance of the bond. Details on the bonding forces are shown in (IV).

4.3 Time

Since it is not possible to calculate all the interactions every particle experiences analytically, some approximations must be made in every simulation. Here the main approximation is that time is divided into steps, during which no interaction force changes. Between each step, new interaction forces are calculated. Ideally, when the time step applied nears zero, this approach would give exactly the right interactions, provided of course that the models used for describing the forces were exactly correct. This, however, would also require infinite time for the calculations. Therefore, a compromise between accuracy and speed must be made.

The time step can be relatively large when no collisions occur. The limit here is that if single particles move distances close to their own radii, they could miss a collision completely. See Figure 2 for illustration.

When the actual collisions do occur, the requirement for a time step becomes much stricter. During the collision, the force a particle experiences increases all the time as it travels further. Thus, it becomes increasingly difficult for the particle to advance each moment. If this advance is, however, divided in to time steps that are too long, it will move to overlap another particle or wall with too little energy lost, or even overlap far more than it was supposed to. This would create excess repulsion between the interacting bodies, which would create too much energy in the simulation. With the time steps clearly too large, this will cause the collisions to produce energy instead of reducing it, thus making the simulation unstable. See Figure 3 for illustration of the problem.
Figure 2: Time step is too large, so a collision is missed.

Figure 3: Above: accurate simulation of a collision. Below: Time step is too large, and thus the energy is increased during the collision.
In all the simulations performed here the time step varied in an attempt to maximize the time required and accuracy. The length of the time step was chosen for each step, and after evaluation of the step it was either canceled and reprocessed with a smaller time step, or accepted, and the next step was processed with a slightly longer time step.

The rules for accepting a time step were:

- **Maximum change in force** A value for maximum change $\Delta F_{\text{max}}$ in any force was set. If any interaction within a simulation was greater than $\Delta F_{\text{max}}$, the time step was rejected. This prevented very steep potential curves from creating the problem illustrated in figure 3.

- **Maximum change in location** A value for maximum change in particle location $\Delta X_{\text{max}}$ was set. If any particle moved between single steps more than $\Delta X_{\text{max}}$, the time step was rejected.

- **Minimum time step** In some exceptionally strong collisions the time step required to meet the two earlier rules can become very small. This could cause a single collision to take most of the simulation time. Yet, as a single collision, it does not necessarily enhance the accuracy of the total simulation very much. Therefore, a minimum time step $\Delta T_{\text{min}}$ is set. If the length of the time step decreases to this, it will be accepted in any case.

In addition to these rules for decreasing the time step, a set maximum time step $\Delta T_{\text{max}}$ is also defined. A static situation in which every particle is at rest would not break any of the given rules, no matter how large the time step. In a simulation, however, the state is never completely static, because the interactions are divided into time steps. Thus, particles oscillate slightly even in a resting position. If $\Delta T_{\text{max}}$ were $\infty$, the time step would increase until one of the rules would break, thus making the static situation unstable.

### 4.4 Energy within the simulations

Energy in the simulations exists in four different forms: potential, kinetic, rotational and internal energies. The total energy inside the simulation is thus presented by Equation 3

$$E = E_p + E_k + E_{\text{rot}} + E_i,$$

where $E$ is the total energy inside the simulation and $E_p$ is the potential energy, $E_k$ the kinetic energy, $E_{\text{rot}}$ the rotational energy, and $E_i$ the internal energy.

Some of the energy is lost from the simulation, due to the inelastic collisions and friction. This energy is represented as $E_{\text{Lost}}$. If the simulation ran without any error in energy conservation, the sum of the energy inside the simulation and lost energy would always equal the initial energy. Any difference in the initial energy and the sum of $E$ and
$E_{\text{Lost}}$ would thus be a result of the inaccuracies in the simulation. This is represented using Equation 4

$$E_{\text{start}} = E + E_{\text{Lost}} + E_{\text{Err}},$$

where $E_{\text{Err}}$ is the energy gained or lost due to simulation inaccuracies.

There is a constant conversion between different forms of energy. The energy flow during a simulation is described in Figure 4. Initially, all energy is potential or kinetic energy. During the simulation these are converted to other forms of energy, including rotational, internal, and lost energy. Some simulations have active outside forces, such as vibrating containers, or moving punches. These bring additional energy into the simulation.

![Energy Flow Diagram](image)

Figure 4: The flow of energy during the simulations. Lost energy is the energy removed from the simulation due to friction and inelastic collisions. Other forms of energy still contribute to the simulation and are, in principle, measurable.

Methods for acquiring values for $E_p$, $E_k$, $E_{\text{rot}}$, and $E_i$ are described below.

### 4.4.1 Potential energy

The potential energy is determined using Equation 5

$$E_p = \sum_{n=1}^{k} h_n m_n g,$$

where $E_p$ is the total potential energy, $k$ the number of particles, $h_n$ the distance of the center of mass of the nth particle from ground level, $m_n$ the mass of the nth particle, and $g$ the gravitational constant. The ground level is defined as the bottom of the container in the simulation.
4.4.2 Kinetic energy

The kinetic energy is determined with Equation 6

\[ E_k = \sum_{n=1}^{k} \frac{1}{2} m_n |\vec{v}_n|^2, \]  

(6)

where \( E_k \) is the total amount of kinetic energy in the simulation and \( \vec{v}_n \) the velocity of the \( n \)th particle as a three dimensional vector in space. Kinetic energy concerns only the velocity of the particle center of mass.

4.4.3 Rotational energy

The rotational energy describes the energy involved in internal rotation of the particles. Its total value is determined using Equation 7

\[ E_{rot} = \sum_{n=1}^{k} \sum_{j=1}^{3} \frac{1}{2} J_n |\omega_j|^2, \]  

(7)

where \( E_{rot} \) is the total amount of rotational energy, \( j \) describes one of the three axes of rotation, aligned to the \( x \), \( y \), and \( z \) coordinates, \( J_n \) is the moment of inertia for particle \( n \), and \( \omega_j \) the rotational speed along axis \( j \). Since all particles are considered ideal, homogeneous spheres, the moment of inertia for all axes for particle \( n \) can be acquired from Equation 8

\[ J_n = \frac{2}{3} m_n r_n^2, \]  

(8)

where \( r_n \) is the radius of particle \( n \).

4.4.4 Spring energy

Spring energy describes the stored pressure and strains in the particles and walls. Since particles and walls are nontransforming objects, the kinetic energy lost when particles are pressed together is converted to spring energy. The amount of kinetic energy transformed to internal energy is determined by the particle elasticity \( (B) \). If \( B \) is equal to 1, all of the kinetic energy is conserved and is transformed from kinetic energy to internal energy. However, if the factor is 0, then none of the energy is conserved. Energy bound as spring energy in the simulation is dependent on the amount of contacts between particles and between particles and walls. The actual form of the interaction is dependent on the choice of potential for the interaction. These are considered in more detail in subsection 4.2.4, and in (I) for the spring approximation and (IV) for the exponential approximation.
4.4.5 Lost energy

In addition to the forms of energy described above, some of the energy is lost during the simulation. In these simulations there are two factors that remove energy from the simulation: inelastic collisions and frictional forces. The degree of energy conservation during the simulation can be checked by recording the amount of energy expected to be lost.

Energy is always lost during collisions that are not completely elastic. In a completely elastic collision, the amount of energy stored within the spring potential can be calculated, based on the interacting potential. In an inelastic collision, only part of that energy is transformed into internal energy. The elasticity of the collision (determined by particle elasticity \( B \)) determines the fraction preserved. Since no energy is lost during a completely elastic collision, the energy lost during a nonelastic collision can be computed from the difference of internal energy gained between the elastic and inelastic collisions.

The amount of energy lost due to friction is recorded in a similar way: the influence of friction on the rotational speed, velocity, and location of each particle is recorded. The total amount of energy is then calculated, using Equation 3. Subsequently, Equation 3 is used to calculate the rotation in the absence of friction-induced changes in rotational speed, velocity, and locations of particles. The energy difference between these two situations indicates the amount of energy the model has lost due to friction. The \( E_{\text{Lost}} \) in Equation 4 is determined by summing up all the terms of energy lost due to inelastic collisions and friction.

4.5 Particles

During these simulations all the particles were considered symmetrical spheres. The parameters describing a particle were radius, density, mass, elasticity, and friction coefficient. Friction was assumed to be the same between particles and particles and walls. Rest friction and moving friction were also assumed to be the same.

In the packing and mixing simulations, the particle-particle and particle-wall support forces were linear (Equation 1). Parameter \( k \) here determined the final strength of the interactions. In the compaction and breaking simulations, the support forces were exponential (Equation 2). Parameters \( k, a, \) and \( A \) determined the final strength of the interactions in these cases.

4.6 Elementary testing of the system

To use the model in the powder simulation, we needed first to prove that it worked in simple situations involving only one or two particles and a wall contact. These were tested thoroughly at the beginning of this study. Examples of the situations tested are illustrated in Figures 5 and 6.
<table>
<thead>
<tr>
<th>collision type</th>
<th>initial</th>
<th>inelastic</th>
<th>semi-elastic</th>
<th>elastic</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
</tr>
<tr>
<td></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
</tr>
<tr>
<td></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
</tr>
<tr>
<td></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
</tr>
</tbody>
</table>

Figure 5: Illustration of primary collision testing. Arrows on the spheres show the velocity (speed and direction) of the sphere. On the left side is the original situation, and in the right columns the situation shortly after the collision.

The tests performed for the system included collision with a wall (with and without gravity). Depending on the elasticity given to the particle, it was assumed to either bounce off at the same speed, stop at the wall, or bounce off at a lower than initial speed. This was achieved with suitable time steps. Long time steps either caused the particle to bounce off at higher speed than it was supposed to, or in extreme cases go through the wall.

The same experiments were done for particle-particle collisions, with one of the particles stationary, or with both of them moving. Again, based on the elasticity given to the particles, they behaved as expected, when the time steps were short enough. In these tests, we also verified that if the collision was not completely on one axis, the directions of the bouncing particles were in agreement with the conservation of energy and momentum.

The transmission of force was tested by setting series of particles in a line touching each other. Then, one particle was shot into the end of the line. With high elasticity the
<table>
<thead>
<tr>
<th>friction</th>
<th>initial</th>
<th>no friction</th>
<th>high friction</th>
</tr>
</thead>
<tbody>
<tr>
<td><img src="image1.png" alt="Initial" /></td>
<td><img src="image2.png" alt="No Friction" /></td>
<td><img src="image3.png" alt="High Friction" /></td>
<td></td>
</tr>
<tr>
<td><img src="image4.png" alt="Initial" /></td>
<td><img src="image5.png" alt="No Friction" /></td>
<td><img src="image6.png" alt="High Friction" /></td>
<td></td>
</tr>
<tr>
<td><img src="image7.png" alt="Initial" /></td>
<td><img src="image8.png" alt="No Friction" /></td>
<td><img src="image9.png" alt="High Friction" /></td>
<td></td>
</tr>
<tr>
<td><img src="image10.png" alt="Initial" /></td>
<td><img src="image11.png" alt="No Friction" /></td>
<td><img src="image12.png" alt="High Friction" /></td>
<td></td>
</tr>
</tbody>
</table>

Figure 6: Illustration of primary friction testing. Arrows on the spheres represent the velocity (speed and direction) of the sphere. The curved arrows inside the spheres represents the sphere’s angular momentum. On the left side is the original situation, and in the right columns the situation after a moment of simulation.

The line remained stationary, but the particle at the other end of the line was projected away at the same speed as the original came in. With low elasticity the entire line began to move forward at the original speed per particle. The softer the particles were (the more they overlapped each other during the collision), the more the particles separated from each others during this.

Friction was also introduced into the system, and with it the angular momentum and rotation of the particles. This was tested first by placing a particle on a tilted plane under the effect of gravity. Without friction the particle began to slide on the plane without rotation. If the time steps were too long, the particle did not remain on the plane, but began to accumulate energy and to jump. With friction introduced, the particle began to roll down the plane. The rotational speed compared with the particle speed stabilized so that the particle’s surface point facing the plane remained stationary, as expected.

Friction and rotation were also tested by gliding a particle on the plane. When no friction was introduced, the particle simply glided at the original speed with no rotation on
the plane. With friction included the particle begun to roll, stabilizing at constant speed and rotation where the contact point with the floor was again stationary, as expected. When gliding a particle with initial rotation in the direction opposite to that of the velocity, the particle with friction slowed down, and even reversed direction if enough rotational speed was given.

A 2D test was also performed, in which three particles were piled together on a plane. With no friction, the pile immediately collapsed. With sufficiently high friction the two particles on the bottom were able to keep the third above them. A similar 3D test was done successfully with four particles. During all these tests the total energy and momentum were tracked, ensuring that both the laws of conservation of energy, and momentum held.

4.7 Mixing degree

In the course of this investigation, a new method for defining the quality of the mixing process (mixing degree) was presented (II). The mixing degree ($M_s$) described here gives a value describing how closely the current particle configuration resembles the initial configuration, compared with any completely random configuration. A value of $M_s = 0$ indicates that the system is still in its initial state, while $M_s \approx 1$ indicates that the current configuration is no more like the initial configuration than any random configuration. As such $M_s$, unlike most methods applied in studying mixing quality, describes the mixing process and its quality, rather than the quality of the mixing state obtained. Assume we have a mixture of black and white particles, which starts from a configuration that is completely random. No matter how well it is mixed, a method based on sampling would see no change in the situation. If, however, the initial situation would have had the black and white spheres completely separated, a sampling method would indicate that the mixing process improved the mixture significantly. $M_S$ would show in both situations that the mixing process was equally efficient.

In this approach the mixing degree will be determined, based on how much the positions of the surrounding particles of each individual particle have changed compared with the initial situation. The method determines the starting neighborhood for each particle. The starting neighborhood is composed of all other particles, so that the weight of a particle in the neighborhood is inversely proportional to its original distance. The mixing degree is then determined by the change in distance between particles. The change in distance between a particle that started close to the given particle produces a larger effect on the mixing degree, while a particle that started far away produces a smaller effect, no matter how much it has moved. See Figure 7 for description of the particle neighborhood.

The value obtained will then be scaled, using random states. A random state here is defined as a state in which the particles of the initial state occupy the same space as the initial state, but their exact locations within that space are completely random.
Figure 7: Effect of the neighbors and their movement on the mixing degree. For particle A, the movement of particle 1 gives the largest change. Particle 2 moves as much, but is farther away, so its impact on particle A’s neighborhood is lighter. For particle B the movement of particle 1 has the least meaning, since it is farthest away. Particles 2 and 3 are both as close, but number 2 moves more, so that it has the highest effect on particle B’s neighborhood. The total mixing degree is calculated, based on all the particles. Thus, a dislocation which causes changes in most particles will be most meaningful. Group C, for example moves, but the C particles will not change location in respect to each other. Thus, the effect of that movement only counts as a change on the particles around them.

The impact of each particle on all other particles’ neighborhoods is determined, using their initial distance. The weight parameter will be determined as $R^{\beta|\Delta x|}$ where $\Delta x$ is the distance between particles $i$ and $j$, and $R$ a scaling factor of dimension length$^{-1}$. $R$ is a dimensionless variable that can be chosen between 0 and 1 (for proper choice of $R$ and $g$, refer to II.) For the situation studied, the distance between each particle $i$ and $j$ may have changed, and the new distance is marked as $x_{nj}$. In considering mixing of the near neighbors, the initial movement away from its original neighbors is most important. Later movement still further has less of an effect. To take this into account the distance moved is scaled with the factor $R^{\beta|\Delta x_{ij}|}$. When these are put together, we obtain Equation 9

$$M_{ij} = R^{\beta|\Delta x_{ij}|}(1 - R^{g|\Delta x_{ij}|}),$$  \hspace{1cm} (9)

where $M_{ij}$ describes the unscaled mixing value of a single particle pair $i$ and $j$. For these to describe mixing of the entire sample, all particle distances must be summed for each particle, and then all the sums for each particle must finally be summed together. Thereafter, this will be scaled, based on the number of particles ($N$). Thus, the mixing degree for the entire system becomes:
\[ M = \sum_{i=1}^{N} \sum_{j=1}^{N} \frac{R^{|x_{ij}|}(1 - R^{|x_{ij} - x_{n_{ij}}|})}{N^2}, \]  

This value \( M \) is a dimensionless value, which will be zero for the initial state. It increases as the state develops further out of the initial order.

The maximum value for \( M \) evaluated this way would be reached when all of the particles are infinitely far from each other, the maximum value being (from Equation 10) while all \( x_{n_{ij}} \)s approach infinity:

\[ M_{\text{max}} = \sum_{i=1}^{N} \sum_{j=1}^{N} \frac{R^{|x_{ij}|}}{N^2}, \]

as \( R^{|x_{ij} - x_{n_{ij}}|} \) becomes zero if \( 0 < R < 1 \) and all \( x_{n_{ij}} \) approaches infinity.

Scaling \( M \) with \( M_{\text{max}} \) would give us a value that ranges from 0 to 1. This could be seen as the absolute value of mixing. This absolute value, however, does not really describe the mixing as such. This value does not reach 1 before the particles have been spread out into a much wider space than the initial volume.

The actual scaling is done by creating several random states. In these the same amount of particles occupy the same container as in the initial state. However, the actual positions inside are completely random and thus different in each random state. We obtained \( M_{\text{rand}} \) intrinsic to the initial state by calculating the mean mixing degree for these random states. \( M_{\text{rand}} \) is an average value of \( M \) for a state with random positioning of particles compared with the initial state (in other words, is completely mixed). The value of \( M_{\text{rand}} \) varies only slightly when calculated from different random states derived a from given initial state. In the case described (II), the mean value for \( M_{\text{rand}} \) was 0.127 with a standard deviation of 0.001. It is worth mentioning that even when the \( M_{\text{rand}} \) is constant for a given initial state, it differs for each different initial state. The \( M_{\text{rand}} \) must be calculated separately each time the setup changes, e.g. if there are changes in the space occupied, or type or amount of particles analyzed. Dividing the mixing value \( M \) for any given state with the corresponding \( M_{\text{rand}} \) gives the scaled mixing degree \( M_s \) as

\[ M_s = \frac{M}{M_{\text{rand}}}, \]

which would be 0 for a state identical to the initial state and around 1 for a state that is as far from the original as a random state is. Less mixed systems would then have a value somewhere between 0 and 1. Since \( M_{\text{rand}} \) is an average of the \( M \) of the random states, some random states have higher values of \( M \) than the average. Thus, values of \( M_s \) exceeding 1 are possible. Considerably larger values could be achieved if the particles in the situation studied no longer occupied the same volume as the random states used in scaling. Another way to achieve a value for \( M_s \) higher than 1 would be if the particles for some reason would arrange systematically so that the initial neighbors end up farther
away than in a random state. This may be possible in some specific cases of segregation. For systems inclined to segregate, the method may be applicable for determining the degree of segregation, if the segregation tendency is taken into account while generating the random states used for scaling. However, further studies are required before the method should be applied to systems of this kind.
4.8 Tablet breaking

When simulating compression, bond behavior could be simulated as a two-point attractive-repulsive force always directed towards or away from the particle centers of mass. After bond formation the particles usually can not move around much, and there are several particles locking them together in a 3D structure.

However, when studying the breaking of such systems, the attractive-repulsive model shows some considerable weaknesses. If a particle is attached to only two others, it is free to roll around one axis (and if attached to only one, in any axis). For this reason, a tablet modeled with this approximation will never break suddenly, but rather behaves somewhat like an amorphous mass (see Figure 8 upper part).

This problem was solved by modifying the algorithm to include strain from torque to the bonds. In this case, not only the distance between the two particles in consideration affects the force that the bond inflicts, but also their orientations with respect to each other. The effect of this addition is illustrated in the lower part of Figure 8. The algorithm itself is explained in more detail in (V).

![Diagram of tablet breaking process]

Figure 8: Upper image: 2D illustration of breakage with attractive-repulsive forces holding the particles together. Lower image: the same with a model that takes the torque into account.
5 Experimental

5.1 Software used

5.1.1 Simulations

All the simulations of this research were performed with a software developed for this investigation. (vpress, currently unreleased). This software was created, using Microsoft Visual C++.

5.1.2 Analysis

Most of the analysis of the data was done in Matlab (various versions 5.3 – R12). Matlab was also used for the evaluation of the mixing degree algorithm (II). Here, a simple system randomizing points in a given space was constructed. For moving the points in random ways, the same system was applied.

5.1.3 Visualization

Blender was used for visualization in some simulations. Python programming language was used for developing the interface between vpress and Blender.

5.1.4 Others

For analysis of the 2D mixing experiment (II), Blender was used to extract images from the videos and Matlab to develop simple image recognition algorithm for tracking the disks in the experiment.

5.2 Instruments

5.2.1 Mixing equipment

In the 2D mixing experiment, the mixing was done with a Heidolph ROTAMAX 120 mixer. The setup is presented in Figure 9.

5.2.2 Tableting machine

The tablets were compressed with an instrumented eccentric tableting machine (Korsch EK0; Erweka Apparatebau, Germany) using flat-faced punches with a diameter of 9 mm (IV and V).

5.2.3 Crushing strength tester

The crushing strengths of the compressed tablets were measured with a Schleuniger-2E (Dr. K. Schleuniger & Co, Switzerland) tablet hardness tester (IV and V).
5.3 Materials

In the 2D mixing experiment (II), the mixing material consisted of 80 rubber discs with a diameter of 22 mm. The middle of each disk was marked with a red dot. Half of the disks were gray, the rest were white.

Tablets compared with simulations (IV and V) consisted of theophylline anhydrate and microcrystalline cellulose pellets. The dry pellets were sieved manually, and those between 0.7 and 1.0 mm in diameter were selected for subsequent experiments. Figure 10 shows examples of the cellets used.

![SEM image of cellets.](image)
6 Results and discussion

6.1 Packing

The effects of friction, radius, elasticity, and spring coefficient were studied with several simulation series (I). Of the properties studied, friction had the clearest effect on packing density (I). The highest packing density acquired was 57% and was obtained with a friction factor of 0. The lowest packing density was 45% and was acquired with the highest friction factor used (0.8). In general, higher friction factors lowered the packing density.

The effect of the friction factor on packing was also examined with a scale 10-fold smaller than that used in other tests (I). The spring forces and gravity were still held the same. Effectively, this simulates a test with similar material, but on a smaller scale. Increasing friction factor still decreased the packing density, albeit the effect was slightly weaker than in the other series.

6.2 Mixing degree

The mixing degree ($M_s$) was introduced here (II) to describe the efficiency of mixing in a given setup. $M_s$ is based on calculating the changes in the initial neighborhood of each particle at the given time. Figure 11 presents six repeated simulations in which the points are randomized one by one, until every particle is in a completely random location compared with the initial position. As the figure shows, the $M_s$ reacts to the percentage randomized systematically.

![Figure 11](image)

Figure 11: The $M_s$ for test sets from 1 to 6 (II) as a function of the percentage of randomized points.
Using $M_s$ as a method to evaluate mixing is mainly aimed at simulations, due to the need for knowing the location of each particle separately. If, however, a method to track at least a few tens of particles is given, it could be applied in real systems. Such a method could include a radioactive tracer or camera (e.g. with transparent glass beads). A small 2D experiment based on camera tracking was presented (II). Figure 12 shows several stages of the mixing, and corresponding calculated $M_s$ of each stage of this experiment.

![Initial state](image1.png) ![Intermediate state](image2.png) ![End state](image3.png)

Figure 12: Mixing experiment with 2D disks. Tracking of each disk was done with a high-speed camera. Calculated $M_s$ is shown under each image.

![Figure 13](image4.png)

Figure 13: Illustration of the mixing degree. All four situations are of the same 2D mixing simulation in different phases; each star and circle represents one point. The $M_s$ and $M_s^2$ are listed under each representation.

$M_s$ also takes into account the changes between similar particles. Therefore, it increases faster than one would intuitively expect (see Figure 13). For this reason, it may
be more intuitive to study $M_s^2$ when doing visual observation, since its value of 0.5 looks more half-mixed.

6.3 Shaking parameters and mixing energy

Higher amplitudes with equal frequency considerably increased the particle traveling-distance during the mixing (III). This is to be expected, since increased amplitude directly increases the distance the container itself moves in each disturbance. The vibration direction had a lesser effect on the total traveling distance. The distance was slightly shorter when the direction was not directly against either axis or the diagonal. Friction had no obvious effect on the distance traveled. Increasing the frequency while keeping amplitude constant generally decreased the traveling distance; however, at high frequencies (400 Hz) the distance was suddenly longer again. With strong vibration, particles initially moved very quickly, and thus most of the moving in this simulation occurred in the first few moments of the simulation. When the frequency was changed while keeping the speed of the vibration constant, the average traveling distance quickly shortened as the frequency increased.

Each of the simulated series showed a considerable amount of mixing during the first disturbance. After this, the mixing advanced similarly in each simulated series. The rapid initial mixing was explained by the different nature of the first disturbance: each particle is at rest and thus gains velocity in the same direction during the first disturbance. In later disturbances, however, some particles will be moving in opposite directions, thus damping the impact. In addition, if the amplitude and speed are intense enough, the first disturbance is the only one in which particles will be fully affected. Later on the particles will not have enough time to fall back to their initial level before they meet the container again. See Figure 14 as an illustration for these separate cases.

![Diagram of mixing process](image)

Figure 14: Illustration of the first disruption (left) and consequent disruptions (middle). If the frequency of the mixing is high enough, the particles will not have time to fall to the bottom before the next disruption sends them back up (right).
Generally, the average length of a particle’s trajectory correlated well with the total mixing degree (III). No clear correlation with the direction of the vibration occurred, however. The simulation with the highest friction studied (0.3) had trajectories as long as those with smaller frictional factors, even if the mixing degrees of these were smaller. When the frequency (with a constant amplitude) was changed, some simulations showed good mixing and yet short trajectories (100 Hz). Others showed poor mixing with long trajectories (25 Hz) and good mixing with short trajectories (200 Hz) (see (III) for details). From these examples, we see that the distance traveled does not necessarily correspond to the quality of mixing.

The particles starting from the bottom generally had shorter trajectories than those starting from the top. On average, the particles starting near the edges also had longer trajectories than those starting near the center. The difference between the center and the edges was, however, smaller than with the bottom and top halves (see III for details).

The energy transferred into the system did not always correlate directly with the degree of mixing. For example, when varying the mixing frequency by a constant amplitude, the lowest kinetic energy for the particles was obtained with the highest amplitude. However, the highest amplitude also resulted in the best mixing in that series.

### 6.4 Tablet compression

Tablet compression was studied within the same simulation environment (IV). A new model for handling the bonds forming between the particles during compression was introduced in these simulations. Comparison between the simulated compression curves and the measured compression curves indicated that the bond model was able to model the final force experienced by the punch very accurately. Figure 15 shows both the simulated and measured compression curves, Figure 16 the difference between these two during the compression.

The force decreased more slowly during release in the simulated tablets. This was caused by the parameters used in the simulation: the particles were made slightly softer and more elastic than the real ones in order to keep the simulation times reasonable.

The internal strength of the bonds during tableting were also examined. Examples of the distributions of bond strengths are shown in Figure 17. The strongest bonds were generally located on the edges of the tablets, and occasionally a stronger area was seen in the very middle of the tablet. This is in agreement with the density distributions available in the literature, although the particle amounts used in these simulations were too small to obtain very detailed images of the variations in the middle part of the tablet.

An estimate for the tablet strength was calculated first, based only on the average strength and amount of the bonds formed in the tablet (III). The change in strength with increasing compression force was similar to the measured changes in strength. The standard deviation obtained this way was, however, clearly smaller than in the experiments.
Figure 15: Simulated (left) and measured (right) compaction curves. The red, black, green, and blue curves indicate different compaction forces. The red curves show the lowest compaction force, and the blue, the highest.

Figure 16: Simulated compression force, as a function of the compression force measured, is shown as a black curve. The gray, dashed curve shows the perfectly linear correlation.
Figure 17: Examples of the bond strength distributions of the simulated tablets. These distributions were obtained from the highest compression force, resulting in a tablet thickness of 3.17 mm. Images have been mirrored horizontally from the middle.
This was largely due to the fact that measured strength is a factor of only the weakest bonds in the middle axis of the tablet. Often the first breakages are enough to break the whole tablet.

6.5 Tablet breaking strength

Tablet breaking was simulated (V), to determine the strengths from simulated tablets, which were comparable with the measured breaking strengths. The bond-strength model used caused the tablets to break in a fashion very similar to that of the measured tablets. Figure 18 gives examples of measured tablet breaking and Figure 19 of simulated tablet breaking. Figure 20 shows how the strains in the bonds are distributed. The strain is similar to the results in the literature in that when the tablet is broken by lateral stress, it is actually torn apart rather than crushed. Figure 20 shows that most strained bonds in the vertical axis are indeed pulled rather than pushed.

Figure 18: Examples of a measured tablet breaking.

As shown in (V), the standard deviations in the simulated tablet strengths were very similar to the measured deviations, once they were obtained by simulating the actual breaking phenomena. This was also speculated in (IV), where the tablet strength was determined roughly from the average strength of the bonds in the tablet.
Figure 19: Examples of simulated tablet breaking. Each line represents a bond between two particles.

Figure 20: An example of the bond strain distribution within a tablet a moment before it breaks. The blue lines indicate bonds that are stretched and the red lines bonds that are pushed together. On the left side only the strained bonds, on the right side only the pushed bonds are illustrated. The middle image shows all the bonds.
7 Conclusions

The simulated packing densities were realistic (I). As such, we can conclude that the forces considered here (gravity, friction, support forces) are enough to describe the packing of ideal spheres. When only these interactions are considered, friction has a greater effect on final packing density than elasticity, size, or hardness.

The value $M_s$ presented in (II) describes well the mixing process and is in line with previous methods. While aiming at simulation, it can in principle be applied in experimental study, with proper methods for tracking some or all particles.

Differing results are acquired when mixing powder with vibrations that differ in amplitude, frequency or direction. However, stronger vibration does not automatically result in better mixing. We showed (III) that even when applying only a few interactions (gravity, friction, support forces) some vibrations resulted in better mixing of the powder, with less energy spent.

Linear support forces between particles cannot explain the compaction pressure development, but a linear plus exponential force gives an acceptable match. The bond model applied in these studies resulted in both equal compression strength curves in tableting and equal variation in crushing strength. The simulated tablet strength distribution also resembled in large scale the density distributions found in the literature.

The variation in crushing strength between identically compressed tablets can be explained by differences in the orders and orientations of the contacts between particles inside the tablet (IV). Crushing the same simulated tablet in different orientations produces as much variation as crushing different simulated tablets of identical, but random packing. The variation is similar in scale to that in the experiments.

In the light of these studies, we conclude that simulations can be used to further fundamental understanding of the behavior of granular material and explain the phenomena observed in them. The simulation environment established here appears to be applicable for research on powder technology.
References


