Cohesion and Interparticle Forces

E.F. Hobbel
Cohesion and Interparticle Forces

\( \dot{c} = 9,000 \)

\( \Delta t \text{ d.s.s } 1 \text{ mgy} \)
Dit proefschrift is goedgekeurd door de promotor prof. B. Scarlett M.Sc.
Summary

In this investigation a number of properties connected with the cohesive behaviour of a powder have been analysed and related to the interparticle forces.

The tensile strength and cohesion of compacted systems, and the cohesion of an aerated system, were the properties studied experimentally. The mechanisms which give rise to the behaviour observed have been clarified and suitable models are suggested. These models are based on the idea that all the interparticle interactions are dispersely distributed.

The behavioural parameters of a powder are related to those of the constituting particles. For a fundamental characterisation of powders a morphological analysis is most suitable. In this investigation a fast algorithm has been designed which determines an approximate polygonal form of the particles. It is shown that the polygonal description will be more powerful than the transformation of the polar representation of the contour in spherical harmonics. The algorithm is demonstrated on a stand alone PC, and can readily be implemented in image analysers.

The distribution function of the normal separation force of single particles adhering to a wall has been experimentally investigated in a centrifuge set-up. Both the Log-Normal and the Gamma Distribution were adequate for a variety of experimental conditions.

A Hollow Dutch Cell was built in which a powder can be subjected to a state of stress in which one of the principal stresses is tensile. Two modes of failure were observed.

Assuming a Gamma distribution of the interparticle forces, it is shown that the maximum force measured in a tensile test does not coincide with the breakage force, but can be substantially lower.

The fracturing of a powder cake is analysed in detail and a crack propagation theory is adapted for powders to explain the phenomena quantitatively.
The yield locus, which describes the resistance of a powder shear plane to plastic flow, leads to the concept of cohesion. The formation of shear planes has been investigated using a Coaxial Shear Tester. In modelling the behaviour of the shear plane the mechanisms which lead to a maximum value of the resistance during deformation are different than those for crack propagation. No apparent simple connection between tensile strength, as measured in a tensile tester and cohesion, as measured in a shear tester can be established.

For aerated systems a viscometer was designed which could detect the resistance to shear without disturbance to the structure. A model, based on Coulomb friction and increasing compressive stress, correlates the results well and enables the differentiation of the total torque recorded into the stress on the wall and base of the cylinder. A value of cohesion, similar to that defined from the yield locus can be defined for aerated systems. This value is dependent on the gas flow.
Samenvatting

Een aantal aspecten van het cohesief gedrag van poeders zijn nader onderzocht en verklaard met de krachten, die op deeltjes niveau werken.

Als voorbeelden van macroscopisch gedrag zijn de treksterkte en de cohesie van gecomprimeerde systemen en de frictie en cohesie van geaerende systemen nader onderzocht. De achterliggende microscopische mechanismen zijn geïdentificeerd en waar mogelijk gequantificeerd. Steeds zijn de modellen zoveel mogelijk gebaseerd op de fundamentele grootheden op deeltjes niveau en op de wijze waarop deze grootheden gedistribueerd zijn. De basis gedachte is dat het gedrag van poeders geheel te verklaren is m.b.v. de gedragingen van de afzonderlijke deeltjes.

De gedragingen van deeltjes worden voor een groot deel bepaald door de relevante vorm van de deeltjes. De meest fundamentele beschrijving van de morphologie is gebaseerd op de fysische lengtematen van het deeltje. Een algoritme is ontworpen om de polygonale basisvorm van een deeltje te bepalen en deze aan de ruwe contour te fitten. Deze techniek kan de basis zijn van hogere order eigenschappen bepalingen en levert veel meer fundamentele informatie op dan bijvoorbeeld de onschrijving naar polaire harmonischen. De techniek is op een stand-alone PC gedemonstreerd en is klaar om in beeldverwerkers geïmplementeerd te worden.

De kansverdeling van de kracht, nodig om een deeltje van een wand te trekken, is met een centrifuge gemeten. Een beeldverwerker classificeerde de deeltjes in nauwe grootteklassen en berekende de individuele verwijderingspercentages. Zowel de log-normaal als de gamma distributie bleken adequaat voor een aantal verschillende testcondities.

Om aan poedermonsters gelijktijdig trek en drukspanningen op te leggen is een holle Dutch Cell ontworpen en getest. Twee bezwijk toestanden werden geobserveerd, welke afhankelijk zijn van de totale spanningstoestand.

Gebruikmakend van de verdeling van krachten tussen de deeltjes is aangetoond dat de treksterkte van een poeder kleiner is dan de theoretische breuksterkte gebaseerd op de sommatie van krachten.

De opbreking van een poedermonster in een treksterkte meter is in detail onderzocht en de geassocieerde verschijnselen zijn verklaard met een aangepaste scheurtheorie voor cohesieve poeders.
De cohesie is gedefinieerd als een punt op de bezwijklijn, welke de maximale weerstand van een poeder tegen afschuiving weergeeft. De formatie en de mechanismen, die de hoogte van de weerstand bepalen zijn experimenteel en met modellen onderzocht. Een afschuiptest, waarin de afschuifzone coaxiaal is met de as van draaiing, de Coaxial Shear Tester, is voor dit doel ontworpen en getest. De modellen voor de schuifzone en die, eerder ontworpen voor de trekmetingen hebben geen gemeenschappelijke basis en de hypothese dat de treksterkte van een poeder een extrapolatie van de bezwijklijn zou zijn, wordt dan ook door de modellen niet ondersteund.

Voor het onderzoek van het afschuifgedrag van geaereerde systemen is een viscometer ontworpen en getest, welke de fragile structuren van het bed nauwelijks verstoort. De gemeten torsie op de as van de cilindrische probe kan, met behulp van een Coulomb model voor de schuifspanningen in de structuur, omgezet worden in een wandschuifspanning. De Coulomb aannemer leidt tevens tot de definitie van een cohesieve kracht van de structuur, welke afhankelijk blijkt van de superficiële gassnelheid.
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Chapter 1. General introduction

Particle technology is ancient. The engineering knowledge dates back to early man, who made slurries and pastes for painting and food, and later ground, molded, mixed and sintered particles for pottery. However rules of thumb and the knowledge of experienced individuals were the only sources of information to control and design processes. The modern need for more precise prediction of powder behaviour creates the requirement for a more sophisticated description of the materials. Models of the flow behaviour of particle systems must be based on fundamental properties in order to be more predictive rather than just correlating behaviour.

The field of particle technology can roughly be divided into four area's:

- particle characterisation
- particle system mechanics
- physical and chemical interaction
- technological application of particle systems

To come to a properly designed application an investigation of all these subjects is necessary.

1.1 Basic definitions

A particle is a (small) coherent volume of a phase.
A particle system is a set of particles which directly or indirectly interact with each other.
A powder is a particle system of solid particles in a gas or a vacuum which contains little or no free liquid.
If the liquid content is high, then, dependent on the amount, the system is called a paste or a slurry.

Since a powder is a set of individual particles truly identical powders will not be found. However, if the macroscopic behaviour of two powders is similar for all situations within certain close limits the powders are called identical. The relevant statistical averages of the properties of the two sets will be equal.

All systems with identical statistical averages can be joined into a group, called powder "X". The use of the term powder in this way corresponds with the use of the term material for molecular sets.
1.2 The scope of this research

This research is to investigate the effect of interparticle forces on the macroscopic powder behaviour. The microscopic approach to modelling the processes occurring in a particle system must eventually yield an explanation of the macroscopic phenomena on the boundaries of the system. To limit the scope of this work a number of subjects have been selected which are related to the concept of cohesion.

Two questions in particular dominate this work:

- what is the relationship between the interparticle forces and the cohesion,
- how does cohesion relate to other macroscopic properties, are influenced by the interparticle forces.

1.3 cohesion

Cohesion is a generally accepted indicator of the mechanical behaviour of a powder. However, contrary to field of non-disperse systems no unique definition of Cohesion is possible for powders. Also at the microscopic level of interparticle forces no parallels with intermolecular forces can be drawn. Cohesion in particle systems is defined operationally and the magnitude of the parameter is dependent on the process in which it is determined.

The strict distinction between cohesion and adhesion, which can be made for materials, is based on the phenomenon of similar phases or dissimilar phases attracting each other. This distinction cannot be made for the particles in a powder. One way to define these terms in relation to powders is to apply adhesion to the situation of small particles clinging to a large one and cohesion to the attraction of particles of similar size. This allows for a gradual transition between the concepts. Particles clinging to a wall of identical material can thus be said to be adhering.

In this thesis cohesion is defined by the flow behaviour of a powder. The maximum resistance against shear, whilst the powder is not normally loaded, is by definition the powder cohesion.
Fig 1.1: The relationship between the different topics. Characterisation of particle morphology, and the distribution of interparticle forces are building blocks for microscopic models and simulations. The microscopic predictions should be compared to the macroscopic results of powder testing equipment. Numbers refer to the chapters where these topics are addressed.
1.4 Synopses

The basic idea in adopting a microscopic approach to powder behaviour is that it can be used to explain all the macroscopic behaviour when the individual responses of any two contacting particles in the set are known.

At this moment a deterministic model of an entire set is not feasible, but to the opinion of the author the basic techniques which will enable such approach need to be examined and brought to a point where they can readily be implemented.

A statistical model is less ambitious but here also the problems are overwhelming. When exact modelling is difficult an approximate model based on distributions and assumptions which approach generally the relevant behaviour of submechanisms may also prove to be valuable.

A number of sub-problems are identified and studied. The hierarchical flow of information of one subject to the other is schematically shown in fig 1.1. The chapters in this work ordered sequentially according to this scheme.

Chapter 2.: Morphology. Any more detailed study of the possible interactions between particles must include the morphology of the particles. A convenient way to represent this morphology will be discussed. This may aid relating computer simulations of the processes occurring during deformation of particle sets to real particle sets.

Chapter 3.: The interparticle forces are both attractive and frictional forces. Since the form of the distribution of these forces is the basis for all subsequent models, experiments have been conducted to select the most suitable.

Chapter 4.: A theoretical analysis is made to relate the distribution of the interparticle forces to the macroscopic forces on the boundaries.

Chapter 5.: In a stress field in which one of the principal stresses is negative the mechanism of yield is experimentally examined.

Chapter 6.: The tensile strength of an agglomerate is believed to be related to its cohesion. Experimental work is done to clarify the mechanisms which determine the tensile strength as determined by a tensile test.

Chapter 7.: An analysis of the mechanisms which define the yield locus is carried out. This model relates directly to the concept of cohesion. Experimental work in a coaxial shear test show the development of shear planes.
Chapter 8.: When the particles are not compacted but aerated, the flow behaviour is influenced by fluid drag forces. The relationship between the interparticle forces and the cohesive behaviour in these systems is of interest.

Chapter 9.: Conclusion.
Chapter 2.: Particle Characterization

2.1 Introduction

Any model which will eventually successfully explain and predict powder behaviour must consider the diversity of size and shape of the particles within the powder. The analysis of particle morphology is, to the opinion of the author, a main building block to provide detailed information for microscopic simulations and models. The different methods of characterization are here examined with that particular goal in mind.

Two fundamentally different methods of particle characterization can be distinguished: mathematical description of their morphology and characterization by their interaction with some environment. The latter method measures a parameter of the particles which is most closely related to the process under consideration, if the proper interaction is chosen. However, such parameters lack generality, are specific to one process and cannot easily be extended to a different process. The parameters are therefore limited and can be used only as a general indicator or, more strictly, if the process and the measuring method coincide.

A mathematical description of the morphology is general, but a calculation of the interaction in a process directly from the morphology of each separate particle has only been done for the simplest of interactions, such as spheres settling in a Newtonian fluid. Nevertheless, a general description will gain more acceptance as more powerful computers can do the enormous amount of work necessary to calculate the mean interaction of a given morphology. To start this approach first of all a mathematical description of the particle form is needed. Meloy [16] offers the following opinion:

"What is needed is a theory of Particle Morphology which not only defines, unequivocally and mathematically, the particle size, its aspect ratio, but also yields a wealth of information about the particle, which in turn can be used to predict the physical properties of the particle..."

2.2 Characterization by chemical/physical interaction

The physical/chemical characterization methods which are currently used have recently been reviewed and their advantages and disadvantages discussed by Barth[2], Beddow[3] and Davies[7]. Earlier papers were published by Allen[1], Kaye[13], and Pahl[20].
When the particle size is calculated from its behaviour in a particular process, this length parameter is most suitable to predict the behaviour in a similar situation. Care should be taken not to assume that the interaction size is a real physical length, or that this size can be used to accurately predict the behaviour in a different process. For instance, a sieve diameter can not be used to predict the diffraction pattern.

Using a stylus a contour can be constructed around the perimeter of a particle. Using different radii of the tip of the stylus different contours for one perimeter will be found. The length of the contour will increase as a smaller radius can follow the fluctuations in the perimeter more closely [23].

Mandelbrot compared this phenomenon to the mathematical concept of self similarity. Kaye [13] has applied it to particle outlines. The analogy is not complete. For mathematically defined fractals on each scale of scrutiny the same underlying complexity is found. For real particles no true self similarity may exist: on different scales different processes may determine the complexity, and when coming to the level of resolution of the method with which the perimeter has been obtained, the complexity disappears. Correlations of the log of the length of perimeter against the log of the sampling length will be scatter plots in which only local trends can be discerned.

The subjects of interest related to cohesion are the effects of morphology on adsorption of water, on interparticle force, on the formation of contact areas and the space a particle needs to move about. For these processes different relevant contours may be important. Looking at this last aspect, as with the stylus, the particle contour will be dependent on the geometry of the interacting particle. Small particles are able to penetrate concave portions of the particle when large ones cannot. The physical tests available at this moment do not test on the average relevant interaction size directly. The results of the tests done to characterize the powder samples in this chapter are of neccessity only size indicators.
2.3 Characterization by morphology

The characterization of a particle by describing its morphology is complicated and requires a lot more effort than using the physical interaction. The advantage is that the result is general and can, theoretically, lead to calculation of any interaction parameter.

In order to reduce the amount of raw data involved with particle contours data reduction is of paramount importance. This inevitably causes loss of information and care must be taken to preserve the features relevant for the process in consideration. In this section a number of data reductions shall be discussed.

2.3.1 Form and process of formation

Dependent on the process of formation and the nature of the material a number of characteristic contours may be expected. We may distinguish between primary processes which lead to the formation or primary particles and subsequent secondary processes which modify the form of those primaries or build larger particles using the primaries as basic units. Some processes are briefly reviewed here with respect to the kind of surfaces and contours they would produce.

Some primary processes of formation are:

1) Crystallization
   Due to their regular form, contours of crystals will be polygonal.

2) Surface minimization due to surface tension
   Because of surface tension non crystalline particles tend to have curved smooth forms.

3) Chemical formation
   Chemical growth may form highly irregular surfaces.

Some secondary processes of formation are

4) Breakage
   The profiles of particles created by breakage can be described by a random walk model. Although the majority of the faces are straight they may also be curved.

5) Attrition and abrasion
   Attrition may be defined as small scale breakage which is limited to the outer layer of material of the particle. This process modifies the overall form mainly by smoothing sharp angles formed between large faces. Abrasion is limited to the surface of the particle and changes the small scale rugosity of the skin. The effect is usually smoothing but pitting may also occur.
On Fig 2.1: The form of a contour can be described by the set of vectors pointing at the atoms in the contour.

6) agglomeration
The process of agglomeration creates complex particles. The agglomerates combine the characteristics of the primary particles and the geometry of their three dimensional structure. Projected contours of these agglomerates are also complex and may show artifacts such as sharp reentrant angles, which are no features of the three dimensional form. Agglomerates are best represented by the description of the constituting particles and their relative position in the agglomerate.
2.3.2 Morphology as a data set.

The most general definition of the morphology of a particle is the set of coordinates and forms of all atoms constituting the particle. Assuming that the form of the atoms can be described by simple mathematical surfaces, the entire form of the whole particle can be constructed using this set.

This definition is not a practical one for daily use, since the amount of parameters to store exceeds the limits of memory of ordinary computers. A suitable data reduction must be made. This procedure has to reduce the amount of data as far as possible without appreciably changing the form of the particle, so that the magnitude of the interaction of interest is altered.

The first reduction can be obtained by neglecting the exact description of the position and form of the interior atoms and taking only the outer. See fig 2.1. Interior atoms are those atoms that are not accessible from the outside using an other atom as a theoretical probe.

To measure the actual coordinates of the atoms is a tedious matter. A more convenient approach is to assume that a few representative points on the perimeter, and many atom distances apart, will do as well for the process in consideration.

A more severe step is to use a set of two dimensional projections or slices of a three dimensional object. The projection is a convolution and causes an irretrievable loss of data and may cause artifacts unless a model for the shape is already available. The accuracy of the reconstruction of the three dimensional shape is dependent on the number of projections considered.

A two dimensional projection of the object can also be split into inner and outer portions. Only the contour of the projection is used. For further reduction a high resolution contour can be sampled and stored as a low resolution contour. A number of methods to sample are shown in fig 2.3. Each results in a different set of points on the contour, but if the density is high enough the relevant features of the contour can be captured in all representations.
2.3.3 Mathematical description of the contour

A number of sets can be defined to describe the sample points on the contour, see fig. 2.2. All the sets used in the following discussion are cyclic. Four systems are discussed which may be useful for different applications:

1) the absolute screen coordinate set:

\[
\text{Geometry} = \{ \xi_0, \xi_1, \ldots, \xi_n \}
\]

\[
= \{ \xi_n, \xi_0, \xi_1, \ldots, \xi_{n-1} \}
\] \hspace{1cm} \ldots(2.3.1)

or in short notation:

\[
\text{Geometry} = \{ \xi_i \}
\] \hspace{1cm} \ldots(2.3.2)

The subscript numbers indicate the successive elements along the contour. The numbering is anti clockwise.
2) A set, which is invariant and independent of the choice of the origin, contains the vectors pointing to the nearest anti clockwise neighbour:

\[ \delta \mathbf{r}_i = \mathbf{r}_{i+1} - \mathbf{r}_i \quad i = i \text{ modulo } n+1 \quad \ldots(2.3.3) \]

Geometry = \{ \delta \mathbf{r}_i \} \quad \ldots(2.3.4)

Since the contour is closed the sum of elements must be zero:

\[ \sum_{i=0}^{n} (\delta \mathbf{r}_i) = 0 \quad \ldots(2.3.5) \]

The length of the perimeter is the summation of the magnitude of all the vectors:

\[ \text{perimeter} = \sum_{i=0}^{n} (|\delta \mathbf{r}_i|) \quad \ldots(2.3.6) \]

The shape of a particle can now be defined as the relative geometry obtained by normalizing the relative geometry set with some absolute length. This length parameter can be chosen as the square of the enclosed area of the particle contour or the length of the perimeter or a Feret diameter for example. A length based on area is less sensitive to choice of scale as a length based on the perimeter.

\[ L = \sqrt{\text{area}} \quad \ldots(2.3.7) \]

\[ \delta s_i = \delta \mathbf{r}_i / L \quad \ldots(2.3.8) \]

\[ \text{shape} = \text{geometry}/L = \{ \delta \mathbf{r}_i/L \} = \{ \delta s_i \} \quad \ldots(2.3.9) \]

Local features can be defined to have similar shape by the connecting the endpoints of a subset of subsequent vectors to form a new subparticle and dividing the vectors by the length which will normalize this area.

3) Some other sets are useful in the characterization of a particle shape: the angles between successive vectors, and the length of the vectors. These sets can be used for statistical analysis.

\[ \cos(a_i) = \frac{\delta \mathbf{r}_i \cdot \delta \mathbf{r}_{i+1}}{|\delta \mathbf{r}_i||\delta \mathbf{r}_{i+1}|} \quad \ldots(2.3.10) \]

Angle = \{ a_i \} \quad \ldots(2.3.11)

Length = \{ |\delta s_i| \} \quad \ldots(2.3.12)
Fig 2.3: Four sampling methods to reduce the amount of data

- a. Circumcising method
- b. Line scanning at regular distances
- c. Polar scanning at regular angles
- d. Grid sampling

Fig 2.4: Chain coding. The limited number of orientations allows considerable data compression.
4) An alternative set can be used for crystals. This set contains the distance of a face to the centre of gravity and the relative angle of the normal vector of the face:

\[
\text{Geometry} = \{ h(i) \} \quad \{ \phi(i) \} \quad \ldots (2.3.13)
\]

According to the rule of Wulff the quotient of the surface energy and the distance of a face are constant if the crystal is in equilibrium with its surrounding. Defay [8] discusses this situation.

2.3.4 Data reduction.

2.3.4.1 sampling a two-dimensional image to obtain the contour.

Once a projection of the object has been made the contour must be extracted.

Several sampling methods to obtain estimates of the contour points are used, for instance:

- scanning the picture with lines at a regular spacing
- walking methods
- grid sampling

Each results in a different set of points, irregularly spaced on the contour or, in the case of grid sampling, close to the contour.

Since grid sampling is the most popular method used in modern image analysis instrumentation this technique will be discussed in more detail.

Contours on Pixel areas.

To obtain an image on a pixel field, the light intensity of the image is digitized, in a limited number of grey values, and sampled at regularly spaced points on a plane. An object in the field can be detected if the grey values of the pixels belonging to the object are different from that of the background. Thus the entire grid can be divided into objects and background.

Again inner and outer pixels can be distinguished within the pixels of the detected object. Connecting the outer points results in a contour, which will be located within the proper projection. Connecting the points just outside the object will yield a second, and larger 'outside' contour. The true contour is located within the object defined by the inside and outside contour.

If the difference in length of the inner and outer contour is small each can be used as a good estimate of the true contour. If the difference is large, as can be the case for objects that are
Fig 2.5: contours of coarse (Above) and fine (Below) sand
represented with only a few pixels, an intermediate contour has to be estimated. If the image analyser does not contain this software algorithm, care has to be taken to represent the boundary of an object with sufficient pixels.

The data representation of a boundary in a grid field can be very compact, since only a limited number of different vectors of one pixel to the adjacent pixel are possible. For a square grid the number is possible orientation is eight, which enables representation of the vector with only one byte. See fig 2.4.

2.3.4.2 linearization of the contour.

The easiest way to represent the contour with limited data is to linearize it locally. Only the end points of each linear piece need to be stored. The linearization procedure can take various forms. Two main methods can be distinguished according to the limitations set on the line piece:

a) start and endpoint each on the contour
b) start and endpoint not restricted.

In order to determine whether a part of the contour should be replaced, statistical trend procedures or least square methods can be used. Fast but less sophisticated procedures use fixed size of lines or a fixed number of lines.

2.3.4.3 basic polygons

Looking at a rough shape, intuitively one sees a smooth simple form and superimposed fluctuations. The fluctuations are considered to be roughness, the simple form being the true basic form of the particle, see fig. 2.5. If the basic shape is a polygon, the least squares method can be used to reconstruct it assuming that the rugosities are random fluctuations around the basic shape. Even when the basic form is not polygonal, an approximate polygonal basic shape may be a sufficient representation for the process of interest.

Having obtained the basic polygon, characteristic parts of the contour may be examined in more detail. The polygon may help focusing attention on the proper part. For instance, the sharpness of an angle, indicated by the angles of the polygon, can be investigated with a more sophisticated function.
2.3.4.5 A fast algorithm for fitting a basic polygon

In order to transform these ideas into an algorithm, two problems must be solved.

The contour is not a function, and the minimum distance of a point on the contour to another contour can not be used in a least square criterion. A procedure has to be decided on which allocates a point on the contour to a side of the approximating polygon.

A computer time problem may arises when one tries to fit a set of high-resolution contours using a least square algorithm. If the program has to be applied to a sample of several hundred particles, the time needed for one particle may not exceed a few seconds. This limitation can only be met by a fast computer, a fast algorithm, not too many points on the contour or preferably a combination of these.
the basic polygon

The polygon which represents the basic shape of the particle is the lowest order polygon which approximates the original with an acceptable fit. The acceptability of the fit is dependent on the level of detail that has to be preserved. For each level of discrimination a different basic form may be defined. If no a-priori information about the roughnesses is available step wise reduction of the number of sides, until it is found that further reduction increases the error of fit substantially, appears to be appropriate. Since the polygon must be simple a maximum number of sides must be set, and a number of ten sides appeared to be sufficient to represent the most complex particles.

the fit algorithm

In order to gain speed we have chosen an approximation of the first kind: beginning and endpoint of each line section are located on the contour. This permits a simple procedure to allocate points on the contour to the sides of the polygon: all contour points between the intersections belong to the intermediate line section. The distance between this line section and points on the contour can now be used in an optimising procedure, see fig 2.6. In this work a least square criterion has been used.

The starting position

Various procedures to initiate the starting position can be designed, see 2.7.

1) The contour can be divided into a fixed number of line sections, each section containing equal numbers of points on the contour.

2) Using a criterion for the variance of the contour around the line section, the length of an initial side is determined. Starting from the last point allocated to this line section, the procedure is used again to find an second line section and so on until all points on the contour are processed.

Although this option may not be the fastest, in this work the second method has been used. A low variance criterion was doubled until a initial polygon of a order less than ten was found. This polygon was then used for optimization.
Fig 2.7: Four methods to define an initial approximate polygon

a  trend detection, points on the contour  
b  trend detection, points not restricted  
c  equal number of sample points  
d  equal lengths of polygon sides

Optimization

Once the starting position has been found, the minimum sum of residues can be found iteratively. Consider an intersection of two lines at point $j$. For both a shift of the intersection to a point before and after $j$, the sum of squares for these two lines alone is calculated. Of the three possible positions the intersection which has the lowest residue is chosen, see fig 2.8. Two adjacent intersection points can not be brought together closer and this condition has to be recognised. This procedure is applied in turn to all intersections until no change occurs any more.
mutations

During the optimizing procedure the number of sides does not change. However frequently it appears that after optimizing two neighbouring line sections are aligned and can be replaced by one larger.

Using the following criterion the number of line sections has been mutated:

\[ | l_1 | + | l_2 | \leq \text{crit}^* | l_1 + l_2 | \quad ...(2.3.14) \]
\[ 1.00 < \text{crit} < 1.05 \quad ...(2.3.15) \]

Setting the criterion to 1.05 eliminates all adjacent intersection points. The elimination of a side results in a non optimized polygon and the complete algorithm can be restarted again.

This mutation appeared to be very successful in finding the suitable polygon of lowest order. If the magnitude of the criterion is higher, lower order polygons are accepted.

performance

The algorithm has been implemented on a PC with a clock rate of 8 MHz, using Turbo Pascal. After compiling the program needed for the most complex cases 15 sec. to find the optimum polygon for a contour of 100 points. It is expected that after optimizing the algorithm and the programming on speed the process time can be further reduced to well within 5 sec.
Examples of complex forms analysed with polygons are shown in fig. 2.9 and 2.10.
Fig 2.8: After comparing the lack of fit for three positions of the central point of two linepieces, the central point is moved one position in the direction of the best fit.
Fig 2.9: A set of similar particles with different roughnesses. Fitting may yield different approximate basic forms as more variation is accepted.
Fig 2.10: Composite forms with different roughnesses
2.3.4.5 parameters derived from a polygonal form

All parameters commonly derived from the shape of the object can also be derived from its equivalent basic polygon. Because the geometry has been simplified fast calculation is possible.

For example:

The surface area:

\[
\text{Area} = \frac{1}{2} \sum_{i=0}^{N-1} (x(i)y(i+1) - x(i+1)y(i)) \quad i := i \mod N \quad \ldots \text{(2.3.16)}
\]

The centre of gravity:

\[
K(i) = \frac{x(i)y(i+1)}{6} + \frac{x(i)y(i) + x(i+1)y(i+1)}{3} + \frac{x(i+1)y(i)}{6} \quad \ldots \text{(2.3.17)}
\]

\[
X_{\text{cog}} = \sum (x(i) - x(i+1)) \cdot K(i) / \text{Area} \quad \ldots \text{(2.3.18)}
\]

\[
Y_{\text{cog}} = \sum (y(i) - y(i+1)) \cdot K(i) / \text{Area} \quad \ldots \text{(2.3.19)}
\]

The moments:

Van Otterloo [19] published a fast calculation method for the moments of a polygon:

\[
M_{pq} = \sum_{\alpha=0}^{p+1} \sum_{\beta=1}^{q+1} d(\alpha, \beta; p, q) \cdot T_n(\alpha, \beta; p, q) \quad \ldots \text{(2.3.20)}
\]

\[
d(\alpha, \beta; p, q) = \frac{1}{(p+1)(q+1)} \cdot \frac{\beta}{\alpha + \beta} \cdot \left| \frac{p+1}{\alpha} \right| \cdot \left| \frac{q+1}{\beta} \right| \quad \ldots \text{(2.3.21)}
\]

\[
T_n(\alpha, \beta; p, q) = \sum_{n=0}^{N-1} (\delta x_n) \cdot x_n \cdot (\delta y_n) \cdot y_n \quad \ldots \text{(2.3.22)}
\]

\[
\alpha, \beta = \text{running variables}
\]

\[
p = \text{order of moment in x direction}
\]

\[
q = \text{order of moment in y direction}
\]

\[
\delta x = x_{n+1} - x_n
\]

\[
\delta y = y_{n+1} - y_n
\]

\[
x_n, y_n = \text{coordinate of contour point n}
\]

24
Fig 2.11: The rolling resistance of a particle. The maximum angle of stable tilt is 90-\( \alpha \).

rolling angle

A particle can be characterized on its rolling ability. If it is not perfectly spherical, a stable position can be found in which the particle resist rolling down an inclined plane. The maximum angle at which a particle is stable can be used as a characterizing parameter. See fig 2.11.

For a polygon this angle can be determined easily. The vector from the centre of gravity to each corner of the polygon divides the angles into two parts. The smallest of these angles is the complement of the static rolling angle.

To continue the rolling translation enough energy must be present to lift the center of gravity over the dead point. This minimal energy can also be calculated once the angle between plane and centre of gravity is known.
fig 2.12: Two methods of surface characterization using the amplitude distribution and the autocorrelation function: (a) skew zero, simple random, typical of a ground surface, (b) skew negative, simple random, typical of a lapped surface, (c) skew positive, modulated random, typical of a bead-blasted surface, (d) skew zero, complex correlation, typical of a milled surface, (e) skew positive, random plus periodic, typical of a turned surface, (f) deterministic, etched or ruled standards. Whitehouse(1974).
The angularity of a particle is a measure of its susceptibility to breakage and attrition. One characterization of abrasiveness or flowability could well be based on various parameters of the angularity of the particle. The basic form already indicates the overall angularity of the various angles. A more accurate description can be found when the points on the contour, in the neighbourhood of an angle, are fitted to a parabola or an other suitable function.

2.3.4.6 Surface characterization.

Having obtained the basic shape of a particle the fluctuations around the sides of the polygon represent the roughness of the particle. The contour of the particle can be unrolled and mapped onto a line. This line can be characterized using the methods developed for surfaces.

Whitehouse [23] discusses a number of classifications for the recording of roughnesses by using a stylus. This classification is based on:

- the distributions of the height of the roughnesses
- an autocorrelation function of the fluctuations

Dependent on the type of skewness of the distribution and its periodicity as indicated by the autocorrelation 5 different groups can be defined. These groups are related to history of formation, see fig 2.12. Other possible classifications are based on the sum of squares of the heights of the roughnesses or on the power spectrum. Both the autocorrelation function and power spectrum are based on the Fourier transforms of the signal. The merits of these analyses and the way they should be implemented in computer algorithms are discussed extensively by Press et al. [21].

The connection between the sum of squares and the coefficients of the Fourier series is given by the Parcefal equation for discrete signals, which states that the sum of squares is equal to the average energy in the signal.

\[ \sum_{i=0}^{N-1} h_i^2 = \frac{1}{N} \sum_{\Omega=0}^{N-1} H(\Omega)^2 \quad \ldots \quad (2.3.23) \]
Fig 2.13: The projection of the contour onto a line yields the Feret diameter. Minimum and maximum diameter can be defined, often the breadth is taken perpendicular to the length. The projected lengths need not be equal to actual physical dimensions.

Fig 2.14: Comparison of Feret length and breadth of a circular and square outline. The ratio of $F_l\times F_b$ and $A$ is smaller for rounded forms.
2.3.4.7 Feret Diameters

An absolute length parameter is derived from the contour using one dimensional Feret projections. If the two dimensional projection is, in turn, projected onto a line in the plane, the length of the projection is the Feret length. This parameter depends on the orientation of the line. See fig. 2.12.

The Feret length is the maximal projection. The Feret breadth is the length of the projection onto a line which is perpendicular on the line of maximal projection.

The statistical mean of all Feret projections of a convex contour is related to the length of the perimeter:

\[
\text{Perimeter} = \pi \times \text{Feret mean} \quad \ldots(2.3.24)
\]

Transferring these formulas to three dimensions the projected areas can give an estimate of the surface area of the three dimensional particle (Kaye [13]).

2.3.4.8 Length ratio's

As an alternative to a complete description of the contour only a few characteristic ratio's of lengths can be determined.

**Elongation**

The elongation of a particle is calculated by dividing the maximum Feret length of the particle with its corresponding breadth. For circular outlines this ratio is 1.

\[
\text{Elongation} = \frac{F_l}{F_b} \quad \ldots(2.3.25)
\]

**Angularity indicator**

The multiple of the Feret length and breadth of any triangle or rectangle is equal to twice its area. This ratio decreases steadily for regular polygons, with the number of sides. For the limiting case of a circle the ratio is \(4/\pi\). This latter ratio also applies to all ellipsoids.

Generalizing it can be proposed that the ratio indicates the sharp, triangular or the smooth, ellipsoidal character of a particle. If the particle is smoother the ratio is smaller. See fig. 2.14.

An exception must be made for shapes like thin prismatic needles: in the limiting case the ratio can be 1. Since these particles would also considered to be sharp, classification based on the ratio must account for this case.
table 2.3.1: angularity indicator as a function of the order of a regular polygon.

<table>
<thead>
<tr>
<th>order of the regular polygon</th>
<th>Fl*Fb/Ap</th>
<th>form</th>
<th>range</th>
</tr>
</thead>
<tbody>
<tr>
<td>3,4</td>
<td>2.0</td>
<td>sharp</td>
<td>1.7 - 2.0</td>
</tr>
<tr>
<td>5</td>
<td>1 + 0.2/5</td>
<td>semi-sharp</td>
<td>1.4 - 1.7</td>
</tr>
<tr>
<td>6</td>
<td>4/3</td>
<td>semi-round</td>
<td>1.3 - 1.4</td>
</tr>
<tr>
<td>∞</td>
<td>4/π</td>
<td>round</td>
<td>1.2 - 1.3</td>
</tr>
<tr>
<td>infinite prismatic needle</td>
<td>1</td>
<td>sharp</td>
<td>1.0 - 1.2</td>
</tr>
</tbody>
</table>

No one ratio can distinguish all forms. For example, the angularity ratio fails to circles and half circles. Also equal-sided trapezia are classified as less sharp which is intuitively misleading.

2.3.4.9 equivalent forms

The complex projection can be replaced with an equivalent area of a simple form. This simple form can be represented with only a few parameters.

The following forms are most popular:

1) rectangles or boxes

Rectangles can be represented by length and breadth, boxes require also height. These length parameters are extracted from the original form. For instance, using the maximum Feret length and the corresponding height and breadth, a box is defined which encloses the particle and has the greatest length. Other definitions seek for highest elongation, minimal volume or most stable position (Heywood[11]). Best fitting or equal moments of rotation may also be used.

2) Ellipsoids and circles.

All procedures used for rectangles can be applied to circles as well. (see e.g. Medalia [15]). Circles are especially attractive since they are an one parameter form. Kaye[13] gives a resume of many applicable procedures.

The difference between the equivalent form for one property and an equivalent form, calculated using another property, is informative. The ratios are called form factors. A number of these form factors are based on the area and the length of the perimeter of the projection. Using equivalent circles the
following radii can be derived:

\[
R_1 = \text{Perimeter} / 2\pi \quad \text{...(2.3.26)}
\]
\[
R_2 = \sqrt{\text{Area} / \pi} \quad \text{...(2.3.27)}
\]

Using these two radii a number of shape factors can be defined, such as:

\[
\text{Shape factor}_1 = R_1 / R_2 \quad \text{...(2.3.28)}
\]
\[
\text{Shape factor}_2 = R_1' / R_2' \quad \text{...(2.3.29)}
\]

For a sphere the projection is circular and the shape factors are equal to 1. If a shape factor for any particle is different from one the particle is not a sphere.

3) equivalent polygons.

The polygons fitted to the contour can also be considered as equivalent forms. After fitting the size can be adjusted to match area or Feret length. Since the polygonal form will also preserve information about the contour itself it is a superior description for simulating the interactions of particles in a particle set.

2.3.5 Spherical harmonics.

A current device to characterize the contour of an particle is the transcription of the contour into its spherical harmonics. [4,9,10,14,16,17,18,19,22]. This method has gained attention because of the success of the Fast Fourier Transshape algorithms in other field of signal processing.

To apply a FFT the contour is seen as a periodic signal in a polar coordinate system. The choice of the origin of this system is problematic since the coefficients are not invariant. The centre of gravity is usually used. If the outline has reentrant features a different approach will have to be followed to avoid problems with multiplicity. The sine and cosine series in those systems are non-spherical.

When a discrete periodic signal can be obtained this signal can be expressed as a Fourier series:

\[
N
f(\theta) = \sum_{n=0}^{N} a_n \sin(n\theta) + b_n \cos(n\theta) \quad \text{...(2.3.30)}
\]

or

\[
N
f(\theta) = \sum_{n=0}^{N} a_n \left( \cos(n\theta + \phi_n) \right) \quad \text{...(2.3.31)}
\]

This transcription does not add any information to the signal but the coefficients are believed to be more suitable for further analyses.
analyses of the coefficients

Luerkens et al. [3,14] discussed the problem of the non-invariant Fourier coefficients and derive a number of compounded descriptors, which are invariant with respect to the choice of origin. The result is the area, a normalized power spectrum and the second and third moments of the polar function. Other relationships with commonly used parameters are difficult to find; for example, no analytical relation is known for the length of the perimeter and the Fourier coefficients.

Using the invariant descriptors particle sets can be discriminated, but the classical parameters would do as well. It is the opinion of the author of this thesis that all the applications suggested by Luerkens for spherical harmonics can be used, would be adequately achieved using the polygonal approach.

the power spectrum

On first sight the power spectrum of the harmonics appears to be useful. A number of arguments will be brought forward to show that the use is very limited.

To obtain the spectrum a Fourier transform must be carried out. Using FFT techniques special care must be taken that aliasing does not interfere with the higher harmonics. These might appear to have higher energy content than really exist.

Meloy [16,17] has determined the power spectrum of the cosine series for a number of shapes and finds a scatterplot with a declining trend. This trend is to be expected since the resolution of the contour is limited and cannot contain infinite energies.

According to Meloy this trend is linear on a log-log scale, and he suggests that the slope may represent a fundamental property of the contour.

Analytically, it can be shown that linearity is not present for simple shapes like circles on which a polar triangular or rectangular pulses have been superposed to simulate a single rugosity. Meloy's assumption is, therefore, not generally valid.

An easy but questionable assumption is that the higher frequencies in the spectrum represent the roughnesses, the lower the basic shape.

1) If only a limited spectrum of low frequencies is used to describe the basic shape of a particle, the family of basic shapes is limited to a small set with smooth contours. Analysing particles which do not have smooth basic shapes will result in a spectrum with higher harmonics as well. This can readily be shown by transforming a simple cube. The higher harmonics information arises from both the roughnesses and the
basic shape and cannot be differentiated.

2) When it is assumed that the energy of a spherical harmonic represent the roughness, it is also implicitly assumed that the mechanisms which formed the contour are homogeneous or identical for the entire perimeter. This is true only in a limited number of cases.

The power spectrum of the raw polar data is for these reasons a limited instrument.

Meloy commented that for some shapes it would be better to determine the basic shape, which he called the melamorf, and subtract that shape from the signal. At that time no easy routine to determine the basic shape was available, and the polar representation of a shape does not enable recognition of linear parts in the contour.

Thus using the routine to determine the basic polygon, which has been developed here, the roughnesses may be extracted from the original shape and analysed according to the methods established for planes.

The safe approach would be to analyse each side of the polygon separately and avoid the outer parts of the sides, since the approximation near the intersections of the faces with the contour are most likely to be distorted.

Recently the thesis of van Otterloo[19] has been published. The possibility for detecting symmetries using the Fourier transform is discussed. This work contains a very extensive review and a thorough mathematical basis for the manipulation of contours.

He defines a measure of similarity between contours which is based on corresponding points on both contours. This technique has a number of difficulties and it is not visibly superior over the method of using the distance between contours pieces which was adopted in this work.

Van Otterloo concludes that the harmonics can only contain global information and do not describe local effects. This agrees with what has been discussed above.
2.4 Experimental Section

In the investigation of the properties of powders the following materials have been used:

- fine sand
- course sand
- fine alumina
- course alumina
- limestone

Fig 2.15: The particle size distribution of a coarse alumina powder (bottom) and a fine alumina powder (top).
Fig 2.16: The particle size distribution of the fine sand.

2.4.1 Characterization by Physical Interaction

Table 2.4.1: Equivalent sphere diameters determined by laser diffraction analyses, instrument Malvern, cumulative weight percentage, see fig 2.15,

<table>
<thead>
<tr>
<th></th>
<th>M (w%)</th>
<th>Dp (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fine sand</td>
<td>10</td>
<td>86.3</td>
</tr>
<tr>
<td></td>
<td>50</td>
<td>137.3</td>
</tr>
<tr>
<td></td>
<td>90</td>
<td>209.5</td>
</tr>
<tr>
<td>Course alumina</td>
<td>10</td>
<td>33.5</td>
</tr>
<tr>
<td></td>
<td>50</td>
<td>59.3</td>
</tr>
<tr>
<td></td>
<td>90</td>
<td>102.4</td>
</tr>
<tr>
<td>Fine alumina</td>
<td>10</td>
<td>2.6</td>
</tr>
<tr>
<td></td>
<td>50</td>
<td>9.4</td>
</tr>
<tr>
<td></td>
<td>90</td>
<td>35.1</td>
</tr>
</tbody>
</table>

Table 2.4.2: Equivalent sphere size determined by sedimentation analysis, Andrease pipette, cumulative weight percentage. see fig 2.17

Limestone

<table>
<thead>
<tr>
<th>M (w%)</th>
<th>Dp (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>1.8</td>
</tr>
<tr>
<td>50</td>
<td>4.0</td>
</tr>
<tr>
<td>90</td>
<td>6.2</td>
</tr>
</tbody>
</table>
Fig 2.17: Particle size distribution of the limestone powder

Fig 2.18: Adsorption isotherm for the limestone powder
2.4.2 Morphological Characterization

The Feret diameters and object area were determined by pixel field analyses.

Fl = maximal Feret length
Fb = Feret breadth corresponding to Fl
Ap = area

<p>| Table 2.4.3: Fine sand, based on 131 particles, see fig 2.5, |
|-----------------|-----------------|----------------||-----------------|-----------------|</p>
<table>
<thead>
<tr>
<th>Fl (μm)</th>
<th>Fb (μm)</th>
<th>Ap (μm²)</th>
<th>Fl/Fb (-)</th>
<th>Fl*Fb/Ap (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>mean</td>
<td>174</td>
<td>132</td>
<td>1.60e4</td>
<td>1.35</td>
</tr>
<tr>
<td>s.d.</td>
<td>41</td>
<td>32</td>
<td>0.67e4</td>
<td>0.27</td>
</tr>
</tbody>
</table>

<p>| Table 2.4.4: Coarse sand, based on 155 particles, see fig 2.5 |
|-----------------|-----------------|----------------||-----------------|-----------------|</p>
<table>
<thead>
<tr>
<th>Fl (μm)</th>
<th>Fb (μm)</th>
<th>Ap (μm²)</th>
<th>Fl/Fb (-)</th>
<th>Fl*Fb/Ap (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>mean</td>
<td>299</td>
<td>224</td>
<td>4.65e4</td>
<td>1.36</td>
</tr>
<tr>
<td>s.d.</td>
<td>68</td>
<td>47</td>
<td>1.74e4</td>
<td>0.28</td>
</tr>
</tbody>
</table>

<p>| Table 2.4.5: Coarse alumina, based on 231 particles, see fig 2.19 |
|-----------------|-----------------|----------------||-----------------|-----------------|</p>
<table>
<thead>
<tr>
<th>Fl (μm)</th>
<th>Fb (μm)</th>
<th>Ap (μm²)</th>
<th>Fl/Fb (-)</th>
<th>Fl*Fb/Ap (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>mean</td>
<td>67</td>
<td>54</td>
<td>2.87e3</td>
<td>1.24</td>
</tr>
<tr>
<td>s.d.</td>
<td>22</td>
<td>19</td>
<td>1.90e3</td>
<td>0.16</td>
</tr>
</tbody>
</table>
Fig 2.19: Particle contours of coarse alumina

<table>
<thead>
<tr>
<th></th>
<th>fine sand (%)</th>
<th>coarse sand (%)</th>
<th>coarse alumina (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>sharp</td>
<td>4.6</td>
<td>7.8</td>
<td>0.4</td>
</tr>
<tr>
<td>semi-sharp</td>
<td>77.1</td>
<td>65.1</td>
<td>42.8</td>
</tr>
<tr>
<td>semi-smooth</td>
<td>13.7</td>
<td>21.9</td>
<td>44.2</td>
</tr>
<tr>
<td>smooth</td>
<td>4.6</td>
<td>5.2</td>
<td>12.6</td>
</tr>
<tr>
<td>mean angularity</td>
<td>1.49</td>
<td>1.48</td>
<td>1.39</td>
</tr>
<tr>
<td>mean elongation</td>
<td>1.35</td>
<td>1.36</td>
<td>1.24</td>
</tr>
<tr>
<td>length</td>
<td>174</td>
<td>299</td>
<td>67</td>
</tr>
</tbody>
</table>
2.4.3 Discussion

The morphology of coarse powders has been determined using a microscope and image analyses. A few characteristic length parameters have been chosen to represent the shape. Ratios of these parameters are capable to yield limited information about the shape of a particle.

The analyses of the shape of the courser powders shows that the sand samples are very similar. The alumina is somewhat less sharp and the ratio of elongation lower.

When the particles are small, such as is the case for the BCR powder, the poor resolution of the microscope and the difficulty to obtain representative non-coagulated particles forces us to use the physical interaction methods. These methods are quick but yield convoluted information. The data can be transferred to a set of equivalent spheres, but this method will cause each method to produce its own equivalent size distribution.

2.5 Conclusion

In this chapter various ways of characterizing particles are discussed. A distinction is made between physical and geometrical methods. The physical methods yield convoluted data from which the original shape of a particle is difficult to extract. The geometrical method gives direct information about the absolute positions of representative points on the perimeter of the particle. Therefore, the geometrical method is believed to be more fundamental.

A proposal is made for a practical method for characterizing particle morphology, which, the author believes, contains enough information to enable future accurate simulation and description of packings of particles. In these packings the interactions of particles in shear and tensile fracture situations are to be examined by simulation. The characterization should, therefore, be able to capture the properties of a real powder in mathematical parameters and thus open the possibility to compare simulated and actual behaviour. This, to the opinion of the author, will be a mayor step forward and bring powder technology to a level more comparable with material science. It is recognized that this goal will not be achieved within a relatively short period, but much of the technology needed is already available and, anticipating further increases in affordable computer power, a ten years period seems not unrealistic. Accurate simulation of the effect of shape and size distribution and interparticle forces would permit design of powders, which is the ultimate goal.
Various methods to reduce the amount of necessary data to describe the particles at the level of relevance are discussed. An algorithm is described which is fast and robust and yields a polygon, which fits a 2 dimensional contour with minimal sides for a certain level of acceptable variation. This polygon has been termed the basic shape of a particle and the residual variation of the perimeter around this basic shape the roughness. This roughness can be further processed with the known methods for characterizing roughnesses of planes.

The rough polygon description of a contour is more powerful than the translation of the contour in spherical harmonics. This latter method yields global rather than local information of the shape and roughness of a contour. Furthermore all the parameters which are obtained from a spherical harmonics description can also be obtained from the polygons.

The basic polygon approach opens up a myriad of new ways to processing particle contour data. For example:

- fast reconstruction of 3 dimensional polyhedrons using multiple scans.
- local and directed investigation of specific features of a contour: asperities, sharp corners, pores etc.
- feature recognition/ primary particle recognition
- shape/object recognition
- shape changes
- real particle set simulations: deformations, shear zones, interparticle forces etc.
- data compression

The reconstruction of polyhedrons may be the most desirable property from the viewpoint of simulation. It should also be noted that a large volume of software, handling polyhedrons, is already available as that was developed for CAD applications.
2.6 Symbols

{}   set brackets  
A    area   \( [m^2] \)
a    angle \( [rad] \)
\( c \)  vector to contact  \( [m] \)
crit  fraction \([-]\)
D    diameter \( [m] \)
H    coefficient of power spectrum \([-]\)
g()   general function \( [] \)
h     height of roughnesses \( [m] \)
i,j   pointer \([-]\)
\( Mpq \)  \((p+q)\) order moment of an area \( [m^1(p+1)(q+1)] \)
n    unit normal vector \( [m] \)
R    radius \( [m] \)
\( \xi \)  vector to reference point \( [m] \)
t    unit tangential vector \( [m] \)
\( x,y,z \)  cartesian coordinates \( [m] \)

\( \theta \)   shear stress \( [Pa] \)
\( \Sigma \)  summation sign \( [] \)

subscripts

b    breadth
i    pointer
l    length
p    particle
n    pointer
2.7 References


3.1 Introduction.

A large volume of research has been published about the nature and magnitude of the fundamental forces between molecules and particles. Krupp[17] reviews and summaries many theories and different experimental methods. Zimon[30] published much experimental data and many theories, including Russian literature. Deryagin[7,8] investigated the role of electrostatic forces, arising due to electrical double layers. He also worked on the effect of the deformation of particles. Kingery[14] wrote a standard work in the field of ceramics and contains an enormous amount of information about the chemical side of the binding forces between particles.

In spite of all this vast amount of knowledge it is still impossible to predict accurately the forces between touching particles. The main problem preventing this is the lack of knowledge of:

- the exact magnitude of the molecular forces involved
- the exact morphology of the contact
- the mechanical properties of the material in contact.
- the influence of adsorbed or condensed layers.

Furthermore experimental work shows that an enormous spread, up to a factor of 1000, in the distribution of the total force can occur for particles of the same size. The form and spread of the distribution has not yet been satisfactorily explained.

In this chapter the interparticle forces, as they appear in multiparticle systems are summarized and investigated. The forces can be divided into normal separation forces, or more shortly the adhesion, and the tangential separation forces, the friction. The emphasis of this investigation is on the determination of the correct form of the distribution of the forces, the experimental research being limited to the adhesion.

A number of possible distributions are compared with the experimental results in order to determine the most likely. The distributions tested were the log-normal, the gamma and the Weibull distribution. Positive identification of the form of the distribution could assist further clarification of the processes involved. This, however, was not the case for the set of separation forces measured, and all the models appeared to be adequate, no single model being much better than the other. This being the case, for the subsequent modelling of the forces
in multi-particle environments we have chosen the Gamma model, which proved to be the most convenient for mathematical purposes.

### 3.1.1 Molecular forces of interaction.

Almost any text on surface chemistry discusses the forces between molecules and these intermolecular forces are now briefly summarized. See eg. Gregg[9] or Ponec[23].

The energy of interaction is mainly dependent on the distribution of the electrons on the molecules. For simple molecules one can tabulate:

<table>
<thead>
<tr>
<th>Interaction</th>
<th>Energy Formula</th>
</tr>
</thead>
<tbody>
<tr>
<td>ion/ion</td>
<td>( E \propto - \frac{Z_1 Z_2}{4\pi \varepsilon_0 R} )</td>
</tr>
<tr>
<td>ion/dipole</td>
<td>( E \propto - \frac{Z_1 \mu_2}{4\pi \varepsilon_0 R^3} )</td>
</tr>
<tr>
<td>dipole/dipole</td>
<td>( E \propto - \frac{\mu_1 \mu_2}{4\pi \varepsilon_0 R^4} )</td>
</tr>
<tr>
<td>ion/induced dipole</td>
<td>( E \propto - \frac{Z_1 I}{4\pi \varepsilon_0 R^4} )</td>
</tr>
<tr>
<td>dipole/induced dipole</td>
<td>( E \propto - \frac{\mu_1 I a}{4\pi \varepsilon_0 R^6} )</td>
</tr>
<tr>
<td>induced dipole/induced dipole</td>
<td>( E \propto - \frac{R^2 a^2}{4\pi \varepsilon_0 R^6} - \frac{1}{R^6} - \frac{1}{R^{10}} )</td>
</tr>
</tbody>
</table>

\( Z = \) ion charge  
\( \mu = \) dipole moment  
\( I = \) Ionization energy  
\( a = \) polarizability

This already shows the diversity of forces which may be involved in the total interaction between two particles.

The last force mentioned is the so called London-vdWaals or dispersion force which occurs in the interaction between any two molecules. In absolute magnitude it is the weakest force but, since it is only additive, it is not cancelled by other interactions as is the case for the ionic interactions of a crystal. Thus it is effective over longer distances.

For this reason in the literature on forces between particles other forces are usually neglected and only vdwWaals forces are considered. At short range and when touching this oversimplifies the situation and ionic interactions should also be included.
The force between two particles can be obtained by differentiation of the total energy content of the system with respect to the distance between the particles:

\[ F = \frac{\delta E}{\delta r} \quad \ldots(3.1.7) \]

For the vdwWaals interactions the higher order terms are conveniently neglected and the force is proportional to the inverse of the distance to the seventh power.

\[ F_{vdW} \propto \frac{1}{r^7} \quad \ldots(3.1.8) \]

This expression is valid for a certain distance only (about 500 Å), thereafter the power increases.

The lower limit is given by the vdwWaals radius which is estimated, for non-ionic lattices, to be about 3 to 4 Å.

The attractive forces are counteracted by the Born repulsion forces when the molecular orbitals penetrate. This force increases at a rate dependent on the size of the molecules involved. Born proposed the following approximation:

\[ E_{repulsion} \propto \frac{1}{r^n} \quad \ldots(3.1.9) \]

In this formula \( n \) varies from 5 for small molecules to 12 for large ones. (Handbook of Chemistry F-240) For simple molecules the exponential repulsion is more accurate:

\[ E_{repulsion} \propto \exp \left( \frac{1}{r} \right) \quad \ldots(3.1.10) \]

For the total energy the attractive and repulsive interactions must be added. Lennard and Jones showed that a 6-12 potential function of the following form is a reasonable approximation for some non ionic materials:

\[ E = -4 \epsilon \left( \frac{1}{2} \left( \frac{r_0}{r} \right)^6 - \frac{1}{4} \left( \frac{r_0}{r} \right)^{12} \right) \quad \ldots(3.1.11) \]

\( \epsilon \) : energy term
\( r_0 \) : equilibrium separation distance
\( r \) : distance between centres

For an ionic lattice such as NaCl, the interaction can better be described by a summation of the electrostatic attraction and the Born repulsion:

\[ E = -\frac{e^2}{4\pi \epsilon_0 r} + \frac{B}{r^9} \quad \ldots(3.1.12) \]
3.1.2 forces between particles

Rumpf classified the possible interaction between particles as follows:

1) Attractive forces without contact
   - vdmWaals forces
   - electrostatic forces
   - magnetic forces

2) Attractive forces with contact
   - liquid bridges with a mobile interface
   - viscous binders
   - solid bridges

Both categories will be discussed.

In principle the total force between two particles is found by summing all individual interactions of the molecules and adding the Coulomb forces. This exercise can be simplified by compounding some interactions in macroscopic forces and transforming the discrete system into continuous bodies.

**Surface energy**

The macroscopic description of the forces between the molecules of different phases is achieved by means of the concept of interfacial energy. This concept will be briefly explained in order to point out some aspects easily overlooked.

To any volume of matter in a phase a time average energy content can be ascribed which is determined by the interaction of the molecules in the volume. The matter located at an the interfacial area has an energy which is different from that in the bulk of the phases because of the mixed interactions between the different molecules. The average energy is thus a continuous function, constant in a bulk area and changing at a interfacial area.

In considering this system in two homogenous phases with a mathematically sharp interface Gibbs ascribed any difference of the total sum of energy or the total mass between the idealized system and the real system to the interface itself. Any change of the position of the interface also changes the sum of energy and mass for the idealized system and, since the real system is independent of the choice of the interface, the interfacial energy and mass must also change. This allows the choice of interface according to certain criteria. A useful criterion is the zero adsorption of matter onto the interface.
Defay[6] shows that for such an interface the following general formula is valid:

$$\sigma = \sigma_0 = f - \sum_{i=2}^{i=c} \Gamma_{i,l} \mu_i \quad \ldots(3.1.13)$$

In this formula $f$ is the surface free energy, $\Gamma_{i,l}$ the Gibbs absorbed matter of phase $i$ on $l$ and $\mu$ is the chemical potential of material $i$.

All changes of the macroscopic form of the phase, in which the interface changes its size also cause a change of the total interfacial energy, thus creating a driving force towards the form having the smallest interfacial area.

The main advantage of this representation of the forces is that the force between large sets of molecules can now be calculated from only the area of the interface. The forces of attraction between two particles have also been described in this way. In this case the interfacial area is the area of material which is touching and the interfacial energy is the surface energy of the material of the particles. Kingery[14] shows the application of this concept to various situations. During sintering, for example, the surface energy concept explains many phenomena in a simple way.

The concept of interfacial energy must be treated with care. The surface tension of an adsorbed layer for instance is almost a paradoxical idea since no bulk of the phase is present anyway. All the properties of an adsorbed layer are strongly influenced by the molecular interactions with the material in the neighbourhood, which also means that the geometry of the neighbourhood can have a definite effect. This affects all macroscopic properties of these layers, for example the viscosity, the diffusity, melting point and surface tension. Any calculation with these macroscopic parameters must be conducted carefully.

The effect of the curvature of the interface on the surface energy can used as an example of the effect of the geometry of the neighbourhood. Defay[6] calculated the effect of small radii:

<table>
<thead>
<tr>
<th>log(r)</th>
<th>droplet $\sigma/\sigma_0$</th>
<th>bubble $\sigma/\sigma_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>-6</td>
<td>0.9996</td>
<td>1.0004</td>
</tr>
<tr>
<td>-7</td>
<td>0.996</td>
<td>1.003</td>
</tr>
<tr>
<td>-8</td>
<td>0.968</td>
<td>1.033</td>
</tr>
<tr>
<td>-9</td>
<td>0.755</td>
<td>1.48</td>
</tr>
</tbody>
</table>

48
Bikerman\[2\] does not accept the concept of surface energy for solid phases. He argues that the surface energy must be an intrinsic property of the material and that the excess energy of a solid phase does not possess this property, since it can easily be influenced by a chemical or morphological variation of the surface.

A few of his arguments can be criticized. His calculation of the pressure in a crystal caused by the infinitely sharp angle of two crystal phases is an unacceptable extrapolation of a formula which he should not have used in the first place. Dufay\[6\] discusses the matter of interfacial energy in crystals extensively and shows that the rule due to Wulff should be used instead. For this formula not the radius of curvature is the important parameter but the distance of the crystal face to the centre of gravity.

\[
\frac{a_y}{h_y} = \frac{1}{2}
\]

(Wulff) \hspace{1cm} ...(3.1.14)

\[
\ln \frac{P}{P_0} = 2 \frac{a_y}{h} \frac{v}{RT}
\]

...(3.1.15)

This discussion shows that care must be taken in transferring the ideas used for the liquid phase to the solid phase or to adsorbed phases directly. Different rules apply.
3.2 Theory of particle interactions

3.2.1 normal forces between non contacting particles.

For non-touching particles of all the molecular interactions only the vdwAls forces are taken into consideration. If the objects are sufficiently separated the discrete molecular morphology of the particle can be replaced by a continuous body whose boundaries can be described by a mathematical, continuous function. Integration can represent the summation of the molecular interactions, given the appropriate continuous potential function.

In his solution for two spheres Hamaker[10] used a parameter $A$ for the interaction energy between the continuous volumes. Later Lifshitz starting from different assumptions showed that the same formulae emerge in the macroscopic theory of dispersion albeit with another set of parameters. The Lifshitz interaction parameter is equal to the Hamaker constant if divided by $3/4\pi$. For the most frequently encountered combinations of two objects in vacuum the following formulas apply:

1) half space-half space $P_{vdW} = - \frac{hw}{8\pi^2} \frac{1}{a^3}$

2) half space- sphere $P_{vdW} = - \frac{hw}{8\pi} \frac{R}{a^2}$

3) sphere 1 - sphere 2 $P_{vdW} = - \frac{hw}{8\pi} \frac{R_1 R_2}{a^2 R_1 + R_2}$

$R_i =$ radius sphere $i$

$a =$ distance between boundaries of objects

In an extensive review Visser[29] tabulated values of interaction parameters as determined by several methods. Dependent on the material the parameter $hw$ varies between $10^{-18}$ and $10^{-21}$ J.

For two objects of dissimilar material, not in vacuum but in a medium with a dielectric constant other than 1, another approximation has been found:

$$\bar{w} = \int_0^\infty \frac{\left(\epsilon_1(i\theta) - \epsilon_3(i\theta)\right) \left(\epsilon_2(i\theta) - \epsilon_3(i\theta)\right)}{\left(\epsilon_1(i\theta) + \epsilon_3(i\theta)\right) \left(\epsilon_2(i\theta) + \epsilon_3(i\theta)\right)} \, d\theta \quad \ldots(3.2.4)$$

Krupp[15] showed that the spectra of a large number of materials could satisfactorily be approximated by:
\[ \frac{\epsilon_1(i\theta) - 1}{\epsilon_1(i\theta) + 1} = a_1 \exp (-b i\theta) \] ...(3.2.5)

Using the previous approximation this yields, for two dissimilar materials in vacuum, the following formula:

\[ \bar{w}_{12} = \sqrt{w_{11} \cdot w_{22}} \] ...(3.2.6)

In terms of the Hamaker constants:

\[ A_{12} = \sqrt{A_{11} \cdot A_{22}} \] ...(3.2.7)

If the intermediate medium is not a vacuum, the following formula is commonly used for the Hamaker constant:

\[ A_{131} = A_{11} + A_{33} - 2A_{13} \] ...(3.2.8)

\[ A_{131} = (\sqrt{A_{11}} - \sqrt{A_{33}})^2 \] ...(3.2.9)

and

\[ A_{132} = A_{12} - A_{23} - A_{13} + A_{33} \] ...(3.2.10)

\[ A_{132} = (\sqrt{A_{11}} - \sqrt{A_{33}})(\sqrt{A_{22}} - \sqrt{A_{33}}) \] ...(3.2.11)

Comparing the results of these formulae with those calculated from graphical integration of \[3.2.4\] Visser[29] concluded that a correction factor should be introduced:

\[ A_{132} = c (\sqrt{A_{11}} - \sqrt{A_{33}})(\sqrt{A_{22}} - \sqrt{A_{33}}) \] ...(3.2.12)

He proposes a factor of 1.6 for water and 1.7 for polystyrene. Van den Temple also addresses this problem and suggests a factor 1.5. These are rather crude corrections and a more accurate one is obtained by substituting the approximation for the spectra directly into formula (3.24). See appendix 3.B.

This results in four correction factors which, for most cases, are almost identical.

\[ hw_{132} = C_{12} \cdot hw_{12} - C_{13} \cdot hw_{13} - C_{23} \cdot hw_{23} + C_{33} \cdot hw_{33} \] ...(3.2.13)

\[ C_{ij} = \sum_{k=0}^{\infty} \frac{(a_1a_3+a_2a_3)}{1 + k/2.(1+b_j/b_i)} \] ...(3.2.14)

\[ a_i = \frac{\epsilon_{i0} - 1}{\epsilon_{i0} + 1} \quad b_i = a_i^2/2hw_{ii} \] ...(3.2.15)
**Electrostatic interaction**

Electrostatic attraction may occur when one or both particles are charged or when double layers are present. A net charge develops through breakage, rubbing or collision when the resistivity is so high that on separating the contact between two parts the charge differences cannot equalize in time. This charge leaks away through the air, especially if there is humidity.

For plastics separate islands of positive and negative static charge may be present, even though the net charge on the particle is zero. In that case an electrostatic attraction between particles may occur.

Electrical double layers are created when the work potentials of touching particles are unequal. When the contact is broken, current transfer may occur. see Deryaguin[7,8]. When after breakage of the contact the charge is not equalized a net charge remains. This static charge influences the total force needed for separation. Zimon[30] reports that for a system of copper-dust the net charge and the double layer just compensate and that no influence of the double layers could be detected.

In order to limit the number of variables in this investigation care has been taken to exclude the role of these forces. When different materials were used no measurements have been taken in dry air.

**Ionic interaction**

The ionic interactions are stronger than the vdWaals forces but the positive and negative charges quickly cancel each other at distance. These forces are, therefore, only important at close approach of about 2 lattice spacings.

When two crystal faces approach the chance of a perfect alignment is extremely small. On touching, a more or less chaotic amorphous crystal, with large imperfections and holes, will form. In some cases the lattices will deform and the ions in the lattices will migrate to form a new, energetically more favourable crystal. This process will be slow since it depends on repositioning of the ions and the force of separation may increase steadily for a considerable time. The speed of recrystallisation will be influenced by both temperature and the amount of water present.

The calculation of the total force of the interaction force in these layers is a too difficult and tedious task, and the concept of interfacial energy of the contact is more convenient. This excess energy is larger when the morphology of the interparticle
layer is more chaotic.

A calculation of the surface energy of a ionic and vdWaals lattice show that the energy of the ionic lattice is about ten times greater. A complication in calculating the properties of crystals is that the energy of a face is also dependent on its orientation in the lattice. In table 3.1 the surface energies of a number of crystals faces is shown.

<p>| | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>vdWaals lattice</strong></td>
<td><em>Argon</em></td>
<td>( \Gamma = 0.02 \ J/m^2 )</td>
</tr>
<tr>
<td><strong>Ionic lattice</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LiCl{100}</td>
<td>( \Gamma = 0.41 \ J/m^2 )</td>
<td></td>
</tr>
<tr>
<td>LiCl{110}</td>
<td>( \Gamma = 1.11 \ J/m^2 )</td>
<td></td>
</tr>
<tr>
<td>NaCl{100}</td>
<td>( \Gamma = 0.32 \ J/m^2 )</td>
<td></td>
</tr>
<tr>
<td>CaCO3{1010}</td>
<td>( \Gamma = 0.23 \ J/m^2 )</td>
<td></td>
</tr>
</tbody>
</table>

For chemical interactions some information is available. Ponec[23] cites measurements on the adsorption of SO2 on BaF and states that for this system the dispersion and electrostatic energies are of the same order of magnitude: resp. 13 and 15 kJ/mole. The conclusion must be that the ionic forces should not be neglected since they are at least as important as the vdWaals forces when the particles are in near or true contact. If nevertheless a calculation, assuming vdWaals forces only, formulas (3.1.1-3), is conducted the separation length parameter will be small to compensate for the additional forces.
3.2.2 normal forces between contacting particles

The continuum approach cannot hold for the calculation of the vdWaals attraction forces for particles in near or true contact. Discrete summation of the vdWaals forces for regular crystals yields good results, but usually the order in an interface will be far from regular. As for the ionic forces an interfacial energy may prove to be useful.

A number of points must be taken in consideration in modelling the interaction between real particles:

- Due to the attractive force both objects deform and more material is closer than the original shape implies. A sphere does not touch with one mathematical point, but will deform and an interfacial area of at least several atoms develops. See fig. 3.1.

- The size of the interfacial area may change slowly with time because the systems 'strives' to a lower energy content. Also, internal stresses which are created when the contact is formed may relax when the material deforms plastically.

- Captured molecules, for example water molecules, influence the properties of the interfacial area.

- The geometry of the solid phase near to the contact will promote adsorption and condensation of molecules from the gas.
phase. The curvature of the solid is very high, which results in a large interaction and a very low potential field for the molecules.

When these molecules adsorb, the forces between the particles are influenced in two opposing ways:

a) the molecules increase the interfacial area and enlarge the forces between the particles by bridging and other interactions. Water may promote ionic/dipole forces and may also form hydrogen bonds between the crystal faces.

b) the dielectricum between the particles is changed and the dispersion forces may be shielded by this, thus decreasing their contribution.

Another important point is that the common method of determining the attractive forces is to measure the force of separation of two objects. During the process of separation the geometry of the objects changes and, at the moment of rupture, the geometry is not completely known. The separation process is probably a crack-extension problem, the crack moving in from the sides of the contact area until the system becomes unstable. This process may also be a function of the rate of application of force. A high estimate of this critical force is based on an instantaneous separation of all the interactions across the interface, modified by a factor to account for the inhomogeneities and voids.

**Force-displacement curves for touching particles**

Even when the external compressive forces on the particles is zero, the internal attractive forces will deform the particles and pull them together until balanced by the repulsive reaction due to the deformation. This deformation will be partly permanent, the permanent fraction of the deformation being dependent on the ease of plastic deformation of the material and the specific geometry at the interface. Deryaguin stated that the elastic part of the deformation recovers when the particles are again separated and that consequently the force of separation is not influenced.
Fig 3.2: Force displacement curve for a touching, resting and separating particle.

C point of first contact on approach
B initial equilibrium position
A-B plastic flow in the contact
A-C separation branch
Fe maximum force during approach
Fp maximum force during separation

The force-displacement curve representing two particles approaching from infinite distance, touching, deforming and subsequently separating again can be described in the following general way. See fig.3.2.
The distance is taken between two reference points in the particles: a convenient choice would be the centre of gravity before deformation. This avoids the problem of redefining the distance between the particles when deformation occurs.
The particles are assumed to be spherical before approach, with a volume radius R, and having no net charge. All situations are assumed to be equilibrium states and the deformation of the particles has ceased.

At a large distance the forces between the particles are dominated by the vdWaals forces. The attractive forces can be calculated using the Hamaker solution. If the particles are rough care must be taken to substitute the appropriate mean radius of the sphere or to insert the radius of the approaching asperities. The large exponent of the vdWaals forces causes the material nearest to the area of contact to be dominant and small asperities may dominate completely. (see appendix 3.B.)

Due to the attractive forces the sphere will deform and the surfaces bulge towards each other. As the particles approach closer this deformation increases and, at a certain distance, the interfaces touch. (d touch > 2R) At this point the integration will deviates even if the correct continuous function for the interfaces is used. As the approach continues, two mechanisms occur: some of the material near to the interface is pulled together and increases the total force of attraction, some of the touching material deforms and builds up a repulsive force. When the repulsive force balances the attraction, the particles are in an equilibrium position. Further approach increases the repulsive forces more than the attractive forces.

During the approach, plastic deformation of the materials may have relaxed some of the repulsive forces. Small rugosities may be flattened. These are dynamic processes and the speed of approach may influence the behaviour.

Reversing the process and separating the particles again, the force-displacement curve will deviate from the approach curve if the plastic deformation flow has been large and the influence of dynamic processes, such as crack extension, is more important. It can be speculated that the force of separation will always be higher than that of touching.

**hardness**

The ease of plastic deformation of a material body is expressed by its empirical hardness. A number of methods are used, each giving either a relative or an absolute value.

1- Moh's scale, based on the scratching of 10 selected minerals
2- Vickers, indentation by a pyramidal indentor,
3- Knoop, indentation by a conical indentor,
4- Brinell, indentation by a sphere.
The most useful definition of hardness for modelling is based on the area of plastic deformation divided by the driving force:

\[ H = \frac{F}{a} \text{ (in Pa)} \]  \hspace{1cm} \text{...(3.2.16)}

The hardness on a macroscopic scale may be different from that on the scale of the contact.

For crystals the situation is further complicated because each face has its own specific hardness.

Table 3.2: Hardness of various faces of crystals

<table>
<thead>
<tr>
<th>Lattice indices</th>
<th>Vickers</th>
</tr>
</thead>
<tbody>
<tr>
<td>K2SO4</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>95</td>
</tr>
<tr>
<td>111</td>
<td>100</td>
</tr>
<tr>
<td>120</td>
<td>130</td>
</tr>
<tr>
<td>NH4H2PO4</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>69</td>
</tr>
<tr>
<td>110</td>
<td>73</td>
</tr>
</tbody>
</table>

When the surface is polycrystalline, the hardness varies along the surface. Also at the interface between two grains in the surface the interface itself may have a different hardness than the bulk of the grains.

Pashley[22] showed the variation of the microhardness with the depth of indentation. As the depth increases the hardness decreases. Oxide layers immobilize the dislocations below the oxide surface causing an increase in hardness.

These complications may contribute to the spread in the distribution of separation forces.

Separation force of deformed spheres

Although the Hamaker solution cannot be used for the case of touching particles, the separation force is estimated by substituting an empirical separation constant in the formula. This constant ranges from 3 to 10 Å, which yields a range of a factor of 10 in the prediction. The constant has been justified by pointing to the lattice distance of vdWaals crystals, but this is superfluous since there is no theoretical basis for this constant whatsoever and it should not be seen as a true measure of the average distance between the particles. Substituting such a constant the Hamaker formula can be used for a first approximation of the separation forces.

A number of authors have included the effect of plastic deformation. All have estimated the force of separation on the assumption of instantaneous breakage. Density differences are neglected which certainly will cause underestimation.
Dahneke[5] approximates the geometry after plastic flow with a sphere, from which a segment is sliced. The size of the segment removed is given by using the Hertzian formula for a pressure force, which is assumed equal to the attractive force. Using this approach he derives a simple formula which, at least qualitatively, shows the effect of the hardness of the sphere. However, the separation constant appears with a power of 7 and, in view of the uncertainty of the estimate of this constant, the quality of an a priori estimation of the quantitative effect of the hardness will be poor.

Deryaguin[8] criticizes Dahneke's approach severely and points out that a deformation calculated with the Hertz formula assumes elastic and not plastic deformation. He further states that any deformation which is not plastic will recover when the spheres are separated and this deformation will not contribute in the separation force. For a ideal elastic sphere the recovery will be complete and the force of separation be equal to the force of attraction calculated for a sphere touching at just one point. This however also is an approximation since, at the point of separation, the spheres will necessarily be deformed. Deryaguin investigated more precisely the deformed geometry. The resulting formulae are complex, and are still based on a few assumptions.

Deryaguin[8] also used a different approach to estimate the total interaction. He calculated the forces exerted between two elements which are just opposite in a chosen coordinate system and then sums them. The cross interactions between elements is neglected and his calculations must, therefore, be an underestimate. In an further calculation Deryaguin used surface energies and a remnant interfacial energy for the contact area to find the force of attraction.
Krupp[16,17] combined the Hertzian approximation and a yield criterion to estimate the area of a contact which has plastically deformed and which is still elastic. When the particles are separated the elastic area recovers. The deformed geometry is that of a sphere with an segment removed.

\[
\pi r_0^2 = (\text{Fatt}/H) + 1/3 \cdot K \quad \ldots (3.2.17)
\]

\[
\pi r_1^2 = (\text{Fatt}/H) - 2/3 \cdot K \quad \ldots (3.2.18)
\]

\[
K = \left( \left( \pi \cdot (1-v) \cdot R \cdot H \right) / (2E) \right)^{3/2} \quad \ldots (3.2.19)
\]

\[
r_0 = \text{radius of total area}
\]

\[
r_1 = \text{radius of plastically deformed area}
\]

\[
H = \text{Hardness}
\]

\[
\text{Fatt} = \text{forces of attraction}
\]

A plastic deformation occurs only if the forces exceed a limit:

\[
(\text{Fatt}/H) > 2/3 \cdot \left( \left( \pi \cdot (1-v) \cdot R \cdot H \right) / (2E) \right)^{3/2} \quad \ldots (3.2.20)
\]

A calculation of the plastic and elastic proportions of the area of deformation shows:

\[
S = \frac{r_1^2}{(r_0^2 - r_1^2)} \quad \ldots (3.2.21)
\]

Substituting the Hamaker approach with a separation constant

\[
S = \left( \frac{h \cdot w}{8 \pi^2} \right) \cdot \frac{R}{a} \cdot \frac{4E^2}{(\pi \cdot (1-v^2)^2)} \cdot R^3 \cdot H^3 - 2/3 \quad \ldots (3.2.22)
\]
After making this first approximation of the contact area a new estimate is made of the separation force. Krupp added, for this second approximation, the Hamaker force for the undeformed sphere to a contribution proportional to the plastic area:

\[ F_{\text{att}} = F_{\text{hamaker}} + \pi r_1^2 \sigma_{\text{opp}}. \]  \( \text{(3.2.23)} \)

The plastic area is calculated by:

\[ \pi r_1^2 = (F_{\text{hamaker}}/H) - 2/3 . K \]  \( \text{(3.2.24)} \)

The force per unit area is equated to the vdWaals pressure of the material. Krupp and Sperling[16] state:

\[ F_{\text{att}} = F_{\text{hamaker}} \left( 1 + \frac{P_{\text{vdWaals}}}{H} - \frac{2}{3} K \frac{P_{\text{vdWaals}}}{F_{\text{hamaker}}} \right) \]  \( \text{(3.2.25)} \)

The effect of hardness is included in this formula at least qualitatively. But to be consistent in this approach the plastic area should have been estimated from the attractive force for the particle in its resting position:

\[ F_{\text{att, rust}} = F_{\text{hamaker}} + \pi r_0^2 \sigma_{\text{opp}}. \]  \( \text{(3.2.26)} \)

\[ \pi r_0^2 = (F_{\text{att}}/H) + 1/3 . K \]  \( \text{(3.2.27)} \)

\[ F_{\text{att, rust}} = \frac{(F_{\text{hamaker}} + 1/3 K P_{\text{vdWaals}}) / \left( 1 - \frac{P_{\text{vdWaals}}}{H} \right)}{} \]  \( \text{(3.2.28)} \)

\[ \pi r_1^2 = (F_{\text{att, rust}}/H) - 2/3 . K \]  \( \text{(3.2.29)} \)

This results in a somewhat larger estimate of the separation force. However, the approximations are so that such tedious details may well be neglected.

With this philosophy Krupp simplified his solution, leaving out the K-term:

\[ F = F_{\text{hamaker}} \left( 1 + \frac{F_{\text{hamaker}}}{R.a.H} \right). \]  \( \text{(3.2.30)} \)

and showed that this simple form already follows the qualitative trends. When the deformation is dominant the force is given by:
trends. When the deformation is dominant the force is given by:

\[ F = \frac{F_{\text{hamaker}}}{R \cdot a \cdot H} = \frac{(hw)^2}{H} \frac{R}{(8\pi^2)^2 \cdot a^5} \]  \hspace{1cm} (3.2.31)

For soft materials like polymers and plastics this is an approximate solution. The force is proportional to the inverse of the hardness. The separation constant again introduces a large uncertainty.

effect of roughness on the separation force

The models discussed previously assume smooth spherical particles. In reality such particles are rarely encountered.

Experimental work by Massimilla[19] on catalyst particles showed that the overall behaviour of a particles is subject to the local geometry of the contacts. This geometry is completely controlled by asperities and by interference from small, adhering particles. The mean force of separation for his particles is constant for a range of particle sizes.

A more precise estimation of the separation forces must include a description of the particle surface morphology leading to the possible contact geometry. At this moment only the most general discussion is possible.

When two rough surfaces meet perfect matching will not occur and, in the first instance, the two faces will rest on each others asperities leaving a complicated network of voids between them. Pressure and thermodynamic forces may cause plastic flow. Even when no small particles are present the final contact geometry may be a network of more or less smooth ellipsoidal capillaries running through the interfacial area. The effect of these imperfections will be manyfold.

The capillaries will preferentially adsorb molecules from the gas and water may influence the separation force even at low relative humidities.

When the particles are separating, the voids and the sides of the contact will act as stress raisers and the separation force will be influenced by the exact location of the voids.

non-symmetrical particles

For real particles the force will probably be positioned such that moments occur during the separation and the particle will rotate as it separates. A new stable position may be found if the critical force has not been surpassed, and, in this new position dynamic effects such as the redistribution of adsorbed layers may influence the force later on.
None of the models discussed can include the complications non-spherical rough particles present. These models can therefore be used for a first indication only.

3.2.3 forces of friction between touching particles.

The friction force of a contact between two particles is difficult to measure and not much theory has been published. More is known about larger systems.

The friction between two dry large bodies is well described by a static and dynamic coefficient of friction. The friction law is usually assumed to vary linearly with the normal pressure between the bodies. The flattening of the asperities between the bodies is apparently accounted for in this linearity.

For liquid-solid friction a number of situations can be distinguished, dependent on the amount of water and the morphology of the surface.

Bikerman[2] tabulated 6 different regimes depending on the normal pressure and the amount of liquid. Of these, the first three are the most characteristic.

1- Hydrodynamic Lubrication
2- Boundary Lubrication
3- Dry friction
4- elastic deformation
5- plastic deformation
6- abrasion

1) Hydrodynamic lubrication exists when a thick layer of liquid completely shields the solid and the friction is dependent only on the properties of the liquid.

2) If the liquid layer is thin, the asperities or peaks of the solid hit each other occasionally and boundary lubrication occurs. This situation is badly defined. The properties of the liquid are strongly influenced by the presence of the solid, and the solid-solid interaction is variable.

If the viscosity of the layer is increased two opposite effects may influence the friction. The penetration of the asperities into the layer may be inhibited thus decreasing the friction, the friction increases due to the larger viscosity.

3) With no water present the friction is determined by the rugosity of the solid surfaces and the normal force between the body. The Coulomb friction law is usually valid.

The other three categories are subdivisions of the effects which occur at higher normal forces.

For contacts between particles the Coulomb law is also assumed to be valid. The forces of attraction and the external pressure force are summed to obtain the total normal stress. This stress
Fig 3.3: The forces involved in separating a particle tangentially correlates well with the friction force (Zimon[30]). It must be noted that the relevant forces are the forces in the equilibrium position and not the separation force. All the lubrication effects described above are also assumed to contribute.
Experimental work on tangential separation

Jimbo[13] remarked that it is often not the true friction but the rolling resistance of a contact which is measured in experiments. When using a body force to separate the particles, that force is active on the mass midpoint and not on the plane of sliding. This causes the particle to roll rather than to slide. If the particle is not perfectly spherical, rolling only occurs when it can be lifted across its asperities.

Jimbo was able to show that a simple relation exists between the tangential and normal separation forces, see fig 3.3:

\[ F_{sn} = F_{ad} - \pi \frac{d^3 \rho g d \phi}{6} \sin \phi - \frac{d \phi}{2r_0} F_{st} \quad \ldots(3.2.32) \]

\( F_{sn} \) = normal component  
\( F_{st} \) = tangential component

In this derivation, the adhesive force is assumed to be uniformly divided along the entire surface of the contact. Different geometries thus yield a similar formula but different coefficients.

This formula was experimentally verified by applying a normal vibration to particles adhering to a plate, which is inclined to the gravity field. A reasonably linear correlation was measured and allowed the estimation of the adhering contact area.

\[ F_{sn} = -\frac{d \phi}{2r_0} M_p \sin \phi + F_{ad} \quad \ldots(3.2.33) \]

\( r_0 \) = radius of the adhering area  
\( M_p \) = mass of the particle  
\( \phi \) = angle of inclination

When a purely tangential force is applied to small particles, for which the gravity component is negligible the formula simplifies to:

\[ F_{st} = \frac{2r_0}{d \phi} F_{ad} \quad \ldots(3.2.34) \]

This formula shows, qualitatively, the influence of the two important parameters. The tangential force is directly proportional to the area of contact, which in this definition is determined by the positions of the protruding asperities, and to the normal separation force.
Fig 3.4: The distribution of separation forces for BaSO₄ particles according to Polke and Herrmann (1971), published in Zervheander (1976).

The experimental results published are presented graphically on a log-normal scale and the lines for normal and tangential separation are almost parallel, indicating that the distributions of normal force and tangential force are almost identical.

Measurements of Polke (cited in Zherveander) show that for limestone particles separated from a plate in a centrifuge the normal forces are about 10 times as large and that the distributions are almost identical. This evidence supports Jimbo's formula. See fig. 3.4.

Zimon[30] shows a set of adhesion data, in which the forces of separation are plotted against the inclination of the plate. There is a difference of a factor of 5 between the tangential and normal separation forces.
3.2.4 adsorption of water on contacts.

The adsorption of water on the contact area is controlled by a complex potential field. The near vicinity of the solid at the contact line and in the network of pores in the contact area itself will promote preferential adsorption of molecules.

The contact line itself is not a perfectly sharp angle, but the radius of the geometry is extremely small. The size and morphology of the pores within the contact may vary widely, their size ranging from the smallest possible to rather large ones if the roughness is appreciable before contact.

The behaviour of the pores in the contact can be compared to the adsorption behaviour of pores. In general pores are classified into three categories: macropores >100Å, mesopores 100-10Å, and micropores <10Å. Adsorption is different for each class. Condensation in micropores and ultra-fine pores already occur at relative humidities far below the saturation point.

Considering the process of condensation on the perimeter of the contact area we can see the following sequence of events. At the contact line itself preferential adsorption will start at very low humidities. Since this line is surrounded by solid material the potential field is extremely low and this area will fill with molecules first. As the humidity increases this adsorption continues and the angle of contact fills. Whilst it is filling the water-gas interface increases and becomes less curved. For each humidity there is an equilibrium adsorption between the particles. See fig. 3.5.

Gregg et al. [9] calculate the saturation pressure for a conical microcapillary with Kelvin's law:

\[
\ln p_s = - \frac{2 \gamma r}{r \cos \delta} + \frac{r \gamma}{RT} \tag{3.2.35}
\]

where:
- \( r \) = radius condensed liquid
- \( \gamma \) = surface tension

This approximation will be reliable for the bulk parameters if the liquid bridge is at least 7 molecular layers thick. Substituting a radius of 7 atoms we find a corresponding humidity of 80%.

Extrapolating to smaller radii, a thickness of 2 layers coincides with 30% RH.

Since the interaction with the solid walls is neglected the actual thickness of the layer will be larger making condensation more likely at low humidities.
Fig 3.5  Schematic drawing of the relationship of intercontact and interparticle condensation and relative humidity. Grey area is occupied by water molecules.

- c: contour line of contact
- d: division line between particles
- Ps: micropores
- Pl: macropores
- w: wall
- s: sphere
Experimental work on the onset of condensation in a contact point is extremely difficult since the area is so small compared to the total area of the particles. An adsorption isotherm for the particles has no significant contribution from the behaviour of the contacts.

**continuous water layers between the particles**

When humidity rises it may be energetically more favourable for some materials to adsorb water onto the entire surface of the contact area. In that case the particles loose contact and the water layers force themselves between the particles. The effect of this penetration of water on the separation force is uncertain. According to Vervoorn[26] the penetration will cause a net decrease of the separation force because:

1) the distance between the particles is increased, which causes a decrease in the contribution of the vdWaals force.

2) the dielectric properties of the space between the particles changes and the vdWaals forces are shielded.

On the other hand the force is enlarged by a strong interaction between the solids and the water layer due to the vdWaals forces, the ionic forces and H-binding.

When two systems are considered, one system in which the adsorbed water is present only on the contact line and the other system in which the adsorbed water penetrates between the particles, then the first system will only transform into the second if this transition is thermodynamically favourable. In separating both systems reach the same end condition and thus it may be concluded that if water has penetrated more energy is needed to separate that system. If it is accepted that adsorbed water on the contactline increases the force of separation then it follows that the penetration of water will also increase the adhesive force.

Against this reasoning it can be suggested that non-equilibrium conditions are not taken into account.

The effect of penetrating water layers on the friction is related to the previous problem. The layer itself will decrease the coefficient of friction, but if the normal force increases enough to compensate for the reduction the friction may also increase.

**literature on the effect of humidity**

Some investigations showing the effect of humidity have been published. Summarizing it seems that between 30-60 %RH the normal separation force increases more or less abruptly, increasing further with the humidity until a maximum is reached when the force drops suddenly.
Fig 3.6: The influence of humidity on the separation force of large glass spheres. Chikazawa (1984)
Chikazawa[4] reports data for 500μ glass spheres. He finds almost a step like increase at 30% and at 75% and a maximum at 80% RH. See fig. 3.6. Zimon[30] states that, at 70%RH, capillary condensation occurs due to which the adhesion increases.

The effect of humidity on powder agglomerates is complex. In these experiments combinations of normal, tangential and rotational separations are to be expected and the results cannot be ascribed to only one contribution. This difficulty can be illustrated by the following example.

Vervoorn[26] measured the unconfined yield strength of titaniumdioxid granules. He found that after a initial increase in strength up to a water content of 5 mg/g, the force decreased steadily as the water content of the granules increased to 20 mg/g. If these effects are mainly due to diminished normal separation forces, the decrease of the yield strength must be due to the effect of penetrating water layers. On the other hand, arguing that the normal separation forces can only increase as more water condensate and penetrate, the diminishing force must be mainly due to a decrease in the interparticle friction.

Only a thorough model of the particle system during these tests may clarify the contribution of all the components. This model is not yet available.

3.2.5 Statistical distributions

All models of powder systems which use averages only are bound to fail eventually in explaining and predicting the behaviour of a powder system. Only if the proper distributions of all the relevant processes are included can a more realistic interpretation be expected.

This research has been concentrated on finding the nature of the distribution of the normal separation force under a variety of conditions. This knowledge may help clarify the dominant processes which give rise to the particular distribution.

Classically a number of distributions have proved useful for description of breakage processes.

1- The Weibull distribution.

\[ P( F \geq F_x) = \exp \left( -\left(\frac{F_x}{b}\right)^a \right) \]  \hspace{1cm} ...(3.2.36)

2- The Gamma distribution.

\[ P( F \geq F_x) = \frac{a}{\Gamma(a)} \int_{F_x}^{\infty} x^{a-1} \exp \left( -b x \right) dx \]  \hspace{1cm} ...(3.2.37)
3- The Log-Normal distribution.

\[
P( F \geq F_x) = \frac{1}{a \sqrt{2\pi}} \int_{F_x}^{\infty} \frac{1}{x} \exp \left( -\ln(x) - \frac{b^2}{2a^2} \right) dx
\]  

...(3.2.38)

In these formulas all forces are made dimensionless by dividing by 1 mdyne.

**Table 3.2** Characteristic parameters for various distributions

<table>
<thead>
<tr>
<th>P</th>
<th>Fmean</th>
<th>Fmedian</th>
<th>Fmodal</th>
<th>Variance</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>b \Gamma(1 + 1/a)</td>
<td>b (\ln(2))</td>
<td>b (1 - 1/a)</td>
<td>\left{ \frac{\Gamma(1+2/a)}{\Gamma^2(1+1/a)} - \frac{1}{\Gamma^2(1+1/a)} \right}</td>
</tr>
<tr>
<td>2</td>
<td>a/b</td>
<td>***</td>
<td>(a-1)/b</td>
<td>a/b^2</td>
</tr>
<tr>
<td>3</td>
<td>\exp\left( b + \frac{a^2}{2} \right)</td>
<td>\exp(b)</td>
<td>\exp\left( b - \frac{a^2}{2} \right)</td>
<td>\frac{\exp(2b + \frac{a^2}{2})}{\left( \exp(a^2) - 1 \right)}</td>
</tr>
</tbody>
</table>

The Weibull distribution has been used for breakage which is dependent on the number of flaws in the material. If applied to particle contact area it could be related to the number of irregularities in the area.

The gamma distribution can be considered to be a distribution of the chance which a number of elements with an exponential breakage function yield. Applied to the contact area it could give an indication of the number of contact points in the area which have to break before separation occurs.

The log-normal distribution can be an indication of the proportional effect of a number of normally distributed, independent factors. Applied to the particle contact, variations in hardness, the Hamaker constants and the number and sizes of voids might be responsible.
Fig 3.7 Comparision of the results of various separation methods by Jimbo (1983)
Distributions in literature.

The only distribution, which has been applied in the literature is the Log-Normal. However, it is not mentioned whether another model is also adequate.

Results by Jimbo[13], Polke(Zherveander[]) and Zimon[30] show that this function describes the intermediate region of broad distributions satisfactorily. The fractions of strongest and weakest bonds are more poorly described. The number of strong contacts is usually underestimated.

Jimbo compared several methods used for the determination of separation forces and concluded that the centrifugal method yields the highest values. Vibration methods can transmit normal as well as tangential vibrations at the same time and, therefore, record lower separation forces. See fig. 3.7. All the measurements were approximated by a log-normal distribution.

Polke measured the tangential and normal separation of BaSO4 and irregular glass particles of 10 and 20µ in a centrifuge. On a log-normal paper all the distributions could be approximated by a straight line, but only if the data for the higher forces were disregarded. The standard deviation of the measurements was about 2, diminishing for more polished surfaces to 0.5. F50, the median value, was proportional to the particle size.

3.3 Experimental

Using a centrifuge a series of normal separation measurements have been conducted on electronically classified particles. The sizing with an Image analyser afterwards allows quick measurement of a number of size classes in one run. This also enables more precise estimation of the separation forces since the force of separation is related to the mass of the particles. The distribution of the forces of each class are fitted with three possible distributions and the models tested whether they are adequate.

3.3.1 The equipment

The measuring cell is composed of a set of flat plates, stacked on top of each other in a holder. The holder is placed in a centrifuge and after spinning the plated are processed using an Image Analyser.
Since the plates are located above each other in one run a series of centrifugal accelerations can be applied to the particles on the plates at a fixed specified rotational speed. This speeds up the analysis and the operating conditions during the spinning are equal for all plates.

Before and after centrifuging the plates were positioned under a microscope and scanned with a image analyser. After the scan the particles were classified in size classes with a increment of 2 \( \mu \), and the number of adhering particles counted. The fraction of particles of each size class which had separated was deduced and the corresponding separation force calculated. At least 400 particles per size class entered the analysis.

**Handling and conditioning of the adhering particles**

The particles were deposited by dropping them from just above the plate. The holder was placed into the centrifuge for a dummy run and the particles on the plates were then counted. This eliminated very loose contacts which break by handling of the holder.

The plates were conditioned in an desiccator for at least three days. The humidity in the desiccator was kept constant by using saturated salt solutions and silica gel.

<table>
<thead>
<tr>
<th>Salts</th>
<th>Relative Humidity</th>
<th>Temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>KCl</td>
<td>84.2 % RH</td>
<td>25 °C</td>
</tr>
<tr>
<td>NaBr</td>
<td>57.8 % RH</td>
<td>25 °C</td>
</tr>
<tr>
<td>MgCl</td>
<td>32.8 % RH</td>
<td>25 °C</td>
</tr>
<tr>
<td>Silica Gel</td>
<td>5.0 % RH</td>
<td>25 °C</td>
</tr>
</tbody>
</table>

**Materials**

Two materials were used for the substrates, glass and perspex. The surfaces were rinsed by washing in a ultrasonic bath with soap, washing with distilled water, alcohol, and water again. No special polishing was applied.

Spherical copper particles were used in these tests. Visual inspection using a SEM showed that the surface of these particles to be smooth. Some particles were contaminated with small satellite particles, which had melted against the main particle. These could not be removed, but if the image analyser detected non spherical particles these were excluded from the particle count in the analysis.

The surface of the copper particles has not been reduced and an oxide layer is most likely. This oxide layer will have some effect on the hardness of the particles and on the vdWaal's and ionic forces. Since the main interest is in the form of the distribution of the forces this contamination was accepted.
3.3.2 Measurement of the force distribution:

Five size classes are selected. The series is not continuous because of a deficiency of particles in the 51-53 size range. The classes were:

1) 45-47 μ
2) 47-49 μ
3) 49-51 μ
4) 53-55 μ
5) 55-57 μ

For each class the force applied was calculated by taking the mean mass of each class and multiplying this by the centrifugal acceleration on the substrate.

\[ F = M \cdot w^2 \cdot r \]

The radial position \( r \) is dependent on the position of the plate in the holder, and varied from 18 to 25 cm. The rotational speed could be adjusted to a maximum value of 3000 rpm.

<table>
<thead>
<tr>
<th>layer number</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>radial distance (cm)</td>
<td>9.8</td>
<td>11.3</td>
<td>12.8</td>
<td>14.3</td>
<td>15.8</td>
</tr>
<tr>
<td>( F/F_g ) (1000 rpm)</td>
<td>125</td>
<td>141</td>
<td>158</td>
<td>175</td>
<td>192</td>
</tr>
</tbody>
</table>

When the fraction of particles remaining on the plates is plotted against the force applied a cumulative force distribution is obtained. This distribution can be fitted using cumulative probability models. Using these models force distributions can be plotted and the distribution parameters, such as the mean force and the standard deviation can be calculated.

In the literature often \( F_{50} \), the median value, is reported because it can be obtained accurately from the cumulative distribution. However, for modelling the median parameter does not supply information on the number and magnitude of weak or strong particles. If only one parameter is to be given \( F_{\text{mean}} \) would be a more suitable choice as it relates directly to the overall forces in a set of particles.

More information can be saved by approximating the distribution with a two parameter model and reporting two characteristic parameters which allow estimation of other statistical parameters of interest.
Fig 3.8: The gamma distribution for copper/glass, Dp = 45-47 μ, 5% RH. above separate runs, below the average values.
Fig 3.9: Effect of pressing-on: copper/glass, Dp= 45-47 μ, 5% RH, Log-Normal distribution
Fpressing/Fg: x = 560, ● = 106, o = 1

Fig 3.10: Effect of pressing-on: copper/glass, Dp= 54-56 μ, 5% RH, Log-Normal distribution,
Fpressing/Fg: x = 560, o = 1
3.3.3 Model discrimination

To estimate the standard deviation triple measurements have been conducted with glass/copper conditioned above silica gel. See table 3.C.3 and 3.C.4. See fig. 3.8.

The largest variation occurs near the median force, the standard deviation proportional to the probability of a force. However for the fitting of the probability models this correlation has been neglected and all points have been given equal weight. This effectively results in a relatively low weight to the points in the extremes.

Fitting the cumulative curves with a least square criterion yielded the sum of residues. Using an F-test all three models proved to be adequate.

The highest variance on the median is about 6%, the mean variance 3%. The chi-square for the total set is of the same order of magnitude.

Fig 3.11: The mean and median values of the Gamma and Log-Normal distributions of the forces of separation of copper spheres from glass plates.
Statistical test could not discriminate between the models; no model is significantly better than the others. However for all the measurements the Weibull distribution resulted in the highest chi-square value and can be identified as the least performing model. In the further discussion this model has been discarded and only Gamma and Log-Normal models considered. see table 3.C.6 and 3.C.7

The two models of the distribution will result in different estimates of the distribution parameters. Comparing the estimates of Gamma and Log-normal the following trends can be noted:

- F modal: Log-normal < Gamma
- F median: Log-normal < Gamma
- F mean: Log-normal > Gamma
- st.dev.: Log-normal > Gamma

The least stable parameter is the modal force, the most robust are the mean and median values.

3.3.4 the effect of particle size.

Using the measurements of the parameters for copper particles on glass plates at 5% RH, the effect of size can be compared to literature values. By plotting the median and mean values a certain trend relating force and size can be noticed. This trend does not extrapolate to the origin, however, and it cannot be concluded that force and size are directly proportional for the system in consideration. see fig. 3.11.

The estimates of the Hamaker model and of the Krupp correction for the deformation using 4 Å for the separation constant is too high for both median and mean values. A more suitable separation constant for this combination of materials is about 5.4 Å.

3.3.5 the effect of humidity.

The effect of humidity is profound and follows the general trend reported in the literature.

The mean separation force at 58 % RH is about twice as high as that at 5 % RH, reducing at the higher humidity. At 80 % RH the force is about 1.5 times as high as at 5 %.

Not only the mean value is affected, the standard deviation of the distribution changes drastically also. At 50 % RH the distribution is much broader than at 5 or at 84 %. See fig. 3.12.
Fig 3.12: The distribution of forces for different humidities
1 = 5% RH, 2 = 58% RH, 3 = 84% RH

Fig 3.13: The distribution of forces for glass and perspex
3.3.6 the effect of contact pressure.

To simulate the effect of consolidation due to external compressive forces the particles have been pressed onto the substrate by a known centrifugal force. This means the contacting force is also dependent on the mass of the individual particles. In the further discussion this variation has been neglected and a mean contacting force is reported. See table 3.C.9

The distribution of separation forces after consolidation could also be adequately represented with both Gamma and Log-normal distributions.

The effect of the contacting pressure is most noticeable for the particles which have contact of median strength. These contacts gained most in force. A number of comparative weak contacts still remained and the number of extremely strong contacts did not change appreciably. See figs. 3.9, 3.14.
3.3.7 the effect of hardness of the substrate

To investigate the effect of deformation of the contacts the results obtained for copper particles on both glass and and perspex were compared. In order to avoid influence of electrical forces only the measurements at 84 % RH will be discussed. In this case both Gamma and Log-Normal models are again adequate. As expected the mean and median force and the standard deviation of the distribution are all larger for the perspex/copper system. See fig 3.13.
3.4 Discussion and Conclusion

The theory of adhesion does not allow direct accurate estimation of the forces of interaction for particles. The exact morphology of the particles plays an extremely important role when they are in close proximity or contact (appendix 3.A). Only approximate values can be calculated for the mean value of the separation force due to vDWaals interaction between smooth spheres. In this work a method of calculating the correct interaction energies between different materials is proposed (appendix 3.B).

For models the entire distribution of forces is important. This distribution can only be obtained experimentally. A number of approximate functions for the distribution of separation forces of relatively smooth copper spheres and glass and perpex plate have been examined.

The use of an image analyser to simultaneously count and classify the particles into narrow size classes permits more accurate measurements, and obviates the need to classify the particles into narrow fractions before the analysis. At least 400 particles are counted in each size class. To achieve this about twenty fields of view on each plate had to be scanned.

model discrimination

Three models have been fitted with a least square criterion and all three were adequate according to the F-test. Although statistical methods did not reject any as significantly worse the Weibull distribution had the largest chi-square value of all models for each test conducted. Gamma and Log-Normal models are therefore more likely candidates. These distributions appeared to be adequate for all the conditions tested. These models differ in their estimates of the percentage of strong contacts, their mean and modal values and standard deviation. Estimates for the median and mean values were most robust.

Since the physical basis of the models cannot be yet clarified, depending on the particular interest either of these models may be chosen. This is of great importance for the modelling of the forces in multi particle systems. For some calculations the gamma distribution has convenient properties which allow simple formulas to be derived.

pressing on

Concerning the effect of consolidation on the modal force the following can be speculated. Suppose the distribution contains three types of contacts: normal, chaotic contacts, stronger contacts that happen to fit perfectly, and weak contacts which are disturbed by intervening asperities or dust particles.

When the normal, chaotic contacts are pressed on the plate, the small asperities in the contact are flattened and the area of
contact is permanently enlarged. This results in an increase of the separation force. The stronger particles already fit in perfectly and additional pressure does not increase their area of contact and thus force to the same degree as is the case for the normal contacts. The irregularities causing the weak contacts are large and not destroyed by the consolidating force, thus, these contacts also increase little in their separation force.

effect of humidity

The mean force measured at different values of humidity varies in accordance to the trends published in literature. For both perspex/copper and glass/copper systems the separation force is higher at 50 % RH than at 5 % RH. This can be explained by assuming that capillary condensation occurs in the pores of the contact and on the contact line itself at humidities below 50 % RH. The decrease in force at much higher humidities can be explained with two hypotheses. After the initial filling of the pores and condensation on the contact line further condensation fills up the space between the particles and thus decreases the vdwAals forces due to shielding. The second possible explanation is that the radius of the liquid bridge decreases as more water is adsorbed. The penetration of water layers cannot account for this decrease since this could occur only if other interactions stronger than the vdwAals-forces would compensate.

The form of the distribution also changes with humidity. The effect is analogous to that for the contacting pressure experiments. The weak contacts are not eliminated and the stronger contacts do not increase as much as the normal ones. Assuming the three classes of contacts this can also be used to explain the humidity effect: weak contacts have large pores and condensation does not occur easily, strong contacts have no pores and condensation can only occur at the line of contact. The majority of the contacts have meso and micro capillaries and condensation will influence the separation force greatly.

hardness

The difference in hardness of the substrate affects the force distribution of the contacts. The standard deviation for the softer contacts is larger and weak contacts are not eliminated. This leads again to the proposition that, for some particles, large irregularities which do not flatten weaken the strength of the contacts.

The conclusion is that the lower values of the broad distribution of forces are mainly due to abnormal contacts, formed by chance fitting of the roughnesses or by interfering irregularities. These exceptionally weak and strong contacts are not much influenced by humidity, pressing on or hardness. The overall distributions can, however, be satisfactory described by both Gamma and Log-Normal distribution functions.
### 3.5 Symbols

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<thead>
<tr>
<th>Symbol</th>
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<td>A</td>
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<td>[J]</td>
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<td>a</td>
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<td>[m]</td>
</tr>
<tr>
<td>ao</td>
<td>length parameter</td>
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<td>[Pa]</td>
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**Greek**

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<td>[1/s]</td>
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<td>[J/m^2]</td>
</tr>
<tr>
<td>σ0</td>
<td>surface energy of a plane interface</td>
<td>[J/m^2]</td>
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**Subscripts**

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<tr>
<td>modal</td>
<td>modal value</td>
</tr>
<tr>
<td>median</td>
<td>median value</td>
</tr>
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<td>Attraction calculated according Hamaker theory</td>
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<tr>
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<td>van der Waals forces</td>
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**Abbreviations**

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<th>Description</th>
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<td>RH</td>
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<td>st.dev.</td>
<td>standard deviation</td>
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<td>var.</td>
<td>variation</td>
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<tr>
<td>SSRes</td>
<td>som of squares of residues</td>
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3.6 References


[26] Visser J. Adv., Comparison between Hamaker constants and Lifshitz-van der Waals constants, Colloid Interface Sci. 3 (1972) 331-63


Appendix 3.A

Simulation of rough objects

When calculating the vDWaals forces between real particles the effect of roughness on the forces has to be taken into account. See e.g. Van den Tempel (1972).

In this appendix the interaction of a rough half space and a smooth half space is investigated. The rough half space is represented by the mean porosity at a certain distance from the smooth plate. For the bulk of the space the density is one, where there is no material the density is nil:

\[ \epsilon = F(z) : \begin{cases} 
\epsilon = 0 & z = H_c \\
\epsilon = 1 & z = H_b 
\end{cases} \]  
\[ \vdots \quad (3.A.1) \]

The mean distance between the spaces can be calculated by:

\[ H_{mean} = (H_c - H_b) \int_{H_c}^{H_b} \epsilon(z) \, dz + H_b \]  
\[ \vdots \quad (3.A.2) \]

The interaction of a porous thin plate, \( dz \), with a half space is equal to

\[ P_{vdW} = \epsilon \frac{h w}{8\pi^2} \frac{3}{z^4} \, dz \]  
\[ \vdots \quad (3.A.3) \]

Integration along the distance results in the following expression for the total pressure interaction between the rough and smooth plates:

\[ F_{vdW} = \frac{3hw}{8\pi^2} \int_{H_c}^{H_b} \epsilon(z) \frac{z^{-4}}{z^4} \, dz + \frac{hw}{8\pi^2} \frac{1}{H_b^3} \]  
\[ \vdots \quad (3.A.4) \]

From this formula it is seen that the pressure can not be calculated from the mean distance between the plates.

For example, for a roughness which can be represented as a square wave, it follows that:

\[ \epsilon(z) = 1 \quad z > H_b \]  
\[ \epsilon(z) = c \quad \]  
\[ \epsilon(z) = 0 \quad z < H_c \]  
\[ \vdots \quad (3.A.5) \]

\[ d \text{ roughness} = (H_b - H_c) \]  
\[ \vdots \quad (3.A.6) \]

\[ H_{mean} = (1-c) \cdot H_b + c \cdot H_c \]  
\[ \vdots \quad (3.A.7) \]

\[ P_{vdW} = \frac{hw}{8\pi^2} \left| \frac{(1-c)}{H_b^3} + \frac{c}{H_c^3} \right| \]  
\[ \vdots \quad (3.A.8) \]

for \( c = 0.5 \)
The effect of roughness can be further illustrated by calculating the distance at which the rough space can be approximated by a smooth one. For this the ratio of the rough and the smooth solutions should be close to unity:

\[
S = \left| \frac{0.5H_{\text{mean}}^3}{(H_{\text{mean}}+d/2)^3} \right| + \left| \frac{0.5H_{\text{mean}}^3}{(H_{\text{mean}}-d/2)^3} \right|
\]

\[
= 0.5 \left( \frac{1}{1 + \frac{d}{2H_{\text{mean}}}} \right)^3 + \frac{1}{1 - \frac{d}{2H_{\text{mean}}}}^3
\]

if \( d = 10 \text{ Å} \) and \( H_{\text{mean}} = 10 \text{ Å} \) \( S = 4.1 \)

\( H_{\text{mean}} = 50 \text{ Å} \) \( S = 1.04 \)

At close range the roughness is the dominating factor, even for a very small roughness the mean value can be used only at relatively large distances.

The rough plates can conveniently be modelled using the density function. Although this approach is also valid for rough spheres, another model is very illustrative. Suppose the surface of the sphere is covered with much smaller sferes. Since the sphere making contact will dominate the entire interaction only this is considered.

The force between the large and the small sphere and a smooth plate is equal to:

\[
F = \frac{A}{6} \left[ \frac{R}{Hb^2} + \frac{\delta}{(Hb-2\delta)^2} \right]
\]

Assuming the particle to be large: \( R=100\mu \) and the roughness to be small, \( \delta=0.1 \mu \), \( S \) is the ratio of forces for the the rough and the smooth sphere.

\[
S = 1 + \frac{0.1 (x+0.2)^2}{100 x^2}
\]

\( x= Hb-2\delta = \text{distance between solids (} \mu \text{)} \)

For distances of the order of 100 Å, \( S \) can be approximated by:

\[
S = 1 + 4 \times 5/ x^3
\]

This calculation shows that the roughness already contributes 1 %
of the total force at a distance of 600 Å. For a close approach we can take a distance of \( x = 4 \) Å and this results in a value of \( S = 250 \).

The roughness is thus so dominant that Krupp suggested that, for rough particles, the radius of the asperities should be used rather than the radius of the particles. This radius he estimated to be about 0.1 \( \mu m \), smaller asperities are more likely to be flattened.

If this suggestion is followed it results in a low estimate of the adhering force. A more realistic estimate considers that if these asperities do not flatten the particle is more likely to touch at three points for a stable contact.
Appendix 3.B: Estimation of the Hamaker constants in multi phase systems

An accurate estimate for a correction factor relating the Hamaker formula and the graphical integration procedure can be found in the following way.

rewrite the complex spectrum with:
\[
\frac{\varepsilon_1(i\theta) - 1}{\varepsilon_1(i\theta) + 1} = \frac{G_1(i\theta)}{1 + G_1} \quad \text{(3.B.1)}
\]

\[
\Rightarrow \varepsilon_1(i\theta) = \frac{(1 + G_1)}{(1 - G_1)} \quad \text{(3.B.2)}
\]

by definition:
\[
\int_{0}^{\infty} G_1G_1 \, d\theta = w_{11} \quad \text{(3.B.3)}
\]
\[
\int_{0}^{\infty} G_1G_2 \, d\theta = w_{12} \quad \text{(3.B.4)}
\]

substituting this into the general formula for the approximate interaction constant for 2 different materials with an intermediate third medium results in:

\[
w_{132} = \left[ \int_{0}^{\infty} \frac{1 + G_1}{1 - G_1} \cdot \frac{1 + G_3}{1 - G_3} \cdot \frac{1 + G_2}{1 - G_2} \cdot \frac{1 + G_2}{1 - G_3} \, d\theta \right] - \\
\left[ \int_{0}^{\infty} \frac{1 + G_1}{1 - G_1} \cdot \frac{1 + G_3}{1 - G_3} \cdot \frac{1 + G_2}{1 - G_2} \cdot \frac{1 + G_3}{1 - G_3} \, d\theta \right] \quad \text{(3.B.5)}
\]

This formula cannot be solved analytically but maximum and minimum value can be estimated:

\[
w_{132} \, \text{Low} = w_{12} - w_{23} - w_{13} + w_{33} \quad \text{(3.B.6)}
\]
w_{132} \text{ High } = \frac{w_{12} - w_{23} - w_{13} + w_{33}}{(1 - a_{1a3})(1 - a_{2a3})} ...(3.B.7)

In these solutions \(a_1, a_2\) and \(a_3\) are the maximum values of \(G_1, G_2\) and \(G_3\).

The minimum value is equal to that given by the classical formula for the Hamaker constant. Visser already noted that this estimate does not agree with the results of a graphical integration of the spectra substituted into formula [3.B.5]. He proposed to correct this discrepancy with a constant factor, dependent only on the intermediate material. This is only an approximate correction and not completely satisfactory. In mathematical form, the correct factor would be equal to:

\[
C = \frac{\int_0^\infty \frac{G_1G_2 - G_2G_3 - G_1G_3 + G_3G_3}{(1 - G_1G_3) \cdot (1 - G_2G_3)} \, d\theta}{\int_0^\infty \left( G_1G_2 - G_2G_3 - G_1G_3 + G_3G_3 \right) \, d\theta} \ldots (3.B.8)
\]

Because (3.B.5) can be split into separate contributions a more appropriate way of correcting would be:

\[
A_{132} = C_{12}.A_{12} - C_{23}.A_{23} - C_{13}.A_{13} + C_{33}.A_{33} \ldots (3.B.9)
\]

In this formula each, correction factor can be found by:

\[
C_{ij} = \frac{\int_0^\infty \frac{G_iG_j}{(1 - G_iG_3) \cdot (1 - G_jG_3)} \, d\theta}{\int_0^\infty G_iG_j \, d\theta} \ldots (3.B.10)
\]

Approximation of the spectra

Approximating the spectra each \(G\) can be represented with:

\[
G_1 = a_1 \exp \left\{ -b_1 i\theta \right\} \ldots (3.B.11)
\]

\[
a_1 = \frac{\epsilon_{10} - 1}{\epsilon_{10} + 1} \ldots (3.B.12)
\]
The factor $b_1$ can be chosen such that the following formula is still valid:

$$w_{ll} = \int_0^\infty G_{ll} \, d\theta = a_1 \, d_1 / 2b_1 \quad \ldots(3.B.13)$$

$$=> b_1 = a_1 \, d_1 / 2w_{ll} \quad \ldots(3.B.14)$$

The true function will be higher for low values of $G$ and, conversely, lower for the high values. This causes estimates for the correction factor to be low.

Using (3.B.13-14) the denominator of formula (3.B.10) can be rewritten as:

$$\int_0^\infty \frac{a_1a_2 \exp(-((b_1+b_2)x))}{(1-a_1a_3 \exp(-(b_1+b_3)x))(1-a_2a_3 \exp(-(b_2+b_3)x))} \, dx \quad \ldots(3.B.15)$$

For two similar materials this reduces to:

$$\int_0^\infty \frac{a_1a_2 \exp(-(b_1+b_2)x)}{1-2a_1a_3 \exp(-(b_1+b_3)x)+a_1^2a_3^2 \exp(-2(b_1+b_3)x)} \, dx \quad \ldots(3.B.16)$$

To this form no analytical solution is known. The most similar tabulated equation is:

$$\int_0^\infty \frac{r_1 \exp(-s_1 x)}{1-r_2 \exp(-s_2 x)} \, dx = \sum_{k=0}^{\infty} r_1 \cdot r_2 \frac{s_1+k\cdot s_2}{s_1+k\cdot s_2} \quad \ldots(3.B.17)$$

Approximating (3.B.10) with a function of this form leads to an estimation of the correction factor, $C$:

$$\int_0^\infty \frac{r_1 \exp(-s_1 x)}{1-r_2 \exp(-s_2 x)} \, dx = C \int_0^\infty r_1 \exp(-s_1 x) \, dx \quad \ldots(3.B.18)$$
it then follows that:

\[ C = \sum_{k=0}^{\infty} \frac{s_l \cdot r^2}{s_l + k \cdot s^2} \] \hspace{1cm} \ldots(3.B.19)

High and low estimates according to the form of formula (3.B.17) can be given for formula (3.B.15), \((b_1 < b_2)\):

\[
\int_{0}^{\infty} \frac{a_{l_2} \exp(-((b_1+b_2) x))}{1 - (a_{l_3} + a_{2a_3} - a_{l_2}a_{3_2}) \exp(-(b_1+b_3)x)} \, dx \quad (\text{low}) \ldots(3.B.20)
\]

\[
\int_{0}^{\infty} \frac{a_{l_2} \exp(-((b_1+b_2) x))}{1 - (a_{l_3} + a_{2a_3}) \exp(-(b_1+b_3)x)} \, dx \quad (\text{high}) \ldots(3.B.21)
\]

These are low and high approximations of the low estimate. The low approximation will always be low, the high estimate can be above or below the true value. However, the high approximation will be less than the absolute high estimate of (3.B.7):

\[(3.B.20-21) \text{ and } (3.B.18) \Rightarrow\]

\[ C_{ij} = \sum_{k=0}^{\infty} \frac{(b_i + b_j) \cdot (a_{l_3} + a_{2a_3} - a_{l_2}a_{3_2})}{(b_i + b_j) + k \cdot (b_1 + b_3)} \] \hspace{1cm} (low) \ldots(3.B.22)

\[ C_{ij} = \sum_{k=0}^{\infty} \frac{(b_i + b_j) \cdot (a_{l_3} + a_{2a_3})}{(b_i + b_j) + k \cdot (b_1 + b_3)} \] \hspace{1cm} (high) \ldots(3.B.23)

For the high values this finally leads to:

\[ C_{11} \approx 1 + \sum_{k=1}^{\infty} \frac{(a_{l_3} + a_{2a_3})}{1 + k/2 \cdot (1 + b_3/b_1)} \] \hspace{1cm} \ldots(3.B.24)

and

\[ C_{13} = C_{23} \approx 1 + \sum_{k=1}^{\infty} \frac{(a_{l_3} + a_{2a_3})}{1 + k} \] \hspace{1cm} \ldots(3.B.25)

\[ C_{33} \approx 1 + \sum_{k=1}^{\infty} \frac{(a_{l_3} + a_{2a_3})}{1 + k/2 \cdot (1 + b_1/b_3)} \] \hspace{1cm} \ldots(3.B.26)
table 3.B.1: values for the corrector $C_{ij}$ as a function of $b_i/b_j$ and $a_1a_2+a_2a_3$. For two similar media $a_1$ must be substituted for $a_2$.

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<th>Cij</th>
<th>1</th>
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<th>3/2</th>
<th>1/2</th>
<th>2</th>
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<td>4.28</td>
<td>2.91</td>
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For a number of materials the parameters of the spectra have been read from the graphs published by Visser. The vdWaals constant has been calculated using the new correction factors and compared with the ones Visser tabulates for the graphical integration.

table 3.B.2: Calculation of the correction factors using the parameters for various materials tabulated by Visser.

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<th>hw 10E-20 J</th>
<th>b' b' b'</th>
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<th>C13</th>
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<td>191</td>
<td>2.6</td>
<td>1.1</td>
<td>.6</td>
<td>1.5</td>
<td>1.5</td>
</tr>
<tr>
<td>diamond</td>
<td>0.7</td>
<td>119</td>
<td>2.1</td>
<td>0.88</td>
<td>.4</td>
<td>1.3</td>
<td>1.3</td>
</tr>
<tr>
<td>Al2O3</td>
<td>0.5</td>
<td>64.9</td>
<td>1.9</td>
<td>0.80</td>
<td>.3</td>
<td>1.3</td>
<td>1.3</td>
</tr>
<tr>
<td>MgO</td>
<td>0.5</td>
<td>44.4</td>
<td>2.8</td>
<td>1.16</td>
<td>.3</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>Polystyrene</td>
<td>0.45</td>
<td>27.8</td>
<td>3.7</td>
<td>1.5</td>
<td>.3</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>PMMA</td>
<td>0.45</td>
<td>26.4</td>
<td>3.8</td>
<td>1.5</td>
<td>.3</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>water</td>
<td>0.3</td>
<td>18.4</td>
<td>1.1</td>
<td>1</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>
It is noted that the value reported for Polystyrene by graphical integration may not be valid since the correction is larger than the maximum correction allowed:

\[ C_{\text{max}} = \frac{1}{1-0.45\times0.3} = 1.33 \]

This approach is also valid for media other than water; however the interval between the low and high estimates increases proportionally with the term \( a_3 \). The geometrical mean of the high and low estimates can be used as a reasonable value.

### table 3.B.4: Correctors for various intermediate media

- **C** Visser = 2.2  
  - C 11 H = 3.7  
  - L = 1.8  
  - mean = 2.6

- **Gold/polystyrene/Gold**  
  - C Visser = 1.8  
  - C 11 H = 2.5  
  - L = 1.6  
  - mean = 2.0

- **MgO/polystyrene/Gold**  
  - C Visser = 1.5  
  - C 11 H = 1.6  
  - L = 1.5  
  - mean = 1.5
Appendix 3.C: Tables of constants and measurements.

**Table 3.C.1: Relevant material constants (Krupp, Visser, Dahneke)**

<table>
<thead>
<tr>
<th>Material</th>
<th>hw</th>
<th>E</th>
<th>v</th>
<th>H*</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>-20</td>
<td>10</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>(10 J)</td>
<td>(10 Pa)</td>
<td>( - )</td>
<td>(MPa)</td>
</tr>
<tr>
<td>copper</td>
<td>119</td>
<td>13</td>
<td>0.34</td>
<td>100</td>
</tr>
<tr>
<td>glass</td>
<td>33.5</td>
<td>7</td>
<td>0.20</td>
<td>100</td>
</tr>
<tr>
<td>Perspex</td>
<td>26.4</td>
<td>.3</td>
<td>0.35</td>
<td>10</td>
</tr>
</tbody>
</table>

* approximate values

**Table 3.C.2: Relevant interaction constants. The interaction parameter for the case of intermediate water is calculated with the approximate formula for the graphical integration.**

<table>
<thead>
<tr>
<th>System</th>
<th>hw12</th>
<th>h132</th>
<th>C11</th>
</tr>
</thead>
<tbody>
<tr>
<td>copper/glass</td>
<td>63</td>
<td>13</td>
<td>1.3</td>
</tr>
<tr>
<td>copper/perspex</td>
<td>56</td>
<td>7</td>
<td>1.3</td>
</tr>
</tbody>
</table>

**Table 3.C.3: Prediction of separation forces using Krupp's modification of Hamaker's model as a function of hardness. z = 4 Å, hw = 55 e-20 J, F in mdyne (= 10^-8 N)**

\[ F = \frac{(hw/8\pi)}{(\pi H z3) \frac{D}{2z^2}} \]

<table>
<thead>
<tr>
<th>F (mdyne)</th>
<th>H (MPa)</th>
<th>Dp</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1000</td>
<td>100</td>
</tr>
<tr>
<td>45-47</td>
<td>348</td>
<td>657</td>
</tr>
<tr>
<td>47-49</td>
<td>364</td>
<td>686</td>
</tr>
<tr>
<td>49-51</td>
<td>379</td>
<td>715</td>
</tr>
<tr>
<td>51-53</td>
<td>394</td>
<td>743</td>
</tr>
<tr>
<td>54-56</td>
<td>417</td>
<td>786</td>
</tr>
<tr>
<td>56-58</td>
<td>432</td>
<td>814</td>
</tr>
</tbody>
</table>
### Table 3.C.4: Example of triplicate measurements of the cumulative force distribution, glass/copper, 45-47 μ, 5 % RH

<table>
<thead>
<tr>
<th>Fc</th>
<th>fraction</th>
<th>mean</th>
<th>width</th>
<th>st.dev.</th>
</tr>
</thead>
<tbody>
<tr>
<td>61.7</td>
<td>0.89</td>
<td>0.90</td>
<td>0.02</td>
<td>0.012</td>
</tr>
<tr>
<td>76.2</td>
<td>0.85</td>
<td>0.85</td>
<td>0.02</td>
<td>0.012</td>
</tr>
<tr>
<td>246.9</td>
<td>0.46</td>
<td>0.41</td>
<td>0.09</td>
<td>0.053</td>
</tr>
<tr>
<td>304.7</td>
<td>0.27</td>
<td>0.27</td>
<td>0.05</td>
<td>0.030</td>
</tr>
<tr>
<td>555.6</td>
<td>0.07</td>
<td>0.07</td>
<td>0.01</td>
<td>0.006</td>
</tr>
<tr>
<td>685.5</td>
<td>0.03</td>
<td>0.03</td>
<td>0.01</td>
<td>0.006</td>
</tr>
</tbody>
</table>

### Table 3.C.5: As 3.C.4, particle size 51-53 μ

<table>
<thead>
<tr>
<th>Fc</th>
<th>fraction</th>
<th>mean</th>
<th>width</th>
<th>st.dev.</th>
</tr>
</thead>
<tbody>
<tr>
<td>89.2</td>
<td>0.96</td>
<td>0.94</td>
<td>0.04</td>
<td>0.024</td>
</tr>
<tr>
<td>110.0</td>
<td>0.86</td>
<td>0.85</td>
<td>0.03</td>
<td>0.018</td>
</tr>
<tr>
<td>356.6</td>
<td>0.37</td>
<td>0.32</td>
<td>0.08</td>
<td>0.048</td>
</tr>
<tr>
<td>440.0</td>
<td>0.24</td>
<td>0.19</td>
<td>0.10</td>
<td>0.060</td>
</tr>
<tr>
<td>802.3</td>
<td>0.07</td>
<td>0.05</td>
<td>0.04</td>
<td>0.024</td>
</tr>
<tr>
<td>990.2</td>
<td>0.02</td>
<td>0.01</td>
<td>0.01</td>
<td>0.006</td>
</tr>
</tbody>
</table>

### Table 3.C.6: Example of F-test of three models of force distribution for particle size 51-53 μ.

<table>
<thead>
<tr>
<th>model</th>
<th>SSres/(n-p)</th>
<th>s²</th>
<th>Fcalc</th>
<th>F0.95</th>
</tr>
</thead>
<tbody>
<tr>
<td>weibull</td>
<td>8.3 10⁻⁴</td>
<td>9.1 10⁻⁴</td>
<td>0.9</td>
<td>3.26</td>
</tr>
<tr>
<td>gamma</td>
<td>6.0 10⁻⁴</td>
<td>9.1 10⁻⁴</td>
<td>0.7</td>
<td>3.26</td>
</tr>
<tr>
<td>log normal</td>
<td>6.0 10⁻⁴</td>
<td>9.1 10⁻⁴</td>
<td>0.7</td>
<td>3.26</td>
</tr>
<tr>
<td>a</td>
<td>b</td>
<td>24.6</td>
<td>19.3</td>
<td>29.7</td>
</tr>
<tr>
<td>-------</td>
<td>-------</td>
<td>------</td>
<td>------</td>
<td>------</td>
</tr>
<tr>
<td>2.2</td>
<td>2.3</td>
<td>5.3</td>
<td>8.0</td>
<td>56-58</td>
</tr>
<tr>
<td>0.6</td>
<td>0.7</td>
<td>1.1</td>
<td>1.6</td>
<td>54-55</td>
</tr>
<tr>
<td>0.6</td>
<td>0.6</td>
<td>6.3</td>
<td>8.3</td>
<td>51-53</td>
</tr>
<tr>
<td>8.8</td>
<td>8.5</td>
<td>3.3</td>
<td>3.3</td>
<td>47-49</td>
</tr>
<tr>
<td>7.0</td>
<td>7.6</td>
<td>1.3</td>
<td>7.6</td>
<td>45-47</td>
</tr>
</tbody>
</table>

*Table 3. C. 8: Parameters based on the Gamma and Log-normal distribution for copper/glass, 5% RH.*

<table>
<thead>
<tr>
<th>a</th>
<th>b</th>
<th>24.6</th>
<th>19.3</th>
<th>29.7</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.2</td>
<td>2.3</td>
<td>5.3</td>
<td>8.0</td>
<td>56-58</td>
</tr>
<tr>
<td>0.6</td>
<td>0.7</td>
<td>1.1</td>
<td>1.6</td>
<td>54-55</td>
</tr>
<tr>
<td>0.6</td>
<td>0.6</td>
<td>6.3</td>
<td>8.3</td>
<td>51-53</td>
</tr>
<tr>
<td>8.8</td>
<td>8.5</td>
<td>3.3</td>
<td>3.3</td>
<td>47-49</td>
</tr>
<tr>
<td>7.0</td>
<td>7.6</td>
<td>1.3</td>
<td>7.6</td>
<td>45-47</td>
</tr>
</tbody>
</table>

*Table 3. C. 7: Comparison of stresses (n-p) for all models.*
### Table 3.C.9: Effect of Consolidation on \( SS_{res/(n-p)} \), All Values in \( 10^{-3} \)

<table>
<thead>
<tr>
<th>F/Fg</th>
<th>1</th>
<th>106</th>
<th>560</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gamma</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>45-47</td>
<td>.16</td>
<td>.88</td>
<td>1.55</td>
</tr>
<tr>
<td>47-49</td>
<td>.63</td>
<td>.47</td>
<td>1.03</td>
</tr>
<tr>
<td>49-51</td>
<td>.57</td>
<td></td>
<td>0.75</td>
</tr>
<tr>
<td>51-53</td>
<td>.60</td>
<td></td>
<td>1.27</td>
</tr>
<tr>
<td>54-56</td>
<td>.01</td>
<td></td>
<td>1.83</td>
</tr>
<tr>
<td>Log N.</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>45-47</td>
<td>.70</td>
<td>.70</td>
<td>2.50</td>
</tr>
<tr>
<td>47-49</td>
<td>.88</td>
<td>.70</td>
<td>0.87</td>
</tr>
<tr>
<td>49-51</td>
<td>.35</td>
<td></td>
<td>0.38</td>
</tr>
<tr>
<td>51-53</td>
<td>.60</td>
<td></td>
<td>0.23</td>
</tr>
<tr>
<td>54-56</td>
<td>.06</td>
<td></td>
<td>1.40</td>
</tr>
</tbody>
</table>

### Table 3.C.10: The Distribution Parameters Derived From the Log-Normal Distribution

<table>
<thead>
<tr>
<th>Fc/Fg</th>
<th>1</th>
<th>106</th>
<th>560</th>
</tr>
</thead>
<tbody>
<tr>
<td>Log N.</td>
<td>Fmean</td>
<td>Fmod</td>
<td>Fmed</td>
</tr>
<tr>
<td>45-47</td>
<td>254</td>
<td>95</td>
<td>187</td>
</tr>
<tr>
<td>47-49</td>
<td>283</td>
<td>141</td>
<td>230</td>
</tr>
<tr>
<td>49-51</td>
<td>291</td>
<td>141</td>
<td>230</td>
</tr>
<tr>
<td>51-53</td>
<td>314</td>
<td>193</td>
<td>270</td>
</tr>
</tbody>
</table>
table 3.C.11: example of cumulative force distributions determined at 54 and 84 % RH, particle size 45-47 μ.

<table>
<thead>
<tr>
<th>RH</th>
<th>54%</th>
<th>84%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fc (mdyne)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.00</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>68.95</td>
<td>93</td>
<td>95</td>
</tr>
<tr>
<td>76.17</td>
<td>87</td>
<td>99</td>
</tr>
<tr>
<td>275.80</td>
<td>60</td>
<td>50</td>
</tr>
<tr>
<td>304.68</td>
<td>52</td>
<td>42</td>
</tr>
<tr>
<td>620.56</td>
<td>24</td>
<td>13</td>
</tr>
<tr>
<td>685.54</td>
<td>20</td>
<td>6</td>
</tr>
<tr>
<td>1103.22</td>
<td>4</td>
<td>3</td>
</tr>
<tr>
<td>1218.74</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>1334.89</td>
<td>3</td>
<td>-</td>
</tr>
<tr>
<td>1474.67</td>
<td>2</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 3.C.12: copper/glass, effect of humidity. 5, 58 and 84% RH (Gamma distribution) Forces and standard deviation in mdyne.

<table>
<thead>
<tr>
<th>RH</th>
<th>5%</th>
<th>58%</th>
<th>84%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fmean</td>
<td>Fmed</td>
<td>σ</td>
<td>Fmean</td>
</tr>
<tr>
<td>45-47</td>
<td>245</td>
<td>207</td>
<td>181&quot;</td>
</tr>
<tr>
<td>47-49</td>
<td>265</td>
<td>237</td>
<td>167&quot;</td>
</tr>
<tr>
<td>49-51</td>
<td>300</td>
<td>253</td>
<td>193</td>
</tr>
<tr>
<td>51-53</td>
<td>305</td>
<td>267</td>
<td>195&quot;</td>
</tr>
<tr>
<td>54-56</td>
<td>324</td>
<td>305</td>
<td>171&quot;</td>
</tr>
<tr>
<td>56-58</td>
<td>355</td>
<td>330</td>
<td>227&quot;</td>
</tr>
</tbody>
</table>

" = mean of three runs
Table 3.4: Parameters of the Gamma distribution

<table>
<thead>
<tr>
<th></th>
<th>Mean</th>
<th>Pmed</th>
<th>pmed</th>
<th>35-58</th>
<th>54-56</th>
<th>50-53</th>
<th>49-51</th>
<th>47-49</th>
<th>45-47</th>
</tr>
</thead>
<tbody>
<tr>
<td>26</td>
<td>6.6</td>
<td>71.0</td>
<td>5.0</td>
<td>9.5</td>
<td>4.3</td>
<td>1.3</td>
<td>1.1</td>
<td>1.14</td>
<td>1.14</td>
</tr>
<tr>
<td>9.3</td>
<td>6.8</td>
<td>8.5</td>
<td>0.0</td>
<td>5.6</td>
<td>6.4</td>
<td>4.4</td>
<td>4.4</td>
<td>3.3</td>
<td>3.3</td>
</tr>
<tr>
<td>16</td>
<td>6.9</td>
<td>6.9</td>
<td>0.0</td>
<td>9.0</td>
<td>0.0</td>
<td>9.0</td>
<td>0.0</td>
<td>3.3</td>
<td>3.3</td>
</tr>
<tr>
<td>19</td>
<td>6.4</td>
<td>6.9</td>
<td>0.0</td>
<td>0.0</td>
<td>6.4</td>
<td>1.0</td>
<td>4.0</td>
<td>1.14</td>
<td>1.14</td>
</tr>
<tr>
<td>21</td>
<td>6.0</td>
<td>6.8</td>
<td>0.0</td>
<td>6.4</td>
<td>9.0</td>
<td>6.4</td>
<td>4.0</td>
<td>4.0</td>
<td>4.0</td>
</tr>
</tbody>
</table>

Table 3.13: Copper Perspex, 84% RH.
Fig 4.1: Describing the position of a particle 3 reference points must be defined. The motion of the particle can then be described using the vectors pointing from the old position of these reference points to the new one.

Fig 4.2: The spatial contact distribution can be described using the centre of the particle and the vectors to the contacts, their radius and the normal vector to the plane of contact.
Chapter 4. Microscopic description of multi-particle systems.

When we examine the processes within a powder microscopically, we are investigating the processes at the scale of the individual particles. At this level the exact morphology of the particles, their displacements, the forces on the contacts and the deformation of the particles themselves are important.

Without any understanding of the processes on a microscopic scale, the advances on the macroscopic scale can only be limited, and those models more correlating than explaining. The literature on microscopic models is, however, much less substantial than that for the macroscopic correlations, and a firm ground for building these models is not yet available. For these reasons the microscopic parameters are here defined in a rather elaborate way. Not all these definitions are used, but it is hoped that by explicitly defining them approximations and assumptions can be pointed out. The definitions are close to the practice of computer programming, using arrays and building libraries of sets.

4.1 morphology of a packing

The morphology of a packing is defined as the set of the morphologies of the individual particles (see chapter 2) and their relative position.

To denote the relative positions of all particles in a powder a set must be defined, which contains the positions of three unique reference points for each particle. A possible choice of these vectors could be: 1) the vector to the centre of gravity and 2) the vector to the point on the periphery which is furthest away from the centre. 3) The third point could be chosen to be in a plane perpendicular to the axis of the first two points and furthest away from the centre. See fig. 4.1.

\[
\text{packing1} = \{ \text{rm} \ i \} \quad \{ \text{rm} + \text{rt1} \ i \} \\
\{ \text{rm} + \text{rt2} \ i \} \\
\{ \{ \text{form} \} \ i \} \quad \ldots(4.1.1)
\]

Contact set

When the interaction of the particles mainly exits through its contacts with its neighbours, the set of contacts describes the information relevant to that behaviour within a smaller set rather than the complete morphology set. See fig. 4.2. The contact sub-set for particle i contains the vectors from the centre of gravity to the centers of the contacts and the geometry of the contacts.
Assuming that the geometry of the contacts can be approximated with simple forms, the most simple case would be a circle with radius $R_c$. The normal vector $n_i$ also has to be stored.

\[
\{\text{contacts}\}_i = \{ c_1, c_2, \ldots, c_j \}
\]

\[
\{ n_1, n_2, \ldots, n_j \}
\]

\[
\{ R_1, R_2, \ldots, R_j \}
\]

4.2 deformation

The deformation of a packing may be defined as continuous, in time and space, in which both the morphology of the particles as their relative position may change.

If we limit the discussion to a deformation in which the basic morphology of the particles does not significantly change, the process can adequately be described with the continuous shift of the relative positions of the particles only. This allows for deformation of the contacts, but excludes large elastic deformation of the particles or breakage.

--- deformation of the geometry set.

The continuous deformation of a packing can be described with all changes in the individual positioning vectors.

(To limit the amount of notation of two dimensional formulas are shown.)

\[
\text{packing}(t+\Delta t) = \text{packing}(t) + \{ (v_{xm}, v_{ym}, v_{zm})_i \} \times \Delta t
\]

\[
\{ (v_{xt}, v_{yt}, v_{zt})_i \}
\]

\[
= \{ (r_{m} + d_{m})_i \}
\]

\[
\{ (r_{m} + r_{t} + \Delta t)_i \}
\]

\[
\{ \text{form } \}_i
\]

The displacements of the particles relative to the coordinate system is given by the displacements of the reference points of each particle:

\[
\text{Displacements} = \{ d_{m} i \}
\]

\[
\{ \Delta t \ i \}
\]
These displacements can be divided into a translation of the centre of gravity and a rotation of the particle around its centre:

\[ \text{Translation} = \frac{\Delta m}{\Delta t} \]  

\[ |\text{Rotation}| = \frac{\Delta t - \Delta m}{|\mathbf{r}|} \]

Although rotation does not contribute to mass flow across any boundary when the particle set is rearranged, energy is needed to perform the mutation in position. A simulation of the deformation of the morphology of a particle set, which assumes a homogeneous deformation field without allowing for these internal rotations, will neglect this portion.

4.3 forces

The forces in a packing are transmitted from one particle to the other through their common contacts. The forces can be internal molecular attractions, as was investigated in chapter 2, or forces imposed externally. The distribution of forces in the packing is generally extremely complex, as has been shown in numerous photoelastic experiments. A distinct network develops, with paths of high force intensity running through the packing. While particles in such a path are highly stressed, the particles in between can be completely unstressed.

This distribution of forces is inherent to particle systems and many properties of the system can only be explained using this distribution.

Forces can be represented as vectors. Taking the contacts as the coordinate system, normal forces and tangential forces can be distinguished:

\[ F_j = F_{nj} + F_{tj} \]  

\[ F_{nj} = F_j \cdot n_j \]  

\[ F_{tj} = F_j - F_{nj} \]

Although commonly neglected, torque and torsion forces also will be present. To approximate these forces the contact can be divided into a number of subcontacts, each with individual normal and tangential forces.

A set of forces on a particle(i) would be:

\[ \{ F \}_i = \{ c_1, c_2, ..., c_j \} \]

\[ \{ n_1, n_2, ..., n_j \} \]

\[ \{ F_{n1}, F_{n2}, ..., F_{nj} \} \]

\[ \{ F_{t1}, F_{t2}, ..., F_{tj} \} \]
4.4 forces and deformation

Whenever a single particle is displaced a counteracting force arises. The magnitude of this force is dependent upon the total number of particles that have to be displaced in this process. The expectation of the force is therefore dependent upon the morphology, the relative displacement and the resistance of the contacts against deformation. See fig. 4.3.

Particle displacement will be less the further away they are from the source of displacement. The intensity of forces will diminish as well. This reaction resembles the elastic media, in which displacement and force intensity also diminish with distance of the perturbation. For soil mechanics elastic media have been used extensively and successfully to model pressures in the earth. There are however a number of important differences that may not be neglected:

- the statistical distribution of forces and displacements
- non-connectivity of the particles
  note for instance the possibility of individual rotations of the particles. See fig. 4.4.

In modelling forces and displacements allowance for this non-connectivity leads to somewhat different results. See the calculations in chapter 6.

4.5 Statistical description

The description of the processes occurring in a particle set that keeps track of all the positions and forces on the particles clearly uses an enormous amount of data. To reduce this and to derive formulas which can be used by humans as well the processes can best be described with statistical models. Few distributions have been published however and where none are known we approximate with distributions that seem not unreasonable and capture at least the relevant form of the true distributions. All items discussed in the previous section are now reviewed again to model them with distributions.

morphology

The most common assumption about the morphology of the particle set is that it is built as a stacking of spheres. Several computer simulations have been published on the packing as a function of different size distributions. These algorithms use a uniform set of rules to place each particle and to build a packing sequentially. Compared to reality both the set of rules and the sequentiality will introduce artifacts. The magnitude of these is difficult to measure.
The distribution of a number of parameters can be determined: the number of contacts per particle and the distance between neighbours.

Non-spherical particles introduce many extra degrees of freedom, and necessarily will increase computer time. This inhibits research.

**Force field**

It must be noted that to approximate the distribution of forces not all forces are physically permitted. Combinations of normal and shear forces on the contacts will cause yield until the force is again relaxed to the critical value. The criteria for yield of a contact will also be distributed depending upon the uniformity of the materials and the morphology of the contacts.

The yield criteria inevitably cause the normal and shear force distributions to be correlated. This can be shown as follows:

First, assume a set of identical contacts, each controlled by a linear criterion. If the shear force does not surpass the yield limit, the contact is static.

\[
F_t \leq \alpha \left( F_n + F_c \right) \quad \cdots (4.5.1)
\]

\(F_c\) = internal attractive normal force

If we assume uncorrelated distributions for the normal and tangential forces and calculate the chance for a contact to yield at a tangential force, \(F_t\), we find:

\[
P \left( \text{yield} \mid F_t \right) = P \left( F_n + F_c \leq F_t/\alpha \right)
= \int_{0}^{F_t/\alpha} P \left( F_n + F_c \right) \quad \cdots (4.5.2)
\]

the chance of a contact to yield at a normal force, \(F_n\), is:

\[
P \left( \text{yield} \mid F_n \right) = P \left( F_t \geq \alpha \left( F_n + F_c \right) \right)
= \int_{\alpha \left( F_n + F_c \right)}^{\infty} P \left( F_t \right) \quad \cdots (4.5.3)
\]

It can be seen that chance of a contact yielding increases as the tangential force increases or the normal force decreases.
After letting these contacts yield, the original distributions will be altered for the probability of high shear forces and for low normal forces. The combination of high shear force and low normal force will not occur, and the separate distribution will be correlated. Alternatively, one could start with completely correlated distributions and assume:

$$F_t(i) = \beta (F_n(i) + F_c) \quad \ldots(4.5.4)$$

If $\beta$ is distributed between 0 and $\alpha$, the yield criterion is always satisfied. The assumption made here however, is even less likely than the previous one.

As a third alternative, the distribution of yield criteria can be assumed to be correlated to one of the distributions. See appendix 1 for examples.

More realistic assumptions cause the formulas to become very cumbersome, and for clarity in modelling it was further adopted to use uncorrelated distributions.

4.6 relationship between the microscopic and macroscopic description

One of the goals is to find the relations between microscopic distributions and the observed macroscopic parameters. The first step in connecting the discontinuous particle systems to the macroscopic description is to convert the system in a continuum. This can be done by integrating the variable in question over a volume and by assigning the density of the function to a point in the volume:

$$\bar{F} = \frac{1}{V} \iiint_V W(r) F(r, \phi, \theta) \, dr \, d\phi \, d\theta \quad \ldots(4.6.1)$$

Different weighing functions may provide some fine tuning, but this is usually neglected. In most cases a spherical volume is chosen and the result assigned to its centre. Choosing the correct volume of the sphere poses a difficult problem: if it is chosen too small the variation in two neighbouring points is too large to see it as a continuum. If, conversely, it is chosen too large no discrimination in the system is possible. A minimum diameter of 10 free path lengths seems to be the best compromise for gasses. For powder particle systems a volume containing approximately 1000 particles would compare.

The same formula can be used to calculate the volume expectation of a variable:

$$E(F) = \bar{F} \quad \ldots(4.6.2)$$
Fig 4.3: The displacement of one particle in a packing causes a wide distribution of forces and displacements in other contacts. Only average quantities are proportional to the distance to the source.

Fig 4.4: The average velocity in a plane parallel to the shear zone. Within the zone the motion of the particles is a combination of rotation and translation.
Fig 4.5: To obtain a continuous density field the volume fraction of solids within a sphere is ascribed to the centre of the sphere. For larger spheres the variation between points is smaller. For points near a boundary a proportional volume can be taken.

The expectation of a variable is not physically related to a volume. We can also calculate the expectation of a variable in a plane or along a line. See fig. 4.5.

\[ E(F) = \frac{1}{A} \int_A W(r) F(r, \phi, \theta) \, dr \, d\phi \quad \ldots (4.6.3) \]

\[ E(F) = \frac{1}{L} \int_L W(r) F(r, \phi, \theta) \, dr \quad \ldots (4.6.4) \]

This description is more flexible and is most useful for describing the variation of parameters when layers occur, as is the case in shear zones.
The thickness of a shear zone has been investigated with Gamma-rays by Scarlett et al.[5]. The zone appeared to narrow down to only several (about 10) particles in height. Clearly averaging over a volume of 10X10X10 would cause all small scale detail to be smoothed and the resulting parameters contain not much useful data. Using expectations for the planes parallel to the shear layer does not integrate over the height and preserves the detailed information.

4.6.1 The packing density

The entire complex morphology of the system is usually described as having only two parameters: the contact density and the volume fraction of solids. In this thesis the latter will be called the packing density of the system. See fig. 4.5.

\[ \bar{\epsilon}_s = \frac{1}{V} \iiint_V M(r,\phi,\theta) \, dr \, d\phi \, d\theta \quad \ldots(4.6.5) \]

\( M() \) is the function of occupation of space by solid matter.

The literature often uses the complement of the density rather than the density itself. There is no reason to choose one or the other but for the simplicity of the formulas. In powder mechanics the density is the more natural parameter, in fluidisation the porosity.

\[ \epsilon = 1 - \bar{\epsilon}_s \quad \ldots(4.6.6) \]

The density contains no information about the relative positioning of the particles in the packing. This is an important parameter. Molerus[1] has shown that the resistance of a shear plane is dependent upon the direction of deformation.

If spatial information is to be included, measurements of the positions of the particles must be done. A convenient way involves taking slices parallel to the shear plane. For parallel planes \( A \) a distance \( tz \) apart, the expectation of the spacial correlation can be introduced:

\[ E( \epsilon_{corr}(tx,ty,tz)) = \frac{1}{A} \iint_A M(x,y,z).M(x+tx,y+ty,z+tz) \, dx \, dy \quad \ldots(4.6.7) \]

If the packing is random this function reduces to a constant for sufficiently large \( tx,ty \) or \( tz \).

The processing time and the amount of data can be reduced by taking correlations only at a fixed distance. If the function
values are approximated with a ellipsoid, an unique tensor, describing the correlation, could be assigned to the plane.

\[ tx^2 + ty^2 = R^2, \quad tz = R \]  \hspace{2cm} \ldots(4.6.8)

\[ E(\epsilon_{corr}(tx,ty,tz)) = \epsilon \]  \hspace{2cm} \ldots(4.6.9)

Presently no detailed information about the morphology of shear planes is available, and the formula has not been tested on usefulness. Without any information about the morphology of the shear layer, only limited models are possible.

### 4.6.2 The contact density

The other macroscopic parameter for the morphology is the coordination number. This number is defined as the average number of contacts on a particle:

\[ n \]

\[ k = \frac{1}{n} \sum_{i=1}^{n} K(i) \]  \hspace{2cm} \ldots(4.6.10)

\( K(i) \) is the number of contacts on particle(i).

Computer simulations often yield a minimum number of three contacts on an individual particle, due to the particular set of rules used to make the particle arrangement. In real cohesive powders the minimum number for an individual can be as low as 1. The absolute minimum for the coordination number is equal to \((2n-2)/n\), in which formula \(n\) is the number of particles in the system. For large numbers of particles this reduces to 2.

The spread in the distribution of the number of contacts is related to the spread in the size distribution of the particles.
Fig 4.6: The average state of stress in a volume of powder can be expressed with a tensor if the forces on the sides of a box drawn around a point in the volume obey the following equalities: $\theta_{xz}=\theta_{zx}$, $\theta_{yz}=\theta_{zy}$, $\theta_{xy}=\theta_{yx}$.

4.6.3 Stresses

4.6.3.1 molecular forces and macroscopic stresses

To calculate the average stress on a plane through a particle set we first calculate the contribution of all molecular forces within one particle.

Taking a small volume around a point on the plane, the average force can be calculated with the stress tensor. Integrating along the entire area of intersection yields the total force due to this particle.
\[ F_i(x,y,z) = \iint_A \left( T(x,y,z) \cdot \mathbf{n} \right) \, da \quad \ldots(4.6.11) \]

Similarly, to calculate the total normal force of all particles on the plane, we can integrate \( T(x,y,z) \) over the entire surface; or, if interstitial forces are absent, sum the \( F(x,y,z) \) over all particles.

\[ \bar{F}_n = \sum F(x,y,z) \cdot \mathbf{e}_1 \quad \ldots(4.6.12) \]

\[ \bar{F}_x = \sum F(x,y,z) \cdot \mathbf{e}_2 \quad \ldots(4.6.13) \]

\[ \bar{F}_y = \sum F(x,y,z) \cdot \mathbf{e}_3 \quad \ldots(4.6.14) \]

The average stresses are the forces divided by the entire surface

\[ \sigma_n = \frac{F_n}{A}, \quad t_x = \frac{F_x}{A}, \quad t_y = \frac{F_y}{A} \quad \ldots(4.6.15) \]

If an average stress tensor is to be calculated in a point in a particle system, the following procedure may be followed.
A box is drawn around the point and the stresses on the walls of the box are identified as the parameters of the stress tensor in that point. See fig. 4.6.
The stresses calculated for each face most likely will be different, the variation between facing sides larger when the box is smaller. This fundamental problem of choice of volume has been discussed in 4.1.4.

To overcome the problem of non-corresponding values for opposing planes in a second procedure the stress tensor may be defined as the average expectation of the stresses on only three sides. The forces on three planes which intersect at right angles are calculated and the average expectations for all three is assigned to the intersecting point. See fig. 4.7.

\[
\begin{array}{c|ccc}
\text{side 1 :} & F_{11} & F_{12} & F_{13} \\
\text{side 2 :} & F_{21} & F_{22} & F_{23} \\
\text{side 3 :} & F_{31} & F_{32} & F_{33} \\
\end{array}
\]

\[
\begin{array}{c|ccc}
T_p &=& F_{11}/A & (F_{12}+F_{21})/2A & (F_{13}+F_{31})/2A \\
&=& (F_{12}+F_{21})/2A & F_{22}/A & (F_{23}+F_{32})/2A \\
&=& (F_{13}+F_{31})/2A & (F_{23}+F_{32})/A & F_{33}/2A \\
\end{array}
\]

\ldots(4.6.18)
Fig 4.7: An alternative description of the stress tensor as the average stresses in three perpendicular planes (above). The forces across the planes are resolved in the direction of the coordinate system (below).
4.6.3.2 Contact forces and macroscopic stresses.

Since all forces are transmitted through the particle contacts, the integration of the forces across a mathematical plain can be simplified by directly calculating the projected forces from the forces on the contacts. In this calculation we assume the contacts and the normal and tangential forces to be homogeneously distributed.

When a static packing is intersected with a plane, all particles intersected are the transmitters of the force across that plane. Only contacts that the intersected particles share with non-intersected particles have to be considered.

If the packing is made of a random packing of monosized spheres, the expectation of the number of particles intersected can be calculated with:

\[ E(N/F) = \epsilon_s * / E(A_p) \]  
\[ E(A_p) = \pi D_p^2 / 6 \]  

The resultants of the forces on these contacts normal to the plane will be called the normal forces; those tangentially the shear forces. The sum of normal forces under and above the plain must be equal and opposite. The stress on the plain is equal to the sum of forces on one side divided by the area.

The average total number of contacts is given by the coordination number. The average number of 'external' contacts we will call the reduced coordination number. No estimates of the reduced coordination number are available, and we assume the internal particles to shield of 1/3 of the area available for external contacts:

\[ k' \approx 2/3 \cdot k \]  

We proceed and follow Rumpf[2-4] in his derivation of the relationship of the attractive forces and the overall breakage strength.

We divide the particle in a number of sectors of the magnitude of a contact: \( \delta_0 \). Looking at a single external contact, the chance that a free sector 'i' is chosen is:

\[ P(\text{sector } i) = \delta_0 / S_p \]  

\( S_p \) is the total surface of the particle.
Due to the normal force $F_n$ on this sector the normal pressure on the unit plain $A$, is equal to

$$Q(\text{sector } i) = \frac{F_n}{d\Omega} \cdot \frac{d\Omega}{A} \quad \text{(4.6.23)}$$

Since we assumed homogeneity, the magnitude of $F_n$ is independent of the orientation of the sector. The expected value of the magnitude of the pressure $Q$ on the surface $A$ is equal to:

$$\frac{Sp}{d\Omega} \sum_{i=1}^{N} |Q(\text{sector } i)| \cdot P(\text{ sector } i) \quad \text{(4.6.24)}$$

$$E(Q) = \frac{F_n}{d\Omega} \cdot \frac{d\Omega}{Sp} \cdot \sum_{i=1}^{N} |(d\Omega/A)|$$

$$= \frac{F_n}{Sp} \cdot \frac{2 \cdot A_{\text{Feret}}}{A} \quad \text{(4.6.25)}$$

in which formula $A_{\text{Feret}}$ is the Feret surface of the particle on the plane.
This formula deviates from Rumpf's original formula, because he assumed the surface to be equal to the particle diameter.

For a sphere, the projected Feret surface divided by the total surface is equal to:

$$\frac{A_{\text{Feret}}}{Sp} = \frac{1}{4} \Rightarrow \quad \text{(4.6.26)}$$

this reduces equation 4.6.25 to:

$$E(Q) = \frac{1}{2} \cdot \frac{F_n}{A} \quad \text{(4.6.27)}$$

The total stress on the plane is caused by all intersected particles, and is the result of the force on $k' \cdot N/2$ sectors. When we equate the expectation of all the sectors to (4.6.27), implicitly neglect the difference in orientation of the internal and external contacts.

The final formula for convex particles is:

$$\sigma = k' \cdot \frac{\varepsilon_s}{\pi} \cdot \frac{F_n}{A} \cdot \left( \frac{2 \cdot A_{\text{Feret}}}{Ap \cdot Sp} \right) \quad \text{(4.6.28)}$$

which for spheres reduces to:

$$\sigma = \frac{k' \cdot 3}{\pi} \cdot \frac{\varepsilon_s \cdot F_n}{D_p^2} \quad \text{(4.6.29)}$$

Substituting our guess for the reduced coordination number:

$$\sigma = \frac{2 \cdot k}{\pi} \cdot \frac{\varepsilon_s \cdot F_n}{D_p^2} \quad \text{(4.6.30)}$$
This results in a formula very similar to Rumpf's equation for tensile forces.

Due to the linear relation between macroscopic stress and microscopic force, the mean value of any possible distribution of these interparticle forces may be substituted.

\[ \sigma = k' \frac{3}{\pi} \epsilon_s \bar{F}_{n} \quad \text{Dp}^2 \]  \hspace{1cm} \text{...(4.6.31)}

Calculation with forces normal and tangential to the plane.

Instead of using forces normal and tangential to the contacts we can choose the intersecting plane as the reference system to define the normal and shear forces in the contacts. This facilitates the calculation of the relation between contact forces and macroscopic stress. However, the introduction of the yield criteria is somewhat more difficult.

Due to the normal forces on a particle the stress on the plane is:

\[ \sigma = k' \cdot \frac{N/A}{2} \cdot \bar{F}_{n} \quad \text{...(4.6.32)} \]

And it follows immediately that:

\[ \sigma = k' \frac{3}{\pi} \epsilon_s \bar{F}_{ns} \quad \text{Dp}^2 \]  \hspace{1cm} \text{...(4.6.33)}

A analogous reasoning for the tangential forces uses the mean value of the shear components. In this case the forces may cancel one another, and a zero mean is possible for a symmetrical distribution of the shear forces.

\[ \theta_{xy} = \frac{3}{\pi} k' \epsilon_s \bar{F}_{xy} \quad \text{Dp}^2 \]  \hspace{1cm} \text{...(4.6.34)}

\[ \theta_{xz} = \frac{3}{\pi} k' \epsilon_s \bar{F}_{xz} \quad \text{Dp}^2 \]  \hspace{1cm} \text{...(4.6.35)}

Yield of contacts

For a contact oriented at a inclination \( \beta \) to the plane, yield
will occur when

$$(\cos\beta Fts - \sin\beta Fns) > \mu (\cos\beta Fns + \sin\beta Fts + Fatt) \quad (4.6.36)$$

$$Fts > \frac{(\mu + \tan\beta)}{(1 - \mu \tan\beta)} Fns + \frac{\mu Fatt}{\cos\beta (1 - \mu \tan\beta)} \quad (\tan\beta < 1/\mu)$$

Only when all distributions are uncorrelated the Coulomb assumption also is valid for the average values:

$$\bar{Fts} > \mu' (\bar{Fns} + \frac{\mu}{(\mu \cos\beta + \sin\beta)} \bar{Fatt}) \quad (4.6.37)$$

and relating the averages to the stresses:

$$\theta > \mu' (\sigma n + \mu'' \sigma a) \quad (4.6.38)$$

However these parameters are no longer only material properties but also contain information about the morphology of the packing.

4.7 conclusions

In this chapter the relationship between the distributions of the microscopic properties and the macroscopic state have been investigated. Two methods were discussed. The first one averages the distribution over a volume large enough to damp the largest variations, the second calculates the expected value for a point in an intersecting plane. This latter method may be more meaningful since it has higher resolution when describing the parameters in thin shear planes.

The macroscopic stress generally can be calculated given a distribution of microscopic forces. Its inverse is not true, unless independent knowledge is available about form and the width of the distribution. Only when the distributions are completely determined can the state of the system be uniquely defined with the stress on the boundaries.

Assuming certain distributions and yield criteria, the resulting formulas are unwieldy and complex. It seems this territory is for computers only.

However, it can be shown that the non-uniformity of the forces has a profound influence on the macroscopic behaviour of a powder. The tensile strength of an agglomerate is less as the distribution of forces is larger for a certain mean interparticle attraction force.
4.8 symbols

A area \[ \text{[m]} \]
cl vector from centre to centre of contactpoint \[ \text{[m]} \]
a,b,d parameters in the distribution functions \[ - \]
da projected area of the contact \[ \text{[m]} \]
dm displacement of the centre \[ \text{[m]} \]
d0 area of contact \[ \text{[m]} \]
drt displacement of the referencepoint \[ \text{[m]} \]
dt time \[ \text{[s]} \]
e1,e2,e3 unit vectors \[ - \]
E() Expectation of \[ - \]
F force \[ \text{[N]} \]
Fn normal to the contact \[ \text{[N]} \]
Ft tangential to the contact \[ \text{[N]} \]
Fc attractive force normal to the contact \[ \text{[N]} \]
F(r,\phi,\theta) Forcedistribution in 3 dim space \[ \text{[N]} \]
Fns force normal to intersecting plane \[ \text{[N]} \]
Fts tangential force \[ \text{[N]} \]
Fxy interparticle forces in the coordinate system of the plane. \[ \text{[N]} \]
k coordination number \[ - \]
k' corrected coordination number \[ - \]
K() number of contacts on a particle \[ - \]
L length \[ \text{[m]} \]
M(r,\phi,\theta) Mass distribution in 3 dim space \[ - \]
n vector normal to the contactpoint \[ - \]
P() probability \[ - \]
Q force normal to the intersecting plane \[ \text{[Pa]} \]
r coordinate distance \[ \text{[m]} \]
rm vector from coordinatecentre to centre of gravity \[ \text{[m]} \]
rt1 vector from centre to referencepoint on contour \[ \text{[m]} \]
rt2 vector from centre to referencepoint on contour \[ \text{[m]} \]
R radius \[ \text{[m]} \]
R rotation of the particle around its centre \[ \text{[rad]} \]
T translation of the particle centre \[ \text{[m]} \]
V volume \[ \text{[m}^3] \]
v velocity \[ \text{[m/s]} \]
W weighing function \[ - \]

Greek

\( \alpha \) internal friction \[ - \]
\( \beta \) parameter \[ - \]
\( \Gamma \) Gamma function \[ - \]
\( \varepsilon \) \( 1-\varepsilon \) : porosity \[ - \]
\( \varepsilon s \) volume fraction of solids:density \[ - \]
\( \phi \) coordinate angle \[ \text{[rad]} \]
\( \Theta \) coordinate angle \[ \text{[rad]} \]
\( \mu \) internal friction \[ - \]
\( \sigma \) normal stress \[ \text{[Pa]} \]
4.9 References

4.A: examples of correllated distributions of forces

example a

Assuming the normal forces can be aproximated with a gamma function:

\[ P \left( \frac{F_n}{F} \right) = \frac{a}{\Gamma(a)} \left( \frac{F_n}{F} \right)^{a-1} \exp\left( -\frac{b}{F} \right) \] ...(4.A.1)

a distribution of shear forces after yield of a independent gamma function would be:

\[ P \left( \frac{F_t}{F} < \alpha \left( \frac{F_n + F_c}{F} \right) \mid \frac{F_n}{F} \right) = \Gamma(c,d',x) \] ...(4.A.2)

\[ P \left( \frac{F_t}{F} = \alpha \left( \frac{F_n + F_c}{F} \right) \mid \frac{F_n}{F} \right) = \int_0^\infty \Gamma(c,d',x) \, dx \] ...(4.A.3)

\[ P \left( \frac{F_t}{F} > \alpha \left( \frac{F_n + F_c}{F} \right) \mid \frac{F_n}{F} \right) = 0 \] ...(4.A.4)

This is not a very desirable solution for the distributions. Furthermore, even if we assume this distribution can again be approximated with a second independent gamma function the expression of the distribution of a related parameter, such as the total force on a point contact, is cumbersome.

\[ P \left( \frac{F_n}{F} = x \right) = \Gamma(a,b,x) \] ...(4.A.5)

\[ P \left( \frac{F_t}{F} = x \right) = \Gamma(c,d,x) \] ...(4.A.5)

\[ F = F_t + F_n \] ...(4.A.6)

\[ |F|^2 = |F_t|^2 + |F_n|^2 \] ...(4.A.7)
\[ P( F > x ) = \int_0^x P( F_n ) \cdot P( F_t > (x^2 - F_n^2) | F_n ) \, dF_n \]
\[ + \int_x^\infty P( F_n ) \, dF_n \]
\[ ...(4.A.8) \]

\[ P( F_t > \sqrt{(x^2 - F_n^2)}) = + \int_{\sqrt{(x^2 - F_n^2)}} \alpha(F_n + F_a) \Gamma(c, d, F_t) \, dF_t \]
\[ ...(4.A.9) \]

It seems that analytic solutions are not easily obtained, and that the work on statistical problems in particle systems will have to be carried out numerically by computers.

**example b**

If we suppose that the probability of a normal force which just matches the yield criterium is equal to:

\[ P( \text{yield} | F_n ) = \exp( -b' \cdot F_n ) \]
\[ ...(4.A.10) \]

the distribution of the shear forces is again deduced from a previously uncorrelated function that is changed because of yield of the weaker contacts:

\[ P(F_t) = P( F_t \text{ uncorr.} ) \cdot (1 - P(\text{yield} | F_t) + P( F_n | F_n = F_t/a ) \cdot P(\text{yield} | F_n) \]
\[ ...(4.A.11) \]

\[ = \]

\[ P(F_t) = P( F_t \text{ unc.} ) \cdot (1 - \int_{F_t/a}^\infty P(F_n) \, dF_n ) \]

\[ + P( F_n | F_n = F_t/a ) \cdot P(\text{yield} | F_n) \]
\[ ...(4.A.12) \]

substitution yields:

\[ P(F_t) = \frac{a_1}{\exp(-b_1x)} \int_{F_t/a}^\infty \frac{a_2}{\exp(-b_2x)} \, dx + \frac{a_3}{\exp(-b_3x)} \]
\[ ...(4.A.13) \]

The result of which is not much less complex than that of example a.
4.B: distributions of forces on uniaxial strength measurements

Performing a hypothetical, perfect, uniaxial tensile strength measurement, a bar of powder is uniaxially stretched. The resulting overall normal force per area shall be called the tensile stress.

We will approximate the distribution of microscopic forces with gamma functions. First we assume that the change of sustaining a maximal force \( F \), normal to the endplatens, on a pointcontact is given by:

\[
P(\text{contact}=F) = \frac{a}{\Gamma(a)} F^a \exp(-bF) \quad \ldots(4.B.1)
\]

\[
F_{\text{mean,contact}} = \frac{a}{b} \quad \ldots(4.B.2)
\]

Second, the relation between the average of the distribution of actual forces and the overall stress has been shown above and is:

\[
\sigma = \frac{k' \varepsilon s}{\pi} F_{\text{mean,}/Dp^i} \quad \ldots(4.B.3)
\]

Assuming a distribution for the actual forces, it is highly unlikely that the first distribution matches the second in such a way that all contacts will sustain until they all break at the same time.

Rather it will happen that a combination of a high force will break a weak contact and after redistributing the microscopic forces the new packing will be stable as long as enough contacts remain.

We approximate the changes of encountering a force higher than the strength of the contact with an exponential function.

\[
P(\text{Fact}>F) = \exp(-dF) \quad \ldots(4.B.4)
\]

\( d=\infty \) : no failure

\( d=0 \) : total failure

The fraction of broken contacts is calculated by:

\[
f_{\text{broken}} = \int_{0}^{\infty} P(F) \cdot P(\text{Fact}>F) \, dF
\]

\[
= \frac{a}{b+d} \quad \ldots(4.B.5)
\]
The mean strength of the remaining particle can be written as:

\[
P( F ) = \int_0^\infty \frac{a}{F} \left| \exp(-bF) - \exp(-(b+d)F) \right| dF
\]

\[
= \frac{a}{b \Gamma(a)} \left| F \exp(-bF) - F \exp(-(b+d)F) \right|
\]

\[
\Gamma(a) \left( 1 - \frac{b}{b+d} \right)^a
\]

\[
F_{\text{gem}} = \frac{a}{b} \left| \frac{1 - \frac{b}{b+d}}{1 - \frac{b}{b+d}} \right|^{a+1}
\]

It is clear that the average strength of the remaining contact increases due to the preferential breakage of the weaker ones. Nevertheless the product of the fraction of remaining contacts and their average strength is lower than that calculated if it is assumed that all contacts separate at the same instant:

\[
F_{\text{break}} = \text{fraction remaining} \times \text{Faverage}
\]

\[
= \left( 1 - \frac{b}{b+d} \right)^a F_{\text{gem}}
\]

\[
F_{\text{break}} = \frac{a}{b} \left| 1 - \frac{b}{b+d} \right|^{a+1} < a = F_{\text{gem original}}
\]
The uniaxial tensile test can now be described semi-quantitatively. As the stress increases from nil the breakage parameter 'd' increases as more contacts gradually break. At a certain critical point the increasing stress can no longer be redistributed, and the whole packing fails. The maximal stress is now a function of the parameters of the distribution of contact attraction forces and the breakage parameter. This latter parameter is dependent upon the way the induced forces are transmitted through the packing.

\[
\sigma_{\text{max}} = \frac{k' \epsilon_s}{\pi} \left| \frac{a}{b} \right| \left| 1 - \frac{b}{b+d} \right| / Dp^2 \quad \ldots (4.\text{B}.12)
\]

\[
= \frac{k' \epsilon_s \text{Fbreak}}{\pi Dp^2} \quad \ldots (4.\text{B}.13)
\]

If we assume the breakage parameter is solely a function of the morphology of the packing, an overall attractive force for the powder can be defined.

\[
\sigma = \frac{a}{b^2}, \quad \text{Fmean} = \frac{a}{b} \quad \ldots (4.\text{B}.14)
\]

\[
a = \frac{\text{Fmean}^2}{\sigma} \quad \ldots (4.\text{B}.15)
\]

\[
b = \frac{\text{Fmean}}{\sigma} \quad \ldots (4.\text{B}.16)
\]

\[
\text{Fbreak} = \text{Fmean} \left| 1 - \frac{\frac{\text{Fmean}}{\text{Fmean} + \sigma}}{(\text{Fmean}^2 + \sigma)/\sigma} \right| \quad \ldots (4.\text{B}.17)
\]

For some magnitudes of the parameters we can calculate that:

\[
d = 1/\text{Fmean} \quad \ldots (4.\text{B}.18)
\]

\[
\sigma = \frac{\text{Fmean}}{d} \Rightarrow \text{Fbreak} = 0.5 \text{Fgem} \quad \ldots (4.\text{B}.19)
\]

\[
\sigma = \frac{\text{Fmean}}{2d} \Rightarrow \text{Fbreak} = 0.66 \text{Fgem} \quad \ldots (4.\text{B}.19)
\]

\[
\sigma = \frac{\text{Fmean}}{4d} \Rightarrow \text{Fbreak} = 0.67 \text{Fgem} \quad \ldots (4.\text{B}.19)
\]

A more narrow distribution of attractive forces exhibits a larger force if the breakage parameter is constant: less weak contacts will destabilise the structure.
In the case of a very narrow, almost monodispersive, distribution the following calculation can be made:

\[ \sigma << F_{\text{gem}}^2 \]  

... (4.B.19)

\[ F_{\text{break}} = F_{\text{gem}} \left| 1 - \frac{F_{\text{gem}}^2 / \sigma}{1 + \frac{d}{F_{\text{gem}}}} \right| \]  

... (4.B.20)

\[ \lim_{\sigma \to 0} F_{\text{break}} = F_{\text{gem}} \left| 1 - \exp\left(-\frac{d}{F_{\text{gem}}} \right) \right| \]  

\[ = F_{\text{gem}} \left( 1 - \exp(-d) \right) \]  

... (4.B.21)

\[ F_{\text{break}} \approx 0.67 F_{\text{gem}} \quad \text{d} = 0 \]  

\[ \approx 1/F_{\text{gem}} \quad \text{d} = \infty \]  

... (4.B.22)

Even for a monosized distribution the breakage force is less than the total of interparticle forces.

At this stage it is not yet possible to estimate any value for the parameters of the distributions or for the breakage parameter d. Furthermore, the distribution of forces will be spatially correlated since once a contact has been disrupted the redistribution of forces will concentrate on its immediate neighbours. Because of this concentration the neighbours also have a higher probability of breaking. This results in a local preferential breakage a process which we will further denote as a crack process. Because of these cracks, the maximal breakage force will even further diminish. See chapter 6.

The process of expansion of the hole occurs by definition on the tip of the crack. If we assume that this expansion occurs when the stress concentration on the tip exceeds the local breakage strength, the concentration along the front must be rather uniform.

There is no reason to assume that the morphology of the packing near the tip will vary much, so we also assume it to be uniform. All conditions for a uniquely defined state are thus satisfied. Further scientific research into the cracking mechanisms is encouraged. This seems the way to clarify the relation between microscopic interparticle forces and the macroscopic breakage force in tensile conditions.
Chapter 5. Macroscopic description of yield in multi-particle systems

5.1 Introduction

In the macroscopic approach one tries to find a direct correlation between the overall displacements and the overall forces on the boundaries of the volume which contains the particles. The justification for this approach is the fact that the many individual responses average out if enough particles are involved. The descriptive parameter can be seen as some statistical average of the distribution of responses. This average describes the state of the powder. The observed large variation in the macroscopic response of particle systems, as opposed to the seemingly deterministic response of multi-molecule systems, is explained by noting that usually much fewer particles than molecules are involved in an experiment.

A connection between the microscopic state and the macroscopic state is sought in formulas that average the microscopic properties in the proper way over a certain volume. If the number of particles is low the averaging is of little value, and macroscopic descriptions of volumes which contain less than a few hundred particles, is probably useless.

The macroscopic description is closely related to the quantities observed during experiments. The quantities are measured on the boundaries of the system, which are the walls of the apparatus. These can be rigid, for example made of metal, or flexible, when made of rubber membranes. The volume of the apparatus is typically many times the volume of the individual particles.

5.2 The macroscopic state of a particle system

In the science of soil mechanics an enormous amount of effort has been put in finding the relations between the deformation of large bodies of sand and clay and the forces exerted. The interest being primarily to prevent failure of constructions build on these materials. A compilation of all possible responses of these materials has been made by Hvorslev [4]. He published a plot of the responses in a three dimensional space of porosity, normal stress and shear stress. This concept has been elaborated by many people, one of the most important being Roscoe [7-8]. It must be realized that since porosity is only a very crude description of the morphology of the shear layer this plot is limited in its ability to explain the phenomena.

The crucial concept here is the formation of a uniquely defined state, attained when a shear zone in the powder is sheared at a constant rate. It is thought that, whatever the initial conditions, a constant response is obtained after prolonged
shearing. This critical state is then solely dependent on the normal compressive stress on the plane. All the critical states may be combined to form a line, which increases monotonically in the space of density, normal stress and shear stress. The assumption is then that all the parameters of the system, such as the parameters for morphology and microscopic forces, are constant and reproducible. However, since the response of a particle system is rarely constant, the requirement of constancy may be relaxed to stationarity. Stationarity is used in the sense that all statistical parameters of a signal are constant, but the actual signal may vary in a stochastic way around a mean. Experimental work has been carried out to determine the reproducibility and the stationarity of a cohesive particle system. This work is reported in chapter 7.

The projection of the critical state line onto the two dimensional plane, which is formed by the normal stress and shear stress, is known as the effective yield locus. Since, for these points, the response of the system is stationary, the more descriptive name 'stationary yield locus' is used in this thesis.

5.3 definition of the instantaneous and time yield loci

A yield point is defined to be the maximum shear stress at a certain compressive stress, $\sigma$, which is necessary to overcome the shearing resistance of a stationary shear plane, which has been previously formed under a certain higher, compressive stress, $\sigma_c$. 

\[
\text{yield point}(\sigma) = \Theta_{\max}(\sigma, \sigma_c) \quad \sigma_c > \sigma \quad \text{...(5.3.1)}
\]

The combination of all yield points for one compressive stress $\sigma_c$ is called the yield locus.

If the yield points are measured immediately after the stationary plane has been formed, the line is called the instantaneous yield locus. If the plane is consolidated under the compressive force for a certain time period before the yield point is measured, the time yield locus is obtained.

Usually the yield locus is situated above the stationary line, and the time yield locus is above the instantaneous yield locus.

\[
\Theta_{\text{stat}}(\sigma) \leq \Theta_{\max}(\sigma, \sigma_c, t=0) \leq \Theta_{\max}(\sigma, \sigma_c, t) \quad \text{...(5.3.2)}
\]

Various criteria have been tested for their usefulness in describing the macroscopic pressures during failure in powders, and a number of them seem adequate. Of these the Mohr-Coulomb is the most simple and, therefore, most attractive. It states that the powder will fail along a plane where the normal stress and shear stress are most critical. Using Mohr circles it can be shown that the combination of shear and normal stress which is most critical is situated on the
circle of minimum and maximum principal stress. The intermediate stress is, therefore, of no importance and is ignored in this analysis.

Photoelastic measurements of the stresses on a rod inserted in the shear zone by Mandl et al.[6] suggested that the Hill criterium might be the most suitable for a fully developed shear zone. In this criterium the intermediate stress is also assumed to be of no importance.

Cohesion as a means of characterisation

In general one can state that Cohesion is a monotonic, increasing function of density, consolidation pressure, and consolidation time. The state of the powder may not be completely defined since a combination of lower density but longer consolidation time may increase the cohesion. If time consolidation is neglected, one can characterize the powder system by its cohesion. The normal stress and density are then completely unique.

Technological importance of Cohesion

The characterisation of powders by their yield loci caused great interest when Jenike showed that the loci could be used for technological calculations. From the loci a new function can be derived, the flow function, which enables a prediction to be made whether a powder will flow or not under a compressive stress after being subjected to a certain history of stresses.

Assuming the Mohr-Coulomb mechanism of yield, the maximum principal stress during stationary deformation and the maximum principal stress for an unconfined plane can be calculated for a single yield locus. By plotting these two parameters, taken for a number of different yield loci onto a graph of maximum principal stress and unconfined normal stress the flow function is obtained. Cohesion is important since it defines the lower end of the yield locus, which is used to calculate the unconfined yield stress of the powder. A better defined yield locus will give more a reliable estimate of the unconfined yield stress and of the flow behaviour.
5.4 Cohesion and Tensile Strength

Cohesion is a macroscopic parameter, which is generally used to describe the degree of coherence of the particles in the system. It is defined in a operational way: that is, it is defined by the test done on the powder. If the test is carefully chosen the cohesion can be a meaningful parameter, useful to model the process in which one is interested.

In this thesis the Cohesion is defined as the shearing stress needed to overcome the flow resistance of a shear zone under zero compressive stress, that is it is defined to be equal to the yield point for $\sigma_2=0$:

$$C(\sigma_c) = \Theta_{\text{max}}(\sigma=0, \sigma_c, t) \quad \ldots(5.4.1)$$
However, none of the existing equipment used for yield determination measures this point accurately and it can only be obtained by extrapolating the yield locus obtained at higher compressive stresses to the axis.

No theoretical formula has been found to completely describe yield loci and a number of useful approximations have been proposed. The simplest of these is based on Coulomb's ideas:

$$\Theta_{\text{max}} = \mu \sigma + C$$ \hspace{1cm} ...(5.4.2)

Ashton et al. [1-2] proposed a more general formula, which allows for curved loci:

$$\frac{\Theta_{\text{max}}}{C} = \left(\frac{\sigma - T}{T}\right)^n$$ \hspace{1cm} ...(5.4.3)

Where $T$ is the intercept of the curve on the $\sigma$-axis.

If only a limited amount of data at the higher normal stresses is available, than due to the large variation in that data, often no distinction can be made between the estimate of cohesion according to the Coulomb or the Ashton formula.

To overcome this problem it was suggested that the intercept of the yield locus on the $\sigma$-axis is equal to the uniaxial breakage strength of the yield zone. This enables one to interpolate and achieve a higher reliability of the estimate of the parameters. To justify this it needs to be accepted that the mechanisms which lead to the peak values at positive principal stresses also dominate situations where a negative principal stress occurs. If an abrupt change in mechanism occurs the continuity of the extrapolation is most unlikely.

Looking at the definitions, the intercept represents the situation, in which simple tensile stress ruptures a shear plane without shearing force. In this situation the mechanism is breakage, and no shearing is involved. In the limiting case, in which the shear stress is negligible small, shear could theoretically occur. However, the shearing planes separate almost immediately on introduction of the shear stress.

The normal force just balances all interparticle forces holding the planes together before shear is applied. This situation is highly dominated by the breaking mechanisms as well. When both principal stresses are positive the shear plane is not ruptured during deformation and new contacts are formed during shear. Since the two mechanisms are so different an interpolation of all results with a single formula may not be allowed. To clarify this problem the two mechanisms are examined in detail both experimentally and mathematically.
Using the tensile strength measured in a movable plate tensile tester Ashton implicitly assumed this to be equal to the breakage stress. This assumption is discussed in chapter 6.

5.5 Experimental

The change in yield mechanism at different states of stress has been investigated in a new type of cell, the Hollow Dutch cell. When testing yield mechanisms in this region, where both tensile and compressive stresses must be applied to the powder sample, all the stresses on the boundaries of the machine must be very carefully controlled. Unknown shear stresses on the walls of the apparatus must be avoided. An arrangement has been found which introduces the tensile stress in such a way that most wall shear stresses are avoided.

To this aim a cell with coaxial cylindrical membranes was designed in which the pressure on the inner and outer membranes could be controlled individually. Since the membranes are attached to the cell walls rather than to the piston, this apparatus resembles the one build by Kleverling, and which is referred to as the Dutch cell test. Using this Hollow Dutch Cell theoretically all possible macroscopic states of stress can be introduced in a powder sample.

5.5.1 Description of the HDC.

The apparatus consists of two concentric cylindrical membranes, between which the powder sample is contained. See figs. 5.2 - 5.4. The height of the cell was chosen as long as possible to minimize the wall effects of the end platens.

The membranes are sealed at the top and bottom into rigid containers in which the pneumatic pressure can be varied in order to control the pressures on the membranes. The inner cylinder is restrained against inward displacement by a sturdy metal gauze.

The membranes are made of latex and are approximately 0.2 mm thick. They were made by spraying a latex covering onto a glass moulds. The membranes were covered with talc powder to reduce their stickiness and to protect them against ultraviolet radiation. The coefficient elasticity of these membranes is very low, the increased pressure due to expansion is less than 10 Pa and can be neglected.

The diameter of the inner cylinder is 6 cm, that of the outer 10 cm. The annular powder sample is therefore 2 cm in thickness and its length is 13 cm. Smaller samples can be made by inserting spacer rings.
Fig 5.2: Schematic drawing of the Hollow Dutch Cell.

- a inner membrane
- b outer membrane
- c load cell (Kistler 9203)
- d capacitive displacement meter
- e water manometers
- f pressure inlet
- g bleed control
- h piston
- s sample
- P1 pressure on inner wall
- P2 pressure on outer wall
- P3 pressure on vertical axis
The sample is supported on a ring connected to a force transducer, which records the vertical stress on the base. An additional vertical stress can be introduced on the top of the sample through a ring connected to a piston. On this piston weights can be placed, the resulting displacement is recorded using a capacitive displacement transducer.

The normal stress on both cylinder walls can be controlled independently by varying the gas velocity through two constrictions.
5.5.2 Operating the apparatus

Consolidation

Various consolidating stresses can be applied to the powder sample. For an isostatic consolidation, the pressure on the inner and outer cylinders is equalized and the average vertical stress is adjusted to the same value. For other consolidations the piston is locked to prevent all movement, and the pressure on the outer wall only is increased.

Filling

Filling the apparatus proved to be difficult. The length of the cylinder is too large and the membranes too flexible to allow one to permit filling the cylinder in one operation. A three stage filling procedure was therefore introduced. During each stage the cylinder was filled for 1/3 and a consolidating pressure was applied to the value of about half the final consolidating pressure. By this procedure a free standing uniform sample was formed on which further tests could be made.

testing

This apparatus can be used to make several measurements. Because of the ability to control all the pressures and to create tensile stresses in the cylinder wall, it combines the features of both a tensile tester and a triaxial cell.

The calculation of the stresses, which exists in the powder sample is based on the elastic theory for homogeneous materials. When in equilibrium the stresses depend only on the normal pressures on the cylinder walls:

\[
\sigma_r(r) = \frac{p_1 R_1^2}{R_2^2 - R_1^2} (1 - \frac{R_2^2}{r^2}) - \frac{p_2 R_2^2}{R_2^2 - R_1^2} (1 - \frac{R_1^2}{r^2}) \quad \ldots(5.5.1)
\]

\[
\sigma_\theta(r) = -\frac{p_1 R_1^2}{R_2^2 - R_1^2} (1 + \frac{R_2^2}{r^2}) + \frac{p_2 R_2^2}{R_2^2 - R_1^2} (1 + \frac{R_1^2}{r^2}) \quad \ldots(5.5.2)
\]

\[
\sigma_z(r) = p_3 \quad \ldots(5.5.3)
\]

All the resulting principal stresses are aligned with the cylindrical coordinates of the system.

By assuming the average condition induced in the powder to be equivalent to those induced in a similar elastic cylinder, these equations predict the compressive and tensile stresses. The stress is not constant throughout the sample. The stresses are highest on the inner wall. Yield starts on the inner portion of the sample, and with increasing pressure the yield zone
expands towards the outer wall.

\[ P_1 \geq P_2 \]

\[ \sigma_\theta(R1) = - \frac{p_1 (R_1^2 + R_2^2)}{R_2^2 - R_1^2} + 2 R_2^2 p_2 \leq 0 \quad \ldots \text{(5.5.4)} \]

\[ = - 2.125 p_1 + 3.125 p_2 \quad \ldots \text{(5.5.5)} \]

\[ \sigma_r(R1) = p_1 \geq 0 \quad \ldots \text{(5.5.6)} \]

According to the Mohr hypothesis the powder yields in a plane whose orientation is determined by the maximum and minimum principal stresses. From equations (5.5.4-6) it follows that if the vertical pressure is smaller than the inner pressure the vertical stress is the intermediate one and yield will occur in strictly vertical planes.

If the inner pressure is low and the weight of the column high a transition in the rank of the principal stresses may occur.

5.5.3 tracer experiments

In order to investigate a possible transition in yield mechanisms, a number of simple experiments were conducted using the Hollow Dutch Cell. In these experiments a ring of tracer material was introduced into the powder cylinder. The yield mechanism was inferred from the distortion of this ring after deforming the powder sample.

The powder cylinder was formed in three layers, each consolidated at 3 kPa and the whole cylinder was consolidated subsequently at 7 kPa, isostatically for 15 minutes. Between the second and third layer, approximately 3 cm below the top, a very thin ring of tracer material was introduced, 5 mm from the inner wall. The tracer material consisted of the sample limestone mixed with a small quantity of carbon black.

The piston was locked and the pressures were slowly increased until the required value was reached. The outer surface of the wall was inspected through the membrane to see whether any cracks were visible. After this inspection the pressure was slowly released, the piston removed and the deformation of the tracer ring was investigated by carefully excavating the powder by suction.
5.6 Discussion results

Table 5.6.1: Result of the visual observations of yield using a ring of tracer material.

<table>
<thead>
<tr>
<th>p1</th>
<th>p2</th>
<th>σr</th>
<th>σθ</th>
<th>Result</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>0</td>
<td>3</td>
<td>-6.375</td>
<td>radial cracks</td>
</tr>
<tr>
<td>6</td>
<td>3</td>
<td>6</td>
<td>-3.375</td>
<td>uniform deformation</td>
</tr>
</tbody>
</table>

Because, in this cell, there is no method of recording the strain rate of the inner surface these measurements are relatively crude. They already yield however a wealth of information and it is foreseen that when the cell is further instrumented, the hollow cell can contribute greatly to an investigation of the transition between the different yield mechanisms.

These experiments demonstrate the situation which occurs after applying a stress large enough to cause cracks on the outer wall.

Two situations which are comparable in the edifference of the principal stresses were selected. In both situations one principal stress was negative.

The deformation in the two cases was however completely different. In one case cracks occurred running from top to bottom of the sample, well distributed along the entire circumference. In the other case there were no cracks at all and inspection of the tracer ring found no slip zones either. This result was very surprising since clear slip planes were expected to form.

The deformation of the cylinder was, however, substantial, the displacement of the inner wall being about 0.5 cm. If a network of slip lines is present, it is extremely fine and uniform and cannot be detected by eye.

A situation of quasi-uniform deformation in a simple shear cell was investigated by Josseling de Jong[5]. He assumes that the macroscopic deformation of an element is actually a combination of two sets of parallel slip planes, which have an mutual inclination according with the Mohr hypotheses. The summation of independent displacements along both these slip planes yields any total macroscopic deformation. The resulting force is however not independent of the set of planes created during the consolidation.

If a set of fine planes is created in the sample during the hollow cell experiment, it would be interesting to investigate whether this model is appropriate here. This work has not been further pursued in this thesis.
If the deformation is truly uniform, it can be concluded that the formation of slip planes observed in simple shear machines is probably caused by the geometry of the system.

The cracks arising in the first case were radial, indicating that they were formed perpendicular to the tensile stress. See fig. 5.5. If Mohr's hypothesis is still valid for this situation the yield locus must coincide with a Mohr circle expressing the maximum possible negative stress. It follows that this would allow a multitude of directions for crack propagation for one stress condition. More probably the Mohr hypothesis is no longer valid and a new mechanism must be proposed.

Since the stress field drastically changes once a crack is initiated the interpretation of these results is of necessity qualitative. Until the problem of the redistribution of stress and of stress concentration due to cracks, has been clarified the problem must rest. The work on cracks occurring during tensile measurements, which is described in chapter 6, shows that calculations can be successful and applying these same techniques to the cylindrical case will most likely yield more quantitative results.

5.6 Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
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<tbody>
<tr>
<td>C</td>
<td>cohesion</td>
<td>[Pa]</td>
</tr>
<tr>
<td>p</td>
<td>pressure</td>
<td>[Pa]</td>
</tr>
<tr>
<td>R</td>
<td>Radius wall</td>
<td>[m]</td>
</tr>
<tr>
<td>r</td>
<td>radial distance</td>
<td>[m]</td>
</tr>
<tr>
<td>T</td>
<td>Tensile strength</td>
<td>[Pa]</td>
</tr>
<tr>
<td>t</td>
<td>time</td>
<td>[s]</td>
</tr>
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</table>

Greek

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>μ</td>
<td>coefficient of friction</td>
<td>[-]</td>
</tr>
<tr>
<td>θ</td>
<td>shear stress</td>
<td>[Pa]</td>
</tr>
<tr>
<td>σ</td>
<td>normal stress</td>
<td>[Pa]</td>
</tr>
<tr>
<td>σr</td>
<td>principal normal stress</td>
<td>[Pa]</td>
</tr>
<tr>
<td>σθ</td>
<td>principal tangential stress</td>
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Subscripts

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<tr>
<td>1</td>
<td>inner cylinder</td>
</tr>
<tr>
<td>2</td>
<td>outer cylinder</td>
</tr>
</tbody>
</table>

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5.7 References


Chapter 6. the tensile crack failure of powders

6.1 Introduction

In this chapter we investigate the behaviour of particle systems during the formation and extension of cracks.

The tearing of a powder cake in tensile testers was studied. Some relevant dimensions are defined in fig. 6.1

As indicated in this figure the displacement of both bottom halves is assumed to be equal to the width of the crack.

The chapter is organized as follows. First, the literature is examined, and it is concluded that the existing theories for tensile testers are not adequate. The crack theory for homogenous materials is introduced and experiments are done to compare this theory and the measurements for particle systems.

After noticing an important deviation from theory, a numerical simulation of the stress field in a particle system has been carried out. The results of this simulation show that the form of the stress field is dependent on the properties of the system simulated. A more general set of formulas derived from the formulas in crack theory can be used to describe a range of new systems. A particular solution has been found that does not contradict any of the observations.

Fig 6.1: Schematic drawing of the sample in a tensile tester.

- a crack length
- b displacement of plate = crack width
- F tensile force
- h cake height
6.2 Theory

In Chapter 4 it was argued that a macroscopic tensile force on an array of particles would always lead to 'premature' breaking of those contacts for which the critical breakage strength was surpassed. This causes the creation and the extending of holes in the original packing morphology. These extending holes could be called cracks. The expected force on the particle contacts surrounding a hole will be larger, promoting preferential extension of holes created.

In all existing theories for breakage strength of powder cakes it is assumed that instantaneous failure of all contacts occurs. Models to calculate the breakage force from the interparticle attraction forces, therefore, overestimate this force.

A number of breakage models, relating microscopic forces to the macroscopic ones shall be reviewed.

Rumpf's model [19-20]
Cheng's model [4]
Kanatani's model [13]

These models have been discussed in a number of publications. They are sometimes seasoned with empirical relations for some of the parameters. One of the most interesting models has been proposed by Schubert. This will be discussed in some detail.

Rumpf

Rumpf[19-20] applied the microscopic approach to the breakage phenomenon. He derived a simple relation between the attractive interparticle forces and a breakage strength.

In this derivation some assumptions had to be made. The packing was assumed to be random, the point contacts were distributed homogeneously, applied force on the contacts was strictly normal, and the particles were convex. Although the derivation is not rigorous, the form of the formula is correct (see Chapter 4).

\[ T = k \cdot (1-\varepsilon) \cdot \frac{H}{\pi \cdot D_p^2} \]  

\( T = (1-\varepsilon) \cdot \frac{H}{\varepsilon \cdot D_p^2} \)  

In addition Rumpf introduced an approximate formula for the relationship between the coordinate number and the density. This was chosen in that it simplified the formula even further. This simplicity is the main advantage of the formula.

\[ k = \frac{\pi}{\varepsilon} \]  

\[ T = \frac{(1-\varepsilon) \cdot H}{\varepsilon \cdot D_p^2} \]  

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Tensile strength: $T$
Interparticle forces of attraction: $H$
Porosity: $\epsilon$
Particle diameter: $D_p$
Coordination number: $k$

However, this estimate of the coordination number is questionable for cohesive powders. Also the non-linearity of the formula is a undesirable property. More convenient linear approximations can be found. See appendix 6.A.

Jimbo\textsuperscript{[12]} used Rumpf's equations as a framework for his empirical correlation between porosity and consolidating pressure. These correlations are based on a large volume of measurements performed on different types of tensile cells.

Cheng

Cheng\textsuperscript{[4]} also extended Rumpf's formulas and introduced the concept of the influence of the consolidating pressure on the strength of a contact. To this means he defined an average distance between the particles in contact. He then related this distance to the porosity of the packing. However, this relationship is based on an incorrect assumption. The porosity of a packing hardly changes due to the diminishing distance between the particles in contact. The main cause for a change is a rearrangement of the particles. Furthermore, even if one could calculate a reliable average distance between the particles, this parameter could not be used for calculation of an attractive vDWaals force since this force is not linear with respect to the distance. Qualitively the formulas describe the influence of the various parameters satisfactory.

The procedure to obtain some of the parameters in his model is highly questionable. The extrapolation of a curved line through limited data with a high variation over considerable distance is an exercise to be avoided.

Examining the data published by Cheng it has to be concluded that any parameter obtained in this manner will have a very low reliability.

A short summary of his numerical efforts:

$$T = \frac{a*b}{2} k (1-\epsilon) \frac{F}{(V_p/S_p)} \quad \text{(6.2.5)}$$
\[ F = f\left( t_0 - \frac{D_p}{3} \ast \left( \frac{\epsilon}{\epsilon_0} - 1 \right) \right) \quad \text{...(6.2.6)} \]

- \( a \) number of contacts in plain/
- \( b \) number of contacts per volume
- \( t_0 \) true area of contact/
- \( e_0 \) total area of particle
- \( \epsilon_0 \) average distance between particle when \( T=0 \)
- \( k \) porosity at \( T=0 \)
- \( \kappa \) coordination number

The Warren Spring laboratories have supported this theory with a number of papers by Valentin, Ashley, Cheng and Farley such as [1, 2 and 4].

**Kanatani**

Kanatani[13] relates a macroscopic deformation field to the virtual energy and calculates a force-stress relation. The deformation field is proposed as a tensor which makes the deformation in each point of the field uniform. This is not in concordance with his assumption that there is no deformation of the particles themselves, but all deformation occurs in the point contacts. It seems that due to this all his formulas apply to a homogenous field rather than to particles.

Kanatani further proposed that yield occurs when the average stress state in a volume surpassed a macroscopic yield criterium. Again this assumption does not explain the mechanism at the particle level.

After calculating and discarding all resulting higher terms in his formula, Kanatani compared this result to the results a continuum would yield and concluded that it compares favourably.

\[ \sigma_{ij} = n_j \left( 1 - \epsilon \right) \frac{F_{id}}{D_p^2} \quad \text{...(6.2.7)} \]

- \( \sigma_{ij} \) stress component
- \( F_{id} \) force per unit solid angle
- \( \epsilon \) porosity

**Schubert**

Schubert[22] also extended the theory of Rumpf and introduced the concept of a non homogeneous distribution of the tensile stress along the height of the sample. He assumed that in certain cases the stress intensity diminishes with the distance to the bottom of his tearing machine which is a ball bearing type.

This distribution of forces caused a lower breaking stress as the sample height was increased. This theory explains a number of his experimental results for wet and dry systems.
In this theory instantaneous breakage of the entire surface is still assumed.

Crack theory

In material science the theory of crack formation and stress distributions has been very successful in explaining the behaviour of many technical materials. The stress concentration formulas have been introduced for ceramics as well, but for a very porous cohesive powder agglomerate the validity of such formulas will have to be examined.

The general description of the stress field as it extends from the tip of a crack in a homogenous elastic body is [7-8]

\[ \sigma(r,\theta) = \frac{K_I}{\sqrt{2\pi r}} f(\theta) \quad \text{(6.5.16)} \]

\( r,\theta \) = polar coordinate system
\( r \) = distance from the tip of the crack.
\( K_I \) = stress intensity factor (N/m\(^{3/2}\))
\( f(\theta) \) = function only dependant of the angle relative to the normal of the coordinate system

The singularity on the tip itself can be avoided by redefining the origin of the coordinate system and choosing it a distance \( R \) inside the crack. Material at a distance \( R \) in the new coordinate system is just in yield condition.

\[ \sigma(\text{yield}) = \frac{K_I}{\sqrt{2\pi R}} \quad \text{(6.5.17)} \]

A compilation of many solutions for various geometries have been published by Tada [25]. The geometry resembling our system most is a crack loaded on the location of opening only. For this situation the following solution has been found for the stress intensity:

\[ K_I = \frac{2 F/d}{f(a/h)} \quad \text{(6.5.18)} \]

The influence of a limited height can be expressed as:

\[ f(a/h) \approx 3.52 (1-a/h)^{-3/2} -4.35 (1-a/h)^{-1/2} + 2.13(1 - a/h) \quad \text{(6.5.19)} \]

\( a \) = length of crack
\( h \) = height of sample
\( d \) = diameter of sample
Examining this solution for the stress intensity it is noted, that, for small cracks, the stress intensity decreases as the length of the crack increases. Any crack smaller than a critical one is therefore stabilized; i.e. if it progresses the driving force decreases.

For cracks larger than 0.3*h the situation is reversed and intensity increases as the crack extends: in this region the crack propagation velocity will increase without bounds. Assuming that the critical length of the crack is identical to the length which is just stable, the maximum force occurring can be calculated with:

\[ \frac{F/d}{2} = \frac{\sigma(yield) \pi}{(Ra) f(a/h)} \]  

for \( F_{max} \):

\[ \frac{d}{a} = 0 \Rightarrow a/h \approx 0.27, \ f(0.27)=1.5 \]  

The minimum is found at a fixed ratio of crack length and sample height.

\[ \frac{F_{max}/d}{3} = \frac{\pi}{3} \sigma(yield) \sqrt{a} \]  

It follows that the maximum force is proportional to the square root of the crack length and the yield value of the material in the tip. Since the critical length is directly proportional to the sample height the maximal force is also dependent on the root of the height.

Tada\[25\] shows that for an elliptical crack the formula's can be simplified. The relationship between width and length of an elliptical crack is given by \( R a = b^2 \). From this it follows that:

\[ F_{max} = c \cdot b_{max} \]  

\[ \sigma(yield) \approx 3/\pi \ c \]
Fig 6.2: The stress concentration above the crack causes breakage of weak contacts. These voids are represented with dots in the drawing. Coalescence of these holes may be a dominant mechanism in the crack extension.

Particle systems and crack theory

To apply the crack theory to particle systems a crack has to be first defined. This is somewhat more difficult than it may seem from first appearances.

A definition is sought in connecting all contacts that break under an applied load without intersecting any particles. This will form a three dimensional wire network touching the particles in the wall of the crack which can be interpreted as a wire network of a body filling the gap. The form of that body is by definition the form of the crack. The new contacts to break define the active tip of the crack and constitute a geometry which may be extremely complex and completely different from that normally visualized for a homogenous material.

This working definition does not account for the definition of initial cracks since only contacts broken during deformation are considered. Only after a few contacts near the tip are broken is the crack detected.
The concept of stress intensity in a homogenous system and in a particle system can be compared. Those particles closest to the contacts broken under a load will have to sustain a larger force than others, even after redistribution of the interparticle forces. Therefore, a stress concentration can be defined as the higher statistical forces on those particles close to the tip of the crack. This concentration increases the chances of particle contacts in the tip to break and, because of this, the existing cracks will preferentially grow. However the stress concentration will not be limited to the contacts in the tip only but extend into the bulk, gradually getting less pronounced. In that stress field individual weak contacts may break and caves may form just ahead of the main crack. The propagation mechanism may therefore also include coalescence of the small holes in front of the tip. See fig. 6.2.

The form of the crack walls as they are formed in tensile measurement equipment is irregular, as if the tip of the crack seeks its way along the easiest progression. No complete description has yet been found, but a random walk path seems to simulate the characteristics of the wall rather well. As the packing is more homogeneous appearance of the surface will be smoother. Nothing can be said from first principles about the relation between width and length of the crack. The form will most likely be influenced by the elastic behaviour of the mass and the plastic horizontal deformation during the load cycle.

6.3 Equipment

Two types of equipment are commonly used for the testing of the breakage strength of powders. Both can be used for our investigation of the crack formation, propagation and critical strength in powders. These types are made of two horizontal aligned plates which can be separated and, thereby, impose an opposite directed wall shear stress on the two halves of a powder system. If the plates are translated horizontally on bearings, we will call this a bearing type, if the plates are suspended with strings of blades, we will call it the suspended type. See fig. 6.3.

We built one ball bearing type with low initial friction and we adapted a commercial suspended type, the Hosokawa Cohetester, to compare the effects of different geometry and mechanism of separating the plates. To limit disturbing vibrations from the surrounding the equipment was placed on heavy stone tables, of which the leg were standing on rubber damping pads.
Fig 6.3: Schematic drawing of the two testers used.

a: ball bearing
b: fixed plate
c: moving plate
d: sample
e: force applied by gravitation
f: blade springs
g: cylindrical split sample holder of the Cohetester
6.3.1 The bearing type cell

The main problem with the ball bearing types is the initial static friction. This friction is influenced by the consolidating pressure on the particles. The ideal would be a frictionless non-vibrating system, as might be made from modern precision air bearing systems. However, for this investigation this was not available and the solution was sought with a ball bearing made of stainless steel precision balls. Six of these balls were placed under a flat plate, three balls each in two rows, without using any constriction to hold the balls in place. This minimized the friction.

To prevent the plate from moving in any other direction, the sides of the plate were confined with 3 balls on each side.

The balls were cleaned with alcohol and dust free tissues between measurements to avoid dust or particles influencing the friction. The plate itself was chosen to be as rigid as possible to avoid any problems with elastic deformation during the consolidation. The contact line, where the bottom plates meet, also was cleaned with great care before every measurement in order to prevent sticking due to compressed dust or particles between them.

preparation of the sample

A cylindrical ring, 63 mm wide and 10 mm high, was placed symmetrically on both bottom plates with the plates immobilized with a screw.

The limestone was deagglomerated with a course sieve and deposited in the ring in horizontal layers with a spoon. The top of the ring was equalized by carefully scraping off the excess powder. A smooth lid is pressed lightly on top and the consolidating weight is placed on a centered pin via a hanger. Samples for which the lid was depressed in a non-symmetrical way were rejected before measurement. After a consolidating time of 10 minutes the ring was rotated carefully to overcome the wallstick. The consolidating weight was taken off and, while holding the top lid with one hand, the ring was lifted with the other while slightly rotating. After removal of the ring the top lid was removed by shearing it horizontally to prevent any underpressure to build up and disturb the sample.

This procedure yielded beautiful free standing cylindrical samples.
6.3.2 The suspended cell

The other solution to eliminate friction problems is to suspend the plates. If the length of the strings or blades is long enough, the upward motion for displacements of $10 \mu$ is of the order of Angstroms and is negligible compared to the elastic deformation of the system. However, nothing comes for free and now care must be taken to separate the force to lift the plates and sample, to deform the springs, to tear the particle system itself. The Hosokawa Cohetester has this correction as a built-in option and is, therefore, very useful for this work. This apparatus is equipped with a cylindrical split sample holder.

material

One material was selected for measurements: the BCR-Limestone sample. The mean size of the particles is $5 \mu$, as determined with settling characterization in an Andrease pipet. See also chapter 2.

preparation of the sample

The plates are immobilized with a screw. Care has to be taken not to lock too tightly since the equipment deforms rather quickly and unlocking the plates will cause disturbing vibrations. The limestone is deagglomerated with a course sieve and deposited in horizontal layers in the cylindrical split holder 50 mm in diameter. After equalizing the surface a smooth piston is used to transfer the consolidating weight onto the sample. The weight was placed on top of the piston. After consolidating for 10 minutes the piston was slowly removed while rotating slightly. This prevented the built-up of an underpressure.

6.4 measurements

The measurements were performed in two ways:

- by varying the force on the bottom plate and measuring the horizontal displacement
- by varying the displacement and recording the induced force.

Only measurements of the first kind have been carried out using the ball bearing type. The Cohetester was more suitable for both kinds.
Fig 6.4: Effect of compaction on the appearance of the crack face.
above: high compaction and smooth face
below: low compaction and ragged face
The force was varied with an automatic titrator which controlled the amount of water in a cup connected to the bottom plate. The water was very carefully introduced in the cup in order to avoid any vibrations that may arise when a drop would fall directly on the surface. When increasing the force at a continuous rate the flow rate of water was about 1 gm/min.

The displacement in the suspended cell is controlled by a built-in motor which winds up a string on a shaft. The speed of the displacement was reduced to 0.5 μ/min. At this speed no dynamical effects could be detected. The horizontal displacement in the Cohetester was recorded with a capacitive displacement meter which made it possible to investigate the entire force/displacement curve.

The force transducer in the Hosokawa cell consists of a metal foil with strain-gauges. This transducer was placed between the shaft and the plate or between the cup and the plate.

6.4.1 Measurements with the bearing type cell

The force to overcome the static friction of the ball bearing and the slight tilt was adjusted to a mean of approximately 1.2 grams. The cleaning of the bearing balls and the touching plates ensured a small distribution of this value. The maximum and minimum value was ca. +/- 0.5 grams around the mean. All measurements were corrected with the mean friction. The distribution of the static friction contributes to the total distribution of the tensile strength of a sample.

observation of the faces after separation

The faces of the halves after separation are more regular - more flat - if the consolidating pressure is higher. See fig. 6.4 Usually, only a single line is noticed on the surface of the cake which indicates that the crack progresses from the bottom to the top in a single front. Sometimes the front progresses locally in an inclined plane and a piece breaks off. On the top surface it appears as if an island has been formed. This is a secondary process, and it will most likely not influence the total force recorded.

the distribution of the tensile force

To obtain an estimate of the distribution of the tensile force a large number of identical measurements were performed at a cake height of 9 mm and a density of 0.355. The force varied between 0.08 and 0.12 N, a total spread of 40%. Because the suspicion arose that, in spite of the heavy mounting of the apparatus,
external vibrations may have influenced these measurements, some experiments were repeated during the night. See fig. 6.5 It appeared that there was definitely some influence. The mean force is higher at night and the absolute total spread too. This can be explained if one assumes that the change on being disturbed increases as the time of measurement increases. Since the rate of increase of the force is constant, strong cakes take longer to break and they will therefore have a higher chance of being disturbed. As a consequence of this the distribution is narrower in case of disturbances. Both distributions can be well characterized with both the gamma and the normal distribution.

Table 6.1: Determination of the distribution of the tensile force of a limestone sample in a bearing type, 2 kPa, 9 mm height, $\varepsilon_s = 0.355$.

<table>
<thead>
<tr>
<th></th>
<th>Night</th>
<th>Day</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gamma</td>
<td></td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>71.6</td>
<td>114.7</td>
</tr>
<tr>
<td>B</td>
<td>6.5</td>
<td>12.2</td>
</tr>
<tr>
<td>$F_{\text{mean}}$ (mN)</td>
<td>110</td>
<td>94</td>
</tr>
<tr>
<td>$\sigma$ (mN)</td>
<td>15</td>
<td>7</td>
</tr>
<tr>
<td>Normal</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$F_{\text{mean}}$ (mN)</td>
<td>112</td>
<td>95</td>
</tr>
<tr>
<td>$\sigma$ (mN)</td>
<td>13</td>
<td>8</td>
</tr>
</tbody>
</table>

The influence of the height of the cake

A number of experiments have been carried out at night to determine the influence of the height of the cake on the mean tensile force. The density of the cakes has been held as constant as practically possible. The mean force is directly proportional to the height of the cake. Assuming that the force is proportional to the breadth of the sample, the force per projected area is independent of the height of the cake in the range of 5 to 9 mm. See fig. 6.6
There seems to be a tendency for lower cakes to be weaker: this could be caused by vibrations during the sample preparation. These will more easily disturb the lower cake than the higher ones. It is, therefore, assumed that the higher values are more representative of the actual process of tearing a powder cake. The mean force per area can be stated as:

\[ F/A = 0.18 \text{ kPa}, \quad \varepsilon_s = 0.355 \]

<table>
<thead>
<tr>
<th>height (mm)</th>
<th>N</th>
<th>( \varepsilon_s )</th>
<th>Fmean (mN)</th>
<th>( \sigma ) (mN)</th>
<th>Tmean (Pa)</th>
<th>( \sigma ) (Pa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.0</td>
<td>5</td>
<td>.356</td>
<td>26</td>
<td>11</td>
<td>138</td>
<td>57</td>
</tr>
<tr>
<td>4.5</td>
<td>5</td>
<td>.342</td>
<td>44</td>
<td>13</td>
<td>154</td>
<td>46</td>
</tr>
<tr>
<td>6.5</td>
<td>5</td>
<td>.364</td>
<td>87</td>
<td>24</td>
<td>213</td>
<td>58</td>
</tr>
<tr>
<td>7.5</td>
<td>7</td>
<td>.363</td>
<td>75</td>
<td>17</td>
<td>159</td>
<td>36</td>
</tr>
<tr>
<td>9.0</td>
<td>19</td>
<td>.355</td>
<td>112</td>
<td>13</td>
<td>198</td>
<td>25</td>
</tr>
</tbody>
</table>

6.4.2 measurements with the suspended cell

The suspended cell is easier to work with and yields more information since the entire force/displacement curve is readily obtained.

observation faces after separation

The geometry of the crack front is more complicated in the Cohetester since in this apparatus the sample holder may also impose forces on the sides of the sample. This may cause the crack front to progress, not only from the bottom up, but also from the sides in. When this happens it can clearly be seen on the top of the sample where two independent cracks run from the sides inward until one crosses the other or until secondary cracks combine the two. The stress distribution in this cell is therefore different than that in the bearing cell.
Fig 6.5: Cumulative distribution of tensile forces measured in day time and night time showing the effect of day time disturbances.

Fig 6.6: The measured tensile strength in both testers. dashed lines: distribution of forces in the ball bearing type solid lines: distribution of forces in the cohetester
displacement by force

The response of the system to an increasing or to a constant force has been determined. See fig. 6.7. The following general description of the phenomenon can be given.

As the force increases the displacement at the bottom increases in an increasingly disproportional way. A critical displacement exists after which the velocity of the crack propagation increases very quickly and the force/displacement curve can no longer accurately be followed.

On holding the force constant after a certain displacement, the widening continues, but as long as the critical displacement is not surpassed this creeping phenomenon damps out. The effect of external vibrations noted in the earlier experiment was also detected in these experiments. When a door was slammed somewhere in the building, the creeping velocity was momentarily increased. Also, when the displacement had come to a standstill it would recommence for a certain period. If the displacement already was close to the critical this sudden increase could start the unstable propagation of the crack.

When, after increasing the force on the system, it was suddenly relieved of the pressure, the bottom plate veered back. This is partly due to the elastic recovery of the particle packing and partly due to the gravitational force. On increasing the force again the original point where the force displacement curve was abandoned was completely reproduced. The removal and reintroduction of the stress can be modelled as a reversible process.

The entire force displacement curve is highly reproducible and, in addition, the following interesting observation can be made. When the curve is first determined while increasing the force at a fixed rate, a second curve is made for which the increase of force is stopped for a while at subcritical displacement. When the force increment is stopped the creep phenomenon causes the displacement to continue. On restarting the increase of force the curve would be inclined more strongly until the first curve was reached and the original curve resumed again.

force by displacement

The force displacement curve obtained when, not the force, but displacement is the governing mechanism is identical when the rate of displacement is low enough to exclude any dynamical effects.
Fig 6.7: Displacement due to controlled force. Sample height is 20 mm.

A steady increase of force
B creep phenomena
C unloading
D reloading
E creep rate decreasing to halt
F sudden disturbance
G unstable crack extension
From this it follows that the force displacement curve is a property; when the rate of increase of force is high enough the curve is uniquely defined.

In order to determine the extension of the crack above the bottom plate before the instability occurs the following experiments have been conducted. Thin metal plates of different height have been inserted at the bottom of the crack when forming the powder cakes. These plates were placed vertically when filling the ring and apparently did not change position after consolidation.

The force displacement curves for three different heights of plates have been recorded: 3, 6, 11 mm for a cake of 19.7 mm, and $\varepsilon=0.315$, consolidated with 2 kPa.

Up to a height of 6 mm the curves were identical. At 11 mm the curve was distinctively different. A similar effect can be seen when steel balls are introduced: balls of 6 and 10 mm hardly change the curves, although the 10 mm balls increased the critical displacement. Summarizing, we can now state that the critical crack has macroscopic dimensions, between 6 and 11 mm for a 19.7 mm cake and is more likely to be about 10 mm. The ratio of height of cake and length of the crack is then about 0.5. See fig. 6.8

Fig 6.8: Above: By inserting metal strips at the position of the crack before consolidation the critical crack length can be examined. Alternatively 3 equal sized steel balls were inserted. Below: actual force displacement curves for different strip heights and other inserts:
The influence of the height of the sample.

In this cell the effect of different cake heights on the critical tensile stress was investigated. The mean force is proportional to the height, but at the lower end of the cake heights, the force is strongly influenced by vibrations which inherently occur during the preparation procedure. This can be explained if one assumes that the treatment always introduces cracks of about the same length. If these cracks are much smaller than the critical length no influence is noted.

The mean force per projected area can be given by:

\[ \frac{F}{A} = 0.21 \text{ kPa} \quad \text{for } \varepsilon = 0.315 \]

...(6.4.1)
The absolute spread of the normal distribution is reasonably constant and about 40 Pa. Compared to the bearing cell these values are 16% higher, the spread is lower. If we would correct for the difference in density we could adopt the following correction factor (based on Rumpf):

\[ f = \frac{\varepsilon s_1 (1-\varepsilon s_2)}{\varepsilon s_2 (1-\varepsilon s_1)} = 1.2 \]

\[ \text{...(6.4.2)} \]

This would reduce the difference.
Fig 6.10: The tensile strength as a function of packing density. The solid lines indicate the 95% interval of the measured distributions.

Table 6.3: The influence of height on the force, 2 kPa, 10 min. normal distributions

<table>
<thead>
<tr>
<th>height (mm)</th>
<th>N</th>
<th>εs</th>
<th>F (mN)</th>
<th>σ (mN)</th>
<th>T (Pa)</th>
<th>σ (Pa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.5</td>
<td>19</td>
<td>.301</td>
<td>23.4</td>
<td>12.4</td>
<td>123</td>
<td>66</td>
</tr>
<tr>
<td>4.5</td>
<td>10</td>
<td>.301</td>
<td>33.8</td>
<td>10.2</td>
<td>150</td>
<td>46</td>
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<tr>
<td>5.5</td>
<td>10</td>
<td>.308</td>
<td>58.7</td>
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<td>6.5</td>
<td>8</td>
<td>.347</td>
<td>73.3</td>
<td>9.0</td>
<td>226</td>
<td>28</td>
</tr>
<tr>
<td>7.0</td>
<td>10</td>
<td>.322</td>
<td>71.8</td>
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<td>205</td>
<td>22</td>
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<td>9.0</td>
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<td>.345</td>
<td>85.6</td>
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<tr>
<td>12.0</td>
<td>10</td>
<td>.313</td>
<td>127.2</td>
<td>7.3</td>
<td>212</td>
<td>12</td>
</tr>
<tr>
<td>19.7</td>
<td>10</td>
<td>.315</td>
<td>209.4</td>
<td>3.3</td>
<td>213</td>
<td>3</td>
</tr>
</tbody>
</table>
critical width of the crack as a function of height of cake

The onset of instability can be determined accurately. The error of the order of 0.02 \( \mu \). The height of cake and the critical width correlate strongly; the quotient of width and height is constant:

\[
\frac{b}{h} = 4500 \quad \cdots (6.4.3)
\]

Assuming the ratio of critical width and length of the crack to also be constant it follows that length and height are directly proportional. The measurement with the metal stripes showed this ratio to be about 0.5. From this the ratio of width and length can be calculated to be 2250.
Furthermore since both force and critical width correlate with the height of the sample: (see fig. 6.11)

\[ F \text{ (mN)} = 48 * b \text{ (} \mu \text{)} \]  ...(6.4.4)

<table>
<thead>
<tr>
<th>Table 6.4: The influence of height on the critical width</th>
</tr>
</thead>
<tbody>
<tr>
<td>Height (mm)</td>
</tr>
<tr>
<td>-------------</td>
</tr>
<tr>
<td>3.5</td>
</tr>
<tr>
<td>4.5</td>
</tr>
<tr>
<td>5.5</td>
</tr>
<tr>
<td>6.5</td>
</tr>
<tr>
<td>7.0</td>
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<tr>
<td>9.0</td>
</tr>
<tr>
<td>12.0</td>
</tr>
<tr>
<td>19.7</td>
</tr>
</tbody>
</table>

Influence adsorbed water

Over a period of several days the measurements were repeated to estimate the influence of changes in relative humidity of the environment. The RH varied between 50 - 70%.

The absolute amount of physically adsorbed water is difficult to quantify since the limestone can contain different amounts of more strongly bound crystal water. During a determination with Karl-Fischer reagent for instance, this water is slowly liberated again and obscures the endpoint of the titration.

The results show no direct correlation between force and relative humidity between 50 and 70% RH. It can be concluded that the force changes but that the equilibrium state is attained so slowly, that quick variations in the environmental conditions do not show directly.

<table>
<thead>
<tr>
<th>Table 6.5: The influence of relative humidity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Day</td>
</tr>
<tr>
<td>------</td>
</tr>
<tr>
<td>1</td>
</tr>
<tr>
<td>12</td>
</tr>
<tr>
<td>25</td>
</tr>
<tr>
<td>26</td>
</tr>
<tr>
<td>27</td>
</tr>
</tbody>
</table>
Fig 6.12: Effect of consolidation on force-displacement curves
To eliminate this variation due to changes in the relative humidity the powder was heated for several hours at 120°C prior to the experiments. This material was then used for 2 hours before it was reconditioned.

The influence of density of packing

Equal weights of powder were compressed to different heights to obtain a variation in density. Looking at the force displacement curves it was found that the higher density shows a steeper and higher curve as expected. The highest compression, however, deviates from this expectation; its curve is almost identical to that of the second highest consolidation. See fig. 6.12.

The ratio between width and cake height is surprisingly identical and independent of the density. For the powder dried at 120°C this ratio is different then the powder conditioned in the laboratory atmosphere of a RH 50-70% indicating a more brittle behaviour.

\[ \frac{b}{h} \approx 6800 \]

Table 6.6: The influence of porosity on the stress/strain curve
10 min., dry powder, normal distributions

<table>
<thead>
<tr>
<th>height (mm)</th>
<th>N</th>
<th>( \varepsilon_s )</th>
<th>P (kPa)</th>
<th>b (( \mu ))</th>
<th>h/b</th>
<th>F (mN)</th>
<th>T (Pa)</th>
<th>( \sigma ) (Pa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>15.5*</td>
<td>10</td>
<td>.400</td>
<td>13.30</td>
<td>2.65</td>
<td>5850</td>
<td>305.0</td>
<td>394</td>
<td>27</td>
</tr>
<tr>
<td>16.5</td>
<td>10</td>
<td>.376</td>
<td>9.71</td>
<td>2.46</td>
<td>6700</td>
<td>307.0</td>
<td>372</td>
<td>26</td>
</tr>
<tr>
<td>17.5</td>
<td>10</td>
<td>.354</td>
<td>5.77</td>
<td>2.75</td>
<td>6360</td>
<td>272.2</td>
<td>311</td>
<td>21</td>
</tr>
<tr>
<td>18.0</td>
<td>10</td>
<td>.345</td>
<td>4.28</td>
<td>2.34</td>
<td>7369</td>
<td>211.3</td>
<td>235</td>
<td>13</td>
</tr>
<tr>
<td>19.7</td>
<td>10</td>
<td>.315</td>
<td>1.97</td>
<td>3.03</td>
<td>6500</td>
<td>186.2</td>
<td>189</td>
<td>11</td>
</tr>
</tbody>
</table>

* excluding inclined slip planes

The deviation of the response of the most compressed samples from the general trend may be identified as an artifact of the test system. These samples often yielded along one of two smooth inclined planes. This is in contrast to the samples at other conditions which yielded in irregular vertical planes. The formation of these planes can be explained as follows. Due to the high compressive forces the split holder will be forced apart. This can be detected with the displacement meter. On relieving the pressure it veers back and doing so compresses
the sample in the horizontal plane. If this stress and the
deformation is high enough one or two slip zones inclined to the
horizontal will be formed. When the sample is subsequently loaded
yield along these slip zones apparently occurs at a lower stress
than the formation of a fresh crack plane. In a few samples the
fracture face showed partial slip along an inclined plane. From
this it seems that during yield a small crack is formed, due to a
local disturbance, which extends to the outside of the slip zone.
Once formed the stress concentration on the tip is such that the
crack does not bend back into the slip zone but it develops
vertically until it reaches the surface.

Even when no slip occurs the force for crack formation is limited
by the force to slide along the plane. The measurements at this
high compressive stress (13.3 kPA) can not be compared to those
at lower stresses.

6.5 Discussion and comparison with literature

The force displacement curves and the tests with metal stripes
clearly show that the mechanism in splitting the powder cake in a
the common tensile testers is one of crack formation and
propagation. Theories which start with the premise of
instantaneous failure of all contacts across the failure plain
cannot be used for these instruments.

6.5.1 empirical models

As was said in the theoretical discussion literature can be split
into two main groups: those who describe the system
phenomenologically and those who follow Rumpf's microscopic
approach.

In the phenomenological description three variables are commonly
distinguished: consolidating pressure, porosity and force per
projected area. The most elegant and consistent approach would be
to fit a function of all three variables at once. This is not
done, always two variables are selected and fitted separately to
a model for these two.

Fitting two combinations allows calculation of the third
combination. The residual sum of squares for this combination may
not be the minimum.

An enormous variety of formula's has been published, and no
superior can be selected otherwise than taking the one reducing
further mathematical efforts. A number of functions appeared
adequate for our data.
Empirical correlations
consolidating pressure versus porosity

A number of formulas have been summarized with:

\[ \frac{d \varepsilon}{d P} = -C \left( \frac{x}{y} \right)^z \epsilon \] \hspace{2cm} ...(6.5.1)

After integration this yields a 4 parameter model which allows enormous freedom. Most times a number of parameters are chosen and the other set to zero.

Table 6.7: A number of empirical formula's deducted from 6.5.1

<table>
<thead>
<tr>
<th>Condition</th>
<th>Formula</th>
<th>( y=0, x=0, z=0 )</th>
<th>( y=0, x=0, z=1 )</th>
<th>( y=0, x=0, z&gt;1 )</th>
<th>( y=0, x=1, z=1 )</th>
<th>( y=0, x&gt;1, z&gt;1 )</th>
<th>( y=1, x=1, z=1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( \varepsilon = -C_1 P + C_2 )</td>
<td>( \varepsilon = -C_1 \ln(P) + C_2 )</td>
<td>( \varepsilon = C_1 P + C_3 )</td>
<td>( \ln \varepsilon = C_1 \ln(P) + C_2 )</td>
<td>( \ln \varepsilon = C_1 \ln(C_2 P + 1) + C_6 )</td>
<td>( \ln \varepsilon - \varepsilon = C_1 \ln(P) + C_2 )</td>
<td></td>
</tr>
</tbody>
</table>

Porosity versus Tensile force per Area

Two functions were selected:

\[ T = C_1 \left\{ -\left(1-\varepsilon\right)/\varepsilon \right\} + C_2 \hspace{2cm} n=0 \text{ or } 1 \] \hspace{2cm} ...(4.5.8)

\[ T = C_1 \exp\left(-C_2 \varepsilon\right) \] \hspace{2cm} ...(4.5.9)

and were adequate. The formula with the smallest sum of residues is:

\[ T = 0.476470E6 \exp\left(-11.47 \varepsilon\right) \hspace{2cm} \text{(Pa)} \] \hspace{2cm} ...(4.5.10)
Consolidating pressure versus Tensile force per area

For the relationship between tensile stress and consolidating stress the following formula was tried:

\[ K_2 T = K_1 P \]  \hspace{1cm} \text{...(6.5.11)}

after fitting the best formula is:

\[ T = 0.1363 \, P \quad \text{(kPa)} \]  \hspace{1cm} \text{...(6.5.12)}

Recapitulating the data can adequately be described with:

\[ \ln T = 0.438 \, \ln P + \ln 0.1363 \]  \hspace{1cm} \text{...(6.5.13)}

\[ \ln T = -11.47 \, \varepsilon + \ln (0.476 \, \text{E}6) \]  \hspace{1cm} \text{...(6.5.14)}

and from this also the relation between porosity and consolidating pressure

\[ \varepsilon = -0.438/11.47 \, \ln (P/P_0) - (\ln (136.3/0.476 \, \text{E}6))/11.47 \]  \hspace{1cm} \text{...(6.5.15)}

6.5.2 prediction of Rumpf's microscopic model

The basis for a microscopic model is an estimate of the interparticle forces involved. Since no information about the rolling or sliding friction between particles is available, only the separation force for two particles can be taken into account. For crystalline materials the distribution of forces may be rather wide due to the large variation in contact geometry and intermolecular or interionic forces. If we assume that the separation forces for limestone particles compare to those of other crystalline material of similar size we can use the data obtained by Polke and Herrmann for 10 an 20 μ Barium Sulfate and published by Zaradnicec. See Ch. 3 also. These show direct proportionality of F50 and particle size. Extrapolating that data to 5 μ particles we obtain:

\[ D_p = 5 \, \mu \implies F_{\text{mean}} \approx F_{50} \approx 10 \, \text{mdyne} \]

\[ \varepsilon s = 0.315 \]

Substituting this in Rumpf's model (6.2.3):

\[ \sigma = \frac{\varepsilon s \, F_{\text{mean}}}{1-\varepsilon s \, D_p^2} \]

yields:

\[ \sigma \approx 2 \, \text{kPa} \]

This is a tensile strength which is about 10 times too high.
6.5.3. The crack model

Comparing the predictions of the crack model with the observations obtained for the limestone cakes the following conclusions can be made.

In Agreement:

1- A stabilized crack can exist; creep will diminish as the length of the crack is extended under constant load.

2- A critical displacement can be defined in a particle system as well as long as the load rate is within a certain range.

3- The maximum force is proportional to the critical width of the crack. This is only valid for elliptical cracks.

4- The critical width is independent of the density of packing and directly proportional to the height of the sample.

Against:

1- Different from the prediction of a square root dependency the force is directly proportional to the height of the sample.

This leads to the conclusion that the stress field in a particle system may not be represented by the formulas presented.

On the other hand Schubert has shown a series of measurements on different powder samples in which the tensile strength declines with increasing height. This would agree with the crack theory. However, if his system is controlled by a crack propagation mechanism with a square root dependency, the procedure of extrapolating data obtained at the lowest two cake heights to the axis to obtain the 'true' tensile strength is not valid.

Assuming our measurements for the limestone samples are carefully carried out it follows that the crack theory must be adapted to cohesieve particle systems.
Fig 6.13: The simulation model: nodes are displaced horizontally, the tensile and shear forces modelled with elastic springs. The total force on a node is found by summing the contributions of the 4 connections.
6.6 Simulation of stress field around cracks in particle systems

A simulation has been set up to investigate the stress field around the central crack in a particle system in a tensile tester test. The powder is represented by a grid, its axis aligned with the bottom plates. Half the bottom points are fixed rigid to the left plate and half to the right plate to simulate the no-slip condition. The forces between the grid points arise due to the stretching of the material between horizontal neighbouring points and due to the sliding displacement of material between the neighbouring vertical points. These forces represent the stress condition in the powder.

The mean difference between modelling a homogeneous system and a particle system is the demand of connectivity of the material. In the grid model this is expressed by the fact that when vertical points slide relative to each other only forces between direct neighbours are accounted and changes in distance between other points have no influence on the stress field. Furthermore it is assumed that the shear deformation does not give rise to normal forces or influence the horizontal forces. This restricts the displacement of the knots to the horizontal plane. See fig. 6.13.

The relation between the force on two points 1 and 2 and the stretching of the material between them is assumed to be linear:

\[ F_{12} = \frac{E_{12} \cdot d_{112}}{112} \]  

\( 112 \) = unloaded distance between knot 1 and 2  
\( d_{112} \) = stretch due to load  
\( E_{12} \) = Elastic modulus for material between 1 and 2  
\( F_{12} \) = tensile force on material between 1 and 2

To simulate a crack or a hole in the grid for some horizontal connections, the elasticity constant is set to zero effectively cutting the connection.

The shear force between vertical points is also assumed to be linear:

\[ F_{13} = \frac{G_{13} \cdot d_{x13}}{113} \]  

\( F_{13} \) = shear force on material between 1 and 3  
\( G_{13} \) = shear modulus between 1 and 3  
\( d_{x13} \) = horizontal displacement of 1 relative to 3

173
Fig 6.14: The simulation of the displacement of each node in the lattice. The displacement is represented as the distance between the node and the drawn lines.

This model is flexible enough to investigate the effect of distributions of elastic and shear moduli in a powder. This is not done in this work and all moduli have been set to 1.

The experiments have been conducted in a 30X30 grid. The crack is situated between row 15 and 16. The differences using a larger grid proved to be negligible. The boundary condition is a unit displacement for one half of the bottom points to the left, the other to the right.

The total force on any point can be found by summing all forces of the material around it. The equilibrium can then be found using a relaxation technique. This results in a set of matrices which contain the horizontal displacements of all knots, the tensile forces between them, and the shear forces between layers.
Results of crack simulation

A series of simulations have been carried out to simulate a stress field around a progressing crack at constant crack width. After finding the equilibrium state for a certain crack, this state was used for the starting condition for a crack one layer higher.

To reduce the amount of data only the forces on the material straight above the crack have been used for conclusions. Since the entire system is linear the total force is directly proportional to the width of the crack. This width has conveniently been set to 1 for all simulations. See fig. 6.14.

\[ F_{\text{tot}} = C \times b \]  \hspace{1cm} \ldots(6.6.3)
\[ C = \frac{F_{\text{tot}}}{1} \]  \hspace{1cm} \ldots(6.6.4)

In figs. and some representative results are shown. Forces and displacements are arbitrary.

In figs. 6.15-6.16 the tensile force on the first point above the crack has been plotted as a function of height. Also a number of plots of the distribution of the force along the height of unbroken material show the dependency of distance to the crack tip.

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|}
\hline
length in layers & F_{\text{tip}} & F_{\text{tot}} & \frac{F_{\text{tip}}}{F_t} \\
\hline
1 & 724 & 2155 & 0.336 \\
2 & 508 & 1671 & 0.304 \\
3 & 409 & 1407 & 0.291 \\
4 & 346 & 1224 & 0.283 \\
5 & 303 & 1094 & 0.277 \\
6 & 269 & 983 & 0.274 \\
7 & 243 & 895 & 0.271 \\
8 & 221 & 823 & 0.269 \\
\hline
\end{tabular}
\end{table}

\textbf{table 6.8:} 30x30 grid arbitrary dimensions
Fig 6.15: Simulation of the tensile forces in a powder cake using a 30x30 elastic lattice.

A: contour map of the tensile forces in the sample (arbitrary levels are displayed)

B: the tensile forces as a function of the position in the sample. The magnitude is represented as the distance between the nodes and the drawn lines.
The force at a distance \( z \) from the tip of the crack can be well represented by:

\[
F(z) = \frac{k_1}{(k_2 + z)} \quad \ldots (6.6.5)
\]

where \( z = \text{number of knots above the crack} - 1 \)

\[
F(z) = \frac{0.124}{0.973 + z} \quad \ldots (6.6.6)
\]

If we shift the center of the coordination system a distance \( k_2 \) within the crack the form may be simplified to:

\[
F(r) = \frac{k_1}{r} \quad \ldots (6.6.7)
\]

The square root dependency, which is known to be valid for homogeneous materials, performed significantly worse:

\[
F(r) = a + c \quad \ldots (6.6.8)
\]

relation of total force/force on the tip

When we write the relationship between the stress intensity and total force as:

\[
K_I = \frac{F_{\text{tot}}}{\sqrt{\pi a}} f(x/H) \quad \ldots (6.6.9)
\]

and \( K_I = \frac{F_{\text{tip}}}{\sqrt{\pi R}} \quad \ldots (6.6.10) \)

than substitution yields:

\[
\frac{F_{\text{tip}}}{F_{\text{tot}}} = \frac{1}{\sqrt{x/R}} f(x/H) \quad \ldots (6.6.11)
\]

Two functions for \( f(x/H) \) have been compared for their correlation with the data:

1) \( f() = c_1 (1-x/H)^{-1/2} + c_2 (1-x/H)^{-3/2} + c_3 (1-x/H)^{-3/2} \quad \ldots (6.6.12) \)
Fig 6.16: Simulation of the tensile forces in a sample. The curves associated with the left axis represent the stress distribution as a function of height above the crack tip, for a fixed crack length. The curve associated with the right hand axis represents the stress in the tip itself as a function of the crack length.

\[
\text{F}(x) = \begin{cases} 
  0 & \text{if } x < 0 \\
  \text{something} & \text{if } x > 0 
\end{cases}
\]

2) \(f(x) = c_1 (1-x/H) + c_2 (1-x/H) + c_3 (1-x/H)^2\) \hfill \ldots(6.6.13)

The first one is suggested by the formula valid for this geometry and homogeneous systems (see 6.5.19), the second is a general polynomial function.

Neither formula performed well. The first formula is worst. However, if we replace the square root dependency with a proportional dependency the general polynomial shows a residual error which is a factor of 100 better.

\[
\frac{F_{\text{tip}}}{F_{\text{tot}}} = \frac{1}{x/R} \left\{ c_1 (1-x/H) + c_2 (1-x/H) + c_3 (1-x/H)^2 \right\} \hfill \ldots(6.6.14)
\]
Table 6.9: parameters of the various formulas after fitting with a least squares criterium.

<table>
<thead>
<tr>
<th></th>
<th>(11)+(12)</th>
<th>(11)+(13)</th>
<th>(14)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(c_1/R)</td>
<td>1 e-18</td>
<td>3.2</td>
<td>2.76</td>
</tr>
<tr>
<td>(c_2/R)</td>
<td>-0.749</td>
<td>4e-10</td>
<td>1.91</td>
</tr>
<tr>
<td>(c_3/R)</td>
<td>+1.07</td>
<td>-2.95</td>
<td>-4.61</td>
</tr>
<tr>
<td>(\chi^2)</td>
<td>5 e-4</td>
<td>2 e-5</td>
<td>4 e-7</td>
</tr>
</tbody>
</table>

Clearly formula 6.6.14 is superior. The minimum in the formula is predicted at \(x/H \approx 0.4\).

The force/displacement curve

\[
F_{tot} = \frac{F_{tip} \frac{x}{R}}{f(x/H)} \quad ...(6.6.15)
\]

Assuming the mean force on the tip to be a constant throughout the total experiment the following relation can be derived:

\[
F_{tot} = \frac{c_1 \left( \frac{x}{H} - \frac{x^2}{H} \right)}{1 + c_2 \left(1 - \frac{x}{H}\right) + c_3 \left(1 - \frac{x}{H}\right)^3} \quad ...(6.6.16)
\]

Since \(x/H\) is a constant for the maximum force it also follows:

\[
F_{max}/d = C \times H \quad ...(6.6.17)
\]

and

\[
F_{tip}/d = \frac{f(x/H)}{x/H} R C = k C \quad ...(6.6.18)
\]

\(C\) has the dimensions of a stress and represents the macroscopic description of the interparticle forces present in the tip.
relation of length/width

The relation between the width of the crack and the total force for a given crack length can be represented by:

\[ F_{\text{tot}} = E(x) \times b \]  \hspace{1cm} \ldots(6.6.19)

\[ E(x) = \frac{a_c}{1 + b_c x + c_c x^2} \]  \hspace{1cm} \ldots(6.6.20)

\[ a_c = 0.30 \]
\[ b_c = 0.408 \]
\[ c_c = -0.0103 \]
\[ \chi^2 = 2 \times 10^{-6} \]

This relates width and length of the crack in the following rather cumbersome way:

\[ b = \frac{c \left( x - \frac{x^2}{H} \right)}{c_3 + c_1 (1-x/H) + c_2 (1-x/H)^3} \frac{1 + b_c x + c_c x^2}{a_c} \]  \hspace{1cm} \ldots(6.6.21)

Fortunately for not too large cracks the width is practically proportional to the length. For larger values the length increases faster than the width. Introducing the following approximation:

\[ b \approx k_1 \times (1-x/H) \]  \hspace{1cm} \ldots(6.6.22)

for the critical values it can be seen that width, length and height of the sample are all proportional.

\[ b_{\text{critic}} \propto a_{\text{critic}} \propto H \]  \hspace{1cm} \ldots(6.6.23)

It was shown that the maximal force as well as the critical width was proportional with the cake height. The simulation is in complete agreement with these observations. It must be noted though that since no information about the real elastic and shear moduli have been included the model can only been seen as a useful indicator of possible behaviour.

Comparing the force/displacement model and observations.

Since too little information is yet available concerning the relationship between width and length in a real powder cake, these are assumed to behave according to formula \[6.6.21\] Leaving out higher order terms that formula can be reduced to a three parameter model:
\[ \text{Ftot} = \frac{a' X}{1 + b' X + c' X^2} \ldots (6.6.24) \]

A number of characteristic curves at different densities have been selected and fitted with the above function. The curves correlated strongly with the proposed formula. Since the variance is very small it is strongly influenced by the error in the exact position of the starting point of the curve. Small displacements of the starting point (up to + and - 0.04 \( \mu \)) have been introduced to minimize the residual error. The standard deviation of the model to the actual line was in all cases better than 1.5 % of the maximum force.

table 6.10: The influence of porosity on the stress/strain curve
10 min., dry powder, normal distributions

| p (kPa) | height (mm) | \( \epsilon s \) | \( a' \) (mN/\( \mu^2 \)) | \( b' \) (1/\( \mu \)) | \( c' \) (1/\( \mu^2 \)) | var | sd (%)
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>9.71</td>
<td>16.5</td>
<td>.376</td>
<td>419.3</td>
<td>0.78</td>
<td>0.0509</td>
<td>0.03</td>
<td>0.3</td>
</tr>
<tr>
<td>5.77</td>
<td>17.5</td>
<td>.354</td>
<td>330.6</td>
<td>0.68</td>
<td>0.0485</td>
<td>0.04</td>
<td>0.8</td>
</tr>
<tr>
<td>4.28</td>
<td>18.0</td>
<td>.345</td>
<td>258.0</td>
<td>0.68</td>
<td>0.0487</td>
<td>0.009</td>
<td>0.5</td>
</tr>
<tr>
<td>1.92</td>
<td>19.7</td>
<td>.315</td>
<td>218.5</td>
<td>1.10</td>
<td>0.217</td>
<td>0.03</td>
<td>1.5</td>
</tr>
</tbody>
</table>
6.7 Conclusions

The following conclusions can be distilled from the experimental work:

A particle system yields preferentially on the tip of a crack under the influence of a stress concentration field. The measurements with metal stripes show that the crack progresses far into the cake before the unstable crack growth occurs. The length of the critical crack can be estimated to be between 0.3 and 0.6 of the height of the sample.

For one density the maximum force is proportional to the height of the cake. This proved to be valid for both the bearing type without sample holder and the suspended type with holder.

The critical width of the crack is independent of density and linearly dependent on the height.

conclusions related to the models

The powder behaviour in a tensile tester can not be described with instantaneous breakage of all particle contacts across the failure plane. Most existing models can, therefore, not be applied.

The crack theory proved to be a valid concept for the tearing of powders and explained a number of phenomenon. The prediction of the relation between the force and height of the sample was false indicating that the stress field was not described accurately.

A computer simulation introducing properties specific for particle systems showed that the stress field does change, and that if linear relation between deformations and forces exist, a more useful set of formulas can be derived. The excellent predictions of the model give faith in future exercises in which the stress field for other geometries will be calculated. Knowledge of the stress field in the hollow Dutch cell (CH.5) is dearly needed to clarify the relationships between forces and the crack formation.

The maximum force can be related to the forces in the tip of the crack when a reliable relation between length and width and the stress field has experimentally been found. Early estimates can show the order of magnitude of the relevant constants.
6.8 Symbols

Some symbols are local and have been explained near the formula that uses them. They are not repeated in this list.

A  projected area of the failure face  [m²]

a  length of the crack  [m]

b  width of the crack  [m]

a'  parameter force displacement  [N/m]

b'  parameter force displacement  [1/m]

c'  parameter force displacement  [1/m²]

C  constant  [Pa]

c  constant  [Pa]

Cb  length ratio  [-]

d  diameter of the sample  [m]

dlij  displacement of i with respect to j  [m]

Eij  coefficient of elasticity  [N/m]

F  force  [N]

Gij  shear modulus  [N/m]

H  height of the sample  [m]

KI  stress intensity factor  [Pa/m]

Ki'  stress intensity  [Pa]

lij  distance between knot i and j  [m]

m,n  parameters stress intensity model  [-]

P  compressive stress  [Pa]

q1  constant  [Pa]

q2  constant  [Pa]

q3  length ratio  [-]

R  radius crack tip  [m]

RH  relative humidity  [%]

X  displacement of the tensile tester  [m]

z  distance from tip  [m]

Greek symbols

εs  density  [-]

r,θ  polar coordinates  [m]

σ  normal stress  [Pa]
6.9 References


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Fig 6.17: The coordination number as a function of the density

Appendix 6.A. The coordination number

In cohesive systems an isotropic packing of the particles on microscopic scale is unlikely. The system can better be visualized as a structure with strong fluctuations of the number of particles per volume. In the more extreme cases the packing is made of dense agglomerates and large holes between them. If one wants to estimate the average number of contacts in a volume using the overall density in that volume, the estimate is only independent of the distribution of the density within the volume if the formula is linear. The commonly used estimate of Rumpf (1958) for the coordination number as a function of density does not possess this property.

A formula that would be linear with respect to the overall density is:

\[ k = \alpha \varepsilon_S + \beta \]  

...(6.A.1)
Since each particle must touch at least one other particle the lower limit for the least dense packing of a large set of particles is 2.

After reviewing a number of data from literature Vervoorn[28] finds:

\[ k = 7.7 \epsilon s + 2 \]  

...(6.A.2)

If one limits the range of overall density to \(0.3 < \epsilon s < 0.6\) an approximation which is even more simple is: see fig. 6.17.

\[ k = 12 \epsilon s \]  

...(6.A.3)


<table>
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<th>(\epsilon s)</th>
<th>(\frac{\pi}{(1-\epsilon s)})</th>
<th>7.7(\epsilon s + 2)</th>
<th>12 (\epsilon s)</th>
<th>(\frac{\pi \epsilon s}{(1-\epsilon s)})</th>
<th>7.7(\epsilon s^2 + 2\epsilon s)</th>
<th>12 (\epsilon s^2)</th>
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<td>7.20</td>
<td>4.71</td>
<td>3.97</td>
<td>4.32</td>
</tr>
</tbody>
</table>

Conclusions.

It can be seen from table 6.A.1 that the numerical values for the different formula's do not deviate more than 20 % in the range of the density \(0.3 < \epsilon s < 0.6\). The Rumpf formula always gives the higher estimate.Introduction of a linear relation solves the problem of consistency of the formulas, which is for modelling more desirable.
appendix 6.B. crack theory formulas for particle systems

The simplest adjustment to the existing formulas describing the relation between stress intensity and distance to the tip is introducing a kind of power law. To omit problems the length has been made dimensionless by dividing with a characteristic length \( R \). This does mean a complete redefinition of the constants that were used before. This is a disadvantage. On the other hand the new definition is physically more appealing:

\[
\sigma(r) = \frac{K_i^*}{(r/R)^n} f(\theta) \quad \text{ ...(6.B.1)}
\]

\[
\sigma(\text{tip}) = K_i^* \quad \text{ ...(6.B.2)}
\]

\[
K_i^* = \left| \frac{a}{R} \right|^m \frac{F_{\text{tot}}}{d R} f(a/h) \quad \text{ ...(6.B.3)}
\]

For homogeneous materials the stress field diminishes with the square root of the distance. This leads to the conclusion that the total strength of a cake is proportional to the square root of the height of the cake. The data show a direct proportionality with height, and from this, it is concluded that the stress field in particle systems must be different.

If we assume \( n \) to be equal to 1 and \( m \) equal to minus 1 all observations done with the limestone can be explained. This dependency has been verified in a mathematical model for crack formation in powders.

Other systems which contain liquid bridges are expected to behave differently since the connection between particles is more flexible before breaking. It is anticipated that each system must be characterized with different powers for the stress field around a crack.
estimation of the critical factors

Setting $m=-1$, $n=1$, the resulting formulas agree with the measurements:

$$F_{\text{max}}/d = q_1 \cdot a_{\text{max}} \quad \ldots(6.B.4)$$

Further it follows from the assumption of constancy of the ratio of length and width that:

$$F_{\text{max}}/d = q_2 \cdot b_{\text{max}} \quad \ldots(6.B.5)$$

Consistent with this model the observations show:

$$F_{\text{max}}/d/H = C \quad \ldots(6.B.6)$$
$$b_{\text{max}}/H = q_3 \quad \ldots(6.B.7)$$

According to (6.B.2 & 3) the yield stress on the tip is equal to the stress intensity:

$$\sigma(\text{yield}) = \sigma(\text{tip}) = K_I' \quad \ldots(6.B.8)$$

$$K_I' = \frac{F_{\text{max}} \cdot f(a_{\text{max}}/H)}{d \cdot a_{\text{max}}} = \frac{c_b \cdot F_{\text{max}} \cdot f(a_{\text{max}}/H)}{d \cdot b_{\text{max}}} \quad \ldots(6.B.9)$$

substituting the following experimental data

$$F_{\text{max}}/b_{\text{max}} = 55.46 \text{ kN/m}$$
$$d = 0.05 \text{ m}$$
$$c_b = 1/2200 \quad \ldots(6.B.10)$$

one obtains:

$$K_I'/f(a/H) = 55.46/0.05/2200 = 0.5 \quad \ldots(6.B.11)$$
$$\sigma(\text{yield}) \approx 0.5 \text{ kPa} \cdot f(a_{\text{max}}/H) \quad \ldots(6.B.12)$$

No values for $f(a_{\text{max}}/H)$ are available yet, but an estimate can be made if one assumes that $\sigma(\text{yield})$ should compare to the theoretical instantaneous breakage strength of the particle packing. The defending argument must be that the particles along the crack front break almost simultaneously.
Using the same estimates for the interparticle forces as used before:

\[ \epsilon s = 0.315 \]
\[ k = 12 \epsilon s \approx 4 \]
\[ Dp = 5 \mu \]
\[ F = 10 \text{ mdyne} \]

The corrected estimation for the breakage strength is:

\[ \sigma_{\text{break}} = \frac{(k/3) \epsilon s F}{Dp^2} \quad \ldots(6.B.13) \]

\[ \sigma_{\text{break}} \approx 1.5 \text{ kPa} \quad \ldots(6.B.14) \]

From this it follows that \( f(\text{amax}/H) \approx 3 \) for the limestone packing. The estimates made to obtain this value must be treated with caution. It is more safe to state that \( f(\text{amax}/H) \) is of the order of magnitude of 3 and probably ranges between 1 to 10.

To summarize the calculations: the force per projected area in a tensile tester can be calculated with:

\[ \frac{F_{\text{max}}}{d/H} = \sigma_{\text{break}} \frac{a/H}{f(\text{amax}/H)} \approx 1.5 \times \frac{0.5}{3} = 0.25 \text{ kPa} \quad \ldots(6.B.15) \]
Chapter 7. Shear zones and cohesion in compacted particle systems.

7.1 Introduction and definitions

Since cohesion is defined to be a point on the yield locus, its concept is directly related to the definition of a yield locus. In this chapter the yield locus and the formation of a stationary shear zone in compressed particle systems are investigated, both experimentally and theoretically. To this end a model has been constructed which includes the effect of the change of density leading to the critical state and the fraction of contacts which are in a state of plastic yield. The simulated yield locus is compared to that obtained in shear testers.

An algorithm will be presented which enables more objective measurements when using the standard Jenike Cell. Using this routine, measurements have been made in order to investigate the reproducibility of the yield points. The concept of a stationary state in a shear zone has also been tested in a new type of coaxial shear tester. This instrument was specifically designed to create a well defined shear zone with unlimited strain.

7.2 Force/displacement characteristics of shear zones

As discussed in CH.5, the yield locus can be used to define a macroscopic cohesion in the shear zone. The cohesive strength is the yield point for zero compressive stress.

The major problem with this definition is that this yield point is not measured directly in existing commercial shear testers and always must be obtained through extrapolation of the higher yield points to the axis. Since no single true model for extrapolation is available one tries to find the best empirical line to find an reasonable estimate. Although a number of empirical functions describing the macroscopic force have been suggested, a satisfying relation between the macroscopic cohesion and the microscopic forces does not exist. In this thesis all mechanisms proposed in literature, which occur during shear deformation, are combined in a numerical model. Comparing the outcome it can be discussed which of these mechanisms are the dominant or whether more mechanisms need to be introduced.
7.2.1 yield loci

Two types of force/displacement curves can be distinguished, which are dependent on the starting condition of the sample.

1) a monotonically increasing curve, which becomes stationary after a certain length of displacement,

2) a curve which goes through a maximum and then becomes stationary.

One can only conclude that the critical state is actually reached if one has a model to show either when the transition occurs or when one has a signal long enough to show that stationarity is achieved. Stationary is here used in the sense that all characteristics of the fluctuating signal are constant.

Since translational cells seldom provide a signal long enough in duration for the latter a model must be used to test for the critical state. In theory, rotating shear cells do provide unrestricted strain and one can test the signal directly for stationarity.

The top of the overconsolidated curves provide the yield points. Thus a model is necessary not only to test for stationarity but also to find a theoretical foundation for the maximum in the force displacement curve. This model should explain the relationship between the interparticle forces, the geometry of the particles, and the observed maximum. Only in this way the cohesion might be explained from first principles.

7.2.2 literature

Only a few models have been suggested in the literature to represent the force/displacement curve in a shear tester of the Jenike type based on the individual behaviour of the particles.

Matchett[7] proposed a simple phenomenological model. He first splits the curve into two successive regions. They are an elastic part in which the stress builds up linearly, and a plastic part when the stress surpasses a certain maximum. This stress is related to the number of bonds across the shear plane. He introduced dynamics by assuming that the bonds in the shear plane are broken in a first order process, and are created proportional to a certain driving force.

This is expressed in the following formulas:

\[ \frac{dN}{dt} = \frac{dN}{dt} \text{(create)} + \frac{dN}{dt} \text{(break)} \quad \ldots(7.2.1) \]
\[ \frac{dN}{dt} \text{ (create)} = + r_1 (N_c - N) \quad \ldots(7.2.2) \]
\[ \frac{dN}{dt} \text{ (break)} = - r_2 N \quad \ldots(7.2.3) \]

\( N = \) number of bonds across the plane
\( N_c = \) maximum number of bonds proportional to the normal stress.

The number of bonds during the displacement is:

\[ N(t) = N_\infty + (N_0 - N_\infty) \exp\left(- (r_1+r_2) t \right) \quad \ldots(7.2.4) \]

\[ N_\infty = \frac{r_1}{r_1+r_2} N_c \quad \ldots(7.2.5) \]

Thus the dynamically steady state is reached after an infinite time. It must be noted that the \( N_c \) parameter has no physical meaning in itself. The force in the plastic part of the curve is proportional to the number of bonds.

\[ F(t) = F_y N(t) \quad \ldots(7.2.6) \]

In the elastic region the force increases linearly:

\[ F(t) = E_s t \quad \ldots(7.2.7) \]

the transition to the plastic part occurring when

\[ E_s t \geq F_y N(t) \quad \ldots(7.2.8) \]

Qualitatively this model does describe the observation of a maximum when the number of bonds before shear exceeded the equilibrium number. However, the assumption of only one type of elasto-plastic bond in a powder is too simplistic to use as a base for further modelling. Also a few assumptions in this model are not so consistent.

Firstly, the mechanisms of bond creation and breakage are identical in the elastic region and the plastic one, and secondly, the driving force is not real but more a model of the observations.

Matchett[5-6] elaborated these solutions and published some higher order kinetics as well. He also introduced the idea that not all bonds are active when the shearing is started. This introduces another first order rate which smooths the transition between the elastic and plastic region. The elastic behaviour of the test system itself can be taken into consideration and can influence the height of the peak in a overconsolidated curve. This means that the yield points are dependent on the instrumentation.
Another fundamental approach by Rowe[9] considers particles. He distinguishes two processes which add up to the observed macroscopic force. Firstly he introduces the idea that the particles in the shear zone have constantly to be expanded against the normal force. He pictures this as a load being transferred along a slope of inclination, $\theta$

$$\theta = \tan \beta \cdot \sigma \quad \ldots(7.2.9)$$

Additionally, there is the friction of the particles themselves. Rowe tried to find a criterion for the angle at which the particles would start to slide and formulated a minimum energy concept. This was not very successful and it has not been elaborated by any other researcher.

In an extensive experimental work Mandl et al.[4] investigated the formation of shear zones in coarse powders (>400μ) in a rotating shear cell. He arrives at the following conclusion: if a overconsolidated force displacement curve is corrected for the force due to the net overall expansion of the powder against the compressive normal force, a monotonic curve is obtained. In other words, the maximum in the curve is caused only by the expansion.

7.2.3 a simulation

In a model the assumptions of Rowe[9] and the observations of Mandl[4] can be included. To this end the macroscopic shear force is split up into three components:

- an elastic component which deform the particles and the contacts elastically,
- a plastic part which allows for the friction between the particles as they slide and rotate in the shear zone,
- an expansion part, that contributes the force needed to continuously move the particles out of the shear zone and against the compressive forces.

$$\theta(\text{total}) = + f(\text{elastic}) \theta(\text{elastic})$$
$$\quad + f(\text{yield}) \theta(\text{yield})$$
$$\quad + \theta(\text{expansion}) \quad \ldots(7.2.10)$$

The instantaneous Yield Locus

The model for the instantaneous yield locus is complex since it is related to a dynamic process which has not yet reached an equilibrium state. At this moment the results can only be qualitative and much experimental work remains to be done to quantify some of the mechanisms. The merit of these models is to direct the experimental work and to concentrate work on certain aspects, so that the future effort will be as effective as possible.
elastic/plastic

It is most unlikely that during the build up of stress only elastic or only plastic deformations occur. Assuming that the macroscopic displacement of the boundaries can reflect the microscopic states, we follow the macroscopic elastoplastic models and split the total displacement into two parts:

\[ \delta = \delta e + \delta p \]  
\[ \frac{d\delta}{dt} = v \]  

...(7.2.11)  
...(7.2.12)

The elastic displacement is proportional to the elastic deformation of the shear zone. Furthermore, this macroscopic displacement is modelled as being proportional to the height of the zone and a critical angle of deformation.

The shear modulus is defined as the quotient of maximum shear force and a critical deformation of the layers:

\[ G = \frac{\Theta_{\text{crit}}}{\tan(\alpha_{\text{crit}})} \]  

...(7.2.13)

Relating \( G \) to the maximal elastic deformation we write:

\[ \delta e(t) = H \cdot \tan \alpha = H \cdot \Theta(t)/G \]  

...(7.2.14)

\[ \Rightarrow \delta e(t) = H \frac{\tan \alpha_{\text{crit}} \cdot \Theta(t)}{\Theta_{\text{crit}}} \]  

...(7.2.15)

Both \( \alpha_{\text{crit}} \) and \( \Theta_{\text{crit}} \) are parameters related to the morphology of the particles and the density of the packing.

\[ \alpha, \Theta_{\text{crit}} = f(\varepsilon s) \]  

...(7.2.16)

One can assume that there is a certain distribution of critical forces and displacements leading to the onset of plastic yield. This distribution is not known. However, it is anticipated that the chance of being in plastic yield, for a particular contact, increases as the macroscopic displacement and shear force on the boundaries increases. This assumption can be translated into simple mathematics.
critical elastic limit

The macroscopic contribution of all plastic displacements of the contacts is $\delta p$, those in an elastic state compound into $\delta e$. As the elastic limit of the shear zone is reached, any further deformation must be due to the extension of the plastic strain only. The fraction of contacts in plastic yield is assumed to be related to the macroscopic displacement in the following way:

$$\text{f}_{\text{yield}} = \left| \frac{\delta e}{\delta e, \text{crit}} \right|^n \quad \ldots(7.2.17)$$

The parameter $n$ represents the elastic behaviour of the powder. If $n$ is small some contacts will fail at low stresses and the behaviour will be a combination of elastic and plastic yield.

$$\frac{d \delta p}{dt} = v \left| \frac{\delta e}{\delta e, \text{crit}} \right|^n \quad \ldots(7.2.18)$$

$$\frac{d \delta e}{dt} = v \left( 1 - \left| \frac{\delta e}{\delta e, \text{crit}} \right|^n \right) \quad \ldots(7.2.19)$$

$$\frac{d \delta e}{dt} + \frac{d \delta p}{dt} = v \quad \ldots(7.2.20)$$

The rate of elastic deformation is also directly proportional to the rate of increase of force (7.2.15):

$$\frac{d \delta e(t)}{dt} = \frac{H \tan \alpha}{\theta \text{crit}} \frac{d \theta(t)}{dt} \quad \ldots(7.2.21)$$

force/displacement curve

The observed force can be divided in three components:

1) The force due to the elastic deformation of the contacts and particles.

2) The force due to plastic deformation of the contacts. The elastic resistance grows until a critical displacement has been surpassed and then a constant plastic resistance remains. This resistance is determined by the friction and attraction on the contacts and, macroscopically, on the morphology of the entire packing of the shear zone.
3) Once plastic yield starts, the particles are translating relative to each other and are pushed out of the shear zone. This expansion is a function of the rate of plastic displacement and a mean angle of contact. See also fig. 7.1.

\[
\tan \beta(t) \cdot \frac{d \delta p}{dt} = H \frac{d \varepsilon_s, e}{dt} = \frac{d h}{dt}
\]  \hspace{1cm} (7.2.22)

Suppose that the angle of contact is a function of the porosity only:

\[
\tan \beta(t) = k_1 \varepsilon_s(t)
\]  \hspace{1cm} (7.2.23)

This assumption is questionable since the angle will also depend on the history of deformation of the packing in the zone. This has, nevertheless, for simplicity been neglected. Also the linear relationship will not be valid for a large region of normal forces and density. One point must be stressed. Only if we assume all the processes to contribute linearly can we disregard the variations and calculate simply with the means of each process. A more complete non-linear model will also have to include the effect of the variations.
substitution of (7.2.23) into (7.2.22) leads to:

\[
\frac{d \varepsilon_{s,e}}{dt} = k_1 \varepsilon_s \frac{d \delta p}{dt} \quad \ldots(7.2.24)
\]

When the zone has become stationary the net displacement of the particles is zero. The number of particles pushed out of the plane and those pushed back are, on average, equal.

effect of plastic deformation on shear zone density

To develop the simple model, we neglect the influence of the elastic displacements on the expansion of the zone and attribute all changes in density to the plastic deformation. Formula (7.2.35) shows the relationship between the expanding component of the shear zone and the plastic deformation. On the other hand the normal forces on the zone have a tendency to compress it and move particles into the layer. The rate of density increase due to the compressive forces will also be dependent on the plastic deformation and the amount of free space available for the particles to move into.

\[
\frac{d \varepsilon_{s,c}}{dt} = -k_2 (\varepsilon_{s,\text{max}} - \varepsilon_s) \frac{\bar{F}_{ns}}{F_a} \frac{d \delta p}{dt} \quad \ldots(7.2.25)
\]

In this formula is \(\bar{F}_{ns}\) the mean compressive force on a particle. It can be related to the macroscopic normal stress if a formula is used such as discussed in chapter 4.:

\[
\bar{F}_{ns} = \frac{\pi D_p^2 \sigma}{k'\varepsilon_s} \quad \ldots(7.2.26)
\]

The relevant density of the contacts is likely to be less in the shearing process than that estimated for the tearing process (Ch.4). Equating them is, therefore, an approximation:

\[
k'1 = k_3 \varepsilon_s \quad \left( k_3 \approx 8 \right) \quad \ldots(7.2.27)
\]

The total rate of change in density of the shear zone is the summation of the rate of expansion and compression.

\[
\frac{d \varepsilon_s}{dt} = \frac{d \varepsilon_{s,e}}{dt} + \frac{d \varepsilon_{s,c}}{dt} \quad \ldots(7.2.28)
\]

Substituting and rearranging one can find a formula which includes the theoretical breakage strength of the zone:

\[
\frac{d \varepsilon_s}{dt} = \left( k_1 \varepsilon_s - k_2 (\varepsilon_{s,\text{max}} - \varepsilon_s) \frac{\sigma}{T} \right) \frac{d \delta p}{dt} \quad \ldots(7.2.29)
\]
For steady state:

\[
d \frac{\varepsilon_s}{dt} = 0, \quad d \frac{\delta p}{dt} = v \quad \text{...(7.2.30)}
\]

\[
\Rightarrow \quad \{ k_1 \varepsilon_s v - k_2 (\varepsilon_{s,\text{max}} - \varepsilon_s) \frac{\sigma}{T} \} v = 0 \quad \text{...(7.2.31)}
\]

\[
\varepsilon_s = \frac{\sigma}{\sigma + k_1/k_2 T} \varepsilon_{s,\text{max}} \quad (\sigma > 0) \quad \text{...(7.2.32)}
\]

These assumptions lead to a formula which shows that powders with a small breakage strength have a nearly constant critical porosity. Also when the normal forces are high, \( \sigma > 20 \times k_1/k_2 T \), the compressibility is negligible.

**Yield stress**

The yield force is composed of the forces to overcome the attraction forces, the sliding friction forces and the rolling friction forces. Breakage and abrasion are not considered in this thesis.

These interparticle forces can be related to macroscopic stresses. See ch. 4. The sliding and rolling friction forces shall be combined. The friction is dependent on the normal compressive forces on the contacts and on the interparticle attraction forces. The latter are a function of the normal forces as well.

\[
\theta_{\text{yield}} = \mu \frac{k_1'}{D_p} \varepsilon_s \frac{F_{ns} + F_{\text{att}}(F_{ns})}{D_p} \quad \text{...(7.2.33)}
\]

(7.2.26) (7.2.27) (7.2.33) \Rightarrow

\[
\theta_{\text{yield}} = \mu \frac{k_1'}{k_1} \sigma + k_3 \varepsilon_s^2 \frac{F_{\text{att}}(F_{ns})}{D_p} \quad \text{...(7.2.34)}
\]

It is seen that this relation is linear if the attractive forces increase linear with respect to the compressive normal forces and the density remains constant or when the combination of density and attractive forces is linear.

**Collecting the components**

\[
\theta_{\text{total}} = f_{\text{elastic}} \theta_{\text{elastic}} + f_{\text{yield}} \theta_{\text{yield}} + \theta_{\text{ex}} \quad \text{...(7.2.35)}
\]
\[ \int \theta_{\text{ex}} \cdot \nu = \int \sigma \frac{dH}{dt} \quad \ldots(7.2.36) \]

\[ \theta_{\text{ex}} = k_1 \varepsilon_s \sigma \frac{d\delta p}{\nu \, dt} \]

\[ = k_1 \varepsilon_s \sigma f_{\text{yield}} \quad \ldots(7.2.37) \]

\[ \theta_{\text{elastic}} = \frac{\theta_{\text{crit}}}{H \tan \alpha} \delta e(t) \quad \ldots(7.2.38) \]

The maximum, critical elastic force is bounded by the plastic shearforce:

\[ \theta_{\text{crit}} = \theta_{\text{yield(\varepsilon_s)}} \quad \ldots(7.2.39) \]

The limit of the macroscopic elastic displacement has been correlated to a critical angle of displacement and to the height of the zone:

\[ \delta e_{\text{crit}} = H \tan \alpha \quad \ldots(7.2.40) \]

\[ \Rightarrow \theta_{\text{elastic}} = \frac{\delta e}{\delta e_{\text{crit}}} \theta_{\text{yield(\varepsilon_s)}} \quad \ldots(7.2.41) \]

Summing all the contribution leads to the following description of the total shear force:

\[ \theta = \left( \underbrace{\frac{\delta e}{\delta e_{\text{crit}}} \theta_{\text{yield(\varepsilon_s)}}}_{\theta e_{\text{crit}}} \right)^n + \left(1 - \underbrace{\frac{\delta e}{\delta e_{\text{crit}}} \theta_{\text{yield(\varepsilon_s)}}}_{\delta e_{\text{crit}}} \right)^n \]

\[ + k_1 \varepsilon_s \sigma \frac{\delta e}{\delta e_{\text{crit}}} \quad \ldots(7.2.42) \]

For small deformations the shear zone is nearly elastic:

\[ \theta(t) = \theta_{\text{yield(\varepsilon_s)}} \frac{\delta e}{\delta e_{\text{crit}}} \quad \delta e \ll \delta e_{\text{crit}} \quad \ldots(7.2.43) \]

For larger deformations more contacts become plastic and the expansion of the shear zone sets in until a new equilibrium is reached.
Fig 7.2: Simulation of force-displacement curves.

a: porosity vs. shear as a function of normal load
b: steady state density as a function of normal load
c: force and density curves vs shear
   the maximum force occurs while the density changes.
Numerical results

Some numerical integrations have been carried out to investigate the behaviour of various components introduced in this model. Curves for a compressible powder are shown in fig. 7.2.

The stationary yield locus for an incompressible powder are straight and go through the origin. A compressible powder has an approximately straight locus for higher compressive forces, but it curves back into the origin at lower normal loads.

The maxima in the curves are due to the expansion of the zones. The peak occurs whilst the state of the powder is in transition between two steady states. This shows that the state of the powder is not so well defined for the yield points and may well be dependent on the geometry of the shear zone which the machine imposes on the powder.

The elasticity parameter determines the sharpness of the maximum. If \( n \) is high no expansion occurs until the stress is very near to the yield stress of packing of the original steady state. Due to the elastic behaviour the force displacement curve is straight. Then an abrupt expansion sets in and the maximum is slightly shifted. The sharp decrease in density diminishes the yield stress at the same time and the competing mechanisms produce a sharp maximum. See fig. 7.3.

If the elasticity parameter is low, the plastic behaviour sets in at moderate stresses and expansion occurs throughout the entire curve. The maximum is broad and somewhat lower than for a similar powder with higher elasticity. Comparing the curves to those obtained in the Jenike Cell for the BCR-limestone samples, the cohesive limestone would be modelled with a high elasticity parameter.

The maxima themselves are related in a complicated way to the parameters describing the friction, the attractive forces and the expansion of the powder. The cohesion too is, therefore, a complex variable and is not related only to the attractive interparticle forces.

There is no apparent, simple connection between the process of shearing and the process of tearing. This model does not support the hypothesis that the tensile strength is an extrapolation of the yield locus.
Fig 7.3: Simulation of the yield loci. The effect of increasing interparticle attractive forces. a: zero attraction, b: unit attraction, c: two unit attractions.
7.2.4 Algorithms for curve characterization

In order to obtain an objective and reproducible decision on the stationarity of the signal, an algorithm has been designed and tried. This algorithm replaces the subjective decision of the operator on whether a test run is acceptable or not.

The signal from the shear testers is interfaced with a computer, digitalized and about 100 equidistant points per run are stored on diskette for later use in the algorithm.

To judge the acceptability of the signal the following tests must be carried out:

- An estimate of the deviation of the signal to the stationary end value using an exponential model.
- A test on whether overconsolidation has occurred, thus testing for a maximum.
- An estimate of the variation of the signal around the idealized mean curve.

**Fitting the signal**

Assuming that the signal can be split into two parts, one in which the signal is mainly influenced by elastic deformations and the other mainly by the plastic deformation and densification of the shear zone, the signal has been fitted with two successive exponentials using the least squares method.

\[
F(t) = A_1(1 - C_1 \exp(-B_1 t)) \quad t \leq to \quad \ldots(7.2.44)
\]

\[
F(t) = A_2(1 - C_2 \exp(-B_2 t)) \quad t > to \quad \ldots(7.2.45)
\]

The point of transition can also be determined in various ways; one method which proved to be quick and robust was as follows. After estimating to after the 10th point the exponent was fitted for \((tend = to)\) and for \((tend = to + \delta t)\). When the Chi-square for the extended fits was lower to was set to \(to + \delta t\). This was repeated until the Chi-square increased.

Local increases of Chi-square were allowed when the parameters proved to be within bounds of the previous one \((+/- 10 \%)\) and when Chi-square diminished again with the next step. This ensured that local disturbances were ignored and the length of the first fit not terminated too soon.

The maximum length for the first fit was arbitrarily set to half the signal, in order to ensure that the second fit was based on enough data points. This criterion was seldom met, and the signal then always proved to be underconsolidated.

**estimation of the variation**

The variation of the signal around its idealized curve is estimated with the sum of the squares of the residues around the fit of the second part of the signal with the exponential.
Fig 7.4: The decision algorithm applied to two Jenike tests. The fitted exponentials almost coincide. The first curve was classified as good, the second as underconsolidated. The smaller curves are measurements of yield points.

formula. This is an high estimate.

\[ \sigma^2 = \sigma^2(\text{fit}) \] \hspace{1cm} (7.2.46)

test on deviation from stationarity

The stationary end value of the signal is estimated with the end value of the exponential fit.

\[ F(\text{signal}, t=\infty) = F(\text{exponent}, t=\infty) \] \hspace{1cm} (7.2.47)

The signal is considered acceptably close to the stationary steady state if the mean of the last 10 points is within 2 % of this estimate.

\[ F(\text{exponent}, t=\infty) - 2\% < F(\text{end}) < F(\text{exponent}, t=\infty) + 2\% \] \hspace{1cm} (7.2.48)

test on overconsolidation
The signal is overconsolidated and rejected when the maximum of the signal is higher than the mean of the last 10 points plus the 95% reliability interval estimated with the variance of the signal.

\[ F_{\text{top}} > F(\text{end}) + 1.97 \sigma \]  \hspace{1cm} \text{(7.2.49)}

test on underconsolidation

If the end signal is lower than the acceptable range and the signal is not overconsolidated it is rejected as underconsolidated.

The indication of underconsolidated and overconsolidated is a useful instrument for feedback to the operator trying to find the proper consolidation procedure. See fig. 7.4.

7.3 The Jenike Shear Tester

7.3.1 method and equipment

Jenike's method is the most common method used to measure yield loci. The method and equipment are described in detail in his own work [3].

In this work a cell with identical dimensions was built, the only difference being the force transducer for which a piezo electric crystal was used (a Kistler load cell).

The equipment can be criticized since the stress condition in the cell is not well defined, and the path available to obtain steady state is rather short. These problems make it difficult to find the right procedure to bring the powder to a critical state within the limitations given. Further, the forces measured are not always in agreement with the forces in the shear plane because of the poor transmission of the applied loads [1,8].

Haaker[1] has studied this in detail and has published a number of correction factors for the problems usually encountered. He also finds that, due to a fortunate coincidence the flow function derived from the faulty measurements agrees rather well with the correct one.

Even though the equipment is not the most suitable for precise scientific investigation, its use for industrial and technological designs has been proven. As a practical instrument it will not quickly be displaced with another instrument.
Robustness of the Jenike method

Delft University of Technology participates in a European programme in which 6 expert laboratories characterized a standard limestone sample with a number of instantaneous yield points using the Jenike equipment and method. The results of this programme are a suitable example to discuss the reliability of the estimate of a yield point. See appendix 7A.

After the first evaluations the robustness of the method with respect to small variations in the measurement procedure and to different operators seemed to be small. From this the safe way is to standardize the procedure completely, even on what was considered previously unimportant points.

Standardizing the measurement procedure

The procedure can be split in four parts:

a) filling of the rings
b) precompressing
c) consolidation of sample until steady state is reached
d) measurement of the yield point.

These parts are now explained in more detail.

a) filling of the rings.

Before filling, the top ring is placed against the positioning pins. As an aid to filling, a mould, high enough to provide material for the compression, is placed on top ring.

The set of rings is then entirely filled and the top is levelled with a knife. Excess powder is scraped off. Then the lid is placed on top and the powder compressed with a certain load.

b) precompressing

Since the Jenike cell has a limited distance of displacement, it is necessary to obtain as close as possible the critical packing density. This minimizes the distance needed to come to stationarity. Two methods have been advocated by Jenike: loading the top with an extra weight and twisting the top.

The extra weight is a weight above that which will be used when the consolidation starts.

The twisting movement causes the particles to be compressed even further, but it is not clear how uniform the packing will be. It is known that if the top lid only is twisted, which can be
achieved when the top ring is firmly held in place, a different result is obtained than when the top ring is allowed to rotate with the lid. The twisting action is not easy to standardize, probably a fully automatic machine must be build.

After precompressing for a time long enough to allow any trapped air to escape the mould is removed carefully. The lid is shifted by horizontal movement in order to avoid the build up of underpressure. The excess powder above the top ring is scraped off. A special lid with a pin which transmit the displacement of the force transducer is placed on top and the sample is now ready for consolidation.

c) consolidation to the critical state

To start the consolidation the pin is displaced at a constant rate. When the pin reaches the lid, the force starts to build up and the top ring is moved. The steady state should ideally be reached just before the top ring and bottom coincide. Then the geometry is perfect and the area of contact is precisely known.

The decision on whether the sample has really come to a steady state is made by the operator. The outcome of a series of runs relies on the experience and reliability of the operator.

d) measurement of the yield point

When the steady state is reached the displacement pin is retracted and the powder allowed to come to its resting position. The normal weight is removed and a new lower weight is applied. The displacement is immediately started again and the maximum of the resulting overconsolidated curve is the yield point sought.

7.4 Experimental 1

The improvement and standardization of this procedure

Standardization and improvement of the procedure was proposed on the following points:

- elimination of the influence of the operator, by using an objective algorithm to decide whether an acceptable curve was obtained. The algorithm has been described above.
- homogeneous filling of the rings.
- a more precisely defined precompression

Of these the first one is the most important since only an objective procedure allows one to compare the effects of other changes.
7.4.1 methods of preparation

As stated before there are two means to increase the initial density of the sample, a higher extra weight or increasing the number of twists.

According to Haaker[1] a higher number of twists is beneficial to the formation of a smooth, stationary plane. He also investigated the influence of different levels of extra weight on the transmission of the normal pressure onto the shear plane. If the density is low the rings are likely to touch each other and a certain fraction of the normal pressure will be deviated from the shear plane itself. A higher compression will prevent this. Experimentally he found that an extra weight of about 100% of the normal weight will make the internal pressure equal to the externally applied pressure.

Using this recommendation of extra weight, a single test was carried out 7 times in succession. No special care was taken to fill the rings and the powder was deagglomerated by cutting it with a knife.

The data were analysed with the algorithm, see table 7.1. Curves that were underconsolidated are marked as (---), those which are overconsolidated (+++). Acceptable curves are followed with a (0).

<table>
<thead>
<tr>
<th>N</th>
<th>Østat [kPa]</th>
<th>Density [kg/l]</th>
<th>Curve</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>6.021</td>
<td>1.156</td>
<td>---</td>
</tr>
<tr>
<td>1</td>
<td>6.055</td>
<td>1.161</td>
<td>+++</td>
</tr>
<tr>
<td>2</td>
<td>6.680</td>
<td>1.190</td>
<td>0</td>
</tr>
<tr>
<td>3</td>
<td>6.401</td>
<td>1.176</td>
<td>0</td>
</tr>
<tr>
<td>4</td>
<td>7.085</td>
<td>-</td>
<td>0</td>
</tr>
<tr>
<td>5</td>
<td>7.119</td>
<td>1.156</td>
<td>0</td>
</tr>
<tr>
<td>6</td>
<td>5.957</td>
<td>-</td>
<td>0</td>
</tr>
<tr>
<td>7</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Mean 6.474 1.168 70%
Stand.Dev. 0.498 0.015
S.D. % 7 1.5

Table 7.1: successive tests: 0 twists, 19 kPA precompressing, 9 kPa consolidation.

The 95% interval of the normal distribution will be called the spread. The spread in the average bulk density, derived from the weight only, is about 3%. This is high and according to other procedures some of these samples would have been rejected. A well formed curve is obtained in 70% of the cases. Nevertheless
the spread in the stationary values is large: ca. 14%.

This high value is probably caused by fluctuations in the packing morphology caused by inhomogeneous filling or a inhomogeneous compression by the weights. These fluctuations can be avoided by standardizing the filling procedure. The following procedure enabled a completely fresh operator to achieve the same performance as a very experienced one.

The filling and precompression

1. sieve the powder through a course sieve to break any large agglomerates.
2. Fill the bottom ring for 2/3 with a spoon gradually with layers of powder, levelling each carefully with the spoon.
3. Place the lid on the powder and precompress the layer using the consolidation weight plus the extra weight. Take care the weights do not swing. Do not twist, at least very slightly and if so over an angle of 1 degree at the most. Maintain a fixed time of compression which allows all the air to escape.
4. Remove the weight. Rotate the lid to loosen it from the powder and carefully lift it so as not to disturb the layer.
5. Position the top ring against the pins.
6. Fill bottom and top ring for 2/3, in the same fashion as before. Place layer by layer and equalize the top layer. If, after compression, it happens that the level of the powder coincides with the position of the shear plane the 2/3 must be replaced with 3/4.
7. Repeat step 3 and 4
8. Position the mould.
9. Fill the whole system, layer by layer. Remove excess powder with a knife.
10. Repeat step 3 and 4
11. Remove the mould and scrape off excess powder above the top ring
12. Position the lid with the transmission pin
13. Carefully place the consolidation weight
14. Start the motor
15. Displace the top ring until it is just aligned with the bottom
16. retract the displacement pin until it no longer touches the lid and remove the consolidation weight the yield point
17. Place the normal weight for the yield point measurement on
the lid, taking care that it does not swing.

18. Restart the forward motion of the pin and record the overconsolidated curve until the maximum has passed.

determination of bulk density.

19. Determine the weight of the powder in the cell.

The characteristics of this procedure are:

- the cell is filled in three layers each is separately preconsolidated.
- the twisting action is reduced preferably to zero.
- the precompression weight is about twice the consolidation weight.
- the travel for consolidation is standardized and the geometry of the test system optimal.
- the decision about the acceptability of the measurement is taken by an objective computer algorithm.

7.4.2 Measurement of yield points

A series of 10 consecutive measurements which were carried out by an experienced operator using this procedure are shown in table 7.2. The limestone contained 0.27 % w/w water. This was determined by heating for several hours at 120 °C.
Table 7.2: Results of the new procedure: precompression at 19 kPa, consolidation at 9 kPa and the yield point at 5 kPa

<table>
<thead>
<tr>
<th>N</th>
<th>$\sigma=9$ kPa</th>
<th>$\sigma=5$ kPa</th>
<th>Density [kg/l]</th>
<th>Curve</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\theta_{\text{stat}}$ [kPa]</td>
<td>$\theta_{\text{max}}$ [kPa]</td>
<td></td>
<td></td>
</tr>
<tr>
<td>H6</td>
<td>6.377</td>
<td>4.248</td>
<td>1.224</td>
<td>0</td>
</tr>
<tr>
<td>H3</td>
<td>6.538</td>
<td>4.150</td>
<td>1.205</td>
<td>0</td>
</tr>
<tr>
<td>H10</td>
<td>6.582</td>
<td>4.248</td>
<td>1.220</td>
<td>0</td>
</tr>
<tr>
<td>H7</td>
<td>6.646</td>
<td>4.248</td>
<td>1.215</td>
<td>0</td>
</tr>
<tr>
<td>H5</td>
<td>6.650</td>
<td>4.297</td>
<td>1.200</td>
<td>---</td>
</tr>
<tr>
<td>H2</td>
<td>6.659</td>
<td>4.252</td>
<td>1.215</td>
<td>0</td>
</tr>
<tr>
<td>H8</td>
<td>6.665</td>
<td>4.346</td>
<td>1.151</td>
<td>---</td>
</tr>
<tr>
<td>H1</td>
<td>6.665</td>
<td>4.297</td>
<td>1.205</td>
<td>0</td>
</tr>
<tr>
<td>H4</td>
<td>6.768</td>
<td>4.150</td>
<td>1.215</td>
<td>0</td>
</tr>
<tr>
<td>H9</td>
<td>6.821</td>
<td>4.346</td>
<td>1.205</td>
<td>0</td>
</tr>
</tbody>
</table>

Statistics of the 8 accepted curves:

<table>
<thead>
<tr>
<th></th>
<th>Mean</th>
<th>$\theta_{\text{stat}}$</th>
<th>$\theta_{\text{max}}$</th>
<th>Density</th>
<th>Curve</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>6.632</td>
<td>4.242</td>
<td>1.213</td>
<td>80%</td>
<td></td>
</tr>
<tr>
<td>Stand.dev.</td>
<td>0.129</td>
<td>0.062</td>
<td>0.007</td>
<td></td>
<td></td>
</tr>
<tr>
<td>SD % $\approx$</td>
<td>2</td>
<td>1.5</td>
<td>0.5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The results in table 7.2 show a significant reduction of the spread in the data. The percentage of acceptable measurements is high: 80%. Also the variation in the density is very low. If we compare these results with those obtained by an inexperienced operator, having done no such test before and only instructed in this procedure, the sets are almost identical.
Table 7.3: As table 7.2, Results of the second operator.

<table>
<thead>
<tr>
<th>N</th>
<th>$\sigma=9$ kPa</th>
<th>$\sigma=5$ kPa</th>
<th>Density [kg/l]</th>
<th>Curve</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\bar{\sigma}$</td>
<td>$\bar{\sigma}_{\max}$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B3</td>
<td>6.450</td>
<td>4.053</td>
<td>1.244</td>
<td>0</td>
</tr>
<tr>
<td>B9</td>
<td>6.504</td>
<td>4.297</td>
<td>1.224</td>
<td>0</td>
</tr>
<tr>
<td>B10</td>
<td>6.523</td>
<td>4.297</td>
<td>1.234</td>
<td>0</td>
</tr>
<tr>
<td>B1</td>
<td>6.528</td>
<td>4.297</td>
<td>1.244</td>
<td>0</td>
</tr>
<tr>
<td>B5</td>
<td>6.699</td>
<td>4.346</td>
<td>1.234</td>
<td>0</td>
</tr>
<tr>
<td>B4</td>
<td>6.719</td>
<td>4.248</td>
<td>1.224</td>
<td>0</td>
</tr>
<tr>
<td>B7</td>
<td>6.772</td>
<td>4.346</td>
<td>1.239</td>
<td>0</td>
</tr>
<tr>
<td>B8</td>
<td>6.787</td>
<td>4.248</td>
<td>1.229</td>
<td>0</td>
</tr>
</tbody>
</table>

Mean: 6.673 | 4.254 | 1.234 | 80%
Stand. Dev.:
S.D. % 2 2 1

The two data sets cannot be distinguished on statistical grounds.

The correlation between the data was calculated for the entire set of 16 measurements. The sample correlation coefficient, $C_{xy}$, is given by Himmelblau [2]:

$$C_{xy} = \frac{\sum_{i=1}^{n} (X_i - \bar{X})(Y_i - \bar{Y})}{\sqrt{\sum_{i=1}^{n} (X_i - \bar{X})^2 \sum_{i=1}^{n} (Y_i - \bar{Y})^2}}$$  \hspace{1cm} \ldots(7.4.1)

table 7.4: the correlation coefficient for the shear test data

<table>
<thead>
<tr>
<th>Data</th>
<th>$C_{xy}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>stationary value / yield point</td>
<td>0.2517</td>
</tr>
<tr>
<td>stationary value / bulk density</td>
<td>0.5432</td>
</tr>
<tr>
<td>yield point / bulk density</td>
<td>0.1704</td>
</tr>
</tbody>
</table>
No significant correlation between the stationary values and the yield points could be detected. Prorating, as recommended in some procedures is, therefore, not advisable. It only obscures the data and increases the spread.
The usefulness of prorating is probably only in correcting gross errors, which did not occur in these measurement.

The more severe test for the new procedure is the measurement of the yield points at extremely low compressive stresses. The lowest point determined in the standardization program is the 1 kPa yield point for the steady state of 2 kPa. Twenty consecutive measurements were performed at these conditions.

Table 7.5: Results using the new procedure.
precompression at 5 kPa, consolidation at 2 kPa and the yield point at 1 kPa,

<table>
<thead>
<tr>
<th>N</th>
<th>$\sigma_{stat}$ [kPa]</th>
<th>$\sigma_{max}$ [kPa]</th>
<th>Density [kg/l]</th>
<th>Curve</th>
</tr>
</thead>
<tbody>
<tr>
<td>kalkY17</td>
<td>1.510</td>
<td>1.170</td>
<td>1.127</td>
<td>+++</td>
</tr>
<tr>
<td>kalkY9</td>
<td>1.560</td>
<td>1.221</td>
<td>1.141</td>
<td>+++</td>
</tr>
<tr>
<td>kalkY15</td>
<td>1.610</td>
<td>1.070</td>
<td>1.127</td>
<td>+++</td>
</tr>
<tr>
<td>kalkY19</td>
<td>1.646</td>
<td>1.318</td>
<td>1.132</td>
<td>0</td>
</tr>
<tr>
<td>kalkY18</td>
<td>1.660</td>
<td>1.220</td>
<td>1.107</td>
<td>0</td>
</tr>
<tr>
<td>kalkY3</td>
<td>1.753</td>
<td>1.270</td>
<td>1.146</td>
<td>0</td>
</tr>
<tr>
<td>kalkY1</td>
<td>1.772</td>
<td>1.270</td>
<td>1.146</td>
<td>0</td>
</tr>
<tr>
<td>kalkY12</td>
<td>1.710</td>
<td>1.070</td>
<td>1.141</td>
<td>+++</td>
</tr>
<tr>
<td>kalkY21</td>
<td>1.710</td>
<td>1.120</td>
<td>1.093</td>
<td>---</td>
</tr>
<tr>
<td>kalkY6</td>
<td>1.710</td>
<td>1.320</td>
<td>1.156</td>
<td>0</td>
</tr>
<tr>
<td>kalkY13</td>
<td>1.743</td>
<td>1.070</td>
<td>1.127</td>
<td>0</td>
</tr>
<tr>
<td>kalkY14</td>
<td>1.748</td>
<td>1.070</td>
<td>1.190</td>
<td>---</td>
</tr>
<tr>
<td>kalkY11</td>
<td>1.753</td>
<td>1.221</td>
<td>1.137</td>
<td>0</td>
</tr>
<tr>
<td>kalkY2</td>
<td>1.758</td>
<td>1.221</td>
<td>1.102</td>
<td>0</td>
</tr>
<tr>
<td>kalkY8</td>
<td>1.802</td>
<td>1.416</td>
<td>1.146</td>
<td>0</td>
</tr>
<tr>
<td>kalkY7</td>
<td>1.810</td>
<td>1.120</td>
<td>1.137</td>
<td>---</td>
</tr>
<tr>
<td>kalkY16</td>
<td>1.810</td>
<td>1.120</td>
<td>1.146</td>
<td>0</td>
</tr>
<tr>
<td>kalkY5</td>
<td>1.810</td>
<td>1.270</td>
<td>1.185</td>
<td>+++</td>
</tr>
<tr>
<td>kalkY20</td>
<td>1.860</td>
<td>1.170</td>
<td>1.127</td>
<td>---</td>
</tr>
<tr>
<td>kalkY4</td>
<td>2.021</td>
<td>1.030</td>
<td>1.151</td>
<td>---</td>
</tr>
</tbody>
</table>
Selecting the ten acceptable measurements:

<table>
<thead>
<tr>
<th></th>
<th>1.758</th>
<th>1.221</th>
<th>1.102</th>
<th>0</th>
</tr>
</thead>
<tbody>
<tr>
<td>kalkY2</td>
<td>1.660</td>
<td>1.220</td>
<td>1.107</td>
<td>0</td>
</tr>
<tr>
<td>kalkY18</td>
<td>1.743</td>
<td>1.070</td>
<td>1.127</td>
<td>0</td>
</tr>
<tr>
<td>kalkY13</td>
<td>1.646</td>
<td>1.318</td>
<td>1.132</td>
<td>0</td>
</tr>
<tr>
<td>kalkY19</td>
<td>1.753</td>
<td>1.221</td>
<td>1.137</td>
<td>0</td>
</tr>
<tr>
<td>kalkY11</td>
<td>1.810</td>
<td>1.120</td>
<td>1.146</td>
<td>0</td>
</tr>
<tr>
<td>kalkY16</td>
<td>1.753</td>
<td>1.270</td>
<td>1.146</td>
<td>0</td>
</tr>
<tr>
<td>kalkY3</td>
<td>1.772</td>
<td>1.270</td>
<td>1.146</td>
<td>0</td>
</tr>
<tr>
<td>kalkY1</td>
<td>1.802</td>
<td>1.416</td>
<td>1.146</td>
<td>0</td>
</tr>
<tr>
<td>kalkY8</td>
<td>1.710</td>
<td>1.320</td>
<td>1.156</td>
<td>0</td>
</tr>
</tbody>
</table>

Mean: 1.741 1.245 1.135 50%
Stand.dev.: 0.052 0.095 0.017
S.D. %: 3 8 1.5

Table 7.6: Correlation between the stationary value, bulk density and yield value.

<table>
<thead>
<tr>
<th>Data Correlated</th>
<th>Cxy</th>
</tr>
</thead>
<tbody>
<tr>
<td>stationary value / bulk density</td>
<td>0.3721</td>
</tr>
<tr>
<td>stationary value / yield value</td>
<td>0.1108</td>
</tr>
<tr>
<td>yield value / bulk density</td>
<td>0.3721</td>
</tr>
</tbody>
</table>

As expected, the percentage of acceptable measurements is lower for these difficult conditions. Both underconsolidated and overconsolidated curves occur at the same condition, indicating that minor changes already can change the form drastically.

Whereas the spread of the stationary value is only slightly increased, that of the yield point is dramatically higher at 16 %.

Looking at the correlation coefficients no significant relation can be found. Again prorating cannot be justified.

7.4.3 Discussion

Applying a strict procedure, aimed at forming a very homogenous powder mass at a density close to the critical state, it is possible to obtain a high percentage of acceptable curves within the limited travel of the Jenike cell.
The algorithm based on the exponential approach of the critical state appears to be a good objective decision maker on the acceptability of a curve. The parameters chosen for acceptance have not yet been completely optimised, but the 2% range appears to coincide with the limit which a human operator would use.

In general one can say that the mathematical description of the curves allows comparison and in future expert systems will help the operator decide on more characteristics than is possible at this moment.

Comparison of the results of the standardized procedure with those obtained in the expert laboratories shows that the procedure is at least as good as the experienced operators. It must be noted, also, that in the measurements no screening on grounds other than the form of the curve was allowed.

The spread in the stationary values is 4% for the 9 kPa and 6% for the 2 kPa critical state. The spread in the yield points is 2% and 18% respectively for the 5 and 1 kPa yield points. The first result is better, the second comparable to previous measurements done in this laboratory without the standardized procedure. In absolute values the standard deviation of any point is about 0.1 kPa.

Since no significant correlation between the stationary values and the yield points could be detected, prorating is not justified. Also the bulk density and the yield points are uncorrelated, which agrees with the concept of the formation of a critical zone in which there is a characteristic density independent of the initial density.

However, since it is known that different bulk densities may give different stationary yield values, the range within which the data are uncorrelated must be limited. The simulation incorporates the idea that the height of the shear zone has an impact on the elastic behaviour of the zone. One can speculate that the height of the zone is dependent on the bulk density, the more dense the system is the more narrow the shear zone. This would relate the influence of the bulk density to the yield points.

The simulation shows that with a few reasonable assumptions curves can be obtained which are very similar to those recorded for real powders. A smooth transition between the elastic build up of the force to the stationary friction has been introduced.
Fig 7.5: The Coaxial Shear Cell
7.5 the Coaxial Shear Tester

7.5.1 Limitations of translational equipment

The main practical disadvantage of the translational cells, such as the Jenike cell, is the limited displacement possible. A solution to this problem has been sought in rotating ring cells, which have unlimited travel. The top of the cell rotates relative to the bottom and a horizontal shear plane is formed. In these geometries problems arise because of the difference in the displacement and the rate of displacement between two points in the shear plane at different radii. In the most extreme case, the Peschl cell, two plates rotate above each other and the centre of the powder theoretically does not travel at all.

Some of the problems have been solved by Scarlett and Todd, who tried to estimate the influence of the walls and difference in travel by splitting the top lid into three concentric parts, measuring the force only on the middle one.

The problem is most acute when one tries to reproduce the sequence of events a shear plane experiences in a translational cell. The formation of a stationary plane is not a problem, but the subsequent release of the shear stress of the entire plane is impossible. If the powder near to the centre is sheared by rotating in the reverse way to release the stress, the powder further away travels more and already builds up a reverse shear stress. When the overall stress is reduced to zero, only at one distance is the condition really neutral, all the other positions are still stressed. Wilms et al.[11] discussed this problem in more detail. When the forward motion at a lower normal force is started again, the resulting overconsolidated curve can be seen as compounded from an infinite number of overconsolidated curves each associated with one radial distance.

To avoid these problems a new type of rotating shear cell was built in which the shear plane is coaxial with the axis of rotation. In this case, all points in the central plane of the shear zone travel at the same speed and through the same distance. See fig. 7.5.

This choice of geometry also enables a more clearly defined shear plane to form. In the translational types the form may be lenslike, more thick in the middle and converging towards the walls. Measurements in rotating shear cells by Scarlett and Todd show that the shear zone extends for at least 6 to 10 particle diameters.

The circular geometry of the coaxial cell causes the shear plane to form at the smallest possible radius, where the stress exerted
Fig 7.6: The development of a thin shear zone. Above: the shear zone can be seen as a thin line. Below: after shearing material is easily removed as it simply falls off the shear zone when it is no longer supported.
on the system is largest for a given moment of torque. This radius can be precisely controlled by inserting vanes. The thickness of the shear zone will also necessarily be narrow and one could expect a different force/displacement characteristic in this system as compared to the others.

7.5.2 Description of the Coaxial Shear Tester.

For the benefit of future investigations with various different cells, the Coaxial Shear Tester (CST) has been designed in two independent parts: a separate turntable and an exchangeable cell.

a) The turn table is powered by a motor, driven stepwise and with a variable, controlled speed between the limits 10 - 5000 rpm. A gear reduction slows this speed to 0.01 - 5 rpm, and a further reduction of 1 to 10 can be selected to run the machine in a directly motor controlled mode. Alternatively a mechanical cyclic reciprocal motion up to an angle of 30° can be chosen. A clutch is used to select either the direct mode, which is controllable between the limits 0.0001 - 0.5 rpm, or the cyclic mode, controllable between the limits 1.5 10-5 - 0.08 Hz. The turn table and the central axis rotate and can be used as a platform to mount different cells.

b) The Coaxial Cell

See fig. 7.7. The coaxial cell is built with the following components:

1) a inner ring. This is a hollow cylinder which is centred on the axis with a ball bearing. This cylinder remains stationary when the turntable rotates and the torque on this cylinder is recorded. The cylinder is equipped with a very flexible rubber membrane to transmit a pneumatic pressure to the powder. The membrane is coaxial with the central axis.

2) the outer ring. Also a hollow cylinder coaxial with the central axis. This is fixed to the turntable and rotates with it. On the inner side there is a second membrane, again coaxial, to transmit pressure to the powder contained between the inner and outer ring.

3) the closure between the inner and outer ring is made by a ring of paper which slightly overlaps the gap. Previous designs without the paper but with a ring of felt closing the gap failed due to the quick deterioration of the felt. The friction introduced by this paper was negligible.
Fig 7.7: A schematic drawing of the Coaxial Shear Cell

a  inner membrane
b  outer membrane
c  vanes
d  air inlet (pressure is bleed controlled)
e  load cell
f  shear zone
4) The top lids. The cell is closed on top with two lids, their gap being aligned with the gap in the bottom. These lids were tightened with screws and once fixed the pressure was not controlled any further. A future extension will be to control also this pressure on the lids in order to be able to vary two of the principal stresses instead of one. This can be done with springs as used by Mandl[4] for his ring shear cell, or by loads which are stabilized against tilting around the central axis by ball bearings.

5) The torque transducer. The torque on the inner ring is transmitted to a piezo electric crystal, which is designed for both tensile and compressive stresses. This allows the measurement of cyclic stress patterns.

7.6 Experimental 2

7.6.1 The formation of the shear zone

Using an alumina powder, tracer experiments have been carried out to follow the formation of a shear zone in the coaxial cell. Perpendicular to the zone a narrow band of coloured material, a mixture of carbon black and alumina, was inserted before shear as applied. By using very little carbon black and very narrow band the tracer material was assumed not to change the characteristics of the shear zone. After rotating the deformation of this band could be studied without disturbance by using a vacuum tube to suck away the covering layers until the depth under consideration was reached.

The shear layer appeared to be located at the radius of the vanes. It was perfectly vertical except in the regime close (about 2 mm) to the top and bottom lids where the plane curved towards the actual gap. Since the shear plane was homogenous over most of its length, repeated measurements could be carried out on the formation of the shear plane whilst using the same tracer band. After examination of the tracer band at a certain depth, the hole was filled, the lids replaced and the shear deformation increased. The progression of the formation of the plane could then be monitored by looking at the same tracer band, progressively somewhat deeper. Five to six observations were made on each experiment. See fig. 7.8.

The photographs show that at the beginning of the strain, the material is deformed in a rather large zone. The shear deformation is largest in the middle and gradually diminishes at the ends until it can not be seen. As the macroscopic deformation increases the shear deformation also increases until a certain critical value is surpassed and any further deformation of the material occurs mainly in a very thin zone. The depth of this
Fig 7.8: The shear zone at different heights in the cell.

a  vane
b  tracer material
c  shear zone
Fig 7.9: Initial formation of a shear zone in the coaxial cell
Fig 7.10: Initial formation of a shear zone in the coaxial cell
zone is estimated to be of the order of a few particle diameters. See figs. 7.9-10. In some runs the shear plane did not develop regularly. As a previously formed shear band was blocked a second shear band was formed to release the stress. This mechanism is intriguing but could not be pursued further.

7.6.2 force/displacement curve

The filling procedure for the coaxial shear tester is less elaborate than that for the Jenike cell, the powder being placed in horizontal layers. The lid is closed and fastened, care being taken not to have a higher pressure on top than that which will be used on the membranes.

The force displacement curve of the coaxial cell is comparatively smooth. Since the surface of the shear plane is about 4 times as large as that in the Jenike cell this could be expected. The distance of travel before steady state is reached for a sample which has just been introduced can be compared to that in the Jenike cell. If the load is reduced for a while and then brought back to the original value a steady state is reached almost immediately. The stationary condition could be maintained over a large distance (>1 m).

Whilst the local fluctuations around the mean are very small, a long term fluctuation could be detected when the vanes were not perfectly aligned. The variation in the shear plane, introduced by the uneven placement of these vanes, must be minimized for optimum results.

The form of the force-displacement curve is different than that obtained by using the procedure of Jenike to measure a yield point. A number of procedures have been followed but all yielded the same result. To measure a yield point the stress on the shear plane was first released by slowly turning backwards until the overall shear pressure was zero. The air pressure on the membranes was then released and, after a few moments, was set to the desired pressure. The turntable was started again at the original speed in the forward direction.

A very small first maximum is noted, but after this peak the signal rises monotonically to a higher maximum, as if the sample is underconsolidated. Neither the first peak nor the second maximum can therefore be seen as the yield value. See fig. 7.11. This mechanism is not yet fully understood. At this moment the tests using the coaxial instrument have to be interpreted as stationary measurements. For non-cohesive materials the yield locus and stationary locus coincide and the results of Jenike tester and coaxial tester agree well.
7.6.3 Comparison of the CST and the Jenike Shear Tester

Measurements have been performed in the cyclic mode on a sample of alumina powder which behaves as a Sand-like material. The yield locus in the CST has a low intercept and the angle compares with that in the Jenike cell. At very low membrane stresses the stress due to the lids causes a deviation of the straight locus, if the lids are removed a value of Cohesion can be measured which coincides well with the intercept of the straight part of the locus. See fig. 7.12.

The stationary values for the cohesive BCR limestone samples do not compare well. The results from the CST show a stationary locus with has a rather high intercept and a angle of friction that is smaller than that obtained in the Jenike cell. See fig. 7.13.
7.6.4 Discussion

The coaxial cell shows that a stationary shear zone can be maintained for a large distance of deformation. The coaxial shear plane avoids the problems of the radial geometry of traditional ring cells and this instrument seems, therefore, more suitable for scientific research on shear planes. The formation of shear plane can be studied in detail. The results of this work show that the deformation sets in a rather large region until a critical deformation in a plane has been reached. Thereafter the deformation narrows down to a very thin shear zone.

In the CST stationary values can be determined in a quick and reproducible way. However, comparing the results for cohesive powders with those obtained previously in a Jenike cell, the absolute values do not match well. The reason for this is as yet unknown and since no absolute true test is available the reason may be sought in both instruments.
Fig 7.13: Yield loci of limestone obtained in the BCR programme, in the Jenike cell+ decision algorithm and in the coaxial cell.
Also the form of the force displacement curves when using the procedure for yield point determination deviates from that obtained in other cells. Inhomogeneous stress distributions in both cells or differences in the packing due to different consolidation procedures could be responsible for this. The coaxial arrangement may introduce different stress conditions as the powder expands and contracts due to changes in the macroscopic forces. These effects have not yet been explained, and further investigation is needed.

7.7 Conclusions

The definition of the macroscopic cohesion of a particle system is closely related to the concept of the Yield Locus and the way in which these are determined. The measurements show that the reproducibility of a yield point determination is highly dependent on the procedure followed. Unfortunately there is no absolute way of determining what is the true value for any condition and the only rule we can apply is that a procedure which yields results with a high resolution is more desirable. Related to a high resolution is a small spread in the results.

The known procedures for the Jenike test have been investigated and some improvement to the following points reduced the spread in the results:

- A procedure to make a more homogeneous packing of the powder
- The position of the cell at the moment of measuring the yield point has been standardized.
- The decision on whether a curve is acceptable or not has been automated by using a computer algorithm.

In order to obtain a density close to the critical state thus giving a curve which comes to the steady state quickly enough, the procedure for precompressing suggested by Haaker has been followed. An extra weight of 100 - 120 % resulted in a high number of acceptable curves. The results of the measurements showed that the standardized procedure and the algorithm are as good as the expert laboratories. It can be foreseen that algorithms such as these will aid or even replace the skilled operator in future.

The results further showed that there is no correlation between density, stationary end value and the yield points. This is somewhat surprising since the prorating of the yield point is generally accepted for reducing the spread in the results. Prorating is an undesirable procedure when the data are uncorrelated since it will only result in more scatter of the results.
The formation and the stationarity of a shear zone has been investigated in the coaxial shear tester. The tracer experiments showed that a vertical shear plane can be formed, which provides the means to investigate the processes occurring in a shear plane in which all elements deform at the same rate. The deformation starts in a broad zone, less at the end and highest in the plane determined by the end of the vanes. When a layer has been subjected to a certain critical deformation, all further deformation occurs in that layer only, effectively forming a very small shear layer with a thickness of only a few particles.

On a particle scale, one could visualize what happens: at the beginning of the deformation the particles rearrange somewhat and start forming a new packing morphology. As the deformation proceeds particles have to move outward and the density of the layer decreases. When one layer reaches a critical porosity the resistance on that plane is greatly reduced and all further deformation occurs in that plane only. Due to the coaxial geometry the layer closest to the central axis which is still free to deform will experience the highest shear stress for a given torque. That plane will therefore deform most and come to the critical state first. Obstacles or locally consolidated parts may cause the plane to deviate.

In a translational cell, this driving force is absent and the plane will be more sensitive to local variations in resistance, multiple planes maybe possible. The location of the plane is more obscure, only the start and endpoints being certain. Investigation of a shear plane is probably more easily done in a coaxial cell.

The stationarity of the shear plane could be followed for a large distance of travel. The short term fluctuations are very small, due to the large surface of the shear plane, long term fluctuations can arise when the deviations in the positions of the vanes is much larger then the particles themselves. Well aligned these fluctuations appear also to be small (<5%).

The absolute values of the shear pressure on the zone in the coaxial and the Jenike cell do not compare well, Also the form of the stress displacement during the yield point procedure deviates considerably.

These effect have not yet been clarified, probable causes could be a difference in stress conditions or in the particle packing in the shear zones.

The simulation of the stress-displacement curves yield very similar results to the curves obtained in the Jenike cell. The model is still a qualitative one since the parameters have not yet been quantified for the actual processes occurring in the powder.

The main conclusion of this simulation is that the maximum in a curve occurs whilst the expansion is in progress. The state of
Fig 7.14: Inhomogeneous shear mechanisms: plastic yields starts at the edge and gradually progresses inwards.

The powder lies between the critical state appropriate to the consolidation condition and that for the load at which the yield point is determined. Since the path from one state to the other may not be unique, this state is, therefore, not completely defined and may well be different for various geometries of shear testers.

The microscopic forces of attraction are an important component in the maxima, but the relation is a complex one. The attraction forces determine the cohesion indirectly through their influence on the compressibility of the powder. On a whole, the maximum is determined by a combination of the friction forces between the particles, the attractive forces and the packing of the powder at the moment of yield.

This combination of factors complicates speculation on the influence of a change in any of these components on the yield locus. On the one hand, one could speculate that the adsorption of liquid on the particles might enlarge the interparticle forces and consequently increase the cohesion. On the other hand, the layer of liquid might lower the friction between the particles and thereby diminish the resistance to shear and reduce the angle of friction of the yield locus and also the intercept on the axis.

The model for the stationary locus states that the density of the
shear zone will be dependent on the compressive stress. As the zone densifies the resistance increases and the stationary locus will be convex upward. It further shows that, at higher normal forces, the compressibility diminishes and the stationary yield locus becomes a straight line with a positive intercept.

The form of the instantaneous yield locus is highly dependent on the compressibility too, because of the large influence of the expansion on the maxima. However, this model does not explain the concave yield loci which are encountered in reality. This may be an indication that an important effect has been overlooked.

One probable process may be the non-uniform shearing of the plane. The plastic deformation may start at the edge of the system to progress gradually inward until the entire plane is in motion. See fig. 7.14. This process would effectively lower the expectation of the force at lower normal stresses more.

Looking at the contributions which make up the yield point in this model, we can roughly define three regions. The points near to the consolidation point at high normal stresses, the points near to the cohesion at low normal stresses and the intermediate points. In the high region the expansion of the zone is small and the maxima are mainly due to the friction and attraction between the particles. In the low region the expansion is high but the energy needed to achieve this is low. Thus, here the expansion is also a minor factor. The intermediate region is heavily influenced by the expansion, the combination of load and expansion requiring high forces. Because of this extrapolation from the intermediate region to the extreme regions will be doubtful unless one has the correct formula which describes the mechanisms controlling the yield locus is available.

The cohesion itself is determined by the friction and the attractive forces only. The zone will expand, however, and the exact morphology of the packing at the point of maximum force is as yet unknown. Since it is most likely that some contacts will be broken sooner than others and since there are no normal forces to create new contacts, the shearing at zero compresses resembles the processes during the tearing of a powder system.

In conclusion one can state that the yield points using the Jenike equipment are not uniquely defined and are dependent on the procedure followed. Cohesion, being the extrapolation of these yield points, is, therefore, also not unique. Theoretically it shows that the extrapolation itself may be questionable if the proper function is not known.

Further research will be needed to explain the relation between
this normal tearing process and the shearing tear.

7.8 symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
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<tr>
<td>B1, B2</td>
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<td>C1, C2</td>
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<td>Cxy</td>
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Greek

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<tr>
<td>μ'</td>
<td>friction coefficient</td>
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subscripts

a, att  attraction
e, elast  elastic
p, plast  plastic
c, crit  critical
y, yield  yield
ex  expansion
com  compression
stat  stationary
max  maximum

7.9 References


Appendix 7.A Standardized procedure versus expert laboratories

Six different, internationally known laboratories have carried out a programme to standardize a limestone sample according to their best interpretation of the Jenike test. These measurements were screened: that is samples with a bulk density that was more than 2 % off their mean value were rejected. Since the results have not yet been made public the laboratories are coded.

Table 7.A.1: the results for the 5 kPa Yield Point at the 9 kPa stationary state.

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<td>St.Dev. [kPa]</td>
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<tr>
<td>A:</td>
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<tr>
<td>B:</td>
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<td>0.303</td>
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<td>C:</td>
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<td>D:</td>
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<td>0.150</td>
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<tr>
<td>E:</td>
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<td>F: Delft</td>
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<tr>
<td>mean values</td>
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</tr>
<tr>
<td>Stand.Dev.</td>
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<td>0.068</td>
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| Delft 88             | 6.627   | 0.128    | 4.254   |           |
Table 7.A.2: The results of the 1 kPa Yield Point for the 2 kPa Stationary state.

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<td>C:</td>
<td>1.514</td>
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<td>0.086</td>
<td>0.009</td>
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</table>

Delft 88

1.741 0.052 1.245 0.095
Fig 8.1: The consistency of aerated powders as measured in a powder viscometer. Hobbel and Scarlett(1985).
\( \Omega_1 = \) speed inner cylinder, \( \delta \Omega = \Omega_2 - \Omega_1 \), \( \Sigma \Omega = \Omega_2 + \Omega_1 \).

Above: the torque as a function of the speed of both cylinders.
Below: the consistency regions as a function of gas velocity. 
- A: yield stress
- B: mobilizing region
- C: fluid region
8.1 Introduction

The influence of the interparticle forces on the shear behaviour of aerated systems will be discussed in this chapter. Due to the relatively open structures, that are sustained by the fluid drag, the frictional forces in these systems are very small. Also the structure itself is easily disturbed when a probe is inserted. In order to investigate the shear behaviour of the aerated state, a powder viscometer was used. This viscometer is able to measure the resistance of the shear zone in an aerated powder without disturbance of the structure or gas flow pattern[12]. For this study the instrumentation has been improved to allow continuous torque measurement with strain gauges. The torque measured with this device can be described in terms of normal stresses and shear stresses in the aerated powder, and the behaviour characterized with an yield locus and a Cohesion.

8.2 Theory of aerated and fluidized systems

The line that divides the aerated state and the fluidized state is rather vague. In most literature state of a powder is said to be fluidised when the gas flow exceeds the minimum fluidising velocity. This is, however, an arbitrary definition and does not relate well to the physical reality. In this thesis we shall define the state as aerated when the gas velocity is lower than the fully supported velocity and as fluidised when the velocity is larger than that value. Because of this difference, some confusion may rise in relating to the literature, but the terminology is suggested because a subdivision which has direct physical meaning will ultimately be more useful. The commonly used term 'rheology' will be used to express the shear behaviour of aerated powder.

8.2.1 Literature on rheology

Little has been published on the subject of the rheology of aerated powders[12-14], since the literature has paid more attention to the fluidised state[3,4,7,10,11,18,19]. The aerated state is currently commanding more attention because it is important in explaining flooding phenomena, where the powder flows rapidly due to its expanded state. For the fluidised state Schuegerl et al.[18,19] investigated in detail the behaviour of various systems. He immersed a smooth rotating cylinder into the fluidized bed and recorded the resultant torque. He concluded that the consistency of an fluidised powder can be well approximated with an Eyring
equation. Some deviations from this general trend are discussed. Hobbel et al. [12,13] investigated the general consistency of aerated systems using a Couette viscometer design. The cylinders were made of gauze to eliminate slip and disturbance of the delicate structure. The consistency proved to be a function of the gas velocity and both speeds of the cylinders. In the low shear rate region the resistance diminished as the shear rate increased. This is called the mobilizing region, the decrease in resistance being due to the shearing of the bed and to vibrations introduced by the cylinders. As the shear rate is increased beyond the mobilizing region the resistance is relatively insensitive to the deformation rate, a situation comparable to the Coulomb behaviour of compressed powders. See fig. 8.1. Close to the yield point, where the speed of the cylinders is reduced to practically zero, the behaviour proved to be very interesting but difficult to probe with the device as it was. For this purpose the viscometer was adapted and the results are the subject of this chapter.

In general the rheological behaviour of fluidized powders is strongly dependent on the particle size distribution, the fluid drag, and the interparticle forces[1]. The powder behaviour will be explained using these three terms only. The interparticle forces include both the attractive forces as well as the frictional forces.

The consistency of an fluidized powder is closely related to its fluidisation characteristics as expressed by the pressure drop curve, the expansion behaviour and the contraction when the gas flow is reduced.

8.2.2 Fluidisation classes

The fluidisation characteristics of a powder can be roughly described by Geldart's fluidisation classification. Based on a large number of experimental observations of various powders, Geldart[8,9] showed that 4 sets of data clustered reasonably well if their characteristics were mapped onto a two dimensional space of density difference and mean particle size. He proposed the classification of cohesive powders as the C-class, homogeneously fluidizable as A and inhomogeneously fluidizable or sand-like powders as B-class. See fig. 8.2.

According to Geldart's procedure the A and B classes may be discriminated after examining the pressure drop versus gas velocity characteristic of a powder. If the minimum fluidising velocity and the minimum bubbling velocity are nearly coincident, then it is a B-powder; if there is a wider difference between these two values it can be classified as an A powder. The advantage of this idea is that a first impression of the behaviour can be made based on simple parameters and easily accessible information.
Fig 8.2: Geldart's classification. Above: the original plot by Geldart. Below: a dimensionless representation by Rietema, who defines a cohesion parameter.
Using Geldart's plot various researchers have investigated the criteria by which the various classes can be discriminated. The consensus of opinion is that the behaviour of powders can be explained in terms of the relative importance of their interparticle forces (IPF). This explains the clustering of the data: the density difference and mean particle size are supposed presumably reasonable indicators of the fluid drag and the interparticle forces.

Molerus[15] showed that a reasonable line of division can be drawn between classes A and B by assuming that the criterium is a constant ratio of the IPF and the fluid drag force. The ratio he calculated, however, is such that his statement that the IPF are completely negligible for B-powder, cannot be true.

Other investigators have concentrated on correlating the MF and the MBP with the mean particle size. Estimation of these points would, according to the criteria set by Geldart, also give a prediction of A and B behaviour. Thonglimp[20] has given an extensive review of all the published correlations.

**alternative variables for IPF**

Since mean particle size can only give a rough indication of the forces involved, other definitions may be more useful. One refinement would be to use another weighting factor in order to give a proper estimate of the effect of the distribution of size on the IPF. This suggestion is elaborated in appendix 8.A. Alternatively, an estimation of the cohesive forces involved could be made from macroscopic experimental parameters. Rietema[17] defined a cohesion number using the angle of inclination a bed can sustain at the minimum fluidising velocity. This angle can be determined by extrapolation of the values determined at lower gas velocities. According to the definition of the classes of Geldart one would expect a B powder to bubble at the minimum fluidising velocity and the angle of inclination at the minimum fluidizing velocity to be zero. see Verloop[22]. Using this cohesion number and also a dimensionless number for the fluid drag Rietema derived a new plot in which the boundaries between the classes were defined more clearly. This line of thought has, however, not been further pursued in the published literature. See fig. 8.2. The tilting bed methods have been used to demonstrate the existence of structures in homogeneously fluidized powders [6,16,17].

8.2.3 Characteristic features of aerated/fluidized systems

**Minimum Bubbling Point.**

Since the resistance of a packing to fluid flow is lower when the
Fig 8.3: The relationship between the stresses, the bed height and the pressure drop across the bed for perfect and imperfect powders.
fluid flows through larger channels, there is a tendency to form large channels in the structure of the aerated powder. The role of the IPF in this is to stabilize any network of contacting particles. It therefore counteracts the growth of the larger channels, but it also stabilizes the walls of channels which already exist against collapse due to the compressive stresses caused by gravity. This dualism must be clearly distinguished, because the behaviour of a bed differs with increasing or decreasing gas flow.

The Fluid drag forces and the IPF can be seen as competing mechanisms. When the fluid drag just overcomes the IPF at the MBP, the homogeneous bed can no longer be sustained and it segregates into voids and a dense phase. The location of the MBP is an important indicator of the relative magnitude of the IPF involved. Increasing the IPF results in a higher MBP, if all other conditions are constant.

Fully Supported Velocity

In explaining the behaviour of real powders, the concept of a Perfect Powder is useful. A PP is a powder in which interparticle attractive and frictional forces are both absent. The behaviour of this powder during expansion and contraction illustrates the ideas which form the foundation of many theories. See fig. 8.3. As the gas flow increases, starting from a packed bed, the pressure drop grows linearly until the entire weight of the bed is supported. At that point, the supporting point, the bed expands to release the stress due to the increasing flow. The pressure drop across the bed is constant and equal to the weight per unit area of the bed. Decreasing the flow, the bed contracts until the minimum fluidising point is reached and the powder returns in its original dense packing. The expansion and contraction curves coincide with no hysteresis. This is an important feature of the Perfect Powder.

A real, imperfect powder does have interparticle attraction and friction forces. Any change in the relative position of the particles will be counteracted by these forces. Due to this hinderance the bed resists any change and a marked hysteresis between the expansion and contraction curves will occur. This hysteresis is larger when the IPF are more prominent. The pressure curve is closely related to the bed height since it is proportional to the density of the bed. This pressure curve also shows hysteresis for imperfect powders.

Comparing the expansion branch of the pressure curve of a real powder which has small IPF and that of a corresponding PP with the same particle geometry it can be noted that a small over pressure in excess of the supporting pressure may develop. This situation is not stable, any disturbance may break the resistance
and the bed will expand. Further increase of the gas flow does not change the absolute pressure drop which is equal to the supporting pressure.

A powder with large IPF expands in irregular steps creating a non-reproducible pressure curve. These cohesive powders will not be discussed further, in this thesis attention will be concentrated solely on the A and B powders which have moderate IPF.

The contraction branch of the curve starts at the supporting pressure but deviates from the perfect line at lower pressures. This is due to the resistance of the powder to contraction which results in a less dense bed for the more imperfect powders. When the contraction is further reduced the bed densifies and the pressure drop curve becomes more linear. The point of deviation is called the Fully Supported velocity, and is an important indicator of the relative importance of the IPF. Unfortunately, no correlations have yet been published between the FS and particle size, research has been concentrated on the minimum fluidizing velocity.

The minimum fluidising velocity.

Because nearly perfect powders lose their resistance to flow at the fully supported point, this point was also termed the minimum fluidizing velocity. For a perfect powder this point is accurately determined by the intersection of the linearly increasing portion of the pressure drop curve and its horizontal part.

Harrison proposed that imperfect powders be characterized in a similar way. The minimum fluidising velocity for an imperfect powder is defined by the intercept of a linearized portion of the curve and the horizontal supporting pressure. Since the hysteresis of the curve leads to ambiguity in this definition only, the contraction branch should be used.

For sand-like materials the increasing portion of the curve is reasonably straight for a certain range and the minimum fluidizing velocity can be determined with accuracy.

For more cohesive materials the curvature of the pressure drop curve extends throughout the entire range. A straight line can only be obtained by fitting an arbitrary portion of the curve. The fundamental physical meaning of the intercept itself is obscure. It does not mark any significant change in the behaviour of the powder. To avoid any misunderstanding about the physical meaning of the MF for an imperfect powder, in this chapter the MF will be termed the apparent minimum fluidizing velocity.

The AMF is highly dependent on density at low gas velocities which can be influenced by external vibrations during the
contraction. These vibrations destabilise the structure and, therefore, diminish the influence of the IPF. The AMF shifts towards the lower gas velocities with increasing power of the external vibrations and will, in the limiting case, be equal to the MF of the corresponding perfect powder.

In general it can be stated that the difference between the AMF and the FS, determined under controlled conditions of external vibration, are a measure of the importance of the IPF. If the AMF and FS coincide it is a Perfect Powder.

The mathematical models for powders which are based upon the concept of a PP will necessarily predict a value too low if compared to the AMF. Only after calculation of the stabilization of the bed due to IPF the AMF can be corrected for this difference.

The compressive stress

A force balance shows that if the powder is not fully supported by the fluid drag, an internal compressive stress must act upon the structure. Analogous to the pressures developed in silos, it can be assumed that Janssen's formula is also valid for aerated powders. For vessels with a large width to depth ratio the wall friction can be neglected and assuming that both the pressure drop across the bed and the density are uniform this stress is proportional to the depth of immersion.

\[ \Sigma(h) = | \frac{W}{A} - dP | \cdot \frac{h}{H} \]  \hspace{1cm} \text{...(8.2.1)}

It can be speculated that, for narrower vessels the effect of wall friction is to release pressure of the structure and thus to allow a less dense packing shifting the FS and the AMF to higher velocities.
Fig 8.4: Geometry of the immersion probe. For this project BII geometries were used.

Fig 8.5 The powder viscometer

- a: inner cylinder  Ø 4 x 6 cm
- b: outer cylinder  Ø 8 x 9 cm
- c: measuring gap  2 cm
- d: fluidised bed container
- e: level of the bed
- f: porous metal plate
8.3 Equipment.

The apparatus is a wide gap couette viscometer, schematically shown in fig. 8.4. Its cylinders have been adapted to aerated open structures. Constructed of thin gauze it permits measurements with minimal disturbance of the packing and gas flow pattern. The cylinders do not promote bubbling. See fig. 8.5.

The screen size is large enough to allow free migration of particles through the gauze. Because the particles in the holes form a rough cylinder, additional slip relative to the cylinder is assumed to be minimal and was neglected in the analysis of the results.

The cylinders are submerged in the bed, which is held in a perspex container, 15 cm in diameter. The fluid is introduced through a metal porous plate. The torque on the inner cylinder is recorded using a strain gauge torque transducer which is placed directly above the inner cylinder, thus avoiding any noise from the shaft bearing. The signal from the transducer is transmitted through a set of slip rings to an Analog/Digital converter which is interfaced with a computer, thus allowing a signal sampling rate up to 100 Hz.

The system can run in either a torque or a strain controlled mode. For the torque control the shaft is equipped with a coaxial torque spring which can be loaded by a motor. If strain control is used the shaft is driven directly by means of a pulley and a motor. The speed of the motor can be regulated to rotate between limits of 0.4 and 120 rpm in either direction.

The outer cylinder of the couette is driven directly by a similar motor.

relation stress and torque

In order to relate the stress on the boundaries of the inner cylinder to the torque recorded, the total torque is regarded as consisting of two elements: the stress on the cylinder wall and the stress on its base. There are two complications. First the geometry of the shear zone near to the wall is well known and is cylindrical, whereas that on the base may be slightly curved. Secondly account must be taken of the difference of the direction of the gas flow through the shear zones. Near to the wall the flow is along the shear plane, on the base it is perpendicular to it. For this a correction factor has been introduced whose magnitude is yet unknown.
Fig 8.6: The stresses on the wall and base of the immersion probe.

See fig. 8.6: \( M_{\text{tot}} = M_{\text{wall}} + M_{\text{bottom}} \)

\[
M_{\text{wall}} = \int_0^H (2\pi R) R \theta(h) \, dh = 2\pi R^2 \int_0^H \theta(h) \, dh \tag{8.3.1}
\]

\[
M_{\text{bottom}} = \int_0^R f \pi R^2 \theta(H) \, dr = f \frac{2}{3} \pi R^3 \theta(H) \tag{8.3.2}
\]

Supposing that the compressive stress is directly proportional to the immersion depth and that the Coulomb law is valid, it follows that the relation between shear stress and immersion depth is also linear. On the other hand, it may be that the density of the bed increases as the compressive stress rises and that this effect also will enlarge the increase in friction as the depth increases.

Even if the true relationship is not linear, for small depths of immersion this assumption would be a valid first approximation.

\[
\sigma(h) = A' \cdot h \tag{8.3.3}
\]
Fig 8.7: A slip-stick experiment. A motor loads a torque spring. As the torque increases the rate of deformation of the shear zone increases until the maximum in the consistency curve is reached. Then the resistance against flow diminishes with increasing velocity and the cylinder slips forward.

\[ \theta = \mu \sigma + C \]  
\[ \Rightarrow \theta(h) = Ah + C \]  
\[ \text{...(8.3.4)} \]
\[ \text{...(8.3.5)} \]

In this formula C is the macroscopic cohesion and a function of the stability of the structure. It is dependent on the gas velocity. The parameter A describes the rate of increase of the internal stress.

substituting (8.3.5) in (8.3.1) and (8.3.2):

\[ M_{\text{wall}} := \pi R^2 Ah + 2 \pi R^2 Ch \quad \text{...(8.3.6)} \]
\[ M_{\text{bottom}} = f. 2/3 \pi R^3 Ah + f 2/3 \pi R^3 C \quad \text{...(8.3.7)} \]
\[ M_{\text{tot}} = (\pi R^2 A) h^2 + (2\pi R^2 C + f 2/3 \pi R^3 A)h + (f 2/3 \pi R^3 C) \quad \text{...(8.3.8)} \]
Fig 8.8: A typical signal of the torque transducer during a yield experiment in the viscometer. At a constant rate of deformation the torque is larger than the so-called residual torque, which is measured directly after stopping the motion of the cylinder. The torque is released at a rate which depends on the mobility of the powder.

This shows that the relationship between the depth of immersion and the torque is parabolic. Using data determined for different immersion depths allows estimation of these parameters by using a fitting procedure. Once the parameters are known, it is possible to calculate the relationship between the true stress on the wall and the total torque.

8.4 Experimental materials

In this project two sand powders, a coarse and a fine one, are selected. The characteristics are described in Chapter 2. According to their mean size and density they would be classified as B powders.

Experiments

Three types of experiments were run on the viscometer:

1) slip stick measurements: See fig. 8.7.
When the torque on the inner cylinder is slowly increased at a constant rate, the powder first resists the stress, and the deformation of the shear zone is limited. When the maximum is reached, the zone suddenly yields and the cylinder slips forward thus releasing the stress. The torque recorded against time shows a characteristic saw-tooth pattern. Because of the repetitive slipping, this mode is commonly called the slip-stick regime. The maximum torque recorded is proportional to the yield stress of the powder.

2) continuous measurements. See fig. 8.8.

The consistency plot of the powder can be obtained by direct strain control of the cylinder and recording the resistance as a function of the speed of both the inner and outer cylinder. The signal is typically a dynamic signal, the fluctuation around the mean being dependent on the gas velocity. Previous measurements showed that the consistency of an aerated powder is more complex and not analogous to true fluids, the resistance of the shear zone in some conditions decreasing as the rate of deformation increases. In this project the deformation rate is low, 0.4 rpm, whilst the outer cylinder was kept stationary.

3) residual stress. See fig. 8.9.

When the rotation of the cylinder is arrested after a period of continuous deformation, the stress on the cylinder drops instantly to the residual stress. After this initial drop a much slower relaxation occurs, the rate of which is dependent on the gas velocity.

8.4.1 Visual observations and pressure drop recordings.

To obtain the fluidising characteristics of the samples, their contracting pressure drop curve was recorded. The bed was fluidized vigorously before the gas flow was adjusted to the required lower value. Both the samples are imperfect, their Amf and FS differ somewhat. It was observed that, at a velocity between Amf and FS, small, local, highly active channels were formed. Voids travelled to the surface through these channels. Apparently these structures were stable since some of them existed for an appreciable time. As the gas velocity was slowly reduced, the number of channels increased, but their size diminished, resulting in a smaller fraction of the surface being occupied. Increasing the gas flow, the bubbling through the channels became more severe, breaking up the structures around them and eventually destabilizing the entire bed. At that gas velocity the bubbles did not rise at fixed positions any more, but appeared to emerge randomly over the entire surface. In order to discriminate between these two modes of bubbling the point 8.9
Fig 8.9: Pressure drop and residual yield stress versus gas velocity for alumina samples. 
$\times, + = \text{coarse}, \quad \circ, \bullet = \text{coarse + 10\% fines}$. 

\[ \Delta P / \Delta P_{\text{max}} \quad \text{versus} \quad V_g \quad (\text{mm/s}) \] 

\[ M_y \quad (\text{Nmm}) \]
Fig 8.10: Pressure drop, residual and continuous yield stress versus gas velocity for fine sand.
Fig 8.11: Pressure drop, residual and continuous yield stress versus gas velocity for coarse sand.
where small channels still exist, is called the Local Bubbling Point (LBP), the velocity at which the bubbles appear at random, is called the Random Bubbling Point (RBP).

The LBP is very sensitive to vibrations and to shear: the channels are destabilized and collapse due to the compressive stress.

For a Perfect Powder the LBP and RBP coincide with the AMF and FS point. For both sand samples the FS and RBP coincided, within the resolution of visual observation.

8.4.2 The rheological measurements

The results of the continuous and residual stress measurements are shown in figs. 8.9-8.11.

**stick-slip and continuous measurement**

The typical slip-stick behaviour is more apparent when the gas velocity is substantially less than FS. Close to the FS point slip-stick and continuous deformation cannot be differentiated, the cylinder slips continuously.

**residual stress**

The residual stress is also an indication of the yield strength of the bed. It shows that it is highly dependent on the gas velocity, decreasing when the velocity approaches the FS. Above the FS no residual stress can be measured.

The consequent, slower relaxation is also a function of the gas velocity. Its rate is almost zero for low gas velocities, steadily increasing at higher flows.

Vibration, tapping and other disturbances cause the rate of this relaxation to increase. Above the LBP, where bubbles rising through the channels cause vibrations in the surrounding bed, these pressure fluctuations also aid the relaxation. At the AMF point, no significant changes in any parameter were noted. This was expected.

**immersion depth**

A number of immersion curves were recorded for two cylinders of diameter 4 and 6 cm. respectively, see figs 8.12-8.14. The torque can be adequately represented by a parabola, and is clearly not a linear function of depth. After minimizing the sum of residues with respect to the correction factor, \( f=2 \) appeared to be optimal. In table 8.1. the results of the least square fitting, for \( f=2 \), are shown.

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Fig 8.12: Immersion experiments, the radius of the probe is 4 cm.
  a: vg= 15.6 mm/s,  b: vg=18.0 mm/s,  c: vg=20.4 mm/s.
Fig 8.13: The friction rate parameter versus the compressive stress

Fig 8.14: the cohesion versus the superficial gas velocity
table 8.1: The parameters of function ## for 2 different cylinders and sand 1, B=2.

<table>
<thead>
<tr>
<th>Vg mm/s</th>
<th>R=2</th>
<th>R=3</th>
<th>δP mmH₂O</th>
<th>Σ mmH₂O</th>
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<tr>
<td>15.6</td>
<td>808</td>
<td>8.00</td>
<td>-</td>
<td>98</td>
</tr>
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<td>16.8</td>
<td>498</td>
<td>5.95</td>
<td>-</td>
<td>104</td>
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<td>17.4</td>
<td>507</td>
<td>6.34</td>
<td>-</td>
<td>108</td>
</tr>
<tr>
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<td>368</td>
<td>4.47</td>
<td>288</td>
<td>111</td>
</tr>
<tr>
<td>18.6</td>
<td>90</td>
<td>9.50</td>
<td>167</td>
<td>114</td>
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<tr>
<td>19.2</td>
<td>173</td>
<td>3.59</td>
<td>-</td>
<td>116</td>
</tr>
<tr>
<td>20.4</td>
<td>16</td>
<td>4.07</td>
<td>64</td>
<td>118</td>
</tr>
</tbody>
</table>

8.5 discussion

The measurements on the residual stress show the existence of a structure that can withstand a stress. This structure has a low relaxation speed as long as there are no vibrations to disturb it. Above the LBP one finds a marked increase in the relaxation speed. This relaxation will be faster as the bubbling is more vigorous. The residual stress itself decreases steadily until it completely disappears at the FS.

This total lack of rigid structure is the condition for free inhibited bubbling: the bubbles will emerge randomly distributed across the entire surface of the bed. Therefore the FS point and RBP coincide and each is an indication of the other. Models of the minimum bubbling point, which suppose complete destabilization of the bed, actually model the RBP and not the LBP. Since the LBP is easily identified as the minimum bubbling point this may cause confusion. Probably all correlations for the minimum bubbling point are estimates of the LBP.

To our knowledge no theoretical models for partial bubbling have yet been published in the literature.

When using a tilting fluidised bed to measure the cohesion of a structure the results will depend on both the yield stress and the relaxation speed. If the relaxation is low a definite angle may be noted. As it increases the bed will steadily deform and the conclusion may be that no structure exists. The tilting bed method is therefore limited in its ability to show the structure of an aerated bed.

Extrapolation of data obtained at lower gas velocities to the AMF implicitly assumes that at the AMF compressive stresses are absent, a situation which is true for perfect powders only.
A rotating viscometer does not have these limitations. Relaxation, residual stress and the consistency of the aerated structure easily can be obtained at all gas velocities.

The effect of the gas velocity on the resistivity of B-powders can now be more clearly explained in term of the AMF, LBP and RBP. Chen et al.[5] showed that the resistivity enlarges steadily until a marked change in rate of increase occurs at the LBP. The authors determined this as the minimum bubbling point. At the AMF has no apparent physical implications for this process. When at the FS or RBP the bed becomes fully mobile and collapses due to the loss of structure. This is correlated with an reduction of the resistivity. Thus, the characteristic behaviour of the resistivity follows the fluidizing behaviour.

Comparing the results of the continuous and the residual stress, the residual stress is always somewhat less. Previous measurements at higher rates of deformation showed that the resistance of the bed can be reduced by increased speed of the cylinders. Apperently the consistency plot goes through a maximum, which is located at very low deformation speeds. This accounts for the typical slip stick behaviour as well. As the torque builds up, the rate of deformation increases steadily until a critical rate has been reached. At that point the resistance suddenly reduces and allows the cylinder to slip forward. The maximum stress recorded in the slip stick measurements represent the maxima of the consistency curves.

Analogous to compacted powders a distinct shear layer develops in the packing due to the strain. When starting the slip-stick measurement in a undisturbed packing the very first peak is significantly less then the maxima in steady state. The structure of the undisturbed bed is, therefore, different from the fully developed shear zone. However, the strength of an undisturbed bed will be proportional to the strength of the shear zone, and conclusions derived for the shear zone will qualitatively be valid for an undisturbed aerated packing.

The immersion experiments clearly show the non-linear increase in resistance. see fig 8.12. This agrees with the theory of increasing compressive stress in the bed. Fitting the entire set of data for the minimum sum of errors yielded an correction factor of 2. The mechanism controlling the magnitude of this factor is not yet known. It can be argued that the shear plane does not coincide with the cylinder geometry, and/or that due to anisotropy the resistance of a horizontal plane is fundamentally different from that of a vertical plane.

The parameters in the formulas which results from the assumptions of increasing normal stress and Coulomb behaviour are strongly correlated. When fitting the curves with a least square criterium the variation in one parameter is compensated with an variation
in another. This obscures detection of trends in the parameters. Nevertheless, if the parameter \( A \) is plotted against the overall compressive stress on the powder a linear relation describes the data adequately:

\[
A = k \cdot \Sigma \\
k = 30 \text{ (1/m)} \quad \ldots(8.3.9)
\]

The cohesion is small and the variation large but a trend can be seen when plotted against the superficial gas velocity. The data can be represented with:

\[
C = k_2 \cdot (FS - V_g) \quad \ldots(8.3.10)
\]

Previous experiments with an alumina, A-type powder, support the hypothesis that the yield resistance is in general directly related to the compressive stress, see fig 8.9. These measurements were performed with a less sensitive system and the yield stress could not be measured at higher gas velocities. Further research on these A-type powders may enable us to generalize the findings for the more perfect sand-like powders.

The findings that an aerated powder behaves as a Coulomb solid has implications for the measurement of the properties of rising bubbles in a aerated bed. It cannot be assumed that Newtonian or Bingham consistency is valid. These models will not predict the stresses in the struture above the roof of the void properly. For an accurate calculation the yield stress and the influence of normal stresses on the resistance of the structure must be included. Furthermore, different behaviour of the bubble may be expected when injected in a bed operated under LBP or above LBP.

8.6 Conclusions.

Using a rotating viscometer it can be shown that above the AMF and above the LBP structures exits that can withstand a stress. These structures are under compressive stress, the fluid drag compensating for a portion of its weight. The resulting shear behaviour can be modelled as a Coulomb-solid. In the range of velocities investigated the cohesion for the sand-like sample was small, of the order of 1 to 10 Pa. There is a tendency for the cohesion to reduce as gasflow increases.

The aerated structures are stabilized by the IPF and destabilized by drag forces and vibrations. The higher the gas velocity the higher the rate of relaxation and the lower the residual stress. At the FS the yield stress is zero and the bed is fully mobile. Bubbles can rise through the bed at random and this velocity is termed the RBP. At gas velocities below the RBP localized bubbling may occur, the lowest velocity at which this occurs is
called the LBP.

It is argued that if the MBP is modelled as the velocity at which complete destabilization occurs, as is assumed in the literature, it then must be identified as the RBP and not the LBP. In experimental work the LBP is easily mistaken for this MBP.

It is suggested that a characterization of powders based on the FS rather than on the AMF would have more physical significant meaning for imperfect powders. Using yield stress measurements and pressure drop readings this point can be measured reasonably accurate.

8.7 Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>friction parameter</td>
<td>(N/M3)</td>
</tr>
<tr>
<td>B</td>
<td>friction correlation factor</td>
<td>(- )</td>
</tr>
<tr>
<td>C</td>
<td>cohesion parameter</td>
<td>( Pa )</td>
</tr>
<tr>
<td>D</td>
<td>diameter cylinder</td>
<td>( m )</td>
</tr>
<tr>
<td>h</td>
<td>height of immersion</td>
<td>( m )</td>
</tr>
<tr>
<td>H</td>
<td>height cylinder</td>
<td>( m )</td>
</tr>
<tr>
<td>M</td>
<td>moment on the cylinder</td>
<td>( Nmm)</td>
</tr>
<tr>
<td>My</td>
<td>moment during yield</td>
<td>( Nmm)</td>
</tr>
<tr>
<td>My,residual</td>
<td>moment remaining after halt</td>
<td>( Nmm)</td>
</tr>
<tr>
<td>Mcyl</td>
<td>moment on the wall</td>
<td>( Nmm)</td>
</tr>
<tr>
<td>Mbot</td>
<td>Moment on the bottom</td>
<td>( Nmm)</td>
</tr>
<tr>
<td>R</td>
<td>radius cylinder</td>
<td>( m )</td>
</tr>
<tr>
<td>Vg</td>
<td>superficial gas velocity</td>
<td>(mm/s)</td>
</tr>
<tr>
<td>Ω</td>
<td>rotational speed</td>
<td>(rpm )</td>
</tr>
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<td>σ</td>
<td>compressive normal stress</td>
<td>( Pa )</td>
</tr>
<tr>
<td>S</td>
<td>total compressive stress</td>
<td>( Pa )</td>
</tr>
<tr>
<td>Θ</td>
<td>shear stress</td>
<td>( Pa )</td>
</tr>
<tr>
<td>Amf</td>
<td>Apparent minimum fluidising velocity</td>
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<tr>
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<tr>
<td>RBP</td>
<td>Random bubbling point</td>
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<tr>
<td>V(x)</td>
<td>gas velocity at point x</td>
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<tr>
<td>IPF</td>
<td>interparticle forces</td>
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8.8 References


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Appendix 8.A: a suggestion for a different mean particle diameter

The currently used mean particle size is optimized for the fluid drag across a packing:

$$Dp_1 = \frac{1}{n} \sum_{i=1}^{n} \frac{W(Dp)}{Dp}$$ …(8.A.1)

A more proper weighing for an estimate of the IPF involved could be based on the energy per volume of packing.

The energy of attraction in any particle is proportional to its size, the number of contacts and the size of the contacting particles:

$$E_{\text{adhesion}} \propto \sum_{m=1}^{k} \frac{Dp \cdot Dpm}{Dp + Dpm}$$ …(8.A.2)

The coordination number will not be equal for the different sizes in a disperse system. It is likely to be larger for the relatively larger particles. This can not be accounted for in terms of the absolute sizes. This uncertainty is somewhat compensated by the mean size of the contacting particles, which will be smaller for larger particles. A crude approximation of [8.A.2] will therefore be:

$$E_{\text{adhesion}} \propto Dp$$ …(8.A.3)

$$V \propto Dp^3/\varepsilon_s$$ …(8.A.4)

The volume of the system is proportional to the volume of the particle and the packing density. Assuming this latter to be nearly constant the attraction energy per volume is proportional to:

$$E/V \propto 1/Dp^2$$ …(8.A.5)

The expectation of the mean energy for a distribution of sizes would be:

$$\overline{E/V} = \int_{0}^{\infty} \frac{1}{Dp^2} \cdot W(Dp) \cdot dDp$$ …(8.A.6)

$$W(Dp) = \text{weight fraction of } Dp$$

Recalculating the equivalent size yields:
\[ D_\text{p} = \sqrt{\frac{1}{\left( \int_0^\infty W(D_\text{p})/D_\text{p}^2 \, d\, D_\text{p} \right)^2}} \quad \ldots(8.\text{A.7}) \]

or

\[ D_{\text{p}2} = \frac{1}{\sqrt{\sum_{i=1}^{n} \frac{W(D_\text{p})}{D_\text{p}^2}}} \quad \ldots(8.\text{A.8}) \]

which formula gives relative more weight to the contribution of the fractions of the smaller sizes. For any disperse distribution this formula will give a smaller mean size than \[8.\text{A.1}\].

It is expected that any refinement of the prediction of the IPF by particle mean size will not change the form of the boundaries of Geldart's plot much, only the clustering and the prediction of correlations will be better. No systematic investigation has yet been performed.
Chapter 9.: Conclusion.

9.1 Concluding remarks

In this thesis a number of topics related to the role of interparticle forces on the macroscopic behaviour of powders have been addressed. The main thought in this program is that powder behaviour can be explained by the taking into account particle size and shape and all possible interaction between the particles. This and the external macroscopic conditions forced onto a particle set determine all behaviour.

Because of the intrinsic variability in particle interactions the contactpoints display a wide range of separation forces. Taking these distributions into account a number of common powder tests have been evaluated mathematically and experimentally. One of the questions to be answered is whether it is permissible to use the tensile strength, as determined using a tensile tester, as an extrapolation of the yield locus and thus to obtain a more accurate estimate of the cohesion and the unconfined yield strength.

Experimental investigation of the processes occurring during a tensile test showed that crack formation and propagation is the principal mechanism. Due to the stress concentration in the region of the crack tip, the maximum force recorded at the critical crack length is much lower than that expected for a total, instantaneous breakage of all the interparticle contacts in a plane. The well established crack theory from materials science was tested against the results and later adapted. The quantitative description derived by using this modified crack theory and a computer simulation agreed well with the observed facts.

The processes occurring in a shear zone have been modelled by assuming a variable density within a small region. This causes a friction force when the zone expands against the normal compressive force. Three regions can be distinguished in a yield locus. They are a low region, where the normal force is small and thus the additional friction low, a high region where the normal force is high but the expansion low, and a intermediate region where expansion may be a dominant factor. The extrapolation of the middle region of the yield locus to the axis may, therefore, be questionable if it is not possible to account for the influence of the expansion. The fact that in reality a concave yield locus is measured in the Jenike method may be due to an other mechanism not yet included. Due to the inhomogeneous stress distribution, shear may start at the edges and gradually progress towards the centre.
Comparing the model for shear zones and the model for crack propagation it can be concluded that they have not much in common and that the only relationship between the two is that a high mean interparticle force results in both a high tensile strength and a high value of cohesion. There is no theoretical justification for the extrapolation of the yield locus to the tensile strength.

A machine was built to directly measure the response of a powder on an overall stress condition where one of the principal stresses is tensile and the other compressive. This instrument has the potential to clarify the failure modes of powders in that region and determine whether shear and tensile behaviour occurs. Measurements of the deformation of a powder in the hollow Dutch cell show that crack formation can be seen to occur below a certain critical negative principal stress. Until that limit is reached the yield locus may be defined, below that another process predominates.

Finally, the author feels that with the instrumentation and techniques available three-dimensional computer simulations of particle sets are feasible within a limited number of years. In a number of chapters the basic tools necessary for such an approach have been touched. Image analysis combined with powerful computing facilities will enable us to simulate packings. Powder shear and tensile behaviour both in compacted and aerated conditions can then be examined and assumptions on how to influence and steer the properties to obtain optimum results can be verified.

9.2 Summary of conclusions on the topics addressed.

To summarize, the following main conclusions have been made in the previous chapters.

A) On the Characterisation

Particles can be characterized in two ways: either by using the interaction of the particle with its surroundings or by describing its morphology. The second method is more fundamental.

An algorithm has been developed to determine the basic, polygonal form of a particle. This basic form reduces the amount of data necessary to describe the morphology to the level of relevant features. Using this polygonic description a computer simulation of the displacement of the particles in real powder should be possible.
The alternative spherical harmonics approach is limited in its usefulness to describe local features and the combination of basic form and Fast Fourier Transforms will prove to be more fruitful.

For intrinsically curved particles, the description with a polygon might be modified by a secondary fit using a function for the expected curvature.

B) Distribution of the normal separation force

The use of an image analyser enables one to sift very narrow size classes without going through extensive sizing methods. Furthermore, deviating particles can be rejected during the analysis.

For all the conditions, use of both the log-normal and Gamma distributions proved to be adequate and can be used for further modelling.

C) Relationship of macroscopic and microscopic parameters for multi particle systems

In multi particle systems sliding, rolling and normal separation all occur to a certain degree. This ratio has not yet been experimentally determined and perhaps only simulation of the displacements using particle sets will give more quantitative information.

A model of the relationship between the forces on the contacts and the macroscopic pressures has been discussed. From this it can be concluded that given the overall pressure, the distribution of interparticle forces is not uniquely defined. Only if an additional condition, as for example the yield of all the contacts, is met, is the distribution completely determinant. On the other hand, for each distribution it is possible to calculate a unique macroscopic stress.

Using these distributions it can be shown that the maximum axial force a system can sustain is smaller than the sum of all the interparticle forces. The theoretical breakage strength is, therefore, always higher than the value actually found during a measurement.

A phenomenon which likely reduce the maximum force even further is the occurrence of stress concentration at holes and cracks.

Experimental work on stress conditions in which one principal stress is negative and the other positive showed a change in mechanism. Up to a certain negative stress homogenous deformation is possible, at stresses more negative than that limit the powder fails by cracking. This finding requires further research into the mechanisms which lead to the formation of cracks.

D) Crack formation and propagation in consolidated systems.
The experiments give evidence of a crack propagation mechanism which controls the force recorded in a tensile test. The stress concentration at the tip of a crack reduces substantially the theoretical maximum force. For the limestone samples, the crack extends through half the cake before it become unstable.

A computer simulation showed that the stress field in a powder is different from that in homogeneous materials. By adapting the formulae for crack forces it is possible to describe the observed facts. Further research on the exact form of the stress increase due to the limited size of the cake will enable correlation of the force measured in one instrument with that in one of a different geometry.

E) Cohesion in shear zones

The work on the formation of shear zones in a coaxial shear tester showed a narrowing of the total deformation from a initial broad zone into a small shear zone. This zone remained stationary over a considerable deformation length and it can be concluded that these stable zones are suitable for a characterisation. The force-displacement curves recorded in the coaxial cell were different from those measured in other geometries, the reason for this has not been completely clarified.

The shear zone formed in the Jenike cell is less defined and rather susceptible to changes in the procedure. This leads, in turn to variations in the estimate of the yield loci. The yield loci are thus properties of the geometry of the shear zone tested. For a reliable test the shear zone must be reproducible and have a low variability.

Analysis of the force-displacement curves using a computer algorithm supplies an objective decision on the acceptability of the curves. This algorithm does not perform worse than the operators in expert laboratories. Using this algorithm as a decision criterion, different procedures were tested and the variability of the resulting yield points compared. A combination of a very careful filling procedure and the compression method advocated by Haaker resulted in the test of lowest variability.

The model of the force-displacement curve shows that the maximum of the curve is measured during a dynamical change from one stationary state to the other. The dynamic expansion of the zone against the normal compressive forces contributes largely to the maximum force.

Three regions could be distinguished: a low, intermediate and
high load. The contribution of the expansion is only appreciable in the intermediate region. The contribution is low in the other regions, because in the first region the normal load is low and in the second the expansion is limited.

A rather complicated curve for the yield locus results and an extrapolation of the values of the intermediate regime to the low regime in order to predict the cohesion is difficult if the correct model for the curve is not available.

In this derivation of a model, the assumption has been made that the entire shear zone deforms uniformly. For regions of low normal load deviations may cause the force to be less than expected.

This fact further complicates the curve.

F) Cohesion in Aerated systems

The work on aerated systems shows the gas flow to be the dominant variable for all processes. A structure is stabilized by the interparticle forces, both attractive and frictional, and destabilised by either the drag forces or the gravity forces.

It can be shown that the measurement of the resistance to flow, using a rotating powder viscometer, is a most useful instrument for the characterization of an aerated powder.

The local bubbling point and the random bubbling point can be defined and related to the structure of the bed. The fully supported velocity is seen to be a more appropriate parameter to describe fluidisation than the usual minimum fluidising velocity.

By assuming a proportional dependence of compressive stress on the depth of immersion and assuming also a Coulomb Solid friction behaviour it is possible to divide the total torque into a stress distribution on the wall and bottom of the cylinder.

The internal consistency of the parameters obtained supports the assumptions made.

Generalizing it can be concluded that aerated powders of type B behave like compacted powders, the friction dependent on the normal stress and thus influencing the fluidisation behaviour of the powder.