Porosity: a parameter with no direction

B. Scarlett, M. van der Kraan and R. J. M. Janssen

Phil. Trans. R. Soc. Lond. A 1998 **356**, 2623-2648
doi: 10.1098/rsta.1998.0290

Porosity: a parameter with no direction

By B. Scarlett, M. van der Kraan and R. J. M. Janssen

Department of Chemical Process Technology, Faculty of Chemical Technology and Materials Science, Delft University of Technology, Julianalaan 136, NL 2628BL Delft, The Netherlands

The macroscopic properties of a granular material are controlled and manipulated by controlling its microscopic composition, i.e. the distribution of particle size and shape. The particles then combine to form an array which is usually characterized by the parameter of porosity, the fractional volume of the interstitial pore space. This parameter is a scalar variable which cannot describe important features of the array, such as anisotropy. The importance of the anisotropy is illustrated by the shear behaviour of a powder. Measurements made in a biaxial shear cell are reported. A tensor quantity that describes the array is proposed as well as the techniques for relating this parameter to the particle properties.

Keywords: powder flow; critical state; biaxial cell; yield strength, unconfined; yield locus; particle packing

1. Granular materials

Particulate systems come in many forms and may be called powders, pastes, slurries, emulsions, aerosols or sprays. They all have the common feature that they consist of a disperse and a continuous phase and that the behaviour of the whole system is dependent upon the distribution of size and shape of the disperse phase. The behaviour is also determined by the governing physical and chemical laws, which are also dependent on the particle size regime (figure 1). Particles which are a few micrometres in size are subject largely to surface forces and phenomena. This is the regime of colloids and aerosols. Some recent technologies are based upon particles which are only a few nanometres in size, nanotechnology. In this regime, the structure of the molecules becomes the dominant factor.

The behaviour of particles which are larger than ten micrometres in size, is largely controlled by mechanical forces. The appropriate laws are those of continuum mechanics and the challenge is the complexity of the geometry and of the boundary conditions. Sometimes, collections of particles less than 1 mm in size are called powders, and those greater are called granular materials. This distinction is descriptive rather than scientific, particularly since the most interesting systems contain a range of particle sizes. When the mechanical forces dominate, then the distribution of size and shape and the disposition of the disperse phase are the key variables by which the properties of the bulk material can be manipulated.

Thus, a study of granular materials or powders demands a stereoscopic view, the macroscopic and the microscopic. On the one hand, the eventual interest is not in the behaviour of one particle but in the average behaviour of the total bulk. On the other hand, the macroscopic properties of the bulk cannot be interpreted, and certainly not manipulated, unless they can be related to the properties of the particles

Phil. Trans. R. Soc. Lond. A (1998) **356**, 2623–2648
Printed in Great Britain

TEX Paper

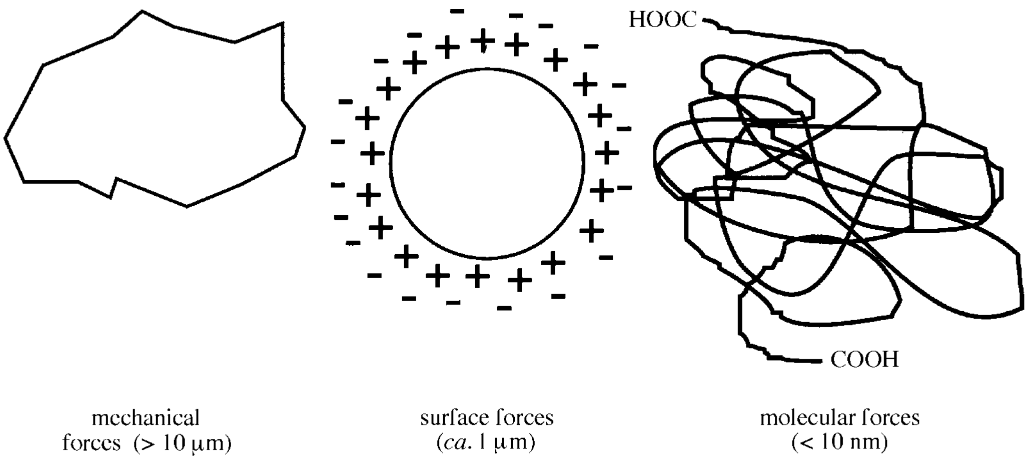

Figure 1. Particle interactions.

which make up the bulk. In this task, we may rightfully plead a special degree of difficulty. In relating molecular properties to those of a gas or a solid, it can be assumed that each molecule is the same. In relating particle to bulk properties, this simplification precludes some of the most important phenomena and certainly excludes the best techniques for adjusting bulk properties. In principle, every particle can have a different size and shape, and, in principle, they may be arrayed in an infinite number of ways. Nevertheless, experiments teach us that the eventual bulk behaviour is usually reproducible, at least to a degree. Thus the relationship between the microscopic and macroscopic properties is not unique; a different array leads to a different relationship. In practice, the particles take up particular arrays which are dependent upon the process conditions and history, and these arrays are themselves dictated by the distribution of particle size and shape. To understand why and how this occurs is to understand the behaviour of granular materials.

A simple example of an array parameter is the most compact state which can be achieved. Thus, the parameter 'tapped density' is frequently measured and used. The tapped density can usually be increased by including a wide range of particle sizes. If a fluid is then passed through the particle array, it exhibits a macroscopic parameter, the permeability to fluid flow. Of course, a smaller porosity implies a smaller permeability, but the sizes of the particles have an independent influence too. Large particles allow a much greater permeability than small particles when packed to the same porosity. The effect of the particles and the effect of the array combine to control the permeability, which is described by the well-known Kozeny–Carman equation. The systematic identification of the state as a separate variable is vital and is illustrated in figure 2. The distribution of particle size and shape controls the porosity, and both combine to determine macroscopic parameters such as conductivity, permeability, fluidizing velocity and flow properties. These macroscopic properties then combine further to control the behaviour in a piece of equipment. A simple correlation between particle-size distribution and the behaviour in some process usually cannot succeed, a model must separate array, bulk properties and then the compounded behaviour.

In this scheme, the first instinct is to characterize the particle array as a concen-

microscopic	state	macroscopic	process
particle size and shape	*porosity*	*fluidizing velocity*	*fluidized bed*
	anisotropy	*conductivity*	
	segregation	*permeability*	*filter*
particle strength	slip planes	*friction angle*	
	chains	*yield strength*	*silo*

Figure 2. Particle interactions.

tration or porosity, i.e. fractional solids or fractional voids. It must be appreciated, however, that the parameter of porosity does not describe all of the important features of the particle arrays which form during a process. Many particle arrays exhibit anisotropy and this explains, for example, the wide variation in permeability which can be caused in a filtration process. Another common phenomenon is segregation. If the distribution of size and shape is not well chosen, then the particles tend to separate during a mixing process, or, alternatively, to mix during a separation process. Thus distinct regions form that have a locally different composition. Usually, these irritating phenomena such as anisotropy and inhomogeneity are combated by the development of more and more ingenious machines. In reality, it is better to recognize that such structures are inevitable, that they must be anticipated and be included in the modelling system. These and other state parameters must be added to the state column in figure 2. There are other factors which must be added to the primary properties, for the same reason. A single particle may not be completely homogeneous, and therefore its size and shape do not completely define its behaviour. As an example, if a particle contains flaws and imperfections, its mechanical strength will be much reduced below that which might be expected from its outline.

2. Particle models

A relationship between macroscopic and microscopic properties requires some model which may range from the empirical to the direct single-particle simulation, but which must include a description of the particle array. Particle models are most successful for dilute systems. In this case, the behaviour of a single particle can be described and it can be assumed that there is no interaction between the particles. Each particle interacts with the surrounding fluid independently, and the total behaviour can be deduced simply by integrating over the range of particle properties and fluid velocities within the system. These assumptions are applicable and successful, for example, to many gas-particle systems where the concentrations are less than 10^{-3} v/v (figure 3).

At higher concentrations, the particles are still not in continuous contact, but their interaction with the fluid is strongly influenced by the presence of the other particles. They may also collide, and these binary interactions also contribute to the resulting behaviour and thus to the appropriate models. At even higher concentrations each particle is in contact with several near neighbours. Although the point contacts may be very small, it is possible for a continuous path to exist through the disperse phase.

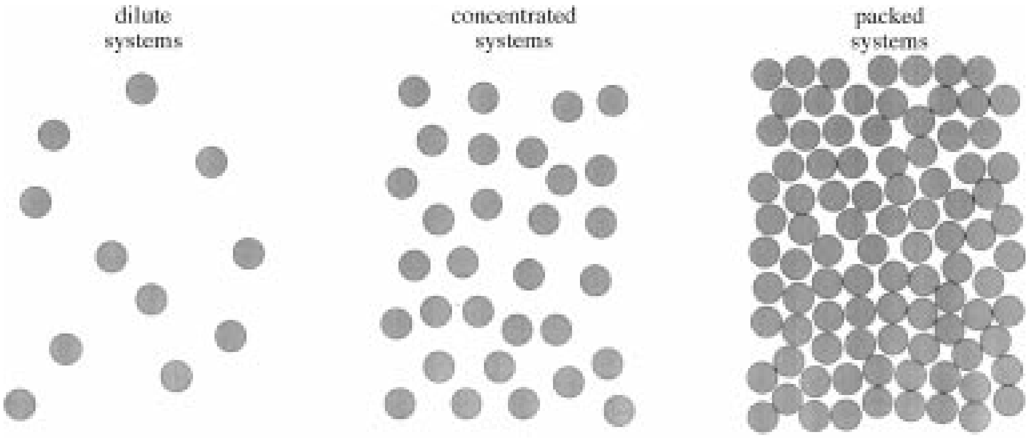

Figure 3. Particle interactions.

In this state, very complex structures can exist and each particle interacts with many neighbours.

It is clear that the modelling becomes more difficult as the average concentration increases. At low concentrations it is almost impossible to disperse the particles randomly, but the variations can be described simply as gradients in the concentration. At intermediate concentrations there is a strong tendency for the particles to segregate, and, thus, regions must be distinguished not only by their concentration but also by the particle sizes present. There is usually some anisotropy of the particle spacing and maybe also of the particle orientation. However, it is in the packed state that the limitations of porosity as a state parameter really become apparent. Properties such as an effective conductivity or permeability are frequently anisotropic, and, thus, the array of the particles must also be anisotropic. Furthermore, it may be that the structure is not characterized by a single anisotropic parameter. Consider the structures shown in figure 4.

The anisotropy of the point-contact structure may well have different axes than that of the pores. A bulk property, such as an effective conductivity or permeability, arises, of course, from the averaging of one or more constitutive equations of transport in the complex geometry. If fluid is flowing through the pore space, then it is that structure which is important. On the other hand, electrical current flows through the solid and it is the array of the point contacts which is the dominant structure. Most properties are dependent upon both of these structures that are, in turn, independently determined by the distribution of particle size and shape. This dependence must be uncoupled if the properties are to be successfully manipulated. The most complex behaviour occurs when the particles move and when each particle can slide and roll relative to its neighbours. Here the structure changes during the strain, and this change is also completely dependent upon the size distribution and shape of the particles. This paper is intended to contribute to the discussion of this problem: the behaviour of powders in shear flow.

3. Shear behaviour of powders

Even in this complex situation, the state of the particle array is usually described primarily as a porosity. The phenomenon of dilatancy was first described by Reynolds

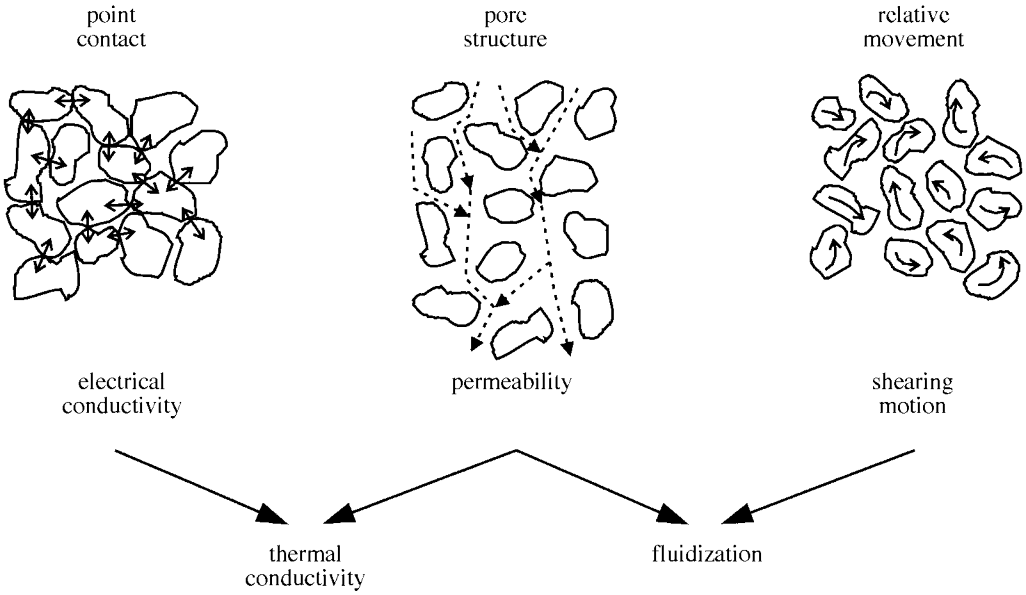

Figure 4. Packed particle structures.

(1885) and leads to the idea of a critical porosity, the porosity at which the material can deform in a steady state. Such a description is summarized in the Hvorslev (1937) diagram (see figure 5), and was the basis of the considerable advances made by Roscoe *et al*. (1958), who plotted the critical state line on this diagram. The axis remains as that of porosity, but it is almost certain that the critical state that is attained after dilation has occurred is not a uniform isotropic packing.

In the process industries, these concepts are commonly used in the design of equipment, primarily of storage silos. A cohesive material is considered to behave as a Coulomb solid, which exhibits a yield strength dependent upon its previous consolidation. The material is usually characterized by a procedure due to Jenike (1961, 1964). A family of yield loci is determined and extrapolated to determine the unconfined yield strength of the powder. This is the parameter that has the greatest influence on the silo design. The critical-state line projects onto the stress plane as the effective yield locus, which is the condition that is attained when the material flows in steady state (see figure 6).

The difference between this line and any yield locus represents the stress necessary to cause dilation, i.e. a change in the state of packing. Thus a free-flowing powder exhibits little dilation, a cohesive powder considerable. In fact, most powders exhibit cohesion, and the yield loci shown in figure 6 pertain to a limestone powder which is certified and adopted as a European, BCR, reference material for shear testing (Community Bureau of Reference). These yield loci are usually determined in a Jenike shear cell, as illustrated in figure 7.

The essential feature of the test is that it is a split cell which ensures that the failure of the solid will be in the form of a slip plane. The cell imposes a steady strain on the consolidated sample and records the stress pattern as the material dilates and fails. Sometimes, a different shear test may be used in which the cell containing the material is in the form of an annulus. This enables the total strain

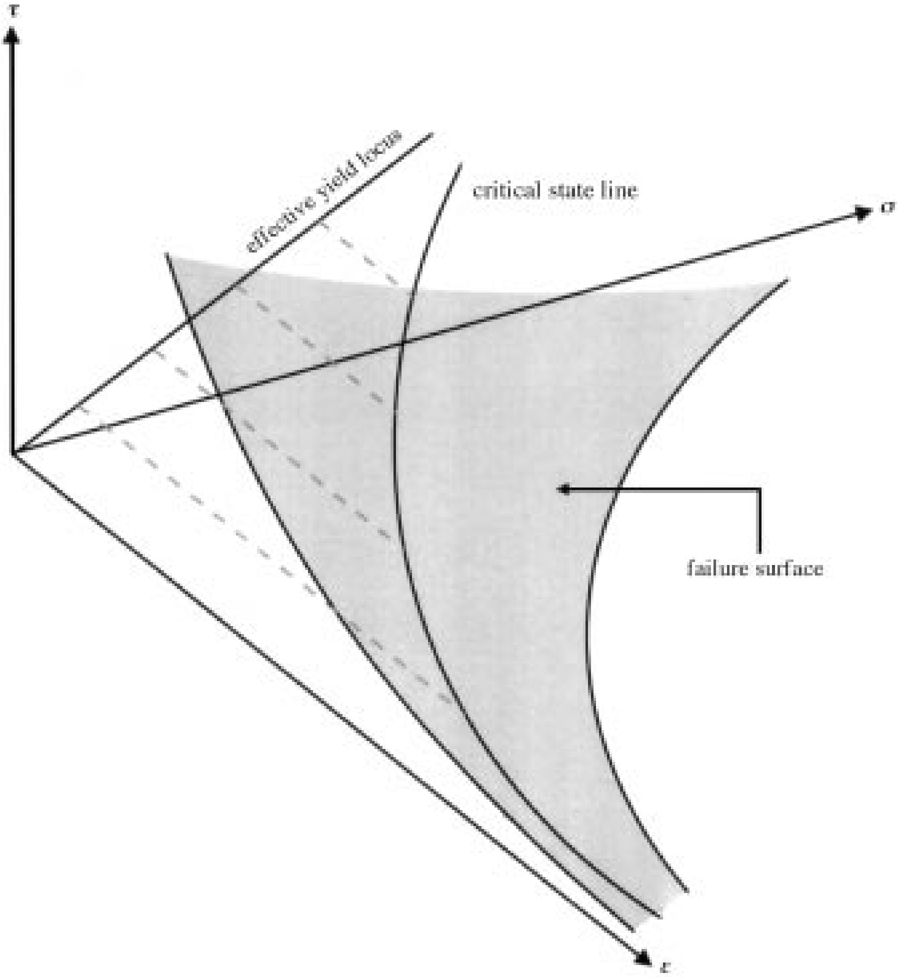

Figure 5. Hvorslev diagram.

to be indefinite. However, the basic geometry is still a split cell with an applied, steady strain and the failure of the material is in the form of a slip plane. Whatever the form of cell used, the basis of the test is to bring the material to the critical state at some level of stress, to reduce the stress and to bring the material again to the critical state. If there was no dependence of the critical state on the stress field, then all the yield loci would collapse onto the effective yield locus and the material would exhibit no cohesion. In this test, all three principal stresses are always positive, that is compressive, and thus the cohesion is best expressed as an unconfined yield strength. This is the value of the maximum principal stress which causes flow at a free surface and that is directed parallel to that surface. Such a state of stress is illustrated in figure 8.

Until this point, the failure mode of the material is by shear. In some circumstances, a stress may be exerted in which one of the principal stresses is negative, that is tensile. In this situation, the material may fail by tensile rupture. Cracks are formed

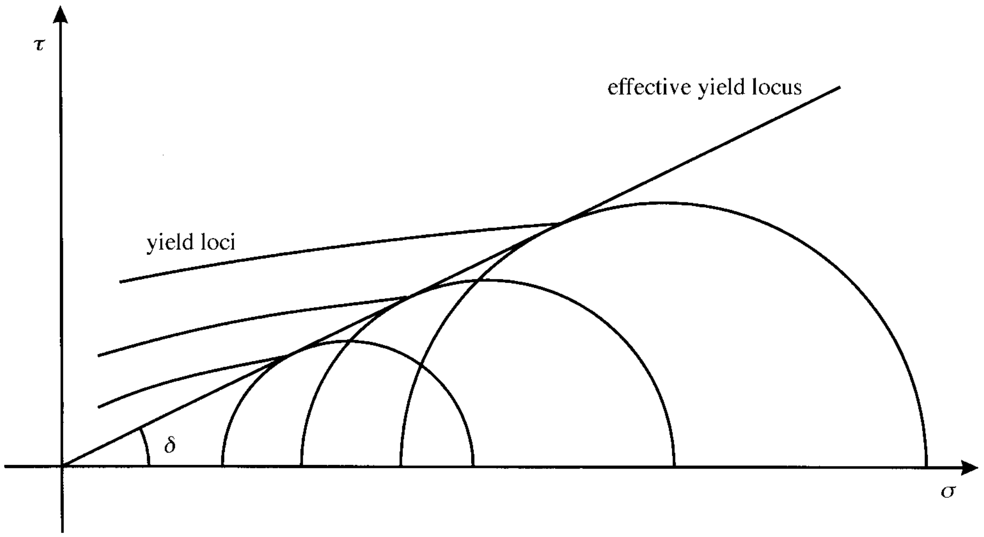

Figure 6. Yield loci.

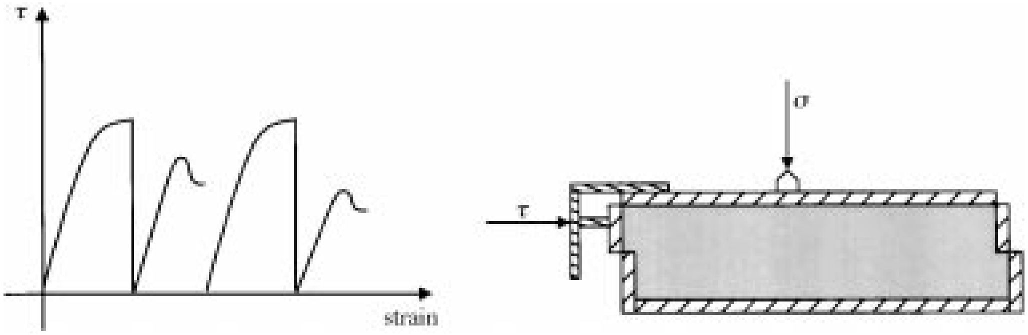

Figure 7. Jenike shear test.

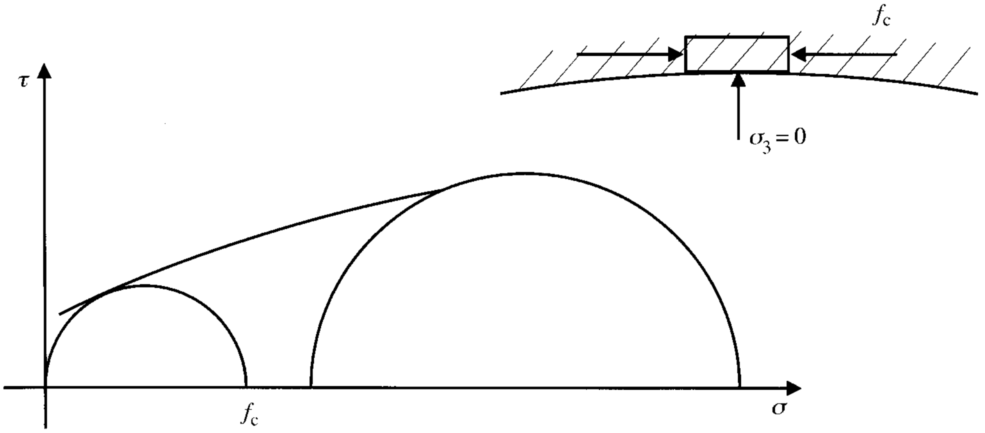

Figure 8. Unconfined yield strength.

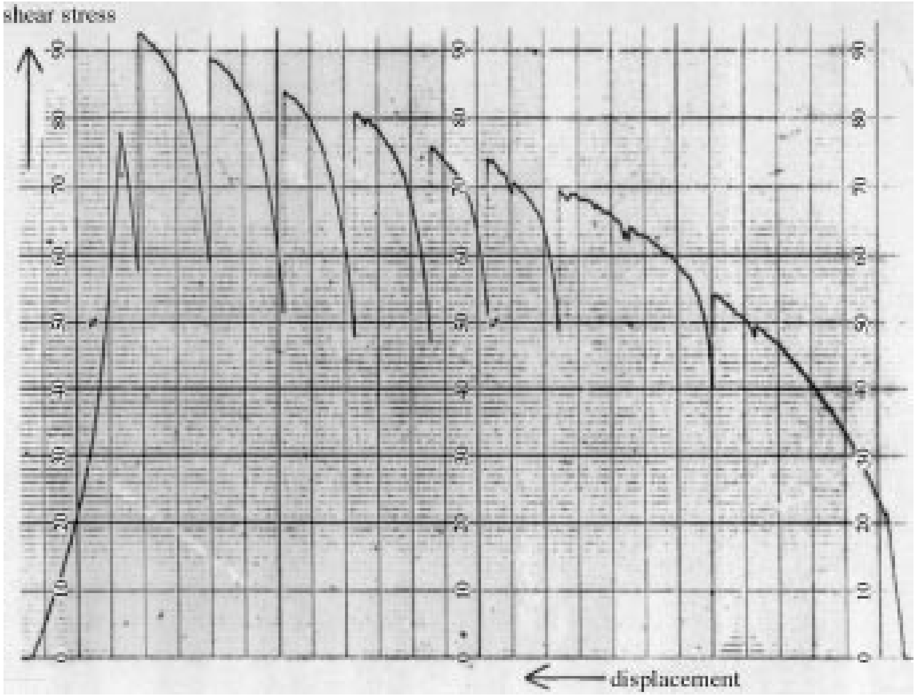

Figure 9. ‘Stick-slip’ behaviour in the Jenike cell

and propagate through the powder bed. Such failure is dependent upon the attractive forces between the particles. In shear failure, in contrast, the particles remain in contact. The attractive forces are small compared to those that are necessary to cause the particles to slide and roll relative to each other, thus those necessary to create and maintain the critical state.

Not all powder samples behave so ideally as the limestone sample when tested in the Jenike cell. Some powders exhibit distinct ‘stick-slip’ behaviour. This is illustrated in figure 9, which shows the stress–strain behaviour of a sample of starch in the Jenike cell.

It is not certain whether this is a characteristic of the Jenike cell or whether it is intrinsic to the particular powder. In this paper, we will suggest that it is, at least partly, a property of the powder. Such behaviour is usually accompanied by a considerable emission of sound and is probably due to particle deformation, particle breakage or both. In any case, it is clear that such powders never attain a critical state, it is not a state which automatically exists for any powder.

4. The biaxial cell

The limitations of the Jenike and similar testers are threefold. The stress measured is a single point, a value of shear stress and of normal stress. The values and directions of the principal stresses are not uniquely determined. The strain is predestined to occur in a narrow shear plane, and deviation from that mode of failure leads to a lower reproducibility in the measurements. The third limitation is that the measurements are always strain controlled. Thus, a predetermined strain is applied and the resulting

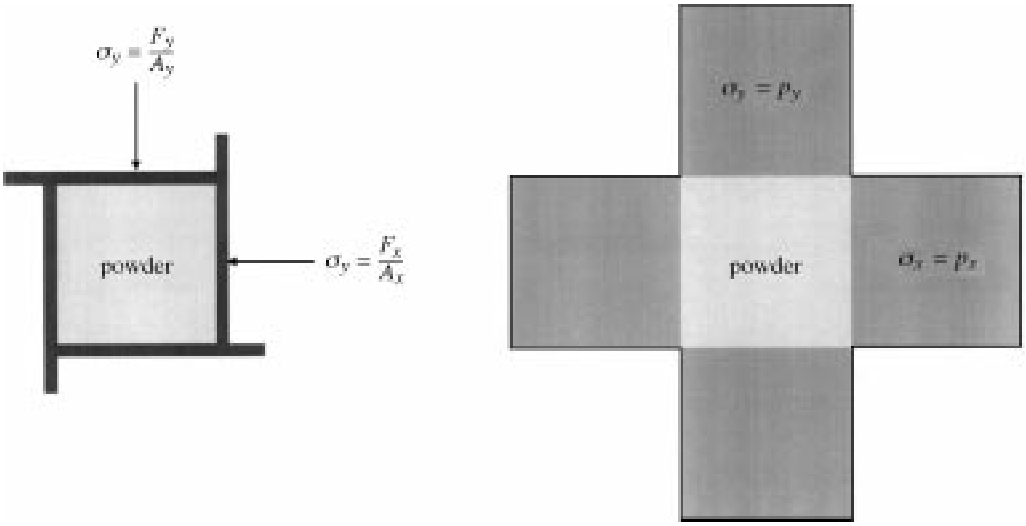

Figure 10. The biaxial principle.

stress is recorded. There is no way to check that an application of the same stress would result in the same value of strain. All these limitations are overcome by the biaxial cell, in which a sample can be deformed simultaneously in two directions which are contrived to be the principal axes of stress. The principle is illustrated in figure 10.

Experimental versions of the biaxial cell have been used for some time in various laboratories. A notable series of measurements were made by Schwedes & Schulze (1990) on a limestone powder using such a cell. These experiments clearly demonstrated that the yield strength of a compacted powder depends on the direction of the principal axes during compaction, relative to those during the subsequent shear. The important fact, which was established by these workers, is that the critical state exhibits considerable anisotropy, and that the response of a powder to subsequent stresses depends considerably upon the relative orientation of the applied stresses and the existing anisotropy.

The cell used by Schwedes & Schulze (1990) has solid walls and the principal stresses are measured by transducers. The cell is operated in a strain controlled mode. An alternative design of biaxial cell was developed by Arthur *et al.* (1977), who applied the maximum and minimum principal stresses through pressurized membranes. This design was further developed by Maltby *et al.* (1995) and has the advantage that the stress can thus be directly controlled, not simply measured. There is no doubt that the biaxial cell offers a considerable improvement in the measurement of powder failure. The hard-wall and soft-wall versions have relative advantages and disadvantages but the soft-wall version offers great flexibility. Such an apparatus was built and used in this work (van der Kraan 1996). The design of the tester is shown in figure 11.

The new feature that has been added is that of feedback control. The deformation of each membrane is detected by an optic fibre, and the pressure is constantly adjusted to maintain a flat profile of each membrane. The relative motion between the membranes is created by stepping motors. Thus, in principle, the test can be programmed to be either stress controlled or strain controlled. The reaction of a powder

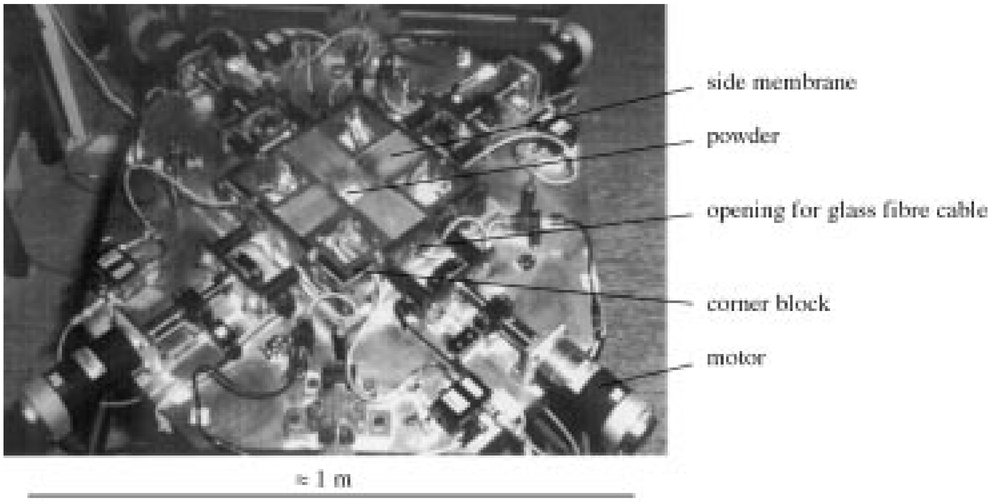

Figure 11. The biaxial cell.

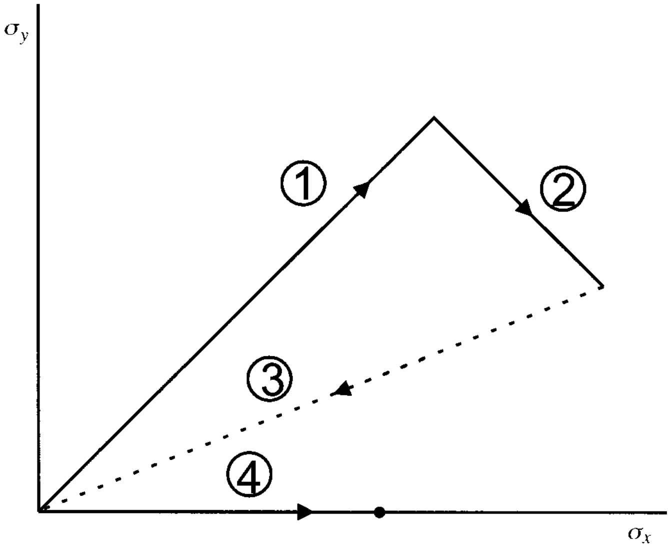

Figure 12. Stress trajectory to determine unconfined yield strength.

to the two modes is different and is instructive. Between these two criteria, and as a more practical test, the cell could be programmed to subject the powder to any anticipated stress–strain path which might be experienced in a particular process.

This shear cell has been used to test the standard BCR-limestone powder. It was first used to determine the unconfined yield strength as a function of the consolidation pressure, that is, the flow function of the powder. The advantage of the cell is that it can be determined directly, no extrapolation is involved. The test was carried out in the stress-controlled mode by the trajectory illustrated in figure 12.

The powder was first compacted biaxially (path 1, figure 12) and was then subjected to a shear stress (path 2), until steady-state deformation was achieved at

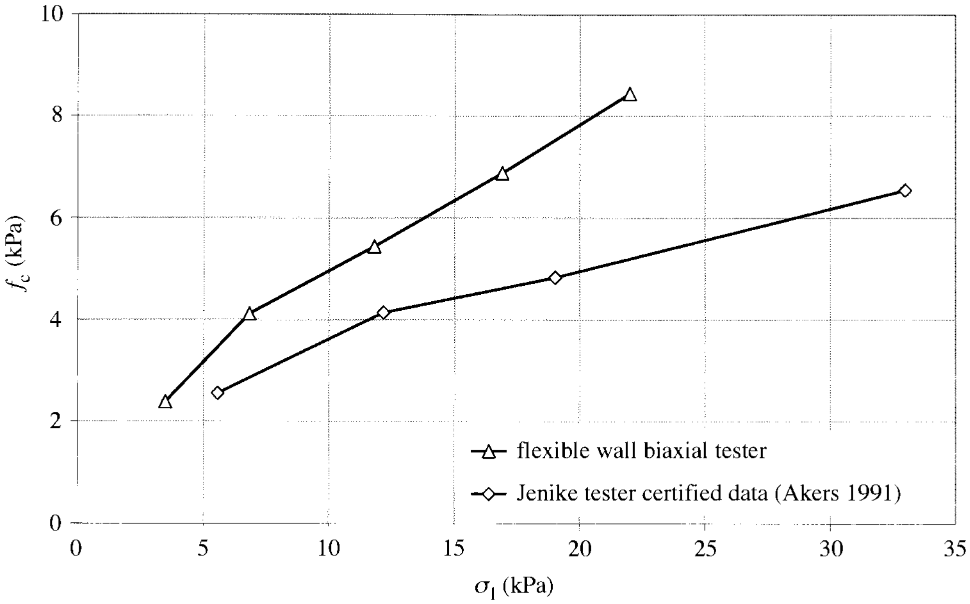

Figure 13. Unconfined yield strength; biaxial cell and reference material.

point A. The stress was then reduced to zero and the powder again subjected to a stress (path 4), in which one principal stress is zero. At point B, the powder again exhibited steady-state shear. Thus, points A and B represent one point lying on the flow function. By repeating the procedure for different compactions, the full flow function was determined. In the trajectory illustrated, the direction of the maximum principal stress was the same during both the pre-shear and the failure. Thus, this condition imitates the procedure that is assumed to occur during the Jenike test, and was expected to yield the largest values of unconfined yield strength. The results of this test are shown in figure 13, where they are compared with the certified results which were obtained with the Jenike tester. The values are, indeed, appreciably larger, and there is every confidence that the differences are real. In the biaxial cell, it was possible to control the direction of the principal stresses during the whole test, whereas this is much more indefinite in the Jenike tester.

A similar stress-controlled path can be used to determine the yield loci of the powder, as illustrated in figure 14. These measurements yield a family of Mohr circles, and the yield locus can be constructed to be tangential to them as shown in figure 15*a*.This curve is compared with the certified results in figure 15*b*. Again, it is seen to indicate appreciably larger yield strengths.

Furthermore, it is common practice to consider a powder to be a Coulomb solid and to fit a yield locus as a straight line represented by the equation:

$$\tau = \sigma \tan \delta + C.$$

This relationship has been assumed for the certified results but it is clear from figure 15*a*, that for this powder, the yield locus is not linear. However, if the results are replotted as normalized deviatoric stress, σ_r/σ_m, against mean stress, σ_m, the yield locus is then almost linear, as shown in figure 16.

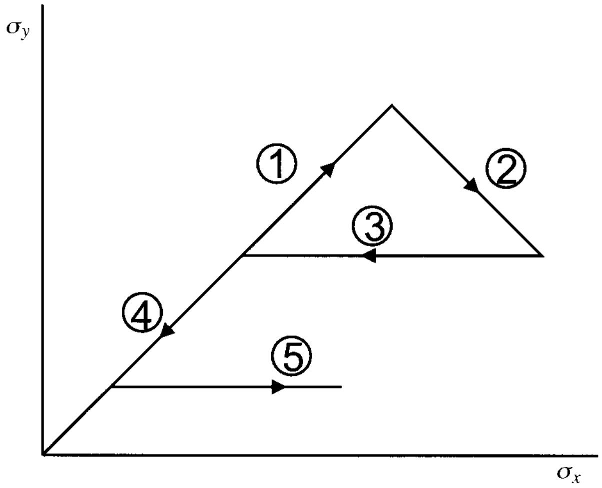

Figure 14. Stress trajectory to determine yield loci.

Figure 16 implies that, for this powder, the yield locus can be represented by the equation:

$$\sigma_{\mathrm{r}} = A\sigma_{\mathrm{m}} + B\sigma_{\mathrm{m}}^2,$$

where

$$\sigma_{\mathrm{r}} = \tfrac{1}{2}(\sigma_1 - \sigma_3), \qquad \sigma_{\mathrm{m}} = \tfrac{1}{2}(\sigma_1 + \sigma_3).$$

We presume that for some powders it may be necessary to take the third or higher powers of the mean stress into account. It should be mentioned that, in these tests, the stress in the third direction was always recorded, and always lay between the other two. Thus, there is every reason to believe that the two controlled stresses are truly the maximum and minimum principal stresses.

The cell can also be programmed to operate in a strain-controlled mode. The strain path illustrated in figure 17 represents a biaxial compaction, followed by shear at constant volume. The limestone powder was tested in this mode at two different strain rates, namely $2.5\times10^{-6}\ \mathrm{s}^{-1}$ and $5\times10^{-5}\ \mathrm{s}^{-1}$. The resulting stresses are shown in figure 18*a*, *b*.

It is clear that the deviatoric stress recorded is smaller at the higher strain rate than at the lower. It does seem that the response time of a powder may be very slow, and that the critical state is attained only after an appreciable time. This is not really surprising considering the billions of particles which are reacting to each other. Thus, tests which are conducted at a strain rate which is too high, do not create the true critical state, the deformation occurs at some pseudo-state.

Although some powders never attain a critical state, they do exhibit the 'stick-slip' behaviour that was observed in the Jenike cell and was illustrated in figure 9. The same starch powder was tested in the biaxial cell. The results of a strain-controlled test are shown in figure 19*a*, *b*. The powder was first biaxially compacted (path 1) and then sheared at constant volume (path 2). The recorded values of the mean and deviatoric stress show clear and persistent fluctuations. During a 'slip', a clear emission of sound occurs and there is a sudden deformation of the membranes outwards, indicating a relaxation of the stress. It is a significant observation that this slip is

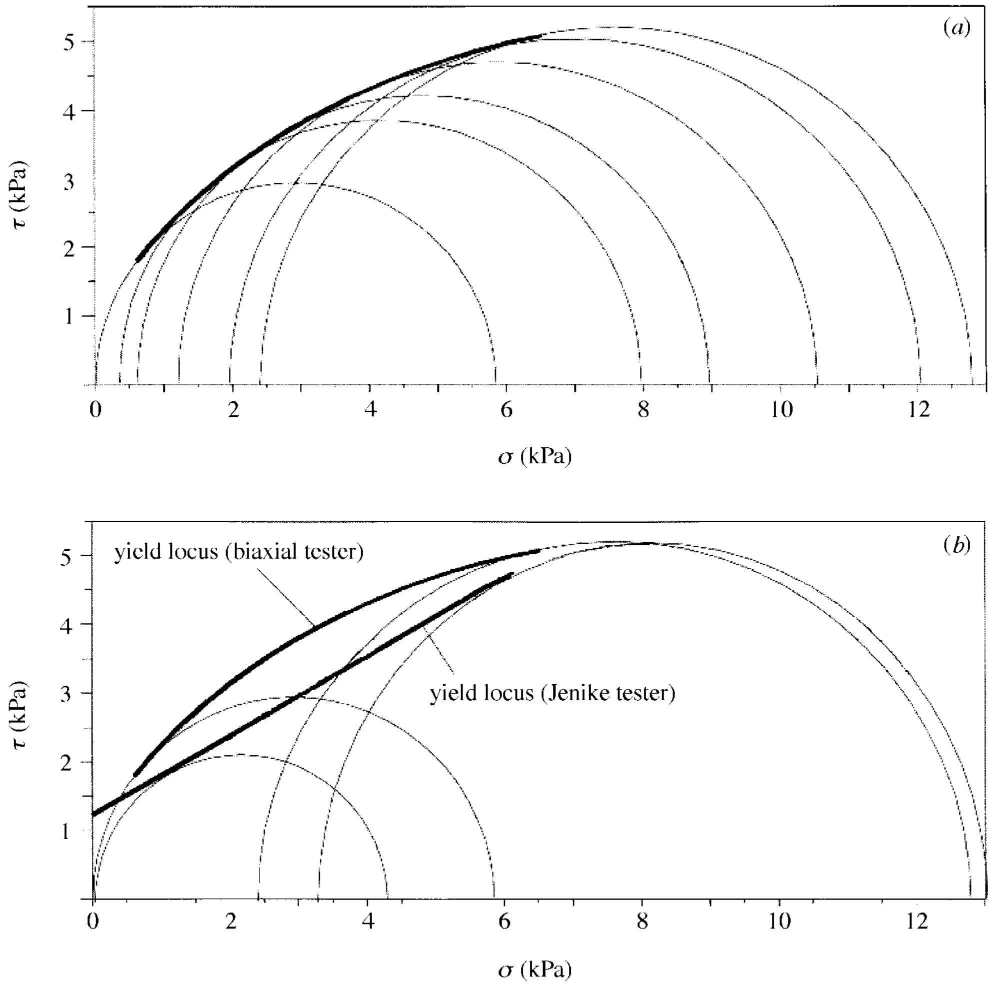

Figure 15. Yield locus; biaxial cell and reference material.

much greater in the direction of the maximum principal axis than that of the minor principal axis.

5. The critical state

It may be concluded that the biaxial cell is an excellent instrument for investigating the flow properties of powdered and granular materials. With further development, it can be programmed to apply any required stress or strain sequence to a sample and to observe the response of that sample. That in itself is a step change in the technology but it is only passive. The question remains, how can we adjust the size and shape of the particles in order to adjust the observed behaviour?

It would seem that the key to achieving that ability is to better understand the critical state. A difficult material must dilate to flow. Free-flowing materials dilate little, cohesive materials much. How does the critical state depend upon the particle properties? Some materials never achieve a critical state; they exhibit oscillating

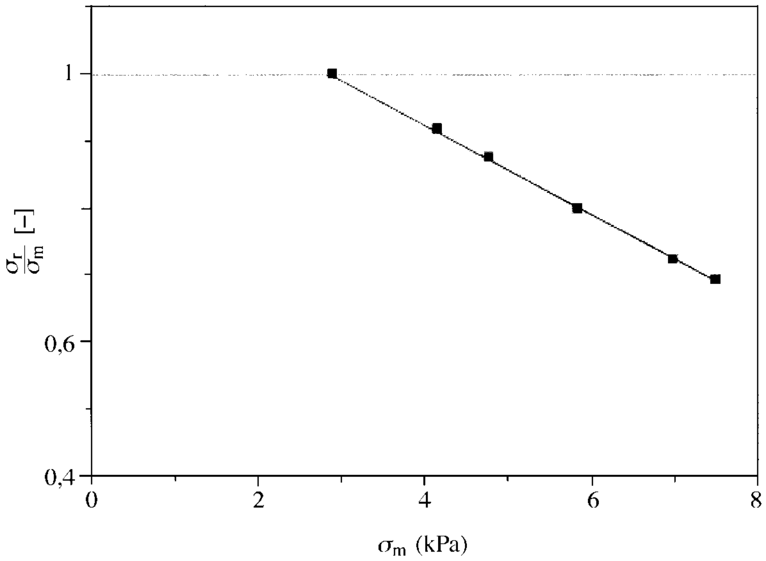

Figure 16. Deviatoric and mean stresses determined in the biaxial cell.

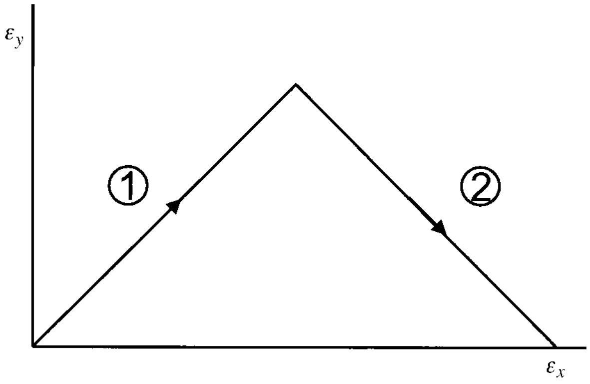

Figure 17. Strain trajectory.

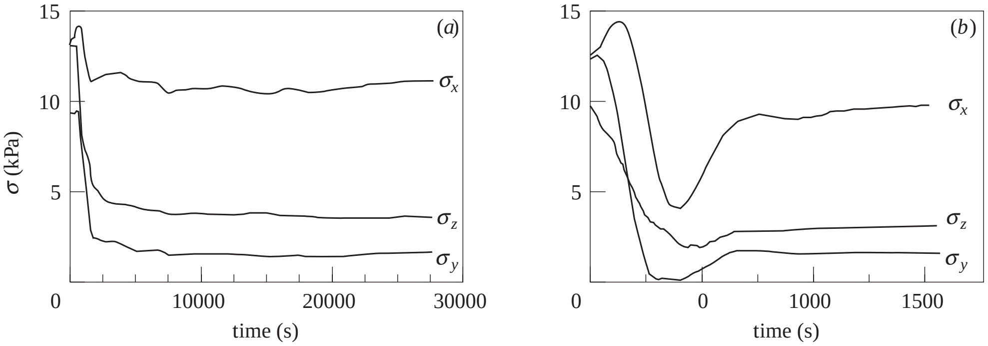

Figure 18. Stresses determined in the biaxial cell at two strain rates.

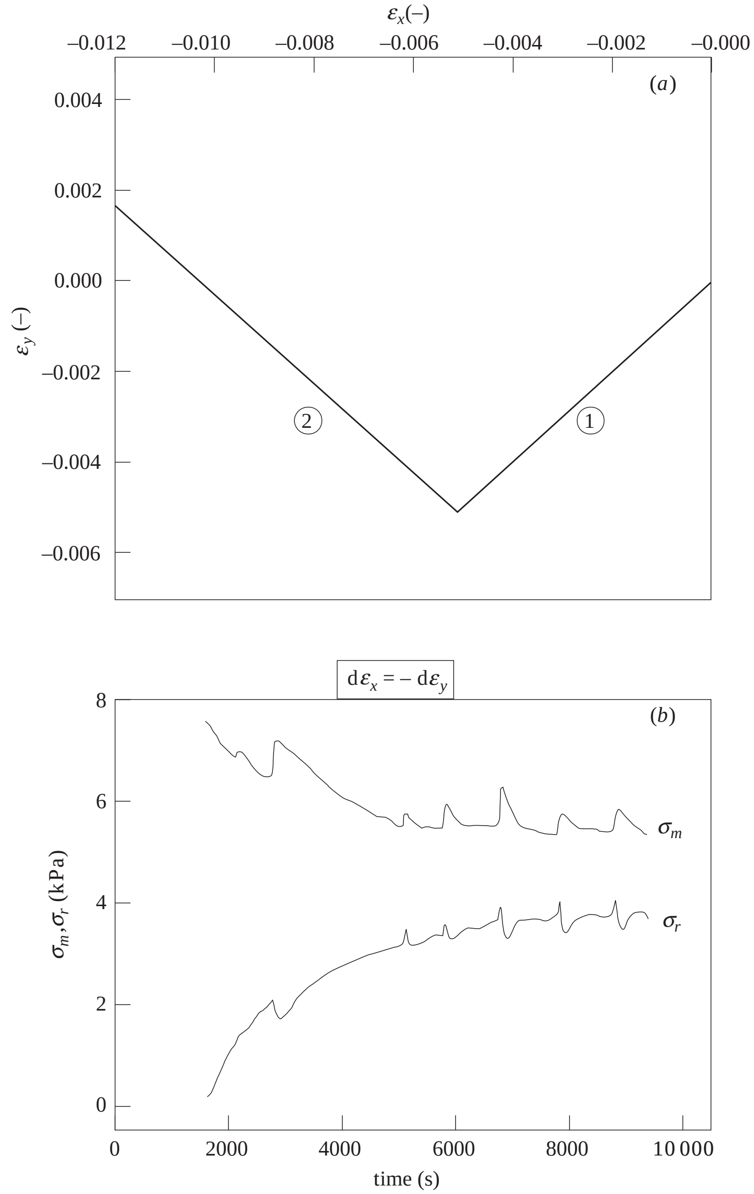

Figure 19. ‘Stick-slip’ behaviour in the biaxial cell.

behaviour. The critical state seems to exhibit anisotropy and thus cannot be completely represented by the scalar parameter of porosity. The need is to define the critical state, to understand the mechanisms which control it and thus to relate it to the primary particle properties.

Scarlett & Todd (1968) proposed a model for critical porosity. It was based upon the characterization of a set of particles by their random chord distribution. We will briefly reiterate the model here. The particles are assumed to be translating and rotating in a shear plane and must attain some minimum porosity, the critical

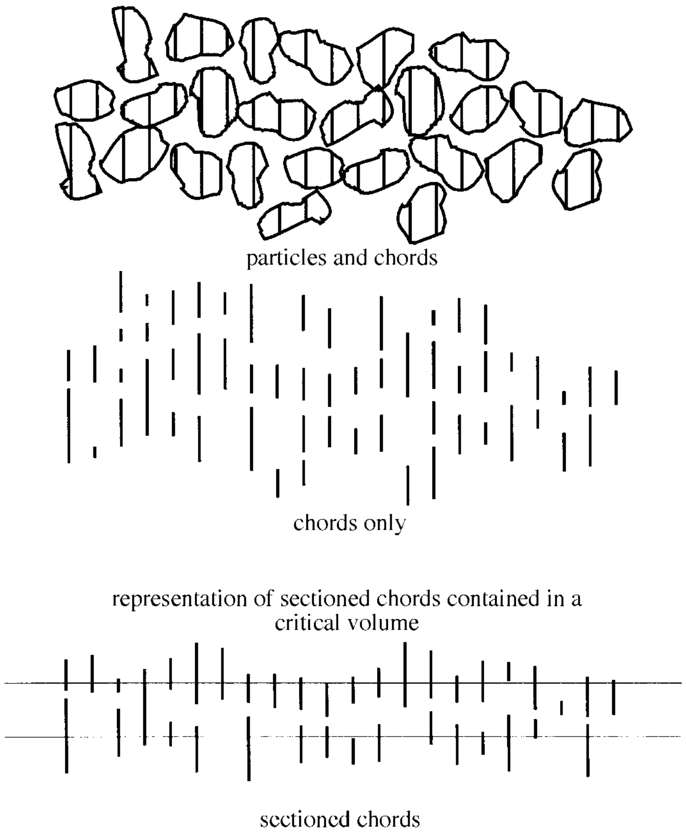

Figure 20. Chord representation of critical porosity.

porosity, in order to be able to maintain the shear motion. Consider now the motion not of the particles but of the random chords that can be drawn within the particles. Furthermore, consider only those chords which are lying instantaneously at right angles to the direction of shear, as illustrated in figure 20.

Some chords can easily pass each other. Others can only pass by moving at right angles to the direction of shear or by disappearing from the plane of view. The first motion represents a translation and was modelled by randomly sectioning each chord. Statistically then, the sectioned-chord distribution must remain constant in the plane of view. The second motion, disappearing from the plane of view, represents a rotation and implies that a long chord must be replaced by short chords of equivalent length. This is because the mass must be preserved. Thus, the postulate was that the random chords must be separated by a distance of at least twice that of the median length of the random-chord distribution. Of course, some distributions could easily attain that state, others not. The distribution shown in figure 21*a* would have to segregate and that in figure 21*b* would exhibit considerable anisotropy. The distribution shown in figure 21*c* would presumably represent a free-flowing material.

Of course, this model did not fulfil all the requirements, but we maintain that it is a basis which can be extended because it did contain the essence of the necessary model. That essence is to consider elements which are smaller than one particle

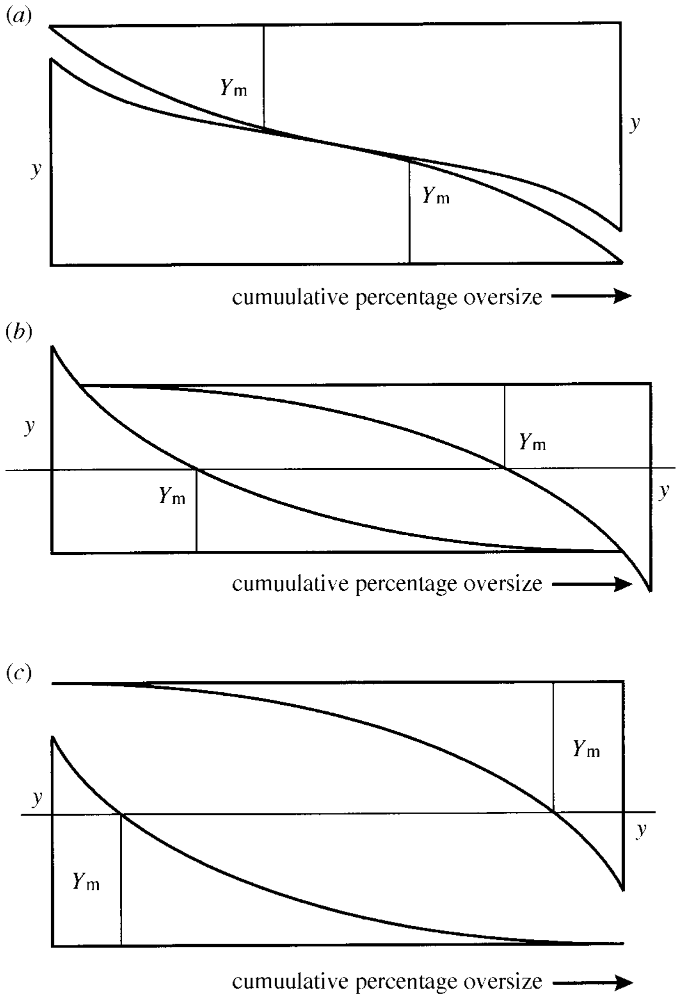

Figure 21. (*a*) Segregating, (*b*) anisotropic and (*c*) free-flowing powders.

and to consider that particles are bundles of smaller elements which have direction. In order to improve the model it is necessary to consider all the mechanisms that can contribute and all the situations which can arise. Consider first some of the mechanisms which can occur and which usually all occur simultaneously to a greater or lesser extent, as illustrated in figure 22.

The particles can exhibit solid-body motion, that is translation and rotation. However, they may also deform, and that deformation may be elastic or plastic. It is quite probable that plastic deformation of the particles is the origin of the 'stick-slip' behaviour. The particles may also break. The breakage may be surface attrition and abrasion or it may be massive fracture (van den Bergh & Scarlett 1991). By attrition and abrasion, the particles become rounder and smoother. At the same time, small particles are produced that act as a glidant between the larger particles. Fracture, on the other hand, leads to smaller but more angular particles. It is almost inevitable that attrition will occur when the particles are sharp, and that fracture will occur

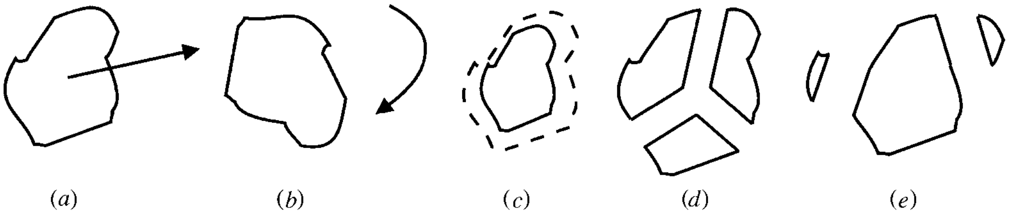

Figure 22. Particle strain: (*a*) translation; (*b*) rotation; (*c*) deformation; (*d*) fracture; (*e*) attrition.

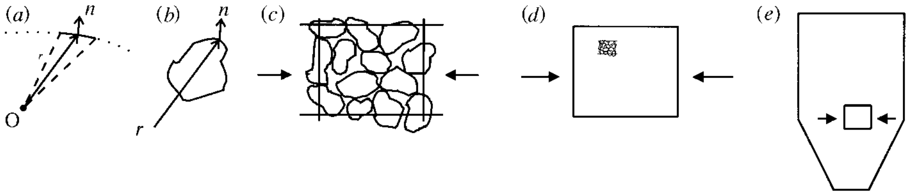

Figure 23. The scale of scrutiny: (*a*) surface point; (*b*) single particle; (*c*) many particles; (*d*) bulk solids; (*e*) equipment.

if the stresses become large. All these effects do occur and lead to a change of the critical state with time and with stress. However, the starting point is the solid-body motion. Translation and rotation of the particles are inevitable, and they are the motions which must be modelled first without the other complications. The limitation of the Scarlett & Todd (1968) model was that it concerned only a shear plane. The instrument that was used to test the model was an annular shear cell where an indefinite continuous shear plane could be maintained. It is clear that such a shear plane is a specially contrived situation. However, within a total system, this is not the only structural feature which may occur. Consider a bulk material to be flowing in a piece of process equipment, for example a silo, and consider the different length scales on which the problem must be examined (figure 23).

On the equipment scale, the flow in the silo is dominated by the effects of the gravitational force, which causes any element of material to interact with that which is supporting it (figure 23*e*). Thereafter, the interaction of the material with the walls of the silo is the key factor, and for small silos this becomes more important than the gravitational force. Such a piece of equipment can only be modelled by considering an element of material which is small compared to the equipment, but which is still large compared to the particle size (figure 23*d*). The variations of stress and of material properties occur then from element to element. It is samples of this size that must be characterized in the shear cell. It is also on this scale that the anisotropy must be formed, since the stresses are averaged on this scale. However, individual shear planes and other failures are constantly forming and ending at several places in such a sample. It must be so, or the equation of continuity would not be obeyed on the macroscopic scale. The best model of this averaged bulk behaviour is still the double-sliding model of Josselin de Jong (Mandl *et al.* 1977). The local motion occurs on a scale of a few tens or hundreds of particle diameters (figure 23*c*). On this scale, the problem is a many-body problem, one of local mechanics and particles rolling and sliding relative to each other. Of course, in come cases the equipment size is only of this order and then it is not realistic to ascribe to the material bulk properties.

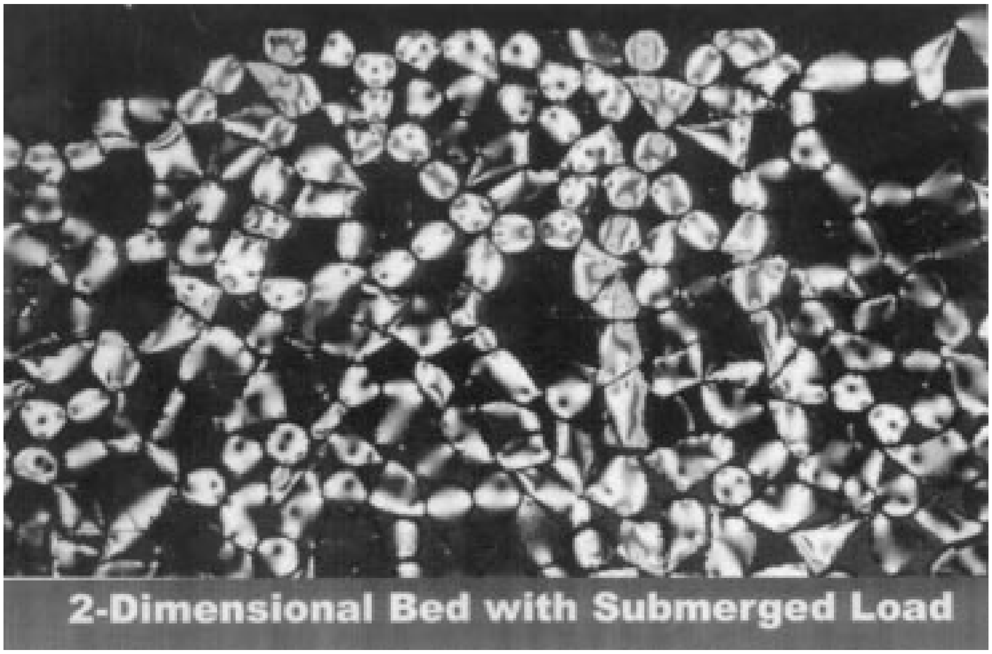

Figure 24. Stress chains in a two-dimensional bed (taken from Barber 1972).

Many people have demonstrated the peculiarity of the stress patterns when relatively few particles are involved, both experimentally and by single element simulation. Figure 24 represents a demonstration of the chains of stress which occur on the level of a few particles made by stressing photoelastic particles. Reducing further, the next scale of scrutiny is that of one particle (figure 23*b*). It is at this level that changes to the particle size and shape occur. The phenomena of deformation and breakage must be modelled at the level of one particle. However, this is not the minimum scale to be considered, since it is not possible to model on that level the dominant effect of the particle shape on the sliding and rolling behaviour.

There are many approaches to characterizing the size and shape of a sample of particles. The most common is to assume that each particle may be considered to be equivalent to a sphere. Such an approach may be instructive but cannot be successful when particle shape is a dominant parameter. More complex ideas include the description of the outline of the particle as a Fourier series (Beddow 1989) or as a fractal (Kaye 1997). The chord length which was used by Scarlett & Todd (1968) is a particularly useful one for isotropic arrays, because the pore space can also be characterized in the same manner. Whatever the simplicity or complexity of the characterizing parameter, the motivation is to attempt to relate or at least to correlate, it to the macroscopic parameters of the particle array. Sometimes this may be successful, but at other times the particle parameter is unsuitable or inadequate. The fact is that the relationship between the microscopic and macroscopic properties is rarely simple, and a single particle parameter cannot be universally the best choice. The characterization of the particles that is appropriate is unique to every problem. In turn, if the appropriate parameter can be defined, a method of measuring it can also be devised.

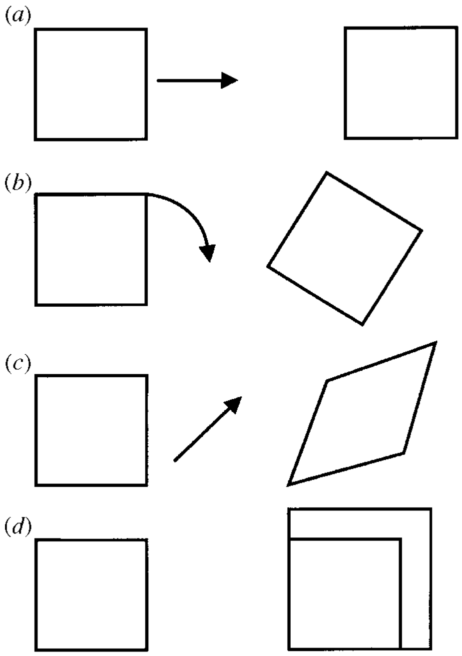

Figure 25. Macroscopic strain: (*a*) translation; (*b*) rotation; (*c*) shear; (*d*) dilation.

Consider the alternative approach. Assume that the position coordinates of every element of particle and every element of pore volume is known. It should be possible to logically integrate and average that information into the parameters which describe any particular problem. For example, the appropriate characterizing parameters should arise by the integration and averaging of the appropriate constitutive equations if the flow of fluid or energy through a static array of particles is under consideration. In this paper, there is no constitutive equation, the concept of 'critical state' is under consideration. The question to be answered is as follows. If a strain is applied to an array of the particles, can they readjust to the new geometry without a change in the total volume of the particles? Consider, then, the four components of strain which the bulk sample can undergo and the manner in which the averaged particle motion can respond to that strain (figure 25).

1. *Translation*: the bulk element may translate. This motion would be achieved if every particle translated by the same distance. No relative motion of the particles is involved. Thus, there is a direct correspondence between the particle motion and the bulk motion in this case.

2. *Rotation*: in this case, each particle may translate and rotate relative to the observer but they do not move relative to each other.

3. *Shear*: the bulk element may shear. Such a deformation would require the particles to move relative to each other, both by sliding and by rolling.

4. *Dilation*: the bulk sample may dilate or contract. This clearly requires a relative motion of the particles. Since the particles slide and roll to achieve shear, it is logical that they will probably also dilate or, of course, contract during shear.

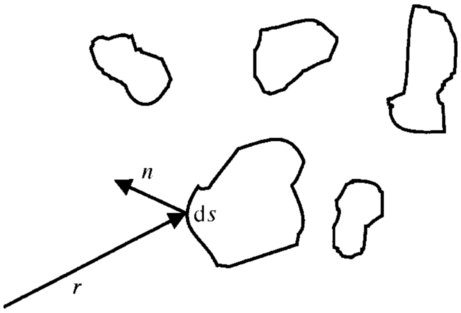

Figure 26. Position vector.

At the microscopic level, each particle may exhibit the same four motions. The shear and dilation motions represent deformation of the particles. If the deformation is plastic, it means that the particles can continuously change shape. It is quite possible that such deformation is the dominant mechanism in the 'stick-slip' behaviour that is observed. In addition, the particles may break, and this undoubtedly occurs at higher pressures. Nevertheless, if a critical state is reached, the implication is that the bulk sample can rotate and shear with no dilation and that this motion is the result of the rigid-body motions of the particles, only translation and rotation. Each small element of every particle contributes to this total motion, but the relative motion is created by the sliding and rolling at the surface of each particle. Presumably, therefore, the motion can be modelled if the position of every element of surface can be followed. Assume a surface element to have a vector position $\boldsymbol{r}$ that is known. The element is itself a vector and thus the unit normal, $\boldsymbol{n}$, must also be known. This is shown in figures 26 and 23*a*.

From these two vectors, a number of tensors of various rank can be constituted and can be integrated over different scales. The basic particle tensor is the dyadic:

$$\mathrm{d}s\,\boldsymbol{rn}.$$

When integrated over the whole surface of one particle, it is a useful particle parameter:

$$P = \int_s \mathrm{d}s\,\boldsymbol{rn}.$$

The scalar invariant of this parameter gives the volume of the particle:

$$|P| = \int_s \mathrm{d}s\,\boldsymbol{r}\cdot\boldsymbol{n} = 3V.$$

Furthermore, when resolved in any direction the parameter still represents the particle volume and now represents the total length of all the chords which can be drawn in that direction multiplied by their cross-section:

$$|P| : \boldsymbol{ii} = \int_s \mathrm{d}s\,(\boldsymbol{r}\cdot\boldsymbol{i})(\boldsymbol{n}\cdot\boldsymbol{i}) = V.$$

The total chord length, of course, must remain the same in every direction, but the distribution of lengths can be different. The relationship applies also to an array of particles, that is, it can be summed over a region containing Q particles. The total

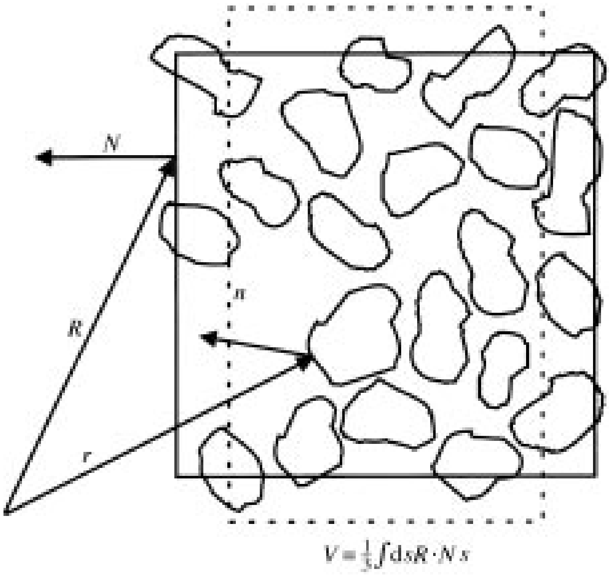

Figure 27. Deforming array.

volume of the particles is

$$V_{\mathrm{p}} = \sum_{Q} \int_{S} \mathrm{d}s\, \boldsymbol{r} \cdot \boldsymbol{n}.$$

The same equation can be applied, of course, to the imaginary region which has been drawn, as shown in figure 27. Thus

$$V_{\mathrm{T}} = \frac{1}{3} \int_{S} \mathrm{d}s\, \boldsymbol{R} \cdot \boldsymbol{N}.$$

The porosity is thus given by

$$1 - \varepsilon = \sum_{Q} \int_{S} \mathrm{d}s\, \boldsymbol{r} \cdot \boldsymbol{n} \bigg/ \int_{S} \mathrm{d}s\, \boldsymbol{R} \cdot \boldsymbol{N}.$$

Here, it is understood that the total surface of the particles includes the intersected surfaces which lie on the boundaries of the region. The porosity could also be defined by resolving both tensors in any direction, $\boldsymbol{i}$. Thus

$$1 - \varepsilon = \boldsymbol{ii} : \sum_{Q} \int_{S} \mathrm{d}s\, \boldsymbol{rn} \bigg/ \boldsymbol{ii} : \int_{S} \mathrm{d}s\, \boldsymbol{RN}.$$

This equation simply demonstrates that the fractional chord in any direction is the same as the fractional volume in any region, which is a well-known stereological result.

The particle tensor, P, is invariant with respect to the choice of the origin. This is easy to demonstrate by moving the origin, A, by a fixed value to B. Then

$$\int_{s} \mathrm{d}s\, \boldsymbol{r}_{\mathrm{B}} \boldsymbol{n} = \int_{s} \mathrm{d}s\, \boldsymbol{r}_{\mathrm{A}} \boldsymbol{n} + \int_{s} \mathrm{d}s\, (\boldsymbol{r}_{\mathrm{B}} - \boldsymbol{r}_{\mathrm{A}}) \boldsymbol{n}.$$

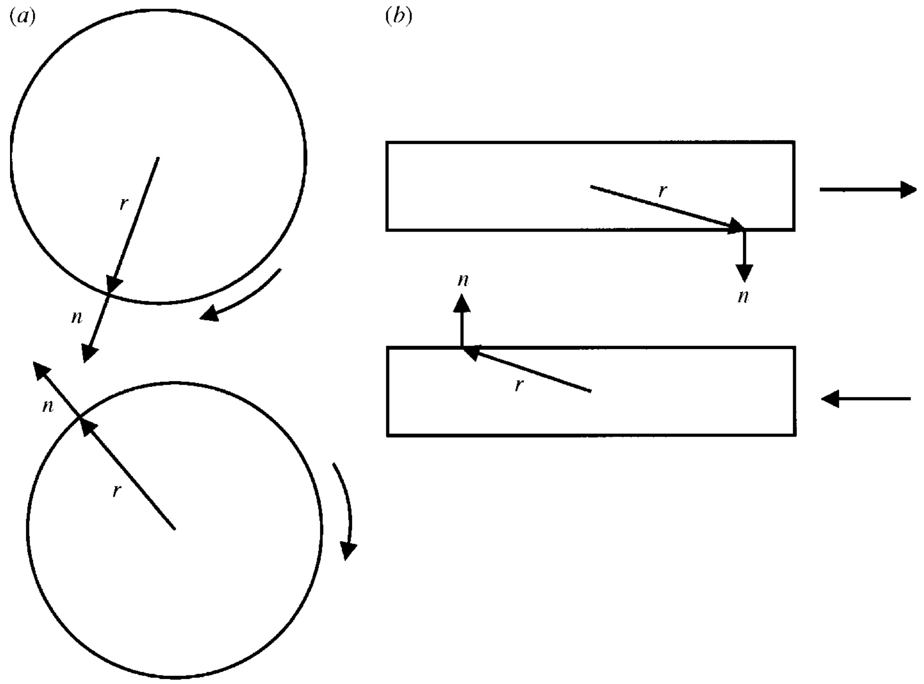

Figure 28. Rolling and sliding.

Since $(\boldsymbol{r}_{\mathrm{B}} - \boldsymbol{r}_{\mathrm{A}})$ is constant, it can be placed outside the integral sign. Then

$$(\boldsymbol{r}_{\mathrm{B}} - \boldsymbol{r}_{\mathrm{A}}) \int_s \mathrm{d}s\, \boldsymbol{n} = 0,$$

since $\int_s \mathrm{d}s\, \boldsymbol{n}$ is always zero for a closed body.

The question arises, is there a special choice of origin which is unique for a single particle? The particle tensor, P, may by split into its symmetric and antisymmetric parts. Thus

$$P = \frac{1}{2}\int_s \mathrm{d}s\,(\boldsymbol{rn} + \boldsymbol{nr}) + \frac{1}{2}\int_s \mathrm{d}s\,(\boldsymbol{rn} - \boldsymbol{nr}).$$

For a single particle, there is one unique point at which the antisymmetric part is zero, i.e. at the centre of rotation. That is the point at which

$$\int_s \mathrm{d}s\, \nabla \times \frac{\mathrm{d}\boldsymbol{r}}{\mathrm{d}t}\boldsymbol{n} = \nabla \times \frac{\mathrm{d}\boldsymbol{r}}{\mathrm{d}t} \int_s \mathrm{d}s\, \boldsymbol{n} = 0,$$

where $\frac{1}{2}\nabla \times (\mathrm{d}\boldsymbol{r}/\mathrm{d}t) = \omega$ is the angular velocity.

In considering the relative motion of particles, it is useful to use this point for each particle since its motion relative to this point is then a simple translation and rotation. The relative motion of the particles is then described by the relative motion of all these points. Consideration of this point immediately confirms some of the obvious facts about the influence of particle shape on the flow behaviour. Particles which do not have a wide distribution of the position vector relative to this point, can roll, those which do can slide (figure 28).

Thus, a set of sheets can shear relative to each other in a slip plane once they are aligned, but cannot respond to a change in the direction of stress. An array of uniform

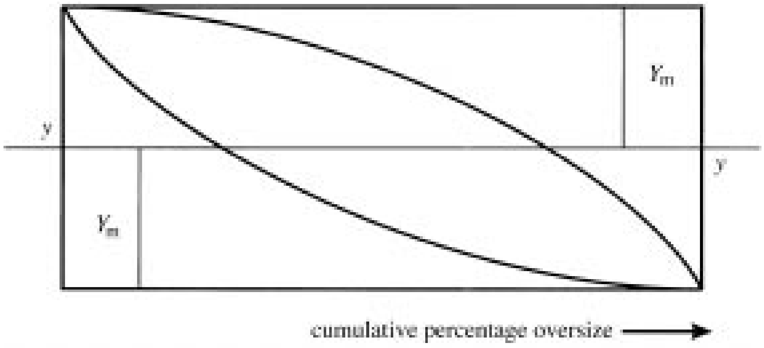

Figure 29. Free-flowing powder. $\sum_P \int_S \mathrm{d}s\, \boldsymbol{rn} = IV$.

spheres, on the other hand, can rotate around each other but do not easily form a slip-plane. The symmetry of the particle parameter, P, defines not only a special point for each particle but also the principal axes, which can be used to define the orientation of the particle relative to some other fixed axes. By integrating over an array of particles, a quantitative measure of the anisotropy is given. Definitions of the centre of rotation also define the chord distribution, which must be considered in manipulating the critical state. It is not the population of all the random chords, it is only those chords which pass through the centre of rotation of the particle. Thus, primary elements which make up the particle, for this purpose, are the tetrahedra that have their apex at the centre of rotation and their base on a surface element. It is suggested that this chord distribution be used to predict the critical state that the particles must attain in order to roll and translate relative to each other. The average spacing now predicted is the average distance between two particles, between their centres of rotation. This refined model allows anisotropy to arise, the particles are not assumed to be randomly orientated. However, a truly free-flowing powder would exhibit no anisotropy and so would have a ‘rotation-chord’ distribution as shown in figure 29.

6. Conclusions

There are two important points which are made in this paper and one important conclusion which can be drawn. It may be worthwhile first to emphasize the important points. There is ample evidence that the state of a particle array is rarely random and isotropic; there is always a degree of structure. This structure forms as a consequence of the process conditions but also as a consequence of the primary properties of the particles, the distribution of size and shape. To understand why and when a structure forms is an essential component of the study of granular mechanics. Ideal models of perfectly mixed and random arrays form a useful basis, but they rarely exist in practice. Segregation, anisotropy and other structure features are not secondary effects, they are often the dominant mechanism. Furthermore, when they are dominant, the only way to overcome them is to adjust the powder properties. It is not possible to mix a strongly segregating powder. A structure parameter such as anisotropy is clearly not a scalar variable and, thus, the parameter of porosity or concentration is inadequate to describe that structure. Of course, the array always has a porosity and that varies, maybe even in a systematic manner. Nevertheless, it is a shadow of the real parameter and is likely to mislead.

The second point is that a powder problem can only be systematically analysed by considering it, simultaneously, at different scales of scrutiny. The single-particle behaviour averages over the scale of many particles to effect a bulk property, and that bulk property varies with time and place within a piece of process equipment. However, the single particle is not the smallest element to consider. The shape and orientation of a particle are as important as its size, i.e. volume. These parameters are clearly not scalar: they are the compound of the position and orientation of each element of surface. Thus it is necessary to consider smaller elements of each particle as the primary unit of the system whose behaviour averages to constitute a bulk property.

The conclusion to be drawn from this paper is that these difficult problems are now amenable to experimental investigation, both by real hardware and by computer simulation. It is already possible to track the position of millions of individual elements, usually particles, through a process, and to add a constitutive equation to that process. The challenge is to compute the process simultaneously at the various scales of scrutiny, which is real parallel computing. The computer has also made it much more feasible to carry out sophisticated experiments in real hardware. Multiple variables can be recorded, correlated and analysed, and the computer can be programmed to control the experiment, in fact to react to the developing situation. This increased power to simulate and to experiment are the right and left hand. It is much easier to look into a simulated experiment and to print out the maximum detail, but in the end these observations can only be verified by real experiments. In this paper an old problem has been revisited. The problem is to understand the complex phenomena which occur when a powdered material flows in shear. The problem is not yet solved but it is probably true to say that the tools now exist to make a solution possible. Those tools are the further development and use of biaxial and even triaxial shear cells and the use of single-element computer simulation models.

References

Arthur, J. R. F., Chua, K. S. & Dunstan, T. 1977 *Géotechnique* **27**, 13.

Barber, B. 1972 Stress distribution in particulate systems. PhD thesis, Loughborough University, Loughborough, UK.

Beddow, J. K. 1989 Morphological analysis of particles and microstructure. *Key Engng Mater.* **29**–**31**, 677–695.

Hvorslev, M. J. 1937 Ingeniorvidenskab. *Skrifter* A **45**.

Jenike, A. W. 1961 Gravity flow of bulk solids. Eng. experiment station, bulletin no. 108. The University of Utah, Salt Lake City.

Jenike, A. W. 1964 Storage and flow of solids. Eng. experiment station, bulletin no. 123. The University of Utah, Salt Lake City.

Kaye, B. H. 1997 Characterizing the flowability of a powder using the concepts of fractal geometry and chaos theory. *Part. Part. Syst. Charact.* **14**, 53–66.

Maltby, L. P., Enstad, G. G. & De Silva, S. R. 1995 *Part. Part. Syst. Charact.* **12**, 16–27.

Mandl, G., Josselin de Jong, L. N. & Maltha, A. 1977 *Rock Mech.* **9**, 95–144.

Reynolds, O. 1885 On the dilatancy of media composed of rigid particles in contact. *Phil. Mag.* **20**, 127.

Roscoe, K. H., Schofield, A. N. & Wroth, C. P. 1958 *Géotechnique* **8**, 22–53.

Scarlett, B. & Todd, A. C. 1968 *Trans. ASME* **68**(MH-20), 1–10.
Schwedes, H. & Schulze, D. 1990 *Powder Technol.* **61**, 59–68.
van den Bergh, W. J. B. & Scarlett, B. 1991 *Powder Technol.* **67**, 237–263.
van der Kraan, M. 1996 PhD thesis, Delft University of Technology.