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Radial voidage variation in randomly-packed beds of spheres of different sizes

K. RIDGWAY AND K. J. TARBUCK

The packing arrangement of the particles in a tablet die, before pressing, has no hitherto been considered as a variable in the tabletting process. Because of the complexity of this subject, the present investigation has been restricted to mono-sized and binary mixtures of spheres in a cylindrical container. The container could be revolved at speeds in excess of 1,000 rpm so that the thickness of an annular layer of water could be measured. From this, the voidage at any radial position could be found. The plot of voidage against distance from the cylindrical wall is a wave form which is damped out after about 5 particle diameters. For packings of mono-sized spheres the oscillations are regular. There appear to be two kinds of binary mixtures, those in which the particle sizes are similar, and those where the diameter ratio is greater than 0·4. The voidages calculated by suitably weighting the values for each component are in agreement with the measured voidages of the first kind but not with those of the second. Because there is an initial distribution of voidage, it is concluded that there must be some radial movement of the particles during the compacting operation.

SINCE the mechanics of random packing of even the simplest forms of particles are not fully understood, any investigation should aim to make measurements on a simple system. Spheres in a simple cylindrical container constitute such a system, and they simulate the situation in a die into which powder or granules have been fed, before compression is applied. For the instance of the packing of mono-sized spheres in a cylindrical container, Scott (1960) has shown that there are two discrete values of the voidage (i.e. the ratio of free space to total space) of the bed, namely 36·3% and 39·9%. These values correspond to "tight", i.e. vibrated or tamped, and "loose", i.e. poured, random packings. The values refer only to the voidage of an infinite array, or alternatively, the central portion of a bed in a large container. They were obtained by extrapolation from the results for a series of beds of known bed diameter: particle diameter ratio. This extrapolation was necessary because at the walls of the container the particle packing is different from that in the bulk. Thus very close to the wall the voidage is high, simply by virtue of the geometry of the sphere and the relative flatness of the container wall (see Fig. 1). The pattern which the voidage follows along a radial line from the wall to the centre of the bed has been determined by Benenati & Brosilow (1962) and Roblee, Baird & Tierney (1958) for a number of values of the (bed/particle) diameter ratio. The present results are in agreement with previous work for mono-sized spheres [see Figs 3 (a), 3 (g), 4 (a) and 4 (g)].

Because the container wall is a smooth surface, the first layer of spheres tend to align themselves in a more or less close packed arrangement. All the spheres in this first layer touch the wall, so that their centres all lie at the same distance from the wall. The first layer is thus well-defined. This arrangement then forms the surface on which the second layer of

From the Department of Pharmaceutics, The School of Pharmacy, University of London, Brunswick Square, London, W.C.1.

spheres forms and consequently this layer is less ordered than the first. Following this principle, each successive layer is more and more random, until eventually the state of complete random packing is reached.

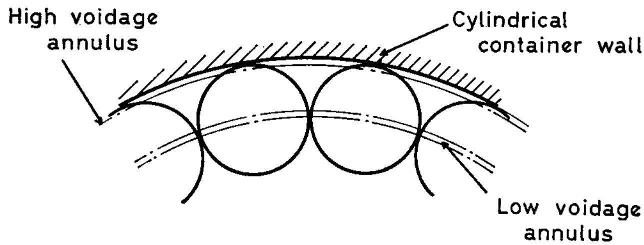

FIG. 1. Two spheres adjacent to a cylindrical wall. The voidage is unity close to the wall and decreases to a minimum at one sphere radius from the wall.

Along any radius of the cylinder there is thus a damped oscillation in the local voidage which can persist for up to five sphere diameters. This fact, it is believed, has not been considered in any tabletting research. The effect is of course dependent upon the size of the powder or granules in the tablet die. For example, Train (1956) used powders of about 200 mesh (aperture 3×10^3 inch) in punch and die sets of 5·68 cm diameter. In this instance the powder may be considered to be bounded by a *flat* die wall. However, many industrial tabletting operations employ 20 mesh granules in dies up to about $\frac{1}{2}$ inch diameter. Here the wall effect may well extend across the entire die charge.

Experimental

The technique used by Roblee & others (1958) and also by Benenati & Brosilow (1960) consisted of filling the interstices of a bed of wooden or lead spheres with a material such as wax or an epoxy resin to form a solid cylinder, which could later be machined on a lathe. The turnings of the annular cuts which were then made were collected and weighed and this enabled the voidage of the annulus to be calculated.

A method adopted by Shaffer (1953) consisted of the incremental filling with water of a packed drum with its axis horizontal. From measurements of liquid level in the drum and the volume of each increment, he was able, by a lengthy calculation, to find what the radial voidage distribution must have been to give the experimental readings which were obtained on *horizontal* layers. However, the calculation had to include the assumption that those parts of the second horizontal increment which were at the outer edges had the same voidage as had been determined for the first horizontal increment. Similarly, the results for the central part of the third increment depended upon taking the values obtained for the first and second. After four increments the results were valueless. We can confirm that this technique has this inherent drawback. It was therefore decided to employ a new technique using centrifugal force. If a packed bed contained in a cylindrical drum were rotated about its axis at sufficient speed any liquid present would form an annular layer at the surface of the cylinder. The required measurement is then the increase in thickness of this annular layer when a known volume of liquid is added. This gives a measure of the voidage in this thin annular region; this measure is completely independent of the voidage in any other part of the bed and there is thus no cumulative error.

The general arrangement of the apparatus is shown in plan in Fig. 2. It consisted of a 6-inch diameter horizontal cylinder of duralumin, mounted in two 7-inch precision ball races which were an interference fit with its outer surface. The accuracy of the voidage determination depends largely on smoothness of rotation of the drum. On rotation by hand the maximum deviation of the inside surface along the length of the drum was $\pm 1{\cdot}8 \times 10^{-3}$ inch. The overall drum length was about 12 inches.

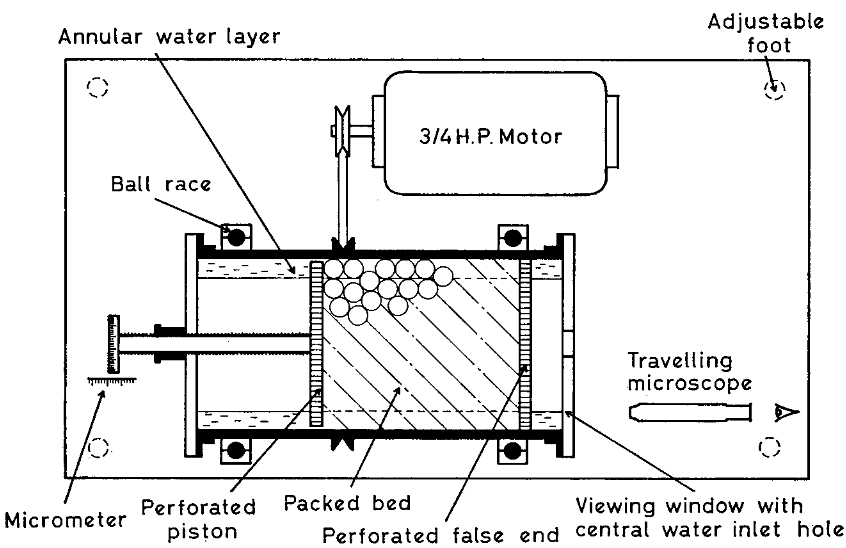

FIG. 2. Plan view of the drum and drive motor. The spheres are compressed into the right hand end of the drum by the piston. Water is added and the water surface in the double window is observed through the microscope.

The required number of spheres are packed into the drum, which can be vibrated by means of an out-of-balance weight rotated by a small subsidiary motor (not shown). The perforated piston is moved in by means of the threaded rod on which it is mounted, until the spheres have been compressed into the minimum volume which they will adopt under these conditions.

The drum initially contains a mixture of ethylene glycol and water of the same density as the spheres. This removes gravitational effects and enables the spheres to be thoroughly mixed before they are compressed by the piston. The glycol–water mixture is then run out and the bed dried by spinning for 10 min. A voidage determination is then made by adding water, whilst the drum is rotating, through an axial hole in the end of the drum. To enable the thickness of the annular water layer to be clearly observed one end of the drum is double, and consists of a plain outer observation window slightly separated from a perforated inner window which forms one end of the packed bed. Both are made of perspex, and the space between them allows a clear view of the liquid surface.

The drum is rotated by a $\frac{3}{4}$ h.p. DC motor and V-belt drive. Drum speeds between 1,100 and 1,400 rpm are used. At these speeds the deviation from circularity of the meniscus is a few thousandths of an inch, and since the difference of two readings is taken, this small error is balanced out. Because of the bulk of the drum and the relatively high speeds, all plugs, bolts, etc., were balanced by similar plugs and bolts diametrically opposite to prevent vibration. Runs were started at 1,100 rpm, and were usually carried through at this speed; when minor resonances occurred, the drum speed was changed to remove them. In addition, when the meniscus was close to the axis of the drum and the centrifugal force became smaller it was necessary to increase the speed to 1,400 rpm. The drum speed was checked frequently by a tachometer. The position of the water surface was determined by a travelling microscope which traversed horizontally along the diameter of the drum.

Results

The voidage at different distances from the wall of the drum was then determined by following the meniscus movement for 50 ml increments of water. This volume represents an optimum. The incremental volume should be kept small in order to give voidage values which approximate to the values at points. However, the change in thickness of the layer is measured more accurately if the volume added is larger. A compromise has to be made, and a change in thickness of 2 mm was chosen.

In this way, radial voidage variation plots have been obtained for polythene spheres of 8, 9, 10, 12 and 20 mm nominal diameter (plots of the latter four are given in Figs 4 *g*, 3 *g*, 4 *a* and 3 *a* respectively). Plots were also obtained for binary mixtures of 10 and 20 mm balls, and for 9 and 12 mm balls over a wide range of mixtures (Figs 3 *b*, *c*, *d*, *e*, *f* and 4 *b*, *c*, *d*, *e*, *f*). Replicate determinations of most of the curves were made. The replications were of two types (*a*) repeat runs on the undisturbed bed of spheres and (*b*) repeat runs after removing the spheres and repacking the bed. The mean deviation of the experimental points from the best curve which could be drawn through them was not greater than 0·02 units of voidage, up to a distance of 4 cm from the drum wall. Beyond this distance the accuracy decreases.

Discussion

The common feature of all plots, and particularly those for mono-sized spheres, is a regular damped oscillation in voidage. A minimum voidage occurs at a distance corresponding to about half a ball radius from the wall, i.e. at the centre of the first layer of balls. The first maximum occurs at something less than one ball diameter and the second minimum at the centre of the second layer ball. Because of the increasing degree of randomness, the waveform is gradually damped out.

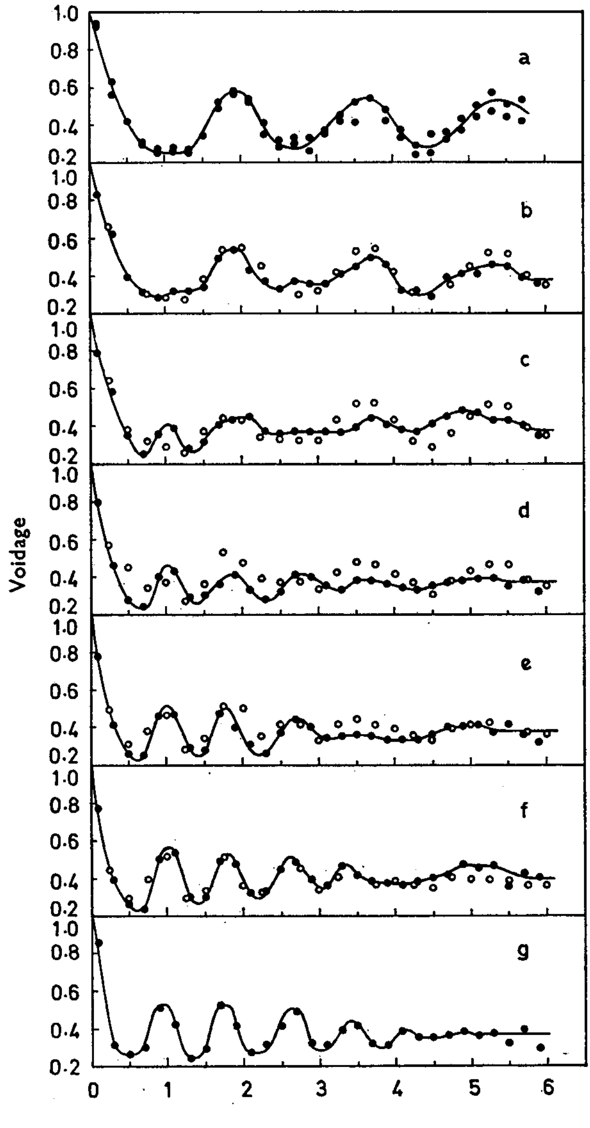

FIG. 3. Radial voidage distribution for mixtures of 10 and 20 mm spheres in a cylindrical container. ●, experimental. ○, calculated by suitably weighting values for the individual components. Mixture composition (% 10 mm by number): a, 0; b, 33·3; c, 58·4; d, 85·7; e, 95·3; f, 99·1; g, 100.

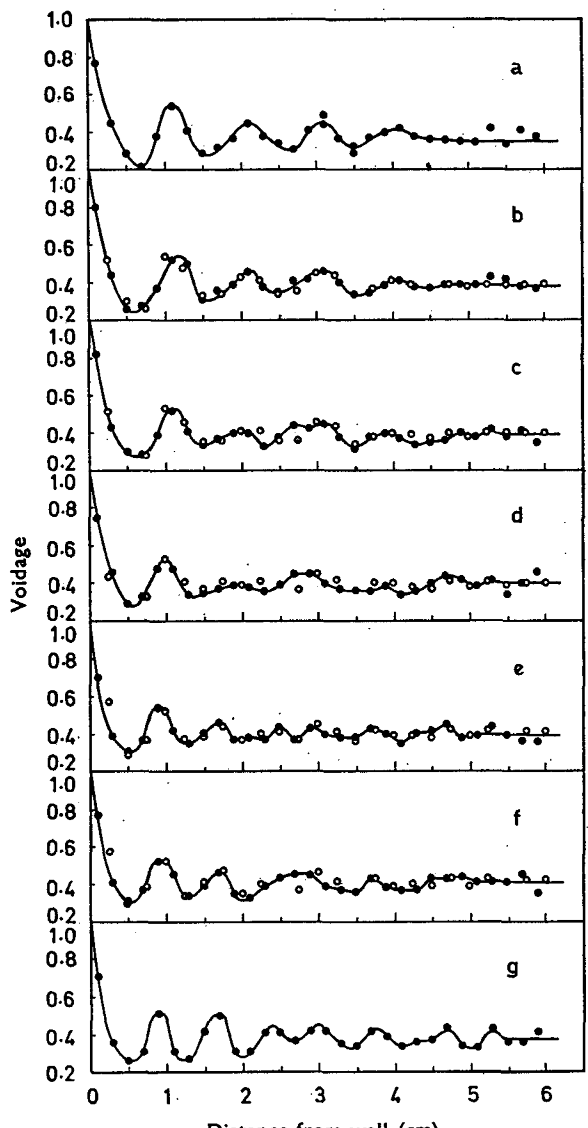

FIG. 4. Radial voidage distribution for mixtures of 9 and 12 mm spheres in a cylindrical container. ●, experimental. ○, calculated by suitably weighting values for the individual component. Mixture composition (% 9 mm by number): a, 0; b, 12·2; c, 31·9; d, 63·7; e, 80·6; f, 89·6; g, 100.

There is an obvious difference between this idealised investigation using spheres in a cylindrical drum and the manner in which non-spherical particles pack in a tablet die. The degree of irregularity of the particle will affect the packing to a great extent. Roblee & others (1958) have shown that with cylindrical particles (length = diameter) the voidage becomes constant after about three particle diameters only. With Berl saddles, a packing especially designed for distillation columns to give a uniform bed, the voidage becomes constant after about one diameter. No such particles exist naturally, however, and it seems likely that for the irregular particles generally encountered (i.e. when the sizes in three perpendicular directions are similar), the voidage variation dies out after about three to five equivalent diameters.

A voidage plot with the distance measured in terms of particle diameters shows that the curves for different sizes of sphere are similar (Fig. 5). Theoretically, these curves should be calculable, but the problem is one of three-dimensional statistics and as such is still unsolved.

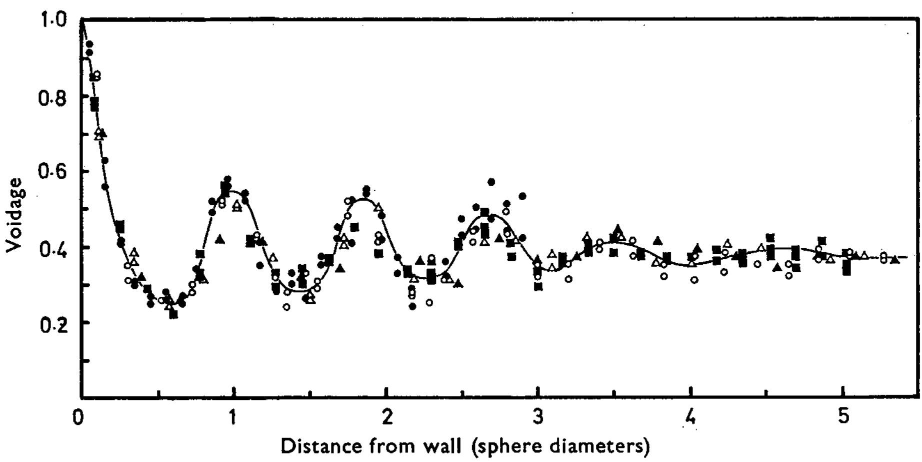

FIG. 5. A plot of radial voidage variation against distance from the drum wall measured in sphere diameters. ▲, 8 mm spheres. △, 9 mm spheres. ○, 10 mm spheres. ■, 12 mm spheres. ●, 20 mm spheres.

The two sets of binary mixtures were chosen to typify instances where the components are of markedly different size (Fig. 3) and where the sizes are similar (Fig. 4). There are two modes of packing for binary mixtures. The smaller spheres can fill the interstices of the larger spheres, and so produce a mixture of low voidage; or, alternatively, the smaller spheres can force apart the large spheres and produce a mixture of higher voidage as in the mixture of 9 and 12 mm balls (Fig. 4). The voidages of these mixtures correspond closely to the voidages calculated from values taken from graphs for the pure components, suitably weighted by the volume fraction.

As would be expected, all the curves for mono-sized spheres are similar, and have five clear minima except for the 20 mm spheres, Fig. 3 *a*, where the ratio of drum diameter to sphere diameter is less than 10 so that five minima cannot be fitted in. The packing thus retains remnants of the layer structure, induced by the wall, up to distances of about five sphere diameters. The mixtures all show a more rapid decay of the oscillation in voidage and it might appear that mixing spheres of different sizes causes a more rapid approach to randomness. That this is not necessarily so is indicated by the close agreement of the calculated points with the irregular curves in most instances. These points are calculated by summing the product of the voidage due to each pure component at any particular distance from the wall and the volume fraction of that component in the mixture. It thus seems that the different sizes of spheres exert their separate effects even when mixed with other sizes.

A useful practical result may well follow from curve 4 *d*, which is for 42·5% by volume of 9 mm spheres mixed with 12 mm spheres. The voidage fluctuations in this particular mixture become small immediately after the first complete oscillation. This could mean that a more uniform fill of a die would result from a mixture of two closely-sized fractions of granules. Whether this correlates with a better tablet in practice has yet to be tested.

It is generally thought (Kamm, Steinberg & Wulff, and others) that in the compaction of a powder mass in a die, there is no significant radial movement when the die walls are well lubricated. When this is so the powder is compacted in the form of a plug, any straight line parallel to the axis of the plug before compaction remaining undeviated after compaction. This, of course, does not mean that there is uniform compaction along the length of the line; Train (1956) has shown that distinct density patterns exist within a compact for a particular compacting pressure. (In this nomenclature, density = fraction of solids present = 1 − voidage.) It seems apparent that if any radial movement does occur, then it depends on the space available within the bed, i.e. on the radial voidage distribution. Since there is generally more free space near to the wall than in the centre of the bed, it would be expected that some outward radial motion must occur when the bed is compressed to a uniform high-density tablet. Furthermore, the compact should still show signs of annular layers of particles, although obviously the waves of the voidage distribution plot will be largely damped out. This formation can be seen in the lead shot compacts of Hersey (1960).

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