Solid phase compaction of polymeric powders: effects of compaction conditions on pressure and density variations

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In a recent communication it was shown that the pressure losses during the solid phase compaction of a polymeric powder could be predicted from a simple relationship of the form:

$$\frac{P_2}{P_1} = (K)^{h/D}$$

where $\frac{h}{D}$ is a function of the geometry of the compact. This has now been explored in more detail and it has been found that for PVdC and PVC the factor K is dependent on the rate of compaction and the die surface finish but is independent of compaction pressure and die diameter. To illustrate the effect of the pressure losses on the structural uniformity of the compacts, microhardness measurements were taken at a large number of points across a section of each sample. A computer plot of hardness contours provided a picture of the homogeneity of the sample which could be related to the compaction conditions and average density of the compact.

Keywords Mechanical behaviour; compaction; pressure; density; microhardness; powder

INTRODUCTION

The moulding of powders is a major industry in fields such as metals, ceramics, pharmacy and agriculture but as yet there has been limited interest in the moulding of polymeric powders. The main reasons for this are the ease with which polymeric materials can be shaped using melt processes and the preference which the moulder has for handling granules rather than powders. However, the fact that plastic powders can be formed cold is arousing interest in some quarters because this could offer considerable economic advantages in terms of energy savings. Recent research[1-4] has shown that the cold moulding of plastic powders may be a viable practical proposition and work is continuing to study the effect of the compaction variables on the quality and structure of the mouldings.

The compaction of powdered materials has been studied for many years[5,6] and there is much information available for metallic and pharmaceutical powders in particular. One of the major problems with cold compaction in a die was identified at the beginning of this century[7] – frictional losses which occur in the die. This causes a variation in pressure within the powder and results in a compact with a non-uniform structure. The nature of the pressure variations is of great importance in relation to the quality of the moulded product and so there have been many attempts to quantify the effect under a range of compaction conditions. For example, Train[8] used manganin resistance gauges embedded in the powder to measure local pressures directly. Others[9-11] have used indirect methods such as the measurement of hardness or density distributions within the compact so that pressure variations may be deduced. This technique has been explored for plastic powders[3] and it was established that a direct relationship exists between hardness number and density, although the nature of the relation depends on the compaction conditions. This is an important result because microhardness measurements across a section of a powder compact are relatively easy to make and when plotted as contours they provide a simple representation of the structural variations within a compact.

When a powder is compacted in a die using a single end punch the most direct evidence of the frictional loss effect in the die is the difference between the applied pressure (P_1) and the pressure acting on the base of the die (P_2). Detailed measurements of the ratio (P_2/P_1) for various powders has occupied research workers[9,12,13] for many years. It has been reported[2] that early trials with two polymeric powders had suggested that a relationship of the form:

$$\frac{P_2}{P_1} = K^{\frac{h}{D}} \tag{1.1}$$

could be used for plastics. The powders were pressed in a cylindrical die of diameter D and the height of the compact produced was h. The term K is nominally a constant although as with many of the properties of plastics it was found to be rate sensitive.

This paper presents the results of more extensive tests

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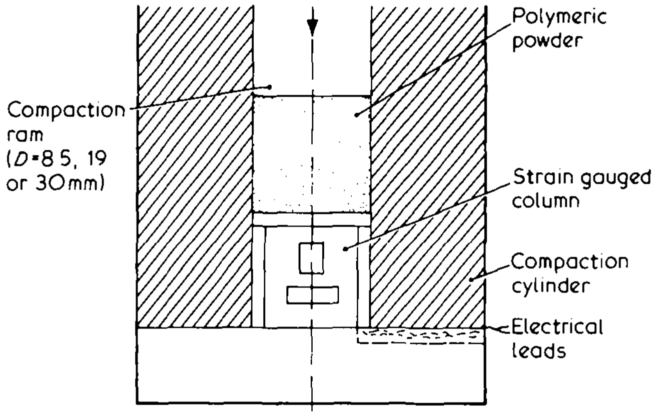

Figure 1 Use of column load cell to measure overall pressure loss due to friction

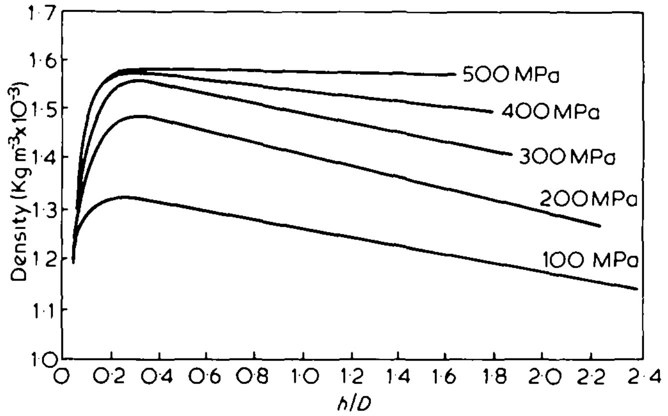

Figure 2 Average density of PVdC compacts as a function of h/D ratio. Die diameter = 19 mm; pressing rate = 8 MPa s^{-1}; bore CLA = 0.22 μm

which confirm the above relationship under a wide range of compaction conditions and sample geometries. For each of the conditions used and values of P_2/P_1 measured, the internal structure of the compact was explored using microhardness measurements and the results are presented in an attempt to correlate structure with the compaction variables.

EXPERIMENTAL

Materials

Two polymeric materials (produced by ICI) were selected for the program of tests. These were Viclan PVdC copolymer (VR 558) and poly(vinyl chloride) (PVC S71/102). The details of these powders, including material and particle characteristics have been given previously[1].

Cylindrical samples were produced in metal dies using single end compaction. The compaction force was applied using a 350 kN testing machine on which the hydraulic system could be controlled to produce a constant pressing rate on the powder.

Three compaction dies were available with diameters of 8.5, 19.0 and 30.0 mm. The surface roughness of each die was controlled accurately at a value of 0.5 μm (CLA as measured by Taylor Hobson method). The die length in each case was sufficient to produce a compact with maximum geometry ratio (h/D) of 2. The upper compaction pressure (P_1) was measured directly on the test machine dynamometer while the lower pressure (P_2) was obtained using a pre-calibrated resistance strain gauge load cell incorporated in the lower spacer of the die as shown in *Figure 1.*

The test pieces were ejected from the dies in the same direction as pressing and the average green density of each compact was obtained by weighing and measurement.

Evaluation of the structural uniformity of the compact and resultant changes with processing conditions was accomplished using a Wallace Microhardness tester. The details of the specimen preparation are given in ref 3. A special computer program was used to plot hardness contours from the values measured across a grid on the cross-section of the sample.

RESULTS AND DISCUSSION

The initial series of tests were carried out using the PVdC material in the 19 mm diameter die. In all of the tests the dwell time at maximum pressure was zero. The effect of non-zero dwell times is discussed later. *Figure 2* illustrates the effect of sample geometry and compaction pressure on the average density of the compacts. The pressing rate was 8 MPa s^{-1} in each case. At each compaction pressure the average densities of the compacts are very low at small values of h/D. As the height of the compact is increased the density increases and reaches a maximum in the region of $h/D = 0.25$–0.3. This dependence of compact density on geometry was first suggested by Balshin[14] although since then there has been little relevance attached to this. For plastics the effect is important, bearing in mind the correlation between strength and average density[1]. The reason for the poor densities achieved at low values of h/D is considered to be insufficient reorientation and sliding of particles in the initial stages of compaction. It can be visualized that in the limit, when the height of the compact (or initial powder fill) is very small (say close to the particle dimensions) then no consolidation of the

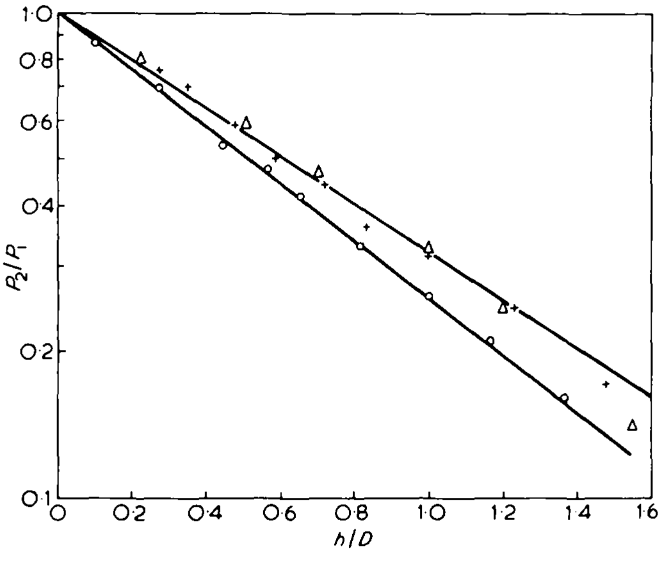

Figure 3 Effect of die surface roughness on pressure transmission in PVdC copolymer. Die diameter = 19 mm; pressing rate = 8 MPa s^{-1}

Symbol	P (MPa)	C.L.A. value (μm)
+	200	0.22
△	500	0.22
○	200	0.5

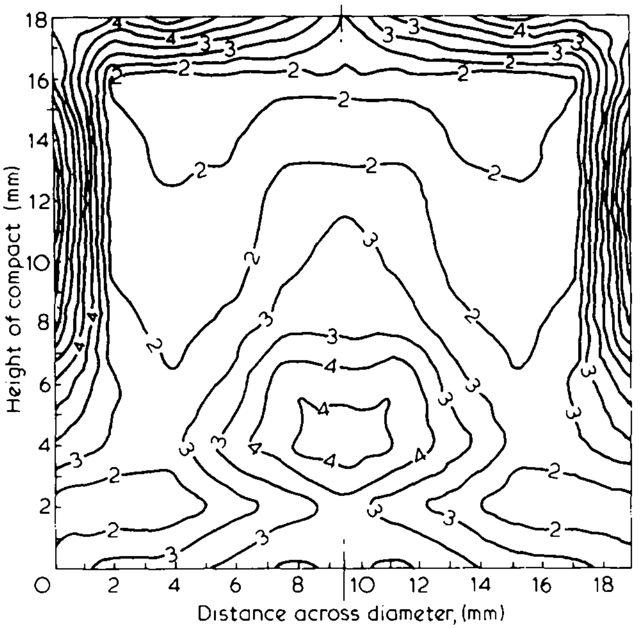

Figure 4 Microhardness distribution for PVdC copolymer compacted at 200 MPa. Rate = 8 MPa s^{-1}

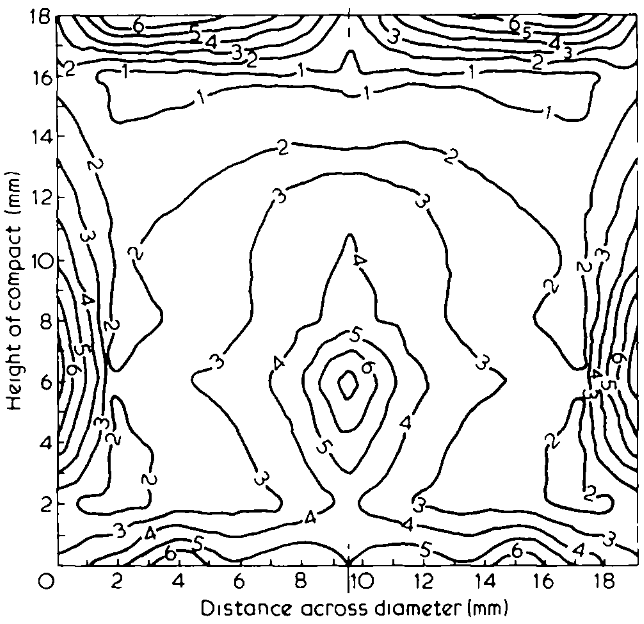

Figure 5 Microhardness distribution. P_1 = 500 MPa. Rate = 8 MPa s^{-1}

particles is possible. As the height of compact increases this effect decreases and the average density increases. However, at a certain point the frictional effects at the powder/die interface start to become dominant and the pressure variations within the powder cause a reduction in the average density of the compact. As the compact height increases this effect becomes more pronounced so that there is a gradual drop off in density as h/D increases.

Figure 3 quantifies the decrease in pressure within the compacts for various values of P_2/P_1. The linear relationships indicated on the semi-log plot in *Figure 3* confirm a relationship of the form given in equation (1.1). It was found that for any particular value of h/D, the pressure ratio is independent of the compaction pressure P_1. However, it has been shown[2] that the pressure ratio (P_2/P_1) is dependent on the pressing rate and a simple modification to (1.1) was suggested to take this into account. It was also found, not surprisingly that as shown in *Figure 3* the pressure ratio depends on the surface roughness of the die. For a CLA value of 0.5 μm the value of K in equation (1.1) was found to be 0.26 for a loading rate of 8 MPa s^{-1}. For the smoother die surface (CLA value of 0.22 μm) it was found that the value of K was 0.315 under the same compaction conditions.

Figures 4–8 show the structural variations within the compacts over the range of pressures, pressing rates and sample geometries discussed above. *Figures 4* and *5* compare the effect of compaction pressure and indicate that in each case the general structural pattern is the same

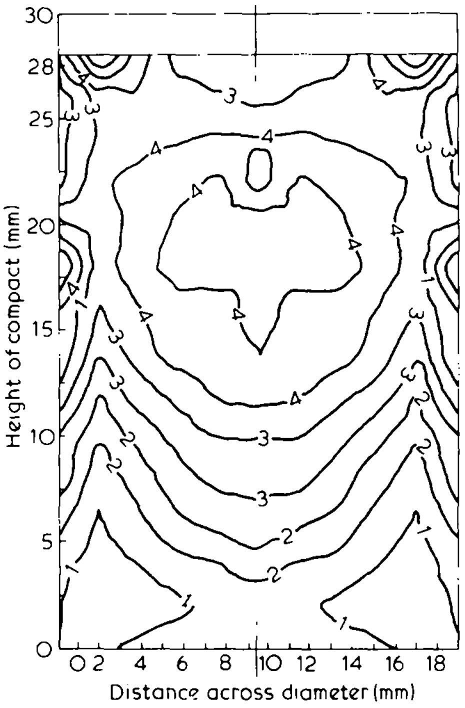

Figure 6 Microhardness distribution after compaction to 200 MPa in a specimen of h/D = 1.45; pressing rate = 8 MPa s^{-1}

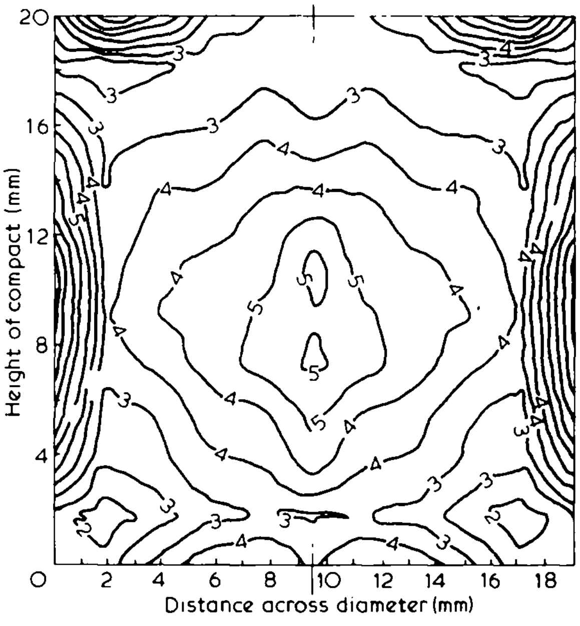

Figure 7 Microhardness distribution of a compact produced at 200 MPa with pressing rate 0.176 MPa s^{-1}

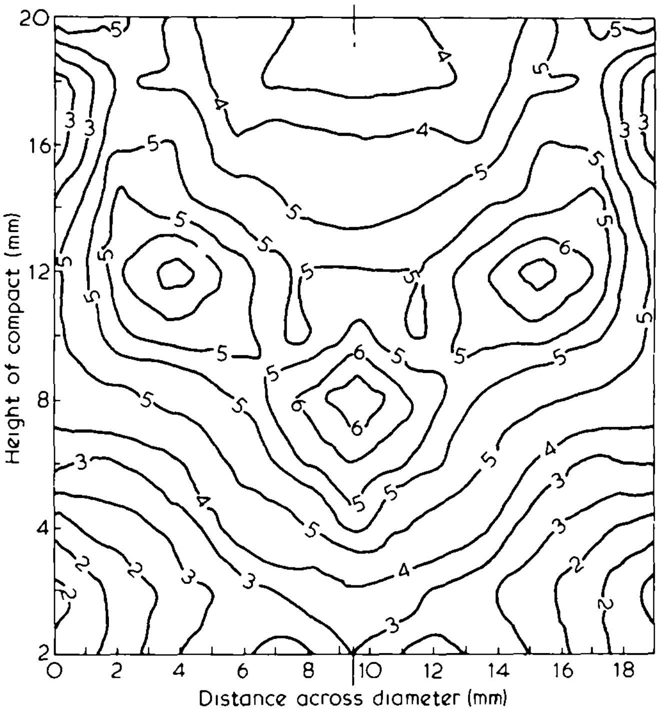

Figure 8 Microhardness distribution P_1 = 200 MPa; pressing rate = 20 MPa s^{-1}

in each case although at the higher pressure it may be seen that regions of greater hardness and hence higher density, are achieved. Increasing the compaction pressure does not improve the homogeneity of the compacts.

The patterns obtained in *Figures 4* and *5* are similar to those obtained by Train[8] using the manganin gauge technique on pharmaceutical powders. Similar variations have also been reported by other workers. The most significant feature is the area of hard (high density) material in the lower centre of the compact. This has been explained[8] in terms of the resolved components of axial and radial forces set up due to the resisting effect of the die walls. McLeod and Marshall[11] measured the density variations within ceramic compacts using autoradiography and explained the regions of high density in terms of the particle movement within the powder mass. In their model it is visualized that a central core of material is forced down into the unconsolidated powder below it. This continues until the reaction from the lower punch exceeds the die wall reaction at which point the material begins to move outwards radially. It is suggested that it is this change of direction which causes the high density zone due to the large shearing forces which are set up in this area.

The reduction in the average density of the compacts which occurs as h/D is increased may be confirmed by comparing the microhardness distributions shown in *Figures 4* and *6*. When the h/D value is increased from 1 to 1.45 the zone of high density material occurs in the upper half of the compact and the lower third exhibits only small values of hardness (and hence density). For this increase of h/D the average density of the compacts has changed from 1405 to 1350 kg m^{-3}.

The effect of pressing rate on the structure of the compacts may be seen by comparing *Figures 4*, *7* and *8*. As with varying the pressure, no improvement in the structural uniformity of the compacts is produced by varying the compaction rate in the region 0.176 to 20 MPa s^{-1}. The highest compaction rate caused the largest area of high density material and this was reflected in the higher average density measured for this condition (1410 kg m^{-3} as compared with 1390 kg m^{-3} at 0.176 MPa s^{-1}). The other feature observed in these Figures is the skin effect on the sides and upper surface of the compacts produced at the lower compaction rates.

In all the tests discussed so far, the dwell time at maximum pressure was zero. Tests at non-zero dwell times showed that the pressure transmission ratio (P_2/P_1) for the powder was not affected for dwell times up to 180 s. However, structural variations did take place when the pressure was maintained on the powder. This was not unexpected because in earlier tests it had been found that dwell time had a more pronounced effect at low pressures than at high. Therefore, since within a compact there is a range of pressures, there is a tendency for the low pressure areas to become more consolidated and thus there is a more uniform density distribution within the compact.

Preliminary trials with PVC have shown[2] that a relationship of the type given in equation (1.1) also applies for this material. *Figures 9* and *10* confirm the similarity in behaviour between PVdC and PVC over the same range of pressures, pressing rates and sample geometries. *Figure 9* shows that there is a maximum densification for an h/D

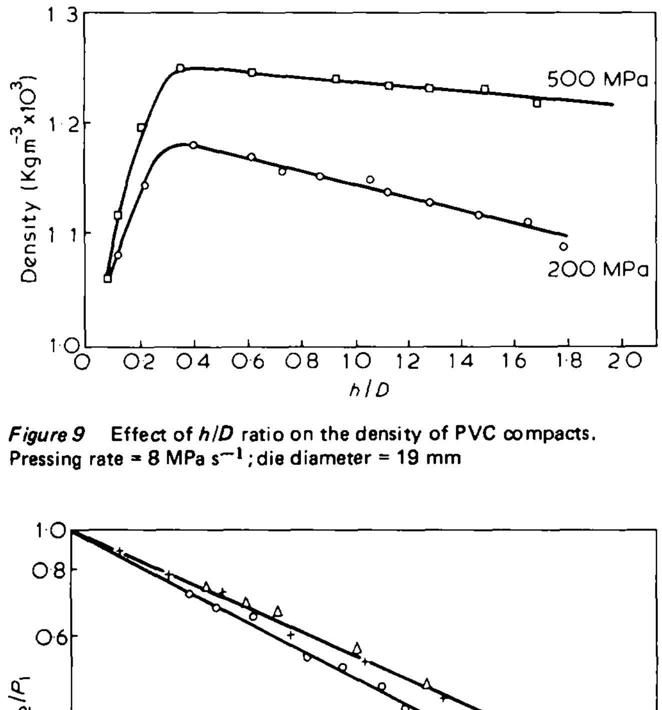

Figure 9 Effect of h/D ratio on the density of PVC compacts. Pressing rate = 8 MPa s^{-1}; die diameter = 19 mm

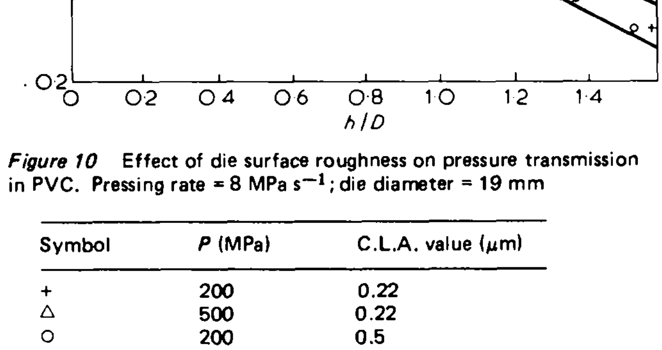

Figure 10 Effect of die surface roughness on pressure transmission in PVC. Pressing rate = 8 MPa s^{-1}; die diameter = 19 mm

Symbol	P (MPa)	C.L.A. value (μm)
+	200	0.22
Δ	500	0.22
○	200	0.5

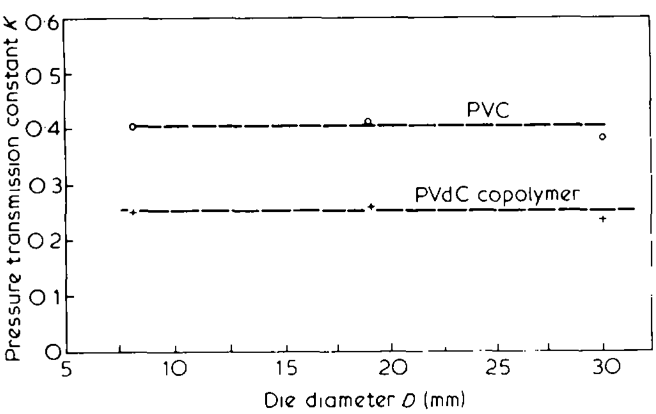

Figure 11 Relationship between pressure transmission constant and die diameter. Pressing rate = 8 MPa s^{-1}

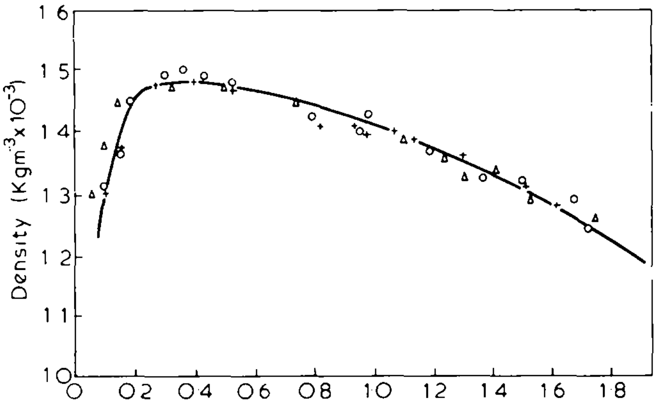

Figure 12 Effect of compact geometry on density on various diameter dies. Compaction pressure = 200 MPa; pressing rate = 8 MPa s^{-1}; bore CLA = 0.5 μm. ○, D = 8.5 mm; +, D = 19.0 mm; △, D = 30.0 mm

ratio of ~0.3. *Figure 10* shows that there is no pressure effect on the pressure transmissibility of the powders but there is a surface roughness effect. It was found that for PVC the value of K in (1.1) was 0.40 when the die CLA value was 0.5 μm (compaction rate = 8 MPa s^{-1}) and this increased to 0.42 when the smoother die (CLA = 0.22 μm) was used. There was also an effect due to the rate of compaction. In the smooth die, K changed from 0.38 at a pressing rate of 0.176 MPa s^{-1} to a value of 0.52 at a pressing rate of 20 MPa s^{-1}. It was found that these values could be predicted using the modified form of (1.1) suggested in ref 2.

In all previous tests the die diameter was 19 mm and the value of h/D was varied by changing the height of the compact. Since the proximity of the die walls has an important influence on the forces experienced within the powder it was decided to examine the effect of using dies of different diameters. Therefore two more dies (of diameter 8.5 and 30 mm) were produced with the same surface roughness as the 19 mm die used for the majority of the tests reported here (i.e. CLA = 0.5 μm). It was found that for both PVC and PVdC there was no significant change in the pressure transmission constant, K, for die diameters in the range 8.5–30 mm (see *Figure 11*). When the average densities of the samples having the same h/D but different diameters were compared, it was found that the values were generally similar in all cases (see *Figure 12*). However, there were some small but significant differences. For example the large diameter die gave consistently higher values of density at the low end of the h/D range. The reason for this is that for the same h/D value in all the dies, the 30 mm die will have a greater height of compact. Therefore, since the low densities were explained in terms of the poorer particle orientation and movement in short compacts, it is not surprising that the 30 mm die gives improved density values. Another interesting result is that the smallest die (diameter = 8.5 mm) consistently gave the highest value for the peak density at the optimum value for h/D. It is suggested that this is a direct result of the proximity, and hence greater influence of the walls, and the forces which they produce in the bulk of the powder.

It is also interesting to compare *Figures 2* and *12* for a compaction pressure of 200 MPa. The results in *Figure 12* were produced in a die with a bore surface roughness of 0.5 μm as compared with 0.22 μm for the earlier results. This emphasizes the effect of bore surface finish because the reduced frictional effects in the smoother die result in higher values of transmitted pressure, particularly at large values of h/D.

The structural variations in PVdC compacts produced in each of the three dies may be seen by comparing *Figures 4*, *13* and *14*. These illustrate the microhardness variations in samples which all have an (h/D) ratio of 1 and which were produced under the same conditions in the 8.5, 19 and 30 mm dies. The immediate conclusion that may be drawn from these diagrams is that not only does the compaction conditions affect the hardness/density relationship[3] but there must also be a geometry effect. The three samples in *Figures 4*, *13* and *14* all have similar values for their average density but the magnitudes of the hardnesses across each section are quite different. In particular, the structure of the 30 mm diameter sample is unusual because it exhibits two hardness peaks on its central axis. McLeod and Marshall[11] have observed this feature during their compaction tests on uranium dioxide powder. They attributed the difference between their

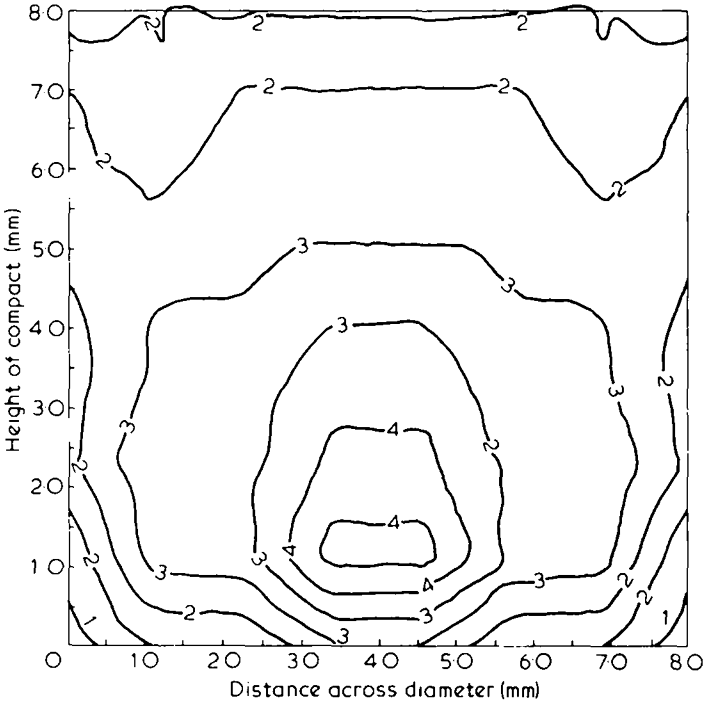

Figure 13 Microhardness distribution P_l = 200 MPa. Pressing rate 8 MPa s^{-1}; h/D = 1; D = 8.5 mm

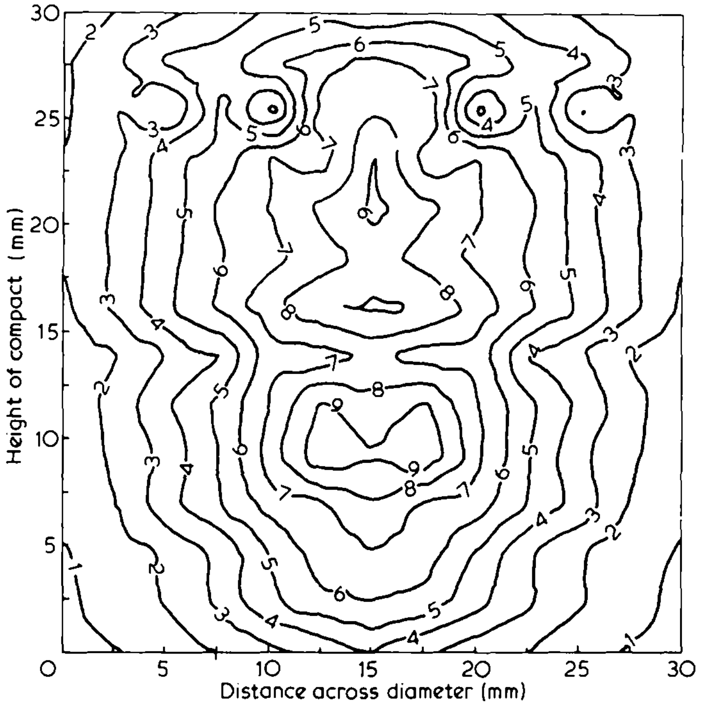

Figure 14 Microhardness distribution P_1 = 200 MPa. Pressing rate = 8 MPa s^{-1}; h/D = 1; D = 30.0 mm

structural pattern and that produced by Train[8] on magnesium carbonate powder as being due to much greater particle movement within their powder to produce the same percentage theoretical density in the two materials. This could also explain the similar effects produced here. In the two smaller diameter dies the conventional type of structural variations are observed, whereas in the large die the powder particles must undergo much greater movement, and hence shearing effects, in order to be compacted to the same density.

CONCLUSIONS

From the series of compaction tests on PVdC and PVC powders the following conclusions have been drawn.

(1) In a uniform section die the axial pressure (P_2) transmitted through the powder may be expressed in terms of the applied pressure (P_1) using the relation

$$\frac{P_2}{P_1} = K^{\frac{h}{D}}$$

(2) The 'constant' K in this equation is a function of the rate of compaction and the bore surface roughness in the die but it is independent of the applied pressure and die diameter.

(3) The average density of a sample produced by solid phase compaction of polymeric powder is markedly dependent on the compaction pressure used and the geometry of the die.

(4) Microhardness measurements across a section of a compacted sample give a good indication of the structural uniformity of the sample.

(5) Increasing the compaction pressure or compaction rate does not improve the homogeneity of the compact.

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