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Cold compaction of polymeric powders

R. J. CRAWFORD, D. W. PAUL
Ashby Building, Department of Mechanical and Industrial Engineering,
The Queen's University of Belfast, Stranmillis Road, Belfast, UK

Using single end compaction in an instrumented cylindrical die, the compaction characteristics of a variety of polymeric powders have been studied. The powders tested range from those which compact very well to those which do not compact at all. Using scanning electron microscopy, the particle size and shape for each powder has been noted and used in conjunction with other physical properties and deformation characteristics to identify the factors which enhance compactability. In general it has been found that a powder with a combination of small particle size, irregular shaped particles and a relatively low bulk modulus is most likely to compact well. As the physical characteristics depart from these desirable conditions the compactability of the powder is reduced.

1. Introduction

The compressibility of plastic powders and granules is of interest in a wide range of production methods. In conventional processing techniques such as injection moulding, extrusion, transfer moulding, etc., cold plastic powder is supplied to the machine and, as part of the normal sequence of events, it is compressed prior to being heated. Thus it is of interest to know such things as the bulk density of the powder, its compressibility, the work needed to compress the powder and how effectively pressures are transmitted within the powder. Surprisingly there is very little of this information available [1, 2] and the effects which particle size, shape, size distribution and material properties, etc., have on these variables are still undefined. It is only recently, with the interest in solid phase compaction as a novel processing method or as a more economic pelletising technique, that the compaction characteristics of plastic powders have been given serious consideration [3–15].

In these studies it has become apparent that there is a large variation in the degree of compaction which can be achieved with different plastics. Some plastics (e.g., polyvinylidene chloride) compact relatively easily into a solid object whereas other plastics (e.g., polypropylene) emerge from a compaction die apparently completely unaffected even by pressures in the order of 700 MPa. Indeed even within the one family of plastics (e.g., polyethylenes) it is possible to get some grades which compact easily whereas others do not compact at all. In other areas of compaction (e.g., powder metallurgy) the compactability of the materials is almost solely dependent on the particle shape. With plastic powders it is probable that this is also an important consideration but there are also a number of additional factors related, in particular, to the inherent properties of the material which are likely to affect the compaction process.

In a previous paper [3] three powders with varying degrees of compactability were studied in detail. However, although these studies on polyvinylidene chloride, polyvinyl chloride and polypropylene provided a useful insight into the role of the compaction variables, they were not conclusive in identifying the main features influencing the compactability of cold plastic powders. With the aid of a computer controlled testing machine to monitor changes in the applied/transmitted pressures and powder displacement during compaction, this investigation has now been extended to examine a much wider range of plastic powders. In addition, scanning electron micrographs have been used to indicate particle size and shape and to supplement information on the bulk density, compacted density, compressibility, compactability and work expended during compaction for each of the powders.

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2. Apparatus and materials

The powders were compacted in a 19 mm diameter hardened steel die using a single moving punch. The die surface roughness was measured using a Taylor Hobson Talysurf which indicated a centre-line-average (CLA) value of 20. The moving punch was forced down on to the powder using a 250 kN Dartec Hydraulic testing machine. This machine incorporated computer controlled feedback so that the powder could be compacted at either a constant loading rate or a constant punch displacement rate. In each case the rate was infinitely variable and was input to the machine via the computer terminal VDU. For the tests described in this paper the rate used was either 8 MPa sec^{-1} or 1 mm sec^{-1}.

In each test the charge of powder (9 g) was accurately weighed and poured into the die. During compaction, the applied force, the transmitted force (to the base of the die) and the punch travel were recorded simultaneously against time. In addition the applied force against punch travel characteristic was recorded automatically on the test machines graph plotter. These graphs were subsequently used to compare the compactability of each powder and determine the work done when the pressure on each powder was increased to 210 MPa. Full details of these characteristics for polyvinylidene chloride are given elsewhere [8]. After each test the die was cleaned thoroughly to prevent deposits of material on the die wall altering its surface roughness and thus affecting subsequent tests.

The bulk density of each of the plastic powders was measured in accordance with Method 501A of BS2872. In effect each powder was poured through a standard funnel into a 100 ml container and when the excess powder had been removed, the weight of powder in the container was used to calculate the bulk density.

The powders tested in the programme were as follows:

(a) Polyvinylidene chloride, ICI type VR558 (PVdC);

(b) Polytetrafluoro ethylene, ICI type G163 (PTFE);

(c) Ultra high molecular weight polyethylene (UHMWPE), (< 0.5% Al_2O_3);

(d) Polyethylene, Gallenkamp type PP560 (PE PP560);

(e) Polyethersulphone, ICI type RVO.35 (PES);

(f) Polyvinyl chloride, BP type P130/1 (PVC P130/1);

(g) Polyethylene, BDH Chemicals (density 924 kg m^{-3}, MFI 8.0) (PE BDH);

(h) Polyvinyl chloride, ICI type S71/102 (PVC S71/102);

(i) Polystyrene, source unknown (powder characteristics given) (PS);

(j) Acrylic, ICI type DA100 (Ac);

(k) Polypropylene – ICI type HW607M (PP).

3. Theoretical considerations

When a particulate mass of material is placed in a rigid die at room temperature and subjected to pressure, there are a number of distinct phases as the material is transformed from loosely packed powder to a closely packed (possibly solid) compact. When the powder is first poured into the die its density is the bulk density of the powder (see Table I). This bulk density is a function of the particle size and shape. With a small particle size material the bulk density is low due to fragile arching and bridging within the particulate system. This effect is more pronounced if the particle shape is irregular. The bulk density of the material does not appear to have any influence on its compactability.

When the compaction punch moves down on the powder the reorganisation of the particles as the pressure builds up, may be divided into several separate stages.

3.1. Particle rearrangement

Initially, as the compaction punch presses down on the powder, there is a phase in which particles, which have been held up lightly by friction or bridges within the powder, will be moved to lower positions. This will occur with very little applied pressure. In a careful study of copper powder having spherical particles, Duffield [16] found that this restacking occurred in the pressure range 0 to 5 MPa. Martin [17] showed that restacking of particles has an important influence on the pressure transmitted through the powder. This effect is particularly important in the later stages of compaction.

After the initial rearrangement of particles at very low pressure, a stage is then reached when further ram movement is not possible without particle deformation. At this point the density of the powder is referred to as the tap density. The extent, in terms of ram travel, of the initial particle rearrangement stage depends on the relative values of bulk and tap density. In general, small irregular

TABLE I Properties of powders studied during solid phase compaction trials

Material	Bulk density* (%)	Particle size (μm)	Particle shape	Work done (Nm)	% punch movement during first 5 MPa
PVdC	21	37	irregular	390	58
PTFE	20	< 10	irregular	85	84
UHMWPE	32	400	regular (spherical)	350	77
PE (PP560)	44	100	irregular	215	63
PES	36	800	irregular	480	51
PVC (P130/1)	42	3	regular (spherical)	280	52
PE (BDH)	33	300	smooth irregular	149	76
PVC (S71/102)	37	150	irregular	306	37
PS	62	200	spherical	310	10
Ac	60	75	spherical	304	18
PP	60	250	smooth regular	245	42

*Bulk density expressed as a percentage of solid density.

particles will experience a greater amount of compaction punch travel than a powder consisting of large spherical particles (see Table I).

3.2. Elastic deformation at contact points

When the tap density of the powder has been reached the particles begin to deform at the contact points in order to resist the applied force. Although the pressure distribution is non-uniform within the powder [5, 6], at any given point the powder particles will be subjected to a steadily increasing compressive force. In the early stages of this densification procedure further restacking may be taking place, especially in unsized materials. Changes in relative positions may involve local elastic contractions in which a particle deforms to pass through a small gap and then recovers when it reaches its new site. During this stage then, densification of the powder is caused by sliding between particles and elastic compression at the contact points.

The extent of this stage in the compaction cycle is dependent on powder failure properties and these are a function of material and powder characteristics. In the Hertz analysis of the elastic deformation of spheres under the action of a force, F, the contact radius, b, was found to be given by the equation

$$\left(\frac{b}{x}\right)^3 = \frac{3}{8}\frac{F}{x^2}\left(\frac{1-\nu^2}{E}\right) \quad (1)$$

where ν is Poissons ratio for the material, E is the modulus of elasticity and x is the diameter of the sphere. As the force, F, is increased the mean pressure, P_m, over the area of contact will increase until it attains such a value that the elastic limit is reached and plastic flow begins. From the Hertz equation it can be seen that the value of F required to cause plastic flow is extremely small. Bowden and Tabor [18] estimated that for a typical copper asperity of radius $r = 10^{-6}$ m, the value of F is less than the weight of a copper particle of 75 μm diameter.

3.3. Plastic deformation at the contact points

As the downward movement of the compaction ram continues the compaction pressure increases and eventually the material around the area of contact is flowing plastically. This is the dominant stage of the consolidation process and its nature and extent are governed by the failure properties of the material. Early transfer to plastic deformation may be found in powders where the particle shape is irregular as in such cases high stresses are formed on small areas of contact between particles, resulting in the elastic limit being overcome at relatively low values of compaction pressure.

Some materials such as brittle ceramic powders and coal [19] fragment suddenly when their yield stress is exceeded. Thus the void spaces within the powder are filled by rearrangement and percolation of the finer breakdown products. Generally it is considered that densification by this means is more difficult to accomplish than by plastic deformation. In their study of the compaction mech-

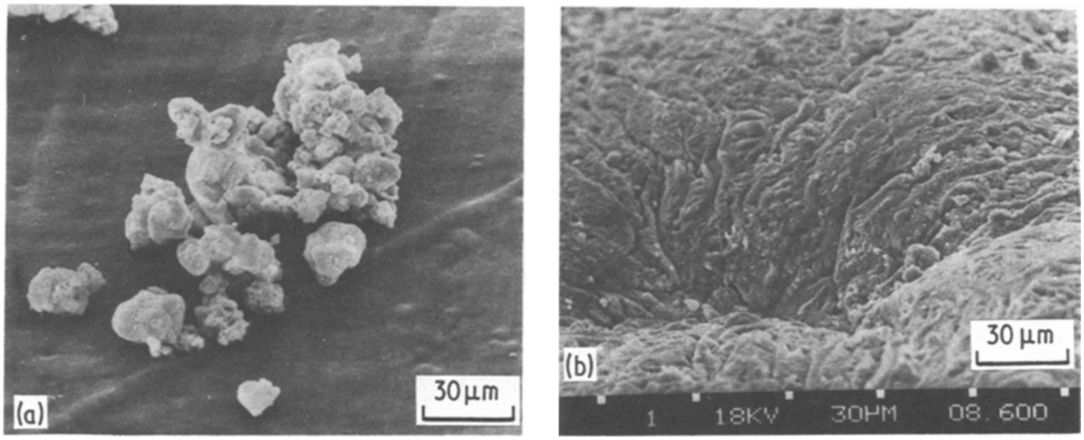

Figure 1 (a) Particles of PVdC before compaction (b) micrograph of PVdC compact.

anics in sodium chloride, sucrose and coal, Hardman and Lilley [19] encountered both types of densification. A detailed microscopic study of their compacts showed that the two effects were not always separable because in coal and sucrose compacts they found evidence of plastic deformation in the small broken fragments which were filling the voids.

3.4. Bulk compression of the material

The final stage of the compaction process occurs when no further reduction in porosity is possible by fragmentation, percolation or plastic deformation. Further densification then involves bulk compression of the material itself.

4. Results and discussion

In this series of tests the bulk density, average particle size and particle shape were determined for a selection of plastic powders prior to cold compaction in a solid metal die. During compaction, the pressing rate was either 1 mm $\sec^{-1}$ or 8 MPa $\sec^{-2}$ and the compaction force (F_1), transmitted force (F_2) and powder displacement were recorded. The object was to identify the powder characteristics which enhance the compactability of the powders so that those which do not compact well in their current state could have their characteristics suitably modified.

In early studies [3–8] the solid phase compaction of PVdC was studied in detail and so it is most convenient to consider it as a basis for comparison with the other materials. PVdC compacts relatively easily into a solid object which has a strength of up to 10 MPa even in the green state. This material has irregular shaped particles which range in size from about 10 μm to 90 μm with an average particle size of about 37 μm, as measured by a Coulter counter [3]. A scanning electron micrograph of the material before and after compaction is shown in Fig. 1. The bulk density of the material is 350 kg m^{-3}, i.e. about 21% of its solid density.

When 9 g of this material was placed in a 19 mm die and the applied pressure was built up to 210 MPa, at a rate of 8 MPa $\sec^{-1}$, the characteristics of Fig. 2 were obtained. It may be seen that in the early part of the test, there is a large amount of powder displacement for relatively little increase in applied force. As a result the test machine has difficulty achieving the set loading rate because it cannot supply sufficient oil under pressure to accommodate the large amount of piston travel. This is not a particular problem of the test machine used but would be common to all hydraulic test machines used in load control mode. The machine used here is probably better than most in this regard because it has an extra large pumping capacity to improve its dynamic performance.

After about 9 to 10 sec the machine is able to control the loading rate accurately to provide the desired 8 MPa $\sec^{-1}$. In the early part of the compaction process the powder is actually being pressed at a rate of about 7.4 mm $\sec^{-1}$ and it may be seen that during this stage the force transmitted to the base of the die starts to build up in phase with F_1. At about 9 sec there is an overshoot of force after which the ratio of F_2/F_1 remains approximately constant. An important point for later discussion is that the powder undergoes about 58% of its total displacement for an applied pressure of less than 5 MPa.

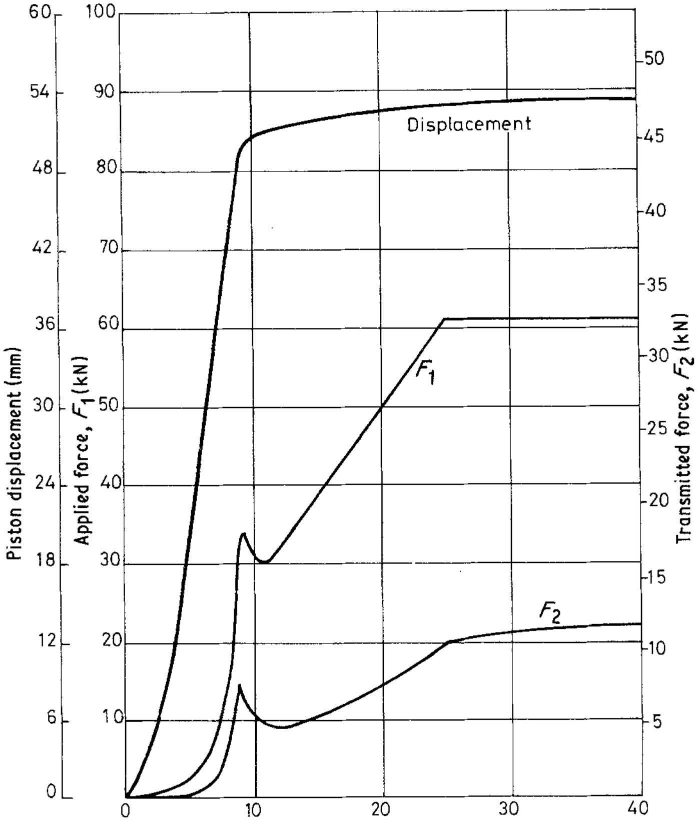

Figure 2 Compaction characteristics for PVdC at a pressing rate of 8 MPa $\sec^{-1}$.

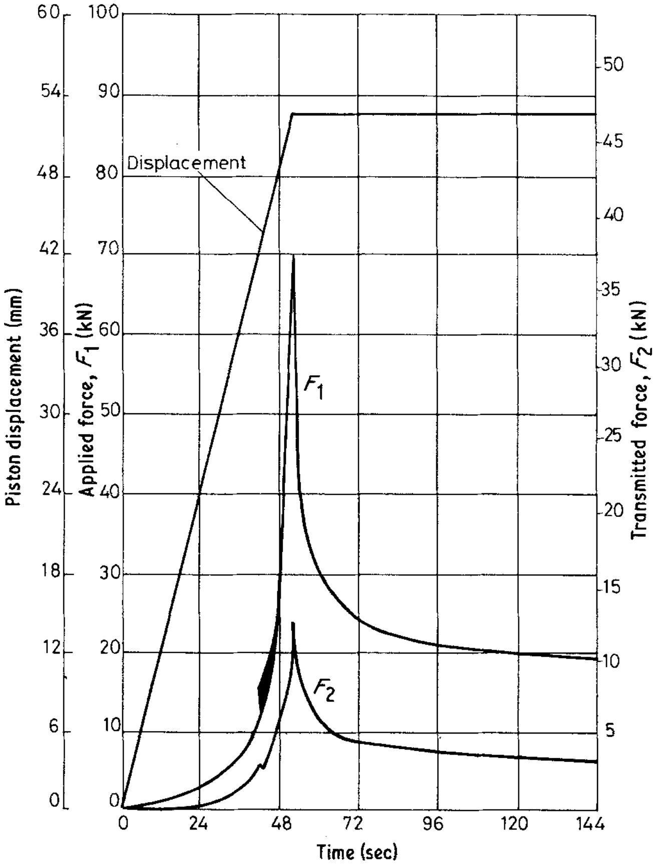

Figure 3 Compaction characteristics for PVdC at a pressing rate of 1 mm $\sec^{-1}$.

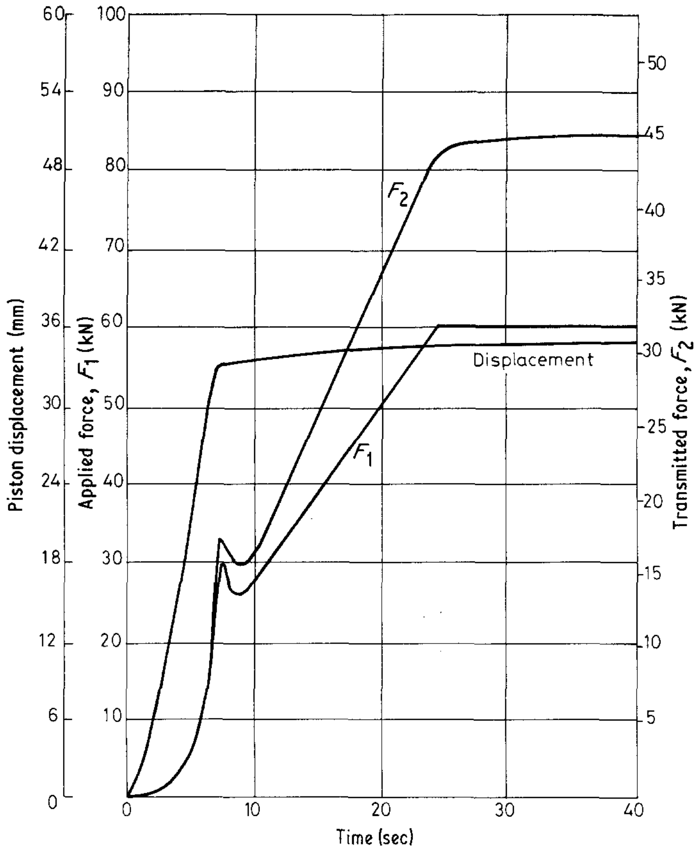

Figure 4 Compaction characteristics for polyethylene (type PP560) at a pressing rate of 8 MPa sec^{-1}.

When 9 g of the same powder was pressed at a controlled rate of 1 mm sec^{-1} the test machine has no difficulty in achieving this rate throughout the test. From Fig. 3 it is clear that the greater part of the powder displacement occurs at relatively low force. When the column of powder in the die has been reduced in height by about 42 mm (80% of total) the applied force has only reached 12 kN (17% of total applied force). At this point there were fluctuations in the force characteristics and subsequently the forces built up very quickly to their peak values. At the end of the test, when the powder displacement was kept constant, there was relaxation of both the applied and transmitted forces. There are several schools of thought regarding the value of the force ratio, F_2/F_1, during compaction of powders. Some workers have reported a constant value throughout the compaction process but generally these tests have been done at relatively low pressures. Figs 2 and 3 indicate that, although the ratio may be constant during some stages of the test, for PVdC the ratio does not have a single value throughout the compaction process. It is also evident that the mode of control (load or punch travel) has an effect on the shape of the F_2/F_1 characteristic.

From the range of powders tested in this programme of work it was possible to identify some other materials which compacted well. These were PTFE, polyethylene (type PP 560), and ultra high molecular weight polyethylene (UHMWPE). The compaction characteristics for these three materials are quite similar and so only those of polyethylene (type PP 560) are included. It may be seen from Figs 4 and 5 the general features of the 8 MPa sec^{-1} and 1 mm sec^{-1} compaction rates are similar to those described earlier. At the constant stroke rate, the ratio of F_2/F_1 is constant all the way through the test and this was also the case for PTFE and UHMWPE. At 8 MPa sec^{-1} the force ratio was constant at one value during the early stages of compaction and then showed a slight variation in the later stages of compaction (increasing as applied force increased). This behaviour was also observed for PTFE and UHMWPE.

The four materials which compacted well have a number of interesting features in common. In particular they all have a low bulk density relative

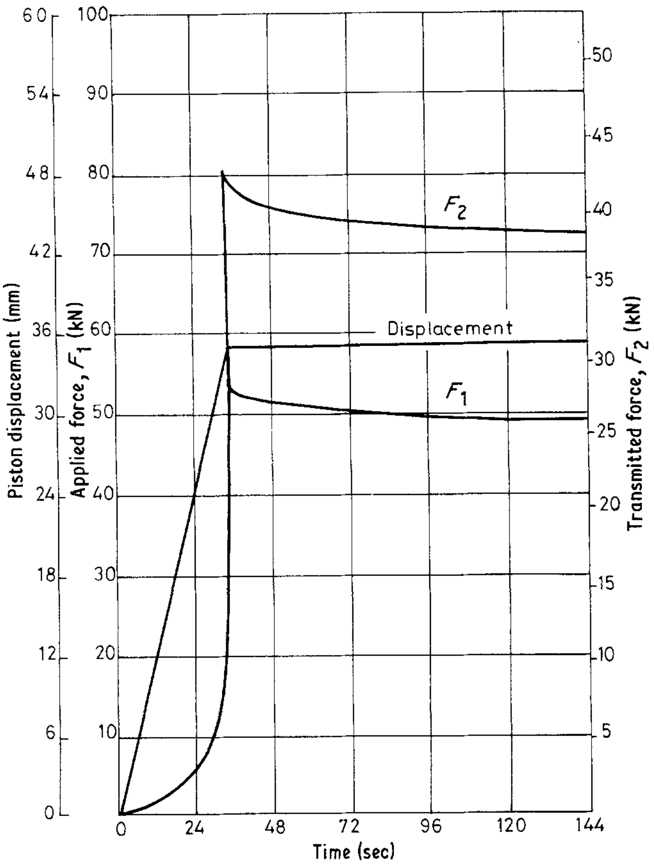

Figure 5 Compaction characteristics for polyethylene (type PP560) at a pressing rate of 1 mm sec^{-1}.

to the density of the solid material and as a result they undergo a relatively large amount of their displacement at a low compaction force. Three of the powders have irregular shaped particles; the exception was UHMWPE which had very smooth almost spherical particles. Two of the powders have fine particles, i.e. PVdC (37 μm) and PTFE ($<$ 10 μm). The other two powders have particle sizes in excess of 100 μm which is not normally regarded as conducive to good compaction. It is interesting that there is no apparent clue to the relative compactability of the materials from their force–displacement characteristics. Fig. 6 shows the wide spectrum of behaviour observed in the four materials and the work done during compaction varies from 85 Nm (for PTFE) to 390 Nm (for PVdC).

The main conclusion from the compaction trials with these widely different materials is that a powder with a relatively low bulk modulus and irregular particles, small in size has the best combination of characteristics to facilitate good compaction. As will be seen in subsequent tests on other powders, the compaction efficiency reduces quite considerably as the properties of the powder deviate from these requirements. The exception at this point appears to be UHMWPE which compacts quite well with only the low bulk density as a desirable characteristic.

The second group of plastic powders which were identified in this programme are those which appear to compact quite well, but break up on ejection from the die or are extremely friable when they are ejected. The powders in approximate descending order of compactability are polyethersulphone, polyvinyl chloride P130/1, polyethylene, polyvinyl chloride (S71/102) and polystyrene. The relevant properties for those powders are listed in Table I. Of these materials, the first two came closest to providing a good solid compact. In each case the product of compaction could be ejected from the die and handled but it had very little strength. It is interesting that these two powders have quite different physical properties but they have quite similar compaction charcteristics. Polyethersulphone approaches two of the desirable properties identified earlier, i.e. relatively low bulk density and irregular particles (see

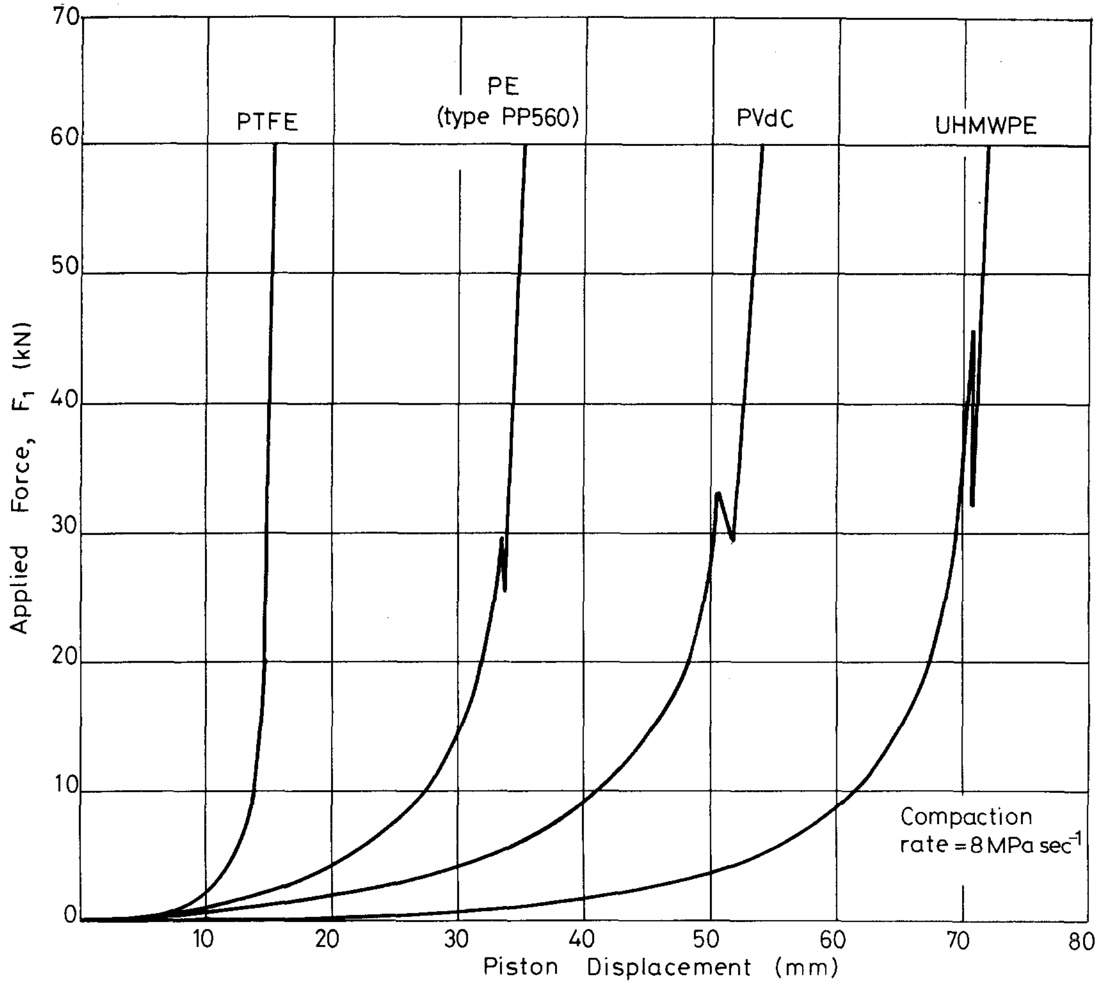

Figure 6 Deformation characteristics for a range of plastic powders.

Fig. 7a), but it has a very large particle size (about 800 μm). A scanning electron micrograph after compaction (Fig. 7b) suggests that the particles tend to lock together to form the green compact. The PVC (P130/1), on the other hand, has extremely fine particles (1 to 3 μm) but they are very regular, almost spherical, which is not normally regarded as a desirable feature in a powder which is to be compacted. Fig. 8a and b show the particles of this type of PVC before and after compaction.

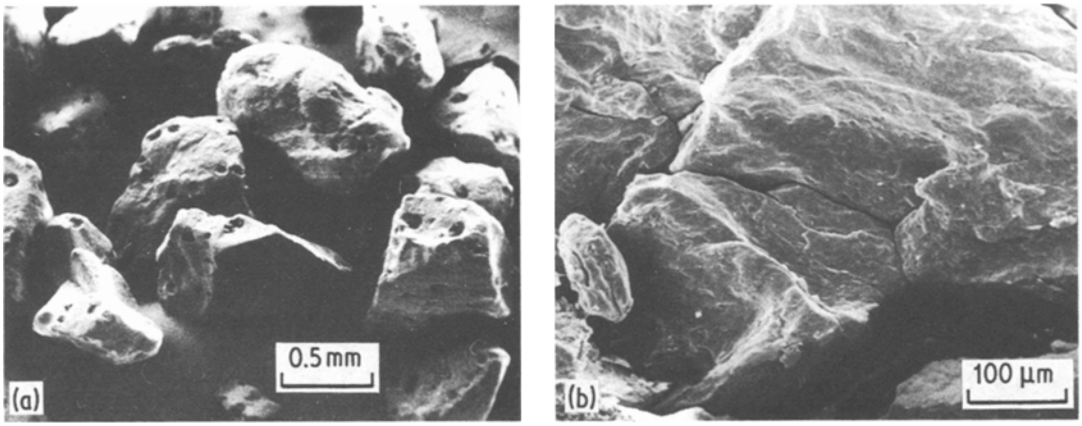

Figure 7 Particles of polyethersulphone (a) before compaction (b) after compaction.

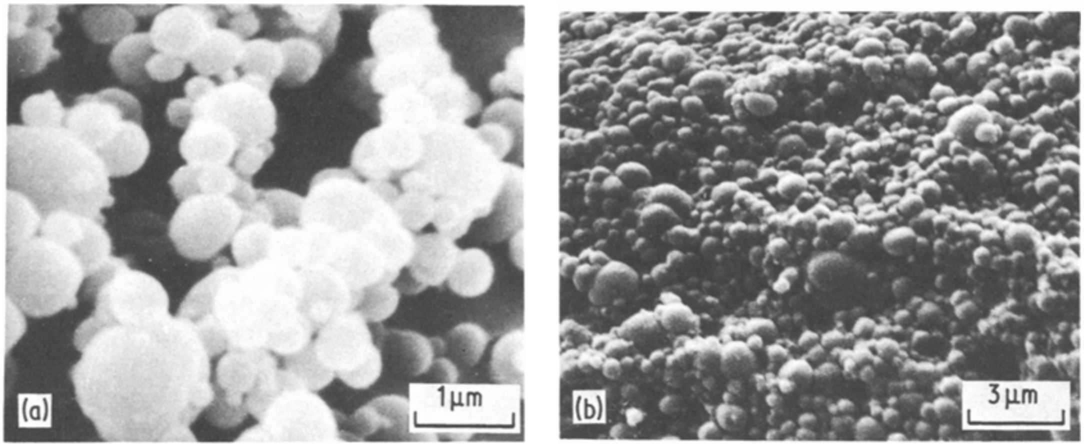

Figure 8 Particles of PVC (P130/1) (a) before compaction (b) after compaction.

It may be seen that in the compacted state the material has a very particulate structure not at all like the more homogeneous structure observed in the relatively strong compacts of PVdC (Fig. 1b).

The next two materials in this poor compaction category, i.e. polyethylene and polyvinyl chloride (S71/102), did not compact nearly as well as the grades of these materials reported previously. In this case the materials appeared, when they were still in the die, to have produced a solid compact but on ejection they broke up into powder. This was also the same for polystyrene. This latter material shows a significant departure on most counts from the physical characteristics of the good compaction powders in the first category. In particular it has large, spherical particles (see Fig. 9) and this, coupled with the high bulk density, suggested that from previous experience that it would have poor compaction behaviour. The force–time characteristics did not indicate any significantly different features from those observed for the good compaction materials. Figs 10 and 11 show typical characteristics for polyethersulphone and polyvinyl chloride (S71/102) and Fig. 12 shows the force-displacement characteristics for all five materials listed in this category. As observed previously, the work done during compaction does not seem to have any particular relevance if other physical properties of the powder (such as particle size and shape) are not conducive to good compaction. The values of work done for the materials illustrated in Fig. 12, vary from about 280 Nm to 480 Nm which is of the same order of magnitude as the work done on the materials which compacted well.

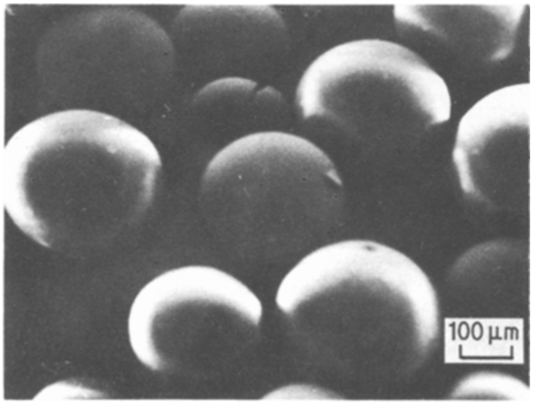

Figure 9 Particles of polystyrene before compaction.

The third category into which the materials were divided was those which do not compact at all. When the pressure was removed from the material in the die, the powder could be poured from the die with no apparent signs of adhesion between the particles. Two materials, polypropylene and acrylic, fell into this category. In both cases the particles were quite regular in shape and in the case of polypropylene, larger than would be regarded as desirable. The acrylic particles were spherical, quite similar to those of polystyrene shown in Fig. 9 although the acrylic powder was finer with an average particle size of about 75 μm. In powder metallurgy every effort is made to avoid regular shaped particles because of their poor compactability. It has been shown quite conclusively that adhesion between particles occurs due to welding at contact points. If the particles are very irregular, ideally star-shaped with

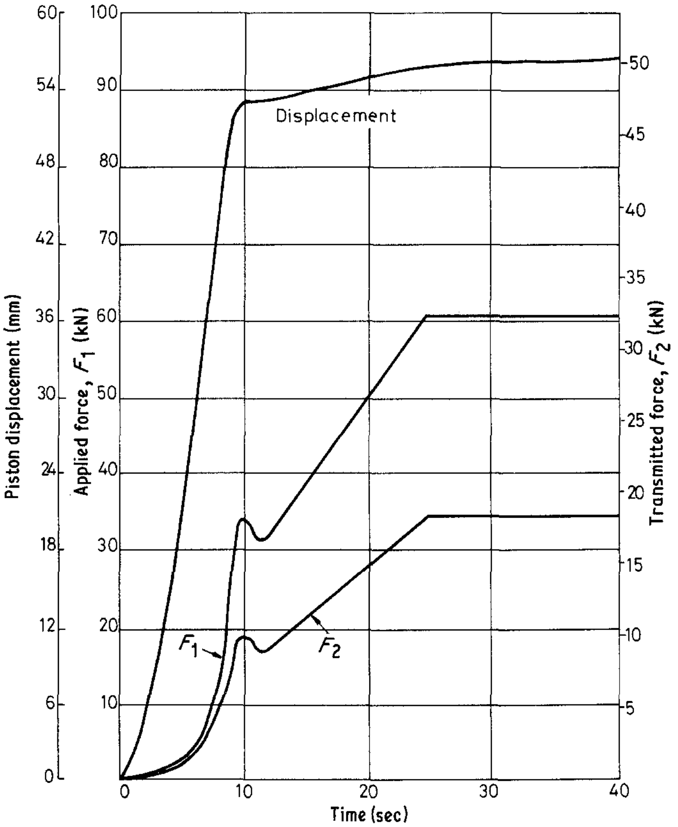

Figure 10 Compaction characteristics for PES at a pressing rate of 8 MPa sec^{-1}.

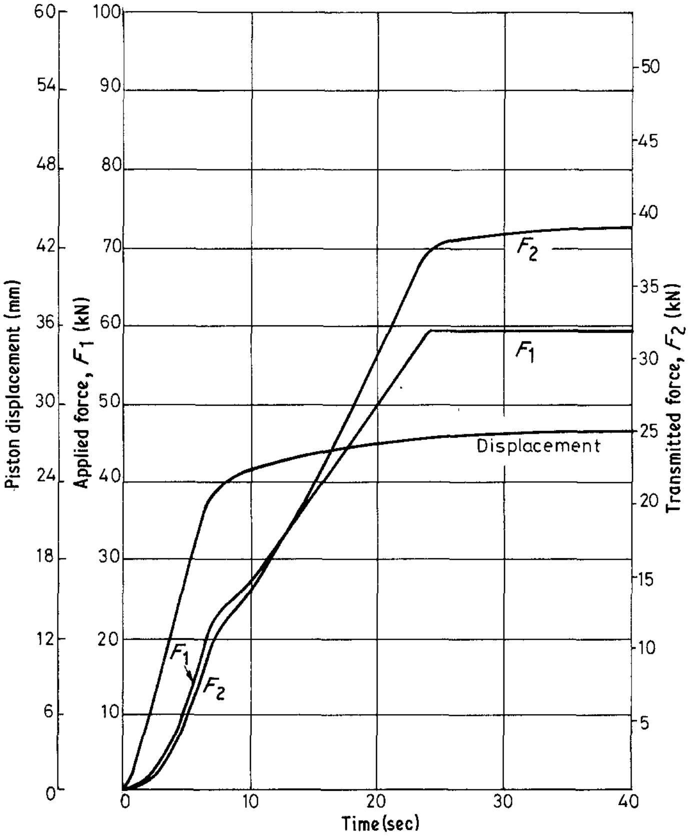

Figure 11 Compaction characteristics for PVC (type S71/102) at a pressing rate of 8 MPa sec^{-1}.

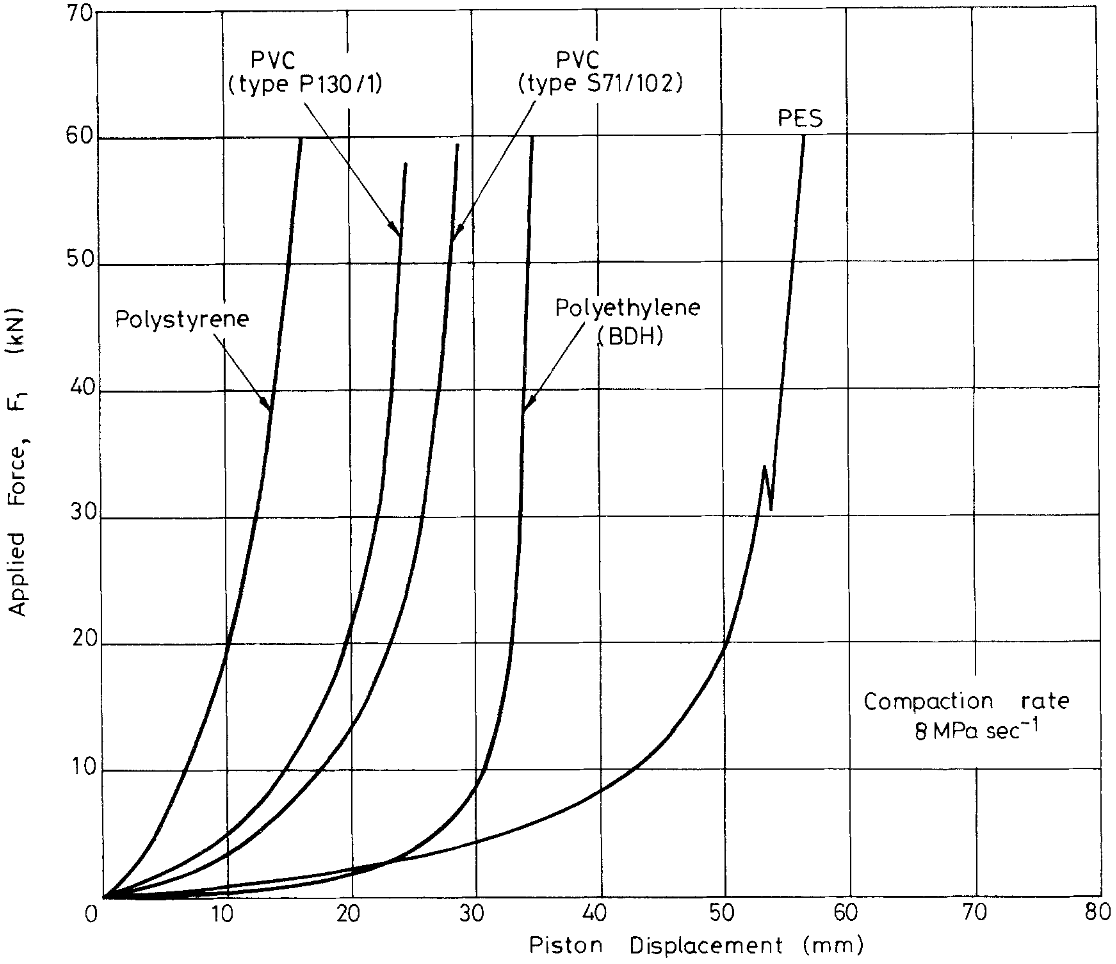

Figure 12 Deformation characteristics for a range of plastic plastic powders.

many sharp asperities, then the high pressures necessary to cause this welding can be generated at the very small area contact points. If the particles are smooth or in the worst case spherical then the pressures at the points of contact are relatively low. In addition, regular shaped particles pack more efficiently when they are poured into the die and thus the subsequent amount of relative movement of particles during compaction is small and there is little frictional head build up in the material. The work done on the materials which do not compact at all is similar to that done on materials which compacted very well (see Table I).

The compaction characteristics for polypropylene and acrylic are also quite similar to those shown earlier for the other materials, except that at a constant loading rate, the smaller amount of powder displacement which occurs for these materials means that the test machine has less difficulty in achieving the desired rate and so no overshoot of load is observed. Typical traces for polypropylene are shown in Figs 13 and 14. With acrylic there was a lot of slip-stick during compaction which was accompanied by fluctuations of applied and transmitted force. These fluctuations may be seen on the force-stroke characteristics shown in Fig. 15.

In an attempt to improve the compactability of polypropylene and possibly validate the suspicions that particle size and shape have a major influence, some of the polypropylene powder was re-precipitated. This was done by dissolving the polymer in 1.5 litres of nearly boiling xylene containing antioxidant, an atmosphere of nitrogen being maintained in the vessel. This solution was then added slowly to well stirred methanol and the precipitate was filtered and washed thoroughly. The polymer was then dried *in vacuo* at 100° C.

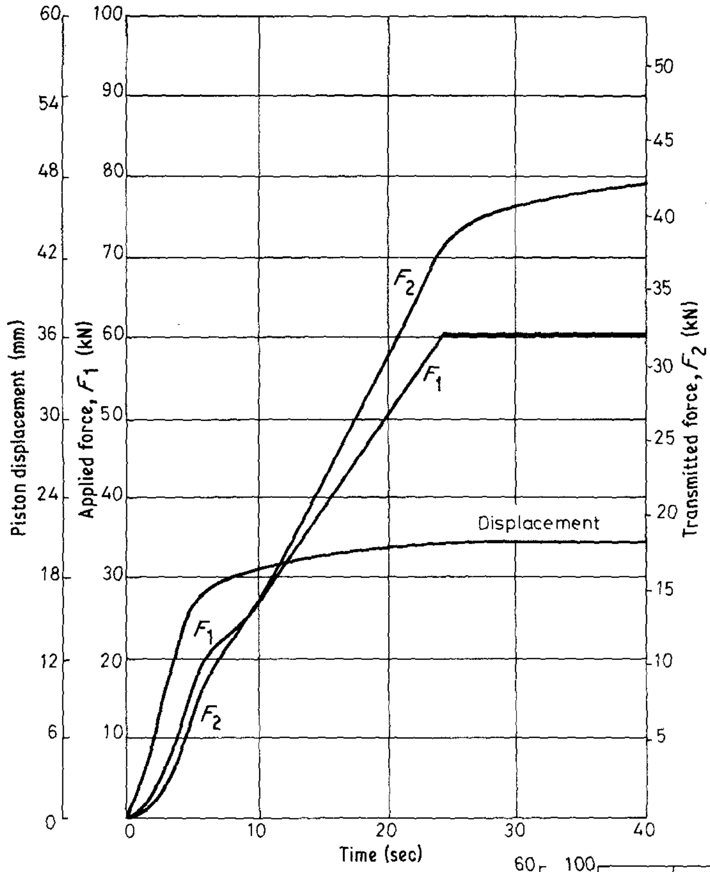

Figure 13 Compaction characteristics for polypropylene at a pressing rate of 8 MPa $\sec^{-1}$.

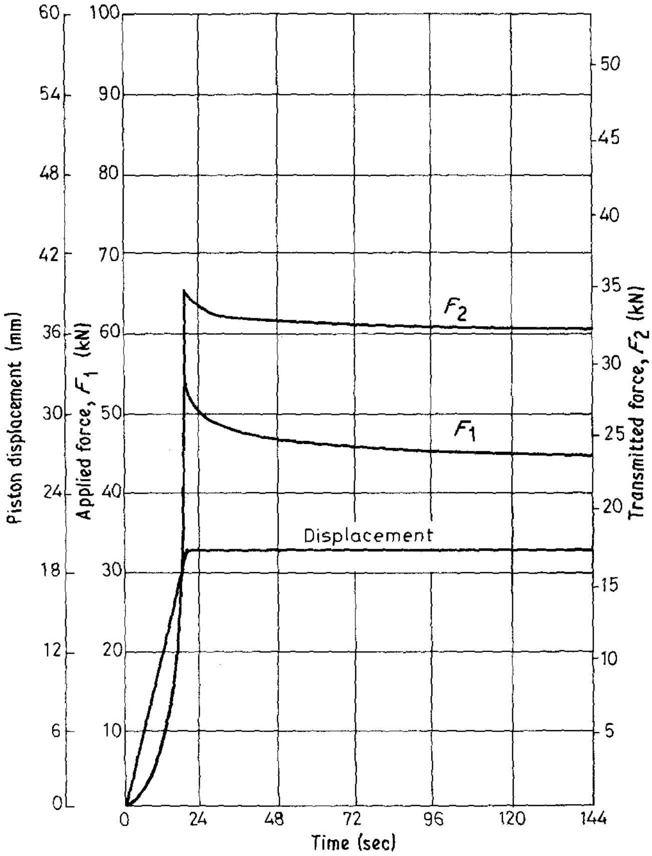

Figure 14 Compaction characteristics for polypropylene at a pressing rate of 1 mm $\sec^{-1}$.

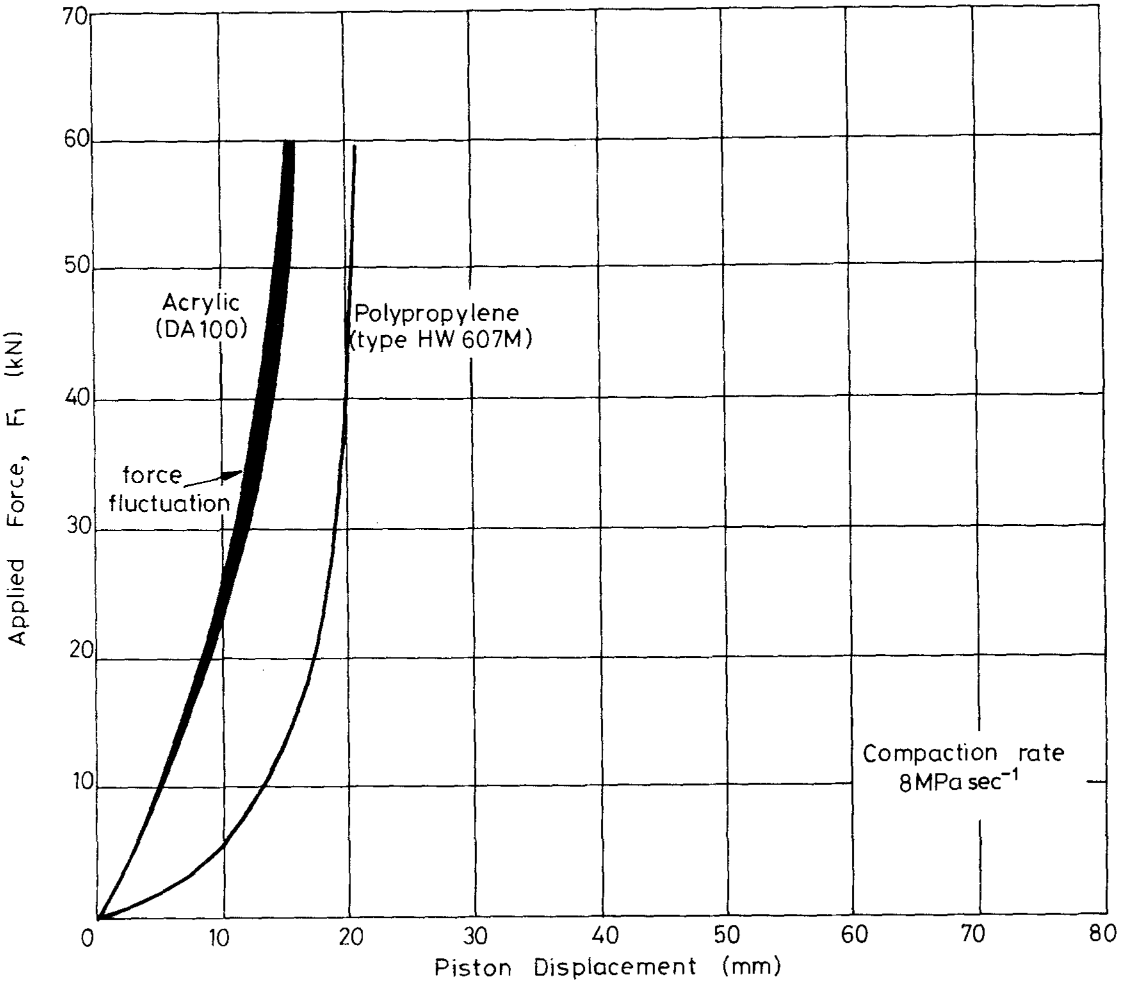

Figure 15 Deformation characteristics for a range of plastic powders.

The resultant polypropylene powder had a very small particle size ($< 5\,\mu$m) and the particles were irregular. These features coupled with a much lower bulk density meant that the powder was well within the range of properties regarded as desirable and indeed the powder compacted extremely well. Similar attempts to investigate the effects of particle size and shape with other polymeric powders are continuing. To date no success has been achieved by simply trying to break up the spherical particles of polystyrene and acrylic into irregular shaped particles.

5. Conclusions

From these cold compaction trials on a variety of polymeric powders the following conclusions may be drawn.

(1) Of the four materials which compacted very well, three exhibited a combination of low bulk density, small particle size and irregular shaped particles. The exception was UHMWPE which compacted very well despite having relatively large, regular shaped particles.

(2) The compactability of the powders tend to decrease depending on the degree of departure from the three desirable features identified above. Generally if two of the features are present then the powder can be compacted but it is weak and friable.

(3) The most undesirable physical property of a powder is spherical shaped particles particularly if they are large ($> 100\,\mu$m). Polymeric powders of this type could not be compacted at all.

(4) The work done on the powder during compaction is similar for materials which do and do not compact.

(5) There is no significant feature on the compaction characteristics for a polymeric

powder which could be used as an indication of compactability.

(6) The value of the ratio of transmitted force to applied force, F_2/F_1, is essentially constant throughout the test for constant stroke rate tests but a small variation in the ratio was observed in constant loading rate tests.

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References

1. R. S. SPENCER, G. D. GILMORE and R. M. WILEY, *J. Appl. Phys.* **21** (1950) 527.
2. D. TRAIN and J. A. HERSEY, *Ind. Chem.* **38** (1962) 77.
3. R. J. CRAWFORD and D. W. PAUL, *J. Mater. Sci.* **14** (1979) 2693.
4. *Idem, Polymer Commun.* **21** (1980) 138.
5. R. J. CRAWFORD, D. PAUL and Y. ADEEBNIA, *European Polymer J.* **16** (1980) 401.
6. R. J. CRAWFORD and D. W. PAUL, *ibid.* **17** (1981) 1023.
7. R. J. CRAWFORD, D. PAUL and D. SPREVAK, *Polymer* **23** (1982) 123.
8. R. J. CRAWFORD, *Polymer Eng. Sci.* in press.
9. T. MAEDA and S. MATSUOKA, *J. Fac. Eng. (Univ. Tokyo)* **23** (1975) 191.
10. D. M. BIGG, *Polymer Eng. Sci.* **17** (1977) 691.
11. G. S. JAYARAMAN, J. F. WALLACE, P. H. GEIL and E. BAER, *ibid.* **16** (1976) 529.
12. L. ERWIN and N. P. SICH, *ibid.* **16** (1976) 841.
13. G. W. HALLDIN and I. L. KAMEL, *ibid.* **17** (1977) 21.
14. H. K. PALMER and R. C. ROWE, *Powder Technol.* **10** (1974) 225.
15. K. UMEYA and R. HARA, *Polymer Eng. Sci.* **18** (1978) 366.
16. F. DUFFIELD and H. GROOTENHUIS, *Iron and Steel Inst.*, Special Report No. 58 (1956).
17. J. J. MARTIN, W. L. McCABE and C. MONRAD, *Chem. Eng. Prog.* **47** (1951) 91.
18. F. P. BOWDEN and D. TABOR, "The Friction and Lubrication of Solids" (Elsevier, Oxford, 1959).
19. J. S. HARDMAN and B. A. LILLY, *Proc. Roy. Soc.* **A333** (1973) 183.

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