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Solid phase compaction of polymers

R. J. CRAWFORD, D. PAUL
Department of Mechanical and Industrial Engineering, Queen's University of Belfast, N. Ireland

A study has been carried out to establish the feasibility of moulding small products and technical parts by cold compaction of finely powdered polyvinylidene chloride (PVdC) co-polymer, polyvinyl chloride (PVC) and polypropylene. The PVdC material compacts relatively easily and it has been found that the strength of the product is affected by various factors, including the particle size, compaction pressure, peak pressure dwell time and the loading rate. Green strength of 11 MPa can be achieved in PVdC and this may be improved three-fold using a post-moulding heat-treatment. The green strength of cold-compacted polyvinyl chloride is relatively low and heat-treatment in a hot-air oven tends to be detrimental. So far no satisfactory way of compacting polypropylene has been found.

1. Introduction

Polymeric materials are usually produced in powder form, which is then converted into pellets by the application of heat and pressure. Processors take these pellets and utilize, for example, an injection-moulding machine, to form them into components using, once again, heat and pressure. The stages involved are illustrated in Fig. 1 along with two interesting alternatives which may be possible using solid phase compaction techniques.

The first alternative is to produce the plastics components by the direct compaction of powder. There are many technical reasons why a cold compaction process would be very important to the modern plastics industry. As no heat has been applied in the production of the component, the material does not suffer any loss in properties and may be recycled as desired. It is also possible to produce plastic components with much thicker sections than can be obtained by injection-moulding since there are no cooling problems. In addition, many of the new high-temperature polymers are difficult or impossible to process by conventional techniques as they retain their solid nature up to the point of degradation. Other polymers, such as polyvinyl chloride, degrade shortly after softening and require stabilizers and modifiers to facilitate processing. High molecular weight polymers, such as ultra high molecular weight polyethylene (UHMWPE), retain a high viscosity when melted which makes moulding very difficult. Similarly, highly filled polymers, such as polypropylene with greater than 30% limestone ($CaCO_3$), are difficult to mould.

There are also economic factors which favour cold compaction moulding of thermoplastic powders. As the compaction is ideally achieved at room temperature there are considerable energy savings in comparison with alternative melt-processing techniques. It is also envisaged that tools and dies used in the production of compacted metallic components could also be used in the production of plastic components.

The second alternative illustrated in Fig. 1 is to produce the pellets by cold compaction, thereby removing one of the heat/pressure stages in the production of the component. Unlike the previous method, it is not necessary to obtain very high strengths because the pellet only needs to be sufficiently strong to withstand handling. If the technical difficulties involved with the very high production rates can be overcome, this method of pelletizing is potentially very attractive. In particular, it can alleviate problems of thermal degradation and the harmful effects of fumes from additives such as fire retardants. It is envisaged that pelletizing by a cold compaction route would offer considerable savings in energy and capital

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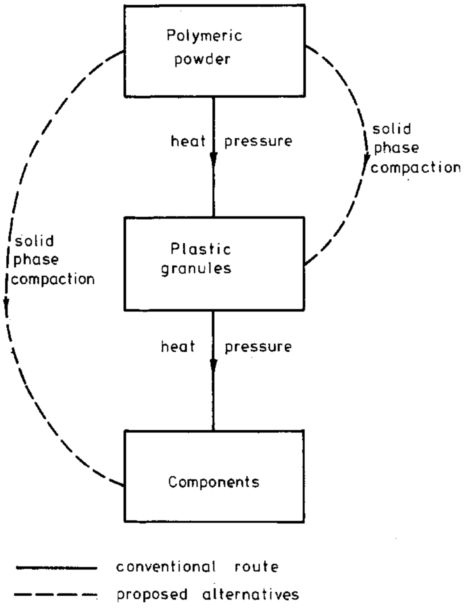

Figure 1 Production of plastic components from polymeric powder.

cost of equipment. It is also likely that the properties of the end product would be improved through the use of a better quality feedstock.

The pressure required to cause the powder particles to adhere together and form a coherent strong mass can be applied in various ways. The simplest method is to place the powder in a die and apply uniaxial compression by means of a ram.

This method has been used by several workers in the compaction of polymeric powders. Maeda and Matsuoka [1] investigated the cold compaction of polyethylene, polyvinyl chloride and polypropylene followed by a heat-treatment in a hot-air oven, boiling water or in a microwave oven. They also evaluated the influence of selected fillers and have demonstrated the feasibility of such a process for commercial components.

Bigg [2] has applied the compaction process to the moulding of thermoset resins, semi-crystalline polymers, and glassy polymers although little or no success could be achieved with the latter.

Jayaraman *et al.* [3] have investigated the pressure powder moulding of glassy polymers, using polystyrene as a model, followed by heat-treatment in a hot-air oven. This raised the mechanical properties of the compact but also caused a significant change in dimensions.

Halldin and Kamel [4] have described a similar process using UHMWPE, while Palmer and Rowe [5] have used polyvinyl chloride compacts to illustrate how mercury porosimetry techniques can help in the understanding of the compaction mechanism.

The objective of the present programme of work at the Queen's University is to examine the parameters affecting the cold compaction moulding process and optimize these in terms of the mechanical properties of the compacts produced.

2. Polymeric powders tested

To facilitate a detailed study of the compaction variables, it was decided to select three materials for the initial programme of work. After discussion with I.C.I. (Plastics Division) the materials chosen were Viclan PVdC co-polymer (VR 558), polyvinyl cloride (PVC 571/102) and a polypropylene co-polymer (HW 607 M). Viclan PVdC co-polymer was chosen because it is difficult to process by conventional methods and yet it has interesting properties in sections thicker than can be achieved by solvent casting. PVC was chosen because of its propensity to thermal degradation and polypropylene was chosen because it is a high tonnage material and it is desirable that any new moulding or pelletizing process should be applicable to it.

2.1. Material characteristics

Since material characteristics such as glass transition temperature (T_g), melt temperature (T_m) and crystallinity, etc., are likely to have an effect on the compaction process, these have been studied for each of the three polymers under investigation.

Tests were performed by differential scanning calorimetry (DSC) to establish the T_g and T_m for each polymer. These traces are shown in Fig. 2. From these it can be seen that Viclan PVdC has T_g at 22° C and further heating shows that this material contains some crystallinity as a double melting endotherm is seen at 115 and 126° C. The glass transition temperature for PVC is about 87° C. The polypropylene material has a high degree of crystallinity exhibiting a small melting peak at about 112° C and a main melting peak at 161° C.

2.2. Particle characteristics

The particle size of powders is of critical importance to the compaction behaviour because it influences the packing density and hence the

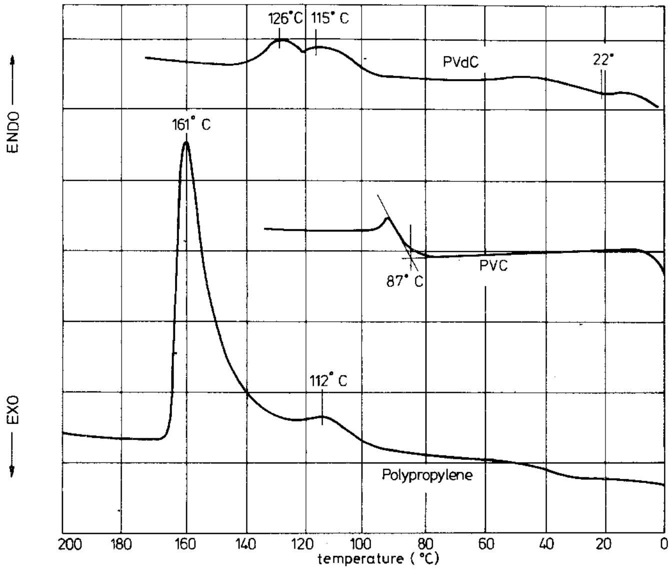

Figure 2 DSC trace for polypropylene PVC and PVdC.

weight of the initial charge of material in the mould. The Viclan PVdC co-polymer, polypropylene and PVC were examined using a scanning electron microscope which showed that the materials consisted of particles of varying size and irregular shape. From the micrograph of Viclan PVdC (Fig. 3) it is estimated that the average particle diameter is 30 μm. It may also be seen from the micrograph that there is considerable porosity and pits on the surface of each particle. Micrographs of PVC and polypropylene (Figs. 4 and 5) indicate a particle mean size of approximately 200 μm. The particles of PVC are much more irregular in shape than those of polypropylene which are almost spherical.

A rapid, automatic determination of particle size distribution can be found from a Coulter Counter (manufactured by Coulter Electronics, Hertfordshire). This size distribution is based on a large number of particles and thus has high statistical accuracy. Viclan PVdC co-polymer was analysed in this way and a particle size distribution ranging from 10 to 90 μm found as shown in Fig. 6.

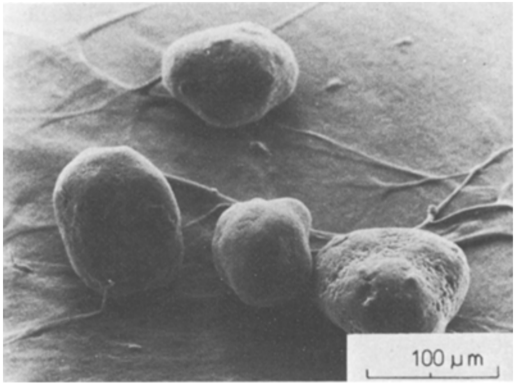

Figure 4 Polypropylene powder.

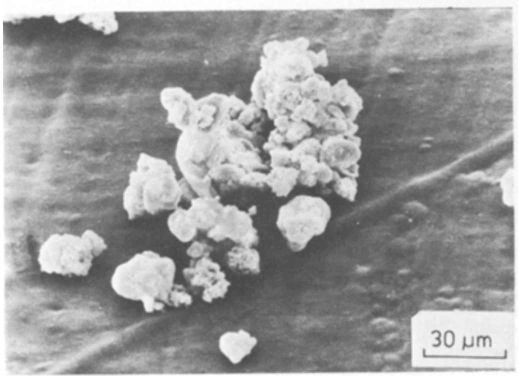

Figure 3 PVdC co-polymer powder

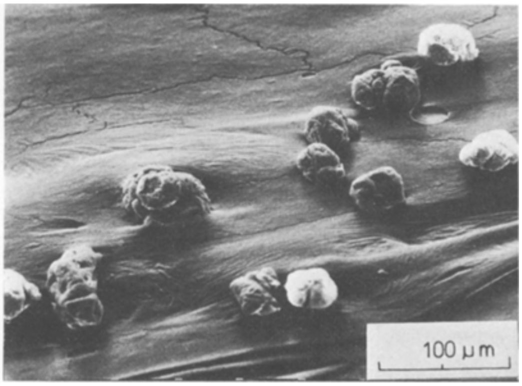

Figure 5 PVC powder.

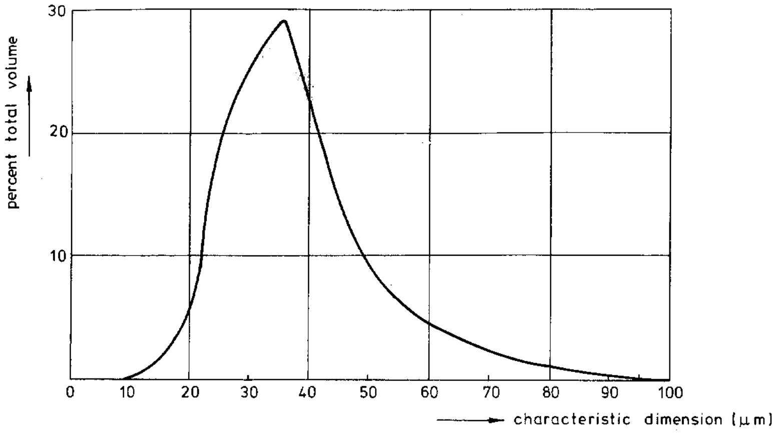

Figure 6 Particle size distribution by Coulter analysis.

TABLE I Initial apparent density of tested materials

Material	Mean particle size (μm)	Apparent density ($g\,cm^{-3}$)
Viclan PVdC	35	0.357
Corvic PVC	200	0.525
Polypropylene	250	0.560

The powder used in all the compaction trials was as received from the manufacturers. However, to ascertain the effect of particle size on compaction, the Viclan PVdC co-polymer was sieved into seven size fractions using an Alpine Classifier (Alpine Machinery Ltd).

The apparent powder density is also important, since it determines the tooling size and storage volume. Apparent density is largely determined by particle size, size distribution and morphology. This was measured in accordance with method 501 A of BS 2872. Table I shows the apparent densities for the three materials under examination.

3. Apparatus and compaction techniques

A die, designed to produce standard • tensile specimens with a pressure area of $400\,mm^2$, was utilized in most of the compaction experiments. The advantage of these specimens (see Fig. 7) is that they can be tested in the compacted state without the need for any machining. This is particularly important in cases where the green compact may be fragile or where it is suspected that any heat produced during machining might modify the properties of the compacted specimen.

The die was mounted on the compacting machine, an Amsler type universal testing machine of 350 kN capacity to achieve static compaction. During each compaction a trace of pressure against time using a chart recorder mounted on the testing machine was taken. A typical example is shown in Fig. 7.

From this plot it can be seen that the three most important parameters during compaction are:

(1) pressure level reached;

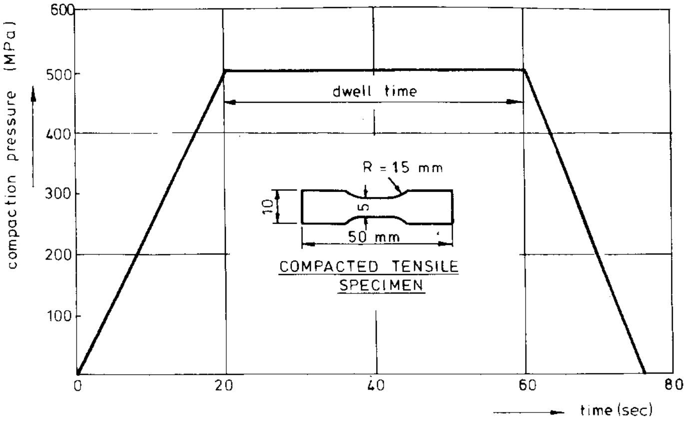

Figure 7 Typical pressure–time profile obtained during static compaction.

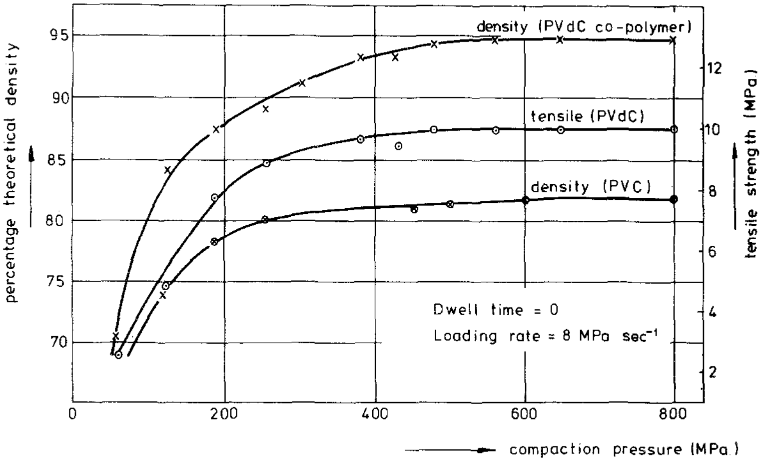

Figure 8 Effect of pressure on density and tensile strength of green compacts of Viclan PVdC co-polymer and PVC.

(2) rate of application of pressure; and

(3) time at peak pressure, i.e. dwell time.

Test pieces were obtained under various conditions of loading rate, pressure and dwell time. The green density was ascertained from measurements of the test-piece volume and weight 24 h after compaction. The time-dependent relaxation of the specimen was also studied over a period of 2 weeks. Tensile testing was carried out on a tensile testing machine at an extension rate of 2 mm min^{-1}.

Some of the green tensile specimens produced under various combinations of the compaction variables were subjected to a heat-treatment in a hot-air oven. The important variables during this heat-treatment were considered to be sintering temperature and duration of treatment. The mechanical properties of the sintered specimens were then measured for comparison with the green specimens.

4. Results and discussion

4.1. Effect of compaction pressure

The effect of compaction pressure on the properties of Viclan PVdC co-polymer and PVC are shown in Fig. 8. It may be seen that in each case there is a terminal densification as the compaction pressure is increased. For PVdC co-polymer this occurs at about 500 MPa, whereas in PVC, terminal densification occurs at 350 MPa. The tensile strength of PVdC co-polymer rises with increase in density and reaches a maximum value of about 10 MPa as shown. The tensile strength of the PVC products were very low, which made testing very

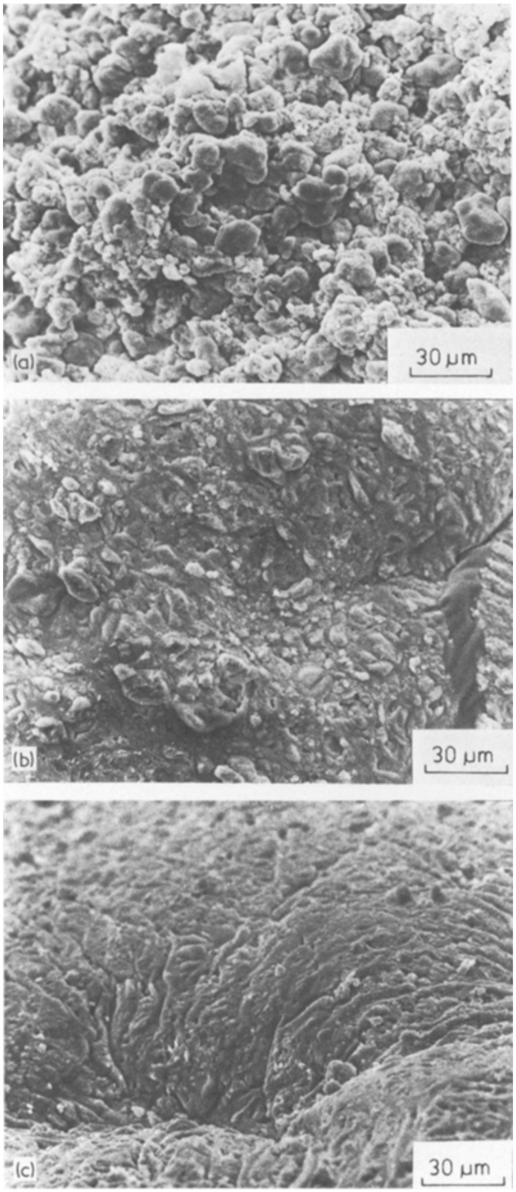

Figure 9 PVdC co-polymer compacted with a pressure of (a) 50 MPa, (b) 310 MPa, and (c) 650 MPa, loading rate 8 MPa sec^{-1} and zero dwell time.

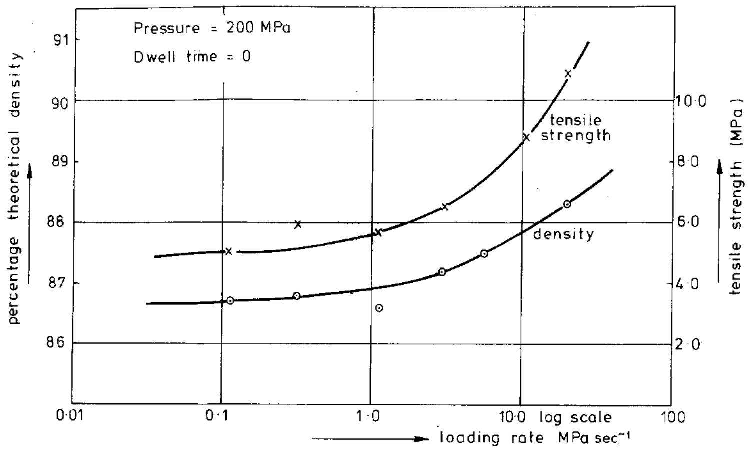

Figure 10 Effect of loading rate on tensile strength and density of Viclan PVdC co-polymer.

difficult and thus no tensile strengths are included.

The particular grade of polypropylene chosen did not respond to cold compaction and even at pressures up to 800 MPa it has not been possible to produce a solid specimen in this material. However, investigations are continuing along several lines in order to improve the compaction characteristics of polypropylene. Since most success has been achieved with PVdC, the initial programme has tended to concentrate on a detailed study of the compaction behaviour of this material.

Fig. 9a to c show the effect of pressure on the microstructure of the compact of Viclan PVdC co-polymer. These scanning electron micrographs substantiate the experimental observation regarding the general increase in tensile strength and density with pressure. In Fig. 9a mutual sliding and reorientating of particles has taken place but discrete particles are very easily seen and there is little or no evidence of frictional heating. The porosity in the compacts seems quite high and the small tensile strength achieved is attributed to electrostatic forces. In Fig. 9b it may be seen that at a compaction pressure of 310 MPa porosity has greatly decreased and the overall average size of particles is smaller. Fig. 9c shows that at 650 MPa there is little evidence from the microstructure of the material that it was ever of a particulate nature.

4.2. Effect of loading rate

The effect of loading rate has been investigated in the static compaction of Viclan PVdC co-polymer and the results are presented in Fig. 10. As the loading rate, to a peak compaction pressure of 200 MPa, is increased from 0.1 MPa sec^{-1} to 20 MPa sec^{-1} the compacted relative green density rises from approximately 1.390 to 1.415. Accompanying this density increase there is also a rise in the green tensile strength from 5.5 MPa to 10.5 MPa. The peak pressure dwell time in all these tests was zero.

Scanning electron micrographs of the microstructure of compacts produced by slow and fast

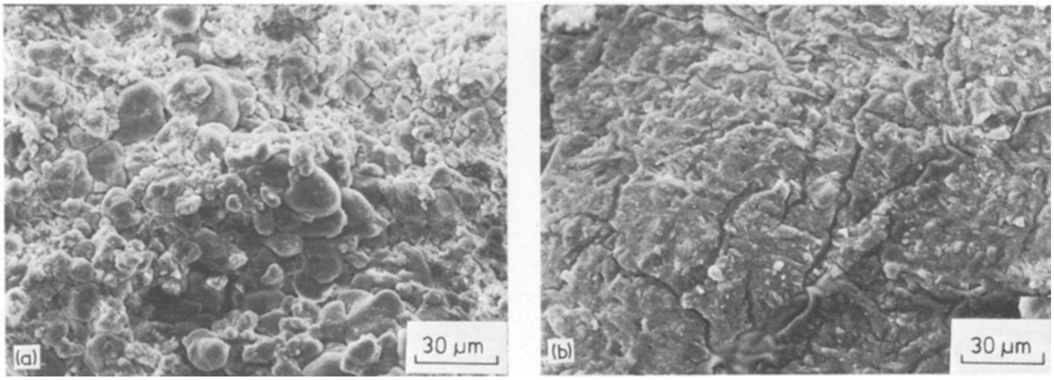

Figure 11 PVdC co-polymer compacted with a loading rate of (a) 0.1 MPa sec^{-1} and (b) 18.5 MPa sec^{-1} at a compaction pressure of 200 MPa, no dwell time.

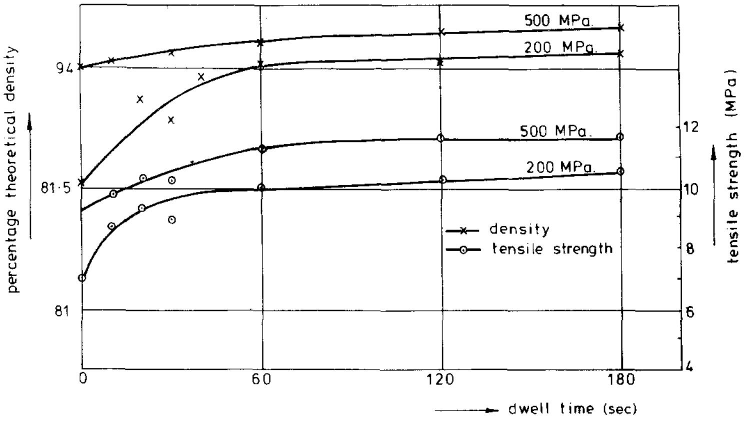

Figure 12 Influence of peak pressure dwell time on density and tensile strength of Viclan PVdC co-polymer.

loading rates are presented in Fig. 11a and b. In Fig. 11a (a slow loading rate), discrete particles are very easily observed. There is no evidence of any flow of the material. This may be because at the slow loading rates any heat generated at the particle interfaces has time to be conducted away into the metal die walls.

In Fig. 11b the microstructure of the compact is barely recognizable as being particulate in origin and it is apparent that there has been some flow in the material. This is probably caused by a substantial rise in compact temperature associated with the more rapid input of energy and smaller conduction of heat to the die walls. This effect must account for the increase in density and increase in tensile strength observed with increased loading rate. From Fig. 10 it may be seen that the tensile strength has not reached a maximum at the highest loading rate available on the Amsler testing machine. It is intended to investigate the effect of further increases in loading rate using a high-speed mechanical forging press and explosive compaction.

4.3. Effect of peak pressure dwell time

Tests were conducted to determine whether or not the dwell time at peak pressure had an influence on the physical properties of the test pieces. Fig. 12 illustrates the effect of peak pressure dwell time on tensile strength and green density for Viclan PVdC. It is apparent that the dwell time under peak pressure is more important at low pressures than at high. At a compaction pressure of 500 MPa with a dwell time of 120 sec the highest green tensile strengths in Viclan PVdC are achieved. This is probably due to increased fusion and more flow between particles.

Fig. 13a and b show the changes in the microstructure of Viclan PVdC when pressed at 200 MPa for dwell times of 0 and 120 sec. At zero dwell time there is little fusion of the particles whereas at a dwell time of 120 sec there is evidence of particle flow and fusion even at this relatively low pressure.

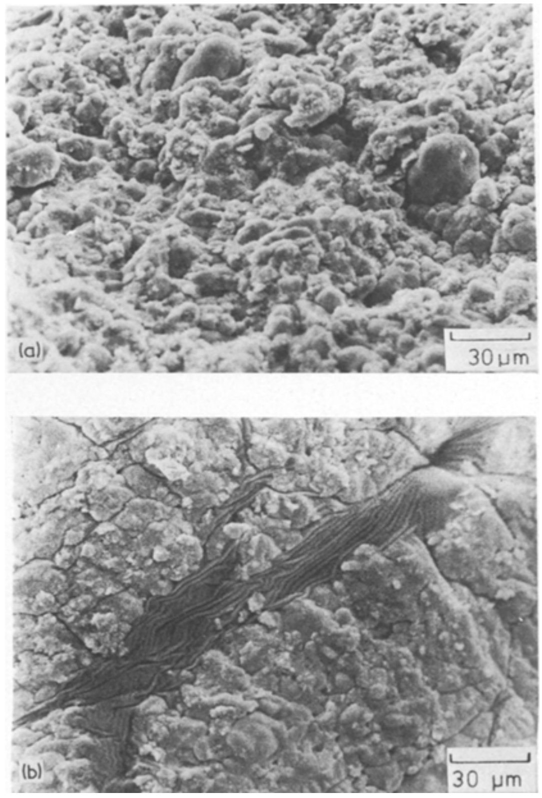

Figure 13 PVdC co-polymer compacted with a pressure of 200 MPa, loading rate 8 MPa sec^{-1} and (a) zero dwell time, (b) 180 sec dwell time.

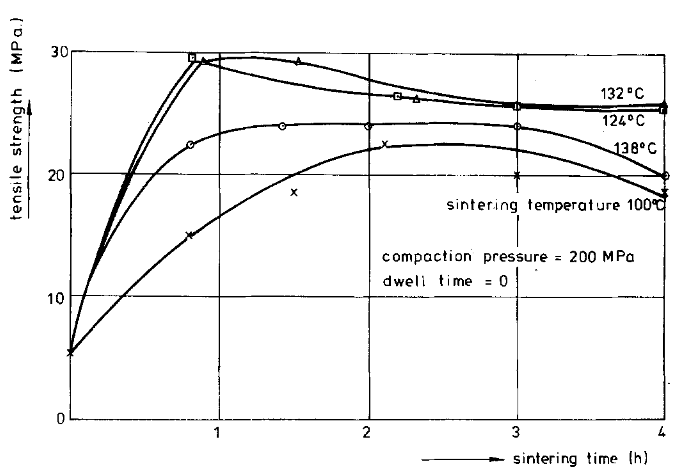

Figure 14 Effect of sintering temperature and time on tensile strength of Viclan PVdC.

4.4. Effect of heat-treatment

Although it is possible to improve the tensile strength of green compacts by increasing the loading rate or the peak pressure dwell time it is apparent that although the maximum value of 11 MPa achieved so far for PVdC would probably be suitable for pelletizing it is still too low for most practical applications. It is essential to increase this value to more acceptable levels and one possibility is to use post-moulding heat-treatment. In the initial study this was carried out in a hot-air oven in the temperature range 100 to 140° C. The effect of this treatment on the tensile strength of Viclan PVdC co-polymer is shown in Fig. 14. This material degrades at temperatures above about 130° C which is borne out by the diminishing returns achieved by sintering at higher temperatures. It is interesting that although tensile strength starts to decrease for sintering temperatures in excess of 130° C, in fact the percentage elongation to break increases three-fold between sintering temperatures of 132 and 138° C. At this latter temperature there are severe bubbling effects on the surface of sintered specimens which are attributed mainly to gas evolution and visco-elastic recovery of the residual stresses induced by the compaction pressure.

In terms of maximum tensile strength, the optimum sintering conditions are obtained at a temperature of 124° C for 45 min (for a compaction pressure of 200 MPa). It has been found that the optimum sintering conditions are influenced by the compaction pressure. For example, using a compaction pressure of 500 MPa had the effect of slightly lowering the optimum sintering temperature. However, there is no significant difference in the sintered strengths of specimens obtained at 200 and 500 MPa. It is apparent, therefore, that high-strength compacts can be achieved by a number of routes. The actual route chosen may be determined from the equipment available or the strength required from the

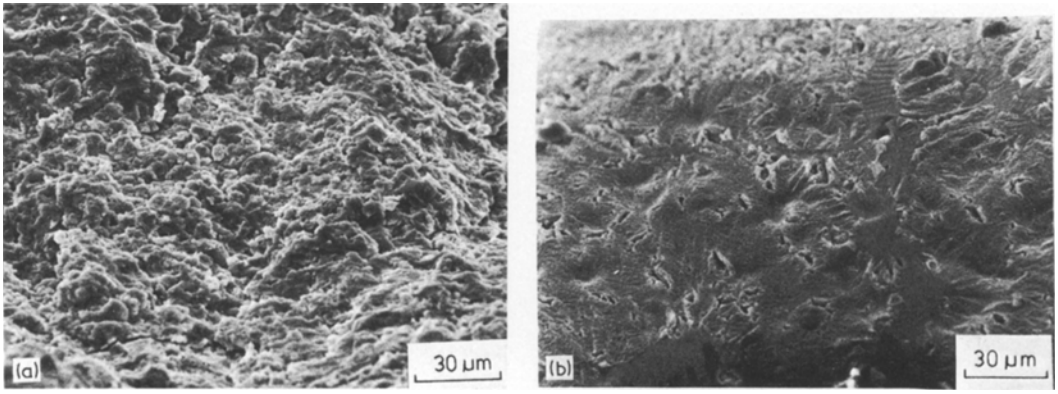

Figure 15 PVdC co-polymer compacted with a pressure of 200 MPa, a loading rate of 8 MPa sec^{-1} and zero dwell time. (a) No sintering treatment, (b) sintered at 124° C.

compact. It is also worth noting that at 500 MPa there is not the same degree of bubbling found on post-sintering examination as was experienced with compaction at 200 MPa, probably because of the reduced internal porosity.

Fig. 15a and b show the microstructure of PVdC before and after sintering. The compaction pressure in each case was 200 MPa with zero dwell time. Fig. 15a shows the green compact and the particulate nature of the structure is evident with little signs of fusion at this pressure. Fig. 15b shows that sintering at 124° C for 45 min improves the homogeneity of the structure and it is undoubtedly the increased flow and fusion between particles which causes the large increase in strength (from 8 MPa to almost 30 MPa at this pressure).

Similar sintering experiments were performed on PVC. Here no significant increase in tensile strength was found by a hot-air oven treatment at temperatures below 87° C, the glass transition temperature of the material. Above this temperature expansion of the compact was observed causing the strength to be reduced rather than enhanced. This is similar to the findings of Bigg [2] on the sintering of glassy materials. The level of porosity in the PVC compacts is quite high (approximately 20%) dure to the large particle size and lack of any flow mechanism during compaction. It is felt that this is a significant factor in the compaction process and if this level of porosity was reduced, an increase in strength during sintering, similar to that reported by Jayaraman *et al.* [3] on polystyrene, might be expected.

4.5. Effect of particle size

To investigate the effect of particle size on the compaction of PVdC co-polymer the powder was classified as described in Section 2. Fig. 16 shows the variation of tensile strength and density with particle size for compaction pressures of 200 and 500 MPa using no dwell time and a loading rate of 8 MPa sec^{-1}. From this figure, it is evident that the tensile strength and density of the green compacts both decrease as the particle size increases.

This may be explained as follows. In a fixed mass, powder with a large particle size will exhibit fewer contacts per unit volume than a powder consisting of smaller particles. As a consequence, at any pressure level the force per contact is considerably greater in the case of the larger particles. It seems reasonable to assume that a stronger compact will result from a large force on the contacts. However, it is also reasonable to assume that the strength will increase with the number of contact points. Thus, as the particle size decreases, two opposing actions are at work on the compact strength. The number of contacts increase which tends to increase the strength, while at the same time the force per contact is decreased which lowers the strength.

These arguments may be used to explain the relationship between tensile strength and particle size shown in Fig. 16. At the finest particle size there is a very large number of contact points and this produces a very strong compact. As the particle size is reduced the number of contacts are reduced but this is compensated by the increased force between the contacts and so there is little change in strength of the compacted specimen. However, as the particle size is increased still further a stage is reached where the decreasing number of contact points dominates. As a result the tensile strength of the compacted specimen decreases.

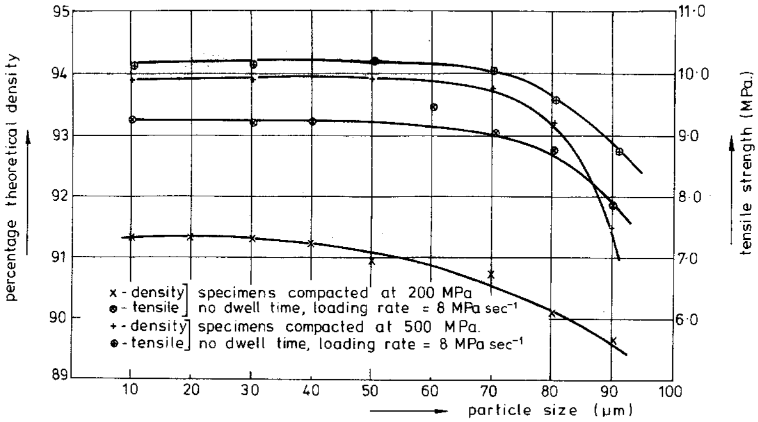

Figure 16 Effect of particle size on density and tensile strength of Viclan PVdC co-polymer.

Maeda [1] and Jayaraman *et al.* [3] have also concluded that a decrease in particle size leads to a stronger compact.

5. Conclusions

The exploratory static compaction tests on three polymeric powders have shown that PVdC (Viclan co-polymer) can be readily compacted into a strong homogeneous structure whereas the grades of polyvinyl chloride and polypropylene used are not amenable to compaction.

From the tests described it may be concluded that:

(1) the density and tensile strength of the green PVdC compacts increase as the pressure and loading rate increase;

(2) an increase in the peak pressure dwell time increases the strength of the PVdC compacts but the effect becomes less significant at the higher compaction pressures;

(3) the strength of the PVdC compacts decreases as the particle size increases;

(4) the strength of the PVdC compacts may be increased by sintering in a hot-air oven;

(5) the ductility of the PVdC compacts increases with sintering temperature but at temperatures above 130° C the considerable increase in ductility is off-set by a reduction in strength due to degradation.

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